Engineered mortar and concrete composites using fibres derived from recycled cardboard

by

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Abstract

The extraction of natural resources for building and construction materials is growing exponentially. Concrete and mortar materials require cement as the primary binder, but cement production negatively impacts the environment. Cement substitution has been studied by many researchers over the years, primarily focusing on industrial wastes as a partial replacement. However, the accumulation of common waste materials is a challenging problem due to the abundance of waste generated across all economic sectors. Due to recent exportation bans, countries are now searching for alternative methods to recycle their common household waste products. Cardboard materials have a limited recycling rate due to the contamination and weakening of the constituent fibres (kraft fibres). This leads to the disposal of cardboard contributing approximately 2.2 million tonnes of waste annually to landfills across Australia. To reduce cardboard waste accumulation, there is potential for kraft fibres (KFs) to be further incorporated into cement-based composite materials. Nonetheless, limited research outputs have been conducted on the successful integration of KFs. KFs are natural fibres which are susceptible to degradation in high alkaline environments, such as cement-based materials. When fibres degrade, the mechanical integrity of the material is compromised, and the service life of the application is often reduced. For the application to be widely accepted in the construction industry, further research is required. As the drive toward the circular economy framework becomes prominent, this research addresses the issue of cardboard waste accumulation and reducing cement consumption in concrete and mortar materials.

This thesis presents a study that integrates matrix and fibre modification techniques on KFs derived from cardboard waste. Silica fume (SF) is used as a fibre pre-treatment, creating silica fume kraft fibres (SFKFs) and metakaolin (MK) as a matrix modifier. Integrating both SF and MK with KFs is a novel concept for minimising the degradation caused by calcium hydroxide in concrete on natural fibres. The target compressive strength was 25MPa, with KFs replacing 5-20% of cement in concrete. Compressive, tensile, and flexural testing was conducted to determine the mechanical capabilities of the novel mix designs. As expected, density and compressive strength declined with increasing fibre integration. However, matrix modification with 5% MK alongside 5% SFKFs increased the tensile strength by 10%. To ascertain the long-term effects of the novel composite, durability experiments were required. These included accelerated ageing simulations of wet and dry cycles, as well as water absorption, water immersion, carbonation, and chloride ion permeability experiments. The ageing simulation results produced a lower mechanical strength for all composite materials, including the control. The integration of 5% MK increased the surface absorption of the fibre composites, whereas 10% SFKF composites exhibited the lowest water absorption at 365 days. Raw KFs had a lower carbonation rate than matrix and modified fibres due to the morphology of the supplementary cementitious materials.

The morphology and microstructure of the fibres were analysed using a scanning electron microscope (SEM) and energy dispersion x-ray spectroscopy (EDS). This application of SF revealed less

degradation on the outer fibre walls, thus increasing service life of the fibre and the corresponding composite. A Fourier transform infrared spectroscopy (FT-IR) was employed to gain insights into the fibres chemical nature. The thermal, calorimetric and combustion attributes of the fibres were measured using thermogravimetric analysis (TGA), differential scanning calorimetry (DSC) and pyrolysis combustion flow calorimetry (PCFC). SFKFs showed a lower heat release capacity, demonstrating a lower combustion propensity compared to raw KFs. Furthermore, a 45% decreased peak heat release rate of SFKFs highlighted the overall reduction in the fire hazards associated with these materials. Moreover, SEM images illustrated advanced petrification on raw KFs compared to SFKFs within the composite. The increase of cement products on the fibre wall contributes to the associated mechanical strength reductions.

In conjunction with the experimental data, life cycle assessments (LCA) and mix design optimisation were used to determine the environmental and economic effects when using waste cardboard KFs. The LCA determined the environmental effect of waste cardboard integration. A sensitivity analysis using a Monte-Carlo (MC) simulation investigated the transportation and energy manufacturing greenhouse gas (GHG) emission variables. Two KF composites were subsequently evaluated. SFKF5 mix design contained 5% KFs and SFKF105 contained 10% KFs with 5% MK. Both composites integrated SF as a fibre modification technique for durability purposes. LCA results of SFKF105 showed savings of 11%, 8%, 4% and 1% for terrestrial acidification potential, global warming potential (GWP), terrestrial ecotoxicity potential (TEP) and human toxicity potential, respectively. SFKF5 revealed savings of 3%, 2% and 4% for GWP, TEP and marine eutrophication potential, respectively. The additional travel requirements of KFs and MK to the cement batching plant for composite production did not surpass the embodied energy and travel emissions of the control. However, this was negated due to the additional energy requirements to manufacture KFs. Optimising the economic and environmental trade-offs via the use of a nondominated sorting genetic algorithm revealed three key regions. Region 1 was the most economical but also created the most carbon emissions. Region 2 was a compromise between both economical and environmental factors and region 3 demonstrated the lowest carbon emissions but also produced the highest costs. Utilising the acquired data from the associated regions produced additional mix designs for mechanical analysis.

The findings of this study provide a detailed understanding of the effect of KFs on cementitious composites when integrated with SF and MK. As a result, material engineers may use the results described herein to incorporate cardboard waste materials into composite designs. The results provide opportunities to utilise the novel composites within low stress grade concrete such as non-structural civil applications, residential slabs, and driveways. In addition, researchers may use similar concepts to integrate other novel waste materials, contributing to the circular economy and enhancing the sustainability of the building and construction sector.

Declaration

"I, Robert Haigh, declare that the PhD thesis entitled *'Engineered mortar and concrete composites using fibres derived from recycled cardboard'* is no more than 80,000 words in length including quotes and exclusive of tables, figures, appendices, bibliography, references, and footnotes. This thesis contains no material that has been submitted previously, in whole or in part, for the award of any other academic degree or diploma. Except where otherwise indicated, this thesis is my own work".

"I have conducted my research in alignment with the Australian Code for the Responsible Conduct of Research and Victoria University's Higher Degree by Research Policy and Procedures.

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List of publications

Based on the research reported in this thesis, the candidate has produced the following papers, which have been published or are under review for publication in various international journals and peer-reviewed conferences.

Journal articles

1. Robert Haigh, Malindu Sandanayake, Yanni Bouras, Zora Vrcelj. 2021. A review of the mechanical and durability performance of kraft-fibre reinforced mortar and concrete, *Construction* and Building Materials, 297, 123759.

2. Robert Haigh, Yanni Bouras, Malindu Sandanayake, Zora Vrcelj. 2022. The mechanical performance of recycled cardboard kraft fibres within cement and concrete composites, *Construction and Building Materials*, 317, 125920.

3. Robert Haigh, Paul Joseph, Yanni Bouras, Malindu Sandanayake, Zora Vrcelj. 2022. Thermal characterization of waste cardboard kraft fibres in the context of their use as a partial substitute within concrete composites, *Materials, 15.*

4. Robert Haigh, Malindu Sandanayake, Yanni Bouras, Zora Vrcelj. 2023. Durability performance of waste cardboard kraft fibre reinforced concrete, *Journal of Building Engineering, 67*.

5. Robert Haigh, Yanni Bouras, Malindu Sandanayake, Zora Vrcelj. 2023. Economic and environmental optimisation of waste cardboard kraft fibres in concrete using nondominated sorting genetic algorithm, *Journal of Cleaner Production, 426.*

Conference papers

1. Robert Haigh 2022. A comparative analysis of the mechanical properties with high volume waste cardboard fibres within concrete composite materials, *Australian Structural Engineering Conference,* 9-10 Nov 2022.

Journal articles under review

1. Robert Haigh, Malindu Sandanayake, Yanni Bouras, Zora Vrcelj. 2023. A life cycle assessment of cardboard waste in stress low-stress grade concrete applications, *Journal of Environmental Management*. (Under review)

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Details of included papers: Thesis by publication

Please list details of each scholarly publication and/or manuscript included in the thesis submission. Copies of published scholarly publications and/or manuscripts submitted and/or final draft manuscripts should also be included in the thesis submission.

This table must be incorporated in the thesis before the Table of Contents.

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Contents

Chapter 1 – Introduction

Cementitious composites have been extensively used in infrastructure projects due to the long service life, low maintenance and high compressive strength. However, the characteristics of the material also include a low tensile strength and low cracking resistance [1]. Therefore, it is common practice to include reinforcement to mitigate these disadvantages. The application of steel reinforcement is a common construction method when enhancing the tensile strength of concrete structures. However, it is not always appropriate depending on the final size and locality of the application. The last several decades have seen a rise in the use of various fibres such as; mineral, natural, glass and plastic within composite materials [2]. Fibres can enhance the reinforcement methods of cement-based materials and reduce the growth of cracks caused by tensile stresses [3]. Natural fibres can be an environmentally beneficial alternative than their synthetic counterpart. Due to climate change, the building and construction industry are urgently searching for alternative methods to become more sustainable. Cement- based materials are a primary concern of the industry due to the excessive greenhouse gas (GHG) emissions created to manufacture cement. To lessen the burden of extracting virgin materials, it is critical to recruit natural fibres from other sources such as waste products. Currently, there is an oversupply of waste created from cardboard materials, where a large volume will result in landfill [4]. Therefore, the utilisation of natural fibres within cardboard products is a viable opportunity to close the loop of resource extraction with a waste material.

Recent studies have demonstrated the benefits of including natural fibres in composite materials [1, 5-7]. However, less research focus has been directed towards the use of kraft fibres (KFs) derived from waste cardboard. The use of short fibres within composite materials has shown increased tensile strength as well as reducing the signs of implied stresses such as cracks. However, there is a decrease in compressive strength due to increased fibre content. It has been suggested that integrating supplementary cementitious materials (SCMs) can compensate the reduction in mechanical strength due to their effective pozzolanic reaction [1, 8-12]. *Savastano et al.* [13] showed further benefits of SCMs with reduced energy manufacturing requirements, decreased carbon dioxide (CO_2) emissions and lowered associated production costs. Research conducted by *de Gutiérrez et al.* [8] shows the transferability of incorporating various SCMs such as; silica fume (SF), ground blast furnace slag (GBFS), Fly ash (FA) and metakaolin (MK). However, the mentioned SCMs have different levels of the pozzolanic reaction and there has not been a determined fibre mix design that is effective across all requirements of a composite material.

Several researchers [14-17] studied the behaviour of KFs in fibre cement board composites. It has been shown that the fibres degraded even when other SCMs were integrated. The researchers incorporated an optimum fibre amount of 8%, 4%, 4% and 8% respectively [14]. However, only *Mohr et al.* [15] used SCMs of SF, MK and GBFS. Their research showed the optimum levels of SF and MK consisted of 30%, while 90% GBFS only showed increased durability factors after wet and dry cycling. *Khorami et al.* [16] integrated waste cardboard and glass fibres within a composite material ;however there has not been a comprehensive study of solely using waste cardboard fibres and other additives to enhance the waste material. Other research has been introduced with concrete composites and *Booya et al.* [18-20] show the effectiveness that kraft pulp fibres can have as a reinforcement agent. However, the literature has not shown a combined overview to mitigate the total degradation effects created. Fibre pre-treatment as a surface modification technique can allow fibres to integrate without this affect. However, this has not been measured previously within concrete composites. The authors predominantly used engineered chemically and mechanically modified fibres which aren't commercially available. Currently, there is a lack of literature on the effects that recycled KFs may have on cement-based composites when surface modification and SCMs are integrated.

The current research on KFs builds upon previous studies and conducts an original investigation when integrating fibres developed from recycled cardboard. It has been demonstrated that this application has potential; however, for successful integration, an effective pre-treatment and chemically altered cement matrix are required. There is limited research available on how to create an ideal material environment to optimize fibre performance. By integrating current knowledge, this research study will strengthen those ideas and develop a comprehensive overview of how to obtain an effective composite matrix for recycled KFs. The mitigation of the negative effects associated with waste cardboard provides an opportunity to diversify current waste management strategies and conform a more sustainable alternative.

1.1 Research significance

The impact of industries on the environment is becoming increasingly important across all sectors of the economy. The endeavour to reduce carbon emissions is evolving, with the larger influences on climate change being at the forefront. These industries include construction, agriculture, transportation, and manufacturing. Research conducted by *Malviya et al.* [21] states that the reduction of synthetic fibre manufacturing can aid towards the minimisation of climate change effects. The use of natural fibres can contribute to the reduction of synthetic products. In addition, the extraction of virgin materials can be reduced, thus reducing further carbon emissions. Similarly, it is critical to repurpose manufactured waste materials for further use in order to reduce the negative impacts on the environment.

Cardboard products are extensively used for common consumer goods; however, the life cycle of the final product is often short. As of 2020-2021, Australia generated approximately 5.8 mega tonnes of paper and cardboard waste, of which 62% was recycled and 38% was disposed of in landfills. Due to the decline in paper and cardboard generation due to online services, the recycling rate also decreased from 66% to 60%, resulting in landfill rates increasing from 34% to 38% [22]. The sheer size and volume of paper and cardboard produced makes them the largest recyclable waste, but also the largest contributor to landfills. [4]. Due to the ban on waste imports from China, significant changes have been made to waste management in Australia. Australia exported 4.4 million tonnes globally, of which 1.4 million tonnes were shipped exclusively to China. Paper and cardboard accounted for 79% of all materials, or 1.1 million tonnes. This figure represents approximately 65% of the total export market for the material in 2017 [22, 23]. Moreover, 35% of all recycled cardboard is too short in size and brittle and will end up in landfill [24]. Therefore, there is an urgent need to develop effective measures to minimise the negative impacts by reusing the materials in alternate applications.

A limited amount of research has been conducted on the incorporation of recycled KFs into cement-based materials, and mainstream practices have not yet been developed. This is primarily due to the difficulty of integrating the hydrophilic fibre material into the hydrophobic environment of common cement mixes [25]. A significant limitation in the integration of natural fibres is the degradation of fibres caused by the high alkaline and mineral-rich properties of cement. [18]. Therefore, a contemporary solution is required to mitigate the negative effects caused by integration of natural fibres for further use. The objective of this research is to develop cement-based composites with recycled cardboard fibres as a reinforcement agent.

Consequently, there will be a reduction in the need to extract virgin materials by reducing the manufacturing of inorganic fibres. Therefore, utilising a waste product that has already been created will reduce the burden on the recycling industry and the environment.

1.2 Research aims

Integration of cardboard KFs cannot be benchmarked against the research currently being conducted on other natural fibres. This is because the lignin and hemicellulose components of KFs within cardboard are mostly removed during the manufacturing (kraft) processes. Therefore, the mechanical behaviour of the fibres when used in conjunction with other materials needs to be thoroughly investigated. This research will experimentally develop and characterise recycled cardboard fibre reinforced concrete derived from waste cardboard. Thus, there will be a combined opportunity to reduce the extraction of virgin materials and the generation of waste. In this way, the research outputs will contribute to environmental, economic, and social benefits that can further facilitate sustainability in the construction industry. The construction industry could also benefit from these innovative practices by changing negative perceptions regarding its consumption of virgin resources.

The aims of the research can be stated as follows:

- Reduce cement consumption in mortar and concrete applications via the integration of recycled KFs.
- Enhance the viability of using recycled cardboard within the building and construction industry.
- Develop a mix design for effective usage of cardboard waste in cement-based composite materials.
- Experimentally examine the physical, mechanical and durability properties of the bespoke cement-based composite.
- Analyse the environmental and economic impacts and benchmark the sustainability aspects of the novel cement-based composite.
- Develop a durable novel cement-based composite that will comply with current building codes and standards within the construction industry.
- Measure the flammability and combustion rate of modified KFs, ensuring the fibre modification technique improves the fibres characteristics for further use in construction.

1.3 Thesis layout

This thesis consists of nine chapters. Each chapter undertakes a focused investigation on the critical aspects of KF integration within concrete and mortar materials. The combination of the varied findings in the subsequent chapters creates a holistic approach to solve the research problem. In **Chapter 1** natural fibre reinforced composites and the limitations associated with using natural fibres in cementitious composites are introduced. In addition, the research significance, and research aims were stated.

In **Chapter 2**, a bibliometric analysis identified a literature gap regarding the use of KFs derived from cardboard waste. The literature review discusses current limitations associated with the integration of KFs into cementitious composites. The review identifies methods that can enhance the fibres durability and improve the mechanical performance of composite designs. The limitations of the research are discussed, and areas of future research focus are identified.

Chapter 3 and **Chapter 4** demonstrate the mechanical performance of KFs derived from waste cardboard within concrete composites. There were multiple mix designs created, including the use of raw KFs, modified KFs using SF and matrix modified composites using MK. **Chapter 3** presents the experimental study of concrete composite mixes featuring the incorporation of 5% fibres. **Chapter 4** centres the attention on the utilisation of 15-20% KF integration. Each experimental program pertained the measurements of compressive, tensile, and flexural strength, highlighting the effect when using waste cardboard KFs in concrete materials. Scanning electron microscope (SEM) demonstrated sufficient SF adhesion on the fibre walls, while the energy dispersive x-ray spectroscopy observations showed adequate dispersion of all composite materials.

Chapter 5 presents the thermal characterization of KFs with an additional mechanical analysis when using 10% KFs within concrete composites. The mechanical results recorded compressive, tensile, and flexural strengths when 10% raw and modified KFs were integrated. The morphology of the fibres was illustrated using a SEM with an energy dispersion x-ray spectroscopy provision demonstrating SF adhesion to the fibre walls. Fourier transform infrared spectroscopy (FT-IR) was employed to gain insights into the chemical nature of the fibres. The thermal, calorimetric and combustion attributes of the fibres were measured using thermogravimetric analysis (TGA), differential scanning calorimetry (DSC) and pyrolysis combustion flow calorimetry (PCFC).

Chapter 6 presents the accelerated aging tests for the evaluation of the durability performance of KF reinforced concrete. The experimental program of accelerated ageing consisted of wet and dry cycling at 20-, 40-, 60-, 80- and 100-day intervals. Mechanical tests were then completed after each ageing cycle to demonstrate the durability of KFs within concrete composites. To complement the findings shown in the accelerated ageing experiments, water immersion, absorption, carbonation, and chloride ion permeability tests were also conducted at 28, 56 and 90 and 365-day intervals. Chloride ion penetration testing demonstrated the porosity of the composites and the ability of ions to transverse within the microstructure of the composites.

Chapter 7 examines the life cycle assessment (LCA) of KFs within concrete materials. The LCA was conducted using the software program "OpenLCA". This chapter presents three key stages of methodology to evaluate the viability and environmental effects of the waste material in concrete. First, the mechanical strength of KFs as a partial cement substitute within concrete materials was evaluated. Secondly, the LCA outputs on key impact categories were analysed. Finally, a sensitivity analysis was conducted on the variability of GHG emissions produced from transport and energy usage. The LCA measured 1m^3 of two novel composites using 10% SF modified fibres with 5% MK as partial cement substitute. The second LCA measured 5% cement substitute with SF modified fibres. Measured impact category results included terrestrial acidification potential, global warming potential, terrestrial ecotoxicity potential, marine eutrophication potential, human carcinogenic toxicity potential, stratospheric ozone layer depletion potential, and mineral resource scarcity. The Monte-Carlo (MC) simulation results provided further information on two variability factors when integrating the bespoke materials within the mix design. Firstly, the measurement of transportation emissions with a range of 10-150km, and secondly, the production emissions including the energy sources and operational machinery.

Chapter 8 presents a multi-objective optimisation using nondominated sorting genetic algorithm, also known as "NSGA-II". This chapter investigates the most optimal amount of KFs to integrate within concrete materials in relation to two objective functions: cost and greenhouse gas emissions. The findings demonstrate the most optimal amount of KFs for concrete integration based on the objective functions. Mechanical testing was conducted on the optimal mix designs obtained from the optimisation results.

Chapter 9 concludes the thesis by presenting the current study's major findings, novel contributions, and direction for future research.

Chapter 2 – Literature Review

2.1 Introduction

The purpose of this chapter is to provide a comprehensive review of the literature pertaining to the mechanical and durability performance of KF reinforced mortar and concrete. Based on a bibliometric analysis, cardboard waste materials rarely appear in concrete and mortar construction materials. Key findings of the literature review demonstrated that the composite matrix and fibre pre-treatment must be modified to enhance the integration of KFs within concrete and mortar materials.

This chapter contains the following paper:

Robert Haigh, Malindu Sandanayake, Yanni Bouras, Zora Vrcelj. 2021. A review of the mechanical and durability performance of kraft-fibre reinforced mortar and concrete, *Construction and Building Materials*, 297, 123759.

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DECLARATION OF CO-AUTHORSHIP AND CO-CONTRIBUTION: PAPERS INCORPORATED IN THESIS

This declaration is to be completed for each conjointly authored publication and placed at the beginning of the thesis chapter in which the publication appears.

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2. CANDIDATE DECLARATION

I declare that the publication above meets the requirements to be included in the thesis as outlined in the HDR Policy and related Procedures – policy.vu.edu.au.

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Signature Date

3. CO-AUTHOR(S) DECLARATION

In the case of the above publication, the following authors contributed to the work as follows:

The undersigned certify that:

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2. They take public responsibility for their part of the publication, except for the responsible author who accepts overall responsibility for the publication;

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A review of the mechanical and durability performance of kraft-fibre reinforced mortar and concrete

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HIGHLIGHTS

- Mechanical values of Kraft fibre composites are often enhanced with supplementary cementitious materials.
- Fibre modification can reduce fibre mineralisation on natural fibre cell walls.
- Waste cardboard remains seldom considered as a partial filler in concrete and mortar materials.

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ABSTRACT

The building and construction industry is one of the leading generators of waste in the worldwide economy. Use of excessive quantities of virgin materials to manufacture building materials is a growing dilemma that needs urgent attention. With the excessive general of waste, research focus has been directed toward the use of waste to substitute and reduce the requirement for immense extraction of virgin materials. After glass and plastic, cardboard is considered as the most prominent recycled waste material that could possess potential use in mortar and concrete applications, thereby reducing virgin material extraction. The current study aims to conduct a systematic review in using cardboard waste in mortar and concrete. A bibliometric assessment of 874 research publications demonstrated that cardboard waste related studies on mortar and concrete remain seldom considered. An analysis of literature indicated kraft fibres within cardboard can be recycled into building materials. The key findings discovered matrix modification and fibre pre-treatment are essential for the enhancement of mechanical and durability properties. Researchers have developed mix designs including supplementary cementitious materials (SCM) to mitigate fibre degradation and enhance mechanical values. However, further research is required to comprehensively analyse an effective material matrix to reduce the degradation caused on the fibre. Therefore, this paper presents key findings of current trends, limitations and future research directions related to the use of recycled cardboard in concrete and cement-based materials.

1. Introduction

Minimising the negative impacts of the construction industry on the environment has been a research focus for many decades. The construction industry is one of the largest sectors that contribute to waste generation in the world [1,2]. Islam et al. [3] highlight that 30–40% of all waste created is due to construction and demolition (C&D) works, even more so in Australia (44%). During the construction phase, a vast proportion of materials used contains a percentage of waste, whereas demolition works can contain 89% waste from all derived materials [4,5]. Various research studies have been conducted on the causative factors and preventive measures of waste generated [5]. These studies found that waste can be linked during all construction phases including; design, procurement, construction activities and lifecycle of the project. Many construction companies have adopted measures to reduce the materials required on site using methods and software such as the lean approach, prefabrication, BIM and CAD. However, these measures are not always suitable due to the fragmented nature of construction activities and the unique nature of each construction project. Although, in recent years BIM technology is becoming increasingly efficient, it is yet to be utilised effectively across the industry, especially in residential building construction.

Households are the third largest industry of waste generation, following construction and manufacturing [6]. The growth in population correlates to the consumption of natural resources as well as the generation of waste materials. Currently, the world

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0950-0618/

generates around 2 billion tonnes of solid waste annually with the forecast of waste materials to increase by 70% globally by 2050. Solid waste treatment and disposal centres generated an estimated total of 1.6 billion tonnes of carbon dioxide (CO_2) equivalent greenhouse gas emissions in 2016. If no improvements are made, this figure is set to increase to 2.36 billion tonnes by 2050 [7]. More than 50% of waste generated from households will result in the addition of landfill [8]. However, countries like Sweden have banned the waste release to landfill, with 98% of the waste being reused or recycled. The waste materials are processed back into useable products or transferred into an incineration system to generate heating and electricity. One tonne of incinerated waste could generate one hour of power for 900 Australian households [9]. Although this system significantly reduces environmental issues of waste disposal, it does not reduce the extraction of virgin materials.

The building and construction industry consumes approximately 31% of all extracted natural resources worldwide [10,11]. Therefore, a contemporary solution must be addressed to minimise the negative effects of waste and resource extraction. Researchers [11–13] have indicated to sustain a viable solution, there must be a direction toward a circular economy. The framework of a circular economy is a system that eliminates a material "end of life" by keeping the material or product "in flow" via effective re-use strategies and methods. This reduces the extraction of virgin materials as well as mitigating the negative effects of waste accumulation [12]. Redirecting waste into building construction products and materials could significantly address both the issues of waste diversion and reduce the extraction of virgin materials. This will have the potential to alleviate environmental issues across the major contributing sectors.

Local Governments are required to combat the increase of waste generation with more sustainable measures. Increasing the cost of waste deposited at local council and landfill sites has been effective in Hong Kong and however, as a result, other issues of illegal dumping increased [14]. Maximising the economic benefits with recycling has shown to minimise waste with materials, especially with materials such as steel and concrete [3,4]. The motivation for improving the environmental impacts are often linked to financial incentives. This is further shown with an annual generation of 8.7 million tonnes of concrete aggregate stockpiled due to financial incentives from local Governments within Australia [15]. The issues of waste development can be drawn to the end-of-life application of the materials. Wu et al. [14] classified waste materials as inert and non-inert components. The list of common construction waste includes timber, metal, paper, cardboard, plastic, rubber, textile, fiberglass, nylon and domestic waste. Inert materials are more commonly reused in land reclamation and site formation works. Those materials include concrete, rubble and soil. However, non-inert materials that are not suitable for land reclamation often end its life cycle in landfill. Those materials include cardboard, fibreglass, nylon, plastic, rubber and textiles. After plastics, cardboard is the highest contributor to landfill [16] and with heavy previous research focus on the former waste type, reusing cardboard is gaining significant popularity in research fields [17]. Research of cardboard waste has remained relatively static due to the concept the material is completely recyclable. However, the progression of experimental research is indicating the fibres contained within cardboard (kraft fibres) can be further used in composite designs. This is shown with researchers using the material as a reinforcement agent or partial filler in both plastic and cement matrix composites.

2. Research significance and methodologies

Fibre composites have been a part of building and construction materials for several decades with synthetic materials at the forefront due to enhanced mechanical benefits [18]. Nowadays, there is a focus towards sustainability with natural fibres (NFs) becoming increasingly researched. The successful integration of NFs within cement-based composites can reduce the manufacturing of other non-natural fibres. There are several research review articles on NFs within composite materials [19,20]. However, none have focused primarily on the integration of kraft fibre (KF). Although many research articles focused on kraft fibres/ pulp, the limitations and barriers have not been explicitly detailed. KFs and NFs have similar reactions when applied within a cement-based composite however, the removal of key amorphous components such as lignin and hemicellulose of NF cause the KFs to react differently when compared to virgin NF. Therefore, the outcome of this review will enable readers to identify current composite design research of KFs and understand the results of those designs via mechanical and durability analysis. This will further allow stakeholders to identify and understand the factors that affect the successful integration of KFs for further use within composite designs.

This research study will begin with a focus on waste fibres used within the building and construction industry and cement-based composite materials using a bibliometric assessment. A bibliometric assessment is a comprehensive science mapping analysis of research publications. The bibliometric assessment is conducted via a bibliography analysis software called "Bibliometrix". The software can highlight current trends of research as well as identifying knowledge gaps within published literature [21]. The search engine used for sourcing publications was Web of Science "WoS", using two main criteria points of "Waste fibre* in Building construction" and "Waste fibre* in cement and concrete". The asterisk was included to broaden the search by finding words that start with the same letters. This is commonly adopted by researchers using the search engine WoS and can also be known as a "wildcard". The time period analysed ranges from 2000 to 2020. The results of the WoS search included journal articles, books, book chapters and conference publications which have researched waste fibre materials used in construction as well as cement and concrete applications. The output of the review is a systematic approach of current published literature as well as the inclusion of results and observations obtained from the bibliometric assessment, as illustrated in Fig. 1. The review aims to highlight current waste fibres currently used in the building construction industry and conduct a thorough viability analysis of mechanical and durability properties of a waste material currently not being utilised.

3. Bibliometric analysis findings

The main information derived from the bibliometric assessment is shown in Table 1. The assessment reviewed 874 documents from 362 journal and book sources. There were 2393 authors and 3232 author appearances. These values represent the numerical distribution between articles, book chapters, review and proceedings paper of the authors and co-authors. The average number of authors per document is represented at 2.74, with co-author ratio calculated via author appearance and documents. This value correlates with the collaboration index as represented at 2.83, indicating the mean number of authors in a multi authored article. These values represent a high collaboration on published research within the fields of waste fibres in building construction and waste fibres in cement and concrete materials.

Fig. 2 illustrates the most frequently published journal sources with the number of documents published. Journal of Construction and Building Materials is the most prominent publication source with the highest publications since 2000. The Journal scope and high number of publications indicate a prominent research focus of experimental studies to improve the integration of waste fibre that can be used within building materials. However, with the

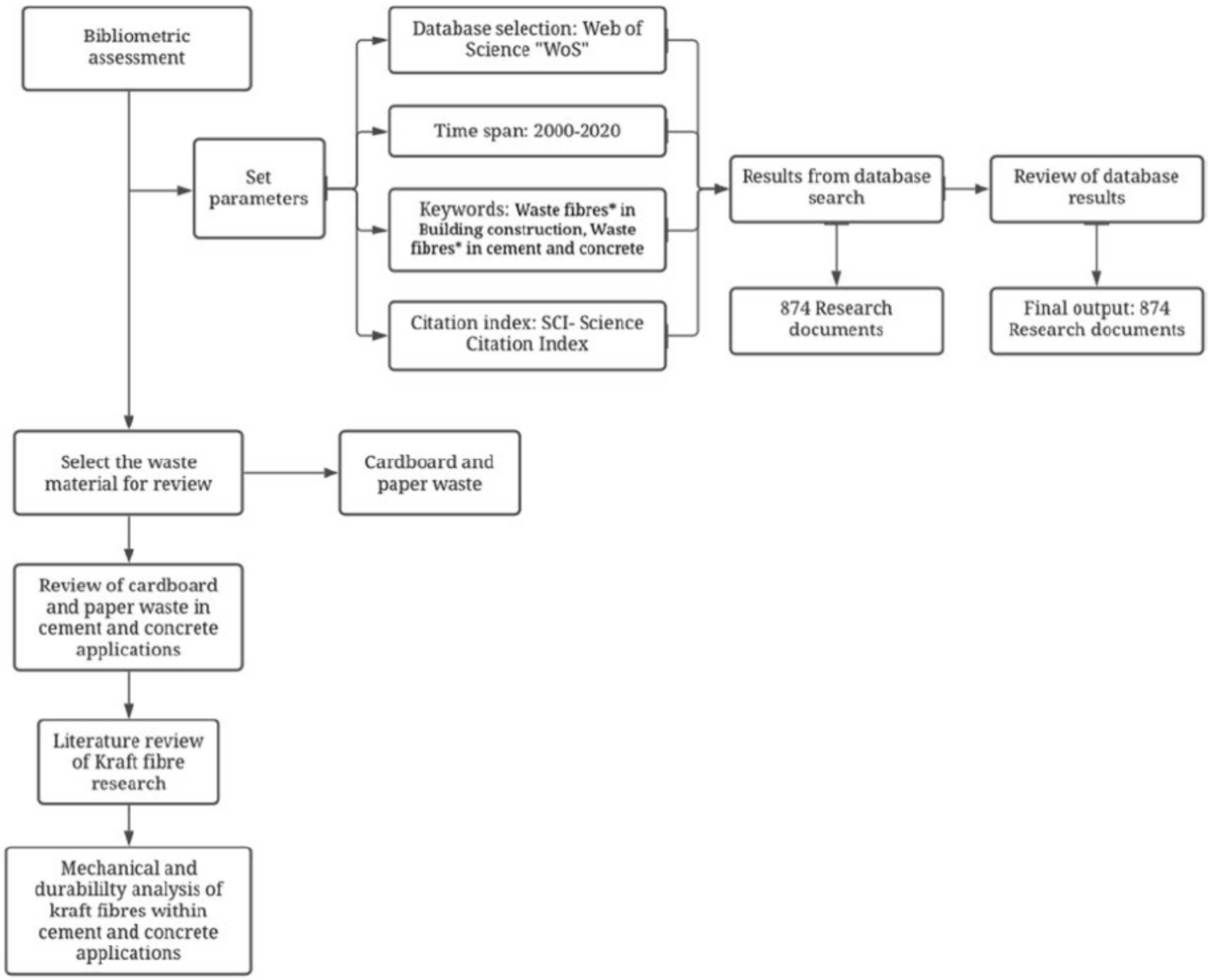

Fig. 1. Research methodology.

Table 1
Main information derived from the bibliometric analysis.

Description	Results
Period	2000–2020
Documents	874
Sources (Journals, Books, etc.)	362
Keywords Plus (ID)	1258
Author's Keywords (DE)	2303
Average citations per documents	12.58
Authors	2393
Author Appearances	3232
Authors of single-authored documents	36
Authors of multi-authored documents	2357
Single-authored documents	42
Documents per Author	0.365
Authors per Document	2.74
Co-Authors per Documents	3.7
Collaboration Index	2.83
Document types	
Articles	620
Article Book Chapter	9
Article Early access	12
Proceedings paper	170
Article Proceeding Paper	11
Review	49
Review early access	3

focus of using natural materials within the building and construction industry, the number of publications in the Journal of Cleaner Production is subsequently rising.

There is an exponential growth of publications in the last 3 years (2017–2020), as illustrated in Fig. 3, which can be related to the further research focus toward sustainability of waste materials within the industry. Published articles were in single digits from the year 2000–2008, with a steady rise following 2009. This can be linked with an increased global awareness of climate change, with significant concerns over air pollution at the 2008 Summer Olympics held in Beijing [22]. In 2009, the Environmental Protection Agency of the United States announced that greenhouse gases endanger the health and welfare of American citizens by contributing to climate change [23]. Following the announcement, the next 8 years exemplified a steady increase in publications until 2017, where publications rose from 55 to 103, then followed with an exponential upward trend to 157–150 annual publications. The significant increase of research can be attributed to the 2030 Agenda for Sustainable Development set out by the United Nations [24]. The previous years' saw the development of the Paris Agreement on Climate Change which set out an urgency among nations around the world. With these global events, researchers identified gaps in literature and experimental studies rose with the aim of reducing greenhouse gas emissions at the forefront.

The thematic word map of the bibliometric assessment is shown in Fig. 4. The data analysed common word occurrences throughout each publication, highlighting the variations of waste materials and other key research focus areas. Due to the search criteria containing cement and concrete, a significant amount of published research articles refers to the topics of mechanical

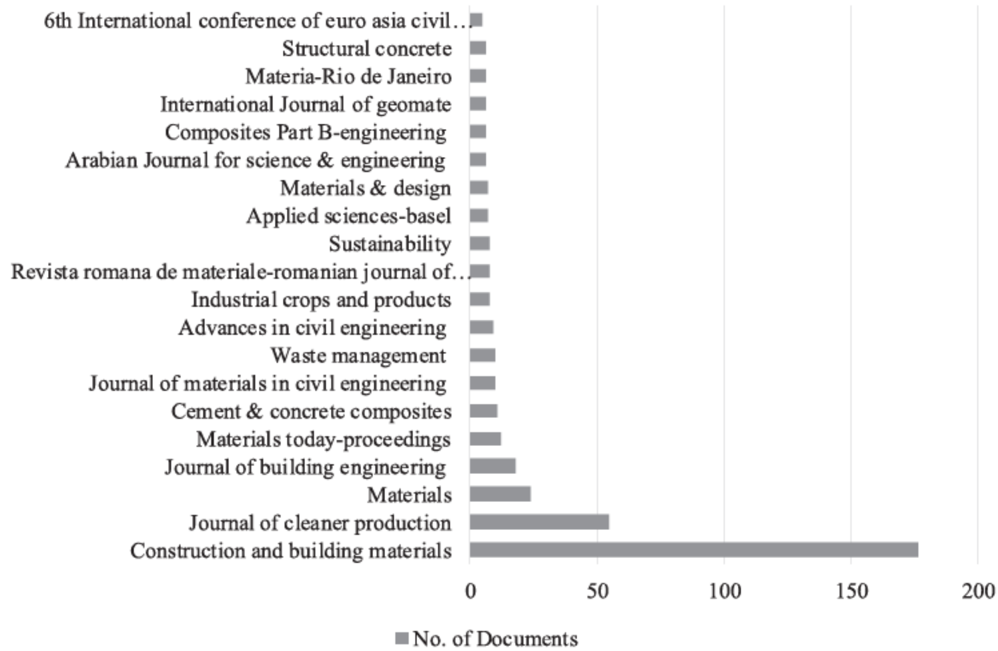

Fig. 2. Publishing source vs number of publications.

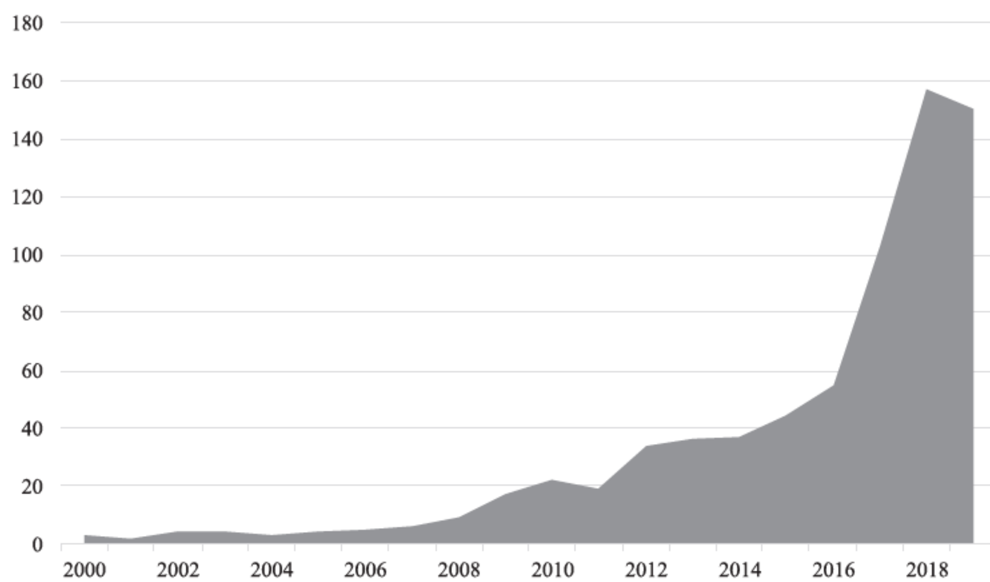

Fig. 3. Annual scientific publications on waste fibres used in building construction materials.

properties and other material mechanics such as durability, workability, compressive and flexural strength. Although extensive research has been focused on the integration of Ground blast furnace slag (GBFS) in cement and concrete materials, the bibliometric assessment has focused primarily on the integration of fibres used with or without partial supplementary cementitious materials (SCM)s. As shown in Fig. 4, Fly Ash (FA) has been the prominent experimental research focus of a partial cement replacement. Glass, plastic, steel, tyre and rubber waste are the other waste materials that are strongly researched areas in building construction materials. In recent years, these materials have mainly used as a filler material in roads and pavement sub layers [25–27]. NFs and cellulose were also mentioned however, only sisal fibres were specifically targeted as the NF used. KFs that are derived from NFs used in the manufacturing of cardboard was not mentioned in this assessment. With cardboard and paper waste significantly contributing to landfill worldwide [28], the bibliometric assessment has indicated a gap in literature for the further use of KFs derived from those waste materials.

3.1. Summary of findings

The bibliometric assessment shows the following key observations and results.

- The results from the bibliometric assessment has shown a prominent research focus toward experimental designs with minimal research attention toward the sustainable and economic benefit of redirecting waste in construction materials.
- When waste materials are integrated as a partial cement replacement, the bibliometric assessment has shown that Fly ash has been heavy researched material when integrated in

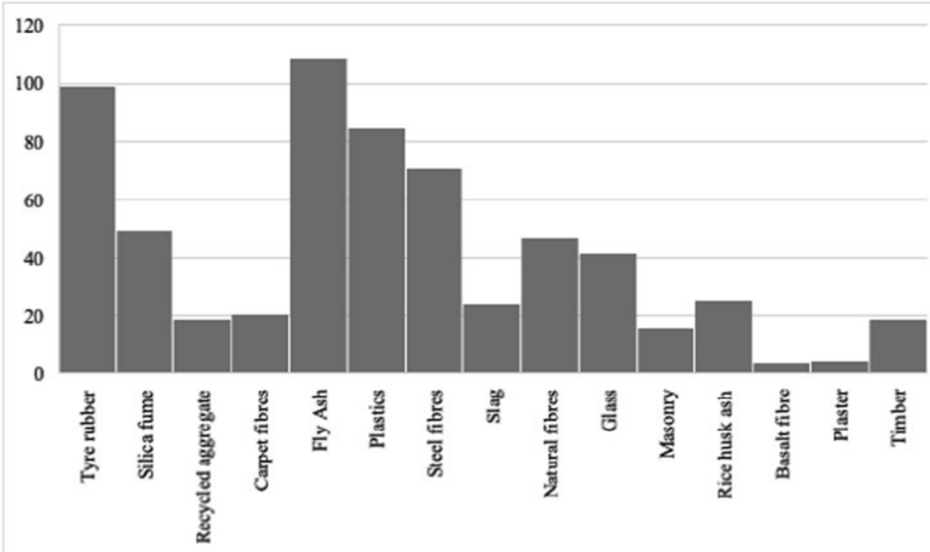

Fig. 4. Thematic map of word occurrences.

concrete materials. Other researched SCMs were Silica Fume and Ground blast furnace slag. However, the former waste materials were not as prominent.

- The primary publication source of waste fibres in building construction and waste fibres within cement and concrete materials is the Journal of Construction and Building materials. This is due to the Journals scope of innovation, focusing on investigative measures such as experimental material designs for building construction materials.
- Common non-inert waste materials such as textiles, cardboard, fibreglass and nylon were not listed within the word occurrences sourced from the 874 research publications.
- The results from the bibliometric assessment did not contain the term "kraft fibres". This shows that significant research has not been conducted on the constituent fibres within cardboard waste material.
- The findings showed that significant research of plastics and tyre rubber have been associated with building construction materials. The waste materials were highlighted in literature as emerging trends of road and pavement sub layers.

The findings from the bibliometric assessment have allowed future researchers to identify influential journal sources and target waste research focus areas. Based on the findings, it was shown that the predominant materials researched were fly ash, tyre rubber and various plastics. Due to the array of listed waste materials, this review will therefore promote the viability of cardboard waste used further in building and construction materials. The bibliometric assessment has identified a gap in literature regarding the use of KFs derived from cardboard waste and therefore a comprehensive analysis is required to inform readers of current experimental trends and opportunities for future research.

4. Kraft fibres

In recent years, growing interest has been directed towards natural fibre composites (NFC) [29]. This has been primarily due to the improved sustainability and biodegradability aspects when replacing synthetic materials. However, the advancement of integrating NFs has been largely disrupted because of the reduced mechanical and durability properties when applied within a cement matrix [19]. NFs can include mineral and various plant species. Each fibre type contains specific material properties and can be used accordingly to the product requirements. Mineral fibres such as asbestos were used extensively as a reinforcement agent due to its ability to deflect the deterioration caused by a high alkaline environment. However, the material has significant health implications on human life [30]. Therefore, the focus on plant fibres has been of interest due to its non-hazardous nature and other significant advantages, as detailed further. A common extraction method to obtain NFs from plants is called kraft processing. This is completed with chemical additives or a thermomechanical process [30,31]. This method is predominant in the paper and cardboard industry and is prevalent across the globe. After the completion of this processing system, the fibres are called kraft fibres (KF) or kraft pulp (KP). These fibres primarily contain cellulose matter and can be the main reinforcement element if used in building and construction material applications.

The integration of KF within cement-based composites has been challenging due to the severe fibre degradation caused by the high alkalinity of the environment [31,32]. A successfully functioning composite material is only truly effective when the materials within the matrix are cohesive and thoroughly bonded. As the stress load transfers within the matrix, it must successfully alternate until it reaches the reinforcement agents within the composite [33]. An example is shown with steel reinforcement within concrete materials. A similar result is required of the fibres micro-mechanics within a composite application. Therefore, significant research has been undertaken to modify the matrix of the composite [34] to allow for the fibres to maintain their strength characteristics. Other research focus areas have involved fibre pre-treatment to mitigate the degradation caused [35]. However, currently composites with KFs are still negatively affected mechanically and a thorough review has not been explicitly examined. In order to attract a significant industry valorisation, the integration of KFs within cement and concrete materials pose a suitable opportunity. This is primarily because of the enormous volume of KFs produced worldwide annually [36]. The present work shows the progress and engineering issues when KFs are used in cement-based composites.

The advantages and disadvantages of KFs are like that of NFs and are highlighted further in Table 2. However, components of NF are removed during the kraft process, this is further explained below. KFs can withhold high tensile strength, improve flexural toughness of composites, enhance crack resistance and reduce fatigue behaviour [37]. However, the ability for the fibres to perform at a high standard is dependent on the environment at which it is applied. Within a cement-based matrix, KFs can deteriorate due to the high alkaline-mineral properties of Original Portland Cement (OPC) [38]. Therefore, the deterioration of KFs must be mitigated to ensure the mechanical and durability properties of the composite are maintained. This is especially critical for the promotion of these fibres to be integrated within cement and concrete composites for a widespread commercial application. It is important to note that a key difference between KFs and NFs is the ability of NFs to withhold moisture in the fibre cell walls. Jongvisuttisun and Kurtis [39] have found that this factor can aid in the mitigation of autogenous shrinkage within the cement paste during the hydration phases. However, as illustrated in Fig. 5, excessive moisture within the fibre cell walls of NFs can lead to premature crack inceptions of the composite microstructure upon evaporation.

KFs have been applied to various composite designs to enhance the reinforcement properties as well as a partial cement replacement. However, as discussed, the successful application of KFs is dependent on several composite dimensions. The main factors that affect the successful integration of KFs within cement-based composites are the material selection within the matrix, fibre percentage and dispersion. When those factors are successfully determined, there should be minimal fibre degradation with a maximum interfacial strength between the fibre and the matrix [19,31]. These factors are all dependent on the final material selected and must be thoroughly examined prior to a mix design phase. The environmental benefits of using NFs have been at the forefront of research focus. This is shown with efforts of minimised cement consumption and reduced energy requirements of other synthetic fibre materials. However, researchers have shown a variety of positive effects when KFs are integrated within composite designs. The benefits are an enhancement of mechanical properties including a reduced crack width when under implied loads. Minimised composite thickness and a reduction of weight in tall structures. There is also a resistance to plastic shrinkage during the hydration phase of cement and concrete materials [41].

4.1. Kraft fibre processing system and natural fibres

The Kraft pulping process is the largest portion of global pulp production [42]. This process (kraft pulping) is the conversion of wood chips or plant materials into a pulp like fibre mass. The main objective is to remove enough lignin to separate the cellulose fibres. There are three main components of plant matter; lignin, cellulose and hemicellulose [43]. The amorphous component of NFs are the lignin and hemicellulose. These components enhance the strength of the fibres by maintaining unity and agglomeration of fibres [44]. Lignin protects two main polysaccharides used within the plant cell walls that are hemicellulose and cellulose. Lignin binds the proteins to reduce enzymatic depolymerisation including cellulolytic and hemicellulolytic enzymes [45,46]. Therefore, when the lignin content is reduced the cellulose fibres no longer agglomerate and the dispersion of fibres multiply. Cellulose is the main component of strength and stiffness within NFs. Cellulose is composed of glucose and formed via rigid insoluble microfibril chains that are integrated by many intra and intermolecular hydrogen bonds [20,38,47]. Hemicellulose surrounds the cellulose chains to strengthen the bond of cellulose components as well as interacting with lignin. The components of natural fibres are shown in Fig. 6.

Table 2
Advantages and disadvantages of Kraft fibres.

Advantages	Disadvantages
Low density and high strength	Lesser durability compared to synthetic materials
Minimal reprocessing	High moisture absorption
Alternative waste sources of KF	Low modulus of elasticity
Non-abrasive material	Higher variability of fibres
Non-hazardous nature	Lower processing temperatures
Biodegradability	Dimensional instability

The kraft process removes the lignin content via the application of hot chemicals within a pressurised vessel or digester. A mixture of alkaline chemicals that has a pH level above 12 are most effective when cooking at a temperature of 170 °C [49,50]. Researchers have reported the use of "white liquor" chemicals such as sodium hydroxide (NaOH) and sodium sulphide (Na_2S) used within the processing system [42,49,51,52]. The wood chips or plant materials are ultimately cooking within the chamber while the chemicals are attacking and dissolving the phenolic material (lignin). The dissolvement of lignin is caused by the hydroxide and hydrosulphide anions which cause the components to fragment into water/alkali soluble or hydrophilic fragments. When the material has reached the appropriate temperature and time parameters, the pulp is then washed, screened, cleaned and dried [50]. Fig. 7 illustrates the fibres transformation during the kraft process.

To measure the residual lignin content within kraft pulp, the term used within the chemical composition is expressed by "kappa number". A lower kappa number means less lignin and is measured within a range of 1–100, this is applicable to all varieties of chemical and semi-chemical pulps [51]. The reduction of lignin and hemicellulose during the kraft process results with a rise in concentration of cellulose pulp [53]. Ma et al. [53] noted that the rise in lignin concentration resulted with a reduction in the fibres mechanical properties. Therefore, the lower content of lignin can enhance the fibres mechanical properties.

Research on the integration of NFs within cement-based materials show that degradation caused on the fibres significantly reduces the lignin and hemicellulose content. There are four main collective processes of alkaline deterioration, these include; degradation of lignin, deterioration of hemicellulose, degradation of hydrogen bonding and alkaline hydrolysis of cellulose microfibrils [38]. The alkaline attack includes the hydroxide anions in cement pore solution to react with the lignin, which causes the lignin to disintegrate. This reaction then results with the release of phenolic hydroxyl groups, reducing the strength of the agglomeration of hydrogen bonds. During the alkaline attack the hemicellulose components convert into fermentable sugar, increasing the hydrophilicity and therefore enhancing the hydrolysis of lignin and hemicellulose into soluble fragments. The fragments are then separated from the fibres thus in turn reducing the strength and durability of the fibres [38]. The removal of these amorphous properties increases the crystallisation and mineralisation on the fibres, these two factors lead to the reduction of fibre strength, fibre embrittlement and lower strain capacity of the fibre. Bonnet-Masimbert et al. [44] and Kochova et al. [54] discuss the importance of removing hemicellulose, lignin and other surface impurities such as fatty acids, pectin, wax and tyloses. The removal of these properties can increase the surface roughness of the fibre which leads to a stronger adhesion and bonding within the matrix. It is important to note that the embrittlement of the fibre is highly dependent on the amount of calcium hydroxide ($Ca(OH_2)$) that is within the matrix.

Although the alkaline attack on lignin and hemicellulose components of the fibre can reduce a level of protection against the cement matrix, it can also significantly decrease the water retained in the fibre cell walls. KF already has a significant level of lignin and hemicellulose removed during the kraft process [30,54]. The

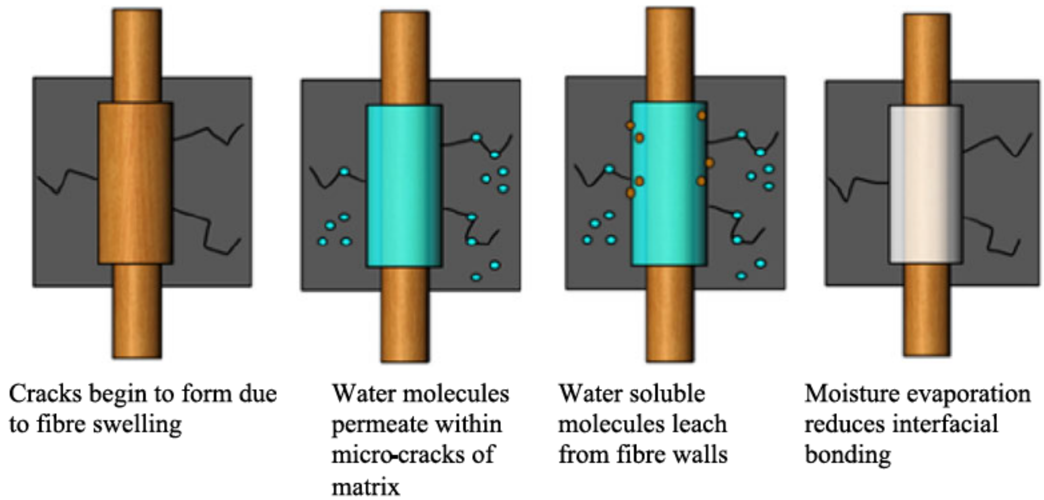

Fig. 5. Consequence of NFs containing moisture within composite matrix. Redrawn from [40].

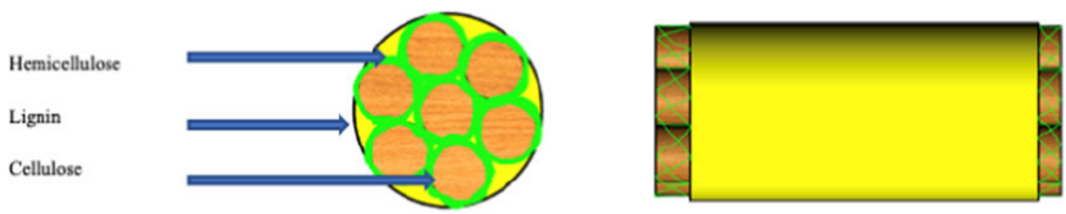

Fig. 6. Components of NFs. Redrawn from [48].

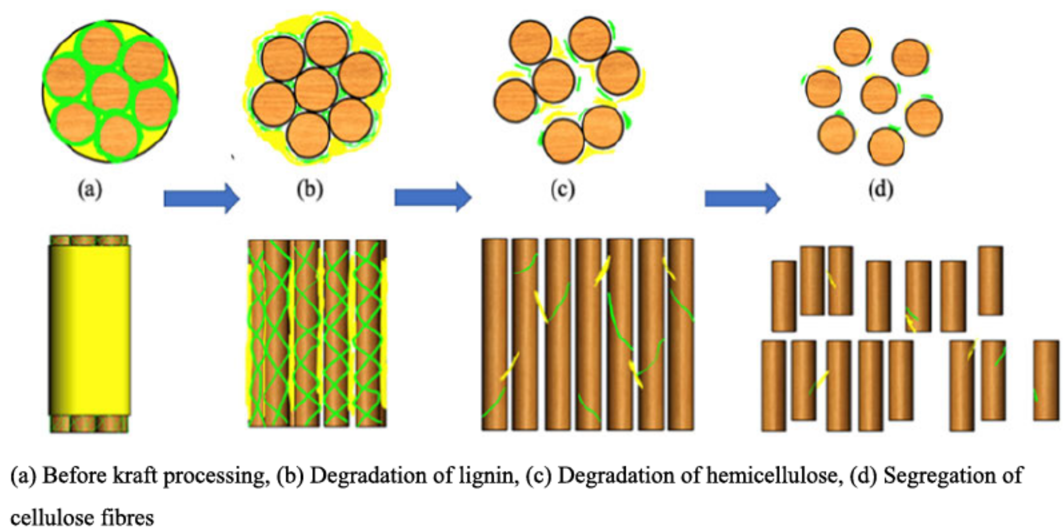

Fig. 7. Kraft transformation process of NFs. Redrawn from [48].

removal of these properties and other impurities will allow a stronger bond within the matrix, as well as reducing the water retained in the fibre [55]. This is further discussed with Bonnet-Masimbert et al. [44] that use NaOH to pre-treat oil palm fibres, in order to remove the unwanted lignin and hemicellulose properties. The removal of these components allow the cellulose microfibrils to align themselves parallel with the applied load [44]. With this effect, it can enhance the load transfer and increase mechanical properties. This ultimately reduces the spiral angle and increases resistance of the fibre. Table 3 summarises the key differences between KFs and NFs.

4.2. Experimental KF research applications

The advancement towards an environmentally friendly option to fibre composite materials has shown prominent researchers focusing on KFs. Table 4 demonstrates the material matrix, type of KF, pulp ratio and research application. This Table focuses on experimental research that has been conducted due to the desired final application of the material being integrated within building and construction materials. The findings and observations show that the incorporation of pozzolanic materials or admixtures has been a common inclusion within the composite matrix. Rarely

Table 3
Key differences of KF and NF.

Factor	KF	NF
Fibre dimensions	Thinner fibres Shorter fibres	Thicker fibres Longer fibres
Fibre coatings	Removal of hemicellulose Removal of lignin	Amorphous protection Fibre impurities (fatty acids, pectin, wax)
Fibre surface	Increased surface roughnessDecreased water retention in fibre walls	Smoother surface area Retention of water in fibre walls
Fibre propensity	Dispersion	Agglomeration

Table 4
Overview of the research on applying kraft fibre pulp in cement composites.

Matrix	Fibril types	Properties		Reference/s
		Fibre Percentage	Application	
OPC- Admixtures	Eucalyptus Kraft pulp	5	Reinforced cement composites	[35,58]
OPC	Sisal Kraft pulp	4.7	Cement roof tiles	[59]
OPC	Eucalyptus Kraft pulp	10	Reinforced cement composites	[60]
OPC	Pine Kraft pulp	8	Reinforced cement composites	[61]
OPC	Pine Kraft pulp w/ PP	1–3	Mortar	[62]
OPC- pozzolans	Pine Kraft fibres	4	Reinforced cement composites	[34,63]
OPC- pozzolans	Pine Kraft pulp	0.039–0.23	Fibre sheet cement composites	[64]
OPC	Pine Kraft pulp	4	Fibre cement beams	[37,65]
OPC- admixtures	Recycled waste Kraft pulp	1–14	Reinforced cement composites	[51]
OPC- admixtures	Recycled waste Kraft pulp w/ glass fibres	4	Reinforced cement composites	[31]
OPC- admixtures	Recycled waste Kraft pulp w/ PP and acrylic fibres	8	Reinforced cement composites	[57]
OPC	Pine Kraft pulp	4	Reinforced cement composites	[66]
OPC- pozzolans	Pine Kraft pulp	2	Concrete	[67]
OPC	Pine Kraft pulp	0.5–2	Concrete and mortar	[41,68]
OPC	Eucalyptus/ Sisal/ Banana Kraft pulp	4–12	Reinforced cement composites	[56]
OPC- pozzolans	Eucalyptus/ Sisal/ Banana Kraft pulp	4–12	Reinforced cement composites	[32,55,69]
OPC- pozzolans	Sisal/ Pine Kraft pulp	4–12	Reinforced cement composites	[70]
OPC	Eucalyptus Kraft pulp	3	Reinforced cement composites	[71]
OPC- pozzolans	Eucalyptus/ Sisal Kraft pulp	1–5	Cement roof tiles	[72]
OPC- pozzolans	Pine Kraft pulp	8	Reinforced cement composites	[73]
OPC- pozzolans	Eucalyptus Kraft pulp	5–15	Reinforced cement composites	[74]
OPC- admixtures	Pine Kraft pulp	8	Reinforced cement composites	[75]
OPC	Pine Kraft pulp	4	Reinforced cement composites	[76]
OPC	Eucalyptus Kraft pulp	6	Reinforced cement composites	[77]
OPC	Recycled waste Kraft pulp	25–33	Concrete blocks	[28]

has the ratio of fibres exceeded 10% and the common application has been reinforced fibre cement composites. The composites have been intended to be used in fibre cement boards and replace other materials such as asbestos and synthetic fibres. This is further shown in Table 5, where the common experimental method is to measure the flexural strength and ageing process of weathered materials. It is important to note that the predominant research has focused on virgin KFs. With a selected number of researchers focusing on recycled KFs derived from waste materials such as cardboard and paper materials [28,31,51,56,57]. Despite the recycling process undertaken to retrieve the KF content, the fibres can remain an effective reinforcement agent when integrated within cement-based composite materials.

5. The mechanical performance of Kraft fibre

The following section describes the factors affecting the successful integration of KFs for further use in cement-based composites. As shown in Table 4, researchers have conducted a variety of mix design experiments with the use of KFs as a reinforcement agent and partial cement replacement within cement-based composites. Due to the inter-connected physical and chemical reactivity of OPC and KFs, the mechanical outcome of those experiments has remained hindered. Table 6 highlights the current strengths, weaknesses, opportunities, and threats (SWOT) associated with the use of KFs within a cement-based matrix.

Cement composites containing NFs typically fail due to the embrittlement of the fibre and will either snap or the fibre will pull out of the matrix. As the load on the material increases, the composite will reach a maximum bearing capacity. The stress is released immediately to other areas of the composite via the formation of micro cracks, as those micro cracks develop larger, other areas will also begin to form additional micro cracks [51]. This process continues until the merging of the micro cracks happens and creates a visible crack in the material [31]. The cracks will form at mid-span where maximum tensile stresses are applied, reducing the ductility of the composite. The stress loading is called 'Bend over point' and as this increase, so does the elongation. The fibres will then be taking the load via a process called crack bridging. The fibres attempt to deflect the failure of that area and transfer the load away from the failing point. However, during the application of maximum stress, fibre pull out is simultaneously occurring. This highlights that the preliminary factor of failure is with the interfacial bond and not the individual properties of the fibre material.

Therefore, it is imperative to enhance the bonding of the material as well as protect the outer layer of the KF from the aggressive nature of an OPC matrix. Fibres that have been immersed in hydrated cement or are within a high alkalinity environment will usually fail due to fibre snapping. However, when the fibres pull out of the matrix, this is due to a less adhesion strength rather than the tensile strength properties of the fibre. The increase of calcium silicate hydrate (C-S-H) can further attach to the surface of the

Table 5
Testing conducted on kraft fibres.

Country of study	Mechanical	Physical	Durability										Reference/s
	Flexural	Compressive	Tensile	Water absorption	Permeable voids/ Porosity	Shrinkage	Thermal	SEM	Wet & dry cycles	Freeze & thaw cycling	Carbonation	Chloride ion	
Brazil	✓			✓	✓			✓	✓				[58]
Brazil	✓			✓	✓			✓	✓		✓		[59]
Brazil	✓			✓									[35]
Brazil	✓							✓	✓				[60]
United States	✓			✓			✓		✓	✓	✓		[61]
United States								✓					[62]
United States	✓						✓	✓	✓				[34]
United States	✓							✓					[64]
United States	✓							✓	✓				[37]
United States	✓								✓				[65]
United States								✓	✓				[63]
Iran	✓							✓					[51]
United Kingdom	✓			✓	✓			✓					[31]
United Kingdom	✓												[57]
Canada	✓	✓						✓	✓	✓			[66]
Canada		✓		✓	✓			✓				✓	[67]
Canada		✓		✓	✓	✓		✓				✓	[68]
Canada		✓				✓		✓					[41]
Brazil	✓			✓	✓			✓					[56]
Brazil	✓			✓				✓					[55]
Brazil	✓							✓					[32]
Brazil	✓			✓	✓								[70]
Brazil	✓			✓				✓					[69]
Brazil	✓							✓					[71]
Brazil	✓			✓	✓		✓		✓				[72]
Brazil	✓			✓	✓		✓	✓	✓		✓		[73]
Brazil	✓			✓	✓			✓					[74]
United States	✓			✓		✓			✓				[75]
Spain	✓	✓						✓	✓				[76]
Brazil	✓			✓	✓		✓	✓	✓		✓		[77]
Pakistan		✓	✓	✓									[28]

fibres and therefore create a stronger bond of the interfacial fibre zone. Silica fume (SF) has been shown to enhance the C-S-H levels within cement materials [45]. Therefore, a prevalent research area has been with the integration of pozzolanic materials [78]. It has been shown that lowering the density of cement-based composites with pozzolanic materials such as Metakaolin (MK), FA and SF can improve the mechanical properties of fibre composites. This is completed when the short fibres are under implied loads and the crack bridging system is enabled. A denser matrix reduces permeable voids and the load transferability to the fibre reacts at a faster rate [31,51,67,78].

Table 6
SWOT analysis of KF composites.

	Physical	Mechanical
Strengths	Adequate Bonding capacity	Fibres bridge the matrix cracks (reducing larger cracks to smaller)
	Reduce free plastic shrinkage	Adequate stiffness and strength
	Enhanced toughness and impact resistance	Decrease thermal conductivity
	Decrease cement consumption	Increase sound absorption
Weaknesses	Fibre fracture	Reduced long term durability
	High water absorption	Fibre pull out
	Lower fire resistance	Complex matrix requirements
	Fibre mineralisation	
Opportunities	Utilising waste cardboard as a source of fibre reinforcement and cement reduction	The formation of an effective crack bridging system with the use of recycled kraft fibres
	Reducing the stockpile of cardboard waste in landfill	Reducing the weight of concrete composites in certain applications of high-rise buildings
Threats	Economically not a viable solution	Severe environmental conditions can reduce the application commercially
	There are insignificant reductions of CO_2 emissions	The integration of waste fibres with concrete materials is complex and troublesome

5.1. Methods to improve composite design

There are two critical elements to maintain the integrity of KFs within cement-based composites. Firstly, to reduce or remove the alkaline compounds. This reduces the degradation that occurs on the cell walls of the fibre, ultimately rendering it weak and ineffective [58]. Secondly, to enhance the stability of the fibres within the cementitious matrix via chemical or physical modification [19]. Incorporating these two modification factors within the mix design of KF cement-based composites will ensure the design life of the material application is maintained to its physical and mechanical requirements. However, the integration of these factors is also limited by several external and internal influences. Examples of external influences include but not limited to weathering conditions, including physical and chemical weathering, location of application, applied loads and forces. These influences can cause the material to deteriorate, accelerating the ageing conditions and significantly weaken the material. Therefore, the primary focus during the mix design is to optimise the internal influences, to inhibit the negative effects caused by external influences.

The internal influences include but not limited to are fibre mineralisation, matrix voids, clumping of fibres, fibre percentage and degradation of fibres. A key component when integrating fibres within a composite material is to ensure the fibre dispersion is distributed consistently. The challenging factor is that KFs have a natural tendency to agglomerate and bond together. The different methods of bonding include interdiffusion, mechanical interlocking, capillary forces, coulomb forces, hydrogen bonding and Van der Waals forces. At a single fibre level, the Van der Waals force results with a self-attraction level that is difficult to segregate from other fibres. This agglomeration consequently concludes with the formation of fibre clusters within the matrix [79]. The clusters cause voids, pockets and ultimately an insufficient fibre dispersion. The presence of voids and pockets, creates weak points when under implied loads. The same issue can promote permeability within the matrix, allowing chemical and moisture transference to occur. Reducing the number of voids is critical to decrease the movement of chloride ions in concrete composites, as chloride ions can enhance carbonation as well as the alkali-silica reaction, ultimately reducing the service life of the material [31,32]. Different materials and chemicals can reduce the clumping of fibres such as SF. Research conducted by Sanchez and Ince [79] show the use of SF separated the fibres due to the small particle size acting as a wedge between carbon nanofibers (CNF). For the composite to remain effective as a reinforcement agent, the fibres must interlock to a certain degree. However, ensuring the dispersion rate is high allows the fibres within the composite to remain effective as a reinforcement agent and take effect during the formation of the crack bridging system [36].

5.2. Fibre modification

Degradation of fibres are due to alkaline hydrolysis and mineralisation of fibre cell walls within OPC matrices [80]. The main cause of high alkalinity is the production of $Ca(OH)_2$ within the OPC matrix. Therefore, reducing the amount of $Ca(OH)_2$ will enhance the longevity of the fibres [19,32,81]. With a high volume of $Ca(OH)_2$, the hydrated cement products can attach themselves to the walls of the fibre and mineralisation can begin to form. This process weakens the fibre and results with fibre embrittlement [62]. Therefore, fibre modification is required to enhance the service life of KFs within the cement matrix. A fibre modification via pre-treatment will reduce the degradation caused that renders the fibres weak and brittle. Due to the removal of NFs amorphous components during the kraft process, KFs remain exposed to these two detrimental factors. Therefore, pre-treatment is an important pre-requisite when integrating KFs within a cement matrix. If KFs are not pre-treated, the cellulose micro-fibrils will be stripped apart and the lumen of the fibre will be saturated with cement hydration products [80]. This results with the embrittlement and failure of the fibre to act as a reinforcement agent. There are several methods of pre-treatment including silane coatings, hornification, autoclave, thermal treatment and chemical treatment [38,58,60,65,75–77,82].

Methods of pre-treatment can include diluting a chemical admixture with water before the application of fibres. This ensures the aqueous solution does not clump or stick to specific areas of the fibre material. Often, the fibres are oven dried before application within the matrix to ensure no condensation remains on the fibre and moisture is completely removed before application [82]. If water is to remain on the fibres it can create an over-supply of moisture within the matrix and can further result with voids, creating an excess in permeability. Dittenber and GangaRao [20] show an improved bond within the matrix can be achieved with prior drying of fibres before application. The hydroxyl groups on the fibre will bond with hydroxyl groups within the matrix, creating hydrogen bonds. This results with water molecules bonding with the hydroxyl groups on the fibre surface, as the water evaporates it can create voids within the matrix, resulting with an ineffective fibre bond. This factor creates a weaker matrix and reduces the bonding between the interfacial zone of the fibre and the matrix of the composite.

Fibre modification has been shown to enhance the interfacial bonding between the fibres and composite matrix [68]. This results in the ability to transfer loads within the composite microstructure when axial loading is applied. Recent studies of concrete composites containing KFs resulted in a reduction in mechanical compressive strength. However, the fibres included unmodified fibres (UF), mechanically modified fibres (MMF) and chemically treated fibres (CTF). When fibres have been modified there is an increased fibre density, resulting with composite compressive strength similar to samples without fibre reinforcement. When fibres weren't modified there was a larger reduction in compressive strength at 28 and 90 days, equalling 28.9% and 17.4% respectively. The increased density of composite materials has shown improved mechanical properties [68]. Previous studies also indicated a reduction in compressive strength due to the integration of KFs [41,66–68]. Research studies with MMFs and CTFs in mortar cubes show a less compressive strength reduction than UFs. Although there is a reduction in compressive strength, samples that contained 4% by weight of cement with UFs showed the largest reduction in strength of 25% when compared to the control sample containing zero fibre integration [66]. This suggests that the UFs do not contain a sufficient fibre matrix bond when compared to modified fibres. When fibre bonding is enhanced, there is an improved load transfer within the microstructure of the matrix. However, other research involving the hornification process of KFs show a 7% increase of compressive strength and 8% increase in flexural properties [76]. KFs have a lower density when compared to other synthetic materials and are expected to induce more voids, rendering a lighter composite [31]. When fibre modification is applied, the voids are reduced, increasing the compressive and flexural strength values.

Other methods of thermal fibre modification were performed by Mohr et al. [63,65]. Their research integrated kraft thermomechanical pulp (TMP) within cement-based composite designs. The results demonstrated TMP exhibits increased first crack strength when subjected to flexural testing due to the micro crack bridging system. However, in the post cracking region of the composite, the strength and toughness decreased. This can be further attributed to the cellulose content of TMP (40–45%) when compared to cellulose content of other KFs (65–80%). When there is less cellulose content, the tensile strength of the composite decreases, and fibre pull out occurs at a higher rate. Tensile strength of TMP is approximately 50–70% of KFs [65]. TMP exhibits a slower degradation when subjected to wet and dry cycles when compared to unbleached and bleached KFs. This is further shown with increased flexural strength of 77.1% and 85.7% and increased toughness properties between 138.6% and 221.7%, respectively, after 25 wet and dry cycles. This research highlights the significance fibre modification can have on composite designs.

When fibres are not modified there is an increased composite porosity, lower mechanical values and a reduction of durability [31]. However, despite these factors, the integration of KFs within a cement-based composite will enhance the flexural properties of the material. Savastano et al. [56] has shown the integration of 12% fibre content by mass, results with flexural strength values between 20 MPa and 25 MPa. These values correlate further with fracture toughness results between 1.0 and 1.5 kJ m^{-2}. Composites containing zero fibre content withhold approximately 12 MPa and 0.5 kJ m^{-2}, respectively. Although, the values are significantly enhanced, the long-term durability of the composite is hindered by the ability of the composite to withhold moisture and deflect the high alkalinity. Composites containing similar values of fibre content (12%) have a water absorption ability of more than double when compared to zero fibre composites. As previously discussed, this factor can lead to severe degradation when water evaporates. Therefore, modified fibres withhold a significant influence on the microstructure of composite materials. This is shown with an enhanced fibre matrix interfacial zone and reduction in mineralisation of the fibre [58].

5.3. Matrix modification

As discussed, OPC contains a high amount of $Ca(OH)_2$ and this factor attributes to the degradation of KFs [34]. It has been shown that reducing the amount of $Ca(OH)_2$ can mitigate the degradation and enhance the materials composition [48,80]. An effective and common approach is to integrate the use of SCMs [19,34]. When modification of the cement matrix is required, the integration of SCMs can lead to enhanced mechanical properties, lower costs and positive environmental impacts [38]. The environmental benefits include a reduction in CO_2 and greenhouse gas emissions, which has lead to the United Arab Emirates mandating the use of SCMs within concrete materials [83]. The integration of SCMs can enhance the service life of final application and reduce the damage of corrosion that typically happens within traditional concrete. Moreover, SCMs are a sustainable answer to reduce the adverse effects caused by the clinker factor of cement. The integration of industrialised by-products such as SF, GBFS and FA can aid in the accumulation of that waste further within cement-based materials [38].

Significant research has been studied on the integration of various SCMs because of their pozzolanic properties [34]. SCMs contain two notable integration characteristics: First, a positive influence on the hydration kinetics of OPC and secondly, a considerable consumption of $Ca(OH)_2$. The natural pozzolanic property within SCMs are an attractive supplement due to the conversion properties of $Ca(OH)_2$, into a desirable product called C-S-H [84]. The chemical properties can counterattack the aggressiveness of the high alkalinity within cement paste. A finely ground pozzolan material primarily consists of siliceous or aluminous, that when in the presence of moisture chemically reacts with $Ca(OH)_2$ to form cementitious properties. The pH level of cement is approximately 12–13, which is high alkaline [38]. Therefore, integrating an appropriate SCM will reduce the pH level and create an enhanced environment for KFs to remain effective.

The most commonly used SCMs include; MK, SF, GBFS and FA [83]. Several researchers have used these materials within research of composite designs due to the improved mechanical properties [34,74,80]. For example, Machado et al. [74] has shown the presence of SF can create a greater amount of hydrated calcium silicate, which aids in the strength of the composite as well as increases the service life when exposed to weathering. However, the researchers noted that when excessive amounts of SF (17.1%) and high values of cellulose fibre (13.5%) are integrated, the porosity of the composite also increases. This can be attributed to the hydrophilic nature of the fibre and the heterogeneous dispersion of SF within the design mix. SF can act as a filler and reduce the porosity of a composite, minimising voids that contain air and moisture [73]. However, the measurement of the materials integration is critical, as excessive amounts can enhance water absorption and reduce durability [74]. Moreover, Machado et al. showed when 10% SF and 10% fibre was integrated, the modulus of rupture (MOR) and modulus of elasticity (MOE) had maximum values at both 28 and 180 days. The results were 12.16–12.48 and 18.15–17.64 MPa respectively. When SF increased, the MOR and MOE significantly decreased at all ages. This further highlights the importance of controlled levels of SCM incorporation. Other results of SF and limestone powder integration have increased MOR to 8.5 MPa. This is a flexural increase of 260% [51]. It is important to note that the flexural strength is marginally improved when SF is integrated, however the critical dimension that SF improves is compressive strength.

The ability for the fibre to maintain its strength is dependent on the applied matrix. For example, GBFS can alter the composites matrix by altering the chemical composition. This is shown with a reduction of Ca(OH)2 as compared to OPC. This factor should increase the fibres ability to withhold its mechanical strength. However, research incorporating the integration of GBFS has shown reductions in the fibre composites flexural capacity [32,69]. This is discussed with Savastano et al. [70], that shows a high porosity within the microstructure of the composite. When GBFS is used within the material, there are values of at least 30% water absorption by mass of the composite. Due to the high porosity of the microstructure, effective crack bridging systems can remain inactive leading to severe reductions in mechanical properties [69]. OPC composite mix designs with KF integration can have sufficient mechanical values, however, the service life and durability of the composite will be severely reduced.

6. Durability enhancement

6.1. Fibre modification

Without fibre modification, hydrated cement products attach themselves to the walls of the fibre and mineralisation begins to form. This process weakens the fibre and causes embrittlement to the fibre walls [34]. Soroushian et al. [61] highlighted petrification can advance due to accelerated ageing cycles. Their research showed that when hydrated cement products fill the fibre cell wall cavities, there is an increase of fibre rupture at fracture surfaces. However, the repetition of wetting–drying cycles increased the flexural stiffness but reduced the flexural toughness without significantly affecting the flexural strength of the composite. Although the results were mixed for freeze–thaw cycles, the outcome on flexural strength was similar. It can be suggested with the integration of SF, there was an increased C-S-H content, causing enhanced durability with the flexural properties of the aged composites. Their research signifies the importance of a combined fibre and matrix modification. Other research conducted by Dittenber and GangaRao [20] show that an improved bond within the matrix can be due to prior drying of fibres before application. Hydroxyl groups on the fibres will bond with hydroxyl groups within the matrix, creating hydrogen bonds. This results with the water molecules bonding with the hydroxyl groups on the fibre surfaces, as the water evaporates it creates voids within the matrix and the fibres bonding decreases. Therefore, drying of the fibres will result with improved bonding due to the elimination of the hydroxyl groups.

As discussed, fibre modification has been shown to increase the durability of KFs when applied within a cement-based composite. Booya et al. [68] show the effectiveness on the durability properties of MMFs and CTFs in contrast to UFs. Due to the long service life of concrete structures, the chloride ion penetration test can assist in evaluating the durability of the material. The less charge passed through the material indicates a better quality of concrete and increased service life. The research findings showed a higher charge of coulombs passed through the composite material when it contained UFs, this results with a lower material durability. The tests subsequently indicated that MMFs and CTFs had a lower charge passed through when compared with UFs. Moreover, the increased chloride ion permeability of UF concrete composites is due to a higher value of interfacial porosity between the fibre and interfacial zone of the matrix [68]. These results were in accordance with other durability testing such as sorptivity index and water absorption. The researchers demonstrated that modified fibres were superior than UFs, when applied in KF concrete composites [67]. Other research of TMP have shown an increased durability within composite designs [65]. This is shown with results derived from wet and dry cycles, indicating that the flexural strength and toughness are superior to UFs. Moreover, when fibres are subjected to modification, the dimensional stability is enhanced but also the mitigation of cement hydration products attaching to the fibre walls. These two factors allow the fibre to maintain its strength [63].

A common durability testing method of composite materials is with the inclusion of wet and dry cycles [61,66,75–77]. This method accelerates the environmental effects that can be imposed on composite materials. When KF composites are subjected to wet and dry cycles, the flexural strength and toughness of the composite are substantially reduced [66]. This is due to the reduced fibre strain capacity. However, modified fibres have shown enhanced mechanical and durability properties. This suggests that the cell wall mineralisation of the fibre was minimised, ultimately increasing the strain capacity of the fibre [38,66]. Booya et al. [66] showed similar flexural strength properties of CTFs after 30 wet and dry cycles (approximately 5.7 MPa). The researchers demonstrated the increased strain capacity when surface modification has been applied to KFs. Overall, from zero to forty cycles, the reduction in flexural strength was shown as 49%, 43% and 33% when composites contained UFs, MMFs, and CTFs respectively. These results also correlate with a reduction in the flexural toughness, 76.6%, 50.1% and 42% for composites containing UFs, MMFs and CTFs respectively. Moreover, when fibres have been modified, there is a greater volume stability of the fibre due to the refinement and pretreatment process [66,77]. The modification of fibres shows a reduction of migrated hydration products within the fibre lumen, predominantly calcium hydroxide $Ca(OH)_2$ [63,65]. Ballesteros et al. [77] have shown the dimensional stability of fibres with hornification treatment. Hornification of KFs show an enhanced fibre matrix interface with a greater specific energy of the composite design [76]. Reducing the percentage of water retained within the fibre, resulted with an increased fibre anchorage system within the matrix. When fibre modification has not been applied, composites containing KFs can significantly absorb water, reducing the durability. This has been shown with Savastano et al. [56] that conducted research using 12% fibre content. Their results demonstrated water absorption had more than doubled. Moreover, due to fibre modification increasing the interfacial zone of the fibre within the matrix, the durability and flexural properties increased [76]. This also in accordance with other researchers that show increased mechanical benefits when fibre modification is applied [41,66–68,75,76].

Freeze and thaw cycles have been shown to alter the fibre bond when compared to wet and dry cycles [61,66]. When axial pressure is applied on the composite, fibre rupture is caused rather than fibre pull out [66]. This is because of an excessive fibre matrix bond within the microstructure of the composite. Flexural strength decreases from zero to forty cycles with reduction results of 20.5%, 23.9% and 29.6% for composites containing UFs, MMFs and CTFs respectively. However, when the results are compared to forty cycles and unaged specimens the reduction shown is 19.5%, 1% and 1% for composites containing UFs, MMFs and CTFs. MMFs contained the better flexural performance when compared to UFs and CTFs. However, the decrease in flexural strength remained the same after ten cycles for CTFs. The freeze and thaw cycles show an increased resistance of flexural strength and toughness when compared to the wet and dry cycling. Researchers have suggested the durability is increased due to the initial resistance of micro-cracks by internal frost pressure [61,66]. Therefore, an increased resistance to freeze and thaw cycles have been shown with modified fibres when compared to UFs.

6.2. Matrix modification

Modifying the matrix of composite designs can aid in the service life of the material [34]. Reducing the level of alkalinity within the matrix can ultimately enhance the durability of the fibre in the composite material. Machado et al. [74] have integrated SF and natural rubber latex to increase the durability properties of KF composites. The results showed that the materials increased fibre strength and prevention of hydration products on the walls of the fibre after 180 days. Moreover, the added effect of carbonation decreased the porosity and permeability. This increased the durability by reducing the water absorption of the composite. These results are in accordance with Urrea-Ceferino et al. [73] who showed the effects of accelerated carbonation on samples with silica as the mineral supplement. After 200 dry and soak cycles, the KF composites exhibited similar MOR measurements than compared with nonaged specimens. This was shown with results of 6.49 MPa–10.9 MPa, and 6.17 MPa–9.39 MPa, respectively. The researchers concluded that the mechanical properties were maintained due to the use of silica as an inert filler. It is important to note that the integrity of mechanical values coincides with the durability characteristics of the composite. This conclusion is in accordance with Mohr et al. [34] that conducted durability studies with SCM composite integration. Their research concluded that SF replacements of 30% or more can eliminate degradation caused on the fibre due to wet/ dry cycling. This is shown with the flexural properties after 25 cycles, 30 and 50% SF composite contained maximum strength values of 200.4% and 159.4% respectively. However, prior to wet and dry cycling only 50% SF composite had similar post cracking toughness to the control. Where 10% and 30% SF composites had 35.4% and 27.4% lower values in toughness respectively. These results are in accordance with Machado et al. [74] that discussed the increase of SF content can increase the porosity of the material, reducing composite toughness. Although the results of SF incorporation are significant, the economic viability is burdened [34].

The benefit of SCM integration can be a denser matrix that can reduces voids and permeability within the composite design [31,51]. This is shown to reduce fibre pull out while minimising water and moisture movement. When the matrix is permeable, chloride ions can integrate faster causing a higher rate of carbonation in concrete composites. However, the integration of KFs can bind chloride ions and prevent the progression throughout concrete. This prevention is a result of the microstructural permeable pores created because of the fibres. This enhances the prevention of corrosion and spalling within concrete applications [68]. Due to the hydrophilic nature of KFs, it is expected that there would be a higher water absorption within the mix and therefore reduce the cracks formed from plastic shrinkage. Booya et al. [67] incorporated SCMs to mitigate the loss of compressive strength within concrete KF composites. KFs were integrated with SCMs including FA, MK, SF, Pumice powder and GBFS. The compressive strength was measured before and after the addition of SCMs, with MK achieving the highest compressive strength at all ages of testing. It is important to note that the replacement value of cement with SCMs was 10%. There was also a significant reduction in permeability with MK and SF due to the transformation of large pores into fine pores. This was shown to be caused by the pozzolanic reaction between MK and SF during early hydration, resulting with a denser composite.

A benefit of reducing the sorptivity index with matrix modifications can be shown with the increased durability of the composite material. This has been shown in KF concrete composites with the integration of SCMs [67]. Cellulose fibres are known to absorb water and cause fibre detachment and microcracks within the composite matrix when the water or moisture evaporates. However, when fibres have been modified this factor reduces. When integrating SCMs such as SF or MK, the sorptivity index reduces showing a reduction of 5–15% and 6–16% respectively. However, the integration of FA, GBFS and Pumice powder showed an increased sorptivity with only GBFS showing a reduction in the first 28 days. These results correspond with the water immersion tests also carried out by the researchers [67]. The effect of GBFS integrated with KFs has also been shown in the study by Roma et al. [72] who highlighted a severe reduction in mechanical and durability properties due to the alkaline attack and petrification of fibres. This led to progressive micro-cracks and a reduction in composite toughness. Their research demonstrates that GBFS is not as effective to other SCMs when reducing the degradation caused on KF composites. This is demonstrated with a decrease in first crack strength after 1 wet and dry cycle, containing 10–70% GBFS [34]. The mechanical properties are also hindered prior to wet and dry cycling experiments, with composites showing a 23.3–27.3% decrease in flexural strength. However, composites containing 90% GBFS showed similar mechanical and durability properties. The results were similar even after 25 wet and dry cycles. Although the results of high GBFS content appears promising, the microstructure of the composites has not been significantly discussed [34]. Other research studies conducted by Savastano et al. [55] have shown the high integration of GBFS can lead to a severe lowering of flexural strength (13 MPa 17 MPa) in aged composites when compared to OPC KF composites. The researchers discussed there was an increased level of carbonation followed by leaching and progressive microcrack formations in the composite when using GBFS as a cement replacement.

Other matrix modifications using FA have shown mixed results. Increasing the amount of FA content in the composite design has shown decreases in flexural and durability properties subjected to wet and dry cycling. On the contrary, as the amount of FA increases, the composite toughness increases with wet and dry cycling [34]. Other FA composite research [64] show negligible results when FA is included in the design mix. MK integration has been used as a SCM matrix modifier. The durability properties when 30% MK is integrated is comparable to the control. Initially, after one wet and dry cycle, MK exhibits 56.3% flexural increase. However, after 25 cycles, the material then performs similar to the control. This could be due to the consumption of $Ca(OH)_2$ in early hydration as compared to other materials. Although there is an initial increase of strength when subjected to wet and dry cycles, only after 15 cycles there was a steady reduction [34]. Although MK is not a common SCM that is integrated within KF composites, the material is becoming more commercially used due to the effective mechanical and environmental benefits [83].

Matrix modifications have been shown to enhance the fibres longevity within a composite design. This is further shown when no matrix modifications have occurred and there is a significant decline in mechanical properties as the ageing process continues [56]. Tonoli et al. [60] demonstrates this mechanical failure when using refined KFs. Other research [71] demonstrates the acceleration of fibre absorbing chemical properties such as, C-S-H, portlandite and calcite phases. It was shown that when no modification has occurred, the KFs induce the loss of their reinforcement capacity. Therefore, as the research suggests, further exploration of an effective mix design is required to mitigate the common findings found within the literature.

The summary of the discussed thirty one analysed research publications is illustrated in Fig. 8. The Figure represents all experimental research conducted on KF cement-based composites. Three parameters were measured of SCM integration, mechanical and durability studies. As discussed in this review, the causation factors of degradation have been thoroughly reviewed. While other areas of experimental research have remained seldom. These areas of research include tensile and compressive strength properties, acoustic and thermal measurements as well as sustainability assessments. Utilising the information within this review can direct future researchers on the development of a comprehensive mix design to mitigate the negative effects currently being created. The discussed maximum flexural values are illustrated in Fig. 9. This Figure represents materials containing an age of 28 days, with higher MOR values often containing SCM integration.

7. Conclusion, limitations, and future research

The building and construction industry has been a leading contributor to the generation of waste materials. The key materials used across the entire industry are concrete and cement-based products. The extraction of virgin materials to produce these products, creates significant environmental burdens and depletes

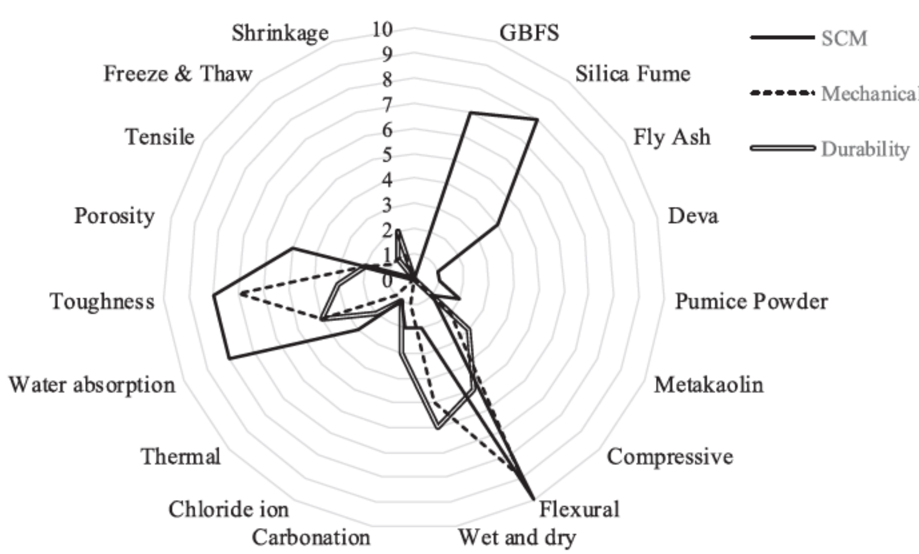

Fig. 8. KF analysed experimental research publications.

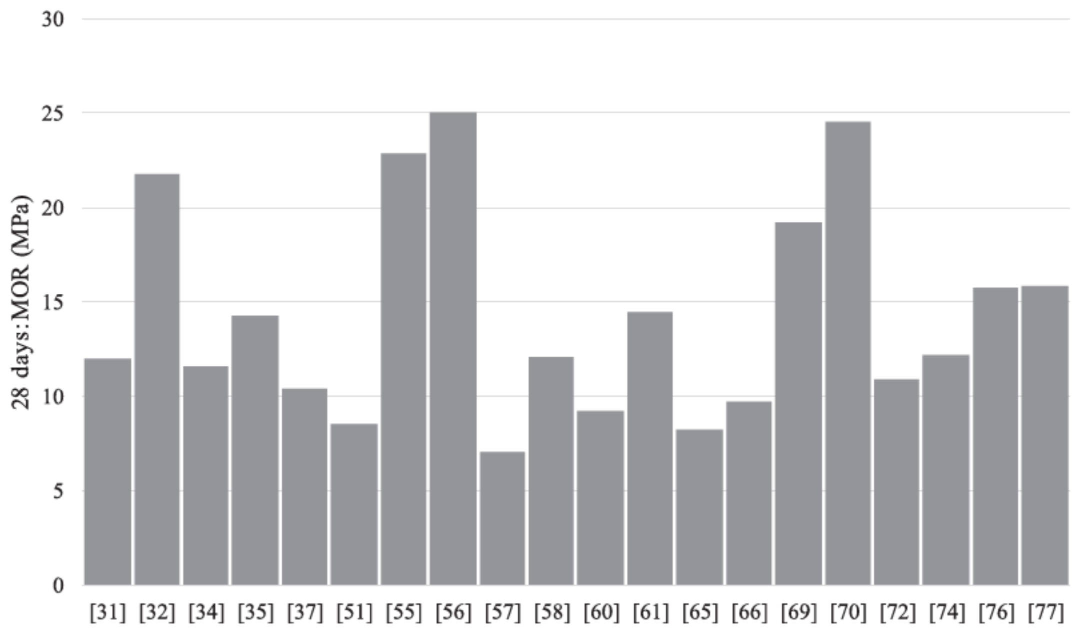

Fig. 9. Mechanical values of KFC.

important resources. The use of waste materials to substitute varying amount of cement percentages have been a research focus for many years. However, not all waste materials have been optimised to their full potential. So far, the valorisation of cardboard and paper waste have been relatively unexplored with minimal integration toward the building and construction industry.

The findings within this current study aimed to review waste fibres used within building and construction and cement/ concrete applications. Initially, a bibliometric analysis was conducted to identify research focus areas of waste fibre materials used in the building construction industry. The bibliometric analysis reviewed 874 documents from the year 2000–2020. The results highlighted influential journal sources with a measured growth in the research focused areas. The results demonstrated key words associated with the reviewed articles and sources, allowing the reader to identify waste research focus areas. These findings can allow future researchers to easily identify current research focus areas and research gaps. Future researchers can review these factors and minimise their search analysis to provide thorough assessments for their results and findings. The limitations and assumptions within this review are:

- The review focused on research conducted within the last two decades. Further findings could be identified from a larger timescale.
- The findings from the bibliometric assessment indicated studies on cardboard waste remained seldom. This was conducted through the Web of Science "WoS" search engine. Other search engines could withhold different studies on the topic of research.
- The review focused on utilising cardboard waste within cement-based materials. There could be other studies using the waste source within polymeric materials.

Despite these limitations, the findings from the analysed articles indicated that an optimised mix design has not been thoroughly evaluated to reduce the degradation caused on the fibres. Currently, researchers have provided critical information demonstrating the causation of mechanical and durability reductions when virgin KFs are applied within a cement-based environment. However, only five research articles have sourced KFs from cardboard waste materials. The current study highlights the need to explore other avenues of fibre and matrix modifications via experimental analysis to overcome the key areas of material degradation. This will enhance the ability to source KFs from other avenues such as cardboard waste to be further used in the building and construction industry.

CRediT authorship contribution statement

Robert Haigh: Conceptualization, Investigation, Writing - review & editing. **Malindu Sandanayake:** Supervision, Review & editing. **Yanni Bouras:** Supervision, Review & editing. **Zora Vrcelj:** Project administration, Supervision, Review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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2.3 Concluding remarks

This chapter presented the mechanical and durability factors of KFs within cementitious composites. The distinguishing properties of natural fibres and KFs were discussed, demonstrating the degradation of hemicellulose components during the kraft process. The causes of degradation on the fibre walls and the reduced mechanical effects on composite materials were also reviewed. The chapter presented future avenues of research to further enhance the integration of waste cardboard within cementitious composites.

In summary, the following conclusions are drawn from this chapter:

- An optimised mix design has not been thoroughly evaluated to reduce the degradation caused on KFs.
- There are minimal experimental investigations using KFs derived from waste cardboard.
- Fibre modification is required for the successful integration of KFs within a cementitious composite.
- Fibre and matrix modifications using silica fume and metakaolin, respectively, have not been used with KFs.
- Raw cardboard fibre integration between 2-8% has shown beneficial mechanical results.

Chapter 3 – Mechanical analysis

3.1 Introduction

In this chapter, the mechanical performance of KFs within concrete composites were experimentally investigated. For evaluation of the compressive, tensile, and flexural strength properties of KFs within concrete composites, three mix designs were used. The findings presented from the literature review discovered the optimal amount of fibre integration was 2-8%. Therefore; 5% fibre integration was selected as a partial cement substitution in concrete. A microstructure analysis was conducted to examine the morphology of the fibre pre-treatment as well as the matrix of the composite material. SF was applied as a wet slurry to the fibres as a fibre pre-treatment. The matrix was also modified using metakaolin, as a partial cement replacement. These materials were integrated based upon the findings from the previous chapter.

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The mechanical performance of recycled cardboard kraft fibres within cement and concrete composites

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ABSTRACT

Global population growth around the world requires significant infrastructure and building developments. The building and construction industry uses excessive quantities of virgin resources for those developments as well as contributing to the waste generated from the residential and commercial sectors. A contemporary solution is required to reduce these negative environmental impacts. In this study, the mechanical properties of cement composites containing kraft fibres (KFs) derived from waste cardboard was experimentally investigated. KFs and metakaolin (MK) were integrated within concrete samples as a cement substitute material. The compressive, flexural, and tensile strength was determined on three mix designs containing 5% raw KFs, 5% surface modified KFs, and matrix modified concrete specimens. Silica fume (SF) was applied to the fibre walls as fibre modification to lower the alkaline zone around the fibre within the cementitious matrix. 5% MK was used as a partial cement substitute to lower the alkaline level of concrete samples. All KF concrete specimens exhibited lower compressive strength properties. However, MK modified samples exhibited the highest tensile strength of 11 MPa. Fibre modified samples had stronger compressive and tensile strength of 20 and 9 MPa, compared to raw KFs. However, raw KFs exhibited a higher flexural strength of 2.5 MPa. The compressive, tensile, and flexural strength of the control were 25, 10 and 2.6 MPa, respectively. Scanning electron microscopy (SEM) observations demonstrated sufficient SF adhesion on the fibre walls, while the energy dispersive x-ray spectroscopy (EDS) observations showed efficient dispersion of all composite materials.

1. Introduction

In recent years, growing climate awareness has placed significant attention on the use of environmentally friendly materials within the building and construction industry [1]. Alternative solutions are required that offer eco-friendly techniques and processes to minimise the impacts of construction on the environment. Researchers have focused on the integration of waste within building materials as a possible solution to aid in the reduction of landfill and resource extraction [2–5]. This method provides a contemporary solution to reduce the burden on the environment and move towards a circular economy [6,7].

Over the past few decades, research studies have focused extensively on experimental investigations using waste materials such as glass, fly ash (FA), plastics, tyres, and demolition waste [2,8,9]. Integration of glass, tyres and plastics were heavily researched on road asphalt pavement construction and bitumen applications [10–12], whereas FA was predominantly integrated in concrete applications [2,5,8,12–15]. Following glass and plastic, cardboard utilises the most volumetric space in the residential recycle bin [16]. In 2018–19, Australia collected 5.9 million tonnes of cardboard waste and with current waste export restrictions to China, the nation is exploring wide range applications of reusing cardboard waste effectively [17]. Minimising the accumulation of cardboard waste will significantly reduce the burden of landfill management. Currently, there is approximately 270 million tonnes of paper and cardboard waste landfilled within Australia [17]. Reducing the addition of these materials to landfill is an urgent requirement.

Kraft fibres (KFs) are the main constituent of cardboard material which are natural fibres (NFs) containing cellulose matter that provide strength in plants and trees [18]. Use of NFs have attracted the attention of researchers due to their renewability and reduced global warming impacts when compared to the use of synthetic fibres [19–23]. NFs also possess other advantages such as non-hazardous, non-abrasive, biodegradability, high strength, low density and low costs [24]. Recent

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research studies of KFs focused on partial cement substitution within fibre cement boards, however, the application was not commercially adopted due to the reduced mechanical effects [25–30]. Other materials that were researched as a partial cement replacement included the use of FA, glass powder, ceramic waste, wood ash, ground blast furnace slag (GBFS) and waste tyres [13,31–35]. Researchers have also investigated supplementary cementitious materials (SCMs) due to the reduction of carbon dioxide (CO_2) emissions and energy consumption in concrete. SCMs have also been shown to improve the strength properties of concrete, reduce the high alkaline levels and lower cement requirements [36,37]. The production of cement contributes significantly to negative environmental impacts, therefore, reducing the cement content in concrete applications has been of research significance. The effect on the environment from cement manufacturing include excessive natural resource depletion, intense energy demands, air, and water pollution. Cement manufacturing industry accounts for significant energy consumption and is responsible for approximately 5% of worldwide annual greenhouse gas emissions [38]. Therefore, to reduce the burden of cement consumption, a contemporary solution is required. To successfully integrate KFs within cement-based composites, an exploration of matrix and surface modification is essential [39]. This is due to the high alkaline percentage of composites during cement usage. Researchers have stated that high alkalinity levels can cause severe degradation to the fibre wall of KFs [4,24,25,39–46]. The degradation process can make the fibre weak and brittle, ultimately reducing the service life of the composite application.

To reduce alkaline percentage within cement-based composites, inclusion of various SCMs such as FA, GBFS, Metakaolin (MK) and Silica Fume (SF) were researched [47–50]. These materials have proved effective as a partial cement replacement when used within KF composites; however, the service life is always reduced due to the adverse effects caused on the KFs. The successful integration of KFs within cement-based composites are dependent on the ability to reduce calcium hydroxide ($Ca(OH)_2$) attacking the fibre walls. Reducing the amount of $Ca(OH)_2$ contained will enhance the service life of the application. MK contains a higher percentage of aluminate (Al_2O_3) than other SCMs and has been shown to enhance the hydration of cement in early ages. This is due to the large generation of heat caused by the rapid conversion of Al_2O_3 within the first few minutes. Several researchers [24,39,51,52] included 10–50% MK substitution with Ordinary Portland Cement (OPC), showing an enhanced durability of NFs. The rapid consumption of $Ca(OH)_2$ reduces the high alkaline percentage that attacks the fibre walls. When compared to FA, GBFS, SF and pumice powder, Booya et al. [24] demonstrates values of 10% MK substitution with OPC can achieve increased mechanical strength while also lowering water absorption and chloride ion permeability. MK was shown to be the most effective SCM as a cement replacement when used in conjunction with NFs [39,52].

Integration of SF with NF cement-based composites was used to increase durability, workability, compressive and flexural strength [24,30,39,51–56]. An increased percentage replacement of SF showed significant compressive strength improvements, especially after wet and dry cycling analysis. 30% and 50% SF showed flexural strength improvements of 200.4% and 159.4% respectively [39]. Mohr et al. [39] noted that 30% or more SF can reduce degradation of NFs significantly, this is in accordance with the findings in [54]. However, large percentages of SF can reduce workability and increase costs. SF can enhance the formation of calcium silicate hydrate (C-S-H). This occurs with the reaction of $Ca(OH)_2$ crystals formed from hydration of calcium silicates, further increasing the production of C-S-H [56]. This can increase the strength of composite materials and enhance the bond between the fibres and matrix. SF has a high silicate dioxide (SiO_2) content, that consumes $Ca(OH)_2$ at later stages, however, as mentioned KFs require the consumption of $Ca(OH)_2$ rapidly to mitigate the degradation. Therefore, to counter the degradation caused on KFs, materials must be selected that can mitigate $Ca(OH)_2$ at both early and later stages during hydration. For this reason, MK and SF have been selected as most suitable for this study.

This paper presents an investigation of the mechanical performance of concrete when waste cardboard has been integrated in the design mix. The findings demonstrate the effect when both matrix modification and surface modification of the KFs has occurred using MK and SF respectively. The compressive, flexural, and tensile properties are measured over the 7, 14 and 28-day period. In addition, the microstructure of the various mix designs are examined using scanning electron microscopy (SEM) to comprehensively analyse the variation in strength of the composite designs.

2. Experimental procedure

2.1. Raw materials and preparation

Waste corrugated cardboard and MK are the main constituent materials used to reduce the consumption of OPC within the mix design of the concrete specimens herein. Waste cardboard was reduced to a pulp material then combined with SF as a wet slurry to create Silica Fume Kraft Fibre (SFKF). This method is graphically depicted in Fig. 1. The moisture is removed from the SFKF via a conventional oven at 20 °C for 8 h. SFKF is then subjected to rotation within a blender mixer. The result is a fibrous material that can be integrated within the concrete mixture. The SF applied to the KFs conforms with the Australian Standard AS/NZS 3582.3 [57] specification of Silica Fume used in cementitious mixtures. MK is used at a 5% percentage level in conjunction with 5% SFKF. The MK used conforms with the ATSM C-618 [58], Class N specifications for natural and calcined pozzolans. OPC is used as the primary constituent for the pozzolanic reactivity of the concrete specimens. The material conforms with AS/NZS 3972 [59]. The composition of the materials is further shown in Table 1. Locally available coarse and fine aggregate were applied, conforming to AS/NZS 1141.6.2 [60] and AS/NZS 1141.5 [61] respectively. Regular portable tap water was used throughout the preparation of the experimental mix design.

2.2. Mix designs

For all concrete mix designs, the target compressive strength was 25 MPa. This is shown to be a primary strength requirement of concrete material applications within Australian residential construction [62]. To comprehensively analyse the reduction of OPC when integrating MK and SFKF, multiple mix designs are formulated as shown in Table 2. Water, fine and coarse aggregates remained the same throughout the various designs with a mass ratio of 0.33, 1.15 and 1.73 per kilogram of concrete respectively. Fibre investigation of SFKF ascertained the effect of fibre modification within the cement matrix. KFs without modification were also investigated to understand the effect of SF on the fibre walls. 5% MK was included to reduce the overall OPC quantity. The mix code is described with the type of fibre, fibre percentage and MK content. For example, SFKF55 correlates to SFKF as the fibre, 5 is the percentage of fibre and 5 is the percentage of MK. When MK is not used in the mix design, only the fibre type and percentage is present, this is shown in KF5 for example. KF corresponds with raw kraft fibre and 5 is the percentage of fibre in the mix design. Current research findings [29,30] demonstrate a 2–8% optimisation of fibre content within concrete and mortar specimens. Their findings discussed the workability of the specimens are compromised when excessive fibre content is within the design mix. Therefore, 5% fibre integration has been conservatively chosen to reduce the negative effects of the KFs within the cementitious matrix. It is important to note that the water ratio remained the same throughout all mix designs. Researchers [63] have recommended to increase their water content to accommodate the increase of fibre integration. However, due to the low fibre content of this research, water absorption rate of fibres was deemed negligible.

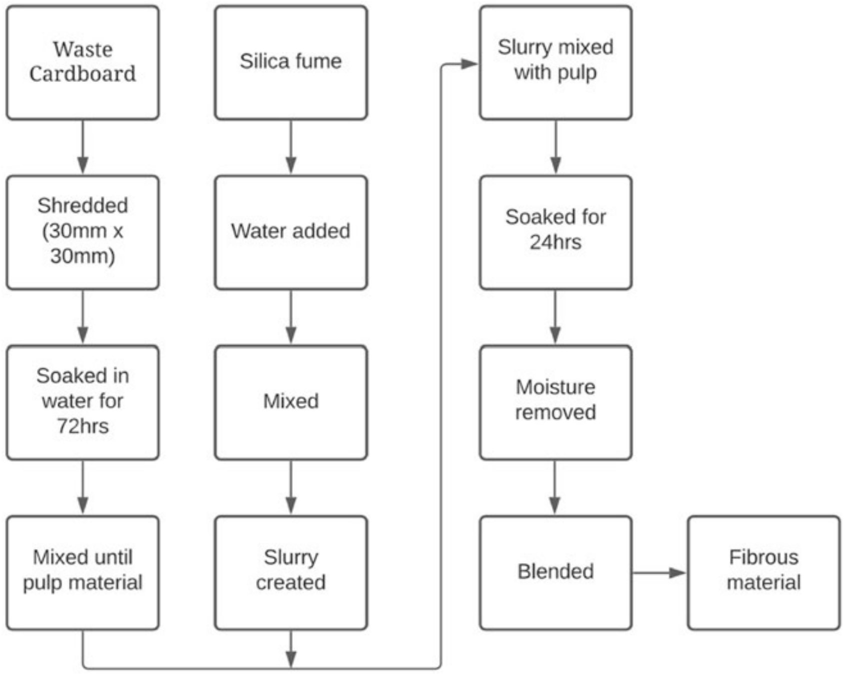


Fig. 1. Methodology of SFKF.

Table 1
Chemical composition of pozzolanic materials.

Chemical	Material component %		
	MK	SF	OPC
SiO_2	54–56	>= 75- <100	19–23
Al_2O_3	40–42		2.5–6
Fe_2O_3	<1.4		
TiO_2	<3.0		
SO_4	<0.05		
P_2O_5	<0.2		
CaO	<0.1		61–67
MgO	<0.1		
Na_2O	<0.05		
K_2O	<0.4		
L.O.I.	<1.0		
Silica, amorphous, fumed, cryst. -free		>=0.3- <1	
$CaSO_4{\cdot}2H_2O$			3–8
$CaCO_3$			0–7.5
Fe_2O_3			0–6
SO_3			1.5–4.5

Table 2
Mix design percentage of altered materials.

% of OPC reduction design materials				
Mix code	OPC	MK	SFKF	KF
Control	100	–	–	–
SFKF55	90	5	5	–
SFKF5	95	–	5	–
KF5	95	–	–	5

2.3. Specimen preparation and curing

The raw materials were mixed using a standard cement or concrete mixer. The materials were dry mixed for 5 min before adding water. This allows sufficient agglomeration of all materials. After adding water, the materials were mixed for an additional 5 min. Upon completion of mixing, the concrete was added to the various moulds in three layers. A steel rod was used to compress the materials twenty times before the additional layers were added to the mould. The moulds were then kept at a constant room temperature of 20 ℃ for 24 h, before added to the curing baths for 7, 14 and 28-days. There were three specimens per test and all samples were checked for their acceptable slump value in accordance with AS 1012.3.1 [64].

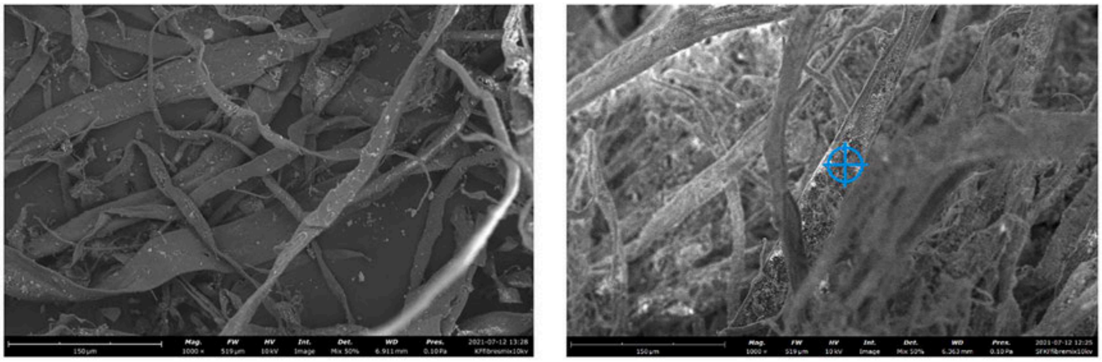
2.4. Testing procedure

The compressive strength test was performed on 200 × 100 mm cylindrical specimens in accordance with AS 1012.9 [65] with a load rating of 20 MPa/ minute. The flexural strength properties were determined on the concrete beams by conducting a four-point bending test. The size of the specimens were 100 × 100 × 350 mm with a load rating of 1 MPa/min in accordance with AS 1012.11 [66]. The tensile strength properties were determined via indirect tensile testing, also known as splitting tensile test. The concrete cylinders were 100 × 200 mm with a load rating of 1.5 MPa/min in accordance with AS 1012.10 [66]. The compressive, flexural, and tensile values were measured at 7, 14 and 28-day intervals, with an average recorded of three samples for each mix design. All mechanical tests were conducted on the Matest C088-11 N Servo-Plus evolution testing machine and Cyber-Plus evolution data acquisition system. The error bars of the recorded values represent the standard deviation on either side of the average. The microstructure of the samples was observed using a scanning electron microscopy (SEM) and an energy dispersive x-ray spectroscopy (EDS). These microstructure observations were conducted on the Phenom XL G2 Desktop SEM, samples were prepared using a diamond cutting saw to a height of 2–4 mm and 4–6 mm in diameter. The EDS analysis determined the chemical composition of the various mix designs.

3. Results and discussion

3.1. Microstructure

The microstructure of the reinforced composites was investigated for each mix design. There was an effective rate of fibre dispersion within the specimens and the mixing method adopted proved to be successful when dispersing the fibre materials within the concrete composites.



(a) Raw kraft fibres (b) Silica fume kraft fibres

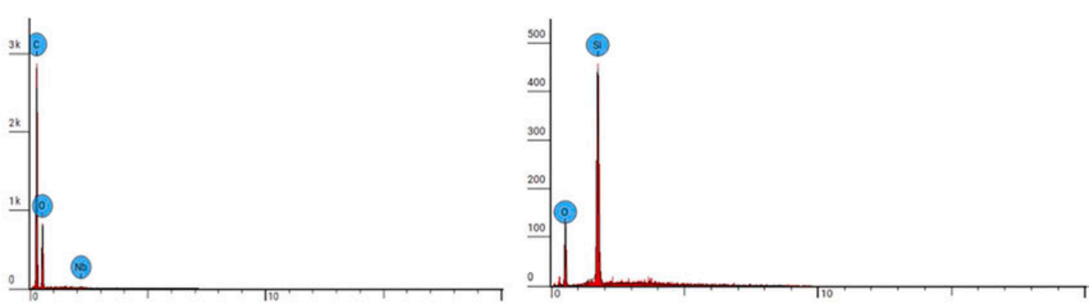
Fig. 2. Recycled kraft fibres modified and non-modified.

Fig. 2 illustrates the raw and modified fibre content. As can be observed, the fibre content various dramatically in size from approximately 10–36 µm. Although the fibre size is not uniform, it does allow the rate of fibre dispersion to increase. This allows the fibre to enter pockets and voids within the microstructure during the formation and agglomeration of the composite materials. The variation of size with KFs is shown in Fig. 2 (a). This is because of the production and procurement of cellulose fibres for manufacturing cardboard products. The raw cellulose fibres are subjected to a complex chemical treatment to produce wood pulp. A caustic soda and sodium sulphide are often used as a liquor to then cook the wood pulp and release the bonding of the fibres. This process removes the lignin attached to the fibres to increase random fibre dispersion and size [30].

The composite samples when substituted with fibre and MK exhibited a drop in pH level. This was shown with pH levels of 11, 12 and 12 for samples SFKF55, SFKF and KF, respectively. Although this is not a significant reduction when compared to pH level of 14 for the control, it does demonstrate the reduction of alkalinity within the matrix environment. This reduction is shown to minimise the cement products attachment to the fibre cell walls and reduce fibre petrification as shown in Fig. 3. It is important to note that the attachment between the SF and composite matrix is critical to establish an anchorage point for the SFKFs. This anchorage point is crucial for the bearing capacity of the fibre to withstand an increase of axial loading on the composite. The interfacial bond of the material is more critical than the mechanical properties of the individual fibre material [56]. SF creates a consumption of calcium hydroxide at later stages which increases the longevity of the fibre. During this process, C-S-H nucleation occurs which provides strength to the cement matrix.

Fig. 2 (b) shows SF particles have attached themselves to the fibre walls sufficiently before integration within the composite material. This is further seen in Fig. 4 that contains the EDS report of the raw KFs and SFKFs. The higher the percentage of SF on the fibre walls will reduce the alkaline zone around the fibre when applied in the OPC composite mix [54]. An additional benefit to modify the fibre with SF is to create an increase of surface area on the fibre. Although this enhancement may be minimal, it is effective when dispersing the fibre in a greater area [67]. The load transfer rate on the fibre is increased when subjected to axial loading because of the surface modification.

The SFKF55 interfacial properties between the fibre and composite matrix are shown in Fig. 3. The samples in the SEM images are segments of the composite material. The Figure illustrates minimal precipitation of SF and cement products on the fibre, highlighting the reduction of alkaline attack on the fibre walls. Fig. 5 illustrates advanced petrification on the non-modified fibre walls. This image depicts severe attachment of hydrated cement products which causes the fibre to become weak and brittle. Fig. 3 demonstrates the matrix of the composite to be dense and rough in texture. This suggests improved bonding of fibres and lower moisture transfer rates. The minimisation of moisture transfer rates can be linked to the reduction of the pore microstructure of the specimens. Similar observations were reported by Booya et al. [24]. During the early stages of hydration, the saturation of gel pore water

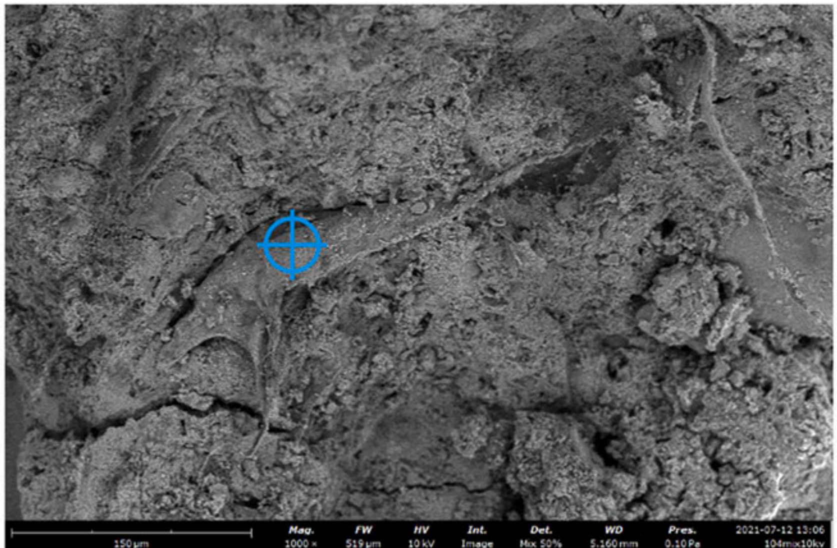


Element Number	Element Symbol	Element Name	Atomic Conc.	Weight Conc.
6	C	Carbon	70.551	63.200
8	O	Oxygen	29.160	34.800
41	Nb	Niobium	0.289	2.000

Element Number	Element Symbol	Element Name	Atomic Conc.	Weight Conc.
8	O	Oxygen	36.294	24.500
14	Si	Silicon	63.706	75.500

(a) Kraft fibre (b) Silica fume kraft fibre

Fig. 3. SFKF55 fibre and matrix analysis.



Element Number	Element Symbol	Element Name	Atomic Conc.	Weight Conc.
6	C	Carbon	33.241	23.500
8	O	Oxygen	53.301	50.200
13	Al	Aluminum	0.630	1.000
14	Si	Silicon	7.923	13.100
20	Ca	Calcium	4.705	11.100
41	Nb	Niobium	0.201	1.100

Fig. 4. EDS report of raw KF and SFKF.

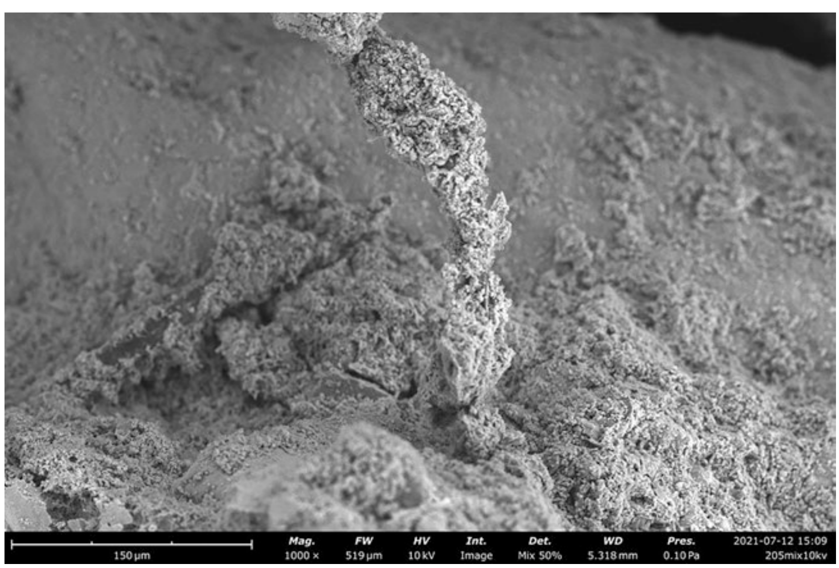


Fig. 5. Total petrification of KF walls.

intensifies as C-S-H growth occurs. This changes the result of the capillary pores located near the KFs. The capillary pores decrease due to the hydrophilic nature of KFs, leaving an increase of consumed water within the matrix of the composite. This results with a decrease in porosity as compared to composites containing hydrophobic fibres [56].

The bonding ability of the SFKFs is graphically depicted with the increase of load transfer as shown in Fig. 6 (b). There are two bonding mechanisms exhibited between the fibre and cement interfacial zone. Firstly, mechanical interlocking between the fibre and hydrated cement products creates an anchorage point on the fibre. Secondly, chemical bonding between covalent O–H bonds and weaker hydrogen bonds [56]. Fibre debonding has been the dominant cause when lower mechanical values are exhibited [24]. Fig. 3 illustrates the embedded fibre, suggesting fibre pull out has not occurred. This demonstrates the increased pore microstructure surrounding the fibre within the composite material. This can be attributed to the SF modification undertaken prior to fibre composite application.

The influence of MK within the composite matrix can be seen in

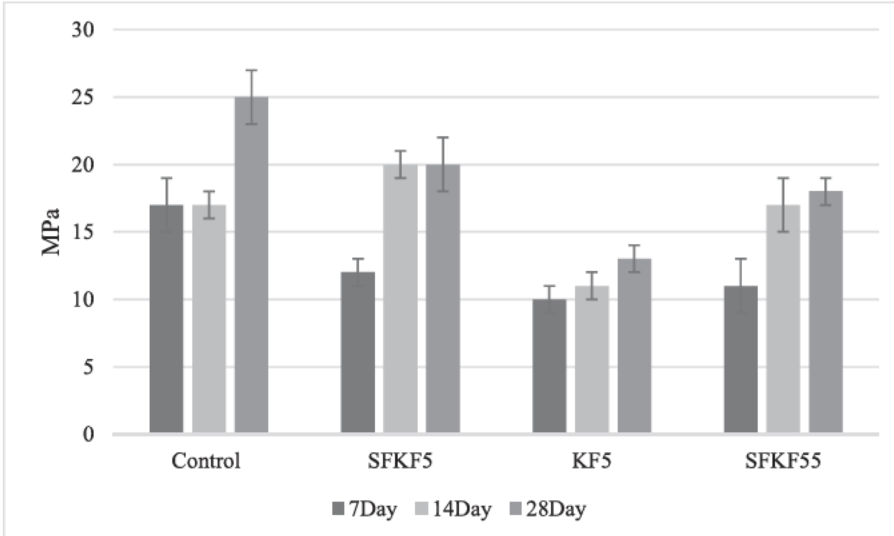


Fig. 6. KF5 & SFKF5 Compressive strength vs strain.

Fig. 3. The fine MK particle size contains a higher surface area than cement. These particles reduce the pore size within the composite, this leads to less voids which also decreases permeability of the binary concrete blend. This further illustrates the increase of strength values for the SFKF55 composite, as hydration occurs MK is consuming $Ca(OH)_2$ during the early stages while SF consumes $Ca(OH)_2$ at later stages of ageing. This is in accordance with researchers that demonstrate the integration of SCMs transforming large pores into smaller pores. This reduces the permeability of concrete specimens. Booya, et al. [24] demonstrates SCM composites containing NFs exhibit lower absorptivity index rate when using SF and MK. Their research reported lower levels of MK integration were sufficient to enhance durability properties when compared to other SCMs such as GBFS and FA.

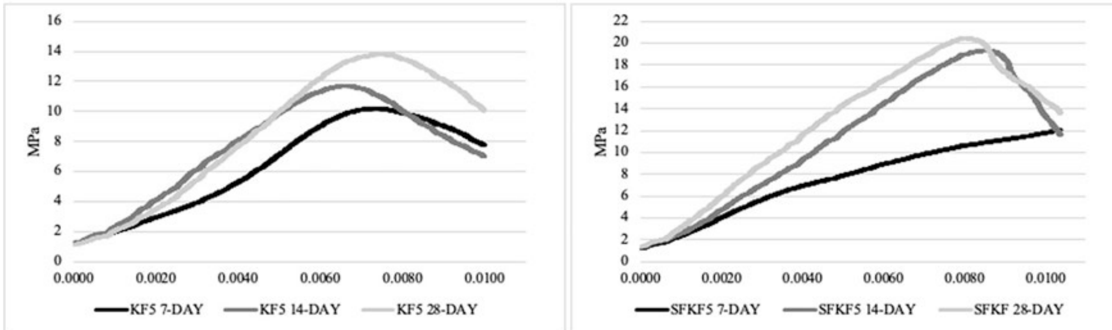
A reduction in permeability is primarily linked to the hygroscopic nature of cellulose fibres. NFs absorb moisture and once the completion of hydration has occurred, the moisture exits the composite material leaving voids and pockets surrounding the fibres. Due to this differential drying occurrence between the fibre and the matrix, the fibre-cement bond is often compromised. Claramunt et al. [68] described a process called 'hornification' to counteract this factor occurring. The researchers demonstrated that with sufficient pre-treatment of fibre material, NFs can show minimal moisture retention even when applied in a high moisture environment. This research attempted to the reduce the hydrophilic action of KFs via the removal of moisture from the fibre once SF was integrated on the fibre walls. With this phenomenon, the fibre had previously encountered two cycles of moisture retention and moisture release, firstly via the production of cardboard products then secondly to produce fibres for this experimental research. Other researchers [56] integrated nano-silica-fume (NSF) to combat the action of water retention with KFs. Their research reported a reduction of porosity and an increase of fibre-cement bonding with the interfacial zone of the composite material. As can be seen in Fig. 3, the fibre is sufficiently embedded within the matrix.

There was sufficient agglomeration of MK and SF fibres within the mix design. This was shown in an area mapping of the composite within the EDS report. The results of the mapping are shown in Table 1. The mapping displayed traces of aluminium that can be accredited for the integration of MK. Silicon can attributed to the SF while the dominant element is calcium that is primarily linked to cement. This mapping further suggests that SF once integrated in a high moisture environment may break away partially from the fibres. This is seen in Fig. 3, illustrating the composition of the fibre wall within the SFKF55 matrix. The fibre walls have traces of calcium and silicon however, the image displays primarily raw carbon of the cellulose matter. Although SF may attach itself adequately in a low moisture environment, once moisture is presented, the material appears to separate partially from the fibre walls.

3.2. *Compressive strength*

The variation of compressive strength for concretes with various fibre types and sample composition are shown in Fig. 7. The coefficient variation of the compressive strength was between 1 and 2 MPa. The standard deviation is shown via the error bars on Fig. 7. The compressive strength of all samples decreases with fibre integration. The inclusion of fibre content in a cement matrix reduces the density and increases voids of composite specimens [24,63]. The compressive strength values of plain concrete (control) measured at 7-, 14- and 28-day intervals were 17, 17 and 25 MPa, respectively. Although the control sample contained the highest value on the 28-day measurement, SFKF5 concrete contained a higher value at the 14-day measurement. SFKF5 concrete strength was 12, 20 and 20 MPa at 7-, 14- and 28-day respectively. The SiO_2 content that attached to the fibre walls has consumed $Ca(OH)_2$ at the later stage. This is shown with a significant increase of strength when compared to the values at the 7-day measurement. The values for KFs without modification are shown with a compressive strength of 10, 11 and 13 MPa at 7-, 14- and 28-day, respectively. These values are significantly lower due to the attachment of hydrated cement products on the fibre walls. This is shown in Fig. 3 where mineralisation has formed on the fibre. This process weakens the fibre and causes embrittlement to the fibre walls [39].

SFKF55 samples exhibited a strength increase of 11, 17 and 18 MPa, at 7-, 14- and 28-day, respectively. The consumption of $Ca(OH)_2$ in early and later stages by MK and SF respectively, has reduced the attachment of cement products on the fibre wall and created a stable environment for the strength of the fibre concrete to increase with time. Although there is no rapid increase of strength properties, the fibres can maintain their strength and durability characteristics within the composite design. The integration of modified and non-modified KFs has reduced the overall compressive strength at the 28-day measurement. This is primarily due to the hydrophilic nature of the KFs which is under duress by the attachment of cement products. KF composites at 7- and 14-days contained the lowest strength which is a 70–58% reduction respectively when compared to the control. Although this is primarily attributed to the attachment of cement products on the fibre walls, researchers have contributed lower mechanical strength due to fibre clumping within



(a) KF5 Compressive strength vs Strain (b) SFKF5 Compressive Strength vs Strain

Fig. 7. Variation of compressive strength.

composite designs [69]. When the fibre composites are subjected to axial loading, there is a load transfer occurring within the microstructure between the fibres and the composite matrix. As the load increases, there is an increase of friction imposed on the fibre walls. If a significant amount of petrification has occurred on the fibre walls, the fibre will fail at a faster rate. This is shown in composite samples in Fig. 6 (a), where the maximum fibre strain is significantly lower than the modified fibre strain in Fig. 6 (b). Several researchers [30,56] outlined the effect of crack bridging during the fracture process of fibre reinforced concrete and mortar samples. Crack bridging is when fibres will span in the voids between two cracks.

Crack bridging induced by fibres can improve the resistance size of crack openings. The formation of microcracks can be minimised with fibres, while mitigating the transference of those microcracks to larger crack propagation. Fig. 6 (b) graphically depicts this with the rapid decline of strength post fibre rupture. The fibres endured the maximum axial loading until failure occurred. Whereas raw KFs contained a lower strength yield with a reduction of formation of crack bridging as shown in Fig. 6 (a). The rounder curve of the gradient in Fig. 6 (a) suggests a slow fibre pull out failure mechanism. The raw KF composite also contains a better residual strain after yielding. Fig. 6 (b) illustrates SFKF composites have an increased bearing capacity, the steep curve after failure suggests a combination of fibre snapping and fibre pull out has occurred. This is further shown with a sudden reduction of residual strain when maximum capacity is reached. Fig. 6 (a) demonstrates this factor by having a lower energy absorption rate which is shown in the area below the curve. The other key factor that attributed to the increase of compressive strength of modified fibres was the increase of density. The ultrafine SF particles that attached to the fibre cell wall has protected the fibre from high alkalinity but also increased the density of the fibre microstructure [54]. This is also in accordance with researchers reporting a 25% compressive strength decrease when fibres were not modified [70]. The increase of strength can be accredited toward the improvement of fibre bonding within the material composition. This is seen in Fig. 4 (b) that illustrate the precipitation of SF primarily on the fibre walls. SF is effective when integrating within a cement environment because of the fine particles that act as a filler within the matrix. The particles reduce the porosity, forming increased levels of C-S-H that decrease the alkaline of the cement [46].

Materials with a higher density often have increased compressive strength. Khedari et al. [69] reported that the reduction in compressive strength can be attributed to the lower density of the properties of NFs. The properties are primarily concerned with the water absorption rate which can lead to an increase of voids and pockets within the composite matrix. When there is an increase of voids in the material, there is a decrease of material density. Therefore, as illustrated in Fig. 7, the reduction of strength with fibre integration is to be expected. Another critical factor is the ratio of materials used within the mix design. Booya et al. [67] reported an increased compressive strength of 19.1–45.6% when reducing the water-cement ratio from 0.5 to 0.35 in fibre reinforced concrete specimens. As previously discussed, the water content remained the same for all samples due to the minimal integration of fibre content.

3.3. Tensile strength

Fig. 8 graphically depicts the variation of tensile strength. The coefficient of variation of the tensile strength was between 1 and 2 MPa. The standard deviation is shown via the error bars in Fig. 8. The development of fibre composition within a modified cementitious matrix is a key factor toward the overall tensile strength bearing capacity. As discussed with the crack bridging effect, an increased level of microcracks form due to fibre pull-out or fibre snapping. Consequently, the axial loading is then redistributed to other fibres. This results with an increase of load bearing capacity. A similar trend can be found with the indirect tensile strength results as seen in the compressive strength behaviour. This behaviour is graphically represented in Fig. 8. However, the maximum strength values were shown with the composite design SFKF55. The values measured at 7-, 14- and 28-day intervals were 7, 10 and 11 MPa, respectively. Although the results were similar to the control, there remained a linear incline that was seen with the compressive strength. The control samples reached 8 MPa at the 7-day interval. This was also shown with the SFKF composite design. However, at the 14- and 28-day measurement, the strength remained the same at 10 MPa. This was quite similar to SFKF, that exhibited a strength value of 9 MPa at both 14-day and 28-day measurement. Research conducted by Toledo et al. [54] shows the immersion of NFs with SF creates a low alkalinity zone around the fibre, reducing the alkaline attack and mineralisation. Researchers agree that the non-cementitious nature of cardboard fibres can reduce the tensile strength within concrete and mortar samples [71]. However, this is specifically in reference to non-modified fibre and matrix designs. Further evidence that KFs require enhancement is shown with the tensile results of the KF mix samples. KF composites contained the lowest tensile strength, which was to be expected due to the embrittlement of the fibre within the high alkaline environment. It is interesting to note that the KF composite specimens although displayed lower strength than the other mix designs, the strength increased with curing time. KF composites measured at 7-, 14- and 28-day showed an increase with 4, 6 and 7 MPa, respectively. This shows that KFs that are not modified reduce the process of continuous hydration within the concrete samples. This can be due to moisture retention on the fibre walls. Moreover, SFKF55 allowed continuous hydration to occur, shown with a strength increase above the KF composites at the 7-day measurement, then a gradual incline between the 14- and 28-day interval. This demonstrates the effectiveness of MK as a binary blend with OPC within concrete and mortar mix designs. MK mortar produces an increase of heat in early stages when compared to 100% OPC. This reaction consumes $Ca(OH)_2$, which protects the fibres while increasing C-S-H gel [72]. Researchers have agreed that MK composites exhibit similar strength to the control, however, the

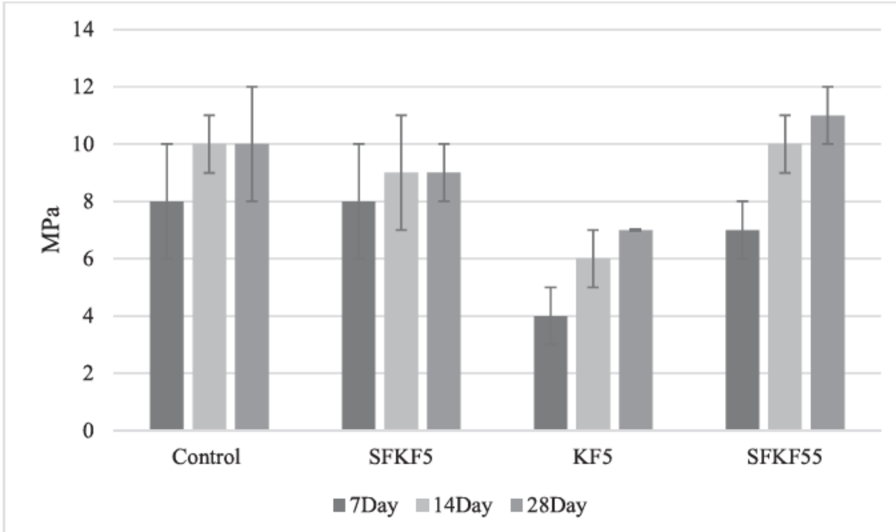


Fig. 8. Variation of tensile strength.

substitution ratio cannot exceed 30% [39].

3.4. Flexural strength

Fig. 9 shows the average maximum flexural strength, also known as the modulus of rupture (MOR). The coefficient variation of the flexural strength was between 0.2 and 0.1, illustrating the data points are close to the mean. The standard deviation is shown in the error bars in Fig. 9 which demonstrates the reliability of the recorded results. All samples subjected to flexural testing with fibre integration contained lower strength properties. This suggested fibre composite specimens withheld the 'balling effect' in flexural beams [30]. The balling effect refers to fibres clamping together causing weak points inside the composite. Fig. 3 illustrates the lack of SF and cement products on the fibre wall. This can also lead to a lack of composite strength, as fibres should be absorbed completely within the composite. As the fibre content increases, there is a lack of hydrated cement products to be dispersed among all surface areas. This leads to a disruption of agglomeration of composite materials which ultimately leads to a weaker matrix.

SFKF5 and KF5 composites contained an increase whereas the control and SFKF55 exhibited the same flexural strength for each composite design at both 7- and 14-day intervals. The control measured 2.4, 2.4 and 2.6 MPa at 7-, 14- and 28-day interval measurements, see Fig. 9. The indirect tensile and compressive strength demonstrated a reduction in strength when raw KF was used, however, KF composite flexural results showed increased strength when compared to all other composite designs. The reduction in flexural strength for SFKF and SFKF55 is most likely due to factors such as a nonuniform dispersion and a weakening response of the binary matrix [63]. Researchers agree that substituting cement with non-cementitious materials within a composite design will reduce the flexural strength of the material [30]. The flexural durability of the fibre reinforced composites can also be shown via the post-cracking behaviour. During the acceleration towards maximum load, the development of micro cracks can be prevented via the agglomeration of fibres caused by the bridging mechanism. This mechanism prevails until the density of micro cracks achieves maximum saturation and the composite cannot withstand additional implied stress [73]. As shown in Fig. 2 (b) the SFKFs are drawn together as a response known as the Van der Waals force [74]. The intermolecular attraction of KFs is increased when SF ultrafine particles are attached to the raw KFs. This attraction intensifies when water is introduced into the mix forming a stronger force of connection [74]. The raw KF composites have exhibited the

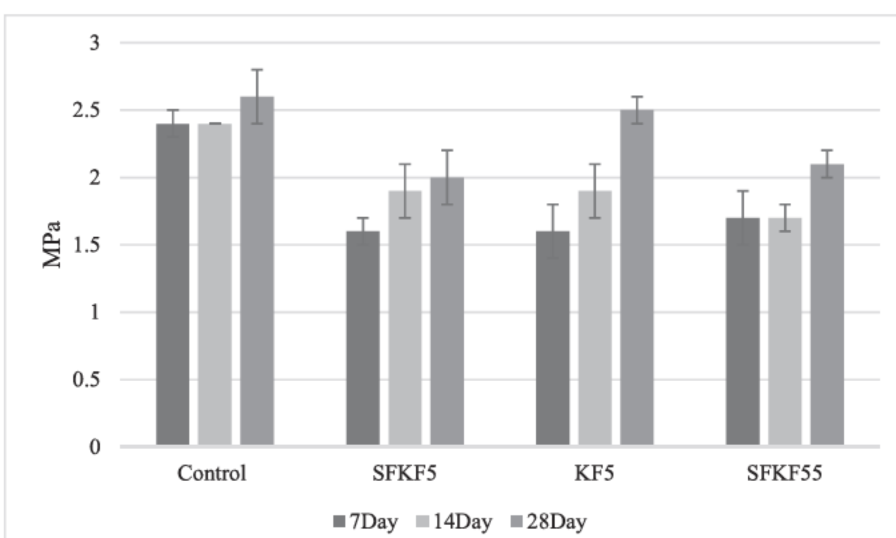


Fig. 9. Variation of flexural strength.

formation of crack bridging to uphold higher strength levels. This is shown with the increase of strength at each interval measurement.

4. Conclusions and future research

This experimental study was conducted to evaluate the effect of waste cardboard with fibre and matrix modifications on the compressive, tensile, and flexural strength properties. The application of SF lowered the alkaline zone around the fibre to prevent attachment of hydrated cement products and mineralisation on KFs. The mechanical analysis of concrete samples containing raw KFs, modified KFs and matrix modified specimens demonstrated that waste cardboard fibres can be adequately integrated within concrete and mortar materials. The experimental procedure remained consistent when applying the different materials. However, the mechanical results demonstrated strength variations per mix design which is graphically represented via error bars. This variation is due to the batching process. The final agglomeration of all materials remained consistent, however; fibre clumping, and fibre dispersion were two factors that were irregular to maintain during the batching process. KFs are a non-homogeneous material within the cementitious matrix and can be inconsistent when compared to concrete and mortar materials. However, the workability of concrete was not compromised when fibre integration occurred, and the matrix bonding of samples was successful. Future research can be directed toward varying the percentage of KFs and the matrix modifier. Further investigations are required on the durability characteristics of the KF composites. The most important outcomes of this research are summarised as follows.

- SFKF composites had a 20% compressive strength reduction at the 28-day measurement, whereas KF composites withheld a 48% reduction. This demonstrates fibre modification using SF can enhance fibre strength within a cementitious matrix.
- Waste cardboard can be utilised further in concrete and mortar materials when being applied to lower concrete strength requirements.
- The integration of waste KFs can reduce landfill size and reserve virgin resource extraction.
- The investigation of the microstructure of the composites illustrated areas of fibre containing no SF or hydration products. This can increase the porosity of the composite leading to a decline in durability characteristics.
- Raw KFs contain a better modulus of elasticity and are more flexible than modified KF. This shows SF can increase the brittleness of the fibre when under flexural strain.
- This experimental study contained dry fibrous KFs. Further investigations are required of KF pulp.
- 5% KF integration exhibited satisfactory strength results for non-structural strength concrete.

CRediT authorship contribution statement

Robert Haigh: Conceptualization, Methodology, Investigation, Writing – original draft, Writing – review & editing. **Yanni Bouras:** Supervision, Review & editing. **Malindu Sandanayake:** Supervision, Review & editing. **Zora Vrcelj:** Project administration, Supervision, Review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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3.3 Concluding remarks

This chapter presented the mechanical performance of 5% KF integration within concrete composites. There were three mix designs experimentally investigated to demonstrate the individual results of raw fibres, fibre pre-treatment and matrix modification within concrete materials. The chapter provided an insight into the composite microstructure along with an analysis of the fibres morphology after silica fume pre-treatment. As a result of the various mechanical strength results, it was determined whether waste cardboard could be used as an alternative to concrete and mortar.

In summary, the following conclusions are drawn from this chapter:

- Waste cardboard can be utilised in concrete and mortar materials when being applied to lower concrete strength requirements.
- By integrating waste KFs, landfill sizes can be reduced and virgin resources can be preserved
- The investigation of the microstructure of the composites illustrated areas of fibre containing no SF or hydration products. This can increase the porosity of the composite leading to a decline in durability characteristics.
- Raw KFs contain a higher modulus of elasticity and are more flexible than modified KF. This shows SF can increase the brittleness of the fibre when under flexural strain.
- Matrix and fibre modified composites have a higher tensile strength when compared with the control.

Chapter 4 – Mechanical analysis of high-volume fibre integration

4.1 Introduction

In this chapter, 15-20% waste cardboard KFs were integrated within concrete composites. Four mix designs were examined to ascertain the strength characteristics of the concrete when a high-volume of cement is substituted. This involved the use of 15-20% SF modified KFs and 15-20% raw KFs. The variation of fibre percentage demonstrated the influence on the compressive strength characteristics. Moreover, this chapter identifies the associated effects when using a large amount of modified and raw waste KFs in concrete composites.

The following paper is included in this chapter:

Robert Haigh 2022. A comparative analysis of the mechanical properties with high volume waste cardboard fibres within concrete composite materials, *Australian structural engineering conference.*

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In the case of the above publication, the following authors contributed to the work as follows:

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Name(s) of Co-Author(s)	**Contribution (%)**	**Nature of Contribution**	**Signature**	**Date**
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4.3 Concluding remarks

This chapter demonstrated a mechanical investigation of 15 and 20% fibre integration when partially substituting cement. Raw and modified KFs derived from waste cardboard were utilised to experimentally investigate the compressive strength characteristics. The results highlight that there is a strength increase when SF is applied as a coating. Using SF as a fibre modification technique has demonstrated sufficient adhesion to the fibres and increased the promotion to be used in further applications.

The main findings of the research are shown below.

- A 25% increase of fibre content does not hinder the compressive strength when SF is applied.
- SF reduces the formation of $Ca(OH)_2$ increasing fibre durability.
- Embrittlement of raw KFs creates a weakening in composite designs.
- Waste cardboard can be utilised as a filler within cementitious composites.
- KFs can be used within non-structural concrete applications.

Chapter 5 – Thermal analysis

5.1 Introduction

In this chapter, the thermal components of KFs were investigated. To understand the morphology of the fibres, the experimental program included the use of a Fourier transform infrared spectroscopy (FT-IR) and a scanning electron microscope (SEM) with an energy dispersion x-ray (EDS) provision. The thermal, calorimetric and combustion attributes were determined using a thermo-gravimetric analysis (TGA), differential scanning calorimetry (DSC) and pyrolysis combustion flow calorimetry (PCFC). This chapter also contains a strength analysis, when integrating 10% kraft fibres within concrete composites. The mechanical results of the composite materials are measured via compressive, tensile, and flexural experiments.

The following paper is included in this chapter:

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In the case of the above publication, the following authors contributed to the work as follows:

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Paul Joseph	5	Supervision, review and editing	[redacted]	8/3/2023
Malindu Sandanayake	5	Supervision, review and editing	[redacted]	8/2/2023
Yanni Bouras	5	Supervision, review and editing	[redacted]	8/2/2023
Zora Vrcelj	5	Supervision, review and editing	[redacted]	7/31/2023

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materials

Article

Thermal Characterizations of Waste Cardboard Kraft Fibres in the Context of Their Use as a Partial Cement Substitute within Concrete Composites

Robert Haigh, Paul Joseph, Malindu Sandanayake *, Yanni Bouras and Zora Vrcelj

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Abstract: The building and construction industry consumes a significant amount of virgin resources and minimizing the demand with alternative waste materials can provide a contemporary solution. In this study, thermal components of kraft fibres (KFs) derived from waste cardboard are investigated. The mechanical properties containing KFs within concrete composites are evaluated. Metakaolin (MK) and KFs were integrated into concrete samples as a partial substitute for cement. Silica Fume (SF) was applied to the KF (SFKFs) with a view to enhancing the fibre durability. The results indicated that there was a reduction in compressive strength of 44 and 56% when 10% raw and modified KFs were integrated, respectively. Raw, fibre and matrix-modified samples demonstrated a 35, 4 and 24% flexural strength reduction, respectively; however, the tensile strength improved by 8% when the matrix was modified using MK and SFKF. The morphology of the fibres was illustrated using a scanning electron microscope (SEM), with an energy dispersion X-ray spectroscopy (EDS) provision and Fourier transform infrared spectroscopy (FT-IR) employed to gain insights into their chemical nature. The thermal, calorimetric and combustion attributes of the fibres were measured using thermogravimetric analysis (TGA), differential scanning calorimetry (DSC) and pyrolysis combustion flow calorimetry (PCFC). SFKFs showed a lower heat release capacity (HRC), demonstrating a lower combustion propensity compared to raw KFs. Furthermore, the 45% decreased peak heat release rate (pHRR) of SFKFs highlighted the overall reduction in the fire hazards associated with these materials. TGA results also confirmed a lower mass weight loss of SFKFs at elevated temperatures, thus corroborating the results from the PCFC runs.

Keywords: kraft fibres; cement composites; waste cardboard; silica fume; metakaolin; thermophysical properties

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1. Introduction

In recent times, researchers have been highly focused on investigating novel sustainable construction materials due to the negative environmental impacts caused by excessive virgin material consumption [1–3]. Global population growth has increased the demand for housing and infrastructure [4,5]. There has been an increased demand for concrete, with worldwide production of cement increasing by more than 24% in the last decade, resulting in approximately 5% of annual global greenhouse gas emissions (GHG) [5,6]. Significant research has been conducted to reduce the volume of cement in concrete and mortar materials due to the high embodied carbon and energy in cement [7]. Several researchers have investigated using supplementary cementitious waste materials as a partial cement substitute within concrete. This has been in order to counter both issues of reducing virgin materials used in cement production and addressing excessive waste generation [8–12].

Due to the abundance of residential waste, these materials are becoming increasingly researched as a potential source of building and construction material. In recent years, there has been a strong interest towards the integration of waste cardboard within cement and mortar materials [13–15]. To effectively use cardboard within a cementitious composite, cardboard must be reduced to either a pulp or fibrous material [16]. The main constituent fibres that give cardboard its strength are called kraft fibres (KFs). KFs are natural cellulose fibres derived from plants and trees [17]. These fibres have advantageous properties such as ease of availability, biodegradability, low density and non-abrasiveness [18]. However, cement has a high alkaline value, which can degrade KFs significantly. Researchers have identified that supplementary cementitious materials (SCMs) can lower the alkalinity and enhance fibre to matrix integration [19,20]. Generally, fibre and matrix modifications are required to enhance the mechanical stability of KFs within a cement environment [14]. This was shown by Booya et al. [21] using silica fume (SF), ground blast furnace slag (GBFS) and metakaolin (MK) to improve the strength and permeability characteristics of fibre-reinforced concrete. Mohr et al. [19] integrated FA, GBFS, MK, SF and calcined diatomaceous earth and volcanic ash (DEVA) within fibre cement boards. Their results demonstrated that binary and ternary blends of SCMs improved the durability of the fibre composites; however, this improvement can be attributed to the significant reduction of calcium hydroxide ($Ca(OH)_2$) and the stabilization of the alkali content. Research integrating KFs within cement-based composites has predominantly focused on the mechanical properties [14], with further research required to understand the thermal characteristics of raw KFs and surface-modified KFs before integration within cementitious composites.

An important factor of natural-fibre-reinforced cementitious composites is the materials' level of decomposition that can occur with increased temperature. Previous research [22] has reported on the thermal properties of natural fibres (NFs) within polar and non-polar composite designs. However, reports on the thermal properties of KFs used in cementitious composite designs are very limited. During the kraft-making process, fibre constituents such as lignin and hemicellulose are removed from the fibre walls [23]. Mild pyrolysis has been shown to mainly effect these elements of NFs, creating a pseudo-lignin that increases the hydrophobic properties [24]. When NF cementitious composites decompose due to increased temperatures, chemical and physical changes can occur. These changes include hydrolysis, oxidation, decarboxylation, depolymerization, debonding and dehydration [25]. Owing to the removal of those fibre elements, further investigations are required of the thermal reactivity when KFs are integrated within a cementitious matrix. The determination of thermal properties can aid towards an appropriate application of KF composites for further use in the building and construction industry. In the present study, in order to accommodate KFs within the cementitious matrix, MK was integrated as the SCM. Dehydroxylation of kaolinite and disordered kaolinite can produce metakaolin and metadiskaolin, respectively. Both types of kaolin can be supplemented for partial cement substitution [26]. These variants of kaolinite generally contain a high percentage of alumina (Al_2O_3), which has been shown to enhance the hydration of cement in the early stages [27]. This factor also increases the durability of NFs because of the rapid consumption of $Ca(OH)_2$, which in turn reduces the alkali attack on fibre walls [19,21,28,29]. When the durability of the fibre is enhanced, the mechanical properties are less hindered by the degradation that can occur due to the high alkaline percentage. Silica fume (SF) was applied as the fibre surface modification to the raw KFs. The integration of SF can increase the mechanical and durability performance of cement composites [13,15]. It can also enhance the formation of calcium silicate hydrate (C-S-H) within the cement matrix, thus increasing the composites' mechanical strength. SF generally contains a high percentage of silicon dioxide (SiO_2), which can consume $Ca(OH)_2$ at later stages of hydration. These factors can also increase the bond between the fibre and the matrix.

This paper aims to evaluate the thermal performance of cardboard, cardboard pulp, and raw and SF surface-modified KFs. The mechanical strength properties of the kraft fibre concrete composite materials are also presented with a view to illustrating and exemplifying the physical attributes of the fibrous material. The main objective of the present study is to demonstrate the successful integration of cardboard waste material as a partial cement substitute within concrete composite materials for further applications within the building and construction industry.

2. Experimental Procedure

2.1. Raw Materials

The main constituent materials used to reduce the consumption of OPC within the mix design of the concrete specimens were waste corrugated cardboard and MK. Figure 1 graphically depicts the sequential process used to reduce waste cardboard into a fibrous material that is viable for concrete integration. As illustrated, the waste cardboard was initially converted into a pulp material and then a specific type of fibre design was combined with SF to create Silica Fume Kraft Fibre (SFKF). The KFs contained an approximate 20 wt. % loading of SF. In the second type of fibre design, only the raw material without any SF attachment (KF) was employed. Moisture was then removed from both fibre designs through drying via a conventional oven at 20 °C for 8 h. The fibres were then subjected to rotation via a blender mixer. The final fibrous material was the result of this eight-step process, and can be subsequently integrated within the concrete mix design. Metakaolin was used within the composites matrix, conforming with the ATSM C-618 [30], i.e., Class N specifications for natural and calcined pozzolans. Silica fume conformed with the Australian Standard AS/NZS 3582.3 [31] specification of Silica Fume used in cementitious mixtures. Ordinary Portland Cement (OPC) was used as the primary constituent for the pozzolanic reactivity and conformed with AS/NZS 3972 [32]. Locally available coarse and fine aggregate were applied, conforming to AS/NZS 1141.6.2 [33] and AS/NZS 1141.5 [34], respectively. The composition of the pozzolanic materials is shown in Table 1.

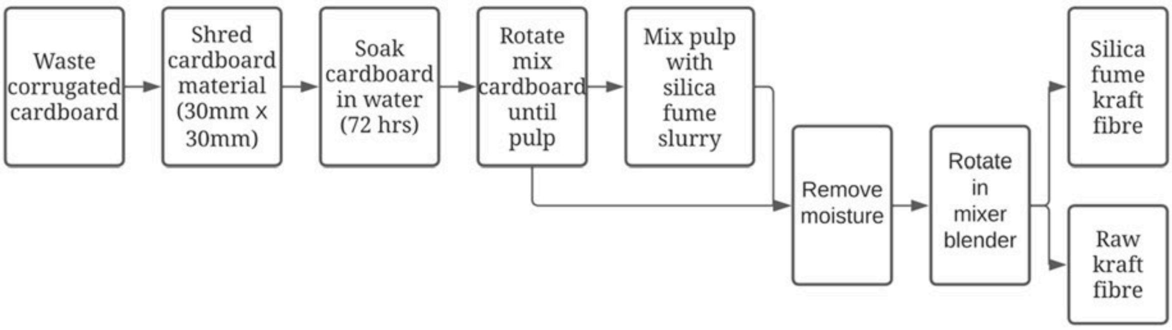

Figure 1. Fibre preparation methodology.

Table 1. Composition of materials adopted from [15].

Chemical	Material Component wt. %		
	MK	SF	OPC
SiO_2	54–56	≥75–<100	19–23
Al_2O_3	40–42		2.5–6
Fe_2O_3	<1.4		
TiO_2	<3.0		
SO_4	<0.05		
P_2O_5	<0.2		
CaO	<0.1		61–67
MgO	<0.1		
Na_2O	<0.05		
K_2O	<0.4		
Amorphous silica, fumes		≥0.3–<1	
$CaSO_4{\cdot}2H_2O$			3–8
$CaCO_3$			0–7.5
Fe_2O_3			0–6
SO_3			1.5–4.5

2.2. Specimen Preparation

A mortar mixer was used to prepare concrete specimens. The raw materials were dry mixed for 5 min before adding water. After adding water, the materials were mixed for an additional 5 min. Upon completion of the mixing process, concrete was poured into various moulds in three separate layers with twenty compactions for each layer using a steel rod to maintain uniform compaction. The specimens were prepared at a room temperature of 20 °C for 24 h, before curing was applied in water baths for 7, 14 and 28 days.

2.3. Mix Design

Raw KFs and SFKFs were thermally analysed to evaluate their degradation characteristics. The mechanical properties of concrete when reducing OPC with SFKFs, raw KFs and MK were analysed. Three bespoke mix designs were analysed in this study, with the mix code correlating to the constituent materials that were used in the concrete. For example, the SFKF105 sample is the integration of 10% SFKFs and 5% MK, whereas, correspondingly, SFKF10 is 10% SF modified fibres and KF10 is 10% raw KFs. The total reduction of OPC within each mix design is shown in Table 2. Water, fine and coarse aggregates remained the same as the control with a mass ratio of 0.33, 1.15 and 1.73 per kilogram of concrete, respectively. The workability of the materials remained consistent during the batching process regardless of fibre integration.

Table 2. Composite constituent materials.

Mix Code	MK	KF	SFKF	OPC
Control	-	-	-	100
SFKF10	-	-	10	90
KF10	-	10	-	90
SFKF105	5	-	10	85

2.4. Testing Procedure

2.4.1. Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) is generally performed in order to gain insights into the thermal and thermo-oxidative behaviour of solid materials. In the present study, the TGA runs were conducted on samples (on powdered materials accounting to ca. 10–15 mg) of waste cardboard (CB), waste cardboard dried pulp (CBDP), raw KFs and SFKFs. Generally, TGA provides data relating to the material's decomposition and can demonstrate the degradation kinetics and char formation propensities. A Mettler Toledo instrument was used in this study, in accordance with ASTM E1131 [35]. The tests were conducted on powdered samples (ca. 10–15 mg) in a nitrogen atmosphere and at two different heating rates, 10 °C min^{-1} and 60 °C min^{-1}, and from 30 to 900 °C, with a gas flow rate of 50 mL min^{-1}. The second heating rate of 60 °C min^{-1} was chosen with a view to directly comparing the results with those from the pyrolysis combustion flow calorimetry (PCFC) experiments.

2.4.2. Differential Scanning Calorimetry (DSC)

The differential scanning calorimetry (DSC) analyses of the samples were performed with a view to identifying phase changes and measuring the energetics of the pyrolytic reactions. For this purpose, DSC tests were conducted on some of the pristine specimens, and fibrous component (KFs) and its admixture with silica fumes (SFKFs). For these measurements, a Mettler Toledo instrument was employed, with the tests conducted in an atmosphere of nitrogen and at a heating rate of 10 °C min^{-1} (from 30 to 550 °C). Accurately weighed samples (ca. 15 mg) were initially taken into aluminium crucibles, with these crucibles subsequently sealed with aluminium lids that contained pinholes, thus facilitating the escape of any gaseous products that were formed, especially at elevated temperatures.

2.4.3. Fourier Transform Infrared Spectroscopy (FT-IR)

Fourier transform infrared spectroscopy (FT-IR) was used to obtain information relating to the chemical nature of the test samples. A PerkinElmer 1600 Series spectrometer was used in this study in accordance with ASTM E1252 [36]. The test specimens were irradiated with infrared rays in the range from 4000 to 600 cm^{-1} in the attenuated total reflectance (ATR) mode. In each case, a few milligrams of the test sample were used as neat and it was mounted onto the diamond crystal stage. A plot of absorbance versus wavenumber was generated for each of the samples, after appropriate baseline correction for each run (number of scans: 16; resolution: 4 cm^{-1}), which aided in identifying the various functional groups present within them.

2.4.4. Pyrolysis Combustion Flow Calorimetry (PCFC)

This technique, also known as "microscale combustion calorimetry" (MCC), is a small-scale calorimetric testing method used to analyse the fire behaviour of various solid materials when subjected to a forced non-flaming combustion, under anaerobic or aerobic conditions. This is conducted in accordance with ASTM D7309 [37]. The seminal work behind this technique was performed at the Federal Aviation Administration in the USA in the late 1990s. This method is assumed to reproduce and decipher the condensed and gaseous parts of flaming combustion in a non-flaming test regime, through a rapid and controlled pyrolysis of the sample in an inert atmosphere (i.e., in nitrogen) followed by high-temperature oxidation (i.e., combustion) of the pyrolyzate components in the presence of oxygen [38].

The main advantage of using PCFC is that only a very small quantity (mg) of a sample is required, and the test method often provides information regarding useful combustion parameters, such as peak heat release rate (pHHR), temperature to pHHR, total heat released (THR), heat release capacity (HRC), effective heat of combustion (EHC) and percentage of char yield. Although PCFC can provide some of the useful data regarding heat-related parameters in the smaller scale, correlation of the available data with real fire scenarios is still limited. In the present work, PCFC runs were conducted in some chosen substrates (mainly step-growth polymers) at 1 °C min^{-1}, applying an FTT microscale calorimeter using method A, i.e., in an atmosphere of nitrogen.

2.4.5. Mechanical Testing

Compressive strength was determined using concrete cylinders of 100 mm diameter and 200 mm length. The load rating applied to the specimens was 20 MPa/min in accordance with AS 1012.9 [39]. The flexural strength was determined using a four-point bending test. The size of the specimens used in this experiment were 100 mm × 100 mm × 350 mm. The load rate applied to the concrete beams was 1 MPa/min in accordance with AS 1012.11 [40]. The tensile strength of the samples was determined via the indirect tensile testing method. The size of the cylindrical specimens used in this experiment were 100 mm × 200 mm. The load rating applied to the concrete cylinders was 1.5 MPa/min in accordance with AS 1012.10 [41]. The mechanical testing equipment used was the Matest C088-11N Servo-Plus evolution testing machine and Cyber-Plus evolution data acquisition system. The tensile, compressive and flexural results were measured at 7-, 14- and 28-day intervals. The average was determined from three specimen samples.

2.4.6. Scanning Electron Microscopy Analysis

A scanning electron microscopy (SEM) with an energy dispersion provision (EDS) was used to analyse the morphological features of the samples. This was conducted on the Phenom XL G2 Desktop SEM operating at 10 kV via 1000 magnitude. Samples were reduced to 2–5 mm in diameter using a diamond cutting saw. The EDS report illustrated the chemical composition of the samples provided.

3. Results and Discussion

3.1. Chemical Characteristics of the Base Materials (FT-IR)

As expected, the test materials (CB, CBDP, KFs and SFKFs) exhibited specific absorptions (i.e., broad signal centred around 3250 cm^{-1} owing to -O-H stretching and -C-H stretching occurring in the region of 2900 cm^{-1}), and generated a typical fingerprint region of a ligno-cellulosic material, in their respective spectrum (a representative spectrum from CBDP is shown in Figure 2). These are measured at approximately 725 cm^{-1}, 1450 cm^{-1}, 2800 cm^{-1} and 2900 cm^{-1}. The peaks at 725 cm^{-1} are correspondent to impurities such as residual waxes and lignin.

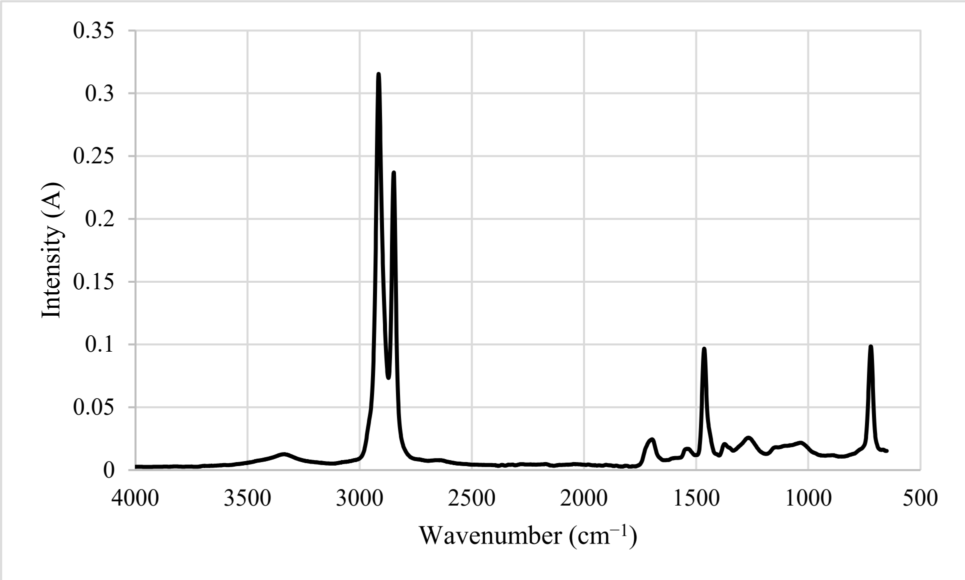

Figure 2. FT-IR spectrum of the dry pulp from cardboard (CBDP), where wavelength (in wave numbers: cm^{-1}) are plotted against signal intensity (in arbitrary units).

3.2. Thermal and Calorimetric Properties (TGA and DSC)

The TGA runs were conducted in an inert atmosphere of nitrogen and at two heating rates, 10 and 60 °C min^{-1}, the latter with a view to replicating the heating rate that was employed for the PCFC runs (i.e., 1 °C s^{-1}). It is relevant to note here that the general profiles of the thermograms for test samples were similar regardless of the heating rate and consisted of the following distinctive steps: 1. Initial loss of water (i.e., mainly desorption of the physically bound water) up to 180 °C; 2. Main chain degradation, including dehydration reactions up until about 400 °C; 3. Secondary degradation of the carbonaceous residue until the end of the run (900 °C). This is also in accordance with researchers stating that there are three main stages of weight loss with natural fibres [23,27]. Firstly, a slight weight loss is exhibited between 50–110 °C due to the evaporation of moisture in the fibre. Secondly, the decomposition of hemicellulose and primary component of lignin at temperatures between 270–360 °C. Finally, maximum weight loss occurs when the cellulose component is degraded heavily between 350–500 °C [27]. Prins et al. [23] also demonstrated that the decomposition of wood fibre occurs between 225–310 °C for hemicellulose, residual lignin between 250–500 °C and cellulose between 305–375 °C. During the manufacturing of KFs for production of cardboard materials, residual components such as lignin are mostly removed via chemical and thermal treatment. It is interesting to note that the mass loss exhibited occurred above 180°C, whereas Urrea-Ceferino et al. [42] conducted TGA experiments on unbleached northern softwood KFs and noted an abrupt mass loss between 105–150 °C. This highlights prior thermal modification during the manufacturing of KFs. The individual mass losses and corresponding temperatures are provided in Table 3, while a typical thermogram (CB at 10 °C min^{-1}) is shown in Figure 3.

Table 3. Mass losses and the amount of residue obtained during the TGA tests.

Sl. No.	Sample (Heating Rate: °C min^{-1})	Mass Loss at 180 °C (wt. %)	Mass Loss at 480 °C (wt. %)	Char Residue at 900 °C (wt. %)
1	CB (10)	7	94	2.4
2	CB (60)	5	77	16
3	CBDP (10)	6	83	6.0
4	CBDP (60)	4	76	16
5	KF (10)	3	72	11
6	KF (60)	4	72	20
7	SFKF (10)	4	59	31
8	SFKF (60)	3	49	44

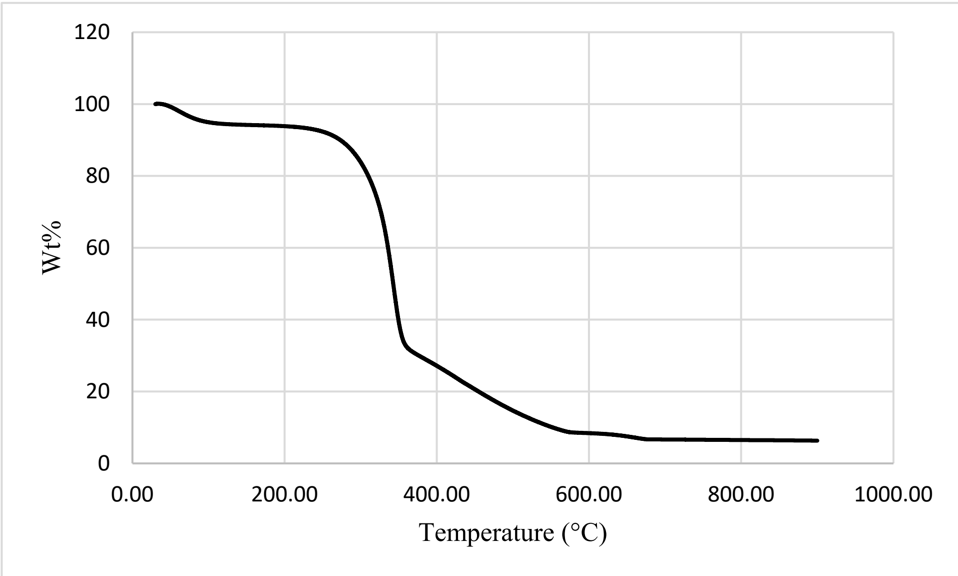

Figure 3. Thermogram obtained for CB sample at a heating rate of 10 °C min^{-1} in nitrogen.

As shown in Table 3, the extent of water retention among the various samples differed, albeit within a relatively narrower range (between 3 and 7 wt. %); however, the corresponding values for mass losses at 480°C varied more widely. It is also interesting to note that these values also seem to be influenced by the heating rates, and this aspect is also reflected in the amounts of char residues that were left after the completion of the runs. Given the presence of the inorganic filler (SF~20 wt. %) in SFKFs, the maximum amount of char residue was exhibited at 900 °C, while the minimum extent of mass losses was recorded at 480 °C. Tonoli et al. [43] conducted an application of acid hydrolysis to whisker fibres and noted a 40% weight loss with temperatures between 150–310 °C. Further degradation was shown to occur within a temperature range of 350–525 °C; however, above 550 °C, no thermal event occurred. This demonstrates that the application of SF modification on the KF walls delays degradation at high temperatures.

As expected, the DSC curves of the samples (for example, SFKFs) showed at least two distinctive peaks which mirrored their corresponding TGA thermograms. Evidently, these represent endothermic peaks that correspond to the loss of physically bound water (between 30 and 180 °C) and main chain pyrolysis reactions (occurring between 250 and 400 °C). A broader endothermic peak beyond 400 °C (and until the end of the run: 550 °C) could be indicative of slow degradation reactions of the residue that was left after the main chain degradation (see Figure 4).

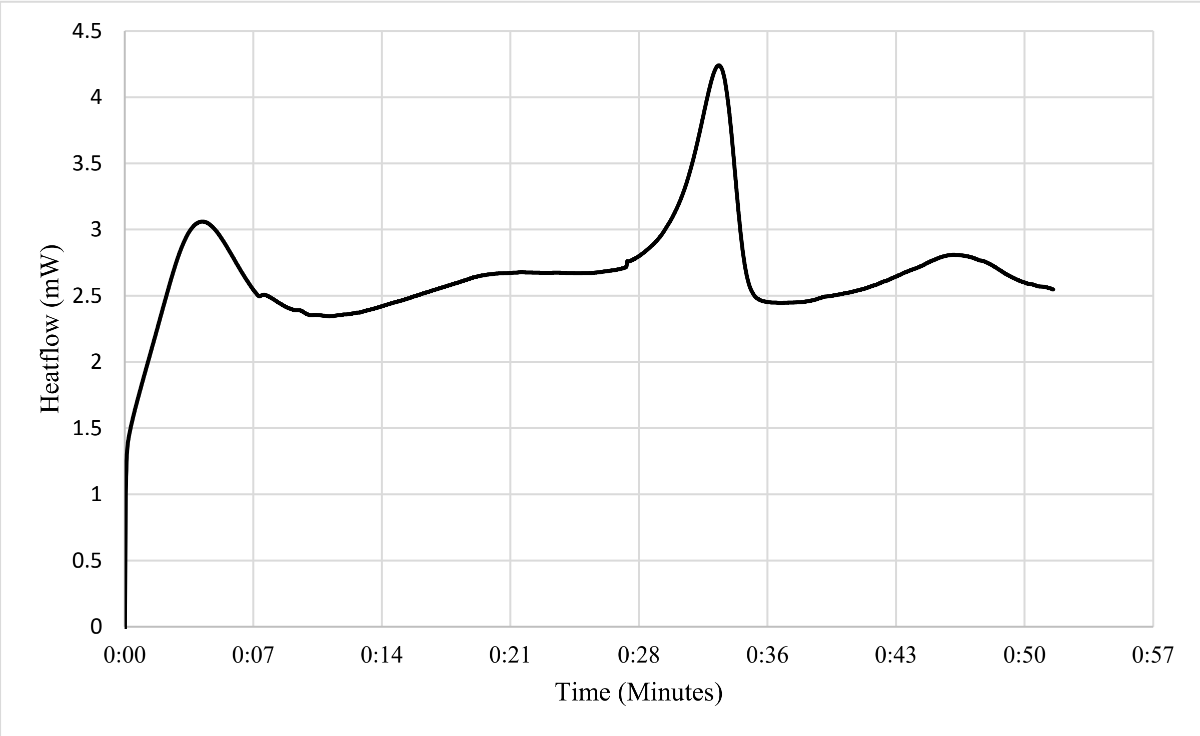

Figure 4. DSC curve of SFKFs showing three different endothermic processes.

3.3. Combustion Attributes (PCFC)

The relevant parameters obtained through PCFC runs are tabulated in Table 4. Generally, the heat release capacity (HRC) is considered as a reliable indicator of the combustibility of a material [38]. Evidently, the HRC value was found to be the lowest for the SFKF sample (so also the corresponding values for THR, pHRR, char yields and EHC). Therefore, it can be concluded that the presence of the inorganic filler, SF, substantially aided in reducing the overall flammability of the fibrous matrix. This factor of fibre modification is critical for the durability of fibre cement composites when exposed to increased or sustained elevated temperatures. Figure 5 illustrates the microstructure of the SFKFs, demonstrating the successful application of SF on the KF walls. The EDS plot further features the attachment of SiO_2 (SF) to the fibrous matrix. Onésippe et al. [24] noted that the inclusion of SF within cement composites can reduce the specific heat by 41%. Integrating SF on the fibre walls reduced the pHRR by more than 45% compared with other fibre types, even though the temperature level was slightly higher. Removing the lignin from the outer cell wall of the fibre, as well as the amorphous components of hemicellulose between the fibres, reduces the overall thermal stability of KFs compared to raw cellulose matter [44]. Increasing the fibre's thermal stability via surface modification can reduce the thermal decomposition rate as well as minimizing the HRC of combustion. The successful integration of pyrolyzed fibres within composite materials is dependent on the temperature applied to the fibres during pyrolysis. KFs are subjected to prior thermal treatment at approximate temperatures of 170 °C via caustic soda pulping. This process delignifies the fibres from the amorphous components, which can cause the morphology of the fibre walls to increase in surface roughness [45,46].

Table 4. The relevant parameters from the PCFC runs.

Sl. No.	Samples	Temp to pHRR (°C)	pHRR (W g^{-1})	THR (kJ g^{-1})	HRC (J g^{-1} K^{-1})	Char Yield (wt. %)	EHC (kJ g^{-1})
1	CB	365	133	9.57	132	17.3	11.6
2	CBDP	367	164	10.1	163	15.0	11.9
3	KF	368	139	9.30	139	22.1	12.4
4	SFKF	371	91.9	6.10	92	47.5	11.5

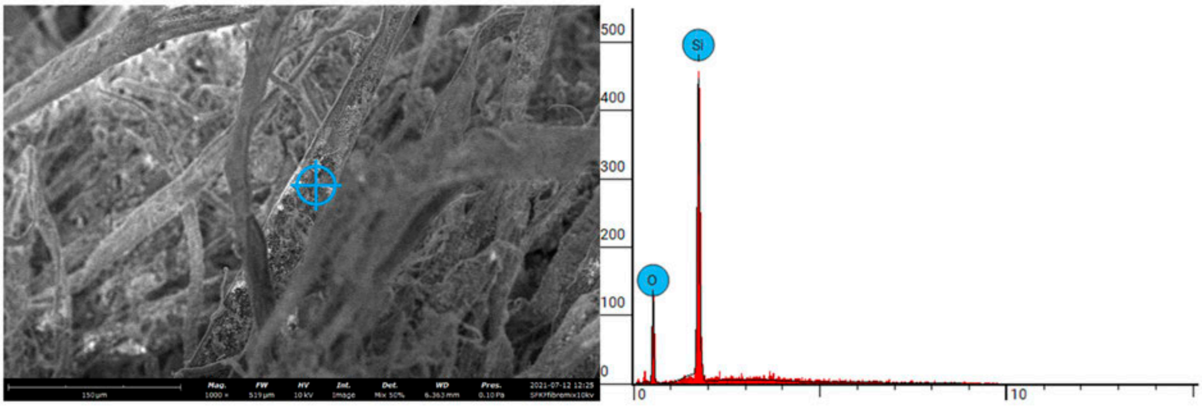

Figure 5. SEM/EDS report of SF on fibre.

3.4. Mechanical Properties

3.4.1. Compressive Strength

Figure 6 depicts the 7-, 14- and 28-day compressive strength. The standard deviation is shown via the error bars in Figure 6. The compressive strength is generally expected to decrease with fibre integration [47]. However, fibre composition within a modified cementitious matrix has been found to be a key factor in increasing the overall compressive strength of the KF composite. This is shown with SFKF105 having a compressive strength of 21 MPa at the 28-day interval. There was a 40% compressive strength increase after the 7- and 14-day intervals. A similar trend is shown with the control. At the 7- and 14-day intervals, the control had a compressive strength of 17 MPa, then a 47% strength increase to 25 MPa at the 28-day interval. Samples SFKF10 and KF10 had a moderate linear incline. Sample SFKF10 had 10, 13 and 14 MPa at the 7-, 14- and 28-day intervals, respectively, whereas KF10 had 8, 9 and 11 MPa at the 7-, 14- and 28-day intervals, respectively. Sample KF10 demonstrated that fibre modification is critical to increase the compressive strength of the cementitious composite. Furthermore, SFKF105 highlights the importance of modifying the matrix. Previously published reports [14,15,48] agree that the compressive strength decreases when fibre integration increases. This is mainly caused via voids and induced pockets within the composite formation. Due to this factor, fibre composites contain a reduced overall density. Therefore, increasing a composite's density can also increase the compressive strength. During the heat of hydration, there is an exothermic reaction within the specimen's matrix. This is specifically related to the reaction of water with pozzolanic materials within concrete, mainly cement [49]. This heat release entails the replacement of weak bonds with stronger ones. When the pozzolanic composition of the fibre cement composites is altered with MK, the exothermic reaction is altered, increasing the chemical bonding of the fibre within the matrix. Figure 6 illustrates the increased compressive strength when also modifying the constituent materials.

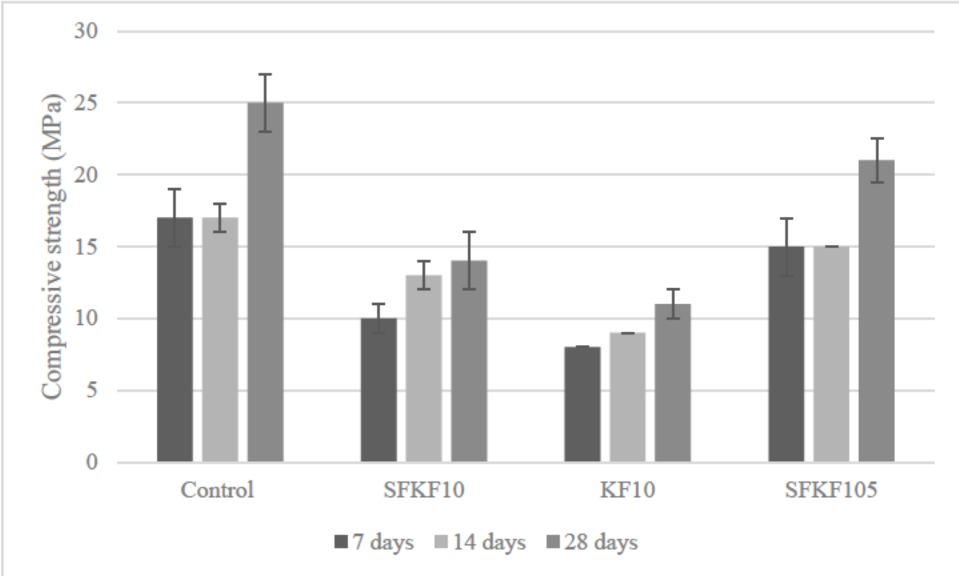

Figure 6. Variation of compressive strength.

3.4.2. Flexural Strength

Figure 7 depicts the 7-, 14- and 28-day flexural strength. The modulus of rupture (MOR) of the composite specimens is depicted with the maximum flexural strength. The standard deviation is shown via the error bars in Figure 7. The control exhibited the highest strengths of 2.4, 2.4 and 2.6 MPa at 7-, 14- and 28-day intervals, respectively. Samples SFKF10, KF10 and SFKF105 withhold the same flexural strength, 1.5 MPa, at the 7-day interval. However, SFKF105 then had a 17% and 25% increase at the 14- and 28-day intervals with 1.8 MPa and 2 MPa, respectively. The integration of MK has been shown to increase the elastic properties of composite materials [50]. This is primarily due to the improvement of uniformity in the matrix. Moreover, the variation of viscosities and alkaline silicates of different cations can also affect the geopolymerization process. This in turn can create variations of the microstructure and influence the mechanical characteristics. It is important to note that raw KFs contained a higher MOR than modified fibres at the 28-day interval. Increasing the fibre content of the composite can also increase the flexural strength due to the crack bridging effect [48,51]. However, previous research showed an increased MOR when only 5% fibre integration occurs. With 5% fibre integration, there was a 25% flexural strength increase of raw KFs compared to SFKFs [15]. In this study, there was a 20% flexural strength increase of raw KFs compared to SFKFs at the 28-day interval. This demonstrated that modifying the fibre with SF can increase the fibre's rigidity under flexural loading. Figure 8 illustrates SEM images of fibres within the concrete composite, with Figure 8A demonstrating fibre pull-out. This occurs when increased axial loading causes the fibre to pull out of the matrix, demonstrating a lack of mechanical and chemical bonding. The portion (see Figure 8B) shows the fibre wall unaffected by the cementitious matrix. Researchers [52] have shown that the primary fault of using natural fibres in cementitious composites is due to fibre deterioration. SF applied to the fibre walls appears to have reduced the deterioration. Figure 8C,D demonstrate the fibre embedded within the concrete composite. Furthermore, Figure 8D shows snapping of the fibre tip. This could be due to excessive loading previously applied to the composite. Generally, fibre composites can fail due to either fibre breakage or fibre pull-out when the load exceeds the composition's tensional, compressional or shear forces [53].

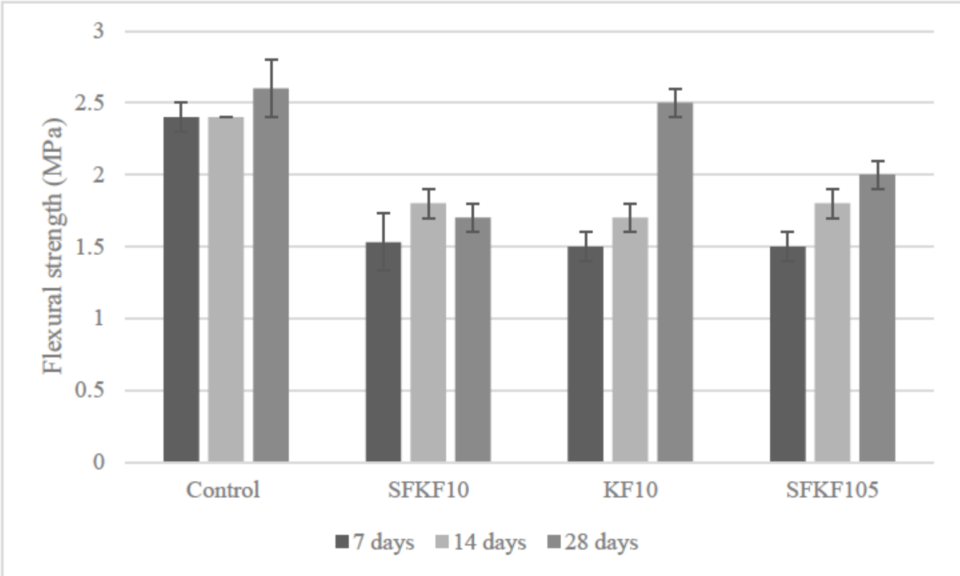

Figure 7. Variation of flexural strength.

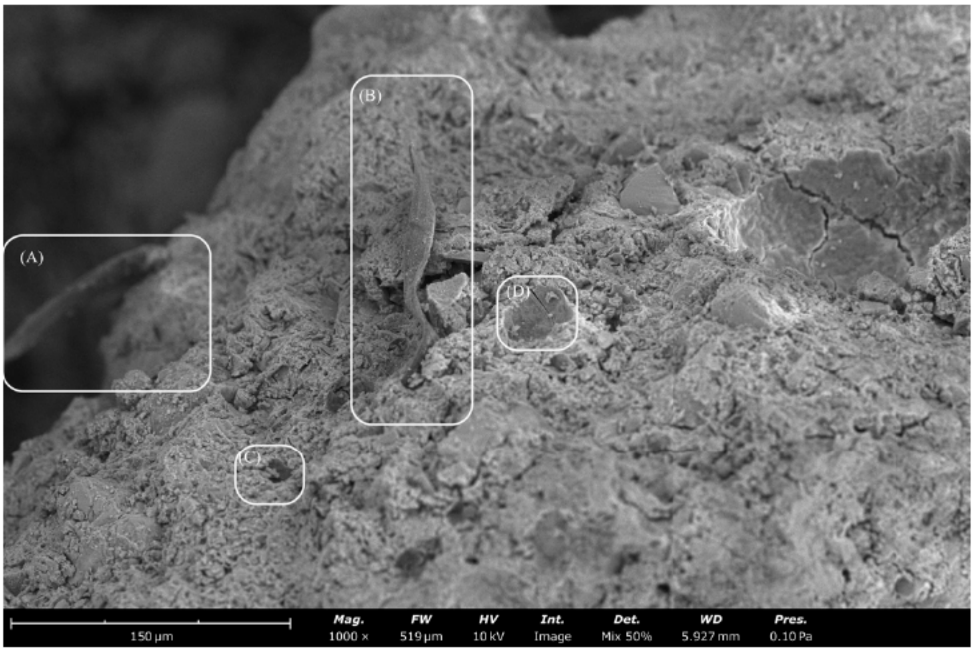

Figure 8. SEM images of fibres within concrete composite. (**A**,**B**) Retained fibre integrity, (**C**,**D**) Fibre snapping occurred.

3.4.3. Tensile Strength

Figure 9 depicts the 7-, 14- and 28-day tensile strength. The coefficient variation of the tensile strength was between 0–0.2 MPa. The standard deviation is shown via the error bars in Figure 9. The fibre composition within a modified cementitious matrix is a key factor towards the overall tensile-strength-bearing capacity. Sample SFKF105 withheld values of 2.1, 2.6 and 2.6 MPa at 7, 14 and 28-day intervals, respectively. It is important to note that the maximum strength at day 28 was higher than the control. This signifies that reducing OPC via matrix modification does not affect the tensile-bearing capacity. Sample KF10 withheld the lowest tensile strength of 1.1, 1.2 and 1.6 MPa at 7-, 14- and 28-day intervals, respectively, although it is important to note that there was a similar linear incline as seen with SFKF10. Sample SFKF10 had moderate tensile strength of 1.7, 1.8 and 2 MPa at 7-, 14- and 28-day intervals, respectively. However, SFKF10 had the highest strength at day 28, which was the weakest for both the control and SFKF105 at their 7-day interval. This further demonstrates that although fibre modification can benefit the tensile strength of the composite, matrix modification is also required to enhance the composite's strength. A previously published work [22] agrees that cementitious-based materials are tension weak and can easily form micro-cracks on the surface. As discussed, modifying the pozzolanic materials within cementitious-based composites can positively affect the exothermic reaction of the material's microstructure. The replacement of weaker bonds with stronger bonds during this phase also enhances the correlation between the constituent materials of the composite. This is shown in Figure 9, with the increased tensile strength of the bespoke mix design.

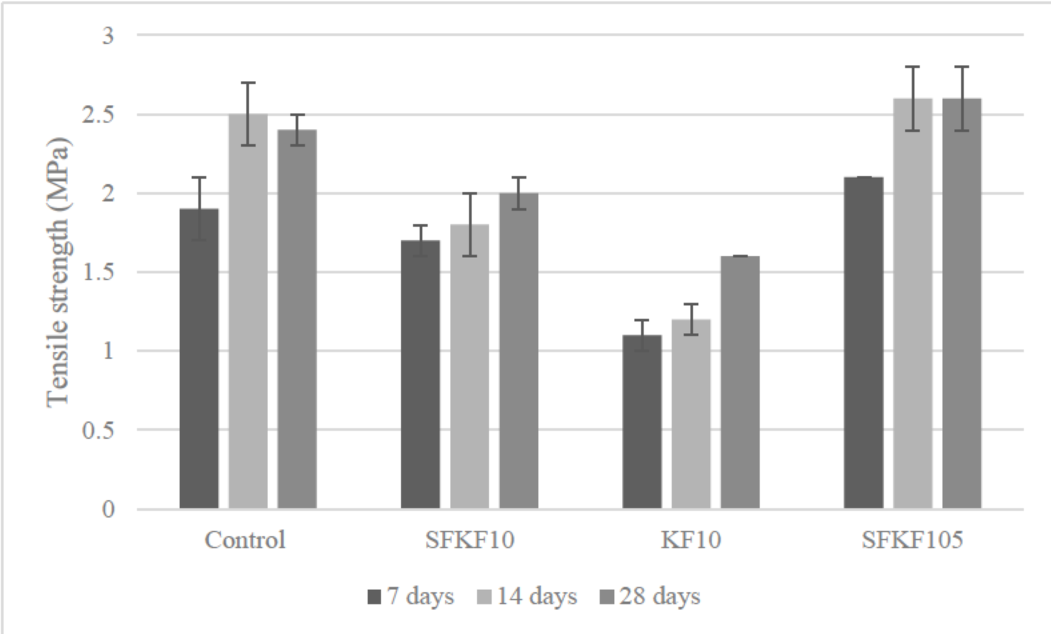

Figure 9. Variation of tensile strength.

4. Conclusions and Suggestions for Future Research

In this study, thermal and mechanical evaluations of fibres derived from cardboard waste within cementitious composites were conducted. Research highlighted the abundance of cardboard waste and a novel approach was defined to reduce potential accumulation in landfill areas, promoting a sustainable approach in construction. SF was implemented as the fibre surface modification technique and MK was used as the matrix modifier to enhance fibre durability. Cardboard waste was subjected to mechanical and thermal processing methods. These processes created KFs, ready for composite integration. Mechanical properties were determined via compressive, tensile and flexural testing methods. Morphology of the fibres was established using an SEM with an EDS provision. FT-IR was employed to illustrate the chemical nature of the materials. Thermal, calorimetric and combustion attributes of the fibres were measured using a TGA, DSC and PCFC, respectively. The SEM/EDS reports illustrated the successful application of SF onto the KF walls. This factor of surface modification using SF exhibited a reduction in the HRC when comparatively analysed with raw KFs, cardboard and cardboard pulp. This also gave SFKFs a relatively lower level of combustibility, which can ultimately lead to an improved durability of the fibres within cementitious composites when exposed to elevated temperatures/fires. The TGA results also favourably compared with the corresponding data obtained from the PCFC runs, especially in terms of the char residues obtained. DSC curves showed a broader endothermic peak beyond 400 °C, indicating a slow pyrolysis reaction following the main chain degradation. The FT-IR spectrum showed signals that are typical of ligno-cellulosic material with a character fingerprint region. The microstructure data revealed that the chemical processing applied to produce KFs does not affect the thermal composition of the cellulose matter. The integration of 5% MK within the cementitious matrix enhanced the compressive, tensile and flexural properties compared to non-modified matrixes. However, compressive and flexural strength was reduced when fibre integration occurred. Tensile strength properties were greater than the control when MK and SFKFs were integrated in the composite. This demonstrated the durability enhancement of surface modification using SF. Future research can be directed towards the fire performance of KF concrete and the reaction of isothermal conditions on the fibre durability. The major findings of this research are summarized as follows:

- Dried cardboard pulp exhibited a higher combustibility rate than its cardboard and fibrous counterpart;
- Inorganic materials such as SF can reduce the overall flammability of natural fibrous materials;
- A 10% KF integration exhibited satisfactory strength results for non-structural concrete;
- Waste cardboard can be utilized within cementitious composites;
- Matrix modification using MK enhances the mechanical strength of fibre-reinforced concrete;
- Using KFs derived from waste cardboard can reduce virgin resources requirement and divert waste in landfill management systems.

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5.3 Concluding remarks

This chapter presented the thermal components of SF modified, and raw KFs. The SF modified fibres showed a lower heat release capacity (HRC), demonstrating a lower combustion propensity as compared with raw KFs. The decreased peak heat release rate (pHRR) of the modified fibres highlighted an overall reduction in the fire hazards associated with fibre composite materials. Previous chapters detailed varied fibre percentages however, this chapter demonstrated the integration of 10% KF within cementitious composites. The mechanical strength results highlighted the importance of modifying the fibre as well as the composite matrix to achieve satisfactory compressive strength results.

In summary, the following conclusions are drawn from this chapter:

- Dried cardboard pulp exhibited a higher combustibility rate than the cardboard and fibrous counterpart.
- Inorganic materials such as SF can reduce the overall flammability of natural fibrous materials.
- A 10% KF integration exhibited satisfactory strength results for non-structural concrete.
- Waste cardboard can be utilised within cementitious composites.
- Matrix modification using metakaolin enhances the mechanical strength of fibre reinforced concrete.

Chapter 6 – Durability analysis

6.1 Introduction

In this chapter, the durability of KF reinforced concrete was comprehensively analysed as a partial cement substitute. Durability testing was conducted, exploring the composites microstructure when integrating KFs derived from waste cardboard. Fibre and matrix modification techniques were utilised via SF and MK, respectively. Wet and dry cycling was applied to the composites as an accelerating ageing technique. Water absorption, immersion, carbonation, and chloride ion penetration testing was also conducted. Porosity analysis of the novel composites demonstrated connecting microstructural system, enhancing the knowledge of durable aspects of potential service life of the composites.

The following paper is included in this chapter:

Robert Haigh, Malindu Sandanayake, Yanni Bouras & Zora Vrcelj. 2022. Durability performance of waste cardboard kraft fibre reinforced concrete, *Journal of Building Engineering, 67*.

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The durability performance of waste cardboard kraft fibre reinforced concrete

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ABSTRACT

Despite the high recycling rate of cardboard, a significant amount of this material is disposed of in landfill areas. The conversion of cardboard waste into kraft fibres (KFs) offers potential sustainability benefits when used in the building and construction industry. This study comprehensively analyses the durability of KF reinforced concrete as a partial cement substitution. Durability testing was conducted, exploring the composites microstructure when integrating KFs derived from waste cardboard. Fibre and matrix modification techniques were utilised via silica fume (SF) and metakaolin (MK), respectively. Porosity analysis of the novel composites demonstrated connecting microstructural system, enhancing the knowledge of durable aspects of potential service life of the composites. SF reduced the water permeability of the novel composites as demonstrated via water absorption and immersion testing. At one year, there was a reduction of chloride ion transference for all fibre composites. Composites were aged via wet and dry cycles, then tested for their compressive and tensile strength at each interval. Compressive strength of all samples peaked at 60- 80 day-cycles. Fibre Matrix modified composites demonstrated a tensile strength of 2.5 MPa when subjected to 100 day-cycles, with the control having 2.7 MPa.

1. Introduction

The building and construction industry is heavy resource reliant, using high carbon emission creating materials across the sector. Globally, construction methods utilise mainly concrete, steel, timber, masonry, and gypsum that are manufactured from extracted raw materials [1]. Due to the compressive strength and durability of concrete, it is the most common material used across residential, commercial, and civil construction [2]. Concrete requires virgin materials including water, coarse aggregate, fine aggregate, and cement. However, the production of cement requires significant processing and energy requirements. Cement manufacturing requires the decomposition of limestone at high temperatures of 1500 °C [3]. The emissions from fossil fuel combustion and raw material usage results in 5–8% of all carbon dioxide (CO_2) emissions from the cement production industry worldwide [4]. Extensive research is currently focused on reducing the acceleration of climate change and minimising the carbon footprint of the building and construction industry [5,6]. Redirecting common waste materials toward construction use has been a contemporary solution. Researchers have focused on using waste plastic and glass within civil construction works of road and pavement sub layers [7,8]. Waste rubber has also been integrated with the prevention of land erosion applications [9]. However, waste paper, and cardboard materials are seldom considered as a building and construction raw material.

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Kraft fibres (KFs) are the main constituent of cardboard and can be recycled 5–7 times before the fibres are too brittle or short to be used again [10]. KFs originate from plants and trees, containing cellulose matter which give the natural fibres their strength characteristics [11]. KFs are considered a green alternative to their synthetic counterpart because of their renewability characteristics [12]. KFs also withhold other advantages such as biodegradability, low costs, high strength, low density, and non-hazardous properties [13,14]. However, synthetic fibres have been used extensively for their ability to negate degradation of high alkaline environments. This is shown within concrete materials, where polypropylene and carbon fibres are placed within pre-cast panels and pavement applications [15,16]. The hydrophilic components of KFs result with the absorption of moisture to the fibre walls [17]. The water absorption adjusts the volumetric size of the fibre and results with the weakening of fibre integration creating fibre degradation within concrete materials [18–20]. Raw KF concrete is a mechanically weak composite material, indicating limited potential applications [21]. Therefore, KFs are often considered difficult to use as a building and construction raw material. Researchers have demonstrated fibre modification techniques to enhance the durability of the fibres within the matrix [18,22]. Mohr et al. [23] applied sodium hydroxide (NaOH) to KFs, increasing the bond between the fibre and the matrix. *Dittenber and Gangarao* [24] dried out natural fibres before application to reduce the hydroxyl groups forming on the fibre walls. This was shown to enhance the strength of the composite design. Increasing the durability of KFs can enhance the service life of the composite as well as expand the potential market application. However, further studies are required to demonstrate the microstructural behaviour of fibre and matrix modified KF composites.

To combat the degradation of natural fibres within cementitious materials, researchers [25–27] have integrated the use of supplementary cementitious materials (SCMs) within composite materials. SCMs show reduced alkaline levels within concrete materials that can aid toward the reduction of degradation of natural fibres [27]. SCMs have been extensively researched due to the reduction of CO_2 and energy requirements when producing concrete [28–30]. Researchers have used SCMs such as fly ash (FA), ground blast furnace slag (GBFS), silica fume (SF) and metakaolin (MK) within cement composite designs. These materials have demonstrated effective mechanical results within natural fibre composites when using as a partial cement substitute [27,31–33]. To enhance the durability of natural fibres within cement composite, a reduced level of calcium hydroxide ($Ca(OH)_2$) is optimal. $Ca(OH)_2$ attacks the microfibrils of natural fibres, minimising the total service life of the composite design [34]. MK enhances the hydration of concrete in early stages because it contains a higher percentage of aluminate (Al_2O_3) than other SCMs [35]. This also reduces the formation of $Ca(OH)_2$ created due to the heating conversion of Al_2O_3 in the first few minutes [33]. Booya et al. [26] demonstrated 10% MK can increase the mechanical strength and composite durability when compared to other SCMs such as FA, GBFS, SF and pumice powder. The integration of SF with natural fibres has also shown to increase the durability and mechanical strength of composite designs [36]. Increased levels of calcium silicate hydrate (C–S–H) are produced when SF is integrated with concrete materials due to the reaction of $Ca(OH)_2$ crystals and calcium silicates during hydration [37]. *Sanchez and Ince* [38] have shown higher levels of C–S–H increase the compressive strength characteristics of cement composites. SF contains high levels of silicate dioxide (SiO2) which consumes $Ca(OH)_2$ at later stages of hydration [39]. This can also reduce the mineralisation occurring on fibre walls and enhance durability properties. The addition of fibre within composites can reduce the formation of micro-cracks and improve the crack resistance [40].

Fibre reinforced concrete can also demonstrate improved durability properties. The steel reinforcement within concrete can be a main source of corrosion [41]. This is due to the transference of chloride ions penetrating the concrete and reaching the steel. Once this occurs, steel will begin to rust and expand. Therefore, reduction in cracks can decrease the permeability of concrete structures. During the curing phase of concrete, microcracks occur due to the change of moisture content within the material. However, fibre integration can reduce the rate of moisture escaping the material and reduce the potential of cracks occurring. Fibre concrete also can reduce the size of cracks due to the crack bridging effect of the fibres. Fibres interlock within the matrix reducing the formation of micro cracks occurring. When there are cracks and voids present within concrete materials, the transference of chloride ions can assist the beginning of deleterious consequences. *Lee and Kim* [42] have shown the integration of 1% cement replacement with cellulose nanocrystal fibre can reduce chloride ion penetration and increase the durability of concrete. Drying shrinkage and water permeability have also been reduced with fibre integration [43]. Porous materials have a higher water absorption potential therefore, the minimisation of pore structure within concrete materials is critical to promote durability. Accelerated ageing with fibre integration in cement composites can also reduce the Ca(OH)2 content, lower porosity, increase precipitation of calcium carbonate (CaCO3) and increase fibre matrix bonding [34]. Researchers demonstrate accelerated ageing of composites via wet and dry cycles does not reduce the mechanical strength of 8% vegetable fibre cement composites [44]. This was primarily due to the addition of silica quartz used in conjunction with natural fibres, increasing the packing density of the matrix. Raw natural fibres in these studies have not undergone thermal and chemical transformations. Whereas waste cardboard fibres have been subjected to complex processing systems during the manufacturing stage.

Therefore, experimental research is required to assess the durability of waste cardboard fibres as a potential filler in concrete materials. To investigate the durability performance of KFs derived from waste cardboard within cementitious composites, this study has utilised three mix designs. Firstly, raw KFs specimens were developed. Secondly, SF was applied to the KFs as a fibre modification technique. Finally, MK was used as a partial cement substitute in conjunction with modified fibres. All fibre integration replaced 10% of cement requirements.

2. Experimental procedure

2.1. Raw materials and preparation

Waste corrugated cardboard and MK are the main constituent materials used to reduce the Original Portland cement (OPC) content required within the mix designs. Waste cardboard was reduced to a fibrous material with the integration of SF as a fibre modification technique. The process begins as waste cardboard was reduced to a pulp material via water immersion and rotating mixing. A SF slurry was then applied to the KFs to create Silica Fume Kraft Fibres (SFKFs). Raw KFs were also used in the mix design to determine the effect SF has as a modifying technique. Both fibres had the moisture removed and were subjected to a rotator mixer, producing a fibrous material. The SF applied to the waste cardboard (KFs) conforms with the Australian Standard AS/NZS 3582.3 [45] specification of silica fume used in cementitious materials. The MK used as a partial cement substitute conforms with ATSM C-618 [46] Class N specification for natural and calcined pozzolans. OPC was used in the mix design as the primary constituent for pozzolanic reactivity conforming with AS/NZS 3972 [47]. Locally available coarse and fine aggregates were integrated throughout the specimens conforming with AS/NZS 1141.6.2 [48] and AS/NZS 1141.5 [49], respectively. Regular potable tap water was used throughout the preparation of specimen designs. A mortar mixer was used to prepare the samples and conducted in accordance with AS/NZS 1012.2 [50]. Specimens were dry mixed for 5 min to allow sufficient agglomeration of all materials. Water was then added for an additional 5 min to complete sample preparation. Upon completion, the concrete materials were then added to the moulds in three layers. A steel rod was used to compress each layer twenty times before the additional layers were added to the mould. Moulds remained in a room temperature of approximately 20 °C for 24 h before being added to the curing baths. Slump tests were conducted in accordance with AS/NZS 1012.3.1 [51].

2.2. Mix design

The target compressive strength of all concrete specimens was 25 MPa. There was a total of four mix designs for the experimental durability analysis. Firstly, the control with no fibre integration secondly, 10% raw KF (KF10) thirdly, 10% SFKF (SF10) and finally, 10% SFKF and 5% MK (SFKF105). All bespoke materials were integrated as a partial cement substitution. Preliminary testing of the fibre composites determined SFKF105 to achieve comparable compressive strength to the control. Therefore, to understand the durability of 10% KF integration, each variable of KF must be assessed. The mix designs and composition of materials are formulated in Table 1 and Table 2, respectively. Water, fine and coarse aggregates remained the same as the control with a mass ratio of 0.33, 1.15 and 1.73 per kilogram of concrete, respectively. The water ratio remained the same for all specimens regardless of fibre integration. The workability of the fibre concrete remained acceptable during the mixing process.

2.3. Testing procedure

2.3.1. Mechanical testing

In accordance with methods of testing concrete AS/NZS 1012.8.2 [52], compressive and tensile testing was performed on 100 × 200 mm cylindrical specimens. The compressive load rating was 20 MPa/minute as determined by AS 1012.9 [53]. Splitting (indirect) tensile testing was conducted with a load rating of 1.5 MPa/minute in accordance with AS 1012.10 [54]. Three samples of each specimen were measured at various ages as discussed below, with the average recorded. The mechanical tests were conducted on the Matest C088-11 N Servo-Plus evolution testing machine and Cyber-Plus evolution data acquisition system.

2.3.2. Wet and dry cycling testing

Accelerated ageing was applied to the concrete composites to analyse the durability of KFs via wet and dry cycling. This method has been widely used to assess the degradation of natural fibres within cementitious matrices [34,37,55–58]. Accelerated ageing tests aim to speed the process of natural weathering via laboratory soaking and drying cycles. Specimens were submerged in 20 ± 5 °C potable water for 23 h, then subjected to 1 h of drying via a ventilated oven at 110 ± 5 °C. Each cycle is complete after one wet and dry cycle, within a 24-h period. Samples were subjected to 20, 40, 60, 80 and 100 cycles. Cycles began at the completion of the 28-day curing process. Upon completion of the listed cycles, specimens are subjected to compressive and tensile testing. This method determines the effect of accelerated ageing on the various mix designs.

2.3.3. Water immersion testing

Water immersion tests were conducted to analyse the porosity of the various mix designs. The experimental program was conducted in accordance with AS/NZS 1012.21 [59]. Test specimens were 100 × 200 mm cylindrical moulded concrete in accordance with AS/NZS 1012.8.2 [52]. After the curing time of 28, 56, 90 and 365-days, the cylindrical specimens were cut into four equal sizes using a diamond cutting saw blade. Once removed from the curing baths, specimens were weighed to the nearest 0.1 g and dried in a ventilated oven of 110 ± 5 °C for 24 h. Upon completion, the specimens were cooled in a desiccator to a room temperature of 23 ± 2 °C and weighed. All specimens were within 1 g of weight than another. The samples were then immersed in potable water with a 50 mm covering at 23 ± 2 °C for 48 h. Subsequently, the samples were removed from the immersion tanks and weighed within 60-s. To determine the immersed absorption rates of the samples, the following calculation was applied:

Water immersion calculation AS/NZS 1012.21 [59]:

Table 1
Cement of each mix design (Kg/m^3).

Mix design	Fibre	Cement	Metakaolin
Control	–	355.9	–
KF10	35.58	320.3	–
SFKF10	35.58	320.3	–
SFKF105	35.58	302.5	17.79

Table 2
Composition of materials adopted from [18].

Chemical	Material component wt%		
	MK	SF	OPC
SiO_2	54–56	$\geq$ 75- <100	19–23
Al_2O_3	40–42		2.5–6
Fe_2 O_3	<1.4		
TiO_2	<3.0		
SO_4	<0.05		
P_2O_5	<0.2		
CaO	<0.1		61–67
MgO	<0.1		
Na_2O	<0.05		
K_2O	<0.4		
Amorphous silica, fumes		$\geq$0.3- <1	
$CaSO_4.2H_2O$			3–8
$CaCO_3$			0–7.5
Fe_2O_3			0–6
SO_3			1.5–4.5

$$Ai = \frac{(M_{2i} \quad M_1)}{M_1} X\ 100\%$$

Where:

Ai = Immersed absorption.

M_1 = Mass of specimen in grams.

M_{2i} = Immersed specimen with surface moisture removed.

2.3.4. Water absorption testing

Water absorption testing was conducted to measure the initial surface absorption of the various mix designs. This testing was conducted in accordance with AS/NZS 4456.17 [60]. Test specimens were 100 × 200 mm cylindrical moulded concrete in accordance with AS/NZS 1012.8.2 [52]. After the curing time of 28, 56, 90 and 365-days, the cylindrical specimens were cut into four equal sizes using a diamond cutting saw blade. A total of twelve specimens for each mix design was measured. Once removed from the curing baths, specimens were weighed to the nearest 0.1 g and dried in a ventilated oven of 110 ± 5 °C for 24 h. Upon completion, the specimens were cooled in a desiccator to a room temperature of 23 ± 2 °C and weighed. All specimens were within 1 g of weight than another. Subsequently, specimens were placed upon stainless steel bars within the measuring tanks. The water did not exceed 3 ± 1 mm above the steel bars. The specimens remained in the water tanks for no longer than 60 ± 1 s and were immediately wiped with a damp cloth and the mass was determined to the nearest gram. To determine the absorption rates of the samples, the following calculation was applied:

Absorption calculation AS/NZS 4456.17:

$$IRA_{gross} = \frac{1000(m_2 \quad m_1)}{A_{gross}} \quad [60]$$

Where:

IRA= Initial rate of absorption, in kilograms per square metre per minute.

M_1 = Mass of specimen in grams.

M_2 = Mass of specimen after 1 min absorption, in grams.

A_{gross} = Gross area of the bed, based on work size dimensions, in square millimetres.

2.3.5. Chloride ion permeability testing

Chloride ion permeability testing was conducted to measure the intrusion of chloride ions entering the microstructure of the varied mix designs. Corrosion is predominantly caused by the intrusion of chloride ions entering the microstructure of concrete materials. This testing was conducted in accordance with ATSM C-1202-9 [61]. Test specimens were 100 × 200 mm cylindrical moulded concrete in accordance with AS/NZS 1012.8.2 [52]. After the curing time of 28, 56, 90 and 365-days, the cylindrical specimens were cut into four equal sizes using a diamond cutting saw blade to a sample size of 50 × 100 mm each. A total of four specimens were measured for each mix design at each varied age mentioned previously. Once removed from the curing baths, specimens were dried in a ventilated oven of 110 ± 5 °C for 24-h. Upon completion, an epoxy coating was applied to the outer surface area not subjected to the sodium hydroxide (NaOH) and sodium chloride (NaCl) solution. The coating was applied three times to resist possible solution penetration during experimental procedures. After coating, the samples were placed into a vacuum desiccator with distilled water covering 50 mm above the samples. The reduced pressure was applied to the samples of approximately 20 ± 5 inch of mercury (Hg) for 18-h. Upon completion, samples were wiped dry and placed into the mounting gaskets. Rubber seals were placed around the samples to prevent solution leakage. Analytical grade NaOH and NaCI solution was injected into each side of the testing chamber separately, with a corresponding percentage of 0.3 and 3%, respectively. A 60-V charge was connected to the solution chamber, measuring the electric flux every 5 min for 6 h. The charge measurement was then calculated via Coulombs. Table 3 shows the values of Coulombs, ranging from negligible to high. Based on the level of charge passed, the resistivity against chloride ion in the concrete demonstrates the porosity of the mix designs microstructure.

2.3.6. Microstructure characterisation

Phenolphthalein spray testing was conducted on the varied mix designs. This method determines a qualitative understanding of carbonation levels within concrete. 0.5 g of phenolphthalein was dissolved within 500 mL of ethanol to create the phenolphthalein solution. The solution was then transferred into a spray-reagent bottle for application on the concrete samples. Test samples were 100 × 200 mm cylindrical moulded cured concrete in accordance with AS/NZS 1012.8.2 [52]. The solution was applied to the various mix design samples at ages of 28, 56, 90 and 365-days. At the respective age of concrete, the samples were cut in half and phenolphthalein spray was applied within 60-s of cutting. Photographs were then subsequently taken to determine the spread of calcium carbonate centres on the cured samples. A novel software was used to measure the darker pixels produced from the photographs obtained after the spray was applied. Photographs were then cropped to image sizes of 300 × 300 pixels. The images were then uploaded to the software and converted into black and white, to represent the morphological feature of the regions that demonstrated colour change. It is important to note that the optimal range of black and white was manually chosen to accurately represent the features of the coloured samples. Upon completion, the software assigns a shade of pixel to represent the areas of carbonation.

The microstructure of the samples was observed using a scanning electron microscopy (SEM) and an energy dispersive x-ray spectroscopy (EDS). These microstructure observations were conducted on the Phenom XL G2 Desktop SEM, samples were prepared using a diamond cutting saw to a height of 2–4 mm and 4–6 mm in diameter.

3. Results and discussions

The results of the composite microstructure and porosity are presented via water immersion, absorption, phenolphthalein indicator, and chloride ion penetration testing. Accelerated ageing results via water immersion techniques are tested via compressive and tensile mechanical testing. These testing methods demonstrate the permeability of the specimens, which can inform future durability performance.

3.1. Microstructure and porosity

3.1.1. Water immersion

The minimum absorption percentage after 28, 56 and 90-day was observed from the control mix, with a rate of 2.28, 1.19 and 1.27% respectively. However, mix design SFKF10 demonstrated the lowest absorption rate at 365 days. Substituting 10% cement with KFs created an increased porosity of the composites. This is shown with the moderate rate of absorption with raw KFs in the initial 90-days, then a spike of absorption at 365-days. The variation of absorption percentage is illustrated in Fig. 1 and the results demonstrate the absorption of all mix designs decreases between 28-day and 56-day periods. This is due to the additional curing of concrete samples. As concrete ages, the microstructural pores constrict with the additional formation of C–S–H [62]. This is further shown with SFKF10 at 365-days, where absorption rates are significantly reduced. The addition of SF as the fibre modification technique increases the formation of C–S–H, which reduces the formation of $Ca(OH)_2$, increasing the fibres durability within the matrix [23]. However, absorption rate of SFKF105 increased after 365-days of ageing. The addition of MK with the modified SF fibres increased the absorption percentage. This is due to the reduction of OPC within the cementitious composite. The pozzolanic properties of OPC within the matrix enhances the bonding properties of the constituent materials. Although MK has been reported to reduce porosity and create a denser matrix [26], the addition of fibres increased the porosity of the composites microstructure. The fibres ability to minimise permeability properties within cementitious composites is dependent on the type of fibre, mix design, fibre length, fibre morphology and curing conditions [63]. KFs are hydrophilic, which enhances their ability to absorb moisture. This characteristic can decrease the bleeding that occurs within cementitious composites during early stages of hydration and curing phases [26]. This factor can reduce shrinkage and cracking occurring because of the interconnected pores and fibres within the microstructure of the composite [63]. Reducing the pore structure of composite materials, reduces the permeability by minimising the connecting flow paths. This can also create a denser matrix which can increase the mechanical strength characteristics. As concrete ages, the compressive strength often increases due these properties [62]. This is also shown with the absorption rate of all mix designs when immersed in water. A significant reduction in porosity is demonstrated at 365-days when compared to the 28-day curing phase.

3.1.2. Water absorption

The testing for composite absorption determines the rate of unidirectional water flow via capillary action [63]. The water

Table 3
Chloride ion penetrability: Based on charge passed [61].

Charge Passed (Coulombs)	Chloride ion penetrability
>4000	High
2000–4000	Moderate
1000–2000	Low
100–1000	Very low
<100	Negligible

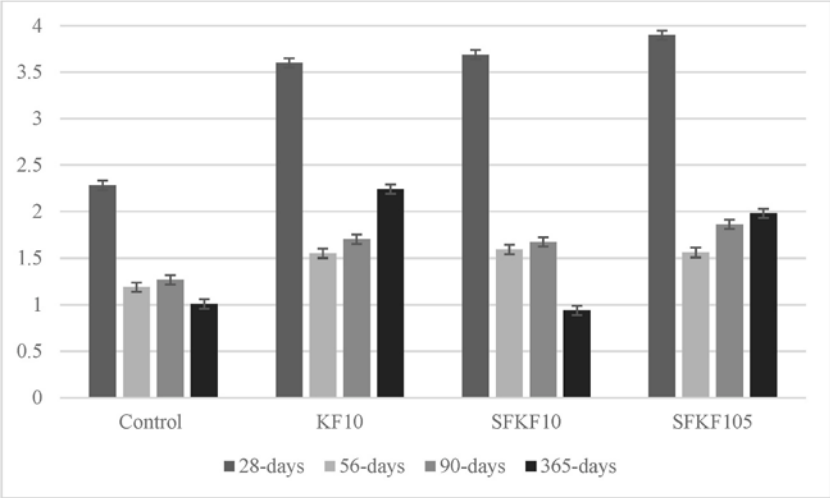

Fig. 1. Water immersion percentage.

absorption percentage after 60 s of exposure is illustrated in Fig. 2. All mix designs had a reduction of water absorption after the initial 28-days of curing. There was a moderate decline with the control mix at 28, 56 and 90-day intervals. However, KF10 and SFKF10 showed a 50% reduction between 56 and 90-day intervals. This is due to the fibres initial absorption of water in early curing stages. Fibre integration can reduce the bleeding that occurs when concrete is formed. Although fibres are integrated within SFKF105, the rate of absorption does not change significantly from 56 to 90-days, demonstrating a 0.17 and 0.16 absorption rate, respectively. This is due to the integration of MK as a partial OPC substitute. The density of MK is lower than OPC and can have an increase of porosity in early ages compared to all OPC concretes [26]. However, as the age of concrete containing MK increases the porosity of the composite decreases [26]. This is shown with a 37.5% decrease from the 90-day interval to MK concrete aged at 365-days. Natural fibres are hydroscopic which demonstrates fibre volume changes due to the absorption of moisture. This factor can promote fibre detachment within composite materials [64]. However, due to the initial thermal and mechanical kraft processing techniques, the fibres ability of absorption is reduced during KF production [17]. In this study, cardboard was reduced to pulp via water absorption and then subjected to additional thermal processes. The thermal processing allowed the shrinkage of fibre volume before being subjected to a wet environment. The linkages of inter-polymer bonds of the cellulose chain were enlarged when water was applied to the fibres [65]. When water evaporates from the composite there is a change in fibre size, promoting voids and gaps throughout the interconnected pores of the material. For this reason, raw KF composites are shown to contain a higher absorption rate compared to non-modified fibre composites. Modified fibres demonstrate a lower absorption rate at all ageing intervals, highlighting an increased stability like the control.

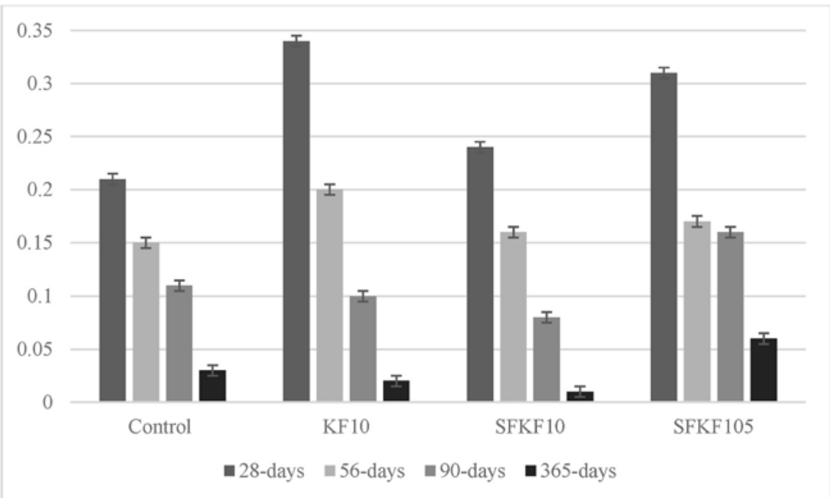

Fig. 2. Water absorption percentage.

3.1.3. Chloride ion penetration

Chloride ion penetration testing was conducted to determine the level of chloride ion movement within the microstructure of concrete materials. The movement of chloride ions can cause significant durability and service life issues of concrete applications. In this experiment, the flow passes through the various mix design samples via the pore solution, which acts as an electrolyte. The interconnected pore system within the concrete affects the passage rate of ions. The more porous the samples are, the higher the rate of flow is to be expected [40]. Therefore, concrete is deemed more durable if there is a reduction in charge via the measurement of coulombs. The charge rating system is shown in Table 3, in accordance with ASTM C1202-19 [61]. As shown in Table 4 and Fig. 3, the highest charge passed was via mix design SF10 with 4830 coulombs at 28-days of age. All samples demonstrated a higher charge rate at 28-days than any other interval age. As the concrete samples aged, the charge rate was reduced. Except for SF10 that had a negligible increase between 90 and 365-day interval. SFKF105 had the lowest charge of 3488 coulombs at 28-days however, the mix design resulted with the highest charge rate of 2563 coulombs at the 365-day interval. This is due to the integration of MK within the mix design. MK has a lighter density than OPC, resulting with a higher porosity [35]. MK consumes $Ca(OH)_2$ at later stages of hydration however, there is an increase of interconnected pores within the microstructure of the constituent materials [66]. The control had a decrease of porosity, with the rating system demonstrating a high to low classification during the testing ages of the specimens. Whereas the remaining mix designs had a moderate rating for their lowest testing classification at 365-day interval. The porous properties of KFs can ease the transport of ions however, there is an increase of interfacial zones between the fibres and the matrix. This increase can enhance the presence of the interconnected pore structure, resulting with a higher charge rate [40]. Raw KFs demonstrated a higher charge rate than SF modified fibres when no other SCM was present. This is due to the additional adhesion SF has within the OPC matrix [64]. The pozzolanic properties of SF enable the material to bond sufficiently to the constituent materials of concrete. When SF is applied to the KF walls, the bond between the fibre and the matrix is also enhanced [64]. In agreement with Booya et al. [63], when fibres are modified it demonstrates an improvement to the chloride ion permeability of concrete materials.

3.1.4. Carbonation indicator

At the respective ages, the concrete was cut transversely, and phenolphthalein indicator was sprayed onto the freshly fractured surface. Photographs recorded the reaction of the spray with the exposed surface, indicating the levels of carbonation that had occurred. This is further shown in Fig. 4. All concrete samples were aged naturally without additional accelerated carbonation tests. This indicated the natural progression of carbonation with the bespoke composite materials. It was reported that the speed of carbonation is reduced when cellulose fibres are present within concrete composites [42]. Ordinary concrete has been shown to demonstrate a higher rate of carbonation [40]. This was further shown in the experimental findings shown in Table 5. At 28-days of curing, the control demonstrated the lowest carbonation percentage. However, the control also had the highest carbonation levels of 45.28% at 365-days. SF10 demonstrated the highest fibre composite carbonation level with 37.79% at the 365-day interval. All mix designs demonstrated an increase over each interval stage except for KF10 that had a marginal difference of 34.67 and 34.19% between 90 and 365-days, respectively. The raw KFs resisted the promotion of carbonation due to the cellulose material absorbing carbon dioxide (CO_2). Matrix and modified fibre composites had a gradual increase during the ageing process. SF modified fibres demonstrated a lower carbonation percentage between 90 and 365-days compared to matrix modified specimens. This is due to the reduced number of varied materials within the composite design. The successful agglomeration of composite materials is critical to ensure uniformity and effective dispersion is maintained during the batching process. Increasing the number of materials can disrupt uniformity and create additional issues such as voids or pockets within the microstructure. Although natural fibres have been shown to reduce the occurrence of carbonation in concrete materials, the addition of MK demonstrates a high increase of carbonation percentages between 90 and 365-days.

3.2. Mechanical and accelerated ageing effects

3.2.1. Compressive strength

The compressive strength of concrete with various fibre types and sample composition are graphically depicted in Fig. 5. The standard deviation is shown via error bars on Fig. 5. The compressive strength of all samples decreased with fibre integration. This was to be expected as KFs contain a lower density than OPC and therefore reduce the total density of the composite material. Fibre integration has been shown to increase permeable voids within composite microstructures [36]. The voids are created due to the hydroscopic characteristic of natural fibres. KFs absorb moisture when being agglomerated with the concrete constituent materials. However, during the curing phase, water molecules evaporate from the KFs within the composite. This creates voids and reduces the interfacial zones between fibres and the respective matrix [65]. When this occurs, the mechanical properties are affected under axial

Table 4
Total charge passed via chloride ion permeability.

Mix design	28 days		56 days		90 days		365 days	
	Total charge (coulombs)	Rating	Total charge (coulombs)	Rating	Total charge	Rating	Total charge (coulombs)	Rating
Control	4291	High	4277	High	2615	Moderate	1927	Low
KF10	4273	High	3086	Moderate	2702	Moderate	2434	Moderate
SF10	4830	High	2250	Moderate	2003	Moderate	2019	Moderate
SFKF105	3488	Moderate	3078	Moderate	2764	Moderate	2563	Moderate

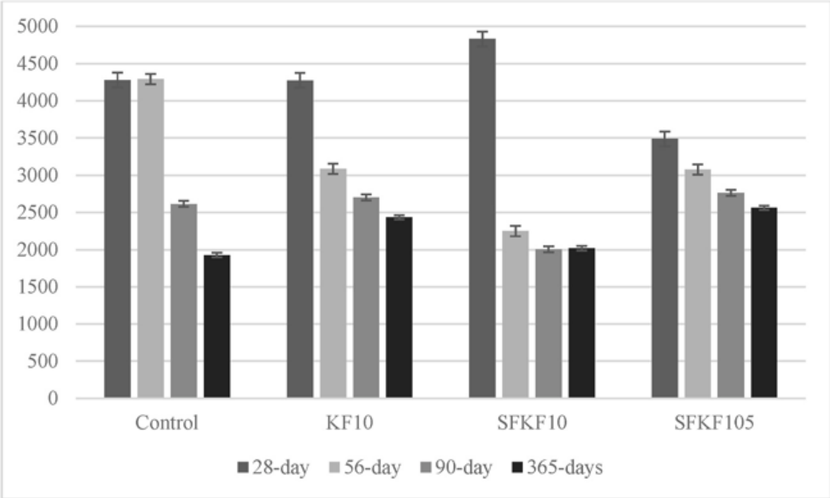

Fig. 3. Charge passed through specimens.

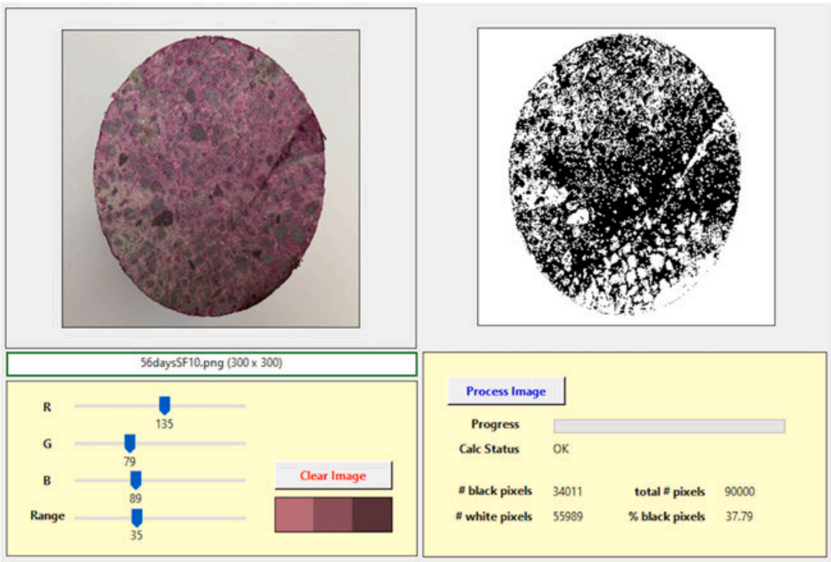

Fig. 4. Software image of SF10 sample at 56-days.

Table 5
Phenolphthalein indicator black pixel percentage.

Mix design	28 days	56 days	90 days	365 days
Control	13.72	19.47	30.89	45.28
KF10	23.48	31.35	34.67	34.19
SF10	15.20	23.71	33.15	37.79
SFKF105	18.51	20.56	27.73	36.80

loading. The compressive strength of the control at 7, 14, 28, 56 and 90-day intervals were the highest. Demonstrating 17, 17, 25, 26, and 30 MPa, respectively. SF modified fibres demonstrated a gradual incline of 10, 13, 14, 17 and 21.5 MPa during the interval ages. Raw KF composites had the lowest compressive strength at all ages. Raw KFs exhibited a significant reduction in strength at 7, 14 and 28-day intervals due to the mineralisation that occurs from OPC products on the fibre walls. This is shown in Fig. 6 where mineralisation has formed on the fibre walls enhancing the occurrence of fibre snapping. Mineralisation increases fibre embrittlement, creating additional voids when fibre failure occurs under compressive loading. The addition of SF on the fibre walls increases the compressive strength of the fibre composite. SF improves the adhesion of the fibres within the matrix as well as reducing fibre volume

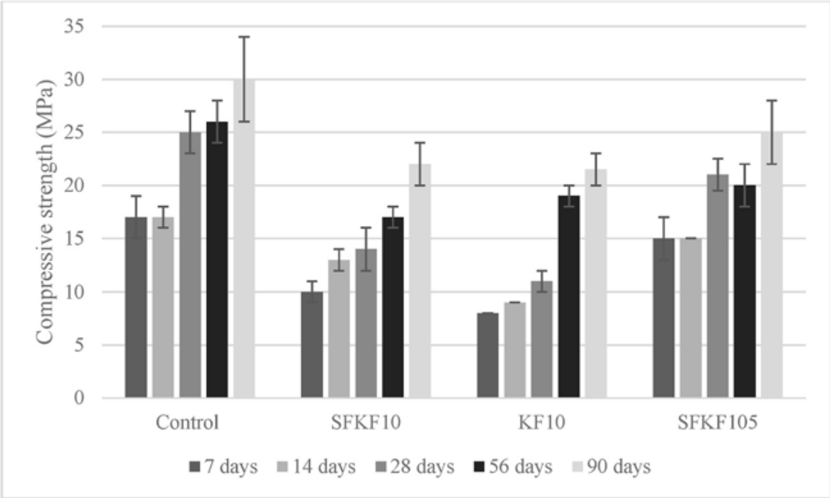

Fig. 5. Compressive strength at various ages.

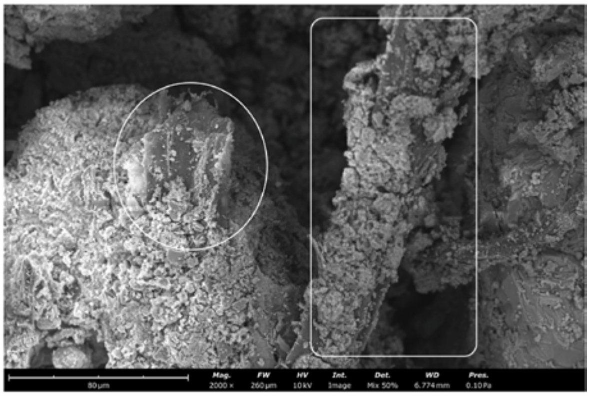

Fig. 6. KF within OPC matrix.

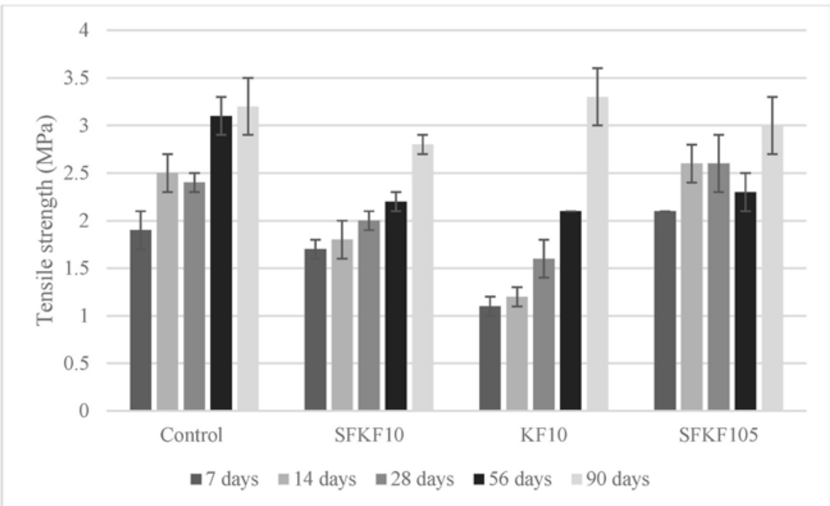

Fig. 7. Tensile strength at various ages.

changes due to absorbed moisture. This is because SF is applied as a coating to the fibre walls, reducing the permeable access of moisture to enter the microfibrils of the natural fibres [64]. SFKF105 demonstrated increased compressive strength compared to the composites with only fibre integration. MK has been shown to increase strength properties among cementitious composites [67]. MK can increase the toughness of the composite which can increase the total strength. MK has been shown to increase absorption rates of water however, the strength of the composite was not hindered. The SiO_2 content that attached to the modified fibre walls has consumed $Ca(OH)_2$ at later stages. This is shown with the increased strength of SFKF10 and SFKF105 at various ageing intervals. The consumption of $Ca(OH)_2$ in early and later stages by MK and SF respectively, has reduced the attachment of cement products on the fibre wall and created a stable environment for the strength of the fibre concrete to increase. This is demonstrated with increased strength values compared to raw KF composites. Although, modified fibres and matrix composition composites do not show rapid increases of strength, KFs can maintain their strength and durability characteristics.

3.2.2. Tensile strength

The indirect tensile strength of the various mix designs is graphically depicted in Fig. 7. The standard deviation is shown via error bars in Fig. 7. SFKF105 demonstrated the highest tensile strength at 7, 14 and 28-day intervals, with strength values of 2.1, 2.6 and 2.6 MPa, respectively. The integration of MK within the composition of the mix design assisted the tensile capacity of the composite. The consumption of $Ca(OH)_2$ in early and later stages of hydration by MK and SF respectively, has reduced mineralisation on the fibre. This is further shown in Fig. 8 which demonstrates minimal cement products attached to the fibre walls. Moreover, the integration of MK highlights the effectiveness the SCM has as a binary blend with OPC. It is important note that the percentage of MK is critical to ensure mechanical properties are not negatively affected. Researchers have demonstrated the substitution ratio of MK with OPC cannot exceed 30% [25]. The control had the highest value of tensile strength at the 56-day interval, demonstrating 3.1 MPa. However, at the 90-day interval there was only a slight increase of 0.1 MPa. Concrete has low tensile strength, requiring the need for steel reinforcement to resist extreme loads. The tensile strength variation can be correlated to the placement of aggregates within the composite. Fibres can form additional strength characteristics however concrete relies upon the constituent materials that cannot aid with increased tensile strength capacities. Both KF10 and SFKF10 had a continual increase of tensile strength over ageing intervals, with a 36 and 22% increase respectively, from 56 to 90-days. The significant rise in strength is due to the fibres ability to agglomerate effectively within the composite material. When fibres interlock within the composite, the axial force that is applied gets transferred throughout the materials microstructure. This is also known as the crack bridging effect [68]. As the composite begins to crack under duress, fibres maintain an interlocking connection to the other side of the crack. Fibre snapping and fibre pull out can occur as the load increases, causing the redistribution of load to transfer to other fibres via the crack bridging mechanism [68]. This increases load bearing capacity of the composite. This can reduce the size of the crack and reduce micro-cracks further developing. Fibres fail when petrification occurs, causing the fibre to become brittle and weak. This is shown with raw KF composites that demonstrate a lower tensile strength compared to modified fibres. SF improves the adhesion between the fibre and the cementitious environment. The SF coating applied to the fibre walls reduces the interfacial zone and enhances a cohesive bond. This bonding mechanism enables a stable environment for the fibres to resist external forces. The increased level of C–S–H from the integration of SF decreases the alkalinity of the matrix which reduces the attack of $Ca(OH)_2$ on the fibre walls.

3.2.3. Wet and dry cycles/accelerated ageing

Accelerating the ageing process is common method to test the durability properties of composite materials [69]. To comprehensively analyse the degradation caused by wet and dry cycles, three KF mix designs were used. First, raw KFs, second SF modified fibres and finally, SF modified fibres with MK as a partial cement substitute. To determine the effect of wet and dry cycles, the values shown in Figs. 5 and 7 demonstrate a baseline of the mechanical strength. The compressive strength values of the various cycles implemented are shown in Fig. 9. The standard deviation of the compressive results is shown via error bars in Fig. 9. The control had the highest compressive strength value of 31 MPa after 20 cycles. However, the subsequent cycle intervals demonstrated a reduction in strength. The initial 20 cycles accelerated the hydration and curing phase of the concrete specimens, increasing significant strength of the control. However, as the ageing process continued, there was a reduction in strength. All composites containing fibre integration had significantly lower strength than the control in the initial 20 cycles. As cycles continued, the fibre composites compressive strength increased, reaching a peak at the 60-cycle interval. Throughout the wet and dry cycle process, composites are subjected to complete water immersion. The composite materials absorb the water via the microstructural pore system, inducing volumetric phase changes of the constituent materials. As discussed, the hydroscopic nature of KFs endure phase changes within the microfibrils of their cellulose chains. When the composites are removed from water and subjected to drying, KFs undergo another volumetric change with the expulsion of water molecules. This repeated process can increase the porosity of the composite via the reduction of the interfacial zone of the fibre and the matrix. The fibres interlocking connection with the matrix becomes weaker and fibre pull out becomes apparent when composites are subjected to mechanical loading. As shown with raw KFs, the increased intervals after 60 cycles reduces the compressive strength. SFKF composites demonstrated a later reduction in strength compared to raw KFs. SF modified fibres decreased in compressive strength after 80 cycles. This highlights the important factor of modifying fibres within cementitious matrices. SF has enabled the bonding of fibres by minimising the attack of $Ca(OH)_2$ in early stages of hydration but also mitigating the volumetric changes caused by water absorption. The coating applied to the fibre walls is primarily SiO_2, which acts as a barrier for significant water penetration. This is also highlighted with the mix SFKF105. SFKF105 demonstrated the highest compressive strength of all fibre composites at 60 cycles. The enhanced matrix via MK minimised the attack of $Ca(OH)_2$ at later stages of hydration, which enabled the fibre to maintain its natural strength characteristics. After 100 cycles, all samples demonstrated a higher increase of compressive strength than the initial 20 cycle interval. There was an increase of strength as concrete ages however, 10–15% cement substitution

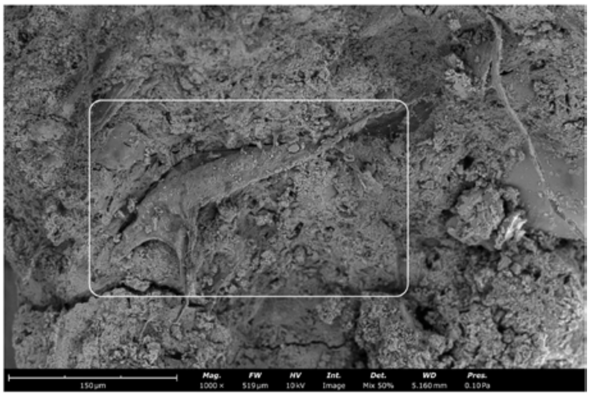

Fig. 8. Fibre without mineralisation (SFKF105).

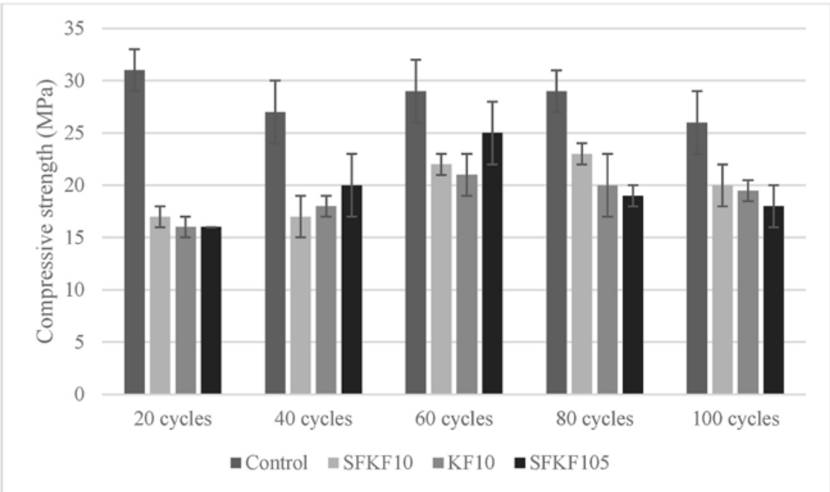

Fig. 9. Compressive strength vs wet/dry cycles.

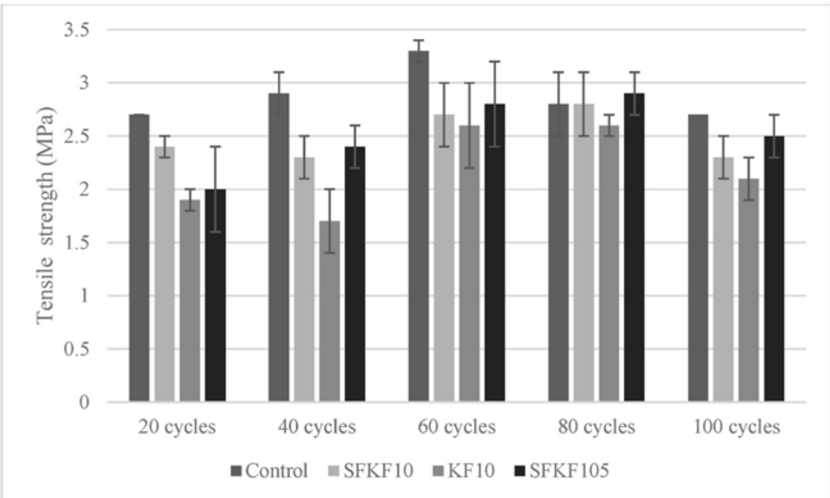

Fig. 10. Tensile strength vs wet/dry cycles.

does alter the physiochemical reaction of constituent materials as concrete ages. It is important to note that the overall compressive strength value of fibre composites can meet non-structural applications.

The indirect tensile strength of all wet and dry cycles is shown in Fig. 10. The standard deviation of the tensile results is shown via error bars in Fig. 10. The control had the highest tensile strength value at the 60-cycle interval. However, there was a steady decline in strength for the remaining 80 and 100 cycles. This was also demonstrated with the compressive strength in Fig. 9. There was a similar trend for fibre composites however, the peak strength began to reduce after the 80-cycle interval. SFKF105 composites demonstrated the highest tensile strength with fibre integration. This was also shown via the compressive strength values. As discussed, the integration of MK enables a stable environment for the fibres to maintain their natural strength characteristics. Wei and Meyer [33] have also noted the integration of MK as a partial cement substitution can enhance fibre composite strength. However, their research demonstrated that the strength was significantly compromised after 15, 30 and 50 wet and dry cycles. This was not shown in the initial 80 cycles due to the integration of SF on the fibre walls. Raw KFs demonstrated the lowest tensile strength, with a significant reduced strength by the 40-cycle interval. At 60 cycles, raw KFs had a 35% increase. This was the highest percentage increase within a 20-cycle period. The raw KFs were exposed within the microstructure of the composite, allowing for volumetric changes to be imposed on the fibre walls. The morphology of the composite's microstructure altered due to the additional interfacial changes occurring around the fibre zones. When natural fibres are near each other within composite materials, there is a van der Waals force pulling the fibres together [38]. This creates the additional interlocking system of fibres which inherently enhances the crack bridging effect as axial loading is applied. As fibres undergo duress from load applications, the fibres will either snap or pull away from the bonds created within the matrix. Therefore, it is critical to ensure a sufficient bond has been promoted and petrification has not occurred from cement products on the fibre walls. Fibre integration prolongs curing phases of cementitious composites. The absorption of water molecules on the fibre walls reduces the bleeding of concrete materials but also releases the absorption of water within the matrix as curing continues [57]. This is an important factor that reduces the dispersion of micro-cracks within the matrix. The promotion of micro cracks and increased porosity can occur via accelerated curing conditions in early stages of concrete development. Minimising the transference between concrete pores can increase the mechanical properties of the composite and increase the durability factors associated with carbonation and chloride ion penetration.

4. Conclusion, assumptions, and limitations

The evaluated durability properties of fibres derived from waste cardboard was experimentally investigated. Matrix and fibre modified composites were analysed for their water absorptivity rates, chloride ion permeability, carbonation, and mechanical strength properties. SF was applied to the fibre walls to enhance the durability of fibres within the high alkaline environment. This reduced the hydroscopic nature of KFs by minimising volumetric changes within the microfibrils of the fibres. The mechanical analysis of concrete samples containing raw KFs, modified KFs and matrix modified specimens demonstrated that waste cardboard fibres can be adequately integrated within concrete and mortar materials. Accelerated ageing via wet and dry cycling demonstrated a reduced mechanical strength of all composite materials as ageing continued. Chloride ion penetration testing demonstrated the porosity of the composites and the ability of ions to transverse within the microstructure of the composites. Each mix design contained a lower Coulomb charge as the concrete specimens aged. However, the incremental charge range varies significantly, allowing moderate ratings to have diverse readings. A limitation of this study was the ageing assessment of fibre composites, further research is required on the thermal characteristics of altered fibre designs and exposure to increased levels of thermal processing. This research assumes the processing and application of waste cardboard is sustainable. The testing methods focused on the physical durability characteristics. Future studies can be focused on evaluating other factors of utilising waste cardboard as a sustainable raw material and dual fibre integration. The most important outcomes of this research are summarised as follows.

- SFKF10 and the control demonstrated the lowest porosity via chloride ion permeability testing
- After 60–80 cycles all composites began to demonstrate a reduction in composite strength
- Raw KF composites demonstrated a lower carbonation percentage than matrix and fibre modified specimens
- Water immersion testing showed SFKF10 had the lowest absorption rate at 365-days
- Water absorption testing showed SFKF105 had the highest surface absorption
- MK increased the cementitious composites porosity
- SF reduced the water permeability to the fibre walls
- Compressive strength increased with time for all composite designs
- Tensile strength increased when no SCMs were integrated
- SEM analysis show a higher percentage of petrification occurring to non-modified fibres

Author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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6.3 Concluding remarks

This chapter presented the durability properties of fibres derived from waste cardboard within concrete composite materials. Matrix and fibre modified composites were analysed for their water absorptivity rates, chloride ion permeability, carbonation, and mechanical strength properties. Accelerated ageing via wet and dry cycling demonstrated a reduced mechanical strength of all composite materials as ageing continued. Chloride ion penetration testing demonstrated the porosity of the composites and the ability of ions to transverse within the microstructure of the composites. Each mix design contained a lower Coulomb charge as the concrete specimens aged. However, the incremental charge range varies significantly, allowing moderate ratings to have diverse readings. The most important outcomes of this chapter are summarised as follows.

- SFKF10 and the control demonstrated the lowest porosity via chloride ion permeability testing.
- After 60-80 cycles all composites began to demonstrate a reduction in composite strength.
- Raw KF composites demonstrated a lower carbonation percentage than matrix and fibre modified specimens.
- Water immersion testing showed SFKF10 had the lowest absorption rate at 365-days.
- Water absorption testing showed SFKF105 had the highest surface absorption.
- The addition of MK increased the cementitious composites porosity.
- SF reduced the water permeability to the fibre walls.
- Compressive strength increased with time for all composite designs.
- Tensile strength increased when no SCMs were integrated.
- SEM analysis shows a higher percentage of petrification occurring in non-modified fibres.

Chapter 7 – Life cycle assessment

7.1 Introduction

In this chapter, a life cycle assessment (LCA) was conducted to see the environmental effect waste cardboard KFs have within low stress-grade concrete materials. This chapter presents three key stages of methodology to evaluate the viability and environmental effects of the waste material in concrete. First, the mechanical strength of KFs as a partial cement substitute within concrete materials. Secondly, the LCA outputs on key impact categories were analysed. Finally, a sensitivity analysis was conducted on the variability of GHG emissions produced from transport and energy usage.

The following paper is included in this chapter:

Robert Haigh, Malindu Sandanayake, Yanni Bouras, Zora Vrcelj. 2023. A life cycle assessment of cardboard waste in low stress grade concrete applications, *Journal of Environmental management (Under review)*.

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2. CANDIDATE DECLARATION

I declare that the publication above meets the requirements to be included in the thesis as outlined in the HDR Policy and related Procedures – policy.vu.edu.au.

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3. CO-AUTHOR(S) DECLARATION

In the case of the above publication, the following authors contributed to the work as follows:

The undersigned certify that:

1. They meet criteria for authorship in that they have participated in the conception, execution or interpretation of at least that part of the publication in their field of expertise;

2. They take public responsibility for their part of the publication, except for the responsible author who accepts overall responsibility for the publication;

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A life cycle assessment of cardboard waste in low stress grade concrete applications

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Abstract

Utilising cardboard waste for the partial substitution of cement within concrete has the potential to yield significant sustainability benefits. Cardboard waste is abundantly available, and a significant proportion of this material is disposed of in landfill. However, conversion of waste cardboard into kraft fibres (KFs) for concrete implementation can be utilized in the building and construction industry. Therefore, identification of sustainability variables associated with cardboard waste in concrete is vital. In this study, two KF composites satisfied the criteria for low stress grade concrete and were subsequently evaluated. SFKF5 mix design contained 5% KFs and SFKF105 contained 10% KFs with 5% metakaolin (MK). Both composites had silica fume (SF) as a fibre modification technique for durability purposes. A life cycle assessment (LCA) determined the environmental effect of waste cardboard integration. A Monte-Carlo simulation was utilised as the sensitivity analysis to investigate transportation and energy manufacturing greenhouse gas (GHG) emission variables. LCA results of SFKF105 had a savings of 11%, 8%, 4% and 1% for terrestrial acidification potential, global warming potential (GWP), terrestrial ecotoxicity potential (TEP) and human toxicity potential, respectively. SFKF5 revealed savings of 3%, 2% and 4% for GWP, TEP and marine eutrophication potential, respectively. The additional travel requirements of KFs and MK to the cement batching plant for composite production did not surpass the embodied energy and travel emissions of the control. However, this was negated due to the additional energy requirements to manufacture KFs. The control, SFKF5, and SFKF105 had an average total of 572, 1023 and 997 kgCO_2-eq/m^3, respectively.

Keywords: Concrete, composites, kraft fibres, life cycle assessment, waste cardboard

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1.0 Introduction

Concrete is the most consumed resource on earth following water [1]. A key constituent material in concrete is cement, and the global production of cement contributes to 5-8% of total carbon dioxide (CO_2) emissions [2]. In 2021, the global consumption rate of cement was estimated to be 4.42 billion tons worldwide [3]. This is a 33% increase of cement consumption over the last decade [4]. Cement production requires significant energy demands from extraction to transportation. During the production of cement, fifty percent of CO_2 emissions are due to the combustion of fossil fuels and the remaining emissions are caused by the calcination process of limestone [5]. The combination of these processes approximately equates to a 0.77 ton of CO_2 emissions released for every one ton of cement clinker produced [6]. However, this value is dependent on the location, manufacturing equipment, energy sources and production efficiency during each stage of manufacturing [7]. Developed countries are continuously investing heavily in infrastructure projects; therefore the demand for cement-based concretes is expected to consistently increase over the next decade [8]. The current consumption rate and requirement of virgin materials in concrete is creating mineral resource deficits in countries such as Hong Kong, Germany and the Netherlands. These countries are now importing basic constituent materials for concrete production [9, 10]. Therefore, there is an urgent requirement to identify alternative materials to supplement traditional constituent materials in concrete while also reducing greenhouse gas (GHG) emissions and energy consumption. To achieve this, alternative processes in concrete production, resource consumption and waste management systems must be reviewed [11, 12].

For many decades researchers have experimented with the partial substitution of cement in concrete with industrial waste materials such as fly ash (FA), silica fume (SF), ground blast furnace slag (GBFS) and glass powder [13-16]. These materials are also known as supplementary cementitious materials (SCM)s and can be mechanically beneficial in concrete due to their pozzolanic properties [17]. Other SCM materials such as metakaolin (MK) are also seen as a sustainable option to cement due to the reduced energy requirements when processing [18, 19]. Researchers have introduced glass and plastics within cementitious materials due to their pozzolanic reactivity with other constituent materials [13]. Waste plastics and glass are becoming popular fillers within road base infrastructure, however, packaging waste materials such as cardboard remain seldom considered [20]. This method of integrating common waste materials can assist the drive toward the circular economy framework [21]. Although there are promotion difficulties to utilise waste materials in concrete due to a reduction in mechanical strength characteristics [22]. There are opportunities to utilise waste as a constituent material in concrete for low-stress grade applications [13, 23]. The use of low stress grade concrete can include; screeding and slab infill, driveways, footpaths, and various concrete landscaping systems [24, 25]. Currently, the building and construction industry promotes the use of waste plastics and glass in infrastructure projects [26, 27]. Table 1 depicts common waste materials in low stress grade concrete. As shown in Table 1, plastics are the dominant waste material being utilised in low stress grade concrete materials. This is further showcased with the use of plastics becoming a common material in civil construction works. However, minimum research has been conducted on the alternative uses of cardboard waste. Despite many experimental investigations, studies have seldom considered benchmarking total environmental impacts of integrating waste materials in concrete. There are environmental benefits when supplementing high GHG emission producing materials in concrete. However, the additional energy requirements of processing and transport requirements can affect the total environmental benefit initially established. Moreover, solely integrating waste materials does not ensure bespoke concrete is sustainable. Key factors to consider of waste integration are the local availability and material abundance. Due to the wide application of cardboard across many industries and 5.92 million tonnes of cardboard waste produced annually in Australia [28]. This research will focus on the integration of cardboard waste in concrete materials to further promote sustainable practices among industry. Researchers [22, 29-31] have focused on utilising kraft fibres (KFs) derived from waste cardboard within cement composites to demonstrate the mechanical and physical characteristics. MK and SF have also been integrated to enhance fibre durability. However, the environmental impact has not been benchmarked when integrating these materials.

Hence, the current study focused to analyse and quantify the environmental sustainability of partial cement substitution with KFs in concrete. A life cycle assessment is widely employed to assess the environmental impacts associated with a product or procedure over its life cycle [32]. The LCA provides a substantial evaluation of the environmental impacts, energy requirements and emission reduction opportunities in various systems and processes. The LCA outcome can assist with making critical decisions toward policies and sustainable investment opportunities [11]. Promotion of novel materials requires benchmarking of the associated environmental benefits. However, among the previous LCA studies conducted on concrete materials [33-36], few have focused on the integration of waste materials that reside in the residential and commercial sector. In addition, no LCA study has been presented with KF integration in concrete materials. To measure the environmental impact of low-stress grade concrete, three strategies will be implemented in the LCA. First, the optimal KF concrete compressive strength is determined of the novel composites via SF fibre and MK matrix modifications. Secondly, LCA impact categories are selected and measured. Finally, a sensitivity analysis of the bespoke composites is conducted to reduce the parameter variables of concrete production. This paper aims to demonstrate the viability of waste cardboard integration and outline the environmental affect when integrated within concrete.

2.0 Research significance and methodology

The detrimental impacts on the environment from various industries are becoming increasingly criticised due to globally accepted climate change reduction targets and the construction industry is no exception. Comprehensively quantifying the environmental impacts of building and construction materials over the whole life cycle is an effective method to benchmark sustainability benefits. Nowadays, the search for locally available alternative materials to replace energy-intensive virgin resources is a popular and an effective approach to achieve sustainable material products that contribute to circular economy [21]. Use of abundant waste materials as a supplementary virgin material can both solve the issues of excessive resource depletion and significant landfills due to extreme waste generation. Use of waste materials in concrete to replace cement and aggregates has been a prominent research focus over the past decade with included materials such as masonry, plastics, glass, FA and GBFS [37-41]. However, research is still exploring the possibility of using novel waste materials in concrete otherwise would end-up in landfills. The world business council for sustainable development (WBCSD) created a cement technology roadmap to identify opportunities of CO_2 emission reductions by 2050. The WBCSD identified four fundamental areas that the cement industry must adopt to achieve emission targets worldwide. These include a source of alternative fuels, energy efficiency, clinker substitutions, carbon capture and storage [42]. Moreover, the integration of alternative waste materials in concrete can ensure the building and construction industry aligns common construction methods with positive environmental change. Therefore, LCA investigations can promote the use of alternative methods based on environmentally positive sustainable results, before large economic investments are required for further development. The use of kraft fibres derived from waste cardboard in concrete materials has not been environmentally benchmarked. Therefore, this research will investigate the mechanical strength characteristics of KFs with a potential application. This will determine the acceptable KF concrete mix design to assess for their environmental characteristics. Moreover, an in-depth sustainability analysis is required to comprehensively determine the effect of sensitivity factors. Figure 1 illustrates this research methodology.

Table 1 Waste materials in low stress grade concrete

Waste material	Partial replacement	Strength (MPa)	Reference
Fly Ash	Cement	25	[43]
Ground blast furnace slag	Cement	17	[43]
Concrete	Coarse aggregate	22	[44]
Glass	Fine aggregate	12-23	[37]
Glass	Coarse aggregate	17- 22.5	[37]
Polypropylene (e-waste)	Coarse aggregate	5-30	[38]
Acrylonitrile butadiene styrene (e-waste)	Coarse aggregate	10-51	[38]
Crumb rubber	Fine aggregate	20- 41	[45]
Ceramic	Fine aggregate	23-28	[46]
Ceramic	Coarse aggregate	24- 26	[46]
Polyethylene terephthalate	Fine aggregate	21-37	[47]
Polyethylene terephthalate	Coarse aggregate	9- 18	[48]
Polyvinyl chloride	Coarse aggregates	16- 46	[49]

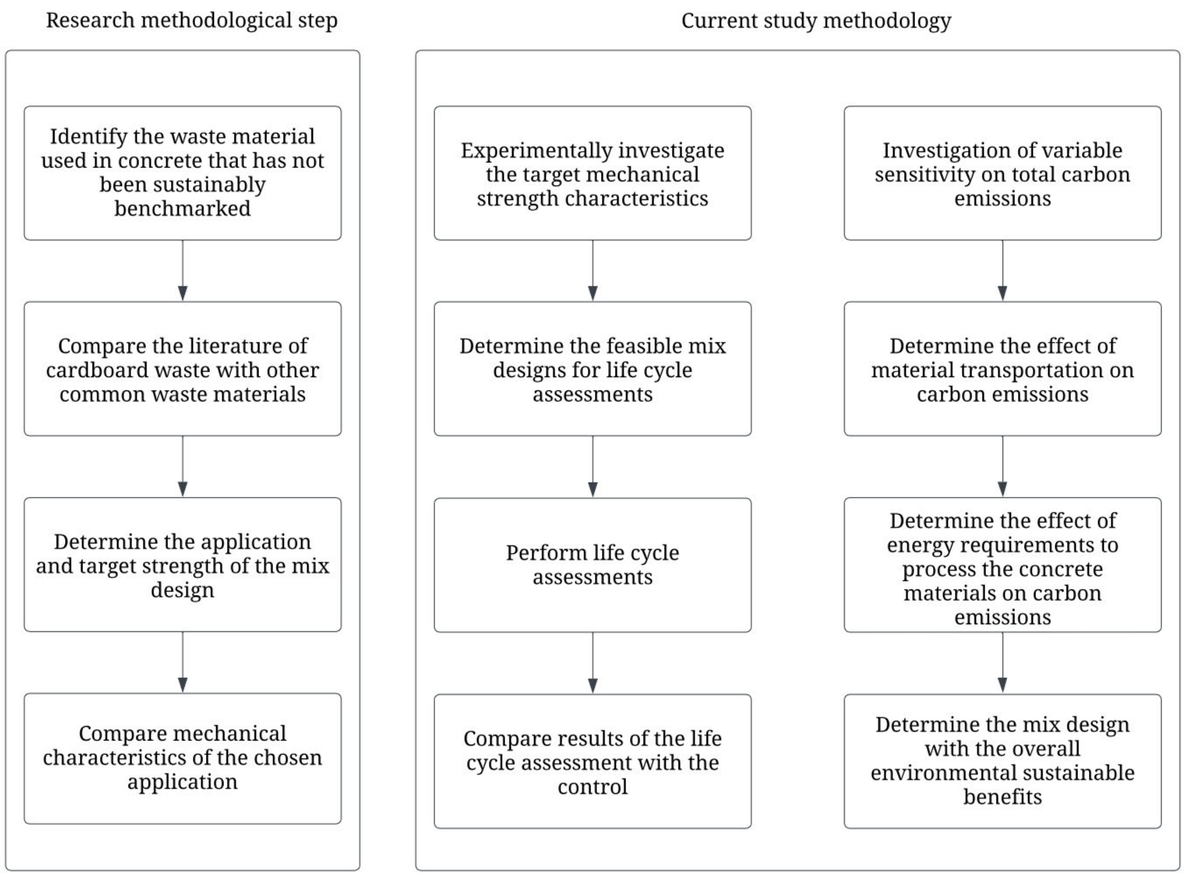

Figure 1 Research process and methodology

3.0 Assessment methodology
3.1 Experimental mechanical testing

Compressive strength testing was conducted on the Matest C088-11 N Servo-Plus evolution testing machine and Cyber-Plus evolution data acquisition system. The constituent materials were mixed via a mortar mixer in accordance with AS/NZS 1012.2 [50]. SF was applied to the fibres as a modification technique conforming with AS/NZS 3582.3 [51]. Matrix modification using MK was used in accordance with ASTM C-618 [52], Class N specifications for natural pozzolans. Fine and coarse aggregates were sourced in accordance with AS/NZS 1141.5 [53] and AS/NZS 1141.6.2 [54], respectively. General purpose cement was the main pozzolanic constituent material conforming with AS/NZS 3972 [55]. The composition of the cement is shown in Table 2. Regular potable water was used throughout all mix designs. All samples were created in a laboratory environmental in accordance with AS/NZS 1012.8.2 [56].The specimens created are to demonstrate viability of the materials being used among industry practices. The processing technique is demonstrated in Figure 2.

Table 2 Composition of general-purpose cement [57]

Material	Formula	Proportion

Portland cement clinker	NA	>92%
Lime stone	$CaCO_3$	0-7.5%
Gypsum	$CaSO_4.2H_2O$	3-8%
Clinker Kiln dust	NA	0-2.5%
Chromium (VI) hexavalent	Cr^{6+}	Trace

3.2 Life cycle assessment

To compare the environmental impacts of concrete prepared with the selected waste material, the LCA approach is required. This study utilised OpenLCA as the primary software in conjunction with Ecoinvent 38 database [58, 59]. The methodology of the current LCA study presented was in accordance with the ISO 14044 [60]. According to the ISO 14044, the LCA methodology has four primary requirements including scope definition, inventory analysis, impact assessment and interpretation.

3.2.1 Scope functional unit, and system boundary

Previous LCA studies have been conducted to compare environmental benefits of integrating various waste materials within cementitious composites, demonstrating their environmental impacts [32, 61-63]. These studies included coffee cup waste, polyethylene bottles and recycled concrete aggregate waste as a supplementary virgin material in cementitious composites. However, according to the recent review and understanding, the current study is the first attempt to investigate the environmental impacts associated with re-using cardboard waste materials as a cement replacement material in concrete. Therefore, the current LCA focused on identifying, comparing, and analysing the environmental impacts related to the production of concrete using cardboard waste as a partial cement substitution. The results aim to demonstrate the viability of diverting cardboard waste from landfills and highlight potential environmental benefits associated. Concrete production of all mix designs was created at a laboratory scale to ensure an effective comparison of the novel materials by reducing unknown variables.

The functional unit for concrete in the current LCA study was taken as $1m^3$. The functional unit provides reference to the input and output data and is an important measurement for the final interpretation of the results [64]. The study considered a cradle-to-gate system boundary as shown in Figure 2 to evaluate and compare environmental impacts. The cradle-to-gate process includes the extraction, transportation, production, and distribution of the final concrete materials to a construction site. The maintenance, usage life cycle and final disposal of the various concrete mix designs are assumed to be the same and therefore are excluded from the system boundary.

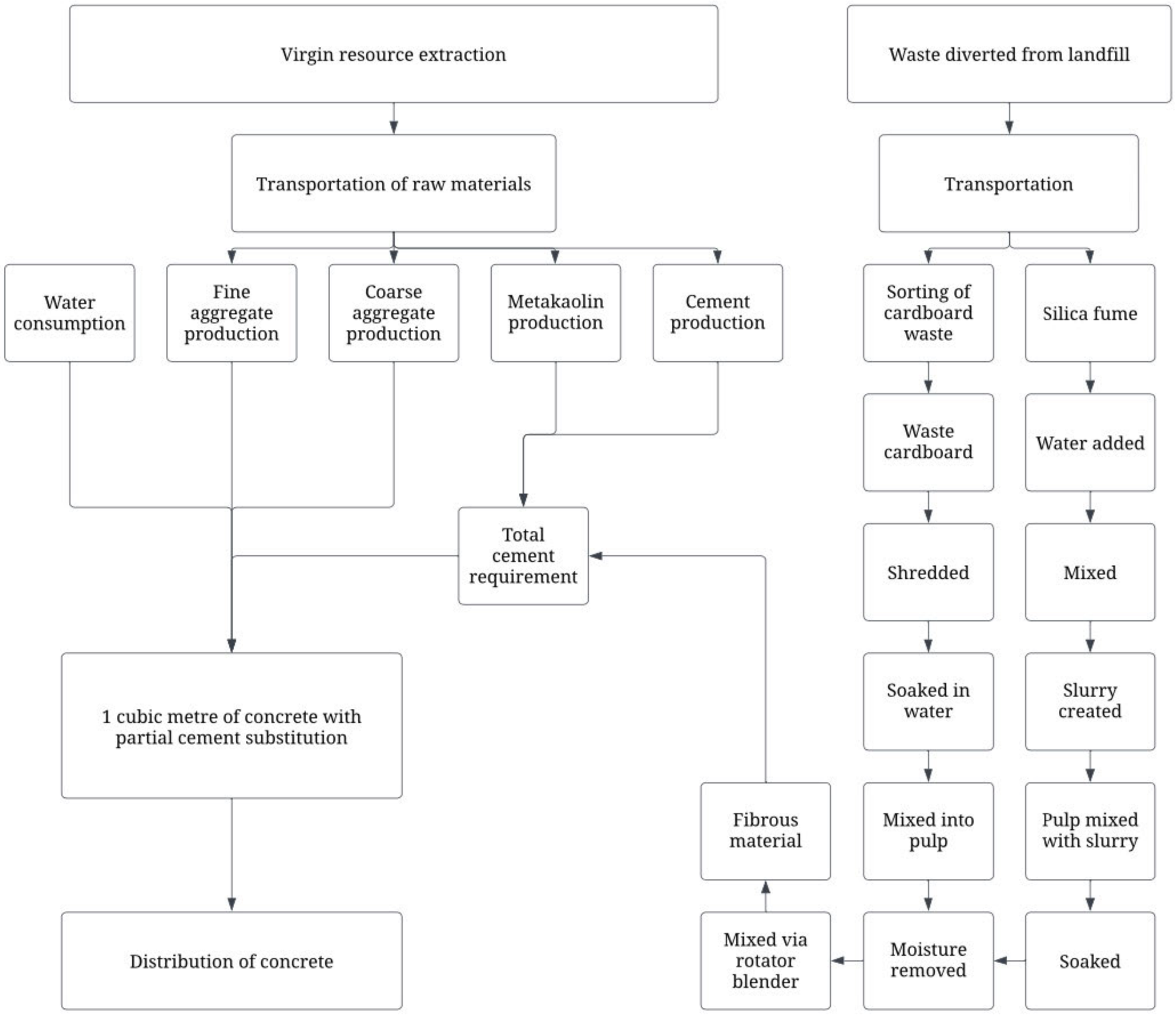

Figure 2 System boundary for the current study

3.2.2 Life cycle inventory analysis

Laboratory process similar to an industry production was used to enable the successful integration of cardboard waste within concrete composites [65]. The cardboard used in this study was extracted from common packaging materials ready to be sent to the material recovery centre. Cardboard waste was subjected to a composition transformation to be used as a constituent material in concrete which included reducing materials to a pulp. It was then transferred into a fibrous material via thermal and blending techniques and the resulting fibres extracted from waste cardboard were called as kraft fibres (KF). In this study, LCA are conducted on three mix designs to compare the environmental impacts for potential use in low stress industry applications such as concrete driveways, screeding and slab infill, footpaths, and various concrete landscaping systems. The mix design compositions are presented in Table 3. The samples included the control, SF modified KFs, and a combination of matrix and fibre modified specimens. It is important to note that surface modification is applied using SF on the KFs at a maximum of 2% loading of the total amount of KFs. This has been shown to be sufficient as a fibre modification technique to enhance the durability of the fibres in high alkaline environments [66].

Moreover, the application of SF can also improve the mechanical strength of cementitious composites, reduce permeability and thermal cracking [67].

In addition to fibre modified concrete samples, matrix modification using metakaolin was integrated at a 5% cement replacement rate. Metakaolin is a highly reactive pozzolanic material obtained by the calcination of kaolin clay. The calcination process involves heating the kaolin clay between 600-850°C. This process transforms the mineral kaolinite into a material with different properties known as metakaolin [68]. The use of metakaolin as SCM is seen to be a sustainable alternative to cement as the energy required is significantly less [19]. The energy requirements and sources of all constituent materials such as fine aggregate, coarse aggregate, MK, and general-purpose cement are shown in Table 4. It is important to note that the energy requirements and sources for the conversion of cardboard waste into KFs are detailed in 4.3.2 Energy emissions.

The concrete mix designs were based on a target compressive strength target of 25MPa. There are additional energy and transportation requirements for the processing and integration of KFs. However, all other materials are assumed to be transported to a one batching plant for concrete production. The mix design code correlates to "SF"- silica fume, "KF"- kraft fibre, the initial number of '10' represents the fibre percentage and the final number '5' representing the amount of MK within the mix design.

Table 3 Composition of various mix design specimens per cubic metre

Mix designs	Cement	SFKF	MK	Fine aggregate	Coarse aggregate	Water
Control	355.9			733.6	1100.5	210
SFKF5	338.1	17.79		733.6	1100.5	210
SFKF105	302.52	35.58	17.79	733.6	1100.5	210

Table 4 Energy inputs for material production (megajoules per kilogram)

Materials	Brown coal	Black coal	Crude oil	Geothermal	Solar energy	Water power	Wind power	Natural gas	Ref
Cement	11.9	26.3	1506.27	5.18	0.24	2.07	0.30	44.1	[58, 69]
MK	11.9	26.3	42.3	-	-	-	-	44.1	[58, 70]
Fine aggregate	11.9	26.3	42.3	0.00003	-	-	-	44.1	[58, 71]
Coarse aggregate	11.9	26.3	42.3	0.00011	-	-	-	44.1	[58, 72]

3.2.3 Life cycle impact assessment

There are three elements of a life cycle impact assessment (LCIA). This is the selection of impact categories; classification of the categories and calculation of the category indicator results. This section quantifies the environmental impacts. The problem-orientated method is adopted for this research and is also known as the "midpoint method". The midpoint method correlates to the environmental categories for which this study will identify, compare, and analyse. Based on the current global environment, the following seven impact categories were selected. Terrestrial acidification potential (TAP100 in KgSO_2-eq), global warming potential (GWP100 in kgCO_2-eq), terrestrial ecotoxicity potential (TEP100 in Kg 1, 4-DCB-eq), marine eutrophication potential (MEP100 in Kg-N-eq), human carcinogenic toxicity potential (HTP100 Kg 1, 4-DCB-eq), stratospheric ozone layer depletion potential (ODP100 Kg CFC-11-eq) and mineral resource scarcity potential (MRS in Kg Cu-eq). The normalisation factors for each of the impact categories is summarised in Table 5. The ReCipe hierarchist (h) Midpoint method was used which combines the eco-indicator 99 and CML baselines. OpenLCA software using the Ecoinvent 38 database was used to model material and energy flows for the system boundary considered.

Table 5 Impact categories and normalisation factors

Impact category	Normalisation factors		
	Unit	Value	Reference
Terrestrial acidification potential	KgSO_2-eq	4.10E+01	[58, 59]
Global warming potential	GWP100 in Kg CO_2-eq	7.99E+03	[58, 59]
Terrestrial ecotoxicity potential	Kg 1, 4-DCB-eq	1.04E+03	[58, 59]
Marine eutrophication potential	Kg-N-eq	4.61E+00	[58, 59]
Human carcinogenic toxicity potential	Kg 1, 4-DCB-eq	2.77E+00	[58, 59]
Stratospheric ozone layer depletion potential	Kg CFC-11-eq	5.90E-02	[58, 59]
Mineral resource scarcity	Kg Cu-eq	1.20E+05	[58, 59]

3.2.4 Limitations and assumptions

The scope, and system boundaries of LCA studies retain limitations and assumptions due to the research direction and desired objectives. The current study has the following assumptions and limitations.

- The study did not consider end of life behaviours with assumption that all the mix designs will have similar end-of-life considerations
- The study assumed the lifespan of the bespoke mix designs equalled that of the control
- The supply of processed waste cardboard is assumed to be locally available in abundance
- Where emission inventories are not available, emission factors are adopted from published literature
- Waste cardboard conversion into fibrous material was assumed to be conducted at the waste recovery centre

- An equal transportation distance of 36 km was used to transport fine, coarse and cement materials to the concrete batching plant
- The mortar mixer used 4 kWh to create 1 cubic metre of concrete
- Medium voltage power supply is used for the mixing of concrete, drying of fibres and blending of cardboard materials
- Conventional treatment of potable water was assumed, and the corresponding emission inventories were adopted from existing databases

3.3 Sensitivity analysis

A Monte-Carlo (MC) simulation is a sampling method to perform an uncertainty analysis of the parameter variables and investigate its influence on the total output [73]. The MC simulation is a technique used in mathematical model that captures the variability of data in a system using probability distribution [74]. In this study, material transport and energy requirements to produce the novel concrete composite materials can be considered uncertain. The scope of the study is to measure the environmental effect of the various mix designs and therefore the sensitivity analysis using the MC simulation will focus on the GHG emissions created. Table 6 demonstrates the GHG emission factors from transportation and energy sources. In this study, the MC simulation utilises the triangular probability distribution. This is a continuous probability used to represent uncertain variables to which values fall within a specific range. It is triangular when plotted on a graph and defined by three parameters (within the range):

- The minimum value
- The maximum value
- The most likely value

The use of the triangular probability distribution is used in this study due to the minimum and maximum values known for energy consumption, with the output of the simulation providing the most likely. This approach assists when assessing the possible range of outcomes and their probabilities, providing valuable insights into the behaviour of complex systems affected by uncertain variables.

Table 6 Input variables used for Monte-Carlo simulation

Variable parameter	Minimum	Maximum	Probability distribution	Reference
Transportation truck emissions	0.161	0.307	Triangular	[75-77]
Coal (black and brown) emissions	0.63	1.63	Triangular	[75-77]
Gas emissions	0.27	0.9	Triangular	[75-77]
Renewable energy emissions	0.03	0.09	Triangular	[75-77]
Construction site distance (km)	10	150	Triangular	[78-80]
Production time (minutes)	60	300	Triangular	[81-83]

Capacity of cement mixer (kWh)	0.8	1.1	Triangular	[84-86]
Capacity of rotator mixer (kWh)	1.2	1.6	Triangular	[87-89]
Capacity of rotator blender (kWh)	1.1	1.5	Triangular	[90-92]
Capacity of oven (kWh)	0.5	1.8	Triangular	[93-95]
Capacity of cardboard shredder (kWh)	0.6	1.4	Triangular	[96-98]

4.0 Results and discussion

4.1 Experimental results

The compressive results of the various mix designs are graphically depicted in Figure 3. The standard deviation is shown via error bars in Figure 3. Two KF composites were chosen that achieved the desired compressive strength of low stress grade concrete. The workability of the composite was not compromised with fibre integration. The results in Figure 3 demonstrate that fibre integration lowered the desired compressive strength. Integrating fibrous materials can create additional voids in the concrete matrix, resulting with a lower mechanical strength when under implied stress. However, the SF content (silicon dioxide) on the fibre walls consumes calcium hydroxide ($Ca(OH)_2$) which can mitigate degradation on the fibres. This has shown to enhance the composite compressive strength and enabled the two samples shown in Figure 3 to be acceptable in this study. SFKF5 and SFKF105 had 20 and 21 MPa at 28-days. The strength of the material was deemed compatible for low-stress concrete applications and therefore a comparison of the material for LCA was deemed appropriate. Figure 4 illustrates the SFKFs used in this study. As can be observed, the fibre content varies in size from approximately 10-36 μm. This is due to the variation of natural fibrous materials that undergo thermal and chemical treatments during the pulping process.

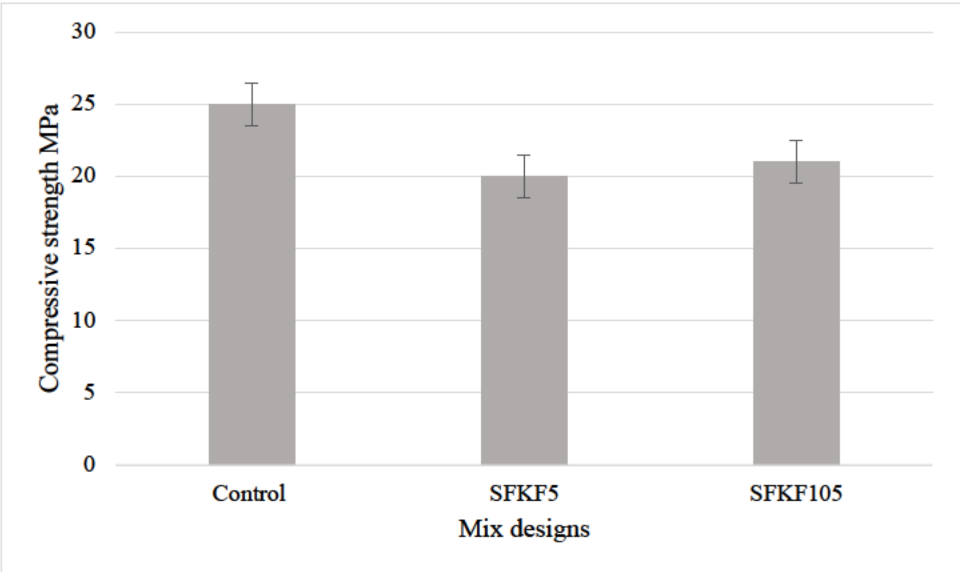

Figure 3 Compressive strength results

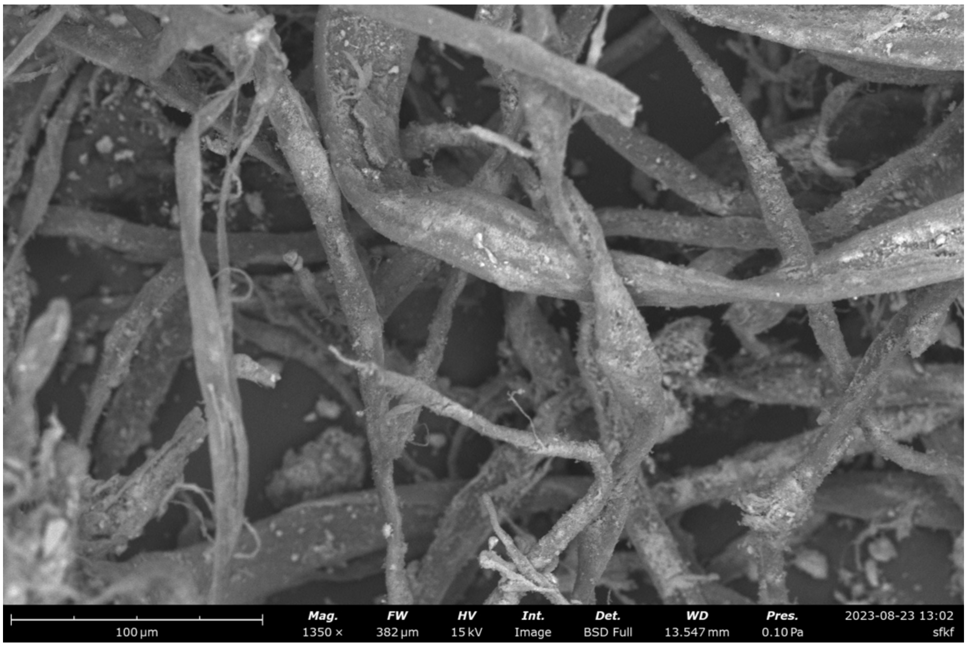

Figure 4 SEM image of SFKFs

4.2 LCA findings

The LCA primary objective was to compare the disparity of results against the environmental categories when integrating waste cardboard as a partial cement substitute in concrete materials. The initial LCA findings included transportation of materials to the concrete batching plant. This study included the city of Melbourne, Australia as a case study to exemplify the findings. Waste cardboard materials were sourced from a local resource recovery centre located 37Km from a concrete batching plant. All other constituent materials for concrete production were sourced 36Km from the nearest quarry. Table 7 demonstrate the results of the various mix designs against the impact categories. Table 8 demonstrates the difference of impact categories between the various mix designs and the control. 5% waste cardboard material integrated within concrete materials demonstrate a negative effect on TAP100. This is shown with SFKF5 having a 0.018075kg. It is important to note that the integration of 5% MK improves the TAP100. All fibre integrated mix designs demonstrated an improved GWP100. This shows integrating cardboard waste materials in concrete can inherently reduce the GWP by reducing cement requirements. The highest contributor toward TEP100 was the control with 658 kg 1, 4-DCB-eq. Again, the higher the percentage of cardboard waste reduced the negative effect of TEP100. The MEP100 was varied marginally compared to the control and fibre mix designs. HTP100 increased with 5% fibre integration. This is due to the extra processing requirement undertaken when converging waste cardboard into a fibrous product ready for composite integration. However, increasing fibre percentage from 5-10% reduces this negative impact marginally. ODP100 increases as the fibre percentage increases. This is primarily due to the increased processing requirements like the negative effect of HTP100. MRS improves when fibre is integrated at all levels compared to the control. However, when MK is utilised as a SCM, the negative effect increases past that of the control. This is because MK is dehydroxylated form of the clay mineral kaolinite [99]. Although abundant in some areas, limestone is still a more common material to source for cement production. Hence, the MRS is higher when MK is integrated in the composite design.

Table 7 LCA results of mix designs

Impact category	Unit	Control	SFKF5	SFKF105
Terrestrial acidification potential	Kg SO_2-eq	7.62E-01	7.80E-01	6.84E-01
Global warming potential	KgCO_2-eq	3.70E+02	3.60E+02	3.43E+02
Terrestrial ecotoxicity potential	Kg 1, 4-DCB-eq	6.58E+02	6.48E+02	6.38E+02
Marine eutrophication potential	Kg-N-eq	5.06E-03	5.17E-03	5.13E-03
Human carcinogenic toxicity potential	Kg 1, 4-DCB-eq	5.61E+00	5.64E+00	5.53E+00
Stratospheric ozone layer depletion potential	Kg CFC-11-eq	6.30E-05	6.20E-05	8.48E-05
Mineral resource scarcity	Kg Cu eq	1.26+00	1.42E+00	1.37E+00

Table 8 Potential environmental savings using KF in concrete

Impact category	Unit	SFKF5	SFKF105
Terrestrial acidification potential	Kg SO_2-eq	-1.81E-02	7.74E-02
Global warming potential	KgCO_2-eq	1.00E+01	2.65E+01
Terrestrial ecotoxicity potential	Kg 1, 4-DCB-eq	9.75E+00	2.05E+01
Marine eutrophication potential	Kg-N-eq	-1.04E-04	-6.49E-05
Human carcinogenic toxicity potential	Kg 1, 4-DCB-eq	-3.35E-02	7.73E-02
Stratospheric ozone layer depletion potential	Kg CFC-11-eq	1.01E-06	-2.18E-05
Mineral resource scarcity	Kg Cu eq	5.34E-02	-1.04E-01

Figure 5 graphically depicts the GHG emissions produced from the various constituent materials in the three mix designs. The emissions are of resource extraction and constituent material manufacturing ready for concrete integration but do not include the production or transportation of the novel concrete composites. As shown, the highest GHG emission producer is cement. The control had the highest percentage of cement and resulted in 335.8 kgCO_2-eq/m^3. SFKF5 and SFKF105 emitted 319.01 and 285.43 kgCO_2-eq/m^3, respectively. As the percentage of cement is reduced, the overall GHG emission is reduced. Cement attributes 90, 89 and 86% of total material GHG emissions for the control, SFKF5 and SFKF105 samples. The high percentage represents an opportunity to potentially reduce GHG emissions with just one constituent material. KFs have not been included in Figure 5 due to the uncertainty factors when producing the material for concrete application. The following section will demonstrate these material variables. 5% MK integration demonstrated 5.77 kgCO_2-eq/m^3. This is significantly less than 16.79 kgCO_2-eq/m^3 for 5% cement integration. Although MK can only be partially substituted for cement due to the reduction of mechanical properties, the material demonstrates environmental benefits, nonetheless.

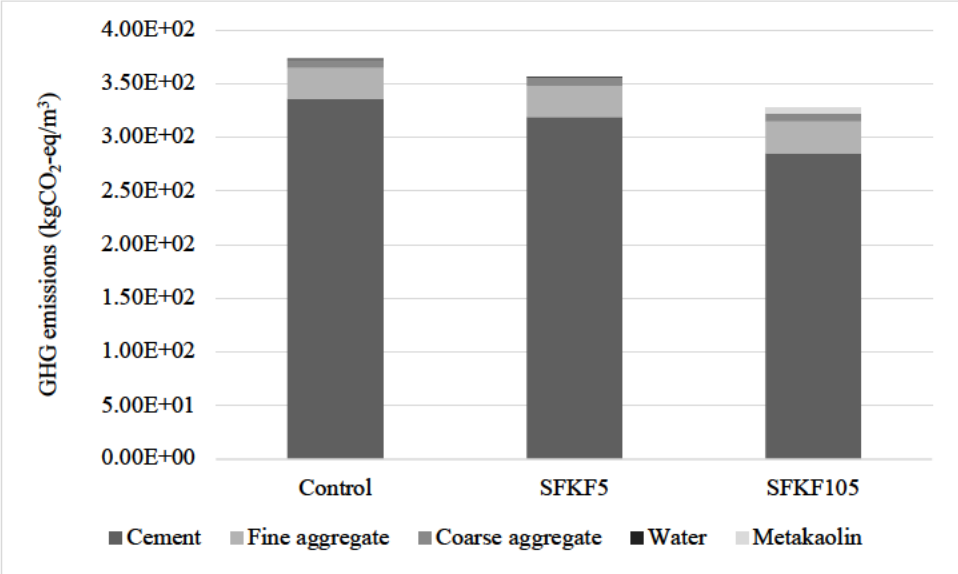

Figure 5 GHG emissions of concrete composites ($1m^3$)

4.3 Monte-Carlo simulation results

4.3.1 Transport emissions

The pre-production transportation is generally considered as a sensitive factor in the LCAs due to the high variable of possible distances and individual transport emissions [61]. Due to the uncertainty of this factor in future case studies, a MC simulation was conducted to analyse the influence of input parameters on the LCA outcomes. The probability simulation graphically depicted in Figure 6 demonstrates the calculations of the variable emission factor of a concrete truck transporting the raw materials to the concrete batching plant with a distance of 10- 150 km. Figure 7 illustrates the location of the raw materials to the concrete batching plant in Melbourne, Australia. The associated transportation emissions are shown in Table 6, with the maximum and minimum values accounted for during the triangular distribution to produce the most likely outcome. The MC simulation performed 10,000 iterations with a confidence level of 0.05. The lower and upper quartile limits of each box is represented on each side of the mean with the minimum and maximum values shown outside of the box. These values are then compared against each mix design to ascertain the GHG emission value that transportation has on the novel composite materials. It is important to note that the material emissions value has remained a constant to ascertain the true emission variability of transportation. For the MC simulation, $1m^3$ was kept desirable to transport as the value would only increase at the same incremental interval. As graphically depicted in Figure 6, the control demonstrates the highest GHG emission value compared to all fibre composites. This is primarily due to the higher cement percentage which has a higher total sample material emission value. The average travel and material emission range of the control, SFKF5 and SFKF105 are 375.28, 362.16 and 339.24 $kgCO_2$-eq/m^3.

As illustrated in Figure 6, there is a larger variable of GHG emissions produced for SFKF105 due to the additional transport requirements of raw materials. For example, the transportation of MK is an additional travel requirement, creating additional emissions. This is shown with a minimum and maximum of 320.89 and 367.44 $kgCO_2$-eq/m^3, respectively. SFKF5 had an average of 362.16 $kgCO_2$-eq/m^3, which was similar to the minimum of the control and the upper quartile of SFKF105. The interquartile range of each sample indicates there are smaller values produced from the 10,000 iterations produced from the MC simulation. This is primarily due to the smaller variability of the 10- 150km transport emissions produced and the constant value of material emissions. Although there is only an 9.6% emission reduction once the material arrives at the construction site for SFKF105, this value can be significant when there is a large concrete requirement. Fibre integration within the composite materials requires additional transportation services from the materials recovery centre; therefore it is expected that higher emissions will be produced from transportation with these materials. The control, SFKF5 and SFKF105 had a total material transportation of 108, 145 and 181km, respectively. The additional distance represents the extra materials required for the concrete batching process. However, minimising the cement requirement in composite materials offset the additional GHG emissions produced from transportation. It is important to note that the emissions produced when manufacturing KFs and the mixing of all constituent concrete materials can vary depending on the resources and location of the project.

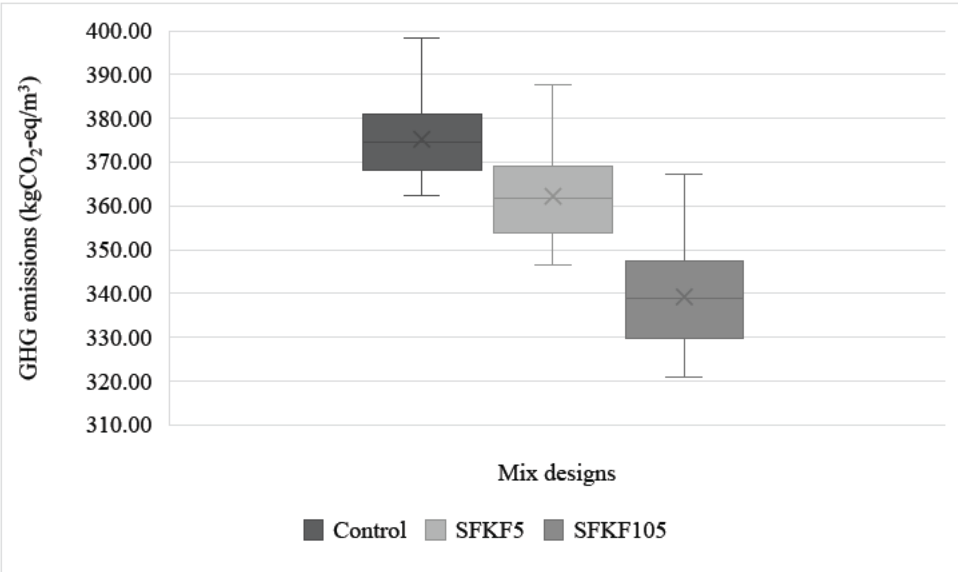

Figure 6 Transportation and material GHG emissions of 1m^3 concrete materials

Figure 7 Travel distances for materials to concrete batching plant

4.3.2 Energy emissions

The energy required to produce concrete materials is considered to be sensitive due to the high variable of energy requirement from different machinery for manufacturing [100]. Due to the uncertainty of this factor in future case studies, a MC simulation was conducted to analyse the influence of input parameters on the LCA outcomes. Figure 8 graphically depicts the variable GHG emissions for the material and the various machinery required to produce the novel composite materials. The MC simulation produced 10,000 iterations with a confidence level of 0.05. Figure 8 details the emissions produced for each type of machine over a variable of 60- 300 minutes. This is due to the uncertainty of user composite production. The machines selected were based on a laboratory scale of concrete and fibre production in line with the scope of the study. As 1m^3 is selected as the functional unit, integrating larger commercial machines would significantly alter the outputs conducted by the LCA. Moreover, a 60- 300-minute time-period was selected to represent a scalable time measurement. The GHG emissions shown in Figure 8 is derived from the MC simulation of the emission factors for the three energy sources. Each machine selected is consistent to use 10, 60 and 30% of gas, coal, and renewable energy sources. The findings illustrated in Figure 8 demonstrate the additional GHG emissions produced to manufacture the composite materials. As shown, there is a significant increase of GHG emissions produced for the novel composite materials when using waste cardboard. The control, SFKF5 and SFKF105 had an average of 525, 897 and 871 kgCO_2-eq/m^3. This represents a GHG emission increase of 42 and 39% for SFKF5 and SFKF105, respectively. This is due to the additional energy requirement when producing ready-made KFs for constituent material integration.

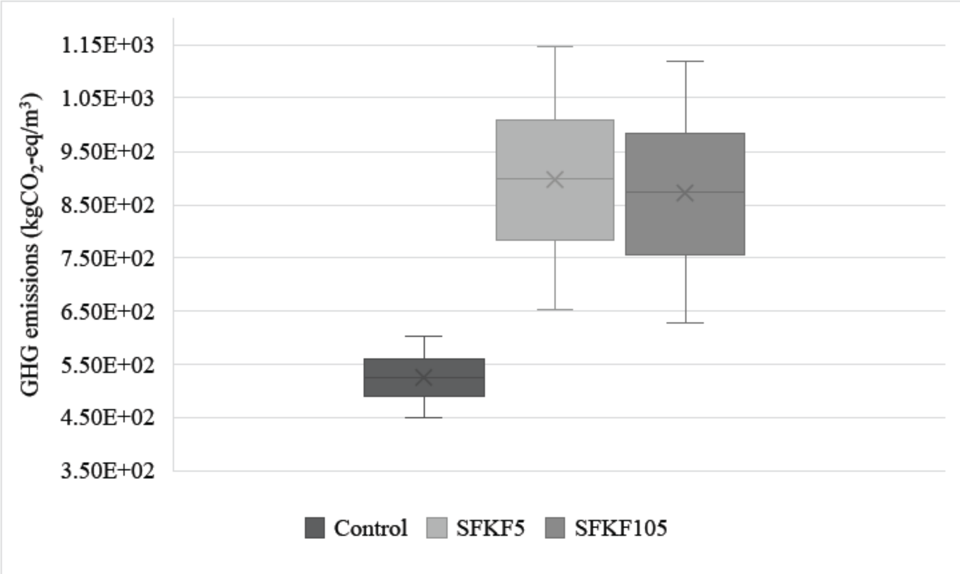

Figure 8 Material and manufacturing GHG emissions for $1m^3$ concrete

Figure 9 illustrates the total GHG emissions produced of all material, transport and energy requirements when manufacturing the composite materials. As shown, waste fibre composites still produce the most GHG emissions within the cradle to gate parameters. The control, SFKF5, and SFKF105 demonstrate an average of 572, 1023 and 997 $kgCO_2$-eq/m^3, respectively. Similar results are produced with the material and manufacturing emissions due to the constant raw material GHG emissions. However, the maximum and minimum values are varied due to the additional transportation requirements. These values extend the scope of the box plot and whisker total data set. SFKF105 has a lower emission value compared to SFKF5. This is due to the reduction of cement when replaced with MK. Despite the additional transportation requirements, small values of cement replacement can demonstrate GHG emission savings.

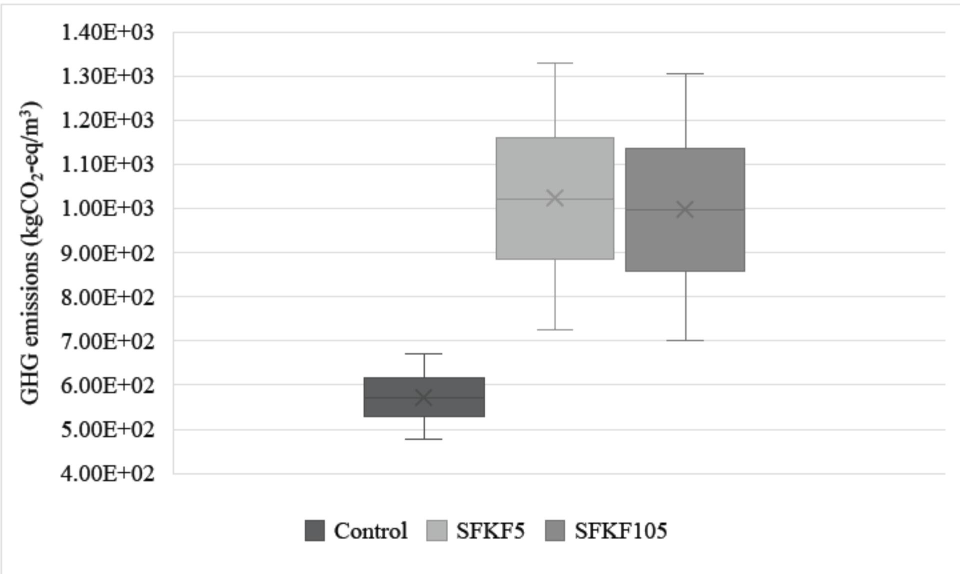

Figure 9 Total GHG emissions for $1m^3$ concrete

Figure 10 demonstrates the variation of fuel source to create the energy required to operate the machinery. As graphically depicted, the primary source of energy is from fossil fuel sources such as gas, brown and black coal. These sources represent 10, 10 and 50% respectively. However, the emission factor remained the same for the variations of coal and have been combined for graphical representation. There is 30% renewable energy sourced for the power of operational machinery. Although there is still a GHG emission factor, this value is significantly lower, as shown in Table 6. Figure 11 demonstrates that the cardboard shredder requires the most energy and ultimately produces the most GHG emissions. The interquartile ranges vary significantly for the various machinery due to the overall energy requirement of each machine. The concrete mixer and cardboard shredder require a high voltage power The increase of power supply requirements increases energy consumption, ultimately increasing the GHG emissions produced. The additional processing of waste cardboard increases the energy requirement and therefore increases the GHG emissions produced with an average of 575 $kgCO_2$-eq/m^3 over a 60–300-minute time-period. This is significant when the concrete mixer produces an average of 157 $kgCO_2$-eq/m^3. However, the critical factor of the energy required is derived from the energy source. As shown in Figure 10, fossil fuel energy is the primary source of power. Local governments worldwide are converting power supplies to greener and renewable energy sources [101]. Therefore, as the source changes to a green energy power supply, the additional power requirements of KFs will not demonstrate larger volumes of GHG emissions produced into the atmosphere.

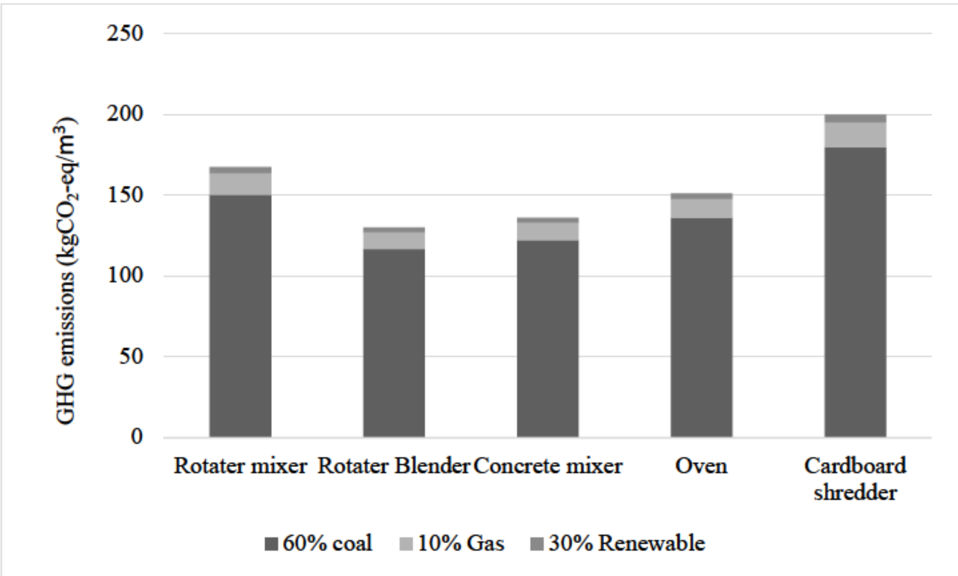

Figure 10 Variations of energy source and their average GHG emissions for 60- 300-minute usage

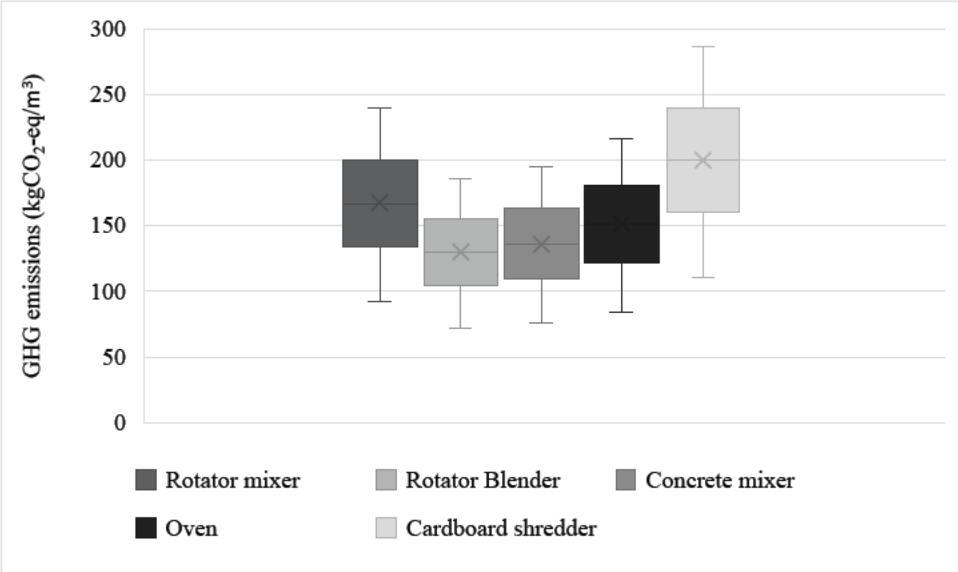

Figure 11 Equipment variation of GHG emissions produced for 60- 300-minute usage

5.0 Conclusion

To search for alternative methods of cardboard waste distribution, this study focused on the integration of waste KFs within low-stress grade concrete. This paper presented three key stages of methodology to evaluate the viability and environmental effects of the waste material in concrete. First, the mechanical strength of KFs as a partial cement substitute within concrete materials was analysed. Secondly, the life cycle assessment outputs on key impact categories were identified. Finally, a sensitivity analysis was conducted on the variability of GHG emissions produced from transport and energy usage. The mechanical performance demonstrated comparative compressive strength in low stress grade concrete, with 5% partial cement substitution using SF modified fibres. A matrix modified composite using MK was also deemed satisfactory for its strength characteristics when using 5% MK and 10% SF modified fibres. The life cycle assessment results revealed for $1m^3$ of SFKF105 had savings of 11%, 8%, 4% and 1% for TAP, GWP, TEP and HTP impact categories, respectively. SFKF5 revealed saving results of 3%, 2% and 4% for GWP, TEP and MRS impact categories respectively. The life cycle assessment of SFKF105 also showed increased negative environmental impacts on MRS, ODP and MEP impact categories. However, the negative impact on the environment in these impact categories is marginally different. Although the additional processing of KFs increased the HTP in SFKF5, this was offset in SFKF105 due to the integration of MK and the increased fibre percentage. The Monte-Carlo simulation provided further information on the variability factors when integrating bespoke materials into the mix design. Initially, the extra requirement of transportation for more constituent materials increased the GHG emissions. SFKF105 revealed an average of 29 $kgCO_2\text{-eq}/m^3$ for material transportation to the concrete batching plant. Whereas transportation of the control produced 59% less GHG emissions. However, this was mitigated by the material emissions originally produced to batch traditional concrete. Ultimately, SFKF105 demonstrated the lowest GHG emissions from cradle-to-gate and the control produced the highest amount of GHG emissions for material and transportation factors. A sensitivity analysis on the energy sources and the operational machinery was also conducted. The results revealed additional energy requirements for the processing of KFs. Due to the additional manufacturing processes to convert waste cardboard into KFs, the original GHG emission material and transportation savings were then eliminated. Moreover, energy sources were compared to see the effect of fossil fuel-based systems and potential GHG emission savings when using renewable energy in the future. This study examined the multiple considerations required when evaluating and classifying what a green material is. The contributions of this study highlight the various considerations required when researching novel materials to be used in cementitious composite systems. As shown, the variability factors can affect what is initially thought to be a sustainable material alternative and potentially create additional negative impacts. However, it is important to note that reducing landfill of waste materials will always be environmentally beneficial. As renewable energy becomes the primary source, opportunities will be more prominent for bespoke materials to be used in the construction industry.

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7.3 Concluding remarks

This chapter focused on the integration of KFs within low-stress grade concrete. The mechanical results demonstrated comparative compressive strength with 5% partial cement substitution using SF modified fibres. A matrix modified composite using MK was also deemed satisfactory for its strength characteristics when using 5% MK and 10% SF modified fibres. The LCA results revealed savings of 11%, 8%, 4% and 1% for TAP, GWP, TEP and HTP impact categories, respectively, for one cubic metre of SFKF105. SFKF5 revealed saving results of 3%, 2% and 4% for GWP, TEP and MRS impact categories respectively. The LCA of SFKF105 also showed increased negative environmental impacts on MRS, ODP and MEP impact categories. However, the negative value in these impact categories is marginally different. Although the additional processing of KFs increased the HTP in SFKF5, this was offset in SFKF105 due to the integration of MK and the increased fibre percentage. The Monte-Carlo simulation results provided further information on the variability factors when integrating bespoke materials into the mix design. Initially, the extra requirement of transportation for more constituent materials increased the GHG emissions. SFKF105 revealed an average of 91.94 kgCO_2-eq/m^3 for material transportation to the concrete batching plant. Whereas the control had 59% less GHG emissions produced. However, this was then offset by the material emissions originally produced to extract and create ready-made materials for concrete production. Ultimately, SFKF105 demonstrated the lowest GHG emissions from cradle-to-gate and the control produced the highest amount of GHG emissions. A Monte-Carlo simulation on the energy sources and the operational machinery was also conducted. The results revealed additional energy requirement for the processing of KFs, however, the highest GHG emission producer was from the concrete mixer. Energy sources were compared to see the effect of fossil fuel-based systems and potential GHG emission savings when using renewable energy in the future.

Chapter 8 – Economic and environmental optimisation

8.1 Introduction

In this chapter, a nondominated sorting genetic algorithm (NSGA-II) was utilised to optimise the economic cost and environmental emissions of concrete incorporating recycled waste cardboard as a partial cement substitute. This chapter presents two key stages of methodology to evaluate the viability and environmental and economic effects of the waste material in concrete. Firstly, the output from the multi-objective optimisation solutions produce the most optimal mix designs for the corresponding objective functions. Secondly, the mix designs produced will be mechanically analysed to determine their compressive values and potential application within the construction industry.

The following paper is included in this chapter:

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2. CANDIDATE DECLARATION

I declare that the publication above meets the requirements to be included in the thesis as outlined in the HDR Policy and related Procedures – policy.vu.edu.au.

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3. CO-AUTHOR(S) DECLARATION

In the case of the above publication, the following authors contributed to the work as follows:

The undersigned certify that:

1. They meet criteria for authorship in that they have participated in the conception, execution or interpretation of at least that part of the publication in their field of expertise;
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Economic and environmental optimisation of waste cardboard kraft fibres in concrete using nondominated sorting genetic algorithm

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A R T I C L E I N F O

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A B S T R A C T

The depletion of natural resources is accelerating due to increased construction activities across the world. The search for alternative materials used in concrete is becoming a prominent measure to reduce greenhouse gas (GHG) emissions in the construction industry. Cement in concrete produces 4–8% of all GHG emissions annually worldwide. Alternative materials such as waste cardboard provide an opportunity to be utilised further in such applications. This study focuses on using nondominated sorting genetic algorithm (NSGA-II) to optimise the economic cost and GHG emissions of concrete incorporating recycled waste cardboard as a partial cement substitute. The multi-objective optimisation solutions produced three key regions that corresponded with trade-offs between the lowest costs, carbon emissions or a combination of both. Region 1 was the most economical but created the most carbon emissions. Region 2 was a compromise between both economical, and environmental factors and region 3 demonstrated the lowest carbon emissions but also produced the highest costs. These regions produced three mix designs which were experimentally analysed for their compressive strength. The results indicated a cost increase of 17, 27 and 36% for three key optimised regions. This is primarily due to increased material costs and further manufacturing requirements when converting waste cardboard into kraft fibres (KFs). Increasing the amount of KFs then reduced the compressive strength from 36 to 56%. However, there was a 7, 18 and 28% GHG emission saving when KFs partially replaced cement.

1. Introduction

The United Nations Paris agreement in 2015 committed to transforming development trajectories towards sustainability. This plan is to ensure global warming does not rise more than 1.5 °C above pre-industrial levels (Nations, 2022). To meet these goals, the United Nations climate action plan requires detrimental environmental emissions to be reduced by 45% by 2030, with a net zero target by 2050. Greenhouse gas (GHG) concentrations reached new highs in 2020, with globally averaged mole fractions of carbon emissions exceeding 410 parts per million (Nations, 2023). Failing to reach these targets may result in increased global temperatures and further catastrophic environmental events worldwide (Zhang et al., 2022a). The building and construction industry is often considered non-sustainable due to excessive generation of negative environmental impacts. This is evident with large creations of waste and excessive GHG emissions produced worldwide from building and construction related activities. A large contributor of these GHG emissions are from natural resource extractions for construction material productions (Appiah et al., 2023). Concrete is the most utilised material in the construction industry and cement production is responsible for 4–8% of annual carbon dioxide (CO_2) emissions worldwide (Shobeiri et al., 2023). Therefore, the search for alternative materials to supplement cement in concrete production is becoming increasingly important. Research studies have focused on developing sustainable concrete by integrating waste to reduce virgin raw materials usage in concrete (Xu et al., 2022; Abera, 2022; Purohit et al., 2022; Kanagaraj et al., 2022). Waste materials such as fly ash (FA), ground blast furnace slag (GBFS) and glass have been extensively explored to partially supplement cement, fine and coarse aggregate in concrete (Afroz et al., 2022; Albitar et al., 2017; Sunayana et al., 2018; Al-Awabdeh et al., 2022; Elaqra et al., 2018). However, vast accumulation of different waste materials across all economic sectors is inherently challenging due to increased costs and limited land availability for recycling and landfill requirements. The circular economy framework includes the elimination of waste, pollution, and circulate materials for the regeneration of nature. This framework is becoming embedded in

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research and development objectives which is supported across local Governments (Dagilienė et al., 2021). This has directed the use of alternative materials in concrete, such as cardboard, that have previously been seldom considered. Cardboard waste materials have often been overlooked due to the inherent problems associated when using a product derived from natural materials. In 2021, cardboard waste in Australia had a 61.2% resource recovery rate, with 55% being recycled and 6.2% directed toward energy recovery. Subsequently this equated to 2.2 million tons (Mt) of cardboard waste in landfills (Pickin et al., 2022). This value is significant due to the volumetric size cardboard waste creates, with 1 ton approximately equates to 1.6 m^3 in size. The challenge of disposal with cardboard waste is further shown as on average, 48% of residential waste in a recycling bin is paper and cardboard materials (Pickin et al., 2022).

Previous research studies have emphasized that replacing virgin materials with waste such as coffee cups can provide environmental benefits and promote sustainable construction practices. The findings demonstrated a possible 10.74% reduction of GHG emissions when fine aggregates are substituted with shredded coffee cup waste (Sandanayake et al., 2022). Mondal et al. (2019) experimentally investigated the use of thermoplastic waste to produce bricks and masonry units (Mondal et al., 2019). Their research improved the thermal resistivity and reduced the density of the material, creating a lightweight cladding option. Other researchers showed the recycling of on-site aluminium waste can reduce carbon emissions by 45% for total building demolition works (Wang et al., 2018). Nascimento et al. (2020) integrated 15–40% waste rubber tyres within pavement systems and the results exemplified that 15% waste utilisation can provide both environmental and economic benefits (Nascimento et al., 2020). Fernando et al. (2021) performed a comparative life cycle assessment (LCA) and life cycle costing (LCC) between FA geopolymer concrete, alkali-activated concrete, and Ordinary Portland cement (OPC) concrete (Fernando et al., 2021). Their research exhibited an increase of 12% and 48% of GHG emissions and initial costs for the FA concrete and geopolymer alkali-activated concrete respectively. This research demonstrates that performing LCA and LCC can provide initial data on the effect of novel concrete materials, however, optimising the mix design is a critical step to avoid unnecessary experimental testing. Heidari et al. (2020) compared the environmental and economic benefits of using concrete materials instead of asphalt paving systems (Heidari et al., 2020). Their research showed a 35% increase of costs when using concrete however, a reduction of approximately 2 million tons of CO_2 emissions and a reduced annual energy consumption of 700,000 GJ when using asphalt paving systems. This study further exemplifies the requirement to perform multi-objective optimisation techniques to display the most sustainable solutions for all objective criteria. Chen et al. (2022) discuss the use of urban waste materials within the development of sustainable concrete. Their research used plastic and glass waste within concrete, highlighting that it is a feasible option when micro-mechanical problems leading to strength degradation have been solved (Chen et al., 2022a). Their findings also detailed that research assessments on the social, economic, and environment have not been transferable to impede large-scale implementation within civil engineering projects. A systematic bibliometric assessment was carried out by Sandanayake et al. (2020) determining key waste materials used in concrete (Sandanayake et al., 2020a). Their findings demonstrated that plastic, glass, FA, GBFS and construction and demolition waste were prominent to replace virgin materials. However, only plastic and glass were common waste materials potentially sourced from the public sector. Researchers have demonstrated that cardboard waste is seldom considered in concrete and mortar products, and that further research is required to promote the waste material in bespoke applications (Haigh et al., 2021).

Research has shown the use of cardboard waste can be integrated within concrete and cementitious materials however, there are additional fibre and matrix modification requirements (Haigh et al., 2022a). This is due to the cellulose chains of kraft fibres (KFs) degrading significantly when exposed to high alkaline environments (Coutts, 2005). Khorami et al. (2013) experimentally investigated waste cardboard using 1–14% KF integration within fibre cement boards (Khorami et al., 2013). Their research findings showed that flexural strength is improved by 250% when 8% KF is integrated alongside 10% limestone powder and 3% nano-silica fume. The additional additives reduced degradation on the fibre walls due to the reduction in OPC. Their findings were based upon trial-and-error investigations. Booya et al. (2018) conducted experimental research on the durability of KF integration within concrete (Booya et al., 2018). The results demonstrated that 2% mechanically and chemically treated fibres can enhance durability properties of concrete composites. Other research has been directed toward the thermal performance, microstructural characteristics, surface fatigue and processing effects of KFs when integrated within cementitious composite materials (Haigh et al., 2022b; Mohr et al., 2006; Savastano et al., 2009; Tonoli et al., 2013). These studies have focused on achieving mechanical objectives while environmental and economic research of KF composites have been seldom considered.

The environmental benefits of waste integration within concrete materials if often measured using LCA. However, the sustainability of a material is often linked with the triple bottom line approach which aims to achieve economic, environmental, and social benefits (Sahu et al., 2023). To ensure transition from research and development into marketable products, industry stakeholders require tools to utilise the information derived from environmental and economic data (Agarwal et al., 2018). Material selection is often based upon the balance of project cost constraints and impeding negative environmental effects. Several studies have attempted to develop tools to measure emissions generated throughout construction phases (Sandanayake et al., 2019; Kayaçetin et al., 2023; Kim et al., 2023; De Wolf et al., 2023). However, these tools do not consider material embodied emissions alongside material costs. Therefore, this study focuses on the optimisation of KFs derived from waste cardboard within concrete materials. Moreover, it is important to find alternative methods to utilise waste cardboard as it is not a common material reused in building and construction. The ability to transform waste cardboard to its main constituent material (KFs), demonstrates the agility of the material for further use. Despite the inherent challenges when using natural materials, alternative methods of integration need exploring. Optimising the amount of KFs within concrete can provide sustainable benchmarks and criteria for further research development. The feasibility of a marketable product using waste materials requires exploration of economic and environmental impacts. Even a small percentage of waste integration can provide beneficial results that promote sustainability of the industry. Optimising the integration of KF concrete between economic and environmental benefits is required prior to the classification and promotion of a sustainable concrete material. This study aims to highlight a comprehensive and holistic understanding of the consequences when integrating KFs in the material development of concrete and mortar materials. Market uptake of waste materials in concrete mixes are limited with cost implications therefore, the study focus is innovative from the perspective to highlight both economic and environmental implications when using these materials. The outcomes can foster a more sustainable approach to the economic prosperity, environmental responsibilities, and social equity of stakeholders within the building and construction industry.

2. Background and research significance

The building and construction industry is a primary contributor to negative environmental impacts, exhibiting excessive resource requirements. Considering the life cycle of materials, significant energy consumption is required from procurement to final disposal. Each stage of material handling can contribute to the creation of carbon emissions, leading to 15% of a buildings total CO_2 output (Sandanayake et al., 2018; Fan et al., 2018; Elkhaldi et al., 2022). Overuse of virgin materials compounds the issue of depleting natural resources and amplifying negative environmental impacts. The substitution of virgin materials with discarded waste products effectively targets the dual concern of excessive waste generation and the consumption of virgin resources.

Material researchers and engineers are challenged to promote the use of alternative materials in concrete due to the large amount of inherent experimental investigations required. The use of destructive and expensive mechanical testings are common to determine viable mix designs for further promotion. Utilising the multi-objective optimisation (MOO) method can reduce avoidable costs and time-consuming tests by determining the most appropriate mix designs for further exploration. The objective of this research is to determine the most optimal amount of KFs within concrete that provide a balance between the economic and environmental benefits. Therefore, the solutions provided by the MOO can promote sustainable benchmarks for waste cardboard concrete integration. Table 1 represents a summary of the reviewed studies on using waste materials in the production of concrete and mortar materials. The results indicate that most optimisation studies have focused on the integration of recycled fine and coarse aggregates. Table 1 also demonstrates that the optimisation of mix designs to maximise mechanical strength characteristics is often the focus of research studies. However, economic restrictions often hinder the transition of novel materials into marketable products. Moreover, the current study attempts to determine the most optimal mix design that minimises negative environmental impacts and reduces costs when using KFs within concrete materials.

Research studies have been conducted utilising various methods of MOO techniques. Liu et al. (2023) conducted research to optimise the mechanical, economic and environmental benefits when integrating recycled aggregate within concrete materials (Liu et al., 2023). The research utilised competitive mechanism-based multi-objective particle swarm optimisation (CMOPSO) and provided good performance when integrating three design scenarios for optimising compressive strength. However, the final optimal solutions in the Pareto set indicated that the four objectives of compressive strength, cost, carbon emission and energy consumption are mutually restricted and cannot be optimal at the same time. Amiri et al. (2022) focused their research toward optimising the mix design when integrating coal waste in concrete (Amiri et al., 2022). This was measured based upon compressive strength and water absorption results using desirability function method as the optimisation technique. The results determined the most optimal material input ratios that provided the acceptable strength and absorption characteristics. Zhang et al. (2020) also utilised MOO via the use of machine learning and metaheuristic algorithms to determine acceptable strength, cost and plasticity levels of concrete materials (Zhang et al., 2020). Their findings indicated that backpropagation neural network has a better performance to determine strength. Whereas the random forest algorithm has a more accurate prediction of slump. However, the Pareto fronts of the bi-objective mixture optimisation problem for high performance concrete was successfully obtained by the MOO model. This research indicates that for bi-objective and tri-objective mixture optimisation problems, the MOO model can serve as a design guide to facilitate decision making with novel composite materials. This is also shown with Zamir Hashmi et al. (2022) that validated experimental data utilising marble waste aggregates and stone dust in concrete materials (Zamir Hashmi et al., 2022). Their research findings demonstrated the optimisation performed through response surface methodology (RSM) predicted the composites to yield the most strength with an error of less than 5%. Moreover, utilisation of the MOO model can yield beneficial future research direction as the most optimal mix design can be provided based upon the research objectives.

Limited research has focused on the experimental studies of KFs within concrete materials however, further promotion is required to highlight the associated benefits with using the waste material as a partial cement replacement. LCA and LCC assessments have been seldom considered of KF concrete therefore, this study aims to address this research gap and provide the most optimal mix design that provides both economic and environmental benefits. The research will also aim to promote the integration of KFs in concrete which can enhance the circular economy concepts with common waste materials.

Table 1
Previous optimisation research using waste materials in concrete.

Waste material	Optimising parameter	Ref
Recycled concrete aggregate	Mechanical	Shaban et al. (2021)
Recycled concrete aggregate	Economic, environmental, and mechanical	Liu et al. (2023)
Ground blast furnace slag and metakaolin	Economic, environmental, and mechanical	Zhang et al. (2022b)
Waste plastic	Economic and mechanical	Shiuly et al. (2022)
Waste coffee cup	Economic and environmental	Sandanayake et al. (2022)
Steel slag powder	Mechanical	Fan et al. (2021)
Waste rubber	Economic, environmental, and mechanical	Mohammadi Golafshani et al. (2021)
Waste plastic and fly ash	Mechanical	Adamu et al. (2021)
Waste glass powder	Economic	Sun et al. (2022)
Coal waste	Mechanical	Amiri et al. (2022)
Ceramic waste	Mechanical	Chokkalingam et al. (2022)
Waste glass and condensed milk can fibres	Mechanical	Ahmed et al. (2022)
Waste oyster shell powder	Environmental and mechanical	Han et al. (2022)
Waste nylon	Mechanical	Arjomandi et al. (2023)
Waste nylon	Mechanical	Nematzadeh et al. (2021)
Waste clay bricks	Mechanical	Abdellatief et al. (2023)
Recycled concrete and recycled steel	Economic and mechanical	Sahraei Moghadam et al. (2021)
Recycled concrete aggregate	Economic, environmental, and mechanical	Rashid et al. (2020)
Waste tyre rubber	Economic, environmental, and mechanical	Zafar et al. (2022)

3. Materials and methodology

The research methodology aims to optimise the KF concrete mix design. The objectives to determine via optimisation will include minimisation of costs and minimisation of GHG emissions. The results from the multi-objective optimisation will be analysed and key regions will be determined that highlight important trade-offs between the economic and environmental objectives of this study. Experimental analysis will verify potential uses of the novel composite based on the provided optimal mix design solutions. Fig. 1 illustrates the research methodology.

3.1. Materials and testing procedure

The materials included within the mix design of the composite material are cement, KFs, metakaolin (MK), silica fume (SF), fine, coarse aggregate and water. KFs were used as a partial cement replacement. Transforming KFs from waste cardboard requires multiple steps to ensure sufficient agglomeration of all constituent materials. KFs were reduced to a pulp material then combined with a SF slurry. The moisture was then removed from the pulp material and subjected to a rotator blender as a fibre dispersion technique. SF was integrated within the composite as a surface modification technique on the KF walls. The SF applied to the KFs conforms with the Australian Standard AS/NZS 3582.3 (AS/NZS-3582.3, 2016) specification of Silica Fume used in cementitious mixtures. MK was used a partial matrix modifier due to the enhanced durability factors of natural fibres within composite materials (Haigh et al., 2023). The MK used conforms with the ATSM C-618 (ASTM-C-618, 2019), Class N specifications for natural and calcined pozzolans. Original Portland cement (OPC) was used as the primary

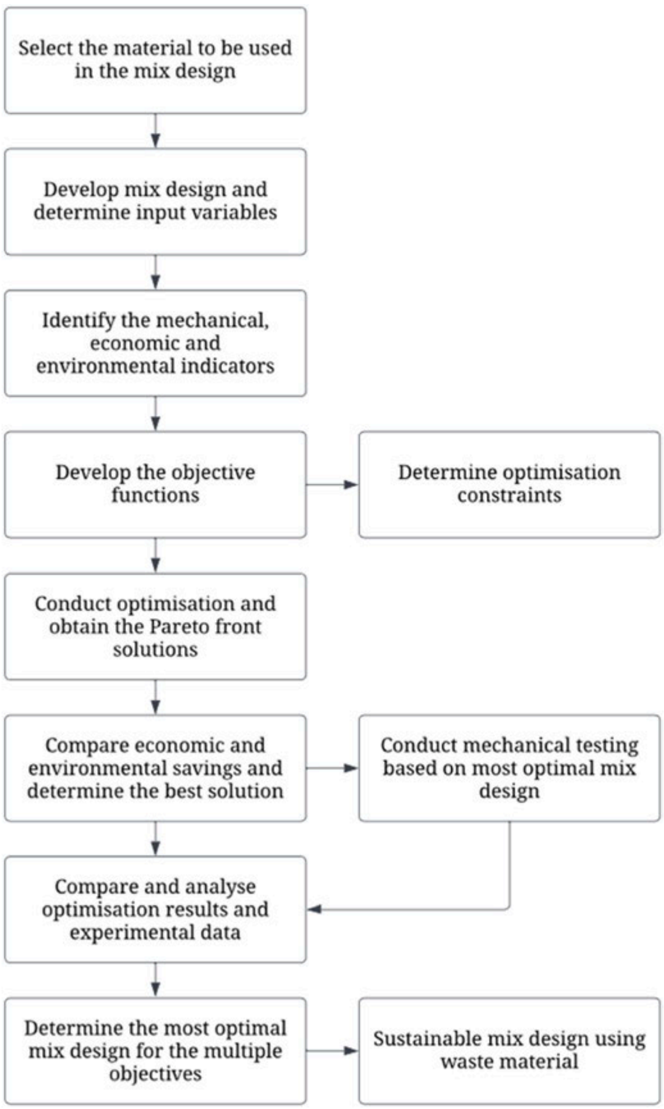

Fig. 1. Research methodology.

constituent for the pozzolanic reactivity of the concrete specimens. The material conforms with AS/NZS 3972 (AS/NZS-3972, 2010). Table 2 summarises the material properties of the pozzolanic materials used in this study. Locally available coarse and fine aggregate were applied, conforming to AS/NZS 1141.6.2 (AS/NZS-1141.6.2, 2016) and AS/NZS 1141.5 (AS/NZS-1141.5, 2000) respectively. Regular potable tap water was used throughout the preparation of the experimental mix design. Table 3 shows the base mix design designed for 1 m^3 of concrete materials.

A mortar mixer was used to prepare concrete specimens. Initially the raw materials were dry mixed before adding water. Upon completion of the mixing process, concrete was poured into 200 x 100 cylindrical moulds in three separate layers. With each layer, a steel rod compacted the layers twenty times to maintain uniform compaction. The specimens were prepared at a room temperature of 20 °C for 24 h, before the application in water baths. The compressive strength test was performed in accordance with AS 1012.9 (AS/NZS-1012.9, 2014) with a load rating of 20 MPa/minute. The compressive values were measured at the 28-day interval, with an average recorded of three samples for each mix design. Compressive testing was conducted on the Matest C088-11 N Servo-Plus evolution testing machine and Cyber-Plus evolution data acquisition system.

Table 2
Chemical composition of pozzolanic materials.

Chemical	Material component %		
	MK	SF	OPC
SiO_2	54–56	≥ 75- <100	19–23
Al_2O_3	40–42		2.5–6
Fe_2O_3	<1.4		
TiO_2	<3.0		
SO_4	<0.05		
P_2O_5	<0.2		
CaO	<0.1		61–67
MgO	<0.1		
Na_2O	<0.05		
K_2O	<0.4		
L.O.I.	<1.0		
Silica, amorphous, fumed, cryst. -free		≥0.3- <1	
$CaSO_4.2H_2O$			3–8
$CaCO_3$			0–7.5
Fe_2O_3			0–6
SO_3			1.5–4.5

3.2. Multi - objective optimisation methodology

The optimal performance of the building and construction of structures can often be conflicting with cost and GHG emission targets. The MOO technique is critical to determine several objectives simultaneously over a feasible data set that are often conflicting. As there is no unique solution that is optimal for all objectives, the MOO requires a trade-off among all objectives to determine the most feasible solution (Afshari et al., 2019). As compared to single objective optimisation, MOO can resolve existing optimisation problems with multiple objectives. The solutions produced from the MOO technique must not be dominated by other feasible alternatives. Therefore, the Pareto-optimal set is most favourable because the set of feasible alternatives are not dominated by other feasible points. The Pareto-optimal set is almost never a single point, rather a set of solutions that can be applied to real-world scenarios (Sandanayake et al., 2022). Nondominated sorting genetic algorithm (NSGA-II) is a common heuristic algorithm to solve two objective functions with limited constraints and variables. NSGA-II uses Pareto-based ranking and niching techniques together, optimising objective functions containing discrete and continuous variables (Sandanayake et al., 2020b).

NSGA-II can handle multiple conflicting objectives, maintain diversity among solutions, and provide a set of Pareto-optimal solutions. Advantages of NSGA-II include diversity preservation, scalability, and convergence to a well distributed Pareto front. This distribution provides decision makers with a range of trade-off solutions enabling key stakeholders to select solutions from the determined trade-offs (Bailey et al., 2023). Chen et al. (2022) utilised NSGA-II to determine the MOO of concrete durability and economic factors (Chen et al., 2022b). Their findings detailed the most optimal mix proportion for concrete considering the two objective functions (durability and costs). Huang et al. (2023) also utilised NSGA-II in conjunction with tree-based ensemble methods for geopolymer concrete (Huang et al., 2023). Their findings determined the trade-offs between strength, costs, and environmental emissions of 676 published mix designs. The developed framework assisted both their bi-objective and tri-objective optimisation problems. Therefore, NSGA-II is selected in the current study to determine the optimum composition of concrete containing KFs with reduced costs and environmental impacts. To minimise the random effect of the optimisation results in NSGA-II, each of the optimisation test problems are solved 100 times. The algorithms have been coded using Python because

Table 3
Base mix design for 1 m^3 of concrete (kilograms).

Water	Fine aggregate	Coarse aggregate	Cement
210	733	1100	355

it can incorporate various function sets.

3.2.1. Constraints and design variables

The main objective of this study is to compare the environmental and economic savings when substituting a percentage of cement with cardboard waste within concrete and mortar materials. To increase the mechanical feasibility factors of KFs within a cementitious composite, matrix and surface modifications are required (Haigh et al., 2021). Therefore, there are additional materials required to substitute a percentage of cement with the waste material. However, it is important to note that all materials to be used as a partial cement substitution cannot exceed the total cement requirement of the control. The following variables are considered to determine the linear constraints.

- Quantity of cardboard waste (KF)
- Quantity of metakaolin (MK)
- Quantity of Silica Fume (SF)

Moreover, the following equations are used to determine the linear constraints.

$$KF + MK + SF + CE = 355 \text{ kgs} \quad (1)$$

$$SF \leq 0.02\ KF \quad (2)$$

Where KF is kraft fibres, MK is metakaolin, SF is silica fume and CE is cement. The total amount of cement in the mix design is 355 kgs. Therefore, total cement substitution with all other material must not exceed 355 kgs. Surface modification is applied using SF on the KFs at a maximum 2% loading of the total amount of KFs. This has been shown to be sufficient as a fibre modification technique (Haigh et al., 2022a). The mix design optimisation problem consists of discrete and continuous variables based on the objective function. For example, the maximum amount of cement replaced by KFs is based upon the desired strength characteristics. Previous research has shown increasing the amount of KFs and MK in concrete beyond 20–25% respectively, significantly reduces the compressive strength (Haigh; Kalpana et al., 2020; Hodhod et al., 2020). Therefore, as shown in Table 4, the upper and lower bounds of KF and MK are set at the former values.

To ascertain the cost and emission factors for KFs on a laboratory scale, the following equations were applied. First, the KF material cost. Where TC is transport, EEC is the equipment energy cost, EC is the equipment cost and LMC is the labour manufacturing cost based upon an 8-h period. These costs are then divided by the kilograms manufactured per hour.

$$(TC + EEC + EC + LMC) / \text{kgs per hour} \quad (3)$$

Secondly, the GHG emission factor for the KF material.

$$(TE + EE) / \text{kgs per hour} \quad (4)$$

Where TE is the transport emissions and EE is the equipment emissions. The total of these values is based on an 8-h period then divided by kilograms manufactured per hour. To ensure the optimised solutions are viable for further exploration, the constraints and assumptions must be identified to ensure the optimisation algorithm is discontinued beyond the desired limits.

Table 4
Upper and lower bounds in kilograms.

KF Upper bound	72
KF lower bound	17
SF Upper bound	1.44
SF lower bound	0.34
MK Upper bound	88
MK lower bound	17

- The total sum of KFs and MK is equal to the total cement requirement of the concrete materials.
- The maximum amount of KF and MK integration is set at 20% and 25%, respectively.
- The control sample remains unchanged of virgin materials to ensure adequate comparison of strength targets are met.
- Transportation costs are calculated as a discrete value based on the distance of travel required.
- Transportation distance from batching plant to construction site is based on 35 km from real-world application in Melbourne, Australia.
- Electricity costs are sourced from current rates of electricity provided from local providers.
- Costs and emission factors of KFs are derived from laboratory process.
- Costs and emission factors of commercial production are used for remaining constituent materials.
- KF material production is assumed to be 100 kg per hour on a laboratory scale.
- System boundary of this study is the cradle-to-gate process.

3.3. Cost calculation equations for material production

The costs associated to produce the 1 m^3 of concrete materials are divided into three major components, cost of labour, cost of materials, and cost of equipment. The costs associated are based on a laboratory production. The production time is assumed to be 1 h based on previous research (Zeyad et al., 2020; Carigi et al., 2023; Hai-Thong Ngoa et al., 2017; Revilla-Cuesta et al., 2023).

3.3.1. Cost of labour

Labour manufacturing cost (LMC) is usually charged via hourly rate. The following equation determines the LMC in AUD, where *LR* is the labour rate and *T* is the time.

$$LMC = LR * T \quad (5)$$

3.3.2. Cost of materials

Raw material cost (RMC) is charged as per unit cost and can be estimated from the following equation. Where FA is the fine aggregate, CA is the coarse aggregate, CE is the cement, WA is the water, MK is metakaolin, SF is the silica fume and KF is the kraft fibres.

$$RMC = FA + CA + CE + WA + MK + SF + KF \quad (6)$$

3.3.3. Cost of manufacturing equipment

There are two parts to the cost of manufacturing equipment. Firstly, the hiring cost of the equipment (EC) and secondly, the equipment energy cost (EEC). The following equations determine the total cost of manufacturing equipment. Where CM is the concrete mixer, OC is the oven cost, CS is the cardboard shredder, BM is the blender mixer, RM is the rotator mixer and T is the time to manufacture the composite materials. For energy costs, the equipment is the capacity of the machinery in kWh then multiplied by time for electrical costs. The electricity rate (ER) is derived from current electrical prices at the time of this study.

$$EC = (CM + OC + CS + BM + RM) / T \quad (7)$$

$$EEC = ((CM\_kWh + OC\_kWh + CS\_kWh + BM\_kWh + RM\_kWh) * T) * ER \quad (8)$$

3.3.4. Transport costs

There are three parts to create the total transport cost (TC). The following equations determines these costs. Where FC is fuel costs, VE is the vehicle costs and DR is the driver costs.

$$TC= FC + VE + DR \tag{9}$$

3.3.5. Total cost calculation

The total cost calculation (TCC) is determined using the following equation.

$$CC = LMC + RMC + EC + EEC + TC \tag{10}$$

3.4. Calculation equations for greenhouse gas emissions

The GHG emissions associated to produce the 1 m^3 of concrete materials are divided into three major components, material emissions, production emissions and transport emissions. The emissions associated are based on a laboratory production. The following references were used to determine the relative GHG emissions (Maddalena et al., 2018; Australian_Government_Clean_energy_regulator, 2023; Environmental Protection Agency, 2014; Our_world_in_data, 2023; IEA Emission factors, 2022; Clavreul et al., 2012).

3.4.1. Material embodied emissions

The material embodied emissions (MEE) is calculated for all materials to be used within the mix design. The material quantities are multiplied by the emission factor. Where ef is the emission factor and ^{q} is the quantity. FA is the fine aggregate, CA is the coarse aggregate, CE is the cement, WA is the water, MK is metakaolin, SF is the silica fume and KF is the kraft fibres. All materials are calculated per kilogram.

$$MEE = FA^{q}*e_f + CA^{q}*e_f + CE^{q}*e_f + WA^{q}*e_f + MK^{q}*e_f + SF^{q}*e_f + KF^{q}*e_f \tag{11}$$

3.4.2. Manufacturing emissions

The following equation determines the manufacturing emissions (ME) for the equipment required. Where CM is the concrete mixer, OC is the oven cost, CS is the cardboard shredder, BM is the blender mixer, RM is the rotator mixer and ef is the emission factor. The capacity of the manufacturing equipment is represented via kWh.

$$ME = CM\_kWh*e_f + OC\_kWh*e_f + CS\_kWh*e_f + BM\_kWh*e_f + RM\_kWh*e_f \tag{12}$$

3.4.3. Transportation emissions

Transportation emissions (TE) are calculated based on the emission factor of the truck (EFT) and the total distance of transportation (KM). The following equation determines these factors. It is important to note that the average distance for materials to be taken to the concrete batching plant and to the construction site is 35 km each.

$$TE = EFT * KM \tag{13}$$

3.4.4. Total greenhouse gas emission equation

The total greenhouse gas emission calculation is determined by the following equation.

$$TEE = RME + ME + TE \tag{14}$$

3.5. Objective functions

The current study is an optimisation to minimise the negative environmental impacts (TEE) and minimise the total composite costs (TCC) for the novel KF composite.

Objective function 1: Minimise, $TCC = LMC + RMC + EC + EEC + TC$[15]

Where RMC is the raw material cost, TC is the transport cost, LMC is the labour manufacturing cost and EC is the equipment cost.

Objective function 2: Minimise, $TEE = MEE + ME + TE$ 16

Where MEE is the material embodied emissions, ME is the manufacturing emissions, and TE is the transport emissions.

4. Results and discussion

4.1. Multi-objective optimisation results

The parameters used to ascertain the optimisation solutions are provided in Table 5. The upper and lower bounds of the bespoke materials correlate to the constraints and design variables of the mix designs. For example, 17 kg of KF or MK is equal to 5% cement requirement in 1 m^3 of concrete. Similarly, 72 kg of KF is equal to 20% of the cement requirement in 1 m^3 of concrete. The non-dominated optimisation solutions are illustrated in Figs. 2 and 3. All material production costs, and environmental emissions are based on 1 m^3 of the novel concrete. The Pareto Front is graphically depicted in Fig. 2. This Figure represents the non-dominated solutions, and the best chromosomes are those that lie within the Pareto Front. The Pareto Front demonstrates the best possible solutions that satisfy the conflicting objectives of the optimisation problem. Fig. 3 represents the best candidate solution to the optimisation problem. The evolution of the best chromosome refers to the improvement of the best candidate solution during the optimisation process. Objective function 1 (TEE) and objective function 2 (TCC) are shown on the Y- and X-Axis, respectively. Moreover, the Y-axis is measured via kilograms of carbon dioxide equivalent ($kgCO_2$-eq) and the X-axis is measured in Australian dollars (AUD). The solutions can be categorised into three dominate regions based on alternative preferences, as highlighted in Fig. 3. Each region contains one solution based on the three region categories. Region 1 provides an optimised solution with the lowest costs however, high GHG emissions. Region 3 provides an optimised solution with the lowest GHG emissions however, the highest costs. Region 2 provides an optimised solution considering both reduced costs and GHG emissions. The optimised solutions lie within these specific regions due to the various trade-offs between the objective function of costs and emissions.

The results indicate that all optimised mix designs are subjected to cost increases. This was to be expected with the additional processing requirements to convert waste cardboard into useable KFs ready for concrete integration. It is also important to note that KF processing costs have been completed on a laboratory scale. Whereas the manufacturing of cement is scaled in large production and associated costs have been reduced through business procurement strategies. This is further shown with the cost of cement being 77, 79 and 69% less than SF, MK and KF, respectively. Nonetheless, regions 1, 2 and 3 have a cost increase of 17,

Table 5
NSGA-II parameters.

Population size	100
Number of iterations/Generations	300
Numbers of decision variables	3
Cross over rate	0.9
Mutation rate	1/Numbers of decision variables
Crossover index	20
Mutation index	100

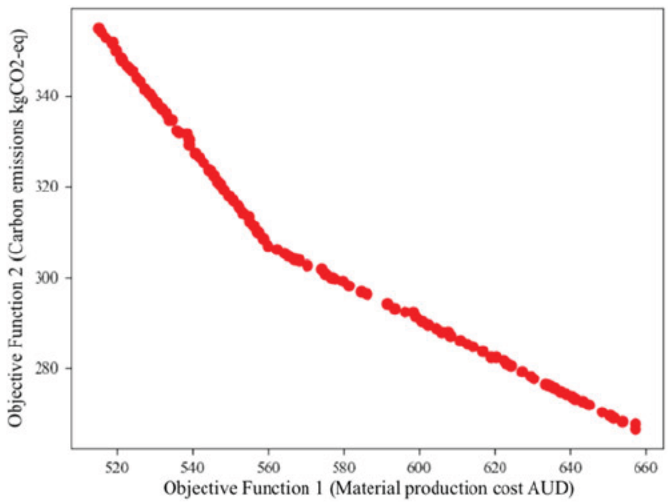

Fig. 2. Pareto front for economic and environmental emissions of novel composite.

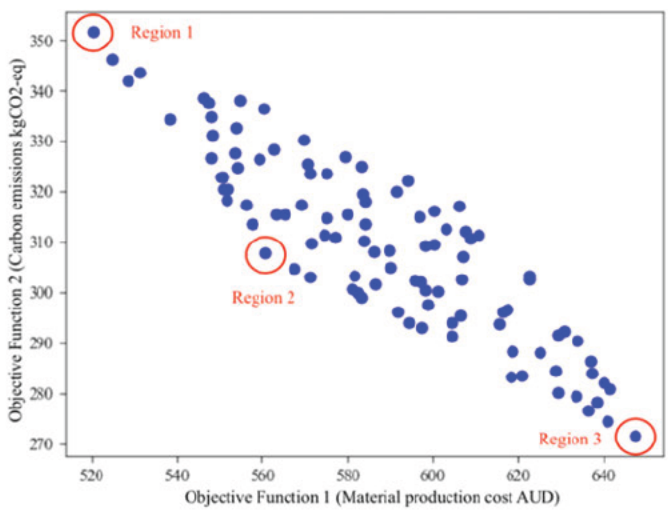

Fig. 3. Evolution of the best chromosome for economic and environmental emissions of novel composite.

27 and 36%, respectively. The optimised results also indicate reduced GHG emissions across all regions. This was to be expected due to the substitution of cement which has a high emission factor. Despite additional emissions created during the processing of KFs, this was counteracted due to the increased percentage of cement substitution. The optimised mix designs for regions 1, 2 and 3 had a total cement substitute of 11, 25 and 43%, respectively. Moreover, increasing the amount of KF, MK and SF then increased the emission savings. Regions 1, 2 and 3 demonstrated GHG emission savings of 7, 18 and 28%, respectively.

The optimised solutions can be used to provide benchmarks toward sustainability goals when integrating novel waste materials in concrete and mortar materials. The selection of the mix design is critical to ensure the targets of the construction project are being met and that feasibility is ascertained. For example, region 1 demonstrates the lowest associated costs. However, the solution provided in region 1 also has the highest GHG emissions, therefore if the project scope was to include sustainable materials as a key performance indicator, then region 3 would be more beneficial. If there were higher allocations for budget, then region 2 would be more optimal as this provides both environmental and economic considerations.

4.2. Experimental analysis

To determine the feasibility of the optimised mix designs from the specified regions for the objective functions above, it is important to analyse the compressive strength. Compressive strength is a key factor that concrete is used in such a wide variety of applications. The non-dominated optimisation solutions provided the relevant input variables of KFs, SF and MK. All other materials were equal to the control. The compressive results are illustrated in Fig. 4. The mix design of the regions when substituting a percentage of cement is shown in Table 6. It is important to note that the workability of the KF concrete was not compromised during the mixing stage. This was determined when KFs did not interfere with water requirements. The pre-treatment of SF on the KF walls negated absorption factors for the natural fibrous material. The novel composite and KF images are shown in Fig. 5. There is an increase of cement substitution from region 1 to 3. As the percentage of KF, SF and MK increase, the compressive strength decreases. As discussed, with the increase of cement substitution there is a decrease of GHG emissions produced. However, as both objective functions are considered to minimise economic and environmental factors for region 2, the compressive strength is below both region 1 and 3.

The results shown in Fig. 4 demonstrate a 36–68% decrease across regions 1 to 3. This is to be expected with the inclusion of less dense materials such as KFs. The reduction in cement can reduce the pozzolanic effect when the constituent materials are mixed. Although MK is considered a supplementary cementitious material, there is a significant increase of aluminate which can cause additional effects within the composite. Primarily, the high percentage of aluminate can reduce the production of calcium hydroxide ($Ca(OH)_2$) which can then decrease the density of the material. Thus, reducing compressive strength. However, reducing the production of $Ca(OH)_2$ has been shown to increase the durability of KFs. It is important to note that the reduced amount of SF has shown an inherent strength reduction of the composite for region 2. The linear constraint, SF $\leq$ 0.02 of KF for regions 1 and 3 show the content of SF at 1.8 and 1.72%, respectively. Whereas region 2 only has 0.5% SF for the total amount of KFs. Despite the reduced amount of MK in region 2, reducing SF demonstrates significant strength reduction.

It is important to note that for regions 1–3, the mix designs provided from the optimal solutions are at the lower and upper bounds of the linear constraints. This is because linear programming seeks to optimise an objective function subject to a set of constraints. In a linear programming problem, the constraints are represented as linear inequalities or equations that limit the possible values of the decision variables. If a constraint is active, the optimal solution will be at the boundary of the feasible region defined by the constraint. However, when a constraint is not active, the optimal solution may be at an interior point of the feasible region. Therefore, the mix designs provided from the optimal solutions are at the lower and upper bounds of the linear constraints because those

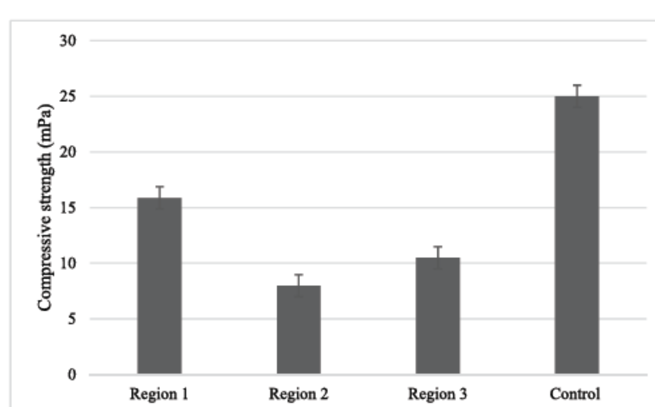

Fig. 4. Compressive strength results for optimised mix design regions.

Table 6
Mix design of cement substitution for 1 m^3 of concrete (kilograms).

Mix design	KF	MK	SF	Cement
Control	–	–	–	355
Region 1	21	17	0.38	316.62
Region 2	72	18	0.36	264.64
Region 3	72	80	1.24	211.76

(a) Composite cylinders during compressive testing

(b) Silica fume kraft fibres

Fig. 5. Sample of the novel composite and KFs.

are the values that satisfy the constraints to optimise the objective functions. This is demonstrated by objective function 1 that has minimal costs at the highest peak of objective function 2. The production cost of KFs is higher than cement because of the manufacturing proficiency of the cement industry. Moreover, as shown in region 3, the higher percentage of KFs indicates a higher cost. However, increasing the amount of KFs reduces the GHG emissions for objective function 2. Only when MK is integrated within the mix design is there a compromise between costs and GHG emissions. MK has less negative environmental effects compared to cement which demonstrates a compromise between objective function 1 and 2 when 5% is integrated within the mix design. The determination of which specific mix design to utilise in real world applications is dependent on the trade-off solutions provided from the three regions. For example, if the strength requirement is not demanding on the application, then region 3 may be most optimal as it saves the most GHG emissions. Alternatively, if costs are a main constraint, then region 1 mix design may be the better solution. Region 1 does provide the highest compressive however, it also produces the most GHG emissions.

Reducing any amount of cement required is more sustainable, despite mechanical strength variations. Therefore, the application of the novel composite will depend on the desired strength. The mix design shown below does not consider additional additives to increase strength, as the scope of the study is to minimise associated costs and GHG emissions when using waste cardboard.

5. Conclusions

Although cardboard waste has a 61.2% recycling rate in Australia, 2.2 Mt amount of cardboard is sent to landfill each year. This is partly due to contamination, but also cardboard materials can only be recycled 2–3 times before the kraft fibres become too brittle and short to be used further. The scope of this study was to conduct a multi-objective optimisation to minimise the economic and negative environmental effects when using kraft fibres derived from waste cardboard. The non-dominated sorting genetic algorithm (NSGA-II) was utilised to produce Pareto front with 100 outputs. The evolution of the chromosome further illustrated three key regions of optimised solutions for further experimental analysis. A comparative assessment of the results indicated that there is a 17, 27 and 36% cost increase when cement is substituted by 12, 27 and 42% kraft fibres, silica fume and metakaolin, respectively. However, increasing the percentage of cement replaced, also increased the savings of GHG emissions produced. This was shown with reduced GHG emissions of 7, 18 and 28% for regions 1–3. The outputs from the optimisation also produced three key mix designs which were further analysed. The three key regions were experimentally investigated for their compressive strength. Similar trends were shown from the objective functions, where the increased amount of cement replaced decreased the compressive strength. Although there was a 36–68% decrease in compressive strength for regions 1–3, the optimised mix designs highlight that the waste material can achieve sufficient strength for non-structural applications. This can be shown in various construction applications such as pavements, top screeds, infill walls, temporary structures, and decorative elements. The findings of this study highlighted the importance of optimising economic and GHG emission savings to promote the use cardboard waste in novel cementitious composites. Limitations of the experimental analysis can be shown with the small number of samples tested, despite being in accordance with Australian Standards. When using fibrous materials in concrete, mass production is required to analyse the microstructure behaviour of novel materials. However, this study focused on the GHG emission and economic savings when using waste cardboard. Further studies can be focused on other environmental factors and increasing the mechanical strength of the composite material. Moreover, this study primarily focused on cement replacement, however additional additives, and other virgin material replacement of constituent materials in concrete could be focused upon. This study can assist decision makers on using alternative waste materials in concrete to promote sustainability aspects of the construction industry. Reducing any amount of waste material resulting in landfill promotes environmental benefits.

CRediT authorship contribution statement

Robert Haigh: Conceptualization, Methodology, Formal analysis, Resources, Data curation, Visualization, Investigation, Writing - original draft. **Yanni Bouras:** Conceptualization, Validation, Supervision, Writing - review & editing. **Malindu Sandanayake:** Conceptualization, Validation, Supervision, Project administration, Writing - review & editing. **Zora Vrcelj:** Supervision, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

No Competing interests were identified.

Data availability

Data will be made available on request.

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8.3 Concluding remarks

This chapter focused on the economic and environmental optimisation when integrating waste KFs within concrete materials. SF and MK were considered in the mix design due to their ability to act as a fibre and matrix modification technique, respectively. The multi-objective optimisation solutions produced three key regions with corresponding mix designs which were experimentally analysed for their compressive strength. The results indicated that there is a 17, 27 and 36% cost increase when cement is substituted by 12, 27 and 42% KFs, SF and MK, respectively. However, increasing the percentage of cement replaced, also increased the savings of GHG emissions produced. This was shown with reduced GHG emissions of 7, 18 and 28% for regions 1-3. The outputs from the optimisation also produced three key mix designs which were further analysed. Similar trends were shown from the objective functions, where the increased amount of cement replaced decreased the compressive strength. Although there was a 36- 68% decrease in compressive strength for regions 1-3, the optimised mix designs highlight that the waste material can achieve sufficient strength for non-structural applications. Moreover, the findings of this study highlighted the importance of optimising economic and GHG emission savings to promote the use of cardboard waste in novel cementitious composites.

Chapter 9 - Conclusion

This research thesis presents a comprehensive investigation of a partial cement substitution using waste cardboard KFs. This study was prompted by the need for a sustainable alternative to cement in cementitious composites. Additionally, waste KFs were investigated in order to provide a solution to the problem of cardboard waste accumulation.

The primary function of cementitious composites is the strength of the material to undertake significant loads. To analyse the viability KFs have within cementitious composites, the mechanical performance was initially investigated. During this process, fibre and matrix modification techniques were created from analysing the gap in literature and then to enhance further knowledge. A multitude of mix designs were created, ranging from 5-20% KF integration. Each experimental program pertained the measurements of compressive, tensile, and flexural strength with raw and silica fume modified fibres, as well as matrix modified mix designs. SEM was utilised to demonstrate the effect all constituent materials had on the KFs. The results illustrated sufficient silica fume adhesion on the KF walls, while the energy dispersive x-ray spectroscopy observations showed adequate dispersion of all composite materials. These results provided an understanding of the physical effect KFs have within the various mix designs. It was found that increasing the fibre content lowered the mechanical strength function of the composites. However, modifying the fibres with silica fume increased the mechanical strength and enabled lower levels of petrification to occur, thus increasing the fibres durability.

Subsequently, the thermal characterization of KFs was analysed. The fibres chemical nature was determined using a Fourier transform infrared spectroscopy (FT-IR). The FT-IR gave insights to KFs exhibiting specific heat flow absorptions. The broad signal illustrated hydrogen bonding stretching, and vinylic hydrogen stretching. This generated a typical fingerprint region of a ligno-cellulosic material, in their respective spectrum. The peaks were correspondent to impurities such as residual waxes and lignin from the kraft process. Due to the previous manufacturing process of KFs within cardboard, it was important to demonstrate the chemical nature of the fibres and determine any impurities on the fibre walls. The thermal, calorimetric and combustion attributes of the fibres were then measured using thermogravimetric analysis (TGA), differential scanning calorimetry (DSC) and pyrolysis combustion flow calorimetry (PCFC). The TGA showed the following distinctive patterns. Firstly, initial loss of water. Secondly, main chain degradation, including dehydration reactions. Finally, degradation of the carbonaceous residue until the end of the run. The extent of water retention among raw and modified fibres differed albeit within a relatively narrower range (between 3 and 7 wt. %); however, the corresponding values for mass losses at elevated temperatures varied more widely. These values were influenced by the heating rates, and this aspect was then reflected in the amounts of char residues that were left after the completion of the runs. The presence of silica fume on the KFs exhibited the maximum amount of char residues and minimum extents of mass losses in lower temperatures. This demonstrated the application of silica fume on the KF walls delayed the degradation at high temperatures. DSC results followed similar patterns to the TGA thermograms. The endothermic peaks corresponded to the loss of physically bound water and main chain pyrolysis reactions. A broader endothermic peak beyond was an indication of slow degradation reaction of the residue that was left after the main chain degradation. The PCFC determined the heat release capacity (HRC) of the fibrous materials. Subsequently, SFKFs showed a lower HRC, demonstrating a lower combustion propensity compared to raw KFs. Furthermore, the decreased peak heat release rate (pHRR) of SFKFs highlighted the overall reduction in the fire hazards associated when using this material in composite designs. TGA results also confirmed a lower mass weight loss of SFKFs at elevated temperatures, thus corroborating the results from the PCFC runs.

The durability performance of KF reinforced concrete was the focus of the proceeding works. The experimental program of accelerated ageing consisted of wet and dry cycling at 20-, 40-, 60-, 80- and 100-day intervals. Mechanical tests were then completed after each ageing cycle to demonstrate the durability of KFs within concrete composites. To complement the findings shown in the accelerated ageing experiments, water immersion, absorption, carbonation, and chloride ion permeability tests were also conducted at 28, 56 and 90 and 365-day intervals. Silica fume modified fibres demonstrated a lower porosity rate via chloride ion permeability testing, exhibiting fibre modification enhanced the durability of the composite. This was further corroborated with the same fibres demonstrating the lowest water absorption on the total composite design. The addition of silica fume on the fibres reduced the total fibre permeability, thus enhancing durability. Composites with fibre content reduced the carbonation process, however, raw fibres exhibited lower levels of carbonation. This was primarily due to the ability of raw KFs to absorb carbon dioxide (CO_2) faster than modified fibres. This also corroborated that silica fume on fibre walls reduced fibre permeability. Compressive and tensile testing at ageing interval demonstrated an increase of mechanical strength. This was initially due to the increased aged of the concrete materials. Tensile strength experiments demonstrated that fibre integration enhanced the strength of the concrete materials. This has been shown due to the fibres ability to interlock within the composite materials and perform the crack bridging effect. Although concrete has a low tensile strength, the integration of fibres demonstrated this ability. Fibre and matrix modified composites demonstrated the highest compressive strength out all fibre mix designs. As expected, the control remained to have the highest compressive strength values due to the increased density value. With both modified fibre and matrix composite mix designs there was an enhancement of total durability by the materials consuming calcium hydroxide at early and later stages. This was initially investigated of each material used for fibre and matrix modification in the preliminary stages of the research.

Following the physical attributes of KF integration within cementitious composites, the sustainability aspects of waste cardboard integration were determined. The life cycle assessment (LCA) was conducted to evaluate the environmental effects when KFs were integrated as a partial cement substitution. This analysed the viability of using the waste material further in cementitious composite designs. LCA outputs on key impact categories were analysed with a Monte-Carlo (MC) simulation utilised as the sensitivity analysis method. The MC simulation included the variability of GHG emissions produced from transport and energy usage from operational machinery. The LCA measured $1m^3$ of two novel composites both using 10% silica fume modified fibres with one composite design incorporating 5% metakaolin as the matrix modifying method. The measured impact category results included terrestrial acidification potential, global warming potential, terrestrial ecotoxicity potential, marine eutrophication potential, human carcinogenic toxicity potential, stratospheric ozone layer depletion potential, and mineral resource scarcity. The initial results of the constituent materials determined that any amount of KF integration demonstrates a saving toward terrestrial ecotoxicity potential. Mineral resource scarcity is also improved with fibre integration. This was to be expected as virgin extraction of raw materials for cement production is lowered with KF utilisation. Hence, the basis of this research. Global warming potential was significantly reduced when KFs were partially substituted for cement. Increasing the amount of KFs inherently increased the savings. However, the MC simulation provided conflicting results when energy was derived from non-sustainable sources. Additional processing requirements to transform waste cardboard into KFs produced further greenhouse gas emissions. This was also shown with additional transportation requirements of the waste material and metakaolin to the concrete batching plant. However, it is envisioned soon energy sources will be more sustainable due current climate action goals and current sustainable trends across all industries.

The multi-objective optimisation determined the most optimal amount of kraft fibres to integrate within concrete materials regarding two objective functions: cost and greenhouse gas emissions. The multi-objective optimisation solutions produced three key regions with corresponding mix designs which were experimentally analysed for their compressive strength. The results indicated that there is a 17, 27 and 36% cost increase when cement is substituted by 12, 27 and 42% kraft fibres, silica fume and metakaolin, respectively. However, increasing the percentage of cement replaced, also increased the savings of GHG emissions produced. This was shown with reduced GHG emissions of 7, 18 and 28% for regions 1-3. The outputs from the optimisation also produced three key mix designs which were further analysed. Similar trends were shown from the objective functions, where the increased amount of cement replaced decreased the compressive strength. Although there was a 36- 68% decrease in compressive strength for regions 1-3, the optimised mix designs highlight that the waste material can achieve sufficient strength for non-structural applications. Moreover, the findings of this study highlighted the importance of optimising economic and GHG emission savings to promote the use cardboard waste in novel cementitious composites.

9.1 Novel contributions

Significant contributions have been made by this research thesis in the subject area of waste cardboard KFs within cementitious composites. Research outcomes are stated as follows:

1. Novel composite mix design using kraft fibres derived from waste cardboard was created.
2. Produced an original modification technique using silica fume on kraft fibres.
3. Illustrated the influence waste cardboard provides as a partial cement substitute within concrete and mortar materials.
4. Demonstrated the use of high and low values of kraft fibre integration within cementitious composites.
5. Discovered silica fume modified kraft fibres have a reduced overall flammability within cementitious materials.
6. Determined matrix modification using metakaolin enhances the durability of kraft fibre composites.
7. Demonstrated silica fume modified kraft fibres have a lower water absorption rate, enhancing durability factors.
8. Illustrated variations of petrification occurrence to non-modified and silica fume modified fibres via SEM analysis.
9. Produced original life cycle assessment results demonstrating savings for terrestrial acidification potential, global warming potential, terrestrial ecotoxicity potential and human toxicity potential impact categories, respectively with kraft fibre integration.
10. An original cost and greenhouse gas emission optimisation mix design using kraft fibres was determined.
11. Determined that waste cardboard can be utilised as a filler within cementitious composites.

9.2 Recommendations and future research

In Chapter two, gaps in knowledge areas of waste cardboard KF integration within cementitious composites were identified. Fibre and matrix modification utilising KFs as a partial cement substitution formed the research objectives of this thesis. The remaining knowledge gaps may be considered in future research projects. The following is a set of recommendations to further the work outlined herein.

1. The effects of extreme heat on kraft fibre cementitious composites remain unknown. Further research can be conducted to provide performance evaluations when using the fibrous materials in concrete. Moreover, the safety and reliability of kraft fibres in concrete when under extreme heat could be used to benchmark natural waste materials in concrete in fire focused research.
2. Spalling investigations with high and low volumes of kraft fibres within concrete materials can also be investigated. Research has been conducted using polymer-based materials to reduce spalling in concrete; however, the effect kraft fibres have on the spalling phenomena remains unknown.
3. Research on the effect kraft fibres have on creep and shrinkage development in concrete materials. Additionally, monitoring these developmental effects via field testing can be beneficial to highlight the potential of kraft fibres used in real-life scenario-based research.
4. Mechanical investigations on the shear strength of composites using kraft fibres in conjunction with other waste materials. As detailed in this report, kraft fibres reduce the compressive strength of concrete. Therefore, the addition of an alternate fibrous material could yield mechanical benefits.
5. The creation of alternative mix designs to accommodate specific strength criteria in building and construction applications. As shown, this research focused on 25MPa concrete materials. However, further research using kraft fibres in higher strength concrete can be investigated.

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