Product Optimisation 2

CCUS Innovation 2.0

Key Knowledge Deliverable 4.4

February 2025

Key Knowledge Deliverable Cover Sheet

This Key Knowledge Deliverable (KKD) has been produced by Imperial College London as part of the Department for Energy Security and Net Zero £1bn Net Zero Innovation Portfolio (NZIP) - CCUS Innovation 2.0 programme. The document is reflective of the status of the project at the time of writing. The material presented could have been subject to change as the project matured. These documents should not be considered a full representation of the final project.

Project Description

This project seeks to further develop and scale a new carbon sequestration process which transforms waste CO2 gas from industrial facilities into valuable construction products. Sequestered CO2 through this process is cheaper than conventional approaches that rely on purification, liquification and offshore or geological storage. The CO2 is stored in the form of a stable mineral which ensures they will be no leakage over time.

The patent-pending technology involves taking globally abundant magnesium silicate minerals and splitting this into magnesia and silica components. Through simple chemical processing two products of high purity are created: a) an amorphous silica that can be used as supplementary cementitious material (SCM) to facilitate low-carbon concrete and b) a concentrated magnesium solution in which CO2 from industrial flues can be sequestered to produce other construction materials.

This CCUS Innovation 2.0 award will be used to increase our technology and commercial readiness level by de-risking and facilitating the development of a pilot facility, in order to demonstrate that the technology is economically viable and deployable at scale.

Description of KKD

This KKD reports on the data collected on the performance of magnesium carbonate products. The aim was to optimise the strength of the products for appropriate applications and maximize durability.

KKDs to be released in full

- D3.4 – Concrete Trials 3
- D4.4 – Product Optimisation 2

KKDs to be released after redactions

- D1.1 – Flue Gas Recovery and Testing 1
- D1.2 – Dissolution Procurement
- D1.3 – Dissolution Operation
- D1.4 – Flue Gas Recovery and Testing 2 & Carbonation Procurement
- D1.5 – Carbonation Operation
- D2.3 – Reagent Regeneration Procurement
- D2.4 – Reagent Regeneration Operation
- D3.2 – Concrete Trials 1
- D3.3 – Concrete Trials 2
- D4.2 – Process Optimisation
- D4.3 – Product Optimisation 1
- D5.2 – Business Development 2 (Supply Chain)
- D5.3 – Business Development 3 (Business Planning)
- D5.4 - Business Development 4 (Commercial Readiness)
- D6.1 – Year 1 Report
- D6.2 – Year 2 Report

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industry.innovation@energysecurity.gov.uk

Contents

Introduction

The carbon mineralisation process used in this research produces nesquehonite (NQ, MgCO3.3H2O), a metastable magnesium carbonate phase. With time, nesquehonite decays to produce hydromagnesite (HM), Mg5(CO3)4(OH2).4H2O, through a dissolution and reprecipitation reaction, as shown in equation 1.

5MgCO3.3H2O → Mg5(CO3)4(OH2).4H2O + CO2 + 10H2O Eq. 1

This transition can be accelerated through the addition of water and elevating the temperature above 60 °C and it is widely reported that this transition is thermodynamically favourable above 50 – 60 °C. The morphological changes that accompany this transition are responsible for the setting mechanism of the materials being developed.

We are investigating the use of these materials as the binder in low carbon boards, as an alternative to gypsum-based plasterboard, and in low-carbon bricks, as an alternative to clay-fired bricks. An overview of the production of these two products is given in Figure 1.

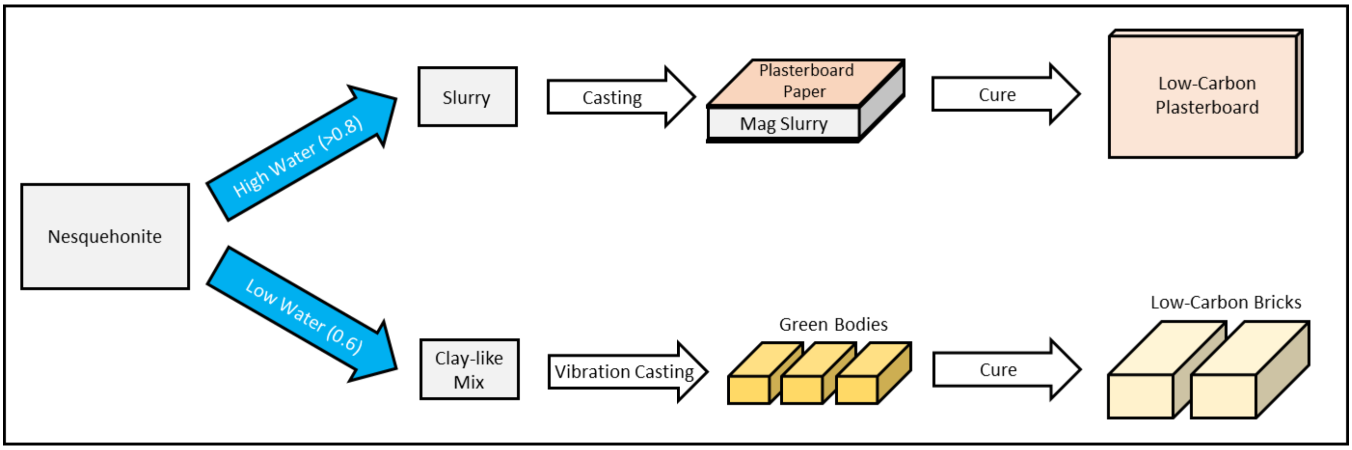

Figure 1. Brick and board production schematic.

Aims and Objectives

This report outlines the intermediate data collected to date on the performance of magnesium carbonate products. The aim of this deliverable is to optimise the strength of the products for appropriate applications and maximize durability. Areas covered include:

Brick/Blocks

Impact of the use of magnesium carbonate aggregates on mix designs and brick/block performance

Impact of the use of milling in the preparation of magnesium carbonate on the performance of magnesium carbonate bricks/blocks

Impact of freeze-thaw testing on the magnesium carbonate brick samples, conforming to British Standard EN 772-18:2011.

Boards

Impact of the water to binder (w/b) ratio and volume of aggregate used on the performance of boards

Impact of different fibre inclusions on the performance of boards

The given data combined with results reported in D4.3 provide an overview of the optimisation processes used to develop the MVPs of blocks and boards.

Methods

Nesquehonite Production

All nesquehonite used in this report was synthesised by combining 2 mol.dm-3 MgSO4 solution with 2 mol.m-3 Na2CO3 solution. The mixture was stirred for 2 minutes, and this produced immediate precipitation of the nesquehonite crystals. The resultant slurry was left at ambient conditions for 4 hours, allowing the nesquehonite to coarsen, before being filtered and washed using tap water. X-ray diffraction (XRD) was used to confirm that the identity of the magnesium carbonate produced was nesquehonite following every synthesis.

Brick/Block Samples

Magnesium Carbonate Aggregate Preparation

Magnesium carbonate paste samples made and tested for previous deliverables 4.2 and 4.3 were saved for use as the starting material for aggregate. These samples were crushed using a pestle and mortar to achieve particles with a range of diameters.

Sieve Analysis

To complete a viable comparison with virgin aggregate, the size distribution of the magnesium carbonate must be the same. To achieve this, a sieve analysis of virgin aggregate was performed. It was passed through 300 μm, 1 mm and 5 mm sieves. The distribution of sand aggregate is given in Table 1. Magnesium carbonate aggregate was sieved and separated to give an identical particle size distribution.

Table 1. Size distribution of aggregate.

Grade	Mass %
> 5 mm	0
1 mm < x < 5 mm	19
300 μm < x < 1 mm	51
< 300 μm	30

Bulk Density

To achieve equivalent v/v ratios of binder to aggregate in samples made for compressive strength testing, the densities of both sand and magnesium carbonate aggregate must be calculated. This was achieved by measuring the volume of a known mass of material using a measuring cylinder. The densities of the two aggregates are given in Table 2.

Table 2. Measured density of virgin aggregate (sand) and magnesium carbonate aggregate.

Aggregate	Density (g/cm3)
Sand	1.74
MC	0.55

X-ray Diffraction (XRD)

The phase composition of the magnesium aggregate used was 100% hydromagnesite. The diffractogram in Figure 2 confirms this, with all peaks being assigned to this singular phase. The use of QXRD gave an amorphous content of <1%. As a result, the aggregate is not reactive and only acts as an inert filler.

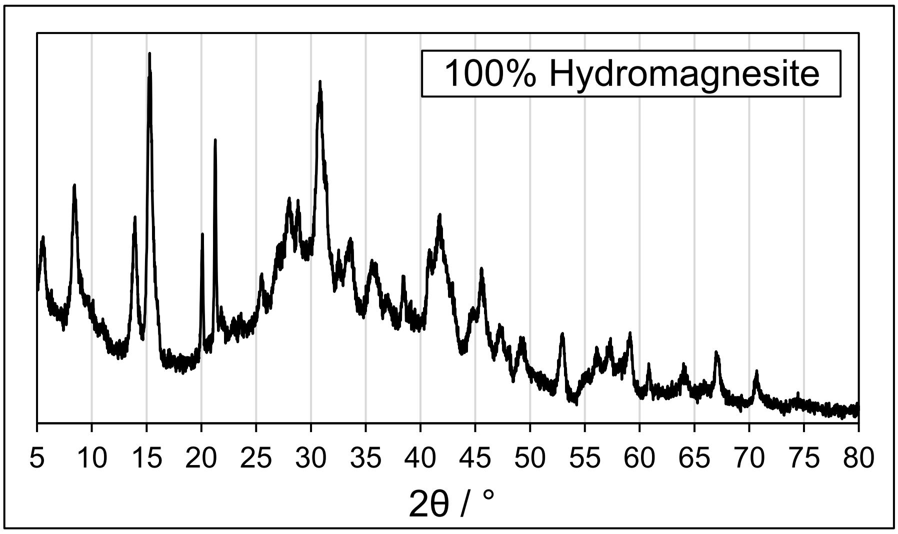

Figure 2. XRD diffractogram showing phase composition of magnesium carbonate aggregate.

Compressive Strength Testing

Magnesium Carbonate Aggregate

For each mix, six 50 x 50 x 50 mm3 mortar cubes were produced and tested using the following procedure.

Nesquehonite and appropriate aggregate (sand or magnesium carbonate) were added to a mixing bowl. The aggregate was added to give a constant volume ratio of aggregate to binder of 1.8. This was dry mixed for 5 minutes using an electric stand mixer, before the appropriate amount of water was added and mixed for a further 10 minutes. The water was added to give a constant flow. This mortar mix was then added to 50 x 50 x 50 mm3 moulds and vibrated using a vibration table for 3 minutes to compact the mix. The cubes were left overnight in moulds for water to be reabsorbed into the internal pore structure of the nesquehonite and the aggregate to give a sufficiently stiff consistency for them to be demoulded using an air gun. The resultant green bodies were placed in an oven at 60 °C for 24 hours to promote nesquehonite dissolution and hydromagnesite reprecipitation. Following this, cubes were placed in ambient conditions until they reached the testing age.

A Controls Group Automatic Compression Tester was used to measure the compressive strength, with a loading rate of 0.3 MPa/s.

Milling of Nesquehonite Starting Material

Nesquehonite was milled according to the procedure optimised in D4.3. Samples were prepared using the procedure outlined in the Magnesium Carbonate Aggregate section. 6 samples were prepared with unmilled nesquehonite, and 6 samples were prepared with milled carbonate. In both cases, the w/b used was 0.6, and the aggregate was sand at a ratio of 4:1, according to the procedure optimised in D4.2 and D4.3.

Durability Testing

Freeze-Thaw Resistance Testing

50 x 50 x 50 mm3 cubes were prepared using the same methodology as described in the Compressive Strength Testing section, according to the mix design given in Table 3. Virgin aggregate was used. These cubes were dried at 40 °C to a constant mass.

Table 3. Mix designs for freeze-thaw testing.

Sample Type	w/b (w/w)	a/b (w/w)	Curing Temperature (°C)	Curing Time (days)	Testing Methods
50 mm3 cubes	0.6	4	60	1 (@60) 28 (@RT)	Freeze thaw testing Compressive strength

Freeze-thaw resistance was determined following the methods given in standards BS EN 772-11 and -21. This consists of immersing the brick samples in water to full absorbance. The samples are then placed into the freezer for 24 hours, before being removed, thawed in water, and returned to the freezer. Samples were taken at 0, 5, 15, 30, 45 and 60 days for compressive strength testing, where the samples were dried at 60 °C until constant mass was reached before testing.

Boards

Flexural Strength Testing

All tested samples were 100 x 100 x 20 mm3 tiles. The main variables investigated were water/binder ratio (w/b) and aggregate binder ratio (a/b). The testing range of these variables was w/b: 0.7, 0.8, 0.9, 1, and a/b: 0, 0.25, 0.5, 1. The samples were prepared following the procedure given below.

Nesquehonite and sand were added to a mixing bowl and dry mixed for 5 minutes using an electric mixer, before the appropriate amount of water was added and mixed for a further 10 minutes. The mix was then added to 100 x 100 x 20 mm3 moulds. No vibration was used. The samples were placed in the oven at 60 °C in moulds for 24 hours to promote nesquehonite dissolution and hydromagnesite reprecipitation. Following this, the tiles were demoulded and placed in ambient conditions until they reached the appropriate testing age. 6 samples of each unique mix were prepared for testing. 3-point flexural strength was measured using a 10 Tonne capacity Instron instrument.

Fibres

All tested samples were 100 x 100 x 20 mm3 tiles. The w/b used in all cases was 0.8 and no aggregate was added. The different fibres selected for testing were short, glass fibres with an approximate length of 1 cm and long typha (plant) fibres of length ~10 cm. These were places in both random orientation and cross-directional orientation. The tiles were prepared following an identical procedure to that used for the Water/Binder Ratio and Aggregate/Binder ratio tests.

Results and Discussion

Boards

Effect of w/b and Aggregate on Performance

The flexural strength data for board samples with varying w/b and a/b is shown in Figure 8. There is an inverse relationship between w/b and flexural strength. In all cases, there is enough water present for the mix to fill the mould without the need for external vibration. This is necessary for the magnesium carbonate to fit into legacy infrastructure as a direct board binder material replacement. This amount of water ensures good compaction can be achieved as well as sufficient nesquehonite dissolution for significant phase transformation to take place. However, increases in w/b beyond the amount necessary for this flowability, increases the porosity within the samples. Therefore, a reduction in flexural strength is observed, as with more conventional binders like cement.

The samples prepared with lower w/b ratios of 0.7 and 0.8, observe a reduction in flexural strength with increasing a/b. This reduction is also commonly seen in practice with cement mortars. Increasing the mass of aggregate means there is less binder to coat these aggregates, fill voids and contribute to overall strength. Therefore, there is a weaker bond between the aggregates and the binder itself, as well as a reduction in flexural strength.

The samples with higher w/b ratios of 0.9 and 1 do not follow this pattern, and show lower strengths with lower a/b. This is because without aggregate inclusions at the higher w/b mixes, there is nothing to mitigate the high percentage shrinkage observed. Therefore, shrinkage cracks were seen, and these had a significant impact on the flexural strengths as the samples then had significant strength reducing flaws.

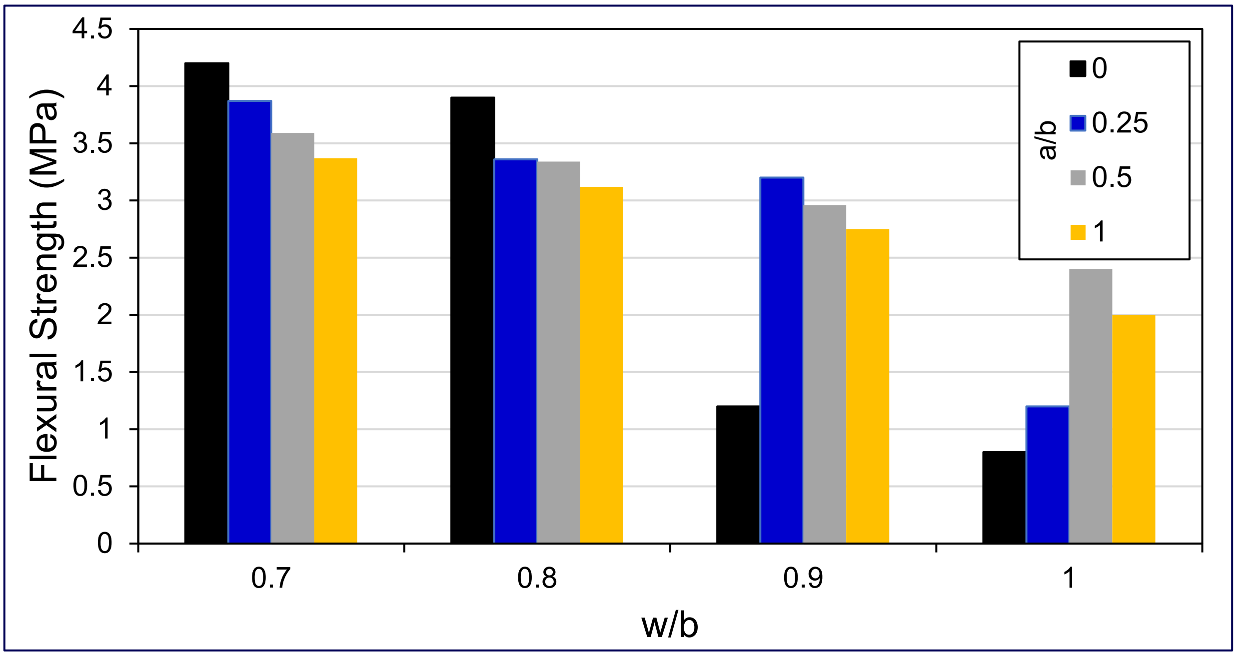

Figure 8. Flexural strength data showing impact of w/b and a/b.

Effect of Fibre Inclusion of Performance

Figure 9 gives flexural strength data from magnesium carbonate board samples containing the different fibres selected for testing. The use of glass and natural fibres in random orientation increases the flexural strength of the board samples by approximately 10-20%. However, the cross-axial natural fibre samples showed a decrease in flexural strength. This is likely due to the layers of fibres having poor adhesion with the binder and easily delaminating. Therefore, the board sample is reduced to binder sections of lower thickness, and lower strength.

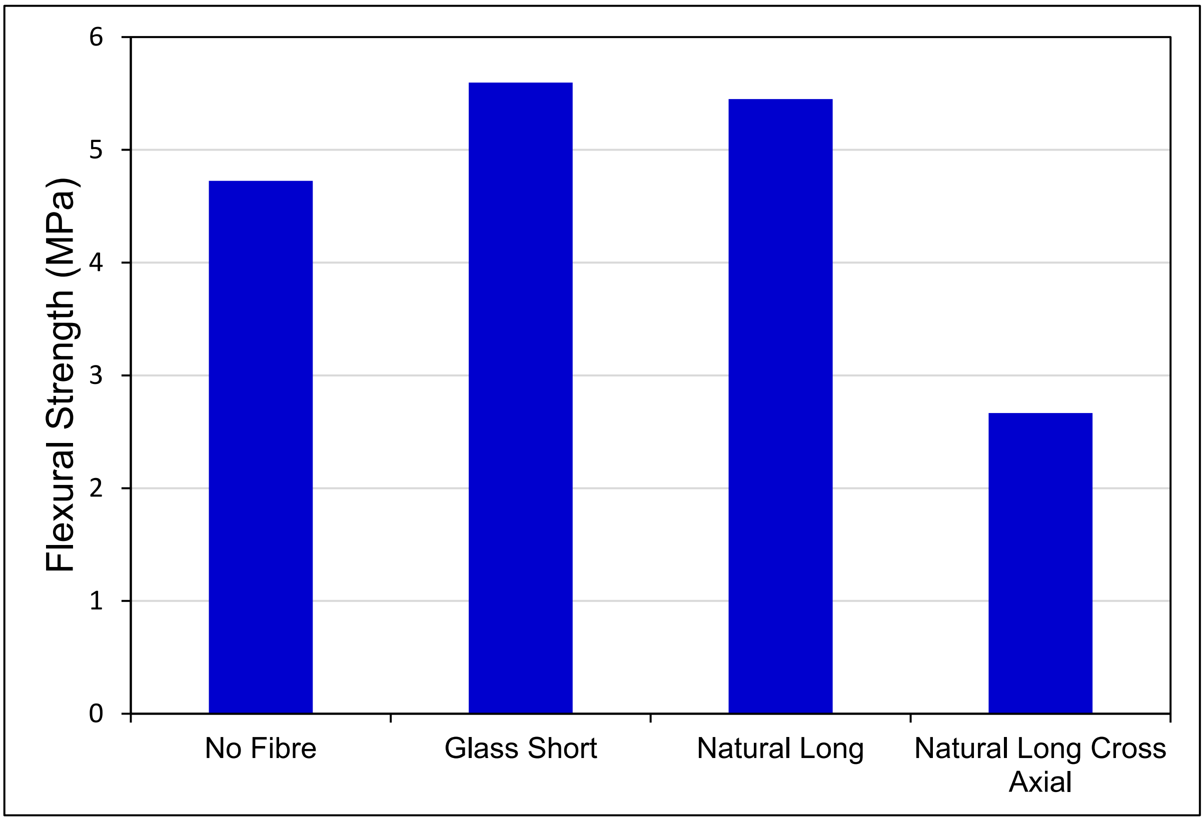

Figure 9. Flexural strength data showing impact of fibre reinforcement.

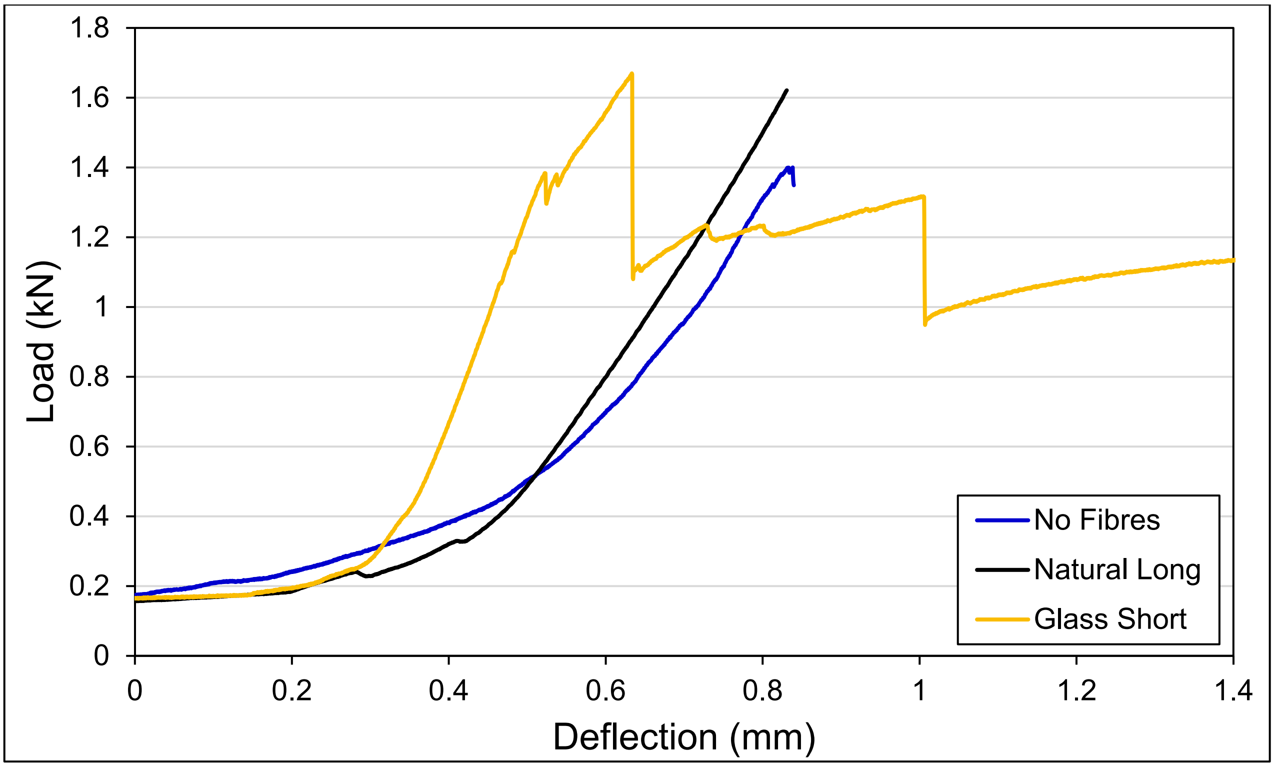

Figure 10. Load vs displacement plots of samples containing different fibre reinforcement.

Figure 10 shows the load/deflection behaviour of the samples with randomly aligned fibres. Without any fibre reinforcement present, the magnesium carbonate boards display brittle failure behaviour. This is also observed with the longer, natural fibres, likely due to the fibres themselves being weak. Therefore, when failure occurs, the fibres snap or split in a brittle manner. However, the samples containing shorter, glass fibres display ductile failure. This indicates better adhesion between the fibres and binder, with increased strength gain due to high energy pull-out of the fibres. This contributes to increased material toughness, as the stress is distributed more effectively, and crack propagation is delayed.

Conclusions

Bricks/Block samples

Compressive Strength

Bricks using magnesium carbonate aggregate exhibit lower compressive strength compared to those made with traditional sand, primarily due to the higher water demand and consequent differences in microstructure.

Milling of nesquehonite significantly enhances compressive strength by reducing particle size, increasing surface area, and promoting a more effective dissolution–reprecipitation process, leading to a denser matrix.

The hydromagnesite bound materials have properties comparable to pre-cast concrete block products. They have insufficient compressive strength to replace sintered clay ceramic bricks.

Durability (Freeze-Thaw Resistance)

Repeated freeze-thaw cycles cause progressive microcracking as water expands on freezing, resulting in a gradual decline in compressive strength.

In contrast, simple water cycling produces minimal degradation, indicating that the freeze-thaw process is the critical factor in durability reduction.

Board samples

Flexural Strength and Mix Design

An inverse relationship exists between the water-to-binder (w/b) ratio and flexural strength; excessive water increases porosity and weakens the matrix.

At lower w/b ratios, increasing the aggregate-to-binder (a/b) ratio reduces flexural strength due to reduced binder coverage and weaker bonding between the aggregate and the binder.

At higher w/b ratios, the absence of aggregate results in significant shrinkage cracking, further reducing flexural strength.

Although the hydromagnesite board samples produced have comparable properties to gypsum boards, further testing will be required to confirm commercial viability.

Fibre Inclusion

The inclusion of fibres (short glass fibres and long typha fibres) shows potential for enhancing the mechanical performance of boards.

The results on fibre reinforced hydromagnesite boards are preliminary in nature and further process development and testing is required to come to definitive conclusions on their commercial viability.