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Influence of additives on strength enhancement and greenhouse gas emissions of pre-cast lime-based construction products

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Influence of additives on strength enhancement and greenhouse gas emissions of pre-cast lime-based construction products

F. J. O'Flaherty[1*] , F. J. Khalaf[1] and V. Starinieri[1]

Abstract

Strength properties of laboratory scale lime-based samples enhanced with additives such as nanomaterials (nanofibrillated cellulose, nanosilica, nanoclay, expanded graphite), hemp & glass fibres, hemp shiv and polyvinyl acetate (PVAc) are determined. Samples were cured for 26 days in air at 20°C / 60% RH after casting before being oven dried for a further two days at 50°C (28 days total). Results show that the nanomaterials on their own had a mixed effect on the strength although $nSiO_2$ as a solo additive performed exceptionally well. The combination of fibres in conjunction with PVAc also greatly enhanced the strength due to increased bond between the fibres and the matrix. In addition, Greenhouse Gas emissions (GHG, $kgCO_2eq$) of an arbitrary block was determined for all composites and compared to the GHG of a commonly used lightweight aerated concrete block. Comparison of the normalised compressive strengths to the different loading conditions as outlined in BS EN 8103 shows that a more widespread use of pre-cast lime composites is possible and without unduly increasing GHG emissions.

Keywords Hempcrete, Nanomaterials, Hemp shiv & fibre, PVAc, Glass fibre, Greenhouse gas

摘要

本文测定了纳米材料（如：纳米纤丝化纤维素、纳米二氧化硅、纳米黏土、膨胀石墨）、大麻纤维和玻璃纤维、大麻杆和聚醋酸乙烯酯（PVAc）等添加剂对实验室尺度的石灰基试件强度的提升作用。试件的养护制度为在20℃和相对湿度60%下养护26天，之后在50℃烘箱中干燥2天（共28天）。试验结果表明，虽然纳米二氧化硅（$nSiO_2$）作为单一添加剂的效果很好，但同时掺加多种纳米材料对试件强度存在综合影响。PVAc可以增强纤维与基体之间黏结作用，因此将纤维与聚醋酸乙烯酯（PVAc）复合使用能明显提高强度。此外，本文测定了所有复合材料砌块的温室气体排放量（GHG，$kgCO_2eq$），并将其与普通轻质加气混凝土砌块的GHG进行比较。根据BS EN 8103标准，将不同加载条件下的试件抗压强度进行归一化并做比较，其结果表明，更广泛地使用预制石灰基复合材料具有可行性，且不会过度增加温室气体排放量。

关键词 工业大麻增强混凝土，纳米材料，大麻杆和纤维，聚醋酸乙烯酯，玻璃纤维，温室气体

1 Introduction

It is well known that a significant portion (approximately 50%) of the global energy consumption and CO_2 emissions are attributed to the building sector in industrialised countries [1] with concrete being the most commonly used building material. However, the cement binder is a non-sustainable material. Cement production is responsible for emissions of nearly one ton of CO_2

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into the atmosphere for each ton of cement manufactured [2]. Sustainable materials, therefore, are required to minimize the environmental impact of buildings [1]. In ancient times, lime-based mortars were used as a binder when building in stone. However, this changed in the nineteenth century as a result of the development of Portland cement. In more recent times, due to the need to develop more sustainable construction products, the use of lime has become popular again [3]. In comparison to cement, lime has a lower firing temperature [4] and hydrated lime absorbs CO_2 when hardening through carbonation.

However, the strength properties of lime as a stand-alone material are questionable for widespread use as a structural material mainly due to its low compressive strength, high shrinkage, and high porosity [5]. Natural Hydraulic Lime (NHL) 3.5 and 5 have compressive strengths at 28 days (3.5 and 5 MPa respectively) that are acceptable at the lower end of loading conditions as specified in BS EN 8103 [6], but these strengths would only allow limited use.

As a result, research was conducted with the aim of enhancing properties such as strength, insulation (thermal conductivity), porosity and shrinkage to make it more suitable as a building material. The material properties mentioned above are all very important from an in-service performance point of view to ensure the material performs effectively. However, this paper concentrates on the influence of selected additives such as nanomaterials (nanofibrillated cellulose, nanosilica, nanoclay, expanded graphite), fibres (hemp and glass), hemp shiv, and polyvinyl acetate (PVAc) on the strength characteristics of the lime composite. The minimum compression strength requirement for existing masonry is 2.9 MPa (BS EN 8103–2 [6]) but construction materials exhibiting these strengths can be only used cautiously with low load levels.

2 Research significance

By developing load-bearing lime composite materials, lime composite units could be off-site manufactured and used in a similar way to existing clay bricks and concrete blocks and, thereby, help reduce the carbon footprint of dwellings. The research employs a first-of-a-kind combination of nanomaterials, hemp shiv and hemp/glass fibres and polyvinyl acetate (PVAc emulsion) to enhance not only the strength but also thermal insulation properties. However, even though the additives can be beneficial for strength enhancement (i.e. > 7.3 MPa, the strength of commonly used lightweight aerated concrete blocks), the paper also establishes if the additives do not adversely effect Greenhouse Gas (GHG) emissions, thereby allowing specification consisting of an

acceptable strength for more widespread use combined with low carbon footprint.

3 Additives

Compressive strength is very important for civil engineering and building applications. Due to its importance, it will be considered the dominant factor that defines which of the additives will be selected to improve the strength characteristics based on the literature to date.

3.1 Nanocellulose (nFc)

Nanocellulose used as a suspension at 7% concentration helped to increase the compressive strength of cement without changing the density of the cement slurry. Due to its specific weight of 1.06 and high water content, water can be replaced by nanocellulose to a small extent. Compressive strength was increased from 16.9 MPa for pure cement to 26.7 MPa using 5% nanocellulose and 0.4 w/c [7]. It is reported that adding 0.15% by weight of nanocellulose fibres leads to a 15% and 20% increase in the flexural and compressive strengths of cement paste respectively due to the high degree of hydration and to the increase in the density of cement paste microstructure [8].

Another study pointed out that microfibrillated cellulose passed 30 times through the refiner and 5 wt. % to wood pulp had an increase in tensile strength of wood pulp compared to a 16-time pass through the refiner; it was raised from 200 to 300 MPa [9].

3.2 Nanosilica ($nSiO_2$)

Nanosilica is another material used to enhance many properties in matrices like cement, lime, biomass, biopolymer and polymer nanocomposites. Among these properties are flexural, compressive and tensile strength. Adding the ultra-fine particles into cementitious materials influences the performance leading to porosity at a nano scale. The nanoparticles of SiO_2 fill the spaces between gel particles of Calcium-Silicate-Hydrate (C-S–H) in cement, thereby acting as a nano-filler [10]. It also changes the interfacial transition zone between cement and aggregate particles due to voids decreasing to nano-size. Properties influenced are strength, durability, shrinkage and the bond between the steel reinforcement and material [11]. The pozzolanic reaction with calcium hydroxide led to an increase in the amount of C–S–H which produced higher density, increased the compressive and flexural strengths and improved durability of the cement mortar. This was achieved by adding $nSiO_2$ to synthesised cement slurry which was used for oil wells. Quantity of 70 g $Na_2SiO_3.9H_2O$ was added to 450 g of distilled water and 54.7 g of surfactant

factor, N-cetyl-N,N,N-trimethyl ammonium bromide (CTAB) and blended with 400 g of water and stirring 15 min. The solution was added to the solution of silicate. The compressive strength increased from 5.5 MPa to 27.6 MPa at 120 h and 87.7°C [12]. It was reported that adding 1 wt. % of $nSiO_2$ to cement, the compressive strength increased 37% in comparison to the pure cement [13]. In another study, nanosilica was added (1, 3, 5, 7 and 10%) to cement: sand, 1: 2.75 and w/c 0.485 at 28 days, the highest compressive and flexural strengths were 36.8 MPa and 5.7 MPa respectively compared to pure cement at 23.6 MPa and 3.6 MPa respectively [14]. Nano-silica was also used to improve the microstructures, mechanical properties and durability performances of ultra-lightweight cement composites (ULCC). The microstructure, durability and mechanical properties of ultra-lightweight cement composite (ULCC) was also improved due to the addition of $nSiO_2$. The compressive strength also increased in addition to water and chlorine ion resistance at 1 and 2% additions of $nSiO_2$ [15].

3.3 Nanoclay (nClay)
Nanoclay has attracted the attention of researchers due to its potential to enhance the mechanical properties of many matrices and polymers. It was found that adding 8 wt. % of nano-metakaolin to cement at 0.5 water/cement, the tensile and compressive strengths improved by 49% and 7% respectively [16]. Another study reported that cement mixed with 1 wt. % of calcined nanoclay (CNC), produced by heating nanoclay (Cloisite 30B) at 900° C for 2 h, decreased the porosity and increased the compressive strength from 53.1 to 74.2 MPa. The compressive strength improved to 69.8 MPa by mixing the cement with 1 wt. % nanoclay [17]. Many percentages of nanoclay (1, 3, 5, 7 and 10%) were added to a cement:sand mixture (0.485 w/c) at a ratio of 1: 2.75. The highest compressive and flexural strengths at 28 days were 37.7 MPa and 5.9 MPa respectively at 10 wt. % nanoclay in comparison to pure cement which was 23.6 MPa and 3.57 MPa respectively [14].

3.4 Expanded graphite (EG)
Expanded Graphite was used in some studies to enhance mechanical properties, but it was primarily used to improve electrical and thermal conductivity properties of construction materials. Expanded graphite was synthesised by preparing expanded graphite/paraffin gypsum composite (EGPG) modified by 1 wt. % carbon fibres, EGPG or gypsum blended with 10 wt. % EG/P (paraffin) and 1 wt. % CF (carbon fibres). The flexural and compressive strengths of EGPG were increased by 65.6% and 6.4%, respectively. Unfortunately, the

thermal conductivity increased 36.0% which is not beneficial in construction materials [18]. Mixing of cement with expanded graphite treated with ozone led to an increase in the bonding between the cement paste and the carbon of EG depending on the effect of gaseous ozone on modification of EG surfaces. Oxygen groups (carbonyl, phenol and carboxyl) were composed leading to this bonding effect and improved the resistance to bending load in the pre-cracking zone [19].

Hydraulic lime (NHL 5) was mixed with graphite which is considered compatible with lime paste and the compressive strength increased three times compared to the neat graphite [20]. Moreover, the porosity and water absorption decreased. Similar to the research by [21], the thermal conductivity also increased but this time up to 80% as a result of this addition.

3.5 Fibres (synthetic and natural)
Fibres are used to improve the mechanical properties of matrices (polymer, cement, gypsum and lime). There are many kinds of industrial fibres like steel, cuprum, polypropylene and carbon fibres. The most common industry fibres are glass fibre. Hemp fibres are natural and are obtained from the hemp plant.

3.5.1 Fibre glass (FG)
Fibre glass is relatively low cost and has been used to reinforce many different materials such as polymers, lead and cement over many decades, and more recently, lime-based materials. Previous research showed that hydraulic mortars containing 2% of glass fibres exhibited the best mechanical performances, either as maximum strength or toughness [22]. It has also been shown that the addition of fibres improves the mechanical properties of cement and lime mortars made from recycled ceramic aggregate [23]. When fibre glass in 50 mm lengths was mixed at 4 wt. % with water resistant gypsum (its compressive strength was 35 MPa from the supplier), the flexural strength significantly improved at 28 days from 4.96 MPa for gypsum to 22 MPa for the composite and the tensile strength from 2.75 to 18 MPa respectively [24]. Flexural strength for cement was also studied. Adding 5 wt. % fibre glass increased the traditional compressive value of cement mortar to 35 MPa for the composite [25]. Additionally, 10–40 mm length of fibre glass was mixed with cement at 2–8 vol. %. The optimum properties of the composite at 28 days were by adding 6 vol. % of fibre glass, flexural strength increased 4–5 times, the tensile strength improved 3–4 times. The impact strength was highly improved 15–20 times compared with the cement alone [26].

3.5.2 Hemp (fibres and shiv)

Hemp fibres (HF)   Hemp fibres are increasingly being used as reinforcements in composite materials e.g. [27, 28], often in place of glass fibres. The fibres are found in the bast of hemp plant and have strengths that are comparable to those of glass fibres [29]. Hemp is a plant described as a quickly renewing construction material, biodegradable, sustainable and eco-friendly. Its stem grows up to 1.2 – 4.5 m with a diameter of 2 cm [30].

The natural fibres found in the hemp plant have mechanical properties that are ideal for use in building composites [31]. They are also used in the manufacture of paper, hemp wool insulation, biosourced plastics and nonwoven thermocompressed market (automobile) [32].

Using hemp fibres with lengths of 5, 10 and 20 mm and ratios of 1, 2 and 3%, compressive strengths in cement-based mortars of 50 MPa and above were achieved in mixtures having 1% fibres with the optimum fibre length being 10 mm [33]. However, in contrast, 2–3% of hemp fibre with a length of 12 mm length gave the best results [34]. Concrete containing hemp fibres was also shown to exhibit a similar performance to concrete mixtures containing polypropylene fibres [35]. Hemp fibres were also shown to be an appropriate substitute for glass fibres for gypsum reinforcement [36].

Away from construction materials, epoxy composites containing hemp and jute fibres were developed for the interior of aerospace and automobile. Mechanical properties improved and the natural fibres were shown to be a viable alternative to synthetic (glass) fibres. Hemp fibres performed better than jute [37].

Hemp shiv (HS)   The use of crushed hemp stem as shives with their fibres (not separated from shives) to manufacture lime-hemp concrete was investigated by de Bruijn et al., [38] but this did not raise the strength properties of the composite. Similarly, a study [39] reported a number of different tests using hemp shiv but the compressive strengths were very low. Other researchers obtained strengths of 0.4–1.2 MPa [40]] and 0.2–0.5 MPa [41]. However, the thermal conductivity results were generally reasonable (which are considered elsewhere, O'Flaherty et al., 2019 [42]).

Research reported that the compressive strength from 10 different laboratories, of samples composed of hemp shiv and cement with citric acid were between 0.32–0.45 MPa [43]. Comparing these to the minimum compressive strength of load bearing materials, lime/shives or cement/shiv composites could only be used as insulators because of their insignificant compressive strength.

Lime mixed with hemp shiv/flax straw was studied and it showed that the lime hemp flax composite was low in strength, density and thermal conductivity but it has good absorptivity. Compressive strength was between 0.41–0.85 MPa and flexural strength was between 0.05–0.24 MPa [44].

3.6 Polyvinyl acetate (PVAc)

It was identified in preliminary testing that there was a lack of bond between fibres used in the mixture and the lime. Polyvinyl acetate (PVAc) was selected as an additive to the mix for this purpose. PVAc is a traditional adhesive which is used for wood connection in the furniture industry for example. Current formulations of PVAc are affordable and nontoxic, but newer formulations with an eco-friendly plasticizer were investigated and their characteristics relating to adhesion, resistance to water, film formation at low temperature, storage stability and absence of volatile organic compounds were established [45].

Polyvinyl acetate resin is a chain of monomers $(C_4H_6O_2)n$ and its density is about 1.03–1.13 g/cm^3. The average density is 1.08 which is close to the density of water and can be easily mixed with water. No mention of adding PVAc to lime based materials was found in the literature.

3.7 Summary of selected nanomaterials and additives for enhancing strength

Nanosilica, nanoclay, nanocellulose and expanded graphite were adopted to study their impact on enhancing the properties such as compressive strength of lime and hemp (fibres or shives) lime composites. In addition, the aim was also to enhance other properties such as insulation (thermal conductivity), porosity, which is linked to breathability and shrinkage, related to potential cracking which would influence the ability of the material to perform structurally. The results of the non-strength enhancements are detailed elsewhere [42].

Some studies discussed improving one or more physical properties like Young's modulus, tensile strength and the impact resistance and provided information on the nanomaterials and synthesis and natural fibres that were used to improve these properties [46]. From the above studies, the best options for developing the mechanical strength is hemp fibre reinforced lime and lime singularly blended with nanosilica, nanoclay, nanocellulose or expanded graphite. Fibre glass was used for comparison to the result of hemp fibres. Preliminary testing identified

the lack of bond between the fibres (hemp fibres and shiv, glass fibres) and the lime matrix, hence PVAc was selected to improve the adhesion.

4 Specimen preparation and test procedure

4.1 Materials

Lincolnshire Lime in the UK supplied the Natural Hydraulic Lime NHL 5 (Singleton Birch Secil, Product Code NHL-SB-5), conforming to BS EN 459 [47]. Its density is 2.7 g/cm^3. Two of the nanomaterials (nSiO$_2$ and nClay) were purchased from Merck in the UK. The nSiO$_2$ was in nanopowder form (spherical, porous) with particle size 5–20 nm (TEM), surface area, 590–690 m^2/g (TEM), bp 2230°C (lit.), mp > 1600 °C (lit.), density 2.2–2.6 g/mL at 25°C and bulk density 0.068 g/mL.

Nanoclay was purchased in powder form with the formula H2Al2O6Si. Its molecular weight is 180.1 g/mol with light tan to brown in colour. Its bulk density is 600–1100 kg/m^3 and average particle size < 25 µm. Expanded Graphite (EG) in powder form was obtained from Graphitene Ltd., Scunthorpe UK. It has a purity > 99.8%, expansion ratio 200 to 300 and particle size 80–200 µm.

Nanofibrillated cellulose was obtained from the University of Maine, Orono, USA. It is a white, odourless 98 w/% dry powder. It has a nominal fibre width of 50 nm with lengths of up to several hundred microns. It has a hydrophilic surface property (31–33 m^2/g (BET)). Its density is 1.5 g/cm^3 in dry powder form.

Hemp shiv was supplied by East Yorkshire Hemp, Driffield, UK. The average size was 15 mm long, width 5 mm and a thickness of about 0.5 mm. Hemp fibre was supplied by Wild Colours, Birmingham, UK and cut by hand into lengths between 5 and 10 mm. The average diameter of hemp fibre is 22 µm. Fibreglass was supplied by East Coast Fibreglass in the UK (chopped, 6 mm long and 13 µm in diameter, white in colour with a density 2.5–2.65 g/cm^3). A Polyvinyl acetate (Evo-Stik Super Evo-bond Waterproof PVAc) adhesive was sourced locally and chosen as it was as an environmentally hazard-free adhesive. It has a white colour in appearance in liquid form. Its specific gravity is approximately 1.05.

All percentages of additives given for hemp shiv, fibres (hemp and glass), nanomaterials (nSiO$_2$, nClay, nFC) and EG are their weights with respect to lime e.g. 20% HS/Lime means 20% of hemp shiv by weight of lime. The % PVAc is also the weight of adhesive by weight of lime.

4.2 Specimen preparation

The materials were mixed using a Hobart mortar mixer. The water/lime ratio was 0.4. However, since the various additives absorb water in the mixture, additional water was added with the aim of achieving a flow of 150 mm on a flow table to correspond to the control sample (as specified in BS EN 1015–3, 1999 [48]) but this was set to 0.5 for all additive containing samples for consistency. Magnetic stirring was used to completely disperse the manomaterial in the water for two hours before adding to the mixture although nClay magnetic stirring continued overnight for better dispersion. Dry mixing was conducted on the lime and hemp (shives or fibres) for five minutes at low speed (62 rpm). The PVAc was first added to the water/nanomaterial mixture and then magnetically stirred for 15 min. It was then gradually poured onto the dry mix of lime hemp (shives or fibres). Mixing was done for 15 min, five minutes on low speed followed by 10 min on high speed (125 rpm). The mixed composite was transferred to a mould and compaction was achieved via a tamping mallet having a tamping surface measuring 40×40 mm. Mould size of 40×40×160 mm was used for casting compressive and flexural strength specimens. The mix proportions are given in Table 1.

4.3 Curing method

The drying method adopted was a mixture of air and oven drying. Immediately after casting, the samples were stored at 20°C and 60% RH. At five days they were demoulded and stored in the same conditions for a further 21 days before drying for two days in a fan assisted oven at 50°C. This method was adopted to optimise the curing process which consisted of air curing, thereby enabling the lime cycle process (carbonation) before ensuring all samples were of a consistent nature before testing. The curing method would be suitable for precast specimens which would be transported to site for construction as opposed to being wet applied in-situ.

4.4 Compressive strengths

The compressive strengths of the specimens were determined using an Instron 3367 with a capacity of 30 kN. Testing was carried out in accordance with BS EN 1015–11, 1999 [49]. Compressive strength testing was conducted on the two broken pieces from the flexural test. Two 40×40 x 5 mm thick metal plates were placed above and below the specimens. Loading rate was 30 N/s. However, since the key theme of this paper is to investigate ways of increasing the compressive strength to make lime based materials load bearing, the flexural strengths will not be considered here.

5 Results

The results of the compressive load tests are shown graphically in Fig. 1 (with error bars). The compressive strengths range between 0.6 MPa and 11.7 MPa. However, modifications are required to the laboratory data to

Table 1 Mix proportions (based on 160 × 160 × 40 mm specimens)

No	% wt. filler	Water/ lime ratio	Lime (g)	Water (g)	Additive 1 (ID & weight, g)	Additive 2 (ID & weight, g)
1	Control (NHL5)	0.4[a]	691.2	276.5		
2	1wt. % nSiO$_2$	0.5	691.2	345.6	nSiO$_2$, 6.9	
3	2 wt. % nSiO$_2$	0.5	691.2	345.6	nSiO$_2$, 13.8	
4	3 wt. % nSiO$_2$	0.5	691.2	345.6	nSiO$_2$, 20.7	
5	0.5 wt. % nClay	0.5	691.2	345.6	nSiO$_2$, 3.5	
6	1 wt. % nClay	0.5	691.2	345.6	nClay, 6.9	
7	2 wt. % nClay	0.5	691.2	345.6	nClay, 13.8	
8	3 wt. % nClay	0.5	691.2	345.6	nClay, 20.7	
9	1 wt. % EG	0.5	691.2	345.6	EG, 6.9	
10	2 wt. % EG	0.5	691.2	345.6	EG, 13.8	
11	5 wt. % nFc	0.5	691.2	345.6	nFc, 34.6	
12	10 wt. % nFc	0.5	691.2	345.6	nFc, 69.1	
13	5 wt. % HF/L	0.5	691.2	345.6	HF, 34.6	
14	7 wt. % HF/L	0.5	691.2	345.6	HF, 48.4	
15	10 wt. % HF/L	0.5	691.2	345.6	HF, 69.1	
16	10 wt. % FG/L + 8 wt. % PVAc	0.5	691.2	345.6	FG, 69.1	PVAc, 55.3
17	10 wt. % FG/L + 12 wt. % PVAc	0.5	691.2	345.6	FG, 69.1	PVAc, 82.9
18	10 wt. % HF/L + 8 wt. % PVAc	0.5	691.2	345.6	HF, 69.1	PVAc, 55.3
19	10 wt. % HF/L + 12 wt. % PVAc	0.5	691.2	345.6	HF, 69.1	PVAc, 82.9
20	20 wt. % HS/L	0.5	691.2	345.6	HS, 138.2	
21	20 wt. % HS/L + 12 wt. % PVAc	0.5	691.2	345.6	HS, 138.2	PVAc, 82.9

Key: *nSiO$_2$* nanosilica, *nClay* nanoclay, *EG* Expanded Graphite, *nFc* nanofibrilated cellulose, *HF* Hemp Fibres, *FG* Fibre Glass, *HS* Hemp Shiv

[a] The design water/lime ratio was 0.4 but the water content was increased in all samples containing additives with the aim of achieving a flow of 150 mm on a flow table. However, this was set to 0.5 for all additive containing samples for consistency
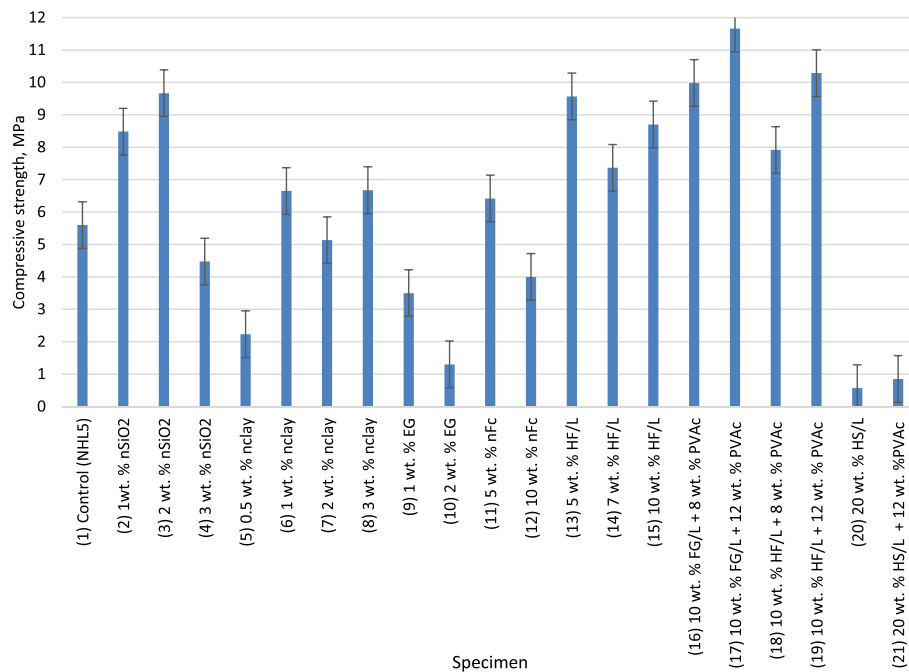
Fig. 1 Mean compressive strength of specimens

standardise them for comparison with existing masonry in terms of strength (Sect. 5.1). The composites GHG emissions are calculated in Sect. 6 and enable the optimum selection of material based on both strength and carbon emissions (Sect. 7).

5.1 Strength comparison with existing masonry

Part 2 of BS EN 8103 [6] provides information on the type of masonry unit to be used and is dependent upon the location of the masonry under certain conditions (internal or external) and size of dwelling (one to three storeys). Three conditions are given, labelled A-C and these are shown in Fig. 2. According to BS EN 8103–2 [6], a block is a masonry unit exceeding 337.5 mm in length or 112.5 mm in height with a minimum bed height of 190 mm. In this paper and for simplicity, it is assumed that the lime-based specimens will be manufactured as blocks so their strength will be compared to competing masonry for that reason. The compressive strength of five types of existing masonry blocks are given in Table 2 (from BS EN 8103–2 [6]) for the different conditions shown in Fig. 2.
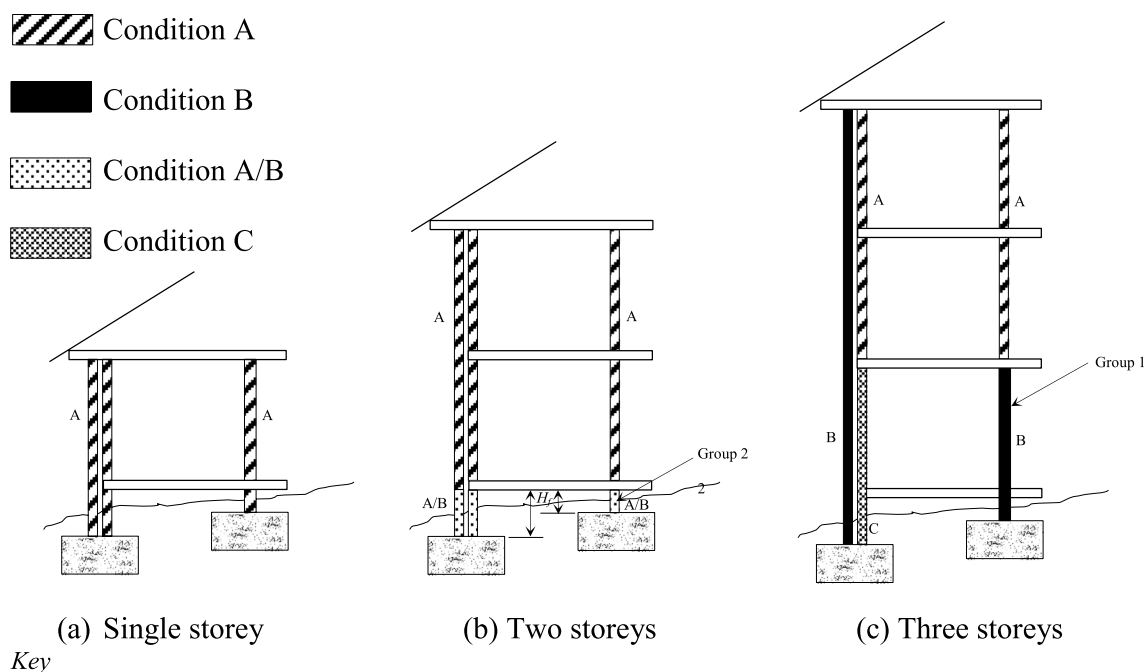

(a) Single storey   (b) Two storeys   (c) Three storeys

Key
Group 1: ≮ 140mm in blockwork and 215mm in brickwork (masonry units having no more than 25% of formed voids, 20% for frogged units)
Group 2: ≮ 140 mm thick in blockwork or 215 mm thick in brickwork below ground floor level if height $H_f > 1$ m (masonry units have formed voids > 25% and < 55%)

Fig. 2 Compressive strength of masonry units for buildings (adapted from BS EN 8103–2 [6])

Table 2 Compressive strengths of blocks used (MPa, refers to strength of blocks only from BS EN 8103–2:2013[6])

Condition	Masonry unit					
	Masonry unit	Clay masonry (BS EN 771–1)	Calcium silicate (BS EN 771–2)	Aggregate concrete masonry (BS EN 771–3)	Autoclaved aerated concrete masonry (BS EN 771–4)	Manufactured stone masonry (BS EN 771–5)
A	Block	Group 1: 5 Group 2: 8	Group 1: 5 Group 2: 8	2.9	2.9	Any unit that conforms to BS EN 771–5 is acceptable
B	Block	Group 1: 7.5 Group 2: 11	Group 1: 7.5 Group 2: 11	7.3	7.3	
C	Block	Group 1: 15 Group 2: 21	Group 1: 15 Group 2: 21	7.3	7.3	

5.2 Normalised compressive strengths

According to EN 1996–1-1:2005 [50][1], the design compressive strength of masonry units (as shown in Table 2) shall be the normalised mean compressive strength, obtained from BS EN 772–1 [51], Annex A. This is either (i) declared by the manufacturer or obtained by conversion of the compressive strength to the normalised mean compressive strength. This applies two factors, the first one modifies the compressive strength based on the curing regime for the particular type of masonry. In this paper, and to better replicate actual conditions to make the results more comparable, it will follow Clause 7.3.2 in BS EN 722–1 [51] (as the air-drying method described is the closest match to the specimens in this paper—store for at least 14 days in the laboratory at a temperature ≥ 15°C and relative humidity ≤ 65%). However, this gives a factor of 1.0 so has no effect on the tested compressive strength. The second factor, the air-dry compressive strength of masonry units is multiplied by a shape factor, *d*, as given in "Table A.1 Shape factor d to allow for the tested dimensions of the specimens after surface preparation" in BS EN 772–1 [51]. Since it is assumed that the specimens are manufactured as blocks, the shape factor *d* is obtained (by interpolation) from "Table A.1" as 0.8. The compressive strengths of the specimens, therefore, as given in Fig. 1 are multiplied by 0.8 to give normalised compressive strengths.

5.3 Application of lime-based blocks in low rise buildings

The normalised compressive strengths of the lime-based samples are given in Fig. 3 along with the minimum normalised strengths of existing masonry blocks at different conditions (from Fig. 2 and Table 2). The minimum specified compressive strength is 2.9 MPa for aggregate or autoclaved aerated concrete and all but five lime-based specimens achieve this compressive strength. This included the two hemp shiv specimens (#20, #21), the latter with PVAc which generally had higher strengths. Both EG specimens were also in this group (#9, #10) along with the 0.5 wt. % nClay, #5. Next highest specified compressive strength was from clay/calcium silicate for Group 1 samples in Condition A at 5 MPa. A further four samples did not exceed this limit including the Control mixture (#1) and specimens with nanomaterials only – 10% wt. nFc (#12), 3% wt. nSiO$_2$ (#4) and 2% wt. nClay (#7). The compressive strength requirement for both the aggregate and autoclaved aerated concrete, conditions B and C was 7.3 MPa whereas the clay and calcium silicate Condition B, Group 1 had similar compressive strength

requirements of 7.5 MPa. In total, a further seven specimens were below this limit consisting of specimens with 5 wt. % nFc (#11), 1 and 3 wt. % nClay (#6, #8), 7 wt. % HF/L (#14), 1 wt. % nSiO$_2$ (#2) and the two 10% wt. hemp fibres (#18, #15), the former with 8% wt. PVAc. The final five specimens had compressive strengths between 7.7 MPa and 9.4 MPa, straddling either side of the clay/calcium silicate blocks, Condition A, Group 2 (8 MPa). Three of the strongest four were fibre based (hemp or glass, #13, #16, #19, #17), the latter three also including PVAc. The 2% wt. nSiO$_2$ (#3) performed exceptionally well as it included just a solo nanomaterial (it was shown earlier [10] that nanosilica acts as a nanofiller and can fill the spaces between the particles of gel of C-S–H in cement. The pozzolanic reaction with the CaOH$_2$ led to an increased C-S–H which increased the strength). It is clear from the results of the normalised compressive strength that although compressive strengths of over 9 MPa is achievable, it is perhaps too much to expect that lime-based blocks with additives can be produced to cover all intended uses as shown in Table 2 (up to 21 MPa). However, the results show that it is possible to produce lime-based products that can be partially used to construct walls of dwellings, be it for one, two or three storeys, and used internally. The results presented in this paper do not take weathering into account, therefore, further research is required on the durability aspects of exposed lime-based materials.

6 Relative Greenhouse Gases (GHG)

Ranking the various samples on strength alone (Sects. 5.2, 5.3, 6.3) gives a sense of the different types of lime-based blocks that can be used to suit the various requirements in BS EN 8103 [6], but it does not take GreenHouse Gase (GHG) emissions of the various specimens into account. This is investigated in the following section to enable optimum specification of lime based materials based on both strength and CO$_2$eq emissions.

6.1 Lime and hemp

A literature search was conducted to determine values of the GHG for hempcrete and the various additives along with a similar value for a reference concrete to enable a comparison to be made. A detailed review into the energy and environmental assessment of industrial hemp for building applications was conducted elsewhere [52] and refers to the values of the GHG of hempcrete that were given by Ip and Miller [53]. It was stated that a Carbon Footprint (CF) study established the life-cycle GHG-emissions associated with a 1 m^2 hemp-lime wall (0.3 m thick) with a 100 year lifetime horizon (100 years was based on the fact that hemp–lime walls are expected to last over

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[1] Updated to BS EN 1996-1-1:2022 and published 30 August 2023' BS EN 1996-1-1:2005 still remains current
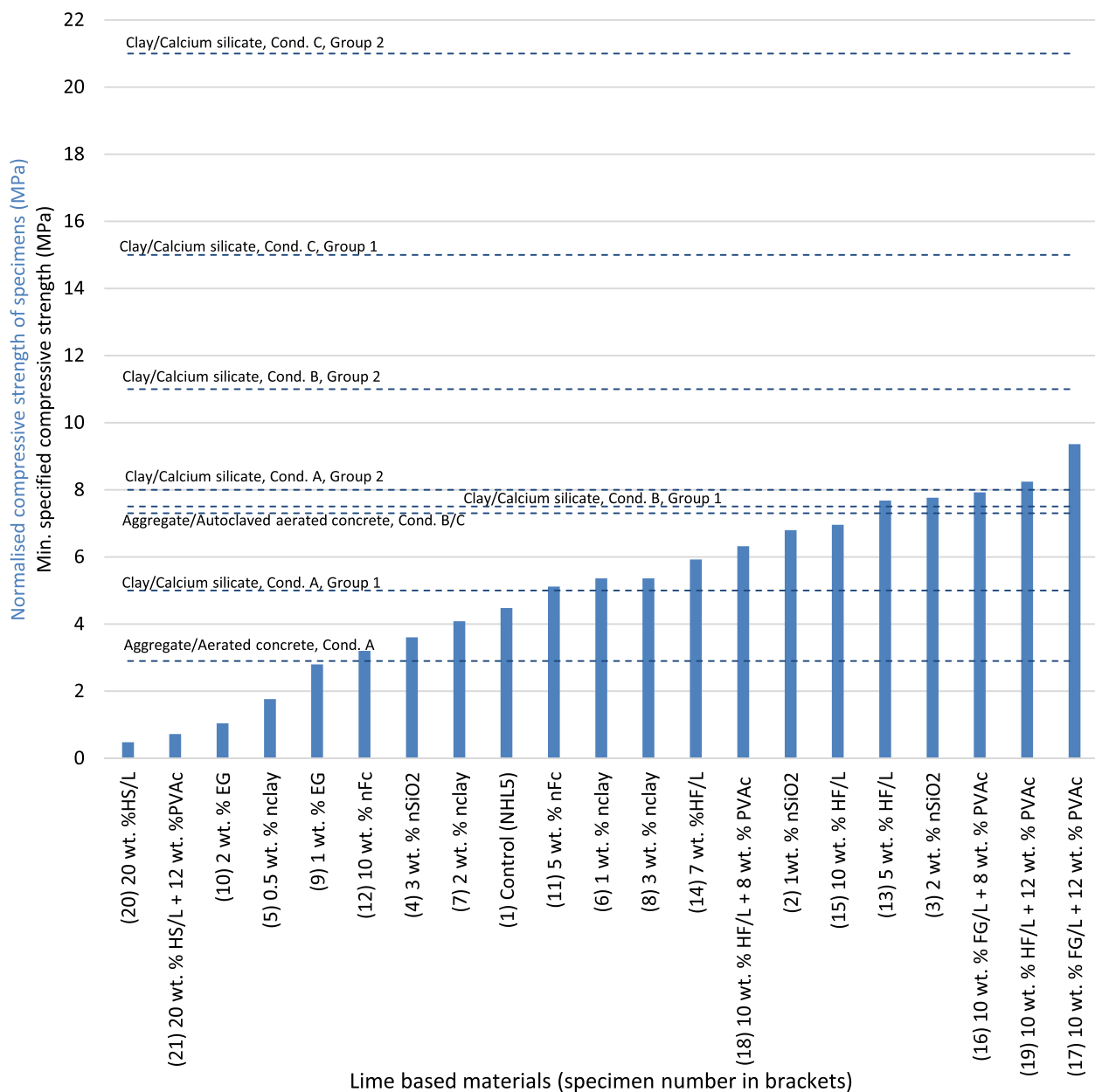
Fig. 3 Normalised compressive strength of lime-based samples (in ascending order) with strengths of existing masonry blocks at various conditions

100 years [54] or similar to that of traditional construction [55]). In addition, this timeframe complies with PAS 2050 [56] and meets the requirements of the assessment framework of the Intergovernmental Panel on Climate Change (IPCC) which bases GHG emissions over a period of 100 years [52]. Three main processes involved in establishing the figures were: i) cultivation of the hemp; ii) hemp processing and iii) construction of a hemp-lime wall. A summary of the positive and negative effects is shown in Table 3. The

main constituents and associated processes as a result of producing hempcrete e.g. lime binder, hemp, timber frame and water are listed for a functional unit ($1 \text{ m}^2 \times 0.3$ m thick) along with the quantities and the emissions, GHG. The transport of the materials to the processing unit and electricity consumed is also included so assumes a cradle-to-gate analysis. The carbon storage as a result of the hemp, timber frame and lime binder is provided in a similar way in Table 3. The analysis shows that 36.08 kgCO$_2$eq was

Table 3 GHG emissions for hemp-lime binder for a functional unit (adapted from [53])

Input	Quantity	kgCO$_2$eq	kgCO$_2$eq/kg
Carbon emissions			
Lime binder – hydrated	37.5 kg	28.7	0.765
Lime binder—hydraulic	7.5 kg	6.48	0.864
Hemp shiv	30 kg	0.18	0.006
Timber frame	0.01 m^3	0.98	
Water	75 kg	0.02	0.00027
Electricity		2.29	
Transport		3.60	
Carbon storage			
Hemp shiv	30 kg	-45.82	-1.527
Timber frame	0.01 m^3	-8.34	
Lime binder	45 kg	-28.55	-0.634
Overall GHG emissions – with timber frame	-36.08 kgCO$_2$eq		

stored per functional unit, determined from a sequestration of 82.51 kgCO$_2$eq and an estimated emission of 46.43 kgCO$_2$eq in the growing and manufacturing of the lime-hemp processes (thereby giving the negative GHG). The same review paper [52] also found that a similar study by Walker and Pavía [57] which again based their findings on a 1m^2 hemp–lime wall but with a thickness of 0.26 m sequesters 14–35 kgCO$_2$eq over its 100-year life span. Another paper [21] reported a GHG of − 108 kgCO$_2$eq/m^3 for hempcrete only without any extra structural supports. This would equate to a pro-rata -32.4 kgCO$_2$eq for the 0.3 m^3 FU by [53].

In this paper, the GHG values for the lime-hemp wall provided by Ip and Miller [53], Table 3 will be used to establish pro-rata equivalent values for a block of arbitrary size of 0.5×0.2×0.3 m which comply with the definition of a block as stated in Sect. 5.1 (and is a size that is commercially available for construction in the UK). A functional unit panel of 1 m^2×0.3 m thick has a volume of 0.3 m^3, the blocks on the other hand have a volume of 0.03 m^3 (0.5 m×0.2 m×0.3 m). The volumetric ratio is, therefore, 10% (100×0.03/0.3) and this will be used to obtain pro-rata emissions where necessary.

6.2 Additives (nanomaterials, fibreglass, PVAc)
The GHG emissions for all additives in the composites (PVAc, EG, nFc, nSiO$_2$, nClay, FG) are given in Table 4. These GHG values in kgCO$_2$eq/kg were converted to kgCO$_2$eq/block and are shown graphically in Fig. 4 (in ascending order of GHG per block). Referring to Table 4, the left-hand section lists the material/additive and

reference and provides a GHG value (in kgCO$_2$eq/kg) for each constituent. On the right-hand section of Table 4, values of GHG are calculated for each individual constituent based on the quantity in the arbitrary block size. The values are algebraically sub-totalled to give a cradle-to gate GHG value for each composite.

6.3 Summary of masonry requirements
The normalised compressive strengths of the lime based samples (Sect. 5.2) and their requirements for use as a construction product (Sect. 5.3) are summarised in Table 5. Referring to Table 5, the Specimens are listed in col. 1 with their normalised compressive strength given in col. 2. The minimum strength requirement is given in col. 3, which enables compliance with none or a number of the five categories as shown in col. 4. The GHG emissions (kgCO2eq/kg) for each specimen are given in col. 5.

6.4 Timber framing and carbon sequestration
Referring to Table 2, it is assumed that specimens with a strength equal or greater than the commonly used autoclaved aerated concrete masonry unit, condition B/C will be assumed to be load-bearing (7.3 MPa). Therefore, all specimens below this level (dashed line in Fig. 4) will be assumed to require timber framing as support, in which case, will utilise the net GHG figures for the timber support as given in Table 3. However, to exclude this negative figure completely for specimens which are load-bearing would not give a true reflection of their eco-friendliness since the absence of a timber support means the tree doesn't need to be felled for processing timber studding.

The type of wood used in timber studding is normally pine, spruce or fir. The carbon sequestration rates of a number of tree species are given elsewhere [64]. Concentrating on say pine trees, the average sequestration is 96.7 kgCO$_2$ per tree per annum (12 species were given). The stemwood volume from a tree of say 30 m×0.3 m diameter is approximately 1.6 m^3 [66]. Therefore, the carbon sequestered by not having to support the lime-based blockwork can be estimated. Research elsewhere [53] used a panel with a functional unit (FU) of 1 m long x 1 m high×0.3 m thick which included two supports, each 100 mm×50 mm. Therefore, it can be assumed that since each block is half the length of the FU (ignoring joints for simplicity), one timber support can be attributed to every block. The thickness is similar (0.3 m) but the height of the block is equal to only 20% of the FU (0.2 m versus 1 m). Therefore, the volume of timber studding apportioned to the block is 0.2 m×0.1 m×0.05 m, or 0.001 m^3. This equates to the equivalent of 0.0625%

Table 4 Influence of mixture constituents on GHG (cradle-to-gate) [53, 58–65]

Material/additive	GHG (KgCO$_2$eq)	Reference	(20) 20 wt. %HS/L	(21) 20 wt. % HS/L + 12 wt. %PVAc	(10) 2 wt. % EG	(5) 0.5 wt. % nClay	(9) 1 wt. % EG	(12) 10 wt. % nFc	(4) 3 wt. % nSiO$_2$	(7) 2 wt. % nClay	(1) Control (NHL5)	(11) 5 wt. % nFc	(6) 1 wt. % nClay	(8) 3 wt. % nClay	(14) 7 wt. %HF/L	(18) 10 wt. % HF/L + 8 wt. % PVAc	(2) 1wt. % nSiO$_2$	(15) 10 wt. % HF/L	Autoclaved aerated concrete masonry	(13) 5 wt. % HF/L	(3) 2 wt. % nSiO$_2$	(16) 10 wt. % FG/L + 8 wt. % PVAc	(19) 10 wt. %HF/L + 12 wt. % PVAc	(17) 10 wt. % FG/L + 12 wt. % PVAc
Hempcrete																								
Lime	0.23	53	2.81	2.81	2.81	2.81	2.81	2.81	2.81	2.81	2.81	2.81	2.81	2.81	2.81	2.81	2.81	2.81	-	2.81	2.81	2.81	2.81	2.81
Hemp fibre	-1.52	53	-	-	-	-	-	-	-	-	-	-	-	-	-1.30	-1.86	-	-1.86	-	-0.93	-	-	-1.86	-
Hemp shiv	-1.52	53	-3.71	-3.71	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Water	2.6E-4	53	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-	0.00	0.00	0.00	0.00	0.00
PVAc	3.36	58	-	5.27	-	-	-	-	-	-	-	-	-	-	-	3.51	-	-	-	-	-	3.51	5.27	5.27
EG	18.3	59	-	-	4.46	-	2.23	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
nFC	1.9	60	-	-	-	-	-	2.32	-	-	-	1.16	-	-	-	-	-	-	-	-	-	-	-	-
nSiO$_2$	5	61	-	-	-	-	-	-	1.83	-	-	-	-	-	-	-	0.61	-	-	-	1.22	-	-	-
nClay	2.5	62	-	-	-	0.15	-	-	-	0.61	-	-	0.30	0.91	-	-	-	-	-	-	-	-	-	-
FG	3.2	63	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	3.90	-	3.90
Electricity	0.22*	53	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	-	0.22	0.22	0.22	0.22	0.22
Transport	0.35*	53	0.35	0.35	0.35	0.35	0.35	0.35	0.35	0.35	0.35	0.35	0.35	0.35	0.35	0.35	0.35	0.35	-	0.35	0.35	0.35	0.35	0.35
Timb-used	-1.34*	64	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3	-	-	-	-	-	-
Timb-avoid	-5.34*	64	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-5.3	-5.3	-5.3	-5.3	-5.3
Total			-1.7	3.6	6.5	2.2	4.3	4.4	3.9	2.6	2.0	3.2	2.3	3.0	0.7	3.7	2.6	0.2	-	-2.9	-0.7	5.5	1.5	7.2
Concrete	0.28	65	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	7.98	-	-	-	-	-
f_c (MPa)			0.5	0.7	1.0	1.8	2.8	3.2	3.6	4.1	4.5	5.1	5.4	5.4	5.9	6.3	6.8	7.0	*7.3*	7.7	7.8	7.9	8.2	9.4

GHG values are kgCO$_2$eq/kg except those marked * which are kgCO$_2$eq/block and are based on an arbitrary block of dimensions 0.50 x 0.20 x 0.30 m
Density of lime taken as 700 kg/m^3 from the product specification data sheet. Lightweight concrete block, 950 kg/m^3. -ve indicate carbon storage; +ve indicate carbon emissions per block

$(100 \times 0.001/1.6)$ of the tree, or 0.06 kgCO$_2$eq to be stored in the tree per annum.

In research by [53], the GHG emissions for processing the timber frame was 0.98 kgCO$_2$eq but the sequestration was 8.34 kgCO$_2$eq, net carbon savings, therefore, being 7.36 kgCO$_2$eq. These figures also indicate that approximately 10.5% of the sequestration is lost as a result of the energy required in post-felling activities. However, it's not clear at what age the tree was when felled and time has an impact on the sequestration process.

Certain species of pine can survive for at least 150 years [67]. However, in some cases, harvesting of the pine trees is done at rotation ages of between 25–30 years [68] with the aim to increase this to 40–50 years to enable better carbon management [69]. Growing pine to even older ages has gained interest due to on-going carbon sequestration. Therefore, if the lifespan of a pine tree is taken as 100 years as per PAS 2050 [56], 6.0 kgCO$_2$eq $(100 \times 0.06$ kgCO$_2$eq) is sequestrated by not having to use timber supports. However, from [53], 10.5% of this would be lost due to its felling/ removal, giving 5.4 kgCO$_2$eq per tree per equivalent studding. On the other hand, if timber supports were to be used, a rotation age of 25 years is assumed so, including felling emissions, 1.34 kgCO$_2$eq $(25 \times 0.6$ less 10.5%) would be sequestered before the tree is felled and processed for timber. These values are shown in Table 4.

6.5 Concrete

With regards to the GHG of concrete blocks, a figure of 0.28 kgCO$_2$eq/kg is given by [65] which is obtained from the Concrete Block Association and included in the ICE Database [70], based on cradle-to-gate analysis. This assumes a lightweight concrete (autoclaved aerated concrete with mix proportions: aggregates 84.7%; cement 8%; PFA; 5% water 2.3%;) with a density of 600 kg/m^3 which is typically used in house building in the UK. In calculating the GHG value in Table 4 and Fig. 4, a search was done on typical lightweight blocks on sale in the UK. The densities ranged between 950 kg/m^3 and 1400 kg/m^3, meaning the mass of the arbitrary block size selected $(0.5 \times 0.2 \times 0.3$ m) would vary. However, a value of 950 kg/m^3 was selected for inclusion in this research resulting in a mass of 28.5 kg. At a GHG of 0.28 kgCO$_2$eq/kg [65], this equates to 7.98 kgCO$_2$eq for the equivalent size concrete block.

7 Relationship between normalised compressive strength and GHG

7.1 Influence of additives on compressive strength and GHG emissions

Referring to the results given in Fig. 4, five different materials exhibited normalised strengths greater than the reference concrete material (7.3 MPa, Aggregated/
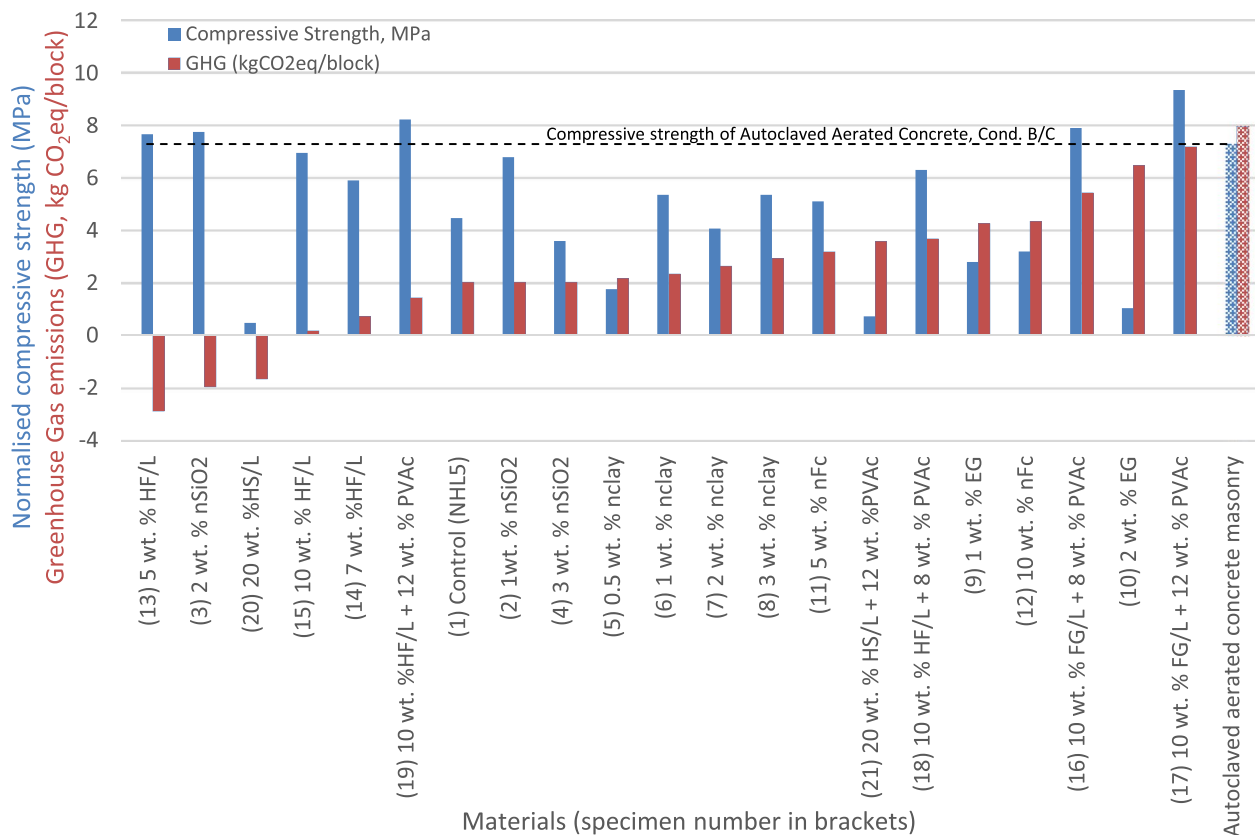
Fig. 4 Comparison between normalised compressive strength and GHG (in ascending order of GHG)

Autoclaved Aerated Concrete, Condition B/C). Three of those materials (#19, #16, #17) all include PVAc and fibres which led to the highest strengths being recorded (7.7 MPa to 9.4 MPa), but the GHG varies considerably since one of the three (#19) includes hemp fibres (lower GHG) whereas the other two contain fibre glass (#16, #17, higher GHG). Although PVAc has a relatively high GHG (5.4 kgCO$_2$eq/kg, see Table 4), this is offset by the combination of carbon storage additives, especially the hemp fibre and avoidance of timber in the load bearing material (a value of 3.36 kgCO$_2$eq/kg was also determined for PVAc [71] but the worst-case scenario was adopted). The other two materials with a normalised strength greater than that of the reference concrete (7.3 MPa) both exhibited negative GHG, #13 had 5 wt. % hemp fibres (7.7 MPa) whereas #3 had an exceptionally high strength (7.8 MPa) considering it was fibreless and contained 2 wt. % nSiO$_2$. The GHG of nSiO$_2$ used was 5 kgCO$_2$eq/kg [61].

A total of seven materials had a normalised strength between that of the Control sample (#1, pure lime using NHL 5, 4.5 MPa and GHG of 2.05 kgCO$_2$eq) and the reference concrete (7.7 MPa). The four strongest of these seven materials all contained either hemp fibre (#14,

#15, #18, the latter also containing PVAc) or nSiO$_2$ (#2). However, although two of the three nSiO$_2$ materials (#2, #3) performed well with regards to strength (6.8 MPa and 7.8 MPa respectively), only the 2 wt. % sample (#3) had a strength higher than the reference concrete material and, therefore, benefitted from a higher carbon storage (timber avoided, -5.34 kgCO$_2$eq/block) as opposed to timber used (-1.34 kgCO$_2$eq/block). The remaining three materials with strengths greater than the Control lime contained nClay (#6, #8) and nFc (#11) and all exhibited similar GHG (2.3, 3.0, 3.2 kgCO$_2$eq/kg). A total of eight materials (#20, #21, #10, #5, #9, #12, #4, #7, see Table 5) had strengths less than the Control (#1) and with GHG ranging from -1.7 to 6.5 kgCO2eq/block. The latter three materials comply with the minimum strength requirement (> 2.9 MPa, Aggregate/Aerated concrete, Cond. A) but the former five exhibit strengths which are unusable as load bearing materials. The two materials with the lowest normalised strength both contained hemp shiv (#20, #21), but the addition of 12 wt. % PVAc to #21 only marginally increased the strength but greatly increased the GHG. Based on these results, it can be concluded that the use of hemp fibres yields the best performing materials when compressive strength and

Table 5 Compliance with masonry requirements

1 Specimen	2 Normalised comp. strength (MPa)	3 Min. strength requirement (MPa)	4 Complies with masonry unit type[a]	5 GHG (kgCO$_2$eq/kg)
(20) 20 wt. %HS/L	0.5	-	None	-1.67
(21) 20 wt. % HS/L + 12 wt. %PVAc	0.7			3.60
(10) 2 wt. % EG	1.0			6.50
(5) 0.5 wt. % nClay	1.8			2.19
(9) 1 wt. % EG	2.8			4.27
(12) 10 wt. % nFc	3.2	2.9	1	4.35
(4) 3 wt. % nSiO$_2$	3.6			3.90
(7) 2 wt. % nClay	4.1			2.65
(1) Control (NHL5)	4.5			2.04
(11) 5 wt. % nFc	5.1	5.0	1, 2	3.20
(6) 1 wt. % nClay	5.4			2.34
(8) 3 wt. % nClay	5.4			2.95
(14) 7 wt. %HF/L	5.9			0.74
(18) 10 wt. % HF/L + 8 wt. % PVAc	6.3			3.69
(2) 1wt. % nSiO$_2$	6.8			2.60
(15) 10 wt. % HF/L	7.0			0.18
(13) 5 wt. % HF/L	7.7	7.3	1, 2, 3, 4	-2.89
(3) 2 wt. % nSiO$_2$	7.8			-0.7
(16) 10 wt. % FG/L + 8 wt. % PVAc	7.9			5.45
(19) 10 wt. % HF/L + 12 wt. % PVAc	8.2	8.0	1, 2, 3, 4, 5	1.45
(17) 10 wt. % FG/L + 12 wt. % PVAc	9.4			7.21

[a] Masonry unit type

1 Aggregate/Aerated concrete, Cond. A

2 Clay/Calcium silicate, Cond. A, Group 1

3 Aggregate/Autoclaved aerated concrete, Cond. B/C

4 Clay/Calcium silicate, Cond. B, Group 1

5 Clay/Calcium silicate, Cond. A, Group 2

GHG are considered, 5 wt. % hemp fibres (#13) being the optimal fibre content.

7.2 Concrete as a load bearing, carbon storage material

Referring to Fig. 4 and Table 4, the inclusion of the carbon storage as a result of timber avoided as opposed to timber used has a big impact on the GHG. This is especially so for materials which benefit from the carbon storage of -5.34 kgCO$_2$eq/block (i.e. compressive strength > 7.3 MPa and is considered load bearing so timber framing is not required). It could be argued that since the reference concrete (see Sect. 6.5) is also load bearing, that, too, could benefit from the carbon storage of timber avoidance and in which case, would reduce its GHG considerably, from 7.98 kgCO$_2$eq/block to 2.64 kgCO$_2$eq/block.

Furthermore, concrete is also known to store carbon via the carbonation process [72] and a paper by [73] analysed the carbon storage of structures containing slag-blended concrete. A 44,000 m^2 building was reported to store 113,000 kg CO$_2$ after 50 years of service, or 2.57 kg CO$_2$/m^2. Relating this to the arbitrary block size of 0.5 m × 0.2 m (ignoring the thickness) gives a sequestration value of 0.26 kgCO$_2$/block for 50 years or 0.52 kgCO$_2$/block if doubled to comply with the Intergovernmental Panel on Climate Change (IPCC) where GHG is calculated over 100 years. The GHG of the block would be reduced even further to 2.16 kgCO$_2$/block. Referring to Fig. 4, this amendment would put it approximately equal with material #5, 0.5 wt. % nClay in terms of GHG.

7.3 Carbon savings for a typical wall

The GHG in Fig. 4 is per single block of dimensions 0.5 m × 0.2 m × 0.3 m. Assuming a 10 mm joint, a wall with dimensions of say 3.57 m × 2.52 m would require 84 blocks (7 × 12) meaning carbon emissions, or carbon savings can be significant. For comparison, the difference

between the block with lowest GHG (#13, -2.89 kgCO$_2$eq) and the gross value of the reference concrete (7.98 kgCO$_2$eq) is 10.87 kg CO$_2$eq/block meaning a difference of 913 kgCO$_2$eq for the 84 blocks used in the arbitrary wall and assuming that the positive influence of the timber avoidance applies only to the lime-based material.

7.4 Analysis of results and selection of optimum materials

It was shown in Sect. 5.1 and Table 2 that different conditions exist which determine the type of masonry to be used for different building sizes (e.g. one, two or three storeys). It was stated in Sect. 5.3 that none of the developed materials underwent durability testing nor were any special attention made to make them weatherproof. Therefore, from that point of view, selection of the optimum lime-based block is founded on higher strength and lower GHG. The compressive strength (7.3 MPa) of the autoclaved aerated concrete masonry was taken as a threshold – the developed materials, therefore, need to at least achieve this strength. Referring to Table 5, the final five materials all achieve this minimum strength. It is significant that four of these materials include fibres, either hemp or fibreglass, and the three strongest materials all contain PVAc in addition to fibres. Although fibres are generally used to limit plastic shrinkage and improve properties such as flexural strength, ductility and durability (particularly freeze–thaw resistance), fibres also have the ability to control crack opening and transfer stress into the adjacent matrix [74]. However, it was reported elsewhere that the short length of the fibres prevent a satisfactory bond with the lime matrix [75]—the length of the hemp fibres in this research was between 5 and10 mm whereas the fibreglass was 6 mm, so can be considered short. Therefore, the addition of the PVAc has had a positive effect in bonding the fibres to the matrix. There is no relevant information in the literature which investigates the use of fibres and PVAc in lime based materials, but there is research which looks at their performance in stabilising soils [76, 77]. In research which investigated the use of polyvinyl acetate in stabilising dune sand, it was found that the polymer physically stabilised the sand via adhering soil particles together. In addition, there was no chemical reaction occurring in the soil [78]. Other research aimed to replace traditional soil stabilizers such as cement found that polypropylene fibres added to sandy soil with additives such as PVAc and clay increased the mechanical strength. This was again reported to be as a result of the adhesion between the particles and PVAc which binds the particles well together, even with fibres present. The incorporation of randomly distributed fibres among the sand grains led to connectivity between the fibres to form a spatial network structure and, thereby, a harder and denser material [79]. The

same, however, cannot be said for material #21 containing hemp shiv and PVAc where the compressive strength was the second lowest at 0.7 MPa. The only other material to make the top five in terms of strength was #3 containing 2 wt. % nSiO$_2$. It was reported in Sect. 3.2 that the nanoparticles of SiO$_2$ fill the spaces between gel particles of Calcium-Silicate-Hydrate (C-S–H), thereby acting as a nano-filler with porosity reduced to a nanoscale [10]. However, two of these materials (#16, #17, Table 5) include fibre glass and PVAc, which lead to a GHG emission of 5.45 and 7.21 kgCO$_2$eq/kg, respectively, and as a result can be disregarded as an optimum material. Therefore, the optimum selection are materials #13 (5 wt. % HF/L) and #3 (2 wt. % nSiO$_2$) which both exhibit negative GHG emissions, -2.89 and -1.96 kgCO$_2$eq/kg, respectively, and both achieve the minimum strength requirement (7.7 and 7.8 MPa respectively). A marginally higher strength (8.2 MPa) was achieved for material #19 (10 wt. % HF/L + 12 wt. % PVAc), but it had the trade off in that the positive influence of the hemp fibre was offset by the negative impact of the PVAc leading to GHG emissions of 1.45 kgCO$_2$eq/kg.

The third best group of materials (see Table 5) all have a compressive strength greater than 5.0 MPa and, therefore, are suitable as a replacement material for Aggregate/Aerated concrete, Cond. A, and Clay/Calcium silicate, Cond. A, Group 1 (referred to as Types 1 and 2 in the table). Some of these materials (#14, #18, #2, #15), which happen to be the four strongest materials in terms of compressive strength, benefit from the inclusion of hemp fibres, nSiO$_2$ and PVAc as described above.

The remaining three materials in this category include nFc and nClay and yielded respectable compressive strengths despite the absence of fibres (and PVAc). In research investigating the influence of polypropylene fibres and nanoclay on the strength of lime treated soil, nanoclay promoted the reaction between lime and soil and produced cementitious materials such as hydrated calcium silicate and hydrated calcium aluminate. The generated cementitious material and nanoclay had the benefit of filling the pores between the soil and the fibre and, thereby, increasing the interfacial friction between the soil and the fibre [80]. The nClay materials, therefore, would be likely to benefit from the inclusion of hemp fibres giving a likely higher compressive strength and possible compliance with the threshold strength (7.3 MPa). nFc, on the other hand, had a strength only marginally greater than the Control NHL 5 lime (5.1 versus 4.5 MPa). The slight increase in strength, however, can be attributed to the microstructural changes that the fibres produce in composite materials and were already understood in concrete with Portland cement. These microstructural changes can be accredited to the

increase in elastic modulus of the crystalline portion of cellulose nanomaterials and their specific surface area. Moreover, the interaction of the surface –OH groups with CSH and portlandite gave an enhanced fibre–matrix bond [81]. Similar to the nClay, the nFc is also likely to benefit from the inclusion of hemp fibres from a strength point of view but their inclusion in the first place can be questioned since material #13 (5 wt. % HF/L) already achieves a strength greater than the compressive strength threshold (7.3 MPa).

The final additive to be considered is expanded graphite, which can be ruled out as a beneficial additive due to its detrimental influence on compressive strength and its high GHG emissions and is used, for example, as a conductive additive in cementitious material [82].

8 Discussion on future studies

The research conducted in this paper was challenging in that it not only attempted to develop load bearing lime based materials, but also considered the carbon footprint of the strengthened lime composites. The main analytical challenge was establishing the carbon footprint of the additives and further work could be done to ensure up-to-date values are available. An example of where variances occur was the carbon footprint of $nSiO_2$. An extremely low value of 0.00084 $kgCO_2eq/kg$ was determined for olivine $nSiO_2$ [83] and in terms of fine aggregate (silica sand), the value was 0.0139 $kgCO_2eq/kg$ [84]. These values appeared out with the values given for the carbon footprints of other nanomaterial e.g. nFc and nClay had carbon footprints of 1.9 and 2.5 $kgCO_2eq/kg$ respectively [60, 62]. Therefore, the value used in the end was 5 $kgCO_2eq/kg$ as this was more in line with other values and is the carbon footprint for silicon dioxide as a food additive [61] but fine tuning of these footprints is required. Regardless, the 2 wt. % $nSiO_2$ composite yield a very impressive compressive strength with a negative carbon footprint and this can be enhanced even further with a review of the carbon footprint of the $nSiO_2$ itself.

The inclusion of PVAc also had a very positive impact on the compressive strengths of the composites. However, despite previous research investigating its impact on the consolidation of sandy soils, its mechanical performance when combined with lime and fibres requires further attention. The chemical reaction between the nClay, nFc and lime also deserves further attention as research to date investigated their interaction between the additives and soil as described in Sect. 7.4.

Future work will also include investigating the use of hemp fibres of different lengths, with a view to further improve the performance of the material since short fibres may not effectively bond with the matrix [75] and

prevent the development of a spatial network structure [79]. Furthermore, the possibility of combining fibres and nanomaterials will also be investigated.

Finally, further research work will also include looking at the sustainability of the proposed materials over their entire lifespan. This will involve considering maintenance, durability, and end-of-life strategies. The effects of the additives on the durability of the proposed products will be investigated by assessing their resistance to freeze–thaw and salt crystallisation cycles, as well as to accelerated weathering.

9 Impact

The results of this work show that it is possible to produce lime-based products that can be used to construct walls of residential and non-residential low-rise buildings, for both new build and home improvement projects. Two of the best performing materials (i.e. #13 and #3, Table 5) have the potential to provide an environmentally responsible alternative to more conventional construction materials, and to contribute to achieving the target set by the UK Climate Change Act to reduce net carbon dioxide emissions by at least 80% from 1990 levels by 2050 [85]. Furthermore, by promoting the use of renewable crop materials in construction, such as hemp, the use of the proposed materials could benefit rural economies through new agricultural markets for farmers and associated industries.

10 Conclusions

The following are the main conclusions emanating from this study. The strengths are obtained from $40 \times 40 \times 160$ mm specimens whereas the GHG values are based on an arbitrary block size of $0.5 \times 0.2 \times 0.3$ m:

- The minimum specified compressive strength is 2.9 MPa for aggregate and autoclaved aerated concrete. All but five samples achieved normalised compressive strengths greater than these meaning more of the conditions in BS EN 8103–2 can be met
- Taking the compressive strength of autoclaved aerated concrete as a cut-off for loadbearing (7.3 MPa), samples with 5 wt. % HF/L and 2 wt. % $nSiO_2$ both exhibited normalised strengths greater than this and both with negative GHG (-2.89 and -0.7 $kgCO_2eq$). Only sample 20 wt. % HS/L also exhibited negative GHG emissions but its strength was negligible
- With regards to the addition of nanomaterials only, specimens containing 2 and 1 wt. % $nSiO_2$ exhibited the highest normalised compressive strength (7.8 and 6.8 MPa respectively). Next highest was 1 and 3 wt. % nClay, both at 5.4 MPa followed by 5 wt. % nFc at 5.1 MPa. The 1 wt. % $nSiO_2$ also benefitted

from a negative timber avoidance since its strength was greater than that of the reference concrete

- Specimens containing fibres (glass and hemp) all exhibited normalised compressive strengths of at least 5.9 MPa. However, those also containing PVAc generally had even higher compressive strengths, the highest value recorded was 9.4 MPa (10 wt. % FG/L + 12 wt. % PVAc). However, a similar sample with HF instead of FG recorded a normalised compressive strength of 8.2 MPa. PVAc and FG had GHG values of 3.36 and 3.2 $kgCO_2eq/kg$ respectively whereas HF was carbon negative at -1.52 $kgCO_2eq/kg$ giving a GHG of 7.2 $kgCO_2eq/kg$ as opposed to 1.5 $kgCO_2eq/kg$ when glass fibre was replaced by hemp fibre

- The use of timber framing has a positive effect with regards to GHG for non-loadbearing walls (-1.3 $kgCO_2eq/block$). This can be increased to -5.3 $kgCO_2eq/block$ if the use of timber is avoided altogether and assumes a 100-year lifespan for carbon storing trees as per IPCC guidelines

- If concrete is assumed to absorb carbon and performs in-service without the use of timber framing as is normally the case, the GHG can be shown to reduce from 7.98 to 2.68 $kgCO_2/block$. However, this is still higher than the best GHG performing materials with a higher compressive strength (5 wt. % HF/L and 2 wt. % nSiO2) at -2.9 and -0.7 $kgCO_2eq/block$

Authors' contributions

All authors contributed to the study design. Laboratory preparation and data collection was done by Khalaf. Research linking enhanced strength to GHG emissions was conceived and conducted by O'Flaherty. Other data analysis were performed by O'Flaherty and Starinieri. Drafts of the manuscript were written by O'Flaherty with previous versions reviewed by Starinieri. All authors read and approved the final manuscript. For the purpose of open access, the authors have applied a Creative Commons Attribution (CC BY) licence to any Author Accepted Manuscript version arising from this submission.

Funding

Availability of data and materials

All relevant data is presented in the main manuscript, any other data will be made available upon reasonable request.

Declarations

Competing interests

The authors have no relevant financial or non-financial interests to disclose.

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