Sustainable Zero-Portland Cement Limecrete Produced from Binary Ground Granulated Blast Furnace Slag and Natural Hydraulic Lime as an Alternative to Standardised Concrete

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ABSTRACT

Concrete, a ubiquitous material in modern construction, faces several fundamental issues, including the cement industry's 8-10% anthropogenic CO_2 emissions (CO_{2e}), that can compromise its sustainability. Therefore, this paper explores novel material combinations of lower carbon binders. Performance issues considered were: volumetric stability; durability; characteristic strengths; environmental impacts; workability; and placement. To address these issues, innovative material combinations of Natural Hydraulic Lime (NHL) and Ground Granulated Blast-furnace Slag (GGBS) are suggested as promising alternatives to traditional cements. Recent changes to BS8500 have allowed for further ternary systems that use GGBS and calcium carbonate thereby giving increased importance to both as ingredients. Combining NHL5 and GGBS can enhance the sustainability of concrete by reducing CO_{2e}, improving resistance to chemical attacks, and maintain overall structural integrity, whilst preserving desirable workability and aesthetic qualities. This research shows the peak mass replacement range of NHL and GGBS in the binary cementitious system at conventional concrete mix ratios, building upon and filling some of the empirical and data gaps. GGBS was used because of its low CO_{2e}, direct cementitious qualities, and to reduce industrial waste. The NHL5 content in concrete was replaced at 10% and 20% increments up to 100% GGBS in concrete to assess the physical properties and mechanical performance. Analysis of compressive and flexural strengths at varying curing ages of 7,14, 28, 91 and 180 days, were conducted for the standard mix ratios of 1:1:2, 1:1:3, 2:1(1:2) and 2:1(2:1). Two curing conditions were examined at 91 days of curing, being submerged in water and in ambient conditions. Increased mechanical performance was produced using a 1:1:3 mix ratio, with the optimum replacement values occurring between 40-60% replacement for all ratios, with the optimal replacement value at 48% and carbon intensity point at 32%, representing the peak mass replacement range and points thus providing evidence and supporting the assertions made from thermodynamic models. The highest compressive and flexural strengths achieved at 31MPa and 2.0MPa by 1:1:3, water cured 40/60, and air cured 60/40, NHL/GGBS samples respectively, being significant gains in strength when compared to either the pure NHL or GGBS binder control concrete samples.

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Introduction and Background

Typical Portland cement is produced from calcined and sintered limestone and clay and other minerals that are heated to a high temperature in a kiln and then ground into a fine powder consisting of the compounds, found in their typical compositions, Table 1.
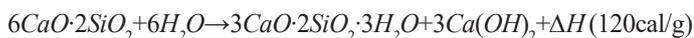
Table 1: Typical Bogue Phases [1,2].

Component	Mineral phase	Cement notation	% of OPC	% of NHL5	Oxide Composition
Tricalcium silicate	Alite	C_3S	60	trace	$3CaO \cdot SiO_2$
Dicalcium silicate	Belite	C_2S	16	45	$2CaO \cdot SiO_2$
Tricalcium aluminate	Aluminate	C_3A	10	2	$3CaO \cdot Al_2O_3$
Tetracalcium aluminoferrite	Ferrite	C_4AF	8	2	$CaO \cdot Al_2O_3Fe_2O_3$

Concrete is an artificial composite rock and is the most broadly used construction material second only to water as the most consumed material generally [3-5]. In 2021 the production of cement, the primary component of concrete totalled 4.3 billion tonnes [6]. Prior to the advent in 1824 of Aspdin's modern Portland cement, Hydraulic Lime was the predominant binder material [7,8].

It is generally accepted that the cement industry accounts for a significant proportion of anthropogenic GHG emissions at 5%, and 5-8% of global CO_2 emissions, with some literature referencing as high as 10-13%, more than three times higher than that of the entire aviation industry [9-18]. There is significant cost associated with cement production with 2% of EU28 block GDP spent on this one activity in 2016, more than the estimated economic growth of 1.9% or the 1.3% of GDP average spent on defence in the block during that year [19-21].

Cement in Europe is typically divided into five classifications; CEM I, CEM II, CEM III, CEM IV and CEM V [22]. CEM I is the closest representation of modern ordinary Portland cement (OPC) being 95-100% OPC clinker with up to 5% minor additions inter-ground, typically gypsum, to prevent flash setting of C3A, whereas the others (CEM II-V) are composite cements containing various other constituents and combination thereof, such as GGBS, silica fume, pulverised fuel ash, limestone fines and other natural pozzolana. These substituted cements typically have lower early age strengths but can develop higher strengths in the long term [23-26].

The material when combined with water undergoes a chemical reaction known as the hydration of cement, which induces the formation of crystalline structures in a gel matrix with the material hardening and strengthening overtime. The hydration kinetics are controlled by the dissolution of the binder materials, diffusion of the reactants and availability of nucleation sites for the growth of the hydrate products [27].

The time to maturity and relationship with curing temperature, environment, and SCMs have long been discussed with various models of cement hydrations produced for OPC concretes [28-34].

Standardised approaches of testing concrete in industry use a relatively narrow range for maturity comparison, an example, having 28-days of curing in $\geq$95%RH at 20± 2°C as a datum, with ambient conditions of 40-60%RH at 20± 5°C compared in this study [35].

It is generally accepted that in ambient conditions OPC based concretes typically achieve most of the ultimate strength within 28days, with little gains thereafter [26,36,37]. The traditional timeframe of 28-days, other than serving as a reference point for description, characterisation, comparison and factorisation, has no intrinsic scientific significance [26]. However some literature has referenced up to 99% of ultimate strength being achieved in this time frame [38].

NHL and OPC Cements

OPC and NHL are produced from the thermal decomposition of limestone. The calcination of limestone to lime, Equation 1, during both NHL and OPC production is identical, resulting in the same amount of CO_2 released and requiring the same amount of energy, the optimal temperature for this has been found to be around 900°C [39-41]. Whereas calcium silicates and aluminates

are produced during clinker calcination at a range of between 850 to 1200°C [42-44]. At this temperature and with the additions of silica, at 65% and above of CaO, belite is produced in addition to lime, highlighted in Figure 1. The sintering temperature required to produce OPC exceeds 1250°C in order to produce alite and the optimum temperature has been found to be 1340°C [45,46].

$$CaCO_{3(s)} \rightleftharpoons CaO_{(s)} + CO_{2(g)} \quad \Delta H_{298K} = \pm 178.8 kJ/mol \ (426 \ cal/g)$$

Equation 1: Calcium Carbonate Decomposition [41].

Figure 1: Clinker Phase Formation Diagram for CaO-SiO_2 [45].

The difference between OPC and NHL production are the quality, quantity and combination ratios of the precursors to clinker firing, the firing temperature and cooling times. The firing temperature is of particular interest as this results in different calcium silicate minerals, including different belite polymorphs, Figure 2, requiring different amounts of energy and thus CO_{2e} from fossil fuel consumption and resulting in differences in stoichiometry.

Figure 2: Belite Polymorph Transformation Temperatures [47].

Natural hydraulic lime is produced from the calcination of marly or calcareous limestones that include 20-30% naturally occurring clay inclusions that provide the alumina and silica.

NHL is classified into three grades, NHL 2, NHL 3.5, and NHL 5, based on their compressive strength and hydraulicity. NHL 2 is the weakest and least hydraulic, while NHL 5 is the strongest, most hydraulic and has the highest leaching resistance of the NHL classifications, which are numbered by the compressive strengths achieved by 28 days [48].

The Bogue phases of NHL5 account for 49%, Table 1, of the mass compared to the 94% of OPC, with the majority of the remaining being a combination of unburnt calcium carbonate ($CaCO_3$) and free lime ($Ca(OH)_2$).

Significant difference in Portlandite and calcium silicate hydrate (CSH) production exists when considering belite, Equation 2, and alite, Equation 3, hydration, with the latter at 3 times that of the former for Portlandite, producing nearly double the heat and requiring a third more water.

Equation 2: Belite Hydration

$$4CaO \cdot 2SiO_2 + 4H_2O \rightarrow 3CaO \cdot 2SiO_2 \cdot 3H_2O + Ca(OH)_2 + \Delta H \ (67cal/g)$$

Equation 3: Alite Hydration

$$6CaO \cdot 2SiO_2 + 6H_2O \rightarrow 3CaO \cdot 2SiO_2 \cdot 3H_2O + 3Ca(OH)_2 + \Delta H \ (120cal/g)$$

The hydration of alite is considered to be responsible for the majority of the strength development in conventional Portland cement based concretes, which is predictable given the bogue phase breakdown, Table 1, and CSH production from alite hydration, Equation 3.

The reaction of the belite mineral is over a much longer period compared to the more reactive alite and although it does contribute a small amount to the early age strength of OPC concrete, its mainly responsible for enhancing the long-term strength development of the concrete that can go on for years, known as densification [49].

Direct Cement Replacement
There is a considerable difference between the embodied carbon of 635kgCO$_2$/t for NHL5 and 819kgCO$_2$/t for PC. This is not taking into account the difference in carbonation between the two materials that would result in the absorption and offset of CO$_2$ from production, reducing the NHL5 figure to 415kgCO$_2$/t [50]. However, this may not be the case when NHL5 is used in conjunction with other SCMs as the carbonation characteristics change.

These embodied carbon figures can be compared to other cement replacement materials and concrete components, Table 2.

Table 2: Comparison of CO$_{2e}$ of cement and SCMs

Study-Material:	[51]	[52]	[53]
	Embodied CO$_2$ (kgCO$_2$/tonne)		
Portland Cement (CEMI)	930	913	860
Ground Granulated Blast Furnace Slag (GGBS)	52	67	79.6
Pulverised Fuel Ash (PFA)	4	4	0.1
Limestone Fines (LF)	32	75	8

Ten GGBS samples from literature were compared, variations in the major oxide compositions of up to 10% in CaO and SiO$_2$ and 15% in Al$_2$O$_3$, were noticed, with large differences in composition when compared to OPC, with on average twice the alumina, a third more silica and less CaO by around 20% [54-59]. A significant difference to OPC cement was the larger proportion of Magnesia (MgO) within most of the GGBS examples, up to near 11% in two samples [55,57].

Eight papers for NHL5 where compared, variability is noticeable in NHL, Significant differences can be seen in ferric and magnesium oxides, particularly in samples and, respectively [60,64]. Standing out as having more than double that of any other, however as a percentage of mass only accounting for, at maximum, less than eight percent [58,60-66]. Lesser but still significant variability is noticeable in the minor oxide compositions of K$_2$O and Na$_2$O, with coefficients of variance at 57.9% and 63.1% respectively. The loss on ignition (LOI) for NHL ranged from 15-20%. The largest mass percentage variance was in CaO with a range from 42-85% of the total mass.

Ternary combinations including cement, NHL and GGBS or other pozzolanic materials have been investigated in the past with many showing performance benefits [58,67-73].

In the European CEM family of cement, CEMII, along with CEMIII and CEMV all incorporate GGBS in various proportions from 6-20% for CEMII/A-S to a maximum of 81-95% for CEMIII/C [22]. GGBS is also used to produce super sulphated cements (SSC), geopolymers and alkali activated materials (AAM) [74,75].

The inclusion of GGBS in NHL-based concrete can lead to improved compressive and flexural strength, as well as allowable durability, while remaining highly permeable [2,76-78].

E.R. Grist and colleagues investigated the potential of natural hydraulic lime-pozzolan concrete (HLPC) and mortars at binder proportions of 50%, 70%, and 85% NHL5, with varied proportions of pozzolana and SCMs in binary and ternary combinations, showing the potential of the material [2,58,77-79]. The binary combination of NHL5 and GGBS was described once in a mortar as a 15% replacement of NHL5 with GGBS in a 1:3 binder to fine aggregate mass ratio and a 0.5 water to binder (w/b) ratio, showing a considerable increase in compressive strength [58]. The later research suggests that combinations of GGBS, and silica fume (SF) can further replace the NHL5 up to 50% in the binder, with concrete compressive strengths of up to 30.5MPa achievable in water and 13.4MPa in ambient conditions [77].

Thermodynamic modelling simulations were performed for the blend of GGBS and NHL5, suggesting that the CSH gel proportion of the hydrate products is highest at 60% GGBS, between two distinct stages of replacement associated with a change in AFm-AFt phases, fully utilised portlandite, and a Ca/Si ratio of 1.91, suggesting this may be the optimal mass replacement for the highest strength [80]. A peak ternary replacement value was suggested at 77% using GGBS and SF for reduced embodied carbon and carbon intensity [2,78].

A study concluded that the carbonation resistance of HPLC is proportional to w/b at different rates depending on pozzolana, and was less than that of conventional CEMI concrete resulting in a higher potential for reinforcement corrosion to occur [77].

The durability of the material and degradation of the mechanical performance occurring via environmental conditions, such as leaching, freeze-thaw action, carbonation, chloride penetration, and sulphate attack are all heavily influenced by the pore structure and permeability of the material, in turn heavily influenced by the w/b ratio, development of hydration products and inclusion of pozzolana and filler materials.

Overall, the addition of GGBS to NHL-based concrete can lead to improved strength and durability properties. However, the optimum dosage and type of SCM may vary depending on the specific application and environmental conditions.

This research sought the peak mass replacement range in the binary binder of NHL5 with GGBS and provide an alternative to conventional OPC concrete of a low strength class for structural grade applications at reduced CO$_{2e}$. Use cases targeted include: unreinforced floor slabs; kerbing; haunches; blinding; unreinforced bases; pad foundations; unreinforced strip footings; and mass-fill concrete.

Experimental Preparation and Sample Production

The experimental programme consisted of the preparation, production, curing, and testing of concrete samples. The tests performed included: chemical analysis and particle size distribution of the component materials during preparation; slump tests for workability during sample production, and: density; flexural strength; and compressive cube testing of the final cured composites [81-85].

Materials

The NHL material, conforming to BS EN 459-1:2015[86], was manufactured in Portugal by Secil, distributed in the UK by Sington Birch and supplied by the merchants Conserv®. The bulk density was determined at 838kg/m³ increasing to 978kg/m3 post vibration consolidation.

The GGBS, identification code: Scunthorpe-GGBS-20230101 conforming to BS EN 15167-1:2006[87], was produced and supplied in the UK by LKAB. The material has a verified to ISO 14025, global warming potential (GWP-fossil), cradle to gate, of 74.4 kgCO$_{2e}$/tonne as declared in the HUB-0602 Environmental Product Declaration (EPD) as per BS EN 15804:2012+A2:2019 [88-90]. This is on the high end of what is found in Table 2. The bulk density was determined at 1011kg/m³ raising to 1224kg/m³ after consolidation under vibration.

The mineral compositions of the materials have been investigated by way of X-Ray: fluorescence (XRF); and diffraction (XRD) tests, Table 3 and Table 4, with primary and residual oxide compositions close to that found in the literature, Figure 3 and Figure 4 [91].

The NHL5 having more than the average by 8.9% burnt lime, 1.9% more silica, 1.1% less MgO, with all other differences less than 1%. The GGBS has more burnt lime, silica and alumina by 5.1%, 2.7% and 1.3%, respectively, and less MgO by 7.4%, with all other differences less than 1%.

Table 3: Oxide Analysis of Constituent Minerals by XRF

	Amorphous oxide analysis (weight %)										
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	SO_3	P_2O_5	Cl-	LOI
NHL5	16.90	4.62	2.92	71.00	1.38	0.83	0.18	1.35	<0.1	<0.1	18.80
GGBS	39.40	11.1	0.31	44.70	1.46	0.43	0.11	1.49	<0.1	<0.1	0.20

N.B. L.O.I. performed at 975°C

Table 4: Crystalline Mineral Phases by XRD

	Crystalline compound analysis (weight %)			
	$CaCO_3$	$Ca(OH)_2$	Ca_2SiO_4	SiO_2
NHL5	38.70	28.70	28.40	4.10
GGBS	100			

N.B. GGBS sample mostly amorphous
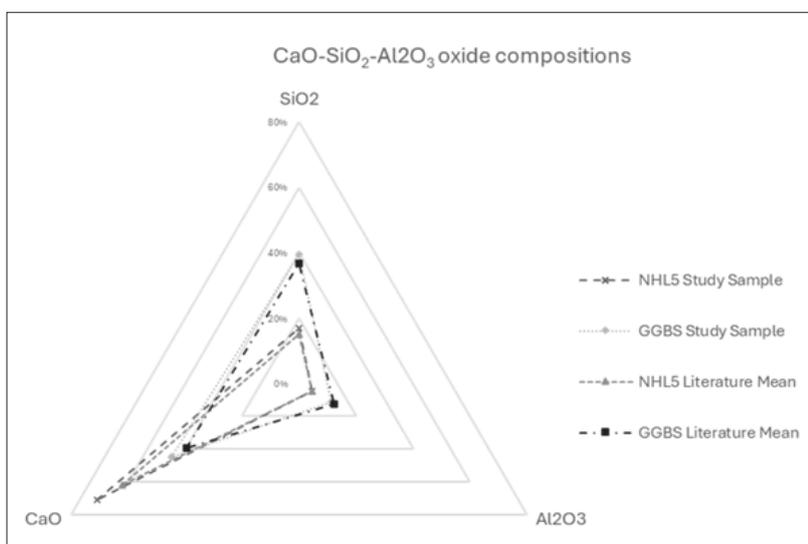
Figure 3: NHL5 and GGBS Ternary Oxide Compositions [54-66].
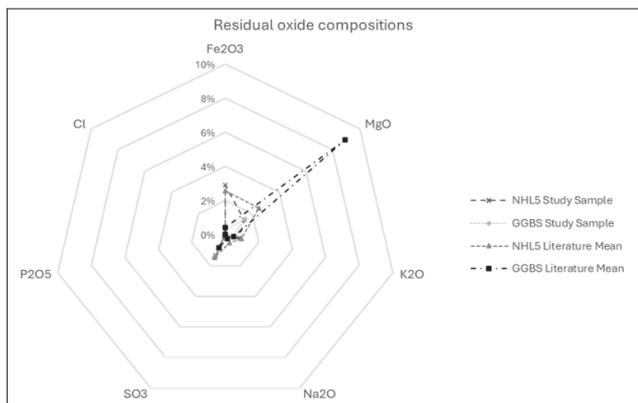
Figure 4: NHL5 and GGBS Residual Oxide Compositions [54-66].

The coarse aggregates (C.Agg) used for concrete production in this study included a quartzite rock described as dark/light brown, grey, white of a rounded and crushed shape graded to 10-20mm in size used for applications such as in concrete, landscaping and filter media sourced by Chas Long & Son Aggregates based in Richmond, North Yorkshire, and extracted from Hackforth Quarry.

The particle size distribution was obtained experimentally using the method stated in BS EN 933-1:2012, Figure 5, using the equipment of BS EN 933-2, having been reduced using the riffle box method in BS EN 932-2 and equipment of BS EN 932-5 [81,92-94]. This process included using sieves complying to ISO 3310-1 and part 2, with additional intermediate sieve sizes to the standard range to increase the data resolution [95,96].

Comparing the binder particle sizes of the NHL5 and GGBS as received by the same sieving method used for the aggregates shows that both have very small particle sizes with 100% passing the 500µm sieve, although the GGBS is much finer, where only 0.7% remain in the pan post 45µm sieve for NHL5, 66.86% remain for GGBS. However, as only 5-6 sieves were used to capture the data, more suitable methods and tools should be sought, such as laser diffraction, optical imaging, or electron microscopy.

On investigation of the British Standards for 'Aggregates for concrete' it was found that the most recent BS EN 12620:2013 has been withdrawn and not replaced by any newer version. Instead, the standard which it replaced, BS EN 12620:2002+A1:2008 has the status of 'Work in hand, Current'. However, the categories used in this paper adhere to the former (most recent) document as they allow for higher precision when categorising for fines content within both coarse and fine aggregates to no other detriment.

The coarse aggregate has a fineness modulus of 6.75, a coefficient of uniformity of 1.62 and coefficient of curvature of 1.0, which

results in the aggregate being considered a uniformly graded medium/coarse gravel, having 1% fines and 2.8% fine aggregate incorporated, Figure 5. The 13% retention on the 20mm sieve, 91% retention on the 10mm, and a three quarter percent on the 63µm sieve indicates the aggregate to meet the Category Gc85/15f1 [97]. The coarse aggregate bulk density was found to be 1365kg/m^3 increasing to 1472kg/m^3 post consolidation under vibration.
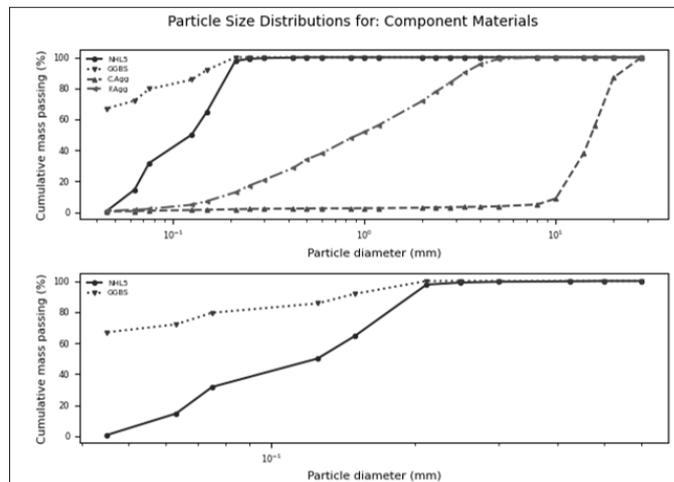
Figure 5: Component Particle Size Distributions

The fine aggregates (F.Agg) used for concrete production in this study included a washed quartzite sharp/grit concrete sand described as dark/light brown of an angular/rounded shape of 0-4mm in size used for applications such as in concrete, screed, bedding or mortar. It was extracted from Hackforth Quarry.

The rectilinear curve between 0.125-4mm, Figure 5, coefficient of curvature, and uniformity, were found to describe the grit sand as well graded, category GF85f3 as per BS EN12620:2013 [97].

Overall, the dry grit sand material resembled 97% a fine aggregate with 1% a coarse aggregate and 2% fines, made of roughly 10% fine sand, 25.5% medium sand, 34.5% coarse sand, and 30% fine gravel. The fine aggregate bulk density was found to be 1243 kg/m^3 raising to 1597kg/m^3 post consolidation under vibration.

The combination of these aggregates resulted in a gap graded aggregate combination with a reduced portion of 4-10mm aggregates resulting in potential for future optimisation.

Sample Production
Four mix ratios were investigated including two conventional concrete binder to aggregate ratios (binder: fine aggregate : coarse aggregate) of 1:1:2 and 1:1:3, and adaptations to the conventional mortar binder to aggregate ratio of 2:1, where the aggregates were subdivided into 2:1 and 1:2 ratios of fine to coarse aggregates, Table 5.

Table 5: Mix Proportions

Mix ID	No. of Sample	Mix Ratio – Binder: Aggregates	Binder Content % (NHL5 & GGBS)	Aggregate Ratio Sand: Gravel	Water to Binder ratio (w/b)
1:1:3	215	1:4	20	1:3	0.4
1:1:2	215	1:3	25	1:2	0.4
2:1(2:1)	102	1:2	33	2:1	0.4
2:1(1:2)	102	1:2	33	1:2	0.4

Concrete mixes corresponding to 34 combinations of the two binders at four aggregate ratios were produced. Two sets (1:1:2 & 1:1:3) at 10% incremental replacement of NHL5 by GGBS and the other two sets (2:1(1:2) and 2:1(2:1)) at 20% incremental replacement. Each mix was used to produce 100mm cube and 100x100x500mm prism samples cast in plastic moulds, having been lightly coated with mineral oil to assist demoulding.

These were used to illustrate differences in the wavelength amplitude and skew associated, with tight and loose, gap-graded, apollonian stacking, and matrix suspension, relating to compressive strength and allowing for easy identification of localised performance peaks.

Despite all the mixes having different water demands when considering water absorption both due to differences in binder SSA and due to aggregate ratios and binder contents, all were produced using the same mass w/b ratio of 0.4, Table 5, resulting in variation in consistency and workability.

Curing Regimes
Samples were cured in both water and ambient conditions to evaluate the influence of curing conditions on compressive strength. All samples followed the same procedure for the initial 3-day period up until demoulding occurred after which some of the samples were submerged in water maintained at 16±3°C and others remained under air-curing conditions within the laboratory environment at 20±5°C at 50±10% RH, for the remainder of the curing periods of 7, 14, 28, 91, and 180 days.

Results and Discussion
Workability
All 1:1:2 samples achieved a slump between 10-50mm, corresponding to slump class S1 with no clear trend associated to the NHL5: GGBS ratio. However, remarks were noted as to the difference in consistency of the two extreme controls with high GGBS described as sticky or tacky and high NHL5 mixes being expressed as like dough [98]. All samples consolidated under vibration satisfactorily. It was noted that the mixes became easier to consolidate toward the 50/50 binder proportions from either the NHL5 or GGBS binder controls.

All 1:1:3 samples failed to achieve adequate slump to be classified by the standard slump test parameters for workability, with all having slump measurements taken as <5mm. However, all samples achieved adequate mechanical consolidation with minimal time on a vibrating table. The same descriptions of the primarily GGBS samples being sticky was given, and primarily NHL samples described as like dough. Additional anecdotal remarks were made upon demoulding and labelling that the primarily NHL5 samples top surfaces were glazed and easy to mark with a pen, whereas all the surfaces of the primarily GGBS samples were slightly chalky/dusty, showed very fine cracks and were slightly absorbent to the pen ink. This would suggest that the surfaces exposed to the air react differently to those encased in the moulds for the NHL5 samples, and that the GGBS samples either were not fully hydrated, experienced shrinkage, surface evaporation, or a combination thereof, during the initial period prior to demoulding. Like the 1:1:2 samples, it was noted that the mixes toward the 50/50 binder proportions consolidated the best under vibration, taking the shortest time and having no externally visible macro voids.

All 2:1(2:1) samples achieved slump classification of S1-S4, Table 6 , These show an increase in slump directly tied to the increase in GGBS proportion to NHL. When compared to the 1:1:2 and 1:1:3 it shows that an increase in the finer binder content to the aggregate, increases workability.

The 2:1(1:2) samples follow the same trend as the 2:1(2:1) however with a larger range, the NHL5 control sample achieved a 30mm slump in the middle of the S1 class and the GGBS control achieving a 190mm slump at the top of S4. A sudden increase in slump value is recognised between the 80% GGBS replacement value and 100% GGBS control, like that occurring with the 1:1:2 mixes.

Table 6: Workability Results

Slump (mm)[class]				
replacement (GGBS %) Mixes				
NHL5 replacement (GGBS %)	2:1(2:1)	2:1(1:2)	1:1:2	1:1:3
0	50 [S1/S2]	30 [S1]	20 [S1]	<5 [n/a]
20	50 [S1/S2]	50 [S1/S2]	30 [S1]	<5 [n/a]
40	90 [S2/S3]	60 [S2]	10 [S1]	<5 [n/a]
60	130 [S3]	60 [S2]	20 [S1]	<5 [n/a]
80	150 [S3/S4]	80 [S2]	20 [S1]	<5 [n/a]
100	160 [S3/S4]	190 [S4]	50 [S1/S2]	<5 [n/a]

Having a wide particle size distribution improves the fluidity and therefore workability of fresh concrete, with higher solid mass fractions and lower viscosity leading to increase in the potential for the minimisation of added water, reduced permeability, and increase load carrying capacity [99].

Density
Density testing was performed to BS EN 12390-7:2019 [83], using the water displacement method (Archimedes' principle). The volume was first calculated using Equation 4, with the density of water taken as 998kg/m^3, assuming a temperature of 20°C.

Equation 4: Volume from Water Displacement

$$V = \frac{m_a - [(m_{st} + m_w) - m_{st}]}{\rho_w}$$

Then the specimen density was calculated using Equation 5, and recorded in kg/m^3.

Equation 5: Density calculation

$$D = \frac{m}{V}$$

The results were as expected given the component densities, PSDs and cementitious material hydration. All but nine samples of the 715 samples tested having density in the normal weight concrete range of 1800-2400 kg/m^3, of which all were very slightly over, six of which were 100% GGBS cement samples, seven of the samples were 91-180 days old, and seven where of the aggregate ratio 1:1:3.

The General Trends Included:
- Increased density over time
- Increased density with increased proportion of total aggregates.
- Increased density with increased proportion of coarse aggregate.
- Increased density with increasing GGBS replacement of NHL5

Compressive Strength

The compressive strength tests were carried out in accordance with BS EN 12390-3-2019, using an IMPACT automatic compression machine, model CT 340, adhering to BS EN 12390-4:2019, Figure 6. After which the maximum load achieved was used to calculate the compressive cube strength, Equation 6 [85,100,101].

Figure 6: Compressive Cube Testing Equipment

Equation 6: Compressive strength

$$f_c = \frac{F}{A_c}$$

Figure 7-10 show the characteristic (average) compressive cube strengths ($f_{ck,cube}$) of NHL5 incrementally replaced by GGBS in the binder of concrete at different aggregate ratios cured in ambient conditions. All aggregate ratio combinations exhibited strengths exceeding the requirements to be classified as at structural strength (10-115MPa), in BS EN 206:2013+A2:2021, for concrete at some percentage of NHL5 replacement with GGBS, with localised peaks between 40-80% at all ages [102]. All combination of NHL5 with GGBS exhibited higher strength than either of the individual materials used as control binders.

NHL5 outperformed GGBS as an individual binder for three of the four aggregate ratios with the exception being that of the 2:1(2:1) mixes in Figure 10.

For all aggregate ratios a significant drop off in strength is observed between the 90% replaced NHL specimens and the 100% GGBS specimens.

The highest average compressive strength achieved by any ambient cured specimen was that of 60% NHL replacement for the 1:1:2 aggregate ratio at 180 days, at 23MPa. The highest average 28-day ambient cured compressive strength achieved was that of the 60% NHL replaced 1:1:3 samples, at 20MPa. The overall highest average compressive strength achieved was that of the 91-day water cured, 60% NHL replaced 1:1:3 samples at 30MPa.

The change from 50% to 60% GGBS represents a ratio drop from 2.06 to 1.82 for Ca/Si, in line with the proposed optimal CSH production from the model and the 55-59% optimal GGBS content derived experimentally in combination with OPC [80,103].
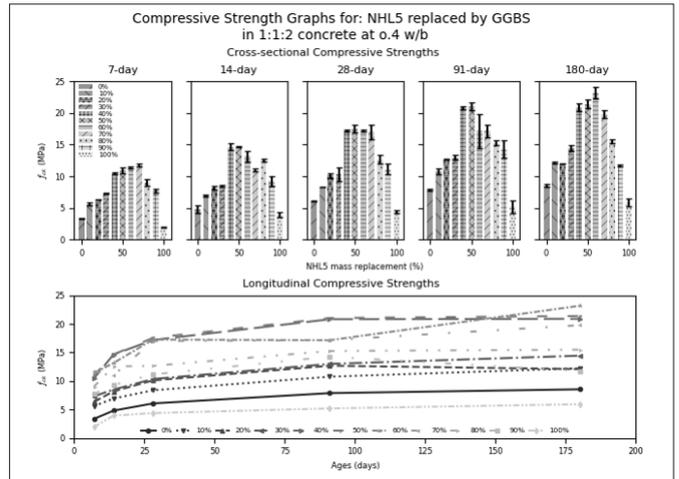
Figure 7: Compressive Strengths of NHL5 Replaced by GGBS in 1:1:2 Concrete at 0.4w/b
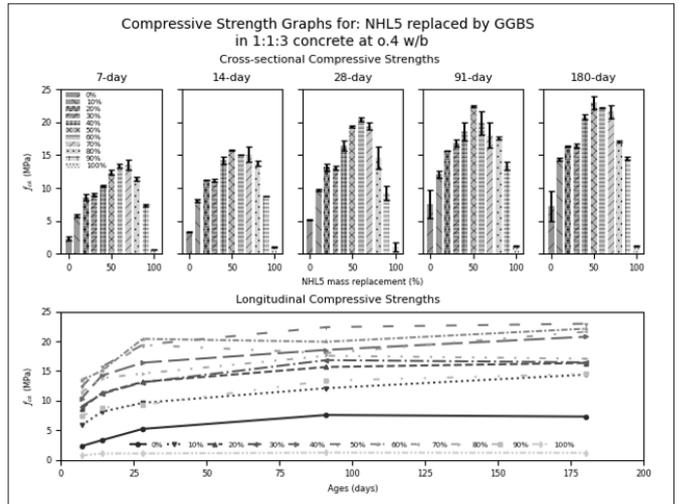
Figure 8: Compressive Strengths of NHL5 Replaced by GGBS in 1:1:3 Concrete at 0.4w/b
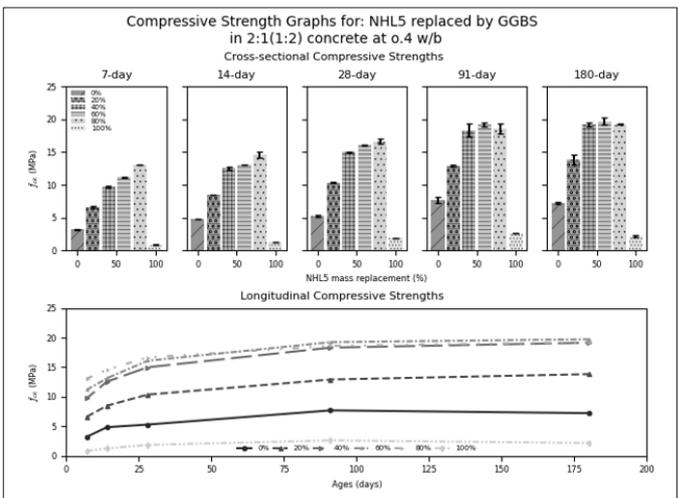
Figure 9: Compressive Strengths of NHL5 Replaced by GGBS in 2:1(1:2) Concrete at 0.4w/b
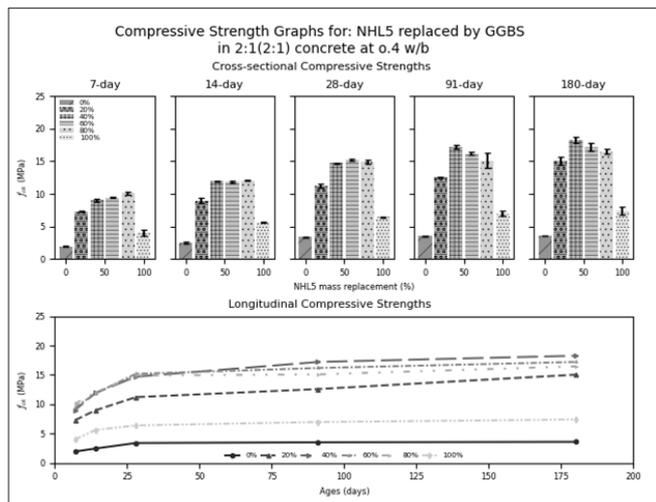
Figure 10: Compressive Strengths of NHL5 Replaced by GGBS in 2:1(2:1) Concrete at 0.4w/b

The NHL5-GGBS composites between 10-90% NHL5 contents for both 1:1:2 and 1:1:3, along with the 20-80% NHL5 contents for both 2:1 mixes showed compressive strengths on par with some of the standardised prescribed concrete (ST) for structural designs [98].

Of the 34 mix designs investigated 26 of them exceeded the assumed 28-day characteristic compressive cube strengths for design of ST1(8MPa), 23 exceeded ST2 (10MPa), 12 exceed ST3 (15MPa), and one exceeded ST4 (20MPa).

Eleven of the water cured 91-day samples from the 1:1:2 and 1:1:3 mixes ranging between 40-70% NHL replacement achieved compressive strengths in excess of the minimum cube strength requirements for class C20/25 being 20MPa. From this one of the samples, from the 60% NHL replaced 1:1:3 ratio 91-day water cured group, achieved a compressive strength of 31MPa achieving the minimum cube strength for strength class C25/30 [102].

The general trends include an increase in strength achieved in the early stages to maturity within the initial 28-day period, with an increasing rate of strength gain with increasing substitution of NHL5 with GGBS up to a roughly 50% replacement level, decreasing thereafter.

Comparing the 7-day strength as a factor of the 28-day strength for each of the mix ratios it can be seen generally 45-80% of 28-day strength is achieved within the first 7 days, with the majority between 60-70%, reducing toward the controls, Figure 11. Showing that the combination of binder materials increases the early age development.

Comparing the 28-day strength as a factor of the 180-day strength for each of the mix ratios, Figure 12, a slight trend toward higher completion of 180-day maturity is achieved with higher GGBS contents, with all samples achieving between 65-95% of 180-day strength by 28-days.

In terms of longitudinal strength progression all mix ratios exhibited general trends of increasing strength with age up to 91days except 1:1:2 and 1:1:3, 60% and 70% replacement samples showing potential dormancy between 28 and 91days. However, both show high errors at 91day data points.

Beyond 91days to 180days: 40%, 50%, 80%, 90%, 1:1:2 samples; 30% and 80%, 1:1:3; and 0% and 100% of 2:1(1:2), exhibit a decrease in strength which can be attributed to either the arrangements of errors from the data points at those time signatures or potential carbonation occurring, or a combination of the two. Whilst 10% and 30% of 1:1:2, and 10%, 20%,40%, 70% and 90% of 1:1:3, do not show a decrease in strength over time.

Many of the ambient cured mixes explored could be utilised in industry for applications akin to the standardised and general-purpose concrete classifications, with the 28-day ambient cured specimens compared to the strength requirements shown in Table 7. However, these classifications are based on water cured strengths with recommended S3 slump classifications for ease of placement. Suitable working practices, curing and consolidation means would be required to use these mixes in similar situations.

Table 7: 28-day Ambient Cured Compressive Strengths Compared to Classification Strength Requirements

| Compressive strength requirement [standardised / generalised classifications]	Compressive strength classes	Mix ratio and replacement values that satisfy the strength requirements for classification at 91 days of water curing			
	[cylinder / cube strengths]	1:1:2	1:1:3	2:1(1:2)	2:1(2:1)
7.5 MPa [ST1/ GEN0]	-	10-90%	10-90%	20-80%	20-80%
10 MPa [ST2/ GEN1]	C8/10	20-90%	20-80%	20-80%	20-80%
15 MPa [ST3/ GEN2]	C12/15	40-70%	40-70%	40-80%	60%
20 MPa [ST4/ GEN3]	C16/20	-	60%	-	-
25 MPa [ST5/ GEN4]	C20/25	-	-	-	-
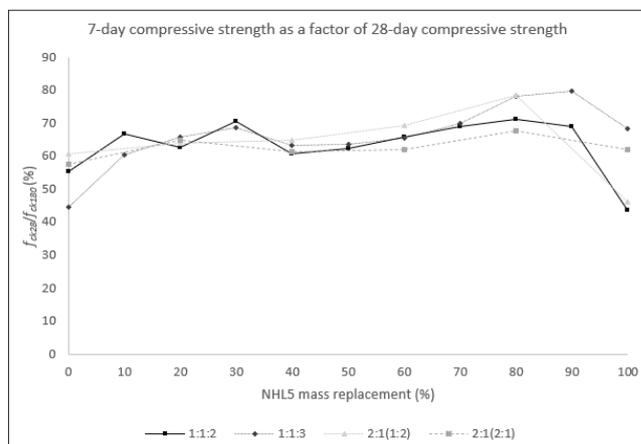
Figure 11: 7-day Compressive Strength as a Factor of 28-day Compressive Strength
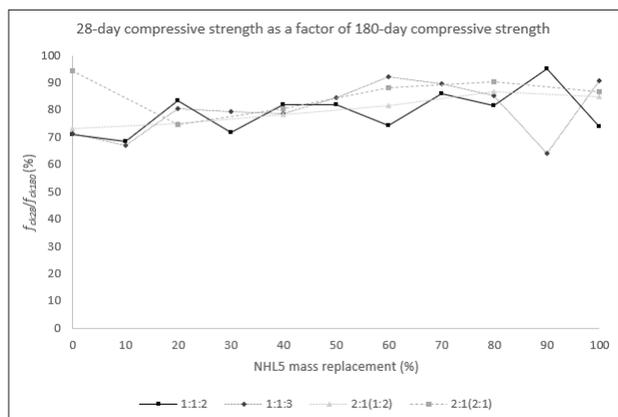
Figure 12: 28-day Compressive Strength as a Factor of 180-day Compressive Strength

When combining the data and evaluating by way of a quadratic best-fit curve an optimal point can be found, Figure 13.
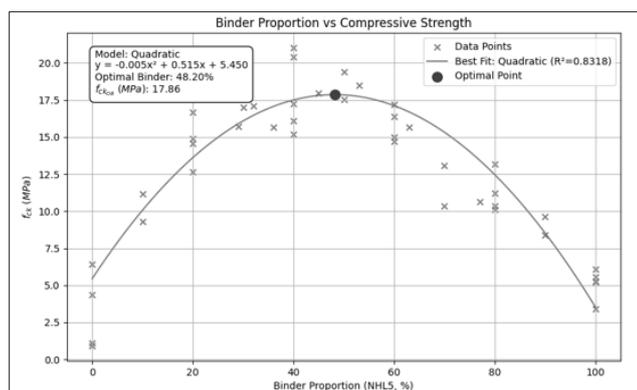
Figure 13: Optimal Compressive Strength Binder Proportion

The optimal replacement value considered for ambient compressive strength is at 48%, in line with the simulations of Wang et al, resulting in a compressive strength projected at 17.9MPa [80].
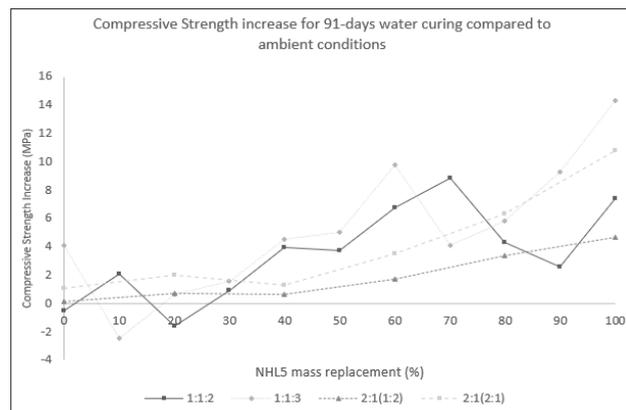
Water Curing Comparison
Studying the effect of water curing on concrete is crucial for understanding its impact on the material's strength, durability, and overall performance. Water curing ensures sufficient moisture for cement hydration, which is vital for achieving desired strength and durability. It enhances the strength development of concrete, making it more robust and resistant to stresses and loads while reducing the likelihood of surface cracks and improving longevity by minimising the ingress of harmful substances like chlorides and sulphates.

Water curing maintains moisture levels by keeping the concrete continuously wet, resulting in more uniform and complete hydration, higher compressive strength, and reduced permeability. In contrast, ambient curing relies on environmental conditions to provide moisture, which can be inconsistent, especially in dry or fluctuating climates, leading to incomplete hydration, lower strength, and higher susceptibility to cracking.

Water curing has significant influence on both blended slag-based cements and NHL5-based composite binders, thus it is reasonable to believe it will also be influential when combining NHL and GGBS[77,104].

Cubes from each mix were cured in water for 91 days and compared to those of 91-day ambient air-cure. A positive trend in increased strength from water curing can be observed with increasing NHL5 replacement by GGBS, Figure 14.

Figure 14: Compressive Strength Comparison of Water and Ambient Curing

The 100% GGBS binder concretes all exhibited the most extreme increase in compressive strengths exceeding a 100% increase in strength when cured in water compared to ambient curing, of 7.4, 14.3, 4.7, and 10.8 MPa for 1:1:2, 1:1:3, 2:1(1:2), and 2:1(2:1) respectively, considerably higher than the 15% difference experienced for PC-slag cements [104].

Generally, the water cured samples had increased compressive strength with the exceptions of 20% and below replacement values.

The findings suggest that when NHL5 and GGBS are combined, either curing immersed in water is beneficial with increasing emphasis applicable to larger replacement values above 30%, or carbonation in ambient conditions is significantly detrimental to the possible strength gain due to hydration. Therefore, these types of concrete may be useful to implement in environments and structures exposed to submerged or waterlogged conditions.

Many of the water cured mixes investigated could be utilised in industry for applications akin to the standardised and general-purpose concrete classifications, with the 91-day water cured specimens compared to the strength requirements, Table 8.

Table 8: 91-Day Water Cured Compressive Strengths Compared to Classification Strength Requirements

Compressive strength requirement [standardised / generalised classifications]	Compressive strength classes [cylinder / cube strengths]	Mix ratio and replacement values that satisfy the strength requirements for classification at 91 days of water curing			
		1:1:2	1:1:3	2:1(1:2)	2:1(2:1)
7.5 MPa [ST1/GEN0]	-	10-100%	0-100%	0-80%	20-100%
10 MPa [ST2/GEN1]	C8/10	10-100%	20-100%	20-80%	20-100%
15 MPa [ST3/GEN2]	C12/15	40-90%	20-100%	40-80%	40-100%
20 MPa [ST4/GEN3]	C16/20	40-70%	40-90%	60-80%	80%
25 MPa [ST5/GEN4]	C20/25	70%	50-60%	-	-

Nevertheless, these mixes may prove useful under certain conditions and constraints, where a longer time to maturity is less consequential with either form work in place for longer or a longer period before full static loading is applied. The mixtures are also well suited for unreinforced precast members and highway elements such as kerbing where this can be produced and cured in a timely manner offsite and for foundations or fill medium.

Mixes conforming to ST1/GEN0 are versatile mixes that could be used for general, non-structural applications such as kerb bedding, drainage works, haunches, blinding, mass and cavity fill. In addition, ST2/GEN1 could be utilised in un-reinforced domestic trench-fill foundation, patios, pathways and house floors. ST3/GEN2 is ideal for unreinforced domestic foundation slabs, floor finishes and general floor slabs. ST4/GEN3 are suitable for surfaces in domestic, agricultural and commercial settings such as house, garages and portal framed sheds. ST5/GEN4 of C20/25 can be used in domestic, commercial, agricultural and light industrial settings for reinforced ground slabs, slab foundations, heavy duty floor surfaces and for structural elements such as reinforced beams and columns.

Flexural Strength Testing
The flexural strength of a material is an element of the tensile strength of the material relating to the resistance in bending. This is a very important property of the material when used in a structural member, such as a beam or column.

The flexural strengths of the samples were determined following BS EN 12390-5:2019 at 28 days of ambient cure [84]. During this test, centre-point loading was applied as per BS EN 12390-5 (Annex A.3), with the force directed down from the centre of the top surface of the prism, whilst it was supported on the bottom in from each end with a free surface throughout the rest of the geometry to allow for bending to occur, Figure 15 [84]. The force and displacement data were taken via a load cell and high precision linear resistor (HPLR) throughout testing, with the load data at failure used to compute the ultimate flexural strength [84].

The loading rates were checked for conformity to the standard using Equation 7, with each samples loading profile checked for conformity to the standards definitions of undue shock, loading continuity, and ±10% rate range limitation.

The average flexural strengths were calculated following Equation 8.

Figure 15: Flexural Testing

Equation 7: Loading rate

$$R = \frac{2bd^2s}{3l} \text{ (N/s)}$$

Equation 8: Flexural strength

$$f_{cf} = \frac{3Fl}{2bd^2} \text{ (N/mm}^2\text{)}$$

The results from the 28-day flexural testing are presented in Figure 16. All combinations of NHL5 and GGBS showed significantly improved flexural strengths when compared to the controls, for all mixes. Two of the mixes failed to produce adequate data for that of the 100% slag samples at both a 1:1:3, and 2:1(1:2), aggregate mix ratio, with one specimen of the 1:1:3 mix breaking during demoulding.

Three of the four mixes investigated had the highest flexural strength achieved at a 40% replacement of NHL5 with GGBS, except for the 1:1:2 mix, occurring at a 60% replacement value, Figure 16. The highest flexural strength achieved was that of the 40% GGBS replacement sample for the 1:1:3 mix ratio at 2MPa.

In the mixes with 100% slag samples, Figure 16, a significant drop in strength is observed between 80% NHL5 replacement and 100%.
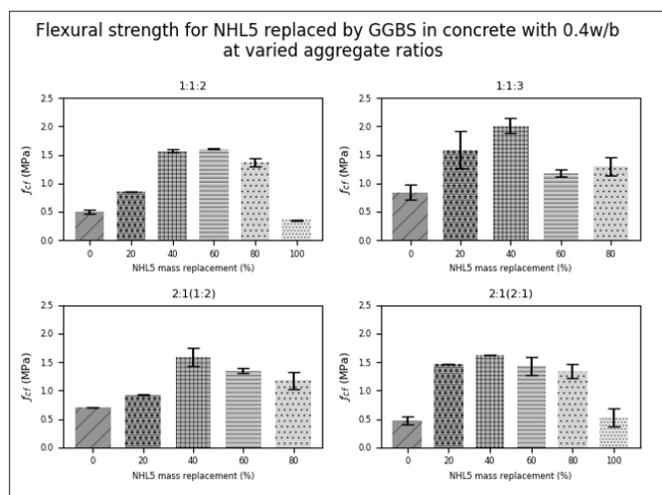

Figure 16: Flexural Strengths for NHL5 Replaced by GGBS

Three of the four mix ratios showed a downward trend in proportions of flexural to compressive strength with increasing NHL5 replacement by GGBS, Figure 17, with the fourth showing a minor increase. For all samples the flexural strength was between 6-16% of the compressive strength at 28 days maturity, with the higher strength samples being between 6-12%.

Comparing these flexural results to general moderate strength lime mortar, at 0.25-2MPa, and OPC concrete at 3.5-5MPa, shows the material to be of intermediary flexural strength, closer to lime mortar, although, to have a closer compressive to flexural strength relationship to OPC concrete (12-16%) rather than lime mortar (40-55%) [105,106].
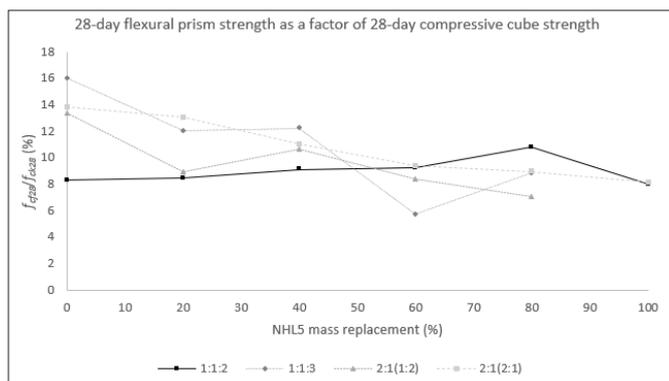
Figure 17: 28-day Flexural Prism Strength as a Factor of 28-day Compressive Cube Strength

Environmental Credentials

Conventional concrete utilises a binder material produced from an OPC clinker which is high in CO_{2e} and embodied energy (E_e). More modern blended binders have incorporated alternative materials to improve these characteristics. The calcination of limestone accounts for the majority of CO_2 emissions during cement production, roughly 60%, with fossil fuel combustion during the firing process accounting for up to 40% of carbon emissions from cement production [107,108].

The production process for cement requires three to six Giga Joules (3-6GJ) of energy per tonne of clinker produced [109]. The CO_2 output from the production of one tonne of cement is roughly one tonne, requiring 1.7 tonnes of raw material to produce a tonne of clinker [110].

Using the estimated figures of each material, Table 9, it is possible to compare the proportioned composites on the low carbon concrete group (LCCG) market benchmark (v3-1-2024) , Table 10 and Figure 18 [111].

Table 9: Assumptions for Benchmarking

Material	NHL5	GGBS	Gravel	Sand	Water
Embodied Carbon (kg/t)	635	67	2	2.6	2
Apparent Density (kg/m³)	975	1237	1472	1603	997

Carbon intensity (CI) can be compared to evaluate efficiency relative to compressive strength without fixed strength class boundaries. For instance, Table 10 shows that some low-strength, high-GGBS mixes with low CI do not classify in the LCCG system, like the 8th CI rank of 2:1(2:1) at 100% GGBS, due to not meeting the minimum C6/8 strength classification in the current LCCG benchmarking system.

The highest CI rank sample achieved a LCCG benchmark rating of 1.1 alongside the samples with the 3rd, 4th and 7th CI ranks, representing the top 0-5% nominal fractile, and 0% nominal embodied carbon fractile of the rating system. The 2nd placed by CI has a worse LCCG rating of 1.2 due to having higher embodied CO_{2e}, alongside the rigidity of the benchmarking system using arbitrary compressive strength boundaries in line with strength classes, when compared to the unconfined CI metric. Similarly, CI ranks 5 and 6, also show the nuance of the LCCG benchmark compared to the more direct CI metric for comparisons, with CI being preferable to define material efficiency but the LCCG framework boundaries being useful in comparing broader material categories.

Eleven of the 34 mixes achieved better embodied carbon scores, Figure 18, of which nine achieved better LCCG ratings, than the UK average, Table 10, with one C6/8, four C8/10, five C12/15, and one C16/20, all with between 90-50% replacement of NHL5 with GGBS.

Table 10: Carbon Intensity and LCCG Benchmarks

Mix Name	NHL5 Binder	Compressive Strength (f_{ck28})	Embodied Carbon (CO_{2e})	Carbon Intensity (CI)	CI Rank	LCCG Market Rating
	%	MPa	kg CO_{2eq} / m³	kg CO_{2eq} / m³ MPa⁻¹		
1:1:2	100	6.1	312	51.5	31	N/A
1:1:2	90	8.4	287	34.2	29	5.4
1:1:2	80	10.1	257	25.4	25	5.4
1:1:2	70	10.4	231	22.3	23	5.4
1:1:2	60	17.2	230	13.4	18	5.4
1:1:2	50	17.5	198	11.3	14	5.1
1:1:2	40	17.2	168	9.7	13	3.3
1:1:2	30	17.0	136	8.0	10	2.1
1:1:2	20	12.7	93	7.4	7	1.1
1:1:2	10	11.2	68	6.1	4	1.1

1:1:2	0	4.4	39	8.9	12	N/A
1:1:3	100	5.2	264	50.4	30	N/A
1:1:3	90	9.6	240	24.9	24	5.4
1:1:3	80	13.2	220	16.7	20	5.4
1:1:3	70	13.1	196	15.0	19	5.3
1:1:3	60	16.4	190	11.6	15	4.4
1:1:3	50	19.4	165	8.5	11	3.3
1:1:3	40	20.4	136	6.6	5	1.4
1:1:3	30	19.4	114	5.9	2	1.2
1:1:3	20	14.6	80	5.5	1	1.1
1:1:3	10	9.3	56	6.0	3	1.1
1:1:3	0	1.1	34	31.3	27	N/A
2:1(1:2)	100	5.3	411	77.6	32	N/A
2:1(1:2)	80	10.4	331	32.0	28	5.4
2:1(1:2)	60	15.0	263	17.6	21	5.4
2:1(1:2)	40	16.1	196	12.2	16	5.1
2:1(1:2)	20	16.7	122	7.3	6	1.3
2:1(1:2)	0	0.9	-	-	-	N/A
2:1(2:1)	100	3.4	391	115.1	33	N/A
2:1(2:1)	80	11.2	327	29.1	26	5.4
2:1(2:1)	60	14.7	259	17.6	22	5.4
2:1(2:1)	40	15.2	188	12.4	17	4.3
2:1(2:1)	20	14.9	119	8.0	9	1.4
2:1(2:1)	0	6.4	48	7.4	8	N/A
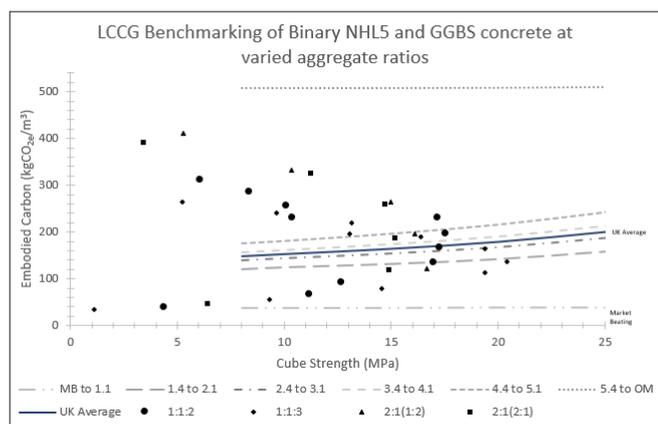
Figure 18: LCCG Benchmarking of Binary NHL5 and GGBS Concrete at Varied Aggregate Ratios

Sustainable concretes to strength class C8/10 are possible using 10-20% NHL5 and 80-90% GGBS in binary binder configurations and have the potential to be used for applications typically requiring concrete to ST1-ST3 or GEN0-GEN2, however plasticisers maybe required to meet recommended workability classifications.

When comparing the ambient CI data for all aggregate ratios, Figure 19, a quadratic model suggests the optimal replacement value considering carbon intensity would be lower than that expressed regarding only strength, in line with the predictions of Grist *et al*, at 32% resulting in a *CI* of 7 $kgCO_{2e}m^{-3}MPa^{-1}$, indicating a significant benefit when compared to PLCs, although at significantly lower compressive strength values [78].
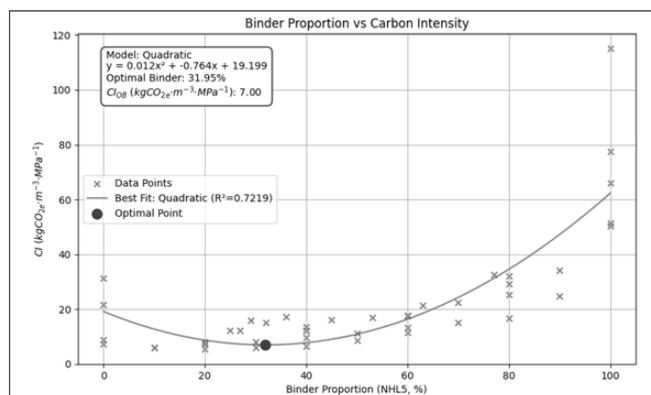
Figure 19: Optimal CI Binder Proportion

The optimal replacement within the scope of the strength class boundaries would be the lower of the two optimal points of 48% and 32% NHL5 for C12/15.

Use cases include: kerbing; haunches; blinding; unreinforced bases; pad foundations; unreinforced strip footings; and non-structural mass concrete.

Thus, there is potential for these materials to be used as an OPC free alternative to conventional concrete. However, the sustainability of these products and efficient use of their constituents would need assessment as the embodied carbon benchmark, material availability and cradle to grave life cycle need to be considered and compared for all: compositions of concrete materials; environments; and use cases [111].

Ongoing research with careful consideration of: material availability; carbonation; and reinforcement corrosion, can enhance both environmental impact and mechanical performance, achieving a product capable of C16/20 (ST4/GEN3) for use in unreinforced floor slabs. Research suggests this could be achieved with the inclusion of plasticisers, the reduction of w/b, in binary binder configurations and/or with the incorporation of other SCMs, and an increase in aggregate proportion. Further substitution using silica fume or organic waste ashes, and the incorporation of superplasticisers has been proposed [2,78].

This research shows binary combinations of NHL5 and GGBS as a binder in mortar and concrete can reduce carbon emissions whilst maintaining performance and thus contribute to improving the quality of infrastructure in the built environment.

Conclusions
Preliminary investigations into limecrete have shown that combining NHL5 with GGBS as a binder can result in a fourfold increase in the characteristic compressive cube strength of associated concretes when compared to individual binder controls.

- The additions of GGBS to NHL5, or vice versa, in concrete can improve the mechanical properties, with the highest 28-day compressive cube and flexural strengths achieved at 21MPa and 2MPa by ambient cured 1:1:3, 60% and 40% GGBS replaced samples respectively. Combining NHL5 and GGBS in a cement-free limecrete can satisfy the characteristic cube strength requirements of structural concretes in the grades of C8-C20. The highest compressive cube strength achieved was that from 60% GGBS replacement at a mix ratio of 1:1:3, cured in water for 91 days. The flexural strength was highest at 40% NHL replacement for the aggregate ratios of 1:1:3 and the two 2:1 mixes with the highest for the 1:1:2 mix being at 60% replacement. The optimum replacement value of NHL5 with GGBS is between 40-60% by mass for all mix ratios and both curing conditions when considering strength.
- The water cured samples show an increase in strength when compared to ambient cured samples with GGBS binder contents above 20%. Given the difference between water and ambient cured results it is suspected that carbonation has a considerable role in the development to strength and maturity requiring further investigation.
- The strength requirements of Standardised prescribed concrete to ST1-4 were satisfied by 26 of the 34 cases. Sustainable concretes to strength class C8/10 are possible using 10-20% NHL5 and 80-90% GGBS in binary binder configurations and have the potential to be used for application typically requiring concrete to ST1-ST3 or GEN0-GEN2, however plasticisers maybe required to meet recommended workability classifications.
- The workability of NHL5 concrete is different to GGBS concrete, and when combined in an aggregate mix ratio of 1:1:2 at 0.4 w/b has the minimal amount of water to achieve an S1 slump class without the use of additives such as plasticisers. Whereas a mix ratio of 1:1:3 requires either additional water or additives at this w/b ratio to achieve an adequate slump measurement >5mm. However, concrete mixes of NHL and GGBS binders with slump measurements inadequate for classification may still consolidate well.
- Thermodynamic modelling simulations based on Gibbs free energy and the law of mass action can be used to evaluate complex chemical systems and accurately predict optimal equilibrium phase assemblages in NHL systems.
- Embodied carbon benchmarking using either CI or the LCCG

system shows that eleven of the suggested mixes can be considered suitable when considering compressive strength and embodied carbon alone.

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