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Compressive Strength and Environmental Impact of Sustainable Blended Cement with High-Dosage Limestone and Calcined Clay (LC2)

Jing Yu[1], Hao-Liang Wu[1,*], Dhanada K Mishra[1,2,*], Gengying Li[1,3], Christopher KY Leung[1]

Highlights:

- Blended cements with 0-80% Limestone and Calcined Clay (LC2) were explored.
- Blended cements with 50%, 60% and 70% LC2 can be classified as 52.5N, 42.5N and 32.5N.
- Blended cements with 50-60% LC2 showed sufficient strength and environmental benefits.
- LC2 had higher cementing efficiency than siliceous fly ash in all replacement levels.
- Blended cement with LC2 had a low heat release rate and total hydration heat.

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ABSTRACT

Replacing half of the clinker by a blend of limestone powder and low-grade calcined clay (in 1:2 wt ratios) has been recently proposed to produce a new version of economical and green cement named Limestone-Calcined Clay (LC2) cement, also known as LC3. The LC2 blend emits much lower carbon dioxide than traditional Portland clinker and has sufficiently high cementing efficiency factor for maintaining compressive strength under high replacement levels. In this study, the feasibility of blending more than 50% of LC2 in cement was explored in order to achieve a greener blended cement. Five LC2 replacement levels were studied, including 25%, 50%, 60%, 70% and 80% by weight of cement. Compressive strength of BS EN 196-1 standard mortar mixes at 3–360 days was evaluated, and the fresh property, hydration heat as well as environmental impact of the blended cement were investigated. The results showed that the blended cements with 50%, 60% and 70% LC2 achieved the compressive strength of 53.6 MPa, 43.9 MPa and 33.4 MPa at 28 days, respectively; thus they fulfill the 28-day strength requirements for 52.5N, 42.5N and 32.5N cements, respectively. The blended cements with 50–60% LC2 had lower material cost index than Portland cement and blended cement with fly ash. In addition, they showed lower embodied energy/carbon emission indices at the early age but higher at the later age as compared to blended cements with fly ash. In the context of the substantial contribution of Portland cement manufacture towards the climate crisis, these findings would help the efforts to reduce the carbon footprint in the construction industry.

Nomenclature

A	Aluminate phase in the binder
C	Cement dosage
C-(A)-S-H	Calcium-(Aluminate)-Silicate-Hydrate
C_3A	Tricalcium Aluminate
Cc	Limestone
CH	Calcium Hydroxide
CI_i	Carbon emission per unit compressive strength at i day (g CO_2/kg/MPa)
$COST_i$	Material cost per unit compressive strength at i day (HKD/kg/MPa)
D_0	Diameter of the cone bottom
D_1	Average diameters of two orthogonal after 25-time drops
EI_i	Embodied energy per unit compressive strength at i day (KJ/kg/MPa)
FA	Fly ash
f_c	Cementing Efficiency Factor
LC2	Limestone-Calcined Clay
LC3	Limestone-Calcined Clay Cement
S	Silica phase in the binder
SCM	Supplementary Cementitious Material (dosage)
Γ	Deformability factor
W	Water dosage

1. Introduction

It is well known that sustainable development by definition requires the world to consume natural resources responsibly so that the ability of future generations to live sustainably is not diminished (Borowy, 2015). As the atmospheric carbon dioxide (CO_2) levels rise exponentially leading to the greenhouse effect that results in global warming and climate change (Boden et al., 2017), the demand for greener construction materials will be increasingly important going forward. The cement industry contributes to 5–8% of the global emissions of greenhouse gas (Boden et al., 2017). Specifically, Portland cement contributes almost one kg of CO_2 per kg of cement produced (Monteiro et al., 2017), which is the primary component of concrete in the predominant construction material today by a long margin.

Production of greener cement and concrete by the use of Supplementary Cementitious Materials (SCMs) (Juenger et al., 2019) and alternative binders (Shi et al., 2019) has been a global effort for several decades. A number of SCMs such as fly ash (Hemalatha and Ramaswamy, 2017), granulated blast furnace slag (Giergiczny, 2019), silica fume (Kadri et al., 2009), cal-

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cined clay (Sabir et al., 2001), rice husk ash (Mosaberpanah and Umar, 2020) and vegetable ash (Martirena and Monzó, 2018) in different combinations, as well as several alternative binders such as magnesia (Wu et al., 2019), have been used for this purpose evidenced by an increasing number of recent publications. However, the amount of common SCMs such as fly ash and slag available globally (being only about 10% of all cements manufactured) is not enough to fulfil the future demand (Scrivener, 2014). The variability in the characteristics of fly ash has also been a particular concern to ensure the quality of concrete (Thomas, 2007).

In this context, calcined clay is the only known SCM material that is adequately available (Ruben, 2016). Ordinary clay with as little as 40% kaolinite which is abundant in earth's crust can be converted into a pozzolanic material known as calcined clay by relatively moderate thermal treatment (Fernandez et al., 2011). The use of calcined clay as a substitution of cement has a long history. In the USA, it was used in making blended cement as early as 80 years ago (Riding and Zayed, 2020). Such application was commonplace in large scale projects involving mass concrete such as the famous Golden Gate Bridge in California which was reported to be in excellent condition proving the benefits of this approach. Use of calcined clay declined around the middle of last century with the gradual rise of coal-fired power plants making low-cost fly ash widely available (Sabir et al., 2001). Calcined clay, on the other hand, transformed into high-reactivity metakaolin, a more value-added product used in high-performance concrete. Metakaolin is characterized by clay sourced with very high fractions of kaolin and purified carefully to get excellent whiteness, calcined at a higher temperature, and ground to a finer particle size (Alice and Mark, 2019). While the beneficial effect of metakaolin on concrete durability was significant, only a small quantity was required to be used for the high cost and performance requirement in niche applications (Siddique and Klaus, 2009).

Recently, calcined clay has re-emerged as a new source of SCM (Tironi et al., 2012). Additionally, calcined clay being a manufactured product provides a much better opportunity for quality control (Cancio Díaz et al., 2017). The research into the use of calcined clay in conjunction with limestone powder has intensified in recent years to develop a new green blended cement known as Limestone-Calcined Clay (LC2) cement, which consists of 30% calcined clay, 15% limestone, 50% clinker and 5% gypsum (Scrivener et al., 2018). Such a cement can reduce the carbon emission of cement production by up to 30%, based on the estimation in Cuba (Sánchez Berriel et al., 2016) and India (Gettu et al., 2019). Normal concrete made with the aforementioned LC2 cement was found to exhibit very similar mechanical properties to Portland cement (Dhandapani et al., 2018). Additionally, LC2 cement has also been used in polyvinyl alcohol fiber (Yu et al., 2020), polyethylene fiber (Yu and Leung, 2020) and polypropylene fiber (Zhu et al., 2020) reinforced strain-hardening cementitious materials, and satisfactory mechanical performance was achieved.

In the LC2 cement system, besides the pozzolanic reaction of Silica phase (S) in Eq. (1) forming additional Calcium-(Aluminate)-Silicate-Hydrate (C-(A)-S-H) phases (Lothenbach et al., 2011), the Aluminate phase (A) from calcined clay can react with Calcium Hydroxide (CH) and limestone ($CaCO_3$, Cc) to produce carbo aluminate (Scrivener, 2014), which also contributes to the strength (Eq. (1)). Thus, it was postulated that the use of LC2 blend would result in enhanced strength at even a high cement replacement level.

$$\text{S} + \text{(A)} + \text{CH} \longrightarrow \text{C-(A)-S-H} \tag{1}$$

$$\text{A}_{(\text{in calcined clay})} + \text{Cc} + \text{CH} \longrightarrow \text{C}_3\text{A}{\cdot}\text{Cc}{\cdot}\text{H}_{11} \tag{2}$$

The development of LC2 cement has involved examining different dosages of LC2 blend and different ratios between the limestone and calcined clay (Antoni et al., 2012). However, there is a lack of experimental research exploring the use of more than 50% of LC2 blend in the binder due to the high water demand as well as possible lower strength gain due to dilution effect. Specifically, in many developing countries like India, the most widely-used cement is low-grade 32.5N cement; therefore, blending higher dosage of LC2 (over 50%) in cement can produce more economical and greener cement while fulfilling the strength requirement.

To fill the aforementioned knowledge gap, this study aims to explore the feasibility of blending a high dosage of LC2 in cement. Blended cements with different LC2 replacement levels (0%, 25%, 50%, 60%, 70% and 80% by weight) were investigated, and the results were compared to plain Portland cement and blended cement with siliceous fly ash. Compressive strength of BS EN 196-1 (BSI, 2016) standard mortar mixes at 3, 7, 14, 28, 90 and 360 days were recorded, and the hydration heat of the mortar was evaluated. Additionally, the environmental impact and cost implication of the resulting cement were discussed; the carbon emission, embodied energy and material cost per unit compressive strength of the blended cement were also quantified.

2. Materials and method

2.1. Ingredients and cement design

CEM I 52.5N Portland cement (BSI, 2011) was used in this study. Siliceous fly ash as per BS EN 450-1 (BSI, 2012) obtained from the National Thermal Power Corporation Limited in Odisha in India (Yu et al., 2018b), and limestone-calcined clay (LC2) blend sourced from India (Bishnoi et al., 2014) were used as the SCM. Specifically, the weight ratio between limestone to calcined clay was fixed at 1:2 in the LC2 blend. The chemical compositions (by X-ray fluorescence spectrometer, JEOL JSX-3201Z), XRD patterns (by PANalytical X'pert) and particle size distributions (by TRI-LASER Particle Size Analyzer) of the Portland cement, LC2 blend and fly ash are shown in Table 1, Fig. 1 and Fig. 2, respectively. SEM images (by JEOL JSM-6390) of the LC2 blend and siliceous fly ash are shown in Fig. 3. Standard sand (BSI, 2016) was used as the fine aggregate.

The BS EN 196-1 (BSI, 2016) standard mortar mix with a water/binder ratio of 0.5 and a sand/binder ratio of 3 was used in this study, to evaluate the strength of the blended cement. Three series of mortar were prepared (Table 2). C100 with plain Portland cement was the control. LC2-Series have five different LC2 replacement levels (25%, 50%, 60%, 70% and 80% by weight of binder), and FA-Series was designed for comparison.

Table 1
Major compositions of Portland cement, siliceous fly ash and LC2 blend.

Oxide percentage	Al_2O_3 (%)	SiO_2 (%)	Fe_2O_3 (%)	CaO (%)	SO_3 (%)	Na_2O (%)	K_2O (%)
Portland cement	4.4	20.2	3.4	63.9	4.7	0.1	0.4
LC2 blend	31.3	45.8	3.4	14.5	1.7	–	0.7
Fly ash	27.5	64.6	3.5	1.1	–	–	1.3

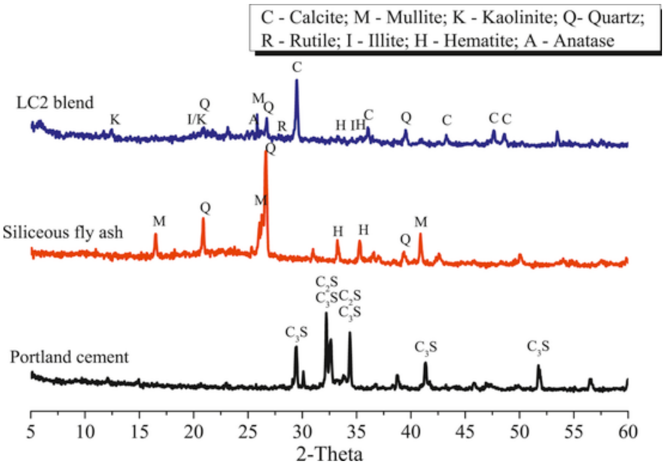

Fig. 1. XRD analyses of Portland cement, siliceous fly ash and LC2 blend.

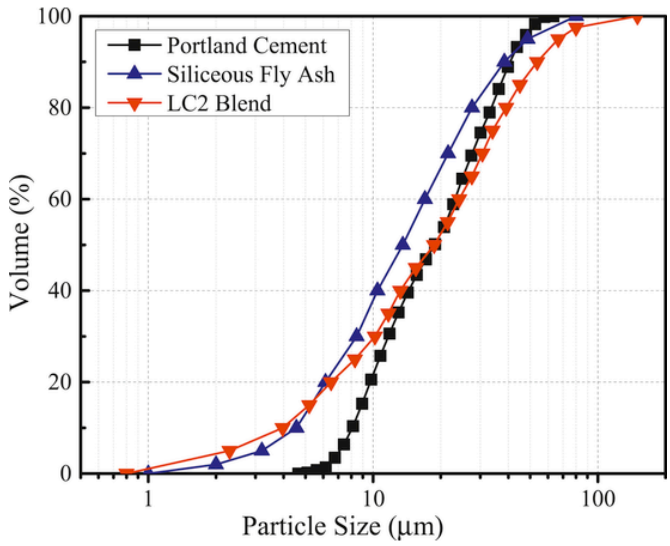

Fig. 2. Particle size distributions of Portland cement, LC2 blend and fly ash. More than 50% of the particles in cement are larger than those in LC2 blend and fly ash.

2.2. Sampling and testing procedure

Mixing. Solid ingredients, including silica sand, Portland cement, siliceous fly ash and LC2 blend were dry mixed in a 4-L Hobart mixer for 1 min at the low speed. Fresh water was then added into the dry mixtures for mixing for 2 min at the high speed.

Fresh property. To evaluate the effect of SCM replacement level (25%, 50% and 80%) on the fresh properties, a flow table test was conducted on the standard mortar as per ASTM C1437 (ASTM, 2007). A 60-mm high truncated cone with a 70-mm top diameter and a 100-mm bottom diameter was put on a flat and smooth table, filled with fresh mortar mixture, and raised upward. Then the table was dropped 25 times in 15 s (ASTM, 2007). The characteristic deformability factor (Γ) was obtained as (Lepech and Li, 2008):

$$\Gamma = (D_1 - D_0)/D_0 \tag{3}$$

where D_1 is the average of the two readings of diameters after dropping, and D_0 is the diameter of the cone bottom (100 mm).

Compressive Strength. Cubic specimens of 40 mm × 40 mm × 40 mm were cast in stainless steel molds on a vibration table (BSI, 2016). The specimens were demolded after 24 h from casting, and then were stored in a curing room (relative humidity of 95 ± 5% and temperature of 23 ± 2 °C) until testing (BSI, 2019b). The test was perform in an automatic compression testing machine with a loading rate of 0.625 MPa/s (BSI, 2019a). Compressive tests were performed in duplicate, and the mean value as well as the standard deviation were reported. The significance of differences between each group was evaluated by the one-way analysis of variance method at the significant level of 0.05 using SPSS.

Hydration Heat. The hydration heat of mortar was measured for 3 days (72 h) at a lab temperature as per BS EN 196-11 (BSI, 2018), with a Calmetrix I-Cal 8000 HPC Isothermal Calorimeter. After mixing, around 200 g of mixture was immediately loaded into an airtight container and transfer to the calorimeter. The heat release data was automatically recorded every 60 s by the system.

3. Results and discussion

3.1. Compressive strength

Fig. 4 summarizes the compressive strength for all the mortar in Table 2 at the ages of 3/7/14/28/90/360 days under standard curing. The overall trend of strength improvement with increasing curing time is expected, which is mainly attributed to the continuous cement hydration and the chemical reactions between the SCM and calcium hydroxide (Eq. (1) and Eq. (2)).

3.1.1. Effect of SCM replacement level

A gradually decreasing trend in compressive strength was recorded for the blended cement with increasing contents of LC2 or fly ash (Fig. 4), which is due to the low clinker content as well as low calcium hydroxide content for SCM reactions. For the LC2-Series, a replacement level of no more than 50% had no significant effect on the compressive strength, which is consistent with the results on compressive strength reported by other researchers (Dhandapani et al., 2018). Specifically, the blended cements with 50%, 60% and 70% LC2 achieved a compressive strength of 53.6 MPa, 43.9 MPa and 33.4 MPa at 28 days, respectively; and they fulfilled the 28-day strength requirements for 52.5N, 42.5N and 32.5N cements as per BS EN 197-1 (BSI, 2011), respectively. Blended cement with LC2 developed significantly higher strength than that with siliceous fly ash under the same SCM replacement level, especially at the early age. In other words, LC2 has higher cementing efficiency than siliceous fly ash, and the cementing efficiency factor at 28 days is further quantified in Section 3.1.3. Specifically, the blended cements with 50%, 60%, 70% and 80% LC2 achieved the compressive strength of 34.2 MPa, 28.2 MPa, 21.7 MPa and 16.3 MPa at 3 days, respectively; these strength values are adequate for many practical applications.

3.1.2. Different strength development rates of LC2 blend and fly ash

Fig. 5 shows the ratio between the 3-day/7-day compressive strength to the 28-day compressive strength for standard mortar with LC2 or siliceous fly ash at different replacement levels. A higher ratio in the figure indicates less strength increase from the early age to 28 days.

The LC2-Series had higher 3 d/28 d and 7 d/28 d strength ratios than Portland cement, while the FA-Series had lower ratios than Portland cement. Specifically, the 3 d/28 d and 7 d/28 d strength ratios are 61.8% and 79.1% in the plain Portland cement, respectively. The average 3 d/28 d strength ratio

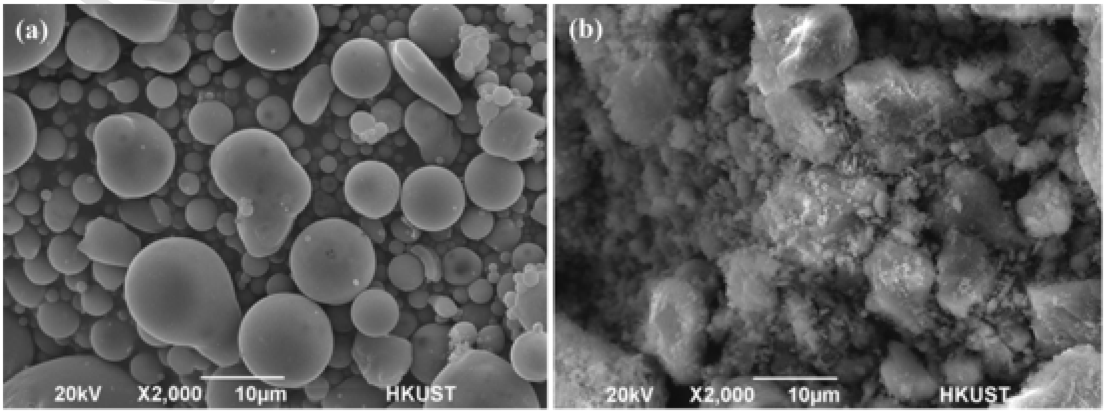

Fig. 3. SEM images at an accelerating voltage of 20 kV and a magnification of 2000 times for (a) fly ash and (b) LC2 blend.

Table 2
Mix design (by weight ratio), where the sand and water ratios follow the standard mortar mix in BS EN 196-1 (BSI, 2016).

Series	Mix	Cement/ Binder	SCM/ Binder	Water/ Binder	Sand/ Binder
Control	C-100	1.0	0	0.5	3
LC2-Series	LC2-25	0.75	0.25	0.5	3
	LC2-50	0.5	0.5	0.5	3
	LC2-60	0.4	0.6	0.5	3
	LC2-70	0.3	0.7	0.5	3
	LC2-80	0.2	0.8	0.5	3
FA-Series	FA-25	0.75	0.25	0.5	3
	FA-50	0.5	0.5	0.5	3
	FA-60	0.4	0.6	0.5	3
	FA-70	0.3	0.7	0.5	3
	FA-80	0.2	0.8	0.5	3

is 64.9% and the average 7 d/28 d strength ratio is 82.3% for LC2-Series (Fig. 5a); the average 3 d/28 d strength ratio is 56.8% and the average 7 d/28 d strength ratio is 72.0% for FA-Series (Fig. 5b). The chemical reaction of the aluminate phase (from calcined clay) in the system (Eq. (2)) is a relatively fast process (Scrivener et al., 2018), and thus it can contribute to the early strength in the system. On the other hand, the pozzolanic reaction (Eq. (1)) of siliceous in fly ash is a slow process that contributes to strength mainly at later stage for the system without chemical activation and under normal curing (Yu et al., 2017). As a result, blending high dosage of siliceous fly ash in cement generally leads to low early strength, which is consistent with the result in the literature (Parashar et al., 2020). Nevertheless, blended cement with LC2 only had less than 10% increment in compressive strength from 28 days to 360 days (Fig. 6). Blended cement with siliceous fly ash had significant strength improvement at the later ages, and the 360-day strength was double in mortar with 60% fly ash and even close to triple in mortar with 80% fly ash as compared to the strengths at 28 days (Fig. 6).

3.1.3. Cementing efficiency factor of LC2 blend

The following equation has been proposed by Bolomey (1936) to estimate the compressive strength (f_c) of Portland cement-based concrete:

$$f_c = a \times (W/C)^{-1} - b \tag{4}$$

where the two constants a and b mainly depend on curing time and cement type. For concrete with SCM in the binding system, the cementing efficiency factor (k) reflects the strength contribution of SCM to an equivalent weight of Portland cement (Smith, 1967). The effective water/cement ratio can be determined as:

$$(W/C)_e = W \,/\, (C + k \times SCM) \tag{5}$$

where W is the water dosage, SCM is the SCM dosage and C is the cement dosage.

With a known compressive strength values (Fig. 4), the cementing efficiency factor of SCM at different replacement levels can be determined by substituting (5) into (4). The regression curve of (4) were determined with the method of least squares (Fig. 7), with a series of compressive strength results of mortar with 52.5N cement (identical to the cement in this study) and different water/cement ratios in (Yu et al., 2017).

This study only evaluated the 28-day cementing efficiency factor of LC2 blend and fly ash (Fig. 8). This factor of LC2 blend is significantly higher than that of fly ash under the same replacement level. The cementing efficiency factor of LC2 blend was found to be about 0.9 for the replacement level of no more than 50%, and it gradually reduced with an increasing replacement level. Specifically, the cementing efficiency factor of LC2 blend could still reach about 0.35 at the 80% replacement level. On the other hand, the cementing efficiency factor of siliceous fly ash was 0.22 at the 25% replacement level, and reduced to about 0.09 when the replacement level was no less than 50%. The change of cementing efficient factor of SCM under different replacement levels should be due to the change of calcium hydroxide concentration in the system, which is controlled by the generation from cement hydration and the consumption from the SCM reactions as discussed by (Yu et al., 2018a).

3.2. Fresh property

Fig. 9 summarizes the characteristic deformability factors (Γ) for some representative mortars in Table 2. The mortars with fly ash had better workability compared to the control, from $\Gamma = 1.10$ for FA-25 to $\Gamma = 1.31$ for FA-80. The deformability factors of LC2-Series decreased with increasing LC2 dosage, which is similar to the trend observed in (Shah et al., 2020). This is because the calcined clay can adsorb a higher amount of water compared with that of cement and fly ash given the much higher specific surface area (Ferreiro et al., 2017). As for mixes with SCM of 25%, 50% and 80% in the binder, the Γ for LC2-Series is 4.5%, 13.4% and 23.7% lower than that for FA-Series, respectively. Regarding the effect of particle shape, the friction reduction of surface particles might be considered (Senff et al., 2009). Calcined clay has high fineness (as indicated in Figs. 2 and 3), which require higher contents of superplasticizers or water to achieve practical workability for real-life applications.

3.3. Hydration heat

Fig. 10 present the heat release rate and accumulative heat of hydration (up to 72 h) normalized per gram of blended cement for some representative mixes listed in Table 2. Three major peaks of isothermal calorimeter curve are clearly shown in Fig. 10a. Compared to the control group with plain Portland cement, the mortar mixes in both LC2-Series and FA-Series showed lower heat release rate and lower accumulative heat of hydration (Fig. 10b) due to less cement content in the system. The increased replacement dosage of LC2 and fly ash could reduce the accumulative heat of hydration, which should be attributed to the dilution effect by more of the available water for the cementitious hydration (Irbe et al., 2018). Generally, the mixes with LC2 had a greater early age pozzolanic strength activity than those with fly ash under the same SCM replacement level, and therefore can lead to higher early compressive strength and total heat of hydration. This result supports the development of compressive strength (Fig. 4).

3.4. Assessment of environmental impact and cost implication

Generally, blending a higher dosage of SCM in cement lowers the environmental impact and the compressive strength simultaneously. To quantify these changes, the cradle-to-gate embodied energy and carbon emission, as well as the material cost for different ingredients are shown in Table 3. Specifically, the energy consumption and carbon emission can vary considerably from plant to plant and according to cement plant processes. Therefore, some representative data for the ingredients from the literature was used in this study. Additionally, the embodied energy and carbon emission for fly ash are almost zero since it is considered as a waste, indicating that the fly ash itself would be more sustainable than LC2 blend unless it is not available or a high replacement levels is required.

3.4.1. Total environmental impact and material cost of blended cement

Fig. 11 shows the embodied energy and carbon emission for LC2-Series and FA-Series. It can be seen that an increased LC2 or fly ash dosage can significantly reduce the embodied energy (Fig. 11a) and carbon emission (Fig. 11b). The LC2 and fly ash contribute 15.6–68.9% and 0.6–6.8% for embodied energy, and 7.1–47.9% and 0.3–3.4% for

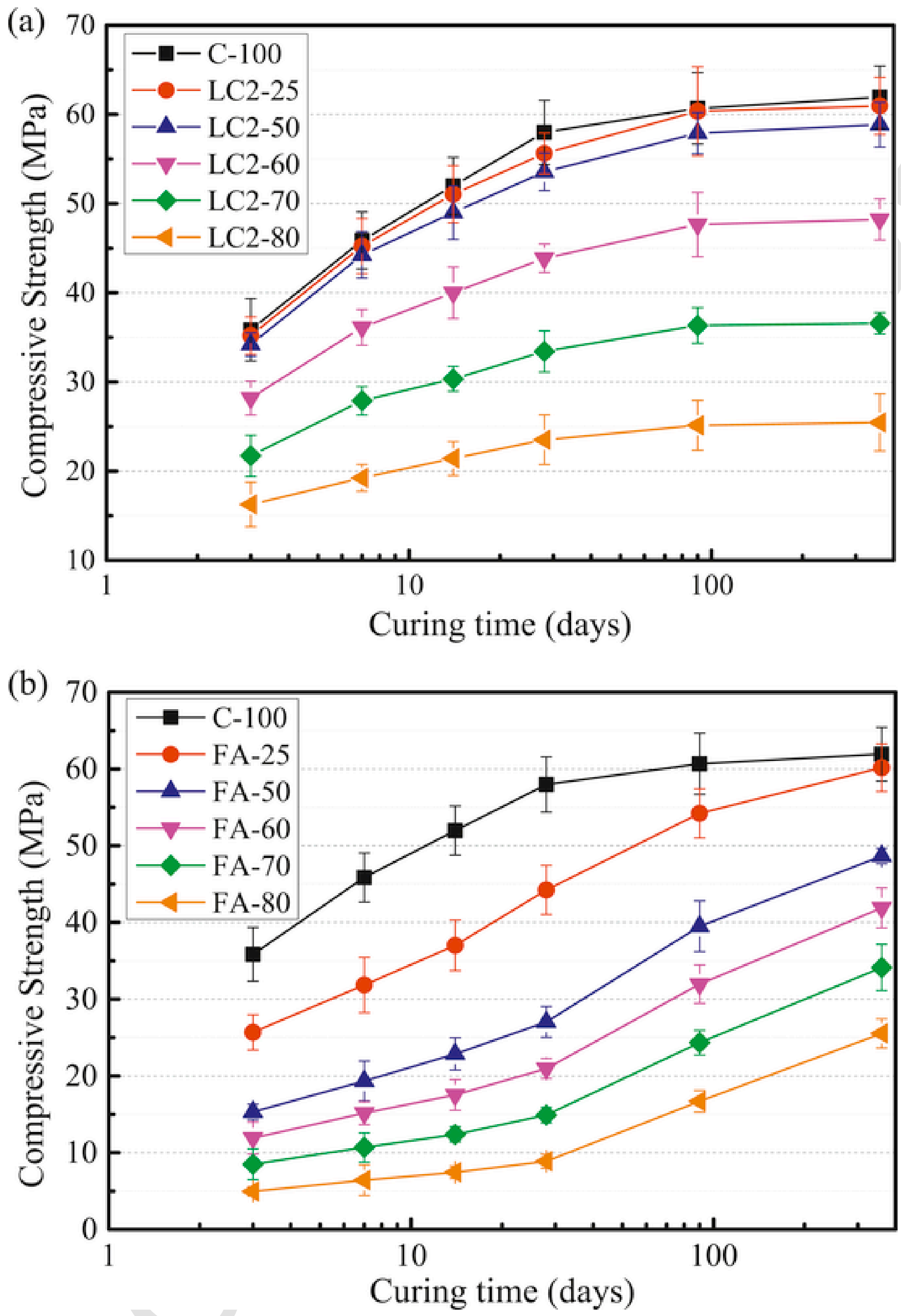

Fig. 4. Compressive strength of standard mortar with SCM at different replacement levels at 3–360 days: (a) LC2-Series; (b) FA-Series. The LC2 blend significantly contributes to the early strength development in the system. Blended cements with 50%, 60% and 70% LC2 achieved compressive strength of 53.6 MPa, 43.9 MPa and 33.4 MPa at 28 days, respectively.

ments, respectively. As compared to the LC2-Series, the FA-Series has 15.1–66.6% less embodied energy and 6.8–45.7% less carbon emission with increasing SCM replacement level from 25% to 80%.

Additionally, compared to commercial cements with identical strength class as per BS EN 197-1 (Table 4), the material cost of LC2-50, LC2-60 and LC2-70 is 25.0%, 21.1% and 11.9% lower, respectively. Since the cost of fly ash and LC2 are identical, thus there is no need to compare the unit-cost per mass of blended cement with SCM at different replacement levels.

3.4.2. Environmental impact and material cost per unit compressive strength

To quantify the environmental impact and the material cost per unit compressive strength for different bended cements, Eq. (6), Eq. (7) and Eq. (8) were used to calculate the embodied energy index (*EI*), carbon emission index (*CI*) and material cost index (*COST*):

$$EI_i\left(\frac{kJ}{kg}/MPa\right) = \frac{Embodied\ Energy\ of\ 1\ kg\ Cement}{i-day\ Compressive\ Strength\ of\ Standard\ Mortar} \tag{6}$$

$$CI_i\left(\frac{g\ CO_2}{kg}/MPa\right) = \frac{Carbon\ Emmision\ of\ 1\ kg\ Cement}{i-day\ Compressive\ Strength\ of\ Standard\ Mortar} \tag{7}$$

$$COST_i\left(\frac{HKD}{kg}/MPa\right) = \frac{Material\ Cost\ of\ 1\ kg\ Cement}{i-day\ Compressive\ Strength\ of\ Standard\ Mortar} \tag{8}$$

where *i* refers to the curing time in days.

The calculation results on EI_i, CI_i, and $COST_i$ for a unit weight of blended cement are shown in Fig. 12. At 28-day age (Fig. 12a), the whole LC2-Se-

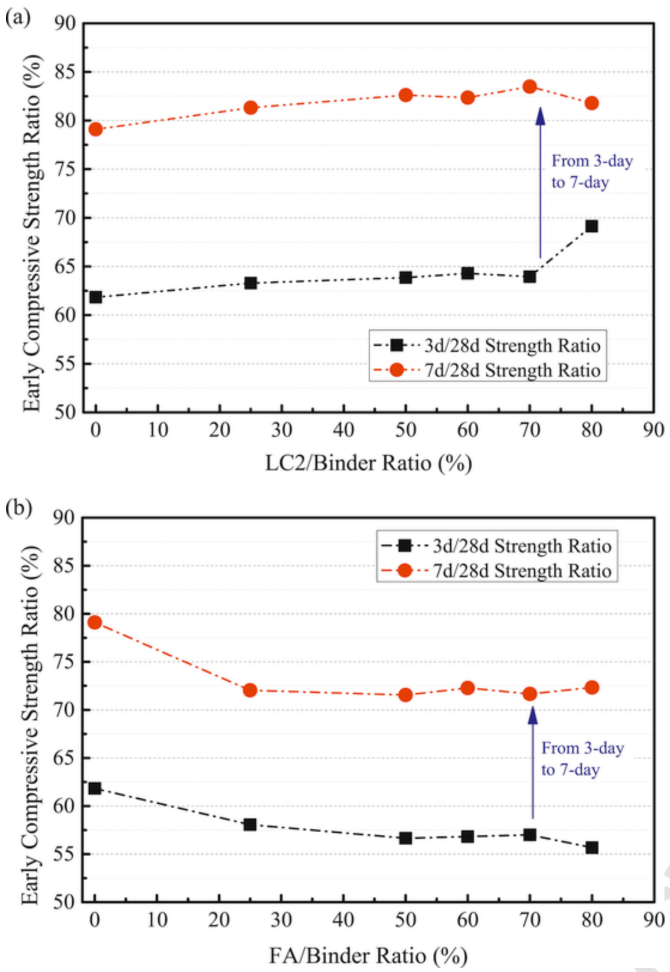

Fig. 5. Early strength to 28-day strength ratios of standard mortar with SCM at different replacement levels: (a) LC2-Series; (b) FA-Series. The LC2-Series has higher 3 d/28 d and 7 d/28 d strength ratios than Portland cement, while the FA-Series has lower ratios than Portland cement.

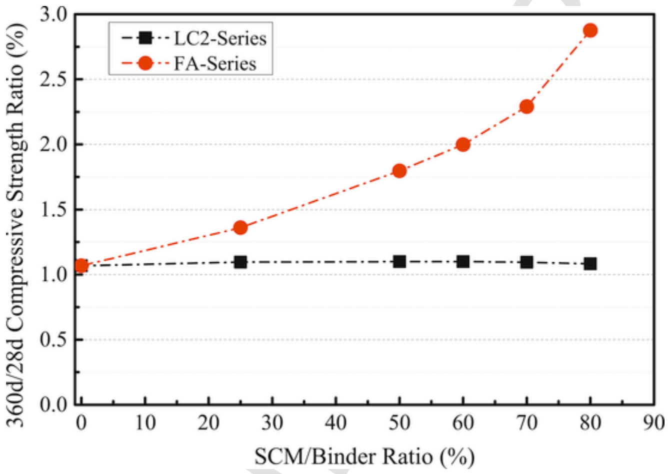

Fig. 6. 360-day strength to 28-day strength ratio of standard mortar with SCM at different replacement levels. Blended cement with LC2 only has less than 10% strength increment and that with fly ash has significant strength improvement at the later age.

ries show significantly lower CI_{28} than the FA-Series and plain Portland cement. Additionally, the LC2-Series have lower EI_{28} and $COST_{28}$ than the FA-Series and plain Portland cement when the LC2 replacement level is no more than 60%, and the lowest value appears for LC2-50. In contrast, the FA-Series have higher EI_{28} and CI_{28} than the plain Portland cement, expect for FA-25. The $COST_{28}$ of FA-Series increases with the increasing fly ash dosage, which is attributed to the obvious reduction of compressive strength in the blend cement (Fig. 4).

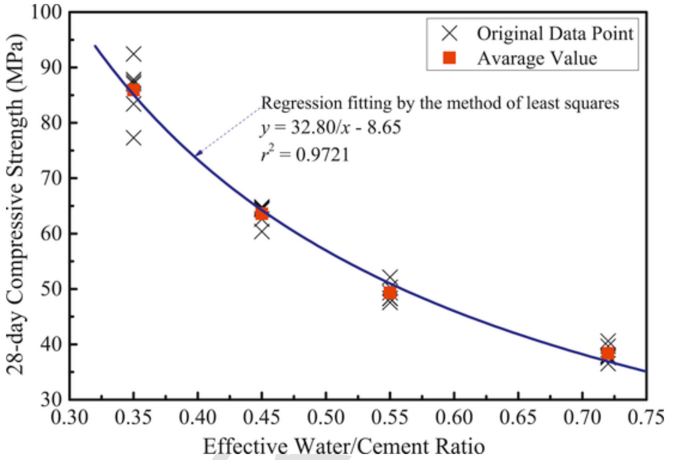

Fig. 7. Regression curve for 28-day compressive strength vs. effective water/cement ratio of mortar with 52.5N cement.

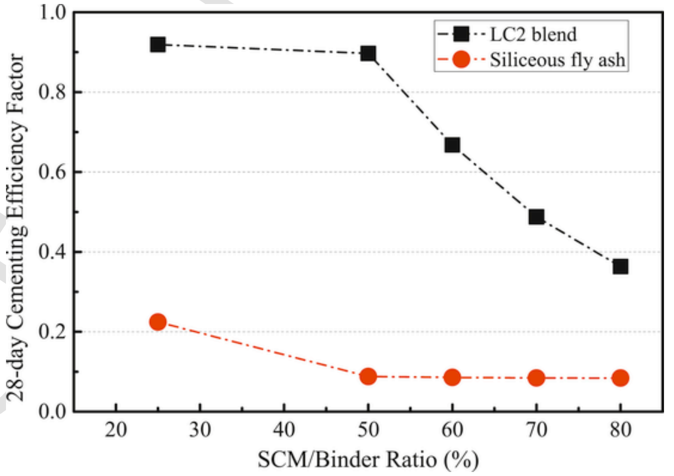

Fig. 8. Calculated 28-day Cementing Efficiency Factor of LC2 blend and siliceous fly ash at different replacement levels. This factor of LC2 blend is significantly higher than that of fly ash under the same replacement level.

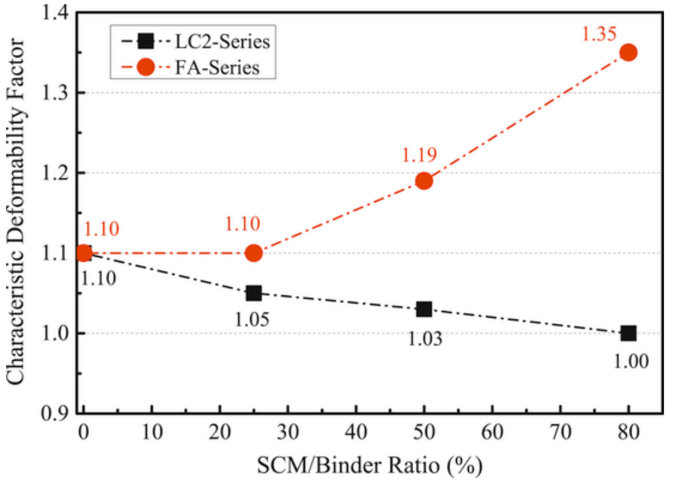

Fig. 9. Characteristic deformability factor of standard mortars for LC2-Series and FA-Series. Blended cement with LC2 can lead to significant reduction of the workability.

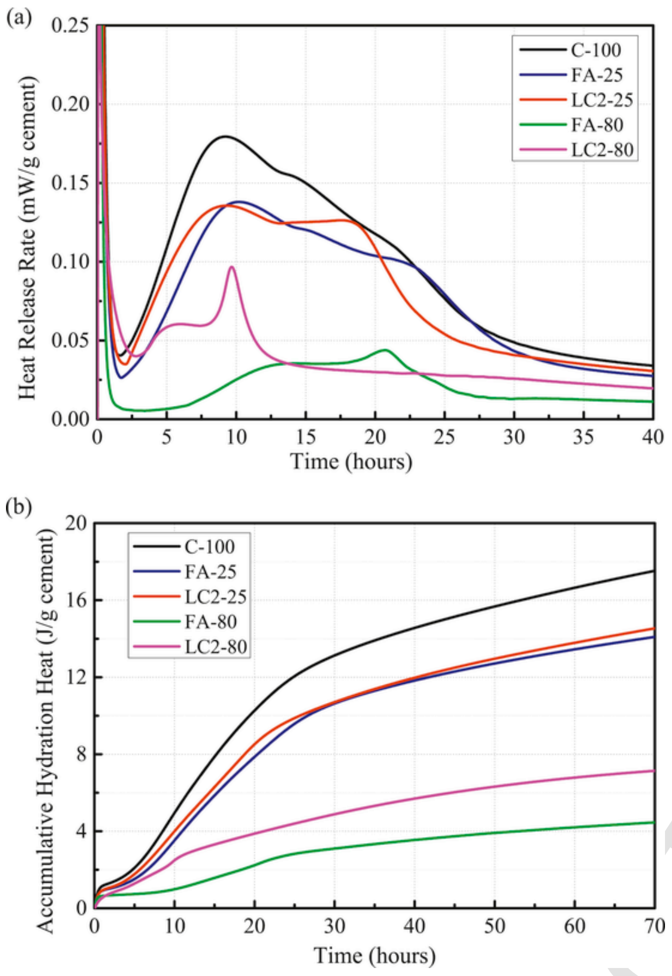

Fig. 10. Isothermal calorimetry results per gram of blended cement: (a) hydration heat release rate at the early stage; (b) accumulative hydration heat up to 72 h. Blended cement with LC2 has lower heat release rate and total hydration heat than ordinary Portland cement.

Table 3
Embodied energy and carbon emission, and material cost of ingredients.

Material	Embodied Energy (MJ/kg)	Carbon Emission (kg eq.CO_2/kg)	Material Cost (HKD/metric ton)
CEM I Portland Cement (52.5N)	5.5 (Hammond and Jones, 2019)	0.912 (Hammond and Jones, 2019)	800
Fly Ash	0.1(Hammond and Jones, 2019)	0.008 (Hammond and Jones, 2019)	400
LC2 Blend	3.04 (Gettu et al., 2019)	0.21 (Gettu et al., 2019)	400 [a]

[a] No commercial data is available yet. The value is assumed here.

At 90-day age (Fig. 12b), the trend of $COST_{90}$ is similar to $COST_{28}$ for both LC2-Series and FA-Series with various SCM replacement levels from 25% to 80%. The trends of EI_{90} and CI_{90} for LC2-Series are also similar to those at 28 days. On the other hand, the EI_{90} and CI_{90} of FA-Series decrease with an increasing fly ash dosage from 0% to 50%, and then keep almost constant for a higher fly ash dosage at 50–80%. Nevertheless, the CI_{90} of LC2-50, LC2-60 and LC2-70 are still lower than those in FA-Series.

At 360-day age (Fig. 12c), the trend of $COST_{360}$ is similar to $COST_{28}$ as well as $COST_{90}$ for both LC2-Series and FA-Series, but the values for cement with ultra-high dosage of SCM (70% and 80%) are very close to each other. The lowest $COST_{360}$ appears for LC2-50, while LC2-25, LC2-60 and FA-25 also show very low $COST_{360}$. The trends of EI_{360} and CI_{360} for LC2-Series are also similar to those at 28 days as well as 90 days, as the blended cement with LC2 only has less than 10% strength increment from 28 days to 360 days (Fig. 6). On the other hand, due to the significant strength development at late stage for FA-Series (Fig. 6), the EI_{360} and CI_{360} of FA-Series decrease with an increasing fly ash dosage from 0% to 80%.

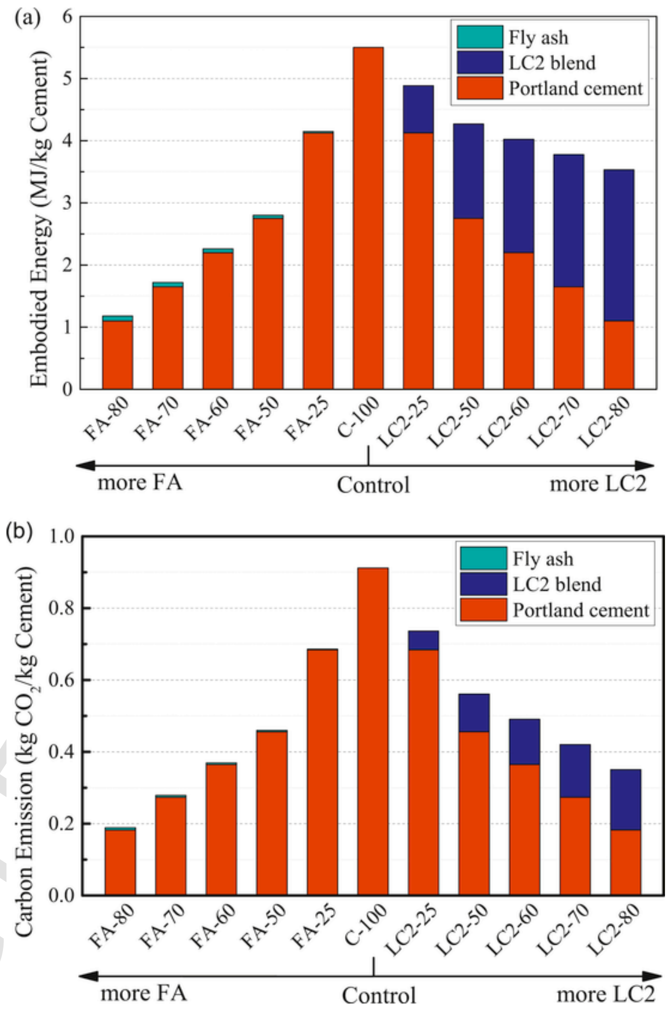

Fig. 11. Unit-mass (a) embodied energy and (b) carbon emission of blended cement with SCM at different replacement levels. Blending a higher dosage of SCM in cement lowers the environmental impact.

Table 4
Comparison on unit-mass material cost of commercial cement and LC2-blended cement.

Strength Class	Type of Cement	Material Cost (HKD/metric ton)
52.5N	Commercial CEM I Cement	800
	LC2-50	600
42.5N	Commercial CEM II Cement	710
	LC2-60	560
32.5N	Commercial CEM II Cement	590
	LC2-70	520

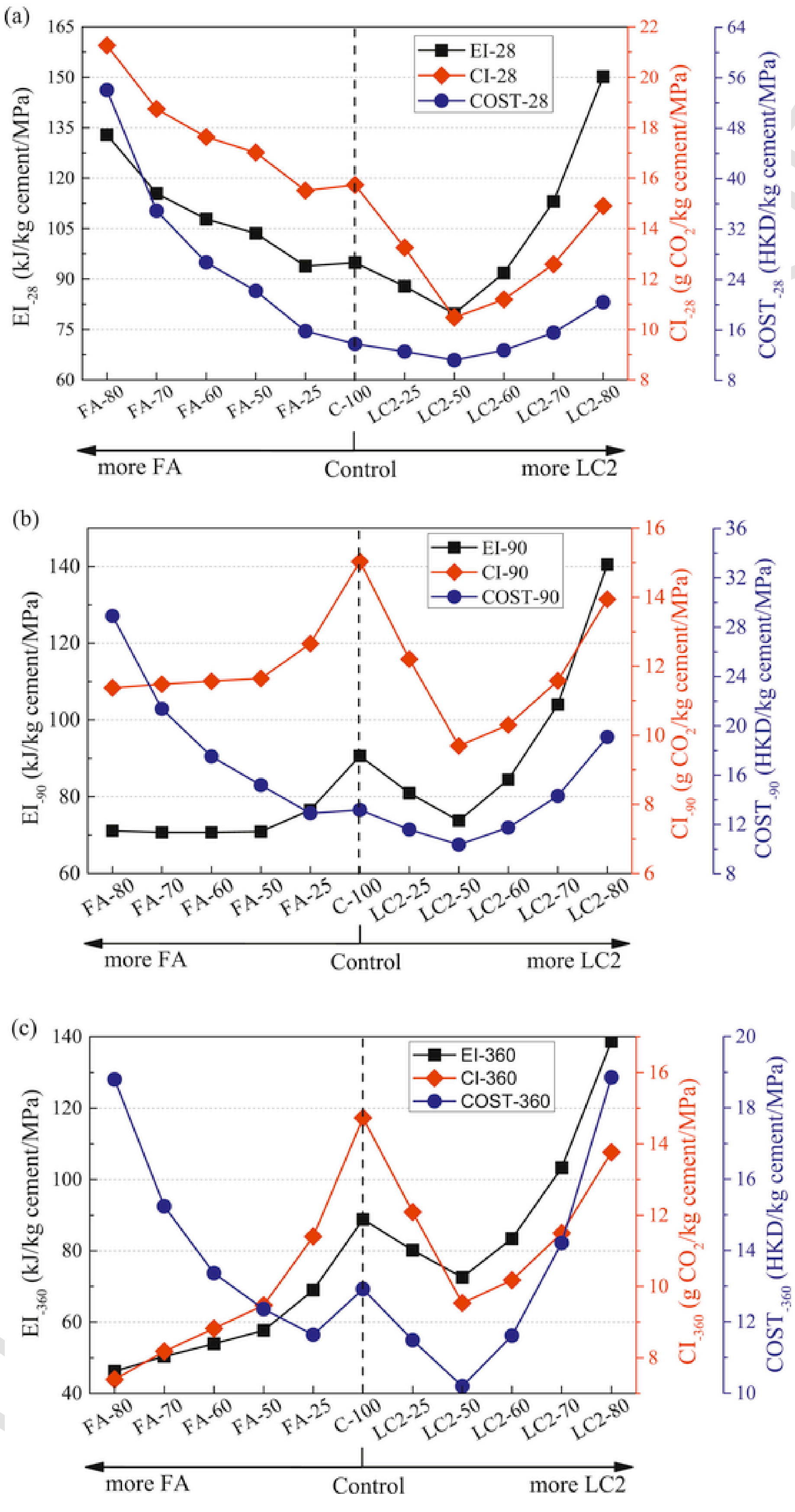
(a)
EI-28
CI-28
COST-28
EI$_{-28}$ (kJ/kg cement/MPa)
CI$_{-28}$ (g CO$_2$/kg cement/MPa)
COST$_{-28}$ (HKD/kg cement/MPa)
165
150
135
120
105
90
75
60
22
20
18
16
14
12
10
8
64
56
48
40
32
24
16
8
FA-80
FA-70
FA-60
FA-50
FA-25
C-100
LC2-25
LC2-50
LC2-60
LC2-70
LC2-80
more FA
Control
more LC2
(b)
EI-90
CI-90
COST-90
EI$_{-90}$ (kJ/kg cement/MPa)
CI$_{-90}$ (g CO$_2$/kg cement/MPa)
COST$_{-90}$ (HKD/kg cement/MPa)
140
120
100
80
60
16
14
12
10
8
6
36
32
28
24
20
16
12
8
FA-80
FA-70
FA-60
FA-50
FA-25
C-100
LC2-25
LC2-50
LC2-60
LC2-70
LC2-80
more FA
Control
more LC2
(c)
EI-360
CI-360
COST-360
EI$_{-360}$ (kJ/kg cement/MPa)
CI$_{-360}$ (g CO$_2$/kg cement/MPa)
COST$_{-360}$ (HKD/kg cement/MPa)
140
120
100
80
60
40
16
14
12
10
8
20
18
16
14
12
10
FA-80
FA-70
FA-60
FA-50
FA-25
C-100
LC2-25
LC2-50
LC2-60
LC2-70
LC2-80
more FA
Control
more LC2

Fig. 12. Comparison of Embodied Energy Index (EI), Carbon Emission Index (CI) and Material Cost Index (COST) per unit compressive strength in blended cements: (a) 28-day, (b) 90-day, and (c) 360-day. Under the same replacement level, LC2-Series showed lower embodied energy and carbon emission per unit compressive strength than FA-Series at the 28-day age, but higher than FA-Series at the 360-day age. Blended cements with 50%–60% LC2 had lower material cost for unit compressive strength than plain cement and blended cement with fly ash.

In summary, though blended cements with 50%–60% LC2 showed slightly lower compressive strength than the plain Portland cement, it has superior environmental and economic benefits by considering the environmental impact and material cost per unit strength. Under the same replacement level, LC2-Series showed lower embodied energy and carbon emission per unit compressive strength than FA-Series at the 28-day age, but higher than FA-Series at the 360-day age.

4. Conclusions

This study examined the feasibility of producing an economical and greener cement by blending LC2 at a high dosage of 50–80% by weight of cement. It extended the earlier research that demonstrated the use of LC2 up to 50% replacement of clinker that produced a cement equivalent to Ordinary Portland Cement of 52.5N grade. Compressive strength, fresh property, hydration heat as well as environmental impact of the blended cement were investigated. The following conclusions can be made:

1) Blended cements with 50%, 60% and 70% LC2 achieved compressive strength of 53.6 MPa, 43.9 MPa and 33.4 MPa at 28 days, respectively; they fulfilled the 28-day strength requirements for 52.5N, 42.5N and 32.5N cements as per BS EN 197-1, respectively.
2) LC2 has a higher cementing efficiency and can develop higher early strength than siliceous fly ash. Specifically, the blended cements with 50%, 60% and 70% LC2 achieved compressive strength of 34.2 MPa, 28.2 MPa and 21.7 MPa at 3 days, respectively.
3) The FA-Series blended cements had 15.1–66.6% less embodied energy and 6.8–45.7% less carbon emission than the LC2-Series with the SCM replacement level from 25% to 80%. However, under the same replacement level, LC2-Series showed lower embodied energy and carbon emission per unit compressive strength than FA-Series at the 28-day age, but higher than FA-Series at the 360-day age.
4) Compared with plain Portland cement, blended cements with 50–60% LC2 had sufficient compressive strength, lower hydration heat, lower environmental impact and material cost per unit strength, but reduced workability.

In the context of the substantial contribution of Portland cement manufacture towards the climate crisis, these findings would help the efforts to reduce the carbon footprint in the construction industry. Ongoing research would reveal the validity of these conclusions under different water/binder ratios and curing conditions. In addition, the use of such blended cements in practical concrete mixes is being currently explored to demonstrate their practical utility. Further research on the volume stability (shrinkage and creep) and durability (particularly the carbonation resistance) of blended cements with high-dosage LC2 is recommended.

CRediT authorship contribution statement

Jing Yu: Investigation, Data curation, Writing - original draft. **Hao-Liang Wu:** Investigation, Data curation, Writing - original draft. **Dhanada K. Mishra:** Conceptualization, Writing - review & editing. **Gengying Li:** Supervision. **Christopher KY. Leung:** Supervision, Resources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence

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