

TL.HKU/0123-21
Design of Durable and Sustainable Concrete with Natural Pozzolana

Final Report

To



Submitted by

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30 November 2021

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1. Introduction

The production and excessive use of ordinary Portland cement (OPC), a key element of concrete is a huge environmental concern due to the resource's consumption and energy use (Celik et al., 2019). The world's cement production has reached to 4.1 billion tonnes in 2020 (Statista, 2021), contributed to 7% of the total global carbon emission (Ventura et al., 2021). The use of sustainable cement or less amount of high emission-intensive OPC is recognized as a most feasible and efficient solution for durable and sustainable concrete production. Replacing OPC with supplementary cementitious materials (SCMs) is a globally acceptable technique to improve the concrete structures and associated environmental impacts. Different types of industrial by-products, waste materials, as well as natural pozzolana, are used in such purposes due to their pozzolanic activity.

Considering the strength and durability of ground granulated blast furnace slag (GBFS), a by-product from the steel industry, is a commonly used SCMs globally. In addition to excellent mechanical performance, the incorporation of GBFS can significantly reduce the environmental impacts. Pulverized fly ash (PFA) produced from coal-fired power plants is also generally used SCM for its pozzolanic properties. The use of certain amount of PFA in concrete showed the good mechanical and durability performance. Similarly, metakaolin is high strength gain clay mineral-based SCM and used in concrete, whereas recently developed limestone-calcined clay cement (LC3) is also in concrete. The use of naturally abundant materials, for instance, natural pozzolana to partially substitute OPC in concrete can a viable solution in many regions.

However, it is essential to explore other potentially alternative SCMs when imbalanced distribution of industrial SCMs is occurred globally (Li et al., 2019). The problem can further be accelerated as the green transition of electricity production may continuously shortage of PFA supply in respect to its demand in many regions of the world. For example, more than half of the total PFA used in Hong Kong is imported, and it will be further increased when coal-based fired plans will gradually be shifted to natural by 2030 according to Hong Kong's climate action plan (Environment Bureau, 2017). In addition, the sourcing of raw materials for concrete mixes is important as the distance may significantly influence the environmental impacts (Göswein et al., 2018; Hossain et al., 2019). In such case, other than commonly used SCMs (e.g. PFA and GBFS), potential sources such as LC3, natural pozzolana, etc. can be effective alternatives to meet the ever-increasing demand of SCMs, particularly for a resource-scarce highly urbanized regions like Hong Kong.

2. Objectives

The key objectives of this project are:

- 1) To examine the mechanical performances and durability in order to evaluate the feasibility of using VA-based natural pozzolana in concrete production, compared to that of its counterparts; and
- 2) To evaluate the comprehensive environmental sustainability of VA-based concretes compared to its conventional counterparts.

3. Key deliverables

- 1) Systematically assessed mechanical performances and durability VA-based natural pozzolana as a replacement to OPC in concrete products to demonstrate its feasibility for being used in Hong Kong's construction industry. The results will produce valuable guidelines for the use of VA for producing sustainable and high strength concrete in Hong Kong.
- 2) Lifecycle assessment (LCA) to provide comprehensive environmental evaluation of VA-based concretes compared to its conventional counterparts which can be used as a reference for sustainable design of concretes. The developed local lifecycle inventory (LCI) and LCA methods can be used as a reference in further LCA of concrete / construction products in Hong Kong in future.
- 3) The technical results, and environmental evaluation would be used as performance benchmarking with respect to its counterparts (e.g. OPC and other industrial supplementary cementitious materials (SCMs) such as FA and GBFS) and they can also be used as a reference in the future.
- 4) Practical guidelines for the concrete producers based on the anticipated results, and this would help promote the sustainability of the construction industry in Hong Kong.

Part I: Technical Properties

1. Material and methods

1.1. Mixture proportions

In this study, six mixtures were cast. The binder volume content and aggregate volume content were set as the same. Table 1 shows the mixture proportions of concrete. The NC in Table 1 uses OPC as the binder and serves as the reference mixture. The SC, PC, and VC replaces OPC partially by 30% GGBS, PFA, and VA by volume, respectively. The VLC replaces OPC partially by 20% VA and 10% LS by volume, and the MLC replaces OPC partially by 20% MK and 10% LS by volume. The raw VA used in this study was sourced from Indonesia. Before being used for production of concrete specimens, the raw VA was ground using a ball mill and sieved to remove larger particles and other debris. After that the VA was dried in oven at 105 °C for 24 h. The OPC complying with the requirement in BS EN 197-1 class 52.5 N [1] was used for concrete production. The chemical characteristics of the OPC, PFA, GGBS, VA, MK, and LS are summarized in Table 2. The pozzolanic reactivity in the table was measured using the modified Chapelle test [2, 3]. The particle size distribution of the OPC, PFA, GGBS, VA, MK, and LS is shown in Fig. 1.

Table 1. Mixture proportions of concrete.

Materials (kg/m ³)	Mixture ID					
	NC	SC	PC	MLC	VLC	VC
Ordinary Portland cement (OPC)	450.0	315.0	315.0	315.0	315.0	315.0
Ground granulated blast furnace slag (GGBS)	0.00	119.4	0.00	0.000	0.0	0.00
Pulverized fly ash (PFA)	0.00	0.00	104.8	0.00	0.00	0.00
Volcanic ash (VA)	0.00	0.00	0.00	0.00	74.3	109.0
Metakaolin (MK)	0.00	0.00	0.00	74.3	0.00	0.00
Limestone (LM)	0.00	0.00	0.00	37.1	37.1	0.00
Coarse aggregates	1015	1015	1015	1015	1015	1015
Fine aggregates	745	745	745	745	745	745
Water	198	198	198	198	198	198
Superplasticizer	2.2	2.3	2.0	5.2	3.0	3.7

[Note: NC: normal concrete produced with OPC; SC: slag concrete with 30% OPC replacement; PC: fly ash concrete with 30% OPC replacement; MLC: concrete with 20% and 10% OPC replacement with metakaolin and limestone, respectively; VLC: concrete with 20% and 10% OPC replacement with volcanic ash and limestone, respectively; VC: natural pozzolana concrete with 30% OPC replacement]

Table 2. Chemical characteristics of cement and SCMs.

Oxides (% by mass)	OPC	GBFS	PFA	MK	LS	VA
Na ₂ O			1.99			1.91
MgO	0.84	6.33	2.70		0.95	0.81
Al ₂ O ₃	3.95	13.81	20.38	43.05	0.08	14.86
SiO ₂	19.88	32.53	50.74	54.67		71.73
SO ₃	5.06	3.16	1.61		0.07	0.05
K ₂ O	0.57	1.13	1.81			2.41
CaO	65.54	40.73	9.37		54.57	1.84
TiO ₂		1.28	1.15	0.69		0.33
Fe ₂ O ₃	2.73	0.31	6.19	0.43	0.03	2.60
MnO		0.17	0.14			0.09
Loss on ignition at 900 °C	1.43	0.54	3.91	1.15	45.43	3.37
Pozzolanic activity (mg of Ca(OH) ₂ consumed per g of pozzolan)	-	-	545	673	-	321

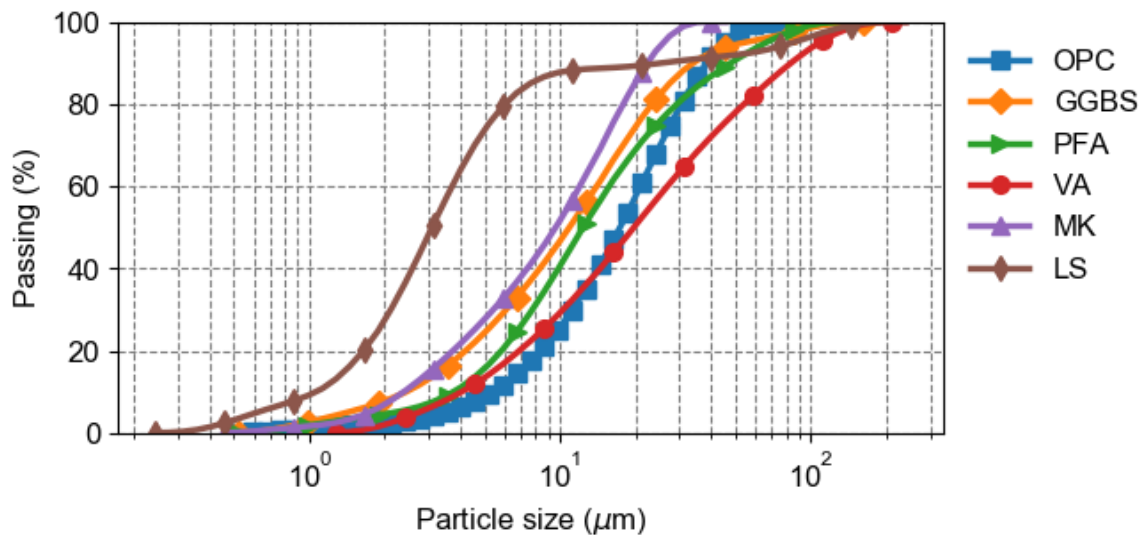


Fig. 1. Particle size distribution of powder materials.

1.2. Test specimens, curing conditions and testing details

Comprehensive series of tests on mechanical and durability properties of these six concrete mixtures such as compressive strength, splitting tensile strength, flexural tensile strength, rate of absorption of water, chloride diffusion coefficient, and alkali-silica reaction vulnerability were carried out.

Compressive strength test was conducted on 100 mm cubic specimens at an age of 3, 28, and 56 days according to BS EN 12390-3 [4]. The setup for compressive strength test is shown in Fig. 2(a). The splitting tensile strength test was conducted on $\Phi 100 \times 200$ cylindrical specimens at an age of 3, 28, and 56 days according to BS EN 12390-6 [5]. The setup for splitting tensile strength test is shown in Fig. 2(b). The loading rate for both compressive strength test and splitting tensile strength test was set to be 3 kN/s. The flexural tensile strength test was conducted on $100 \times 100 \times 500$ prismatic specimens at an age of 3, 28, and 56 days as per BS EN 12390-5 [6]. The setup for flexural tensile strength test is shown in Fig. 2(c). The loading rate for flexural tensile strength test was set to be 1 kN/s. For each type of mechanical strength tests at each age, three specimens were tested and average values were reported. The specimens were demolded one day after casting and then cured in a water tank at about 23 °C until the testing age.

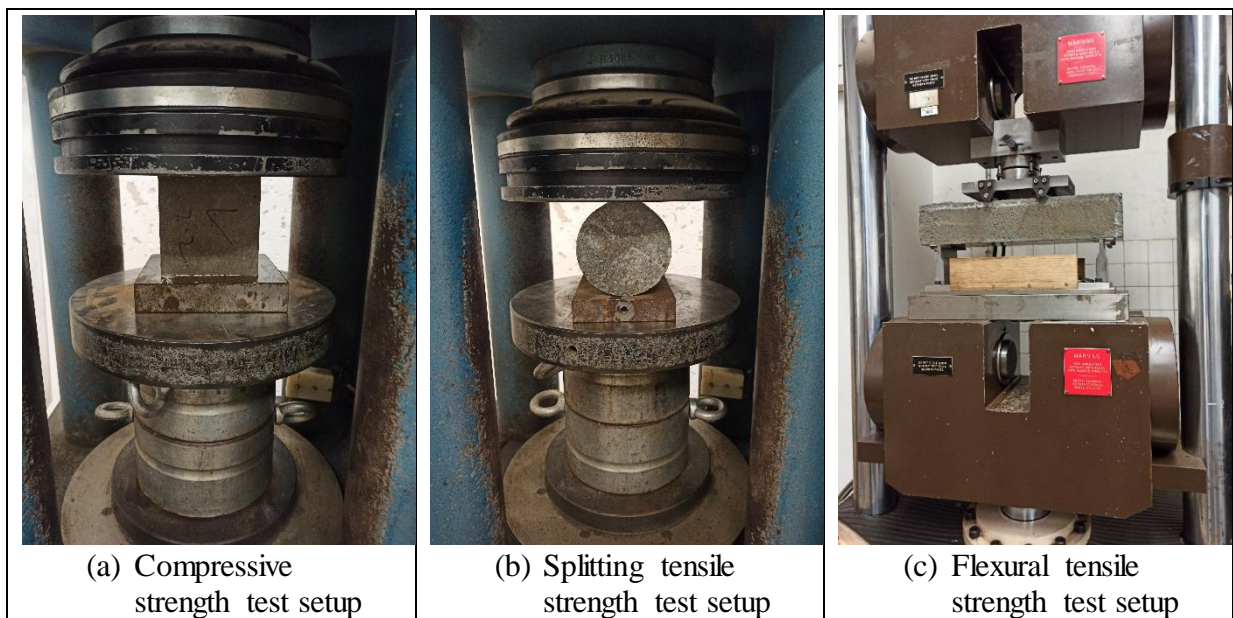


Fig. 2. Mechanical test setup.

The initial and secondary rate of absorption of water of concrete is measured using $\Phi 100 \times 200$ cylindrical specimens as per ASTM C1585 [7]. Three specimens were prepared for each concrete mixture and mean values were reported. The specimens were demolded one day after casting and cured in a water tank at about 3 °C for 28 days. The specimens were then taken out of the water tank and cured in an environmental chamber at a temperature of 50 °C and RH of 80% for 3 days. After that, the specimens were stored in

sealed containers ($T \sim 23^\circ\text{C}$ and $RH \sim 65\%$) for about 56 days before the start of water absorption test.

Mortar mixtures as per the proportions of ASTM C1567, instead of the concrete mixtures in Table 1, were used for investigating the potential alkali-silica reactivity. It should be noted that the replacement percentages of the SCMs in the mortar mixtures are kept the same as those in the concrete mixtures. The main difference was that crushed fine granite aggregates were used in the mortar mixtures, while both fine granite aggregates and coarse granite aggregates were used in the concrete mixtures. For each of the six mortar mixtures, three $25 \times 25 \times 285$ mm prismatic specimens were prepared for the accelerated mortar bar test. Two metal gage studs were embedded at the ends of the specimens to facilitate length measurements. A digital length comparator with an accuracy of 0.001 mm was used to measure the length change of the specimens as shown in Fig. 3. The specimens were demolded one day after casting and cured in water at 80°C for another one day. Finally, the specimens were stored in a 1 M NaOH solution at 80°C for 14 days. The length of the specimens was measured after demolding, 1 day of water bath, and 1, 2, 3, 5, 7, 10, and 14 days submersion in the NaOH bath.



Fig. 3. Apparatus for measurement of length changes.

For bulk diffusion test, one 100 mm cube specimen was cast for each of the six concrete mixtures. The specimens were demolded after one day, and were then stored in water condition for 27 days. After that, the specimens were air-dried for 1 day before sealing the four lateral sides using epoxy resin. The sealed specimens were then immersed in saturated lime water for 3 days before being immersed in 165 g/L NaCl solution for 60 days. The measurement of acid-soluble chloride contents at different depths of concrete specimens can be referred to ASTM C1152 [8].

3. Results and Discussion

3.1. Setting times

Fig. 4 shows the initial and final setting times of the binders of the six types of concrete. It is evident from the figure that the addition of SCMs to OPC increases the initial and final setting times to different extent. In general, the setting times of VLC and MLC are closest to those of NC: the initial setting times of VLC and MLC are respectively 25 and 40 minutes longer than that of NC, and the final setting times of VLC and MLC are respectively 20 and 5 minutes longer than that of NC. It is noted that 10% replacement of VA in VC by LS reduces initial and final setting times by 40 and 30 minutes, respectively. This decrease in the setting times by the addition of limestone can be explained by the effect of particle size on the hydration process. Due to their small size, limestone particles fill the interstices of the cement particles and serve as nucleation sites for the hydration and as a result accelerate the rate of cement hydration. Among the six cementitious binders, the PC has the largest initial and final setting times, which are respectively 270 and 420 minutes.

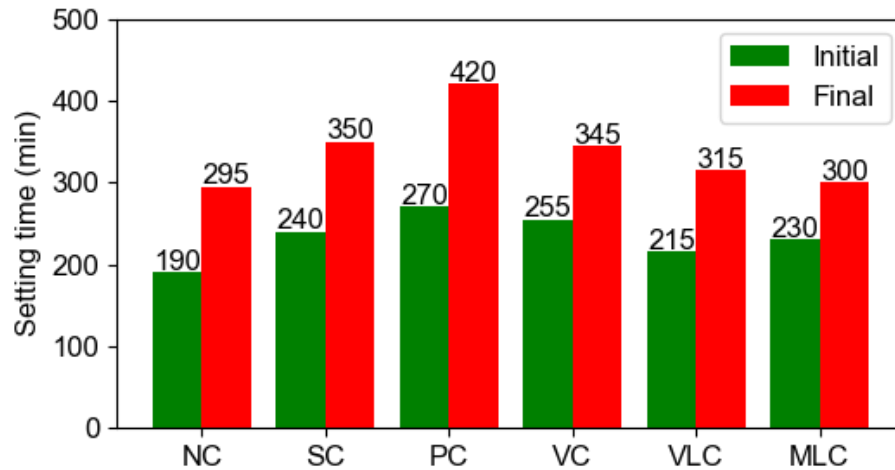
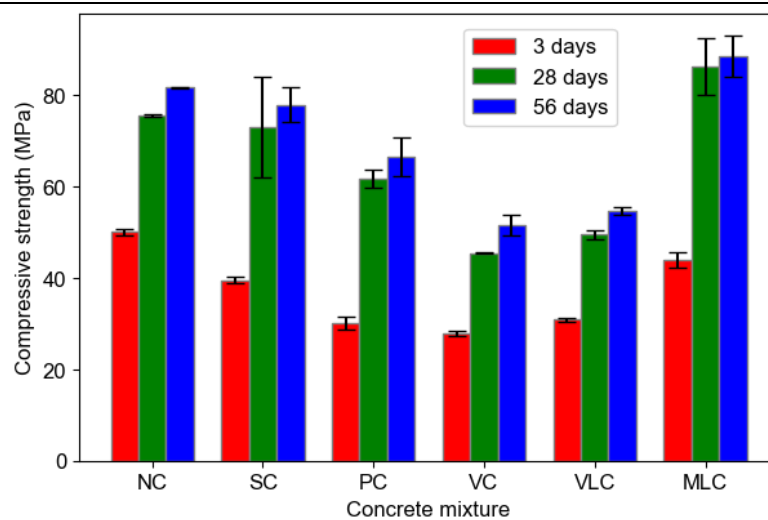


Fig. 4. Initial and final setting times of cementitious binders of the concretes.

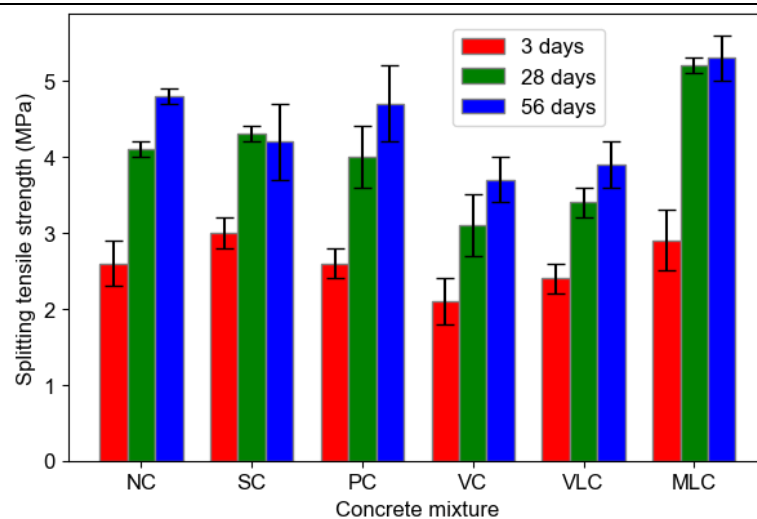
3.2. Mechanical strength

3.2.1. Compressive strength

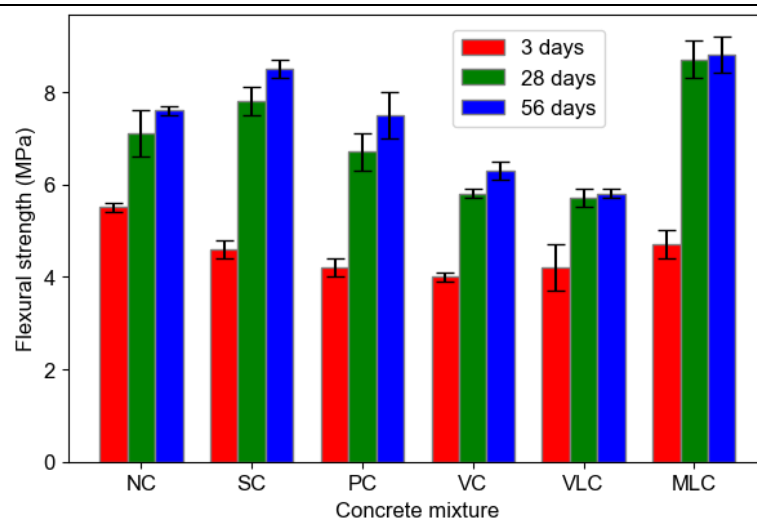
Fig.5(a) shows the compressive strength development of the six concrete mixtures from an age of 3 days to 56 days. The compressive strength of all the six concrete mixtures increases as their ages increase, however, the increase from 28 days to 56 days is very small compared to that from 3 days to 28 days. At 3 days, the NC has a compressive strength of 50.1 MPa, which is respectively 10.6, 19.9, 22.3, 19.2, and 6.1 MPa larger than that of SC, PC, VC, VLC, and MLC. This indicates that the addition of SCM, be it industrial byproduct or natural pozzolan, reduces the early-age compressive strength of concrete. Among the five types of concretes with SCMs, the VC has the lowest 3-day compressive strength and the MLC has the highest 3-day compressive strength.



(a) Compressive strength



(b) Splitting tensile strength



(c) Flexural tensile strength

Fig. 5. Mechanical strength development of the six concrete mixtures.

At 28 days, the compressive strengths of the NC, SC, PC, VC, VLC, and MLC increase by 50.8%, 85.2%, 104.8%, 63.7%, 60.5%, and 96.1%, respectively, compared to their respective counterparts at 3 days. It is noted that all the concretes with SCMs have a larger increase in compressive strength than the NC, which is a result of pozzolanic reaction. The 28-day compressive strength of the MLC becomes 86.3 MPa, which is the largest among the six concrete mixtures. This is probably due to the high pozzolanic reactivity of the MK, which promotes the pozzolanic reaction between pozzola and calcium hydroxide (CH) and furthermore accelerates cement hydration. The high degree of pozzolanic reaction in MLC is evidenced by a very small amount of CH in the MLC binder at 28 days as shown in Fig. 6 and Fig. 7.

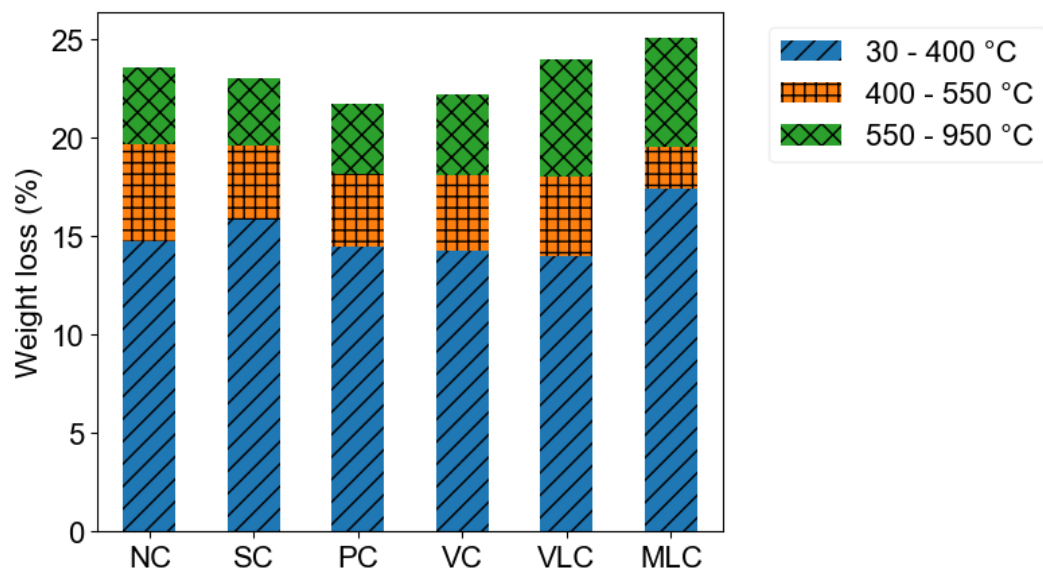


Fig. 6. Distributions of weight loss percentages of cementitious binders at an age of 28 days at various temperature ranges.

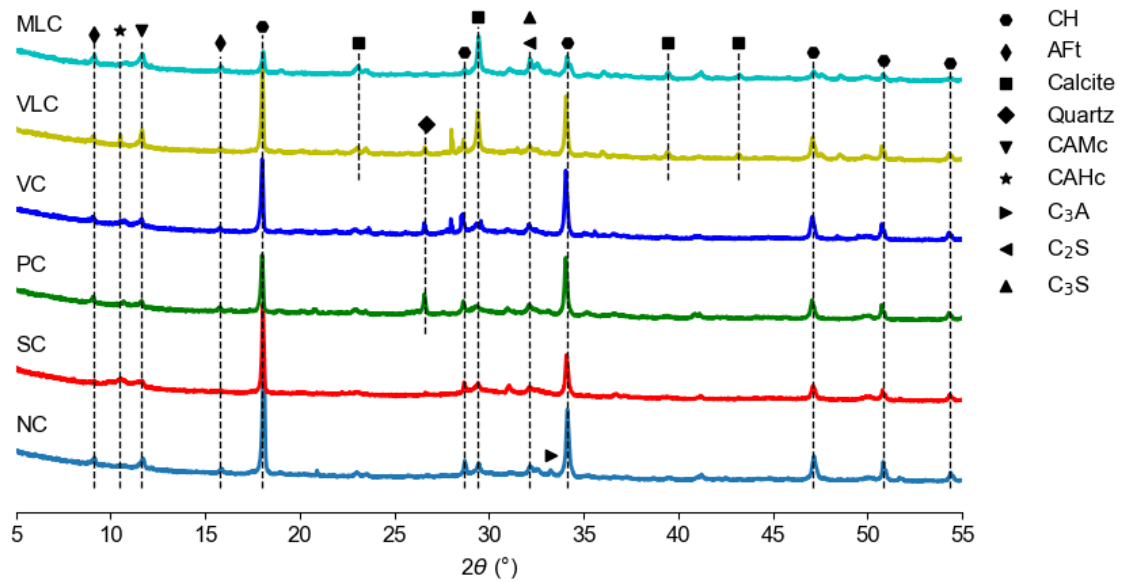


Fig. 7. XRD spectra of cementitious binders at an age of 28 days.

Fig. 6 shows the distributions of weight loss percentages of cementitious binders at an age of 28 days at various temperature ranges. These weight losses of cementitious binders were measured by thermogravimetric analysis (TGA) equipment. The TGA was conducted by heating about 50 mg ground cementitious paste sample from 30 °C to 950 °C at 20 °C/min in nitrogen flow (20 ml/min). The weight loss percentage in between 400 °C and 550 °C corresponds to the CH content in the cementitious binder. As shown in Fig. 6, the CH contents in all the five concrete mixtures containing SCMs are less than that in the NC. Furthermore, the CH content in the MLC is obviously the lowest. This finding is further confirmed in the XRD spectra of these six cementitious binders at an age of 28 days as shown in Fig. 7. As can be seen the figure, the magnitudes of peaks corresponding to the CH phase are the lowest for the MLC.

The 28-day compressive strengths of the NC and SC are comparable and respectively 10.8 MPa and 13.2 MPa lower than that of the MLC. The compressive strength of the PC is in between those of the SC and VLC: 11.3 MPa lower than that of the SC and 12.2 MPa higher than that of the VLC. The compressive strengths of the VLC and VC are considerably lower than those of the other concrete mixtures. This is likely a result of the considerably low pozzolanic reactivity of the VA compared to those of PFA and MK as shown in Table 2. At 56 days, no remarkable change is observed in compressive strengths of the six concrete mixtures.

3.2.2. Splitting tensile strength

5(b) shows the splitting tensile strength development of the six concrete mixtures from an age of 3 days to 56 days. The splitting tensile strength of all the six concrete mixtures increases as their ages increase except for the SC, which shows a very slight reduction from 28 days to 56 days. The 3-day splitting tensile strengths of NC, SC, PC, VC, VLC, and MLC are 2.6, 3.0, 2.6, 2.1, 2.4, and 2.9 MPa, respectively. At 28 days, the splitting tensile strengths of NC, SC, PC, VC, VLC, and MLC increase by 57.7%, 43.3%, 53.8%, 47.6%, 41.7%, and 79.3%, respectively, compared to their respective counterparts at 3 days. It is noted that the splitting tensile strength of MLC is the highest and that of the VC is the lowest among the six concrete mixtures. A bit different from the trend in compressive strength, the splitting tensile strengths of the SC and PC at 3, 28, 56 days are comparable to that of the NC, and the splitting tensile strength of the MLC at 3, 28, 56 days are higher than that of the NC at 3, 28, 56 days. Fig. 8(a) shows that there a good linear correlation between compressive strength and splitting tensile strength of concretes. The splitting tensile strength of concretes is about 1/16 of their compressive strength.

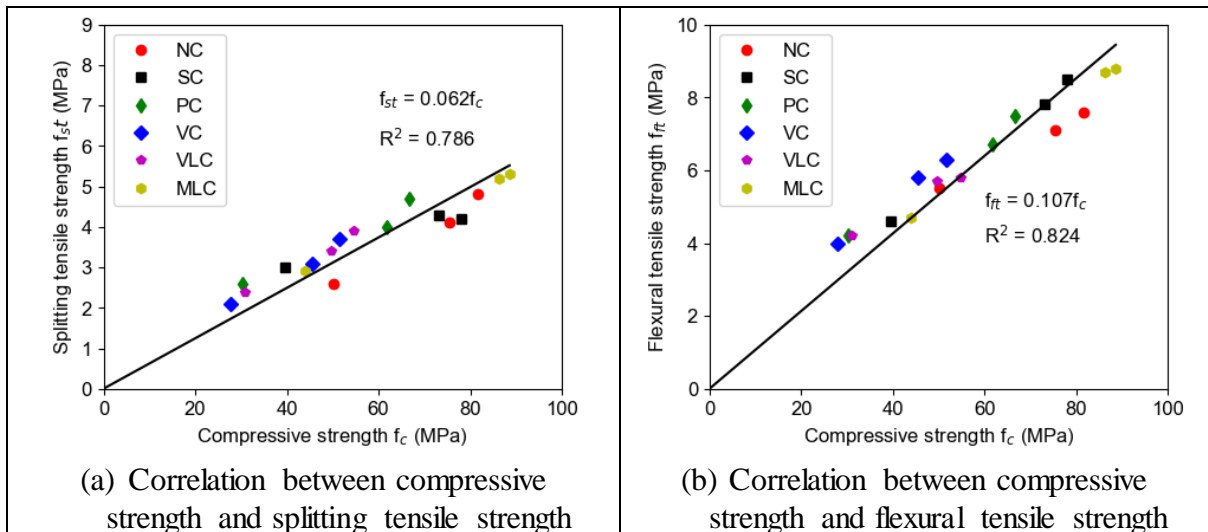


Fig. 8. Relationships between compressive strength and splitting tensile strength and flexural tensile strength of concretes.

3.2.3. Flexural tensile strength

5(c) shows the flexural tensile strength development of the six concrete mixtures from an age of 3 days to 56 days. The flexural tensile strength of all the six concrete mixtures increases as their ages increase, but the increase from 28 days to 56 days is minimal. At 3

days, the NC has the highest flexural tensile strength, followed by the MLC, SC, PC, VLC, and VC. At 28 days, the flexural tensile strengths of NC, SC, PC, VC, VLC, and MLC increase by 29.1%, 69.6%, 59.5%, 45.0%, 35.7%, and 85.1%, respectively, compared to their respective counterparts at 3 days. The MLC has the highest flexural tensile strength at 28 days and 56 days. The flexural tensile strengths of the VC and VLC remain to be the two lowest throughout the testing period. Fig. 8(b) shows that there is a good linear correlation between compressive strength and flexural tensile strength of concretes. The flexural tensile strength of concretes is about 1/10 of their compressive strength.

4. Durability properties

4.1. Rate of water absorption

Fig. 9 shows the initial and secondary rates of absorption of water of the six concrete mixtures. The VC has the highest initial rate of absorption of water, followed by the VLC, PC, NC, SC, and MLC. The replacement of cement by VA and PFA increases the initial rate of absorption of water of concrete, while the replacement of cement by GGBS and MK/LS decreases the initial rate of absorption of water of concrete. However, the replacement of cement by PFA decreases the secondary rate of absorption of water of concrete. The VC has the highest secondary rate of absorption of water, followed by the VLC, NC, SC, PC, and MLC. It is noted that both initial and secondary rates of water absorption of the MLC are much smaller than those of the control mixture, i.e., the NC. The initial rate of water absorption of the MLC is 16.7% that of the NC, and the secondary rate of water absorption of the MLC is 37.1% of that of the NC.

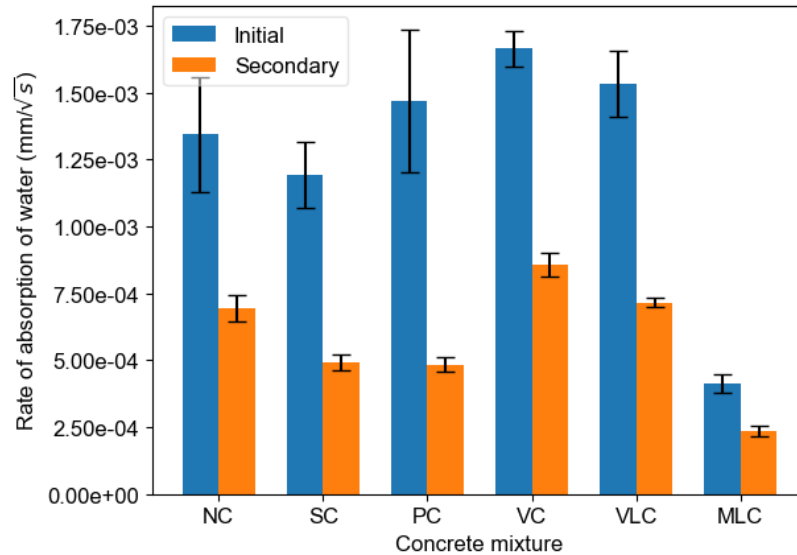


Fig. 9. Initial and secondary rates of absorption of water of the six concrete mixtures.

4.2. Alkali-silica reaction

Fig. 10 shows the expansion of the six concrete mixtures during the entire period of the accelerated mortar bar test. It is noted that the VC and VLC show an obvious shrinkage behavior during the first week. Their shrinkage values peak at the 5th days, and then gradually increase as testing days increase. All the expansion of the six concrete mixtures are far below the ASTM threshold expansion value (0.1%), as shown in the figure. This indicates that all the six concrete mixtures containing granite aggregates are not vulnerable to ASR. Among the six mixtures, the expansion values of the NC and SC at 14 days are the two highest, while the expansion values of the rest four concrete mixtures are minimal at 14 days.

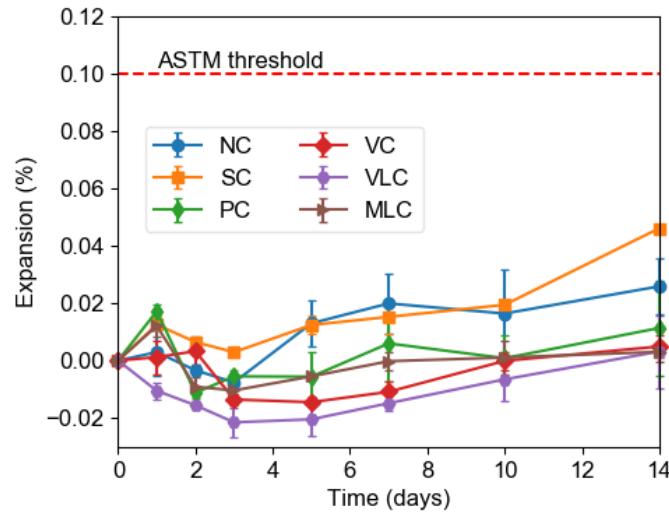


Fig. 10. ASR expansion of the six concrete mixtures.

4.3. Chloride diffusion coefficient

Fig. 11 shows the distribution of total chloride contents in the concrete samples of the six mixtures. As can be obviously observed from the figure, the chloride contents of NC at 6 mm, 8.5 mm, 11.5 mm, and 14.5 mm are obviously larger than the counterparts of the rest concrete mixtures. The chloride contents of MLC at 2 mm, 4 mm, 6 mm, and 8.5 mm are obviously smaller than the counterparts of the rest concrete mixtures. Among the SC, PC, and VC, the chloride contents of VC at inner layers are higher than those of SC and PC.

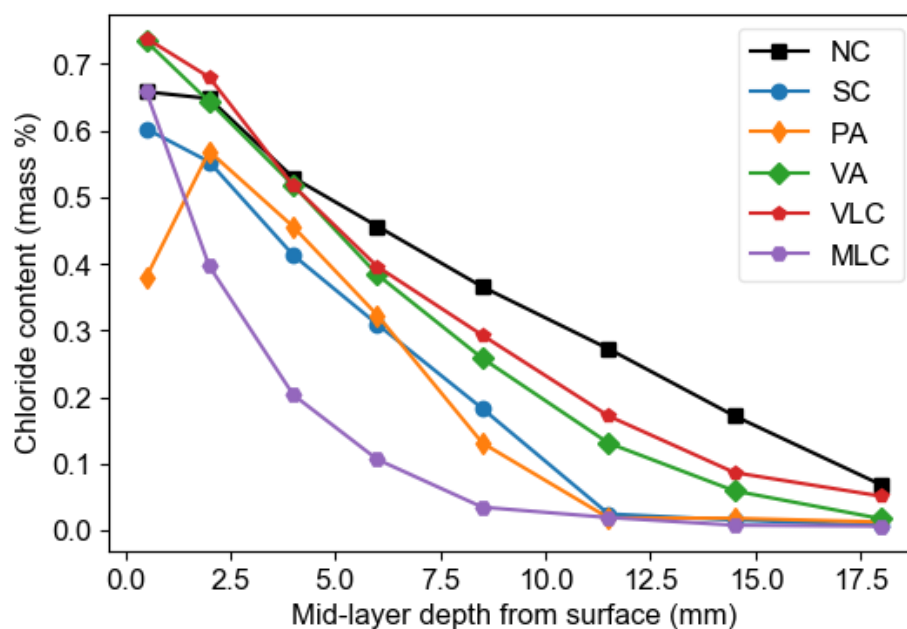


Fig. 11. Distribution of total chloride contents in concrete samples.

Fig. 12 shows the apparent chloride diffusion coefficients of the six concrete mixtures calculated based on least square curve fitting method. As shown in this figure, the apparent chloride diffusion coefficient of NC is much larger than that of the other five concrete mixtures. It is noted that though compressive strength of VC is only half of that of NC, its apparent chloride diffusion coefficient is only about half of the counterpart of NC. But the apparent chloride diffusion coefficient of VC is still larger than that of SC, PC and MLC. Among all the six concrete mixtures, the apparent chloride diffusion coefficient of MLC is notably the smallest.

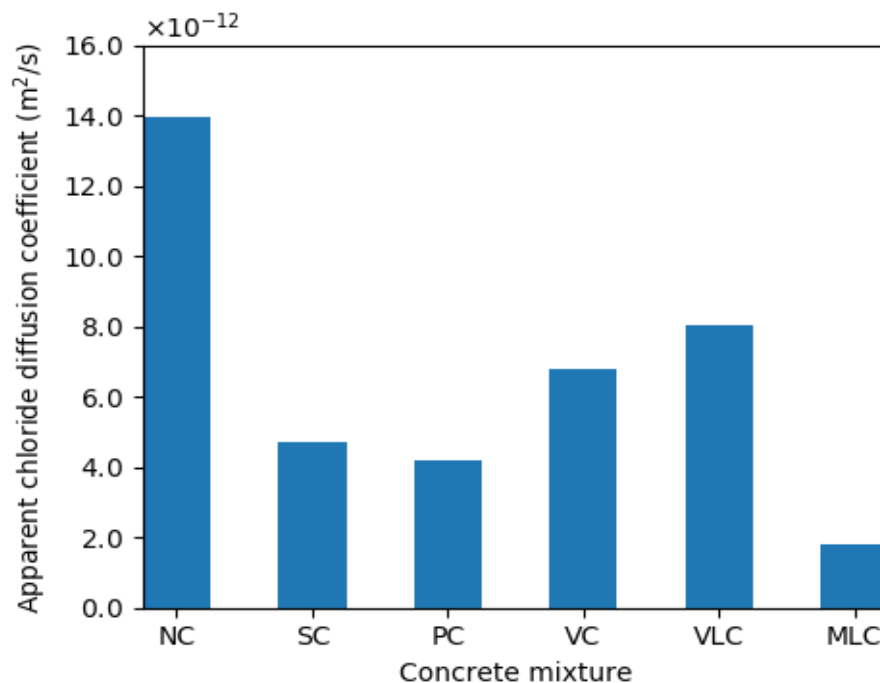


Fig. 12. Apparent chloride diffusion coefficients of six concrete mixtures.

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Part II: Environmental Impacts and Costs Benefit Analysis

1. Materials and Method

1.1. Materials

To evaluate and select the sustainable concrete based on multicriteria analysis, different types of binder materials were used in this study. In addition to locally produced ordinary Portland cement (OPC) and PFA, other SCMs such as GGBS, metakaolin (MK) and volcanic ash (VA) as natural pozzolana together with limestone (LS), were used to design the concretes and comparative analysis. Natural aggregates produced from crushed stone were used as both coarse and fine aggregates, as those are commonly used in Hong Kong. In addition, same sources of superplasticizer and supplied water was used in casting the designed concretes.

1.2. Mix-designs of designed concretes

At first, a total of six different concretes with different binders were designed by considering the same volumetric replacement ratio of OPC by the alternative binders, and same water and aggregates contents. The w/b mass ratio is ranged from 0.44 to 0.47 of the concrete, depending on the density of the binders. The mixtures of the designed concretes are given in Table 1. Concrete produced with only OPC is regarded as normal concrete (NC), whereas 30% OPC replacement by GGBS (SC), 30% replacement by PFA (PC), 20% and 10% OPC replacement with MK and LS, respectively (MLC), 20% and 10% OPC replacement with volcanic ash (VA) and LS, respectively (VLC), and 30% OPC replacement by VA (VC). MLC is also known as LC3 concrete. The considered replacement ratio is mostly acceptable globally, and practicing in Hong Kong (Panesar *et al.*, 2019; Kurda *et al.*, 2018; Yang *et al.*, 2016; Leung and Wong, 2011). The superplasticizer contents were adjusted to obtain the comparable workability. The casted samples (in $100 \times 100 \times 100$ mm standard-sized molds) were cured with tap water, and then 28-day compressive strengths were evaluated according to BS Standard Methods for the designed concretes.

Table 1. Mixtures of the designed concretes.

Materials (kg/m ³)	Concrete codes					
	NC	SC	PC	MLC	VLC	VC
Ordinary Portland cement (OPC)	450.0	315.0	315.0	315.0	315.0	315.0
Ground granulated blast furnace slag (GGBS)	0.00	119.4	0.00	0.000	0.0	0.00
Pulverized fly ash (PFA)	0.00	0.00	104.8	0.00	0.00	0.00
Volcanic ash (VA)	0.00	0.00	0.00	0.00	74.3	109.0

Metakaolin (MK)	0.00	0.00	0.00	74.3	0.00	0.00
Limestone (LM)	0.00	0.00	0.00	37.1	37.1	0.00
Coarse aggregates	1015	1015	1015	1015	1015	1015
Fine aggregates	745	745	745	745	745	745
Water	198	198	198	198	198	198
Superplasticizer	2.2	2.3	2.0	5.2	3.0	3.7
<i>Sum (kg/m³)</i>	2410.2	2394.7	2379.8	2389.6	2387.4	2385.7
w/b ratio	0.44	0.46	0.47	0.46	0.46	0.47

[Note: NC: normal concrete produced with OPC; SC: slag concrete with 30% OPC replacement; PC: fly ash concrete with 30% OPC replacement; MLC: concrete with 20% and 10% OPC replacement with metakaolin and limestone, respectively; VLC: concrete with 20% and 10% OPC replacement with volcanic ash and limestone, respectively; VC: natural pozzolana concrete with 30% OPC replacement]

1.3. Environmental impacts of designed concretes

Environmental LCA is a recognized technique to evaluate the possible environmental consequences of a product, process or system of its whole lifecycle, with four fundamental steps: (i) goal and scope definition; (ii) lifecycle inventory; (iii) impacts assessment; and (iv) results interpretation ((ISO, 2006a,b).

1.3.1. Goal and scope definition

Environmental impacts of the designed concretes produced with commonly used industrial SCMs and potentially alternative SCMs, were evaluated by using LCA technique. The considered system boundary is ‘cradle-to-gate’ (e.g. raw materials acquisition to the production of concrete). In this study, environmental impacts based on the volume (e.g. per 1 m³ concrete production) of the designed concretes was considered as functional unit.

1.3.2. Lifecycle inventory analysis

To conduct the comprehensive LCA of the designed concretes with different alternative materials, lifecycle inventory (LCI) data for various raw materials, energy, and transportation are needed. The raw materials used for the designed concretes with their sourced locations are given in Fig. 1, and the energy requirements for the production of such materials and the studied concretes with their corresponding upstream literature/ database is shown in Tables 2-3.

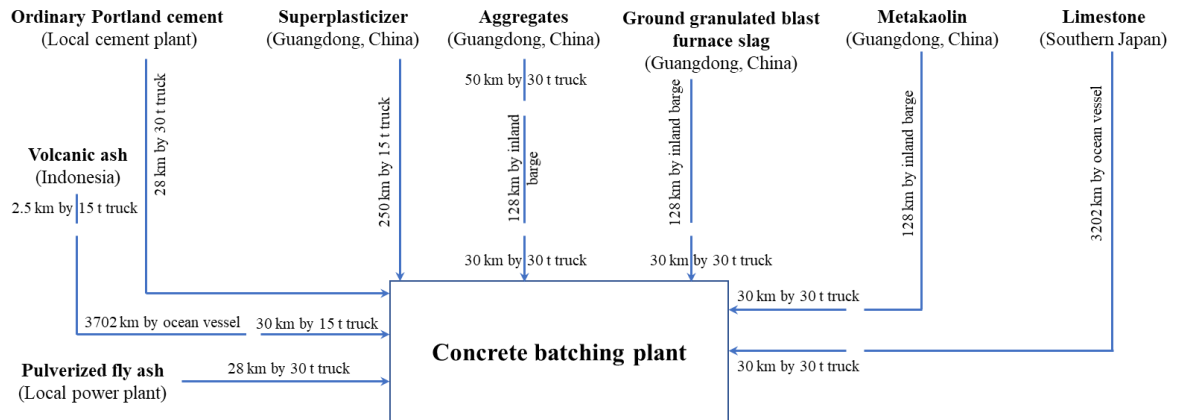


Fig. 1. Transportation of materials for the designed concretes.

Table 2. Raw materials including associated transport for concrete production in Hong Kong.

Materials	Source locations	Distance (km) and transport type	Sources of data	Upstream data/ database
Ordinary Portland cement	Local (average distance from local cement manufacturer to concrete batching plant)	28 km by 30 t trucks	Survey with supplier	CLCD (2010c)
Aggregates (both fine and coarse)	Dongguan in Guangdong Province (China) to Hong Kong Port	50 km by 30 t trucks, and 128 km by inland barge	Survey with supplier	CLCD (2010c); CLCD (2010d)
	To concrete batching plant (averaged)	30 km by 30 t trucks	Estimated	CLCD (2010c)
Pulverized fly ash	Local coal-fired power plant to concrete batching plant (averaged)	28 km by 30 t trucks	Estimated	CLCD (2010c)
Ground granulated blast furnace slag	Guangdong to Hong Kong Port	128 km by inland barge	Hossain <i>et al.</i> (2018)	CLCD (2010d)
	To concrete batching plant (averaged)	30 km by 30 t trucks	Estimated	CLCD (2010c)
Metakaolin	Guangdong to Hong Kong Port	128 km by inland barge	Estimated	CLCD (2010d)
	To concrete batching plant (averaged)	30 km by 30 t trucks	Estimated	CLCD (2010c)
Limestone	Southern Japan to Hong Kong Port	3,202 km by ocean ship (vessel)	Estimated	ELCD (2018)
	To concrete batching plant (averaged)	30 km by 30 t trucks	Estimated	CLCD (2010c)
Superplasticizer	Guangdong to batching plant in Hong Kong (averaged)	250 km by 15 t trucks	Estimated	CLCD (2010c)
VA	VA extraction sites to exported Port	2.5 km by 15 t truck	Survey with supplier	CLCD (2010c)
	Exported Port to Hong Kong Port	3,702 km by ocean ship (vessel)	Survey with supplier	ELCD (2018)
	To concrete batching plant (averaged)	30 km by 15 t trucks	Estimated	CLCD (2010c)

Table 3. Energy consumption and the sources of energy for materials/processes of the designed concretes.

Materials/processes	Energy consumption	Upstream data/ databases
Fine aggregates (crushed stone)	7.57 kWh/t (electricity) and 1.37 L/t (diesel) ^a	CLCD, 2010a,b; CLP, 2016
Coarse aggregates (crushed stone)	6.07 kWh/t (electricity) and 1.37 L/t (diesel) ^a	CLCD, 2010a,b; CLP, 2016
Ordinary Portland cement production	R *	CLCD, 2010e; Hossain et al (2017)
Pulverized fly ash production	9.3 kWh/t (electricity) ^b	CLCD, 2010a; CLP, 2016
Ground granulated blast furnace slag production	72.15 kWh/t (electricity) ^c	CLCD, 2010a; CLP, 2016
Metakaolin production	400 kWh/t (electricity) ^{b,g}	CLCD, 2010a; CLP, 2016
Limestone production	12.7 kWh/t (electricity) ^{b,g}	CLCD, 2010a; CLP, 2016
Superplasticizer production	2500 kWh/t (electricity) ^b	CLCD, 2010a; CLP, 2016
Volcanic ash production	4.93 L/t (diesel for excavation) ^d , 1.89 kWh/t (for processing) ^d and 9.3 kWh/t ^e (for grinding, sieving and drying)	CLCD, 2010a,b; CLP, 2016
Concrete batching	2.5 kWh/t (electricity) for per m ³ concrete ^f	CLCD, 2010a; CLP, 2016

* Referred to the database/references in the right column; ^a Hossain *et al.* (2016); ^b MPA (2009); ^c First-hand data; ^d Dunlap (2003); ^e For further VA processing; ^f Zhang *et al.* (2014); ^g Jones et al. (2011).

1.2.3. Lifecycle impact assessment (LCIA) and interpretations

The raw materials including their transportation and concrete production process were modeled in the SimaPro 9 software, and then the environmental impacts of the designed concrete were evaluated using the IMPACT 2002+ LCIA method (Jolliet et al., 2003). Considering the global significance, four mid-point impact categories such as respiratory inorganics, acidification potential, global warming potential, and non-renewable energy consumption, were selected. Contribution analysis was also conducted to identify the hotspot for future reduction of environmental impacts.

2. Costs evaluation

Due to difficulties of getting the exact costs information for the raw materials from the concrete manufacturers, different statistics, internet sources and first-hand survey data were used in this study (given in Table S2). The costs of OPC and aggregates were collected from the Census and Statistics Department (CSD), whereas the costs for PFA, limestone and GGBS were collected from the Hong Kong Merchandise Trade Statistics (HKMTS). The costs for water was calculated based on the non-domestic water consumption (construction) provided by the Water Service Department (WSD) of Hong Kong SAR. The costs for metakaolin and superplasticizer was collected from internet sources. Considering the range of costs for some materials, average prices were considered. Apart from the (potential) original costs provided by the supplier (assuming the costs of unprocessed VA), an additional 10% costs were included due to processing of materials such as grinding, sieving

and drying for the final use VA in the concrete. The costs related to the raw materials were only considered in this study (excluded the costs of concrete production process including the capital and labour costs, as it assumed those costs would be similar for all of the designed concretes).

3. Multicriteria decision-making process

On the basis of the mechanical strength, environmental impacts and the associated costs, the multicriteria decision-making process was employed to evaluate the performance score for selecting the optimized ones from the designed concretes. In this study, simple additive weighting method was adopted, as it is considered as a simple, mostly applicable and weighted linear scoring method. This method has adopted in several studies, i.e. fiber-reinforced cement composites was evaluated by Akbar and Liew (2021). In this method, the weighted average and performance score are evaluated by considering the normalized value of product and the importance of the selected criteria for the objectives. According to this method, the performance score of concretes designed with different alternative cementitious materials was calculated using the following equation.

$$P_s = \sum_{j=1}^m w_j (x_{ij})_{normal} \quad (1)$$

where, P_s represents the performance score, which was used to calculate the rank of the designed concretes; w_j represents the specific weight (the equal weight (0.167) was assigned to each indicator); $(x_{ij})_{normal}$ represents the normalized matrix of the different indicators (e.g., compressive strength, selected four environmental impact indicators, and the associated production costs of the designed concretes).

$$(x_{ij})_{normal} = \frac{x_{ij}}{\text{Max}(x_{ij})}, \text{ for beneficial criteria, e.g. compressive strength} \quad (2)$$

$$(x_{ij})_{normal} = \frac{\text{Min}(x_{ij})}{x_{ij}}, \text{ for non} \quad (3)$$

– beneficial criteria, e.g. environmental impacts and costs of concretes

4. Environmental sustainability of the designed concretes

4.1. Comparative environmental performance by LCA

The LCA results for per FU (in volume) of the designed concretes are shown in Table 4. In respiratory inorganics, the results showed that NC possess higher impact compared to concretes designed partially replaced OPC with other potential alternatives binders except PC. The results show that 0.342 kg PM_{2.5} eq emission was associated with per m³ of NC production, which is about 5% and 7% higher than that of SC and MLC production. However, much higher saving was observed for VA-based concretes (16-17% compared to NC), as they are responsible for 0.283 – 0.288 kg PM_{2.5} eq for per unit of concretes. (Fig. 2).

In the category of global warming potential, OPC concrete showed the highest CO₂ eq GHG emission compared to all other designs. Results show that about 520 kg CO₂ eq GHG emission was associated with NC, which is 18%, 16% and 19% higher than that of SC, PC and MLC production. This is mainly due to the higher GHG emission factor for OPC production in Hong Kong (Hossain et al., 2017; Zhang et al., 2014). VA-based concretes reduce even higher GHG emission (25%), as they are associated with only 389-392 kg CO₂ eq GHG emission compared to 520 kg CO₂ eq than the NC production. Similar results were reported by Hedayatinia et al. (2019) and Hossain et al. (2021), as those studies reported the potential GHG emission reduction was 24-28% for using 30% VA. MLC production has the similar global warming potential impact with SC, but about 5% lower than the PC production. In addition, about 8%, 11% and 7% lower GHG emission was associated with VLC production compared to SC, PC and MLC, respectively. The corresponding saving was 8%, 11%, and 6% for VC production (Fig. 2). Although similar replacement ratio and locally PFA was used in PC, GHG emission was even higher than GGBS due to allocated upstream emission (the electricity generation from coal-fired power plants).

The results also demonstrated that about 3,616 MJ non-renewable energy was consumed for the production of NC (per FU). The value is 10%, 7% and 9% higher than that of SC, PC and MLC production. The energy consumption for SC, PC and MLC is not significantly varied. VA-based concretes save much higher energy than the OPC concrete (18-19%), as they are associated with 2933-2973 MJ for the production of per unit concrete. The results also showed that VA-based concretes require much lower energy than other alternatives. For example, about 10%, 12% and 11% than the SC, PC and MLC production, respectively. The corresponding saving was 9%, 11% and 10 for VC (Fig. 2). This is because of considerably

lower energy requirements for VA processing (than MK), and the absent of allocated impacts (than industrial SCMs).

Except PC, considerably higher acidification impact was found for the production of NC (Table 4), which is about 11% and 12% higher than that of SC and MLC. The highest acidification impact was for PC (5% higher than the NC) due to the allocated upstream impacts (electricity generation from coal). Compared to NC, about 17% lower acidification impact was observed for VA-based concretes (VLC and VC). The impact was almost similar for both SC and MLC, but lowest for both VLC and VC. Compared to PC, the saving was more than 20% for both VLC and VC.

Table 4. Selected environmental impacts (per m³) for the designed concretes.

Concrete codes	Respiratory inorganics (kg PM _{2.5} eq)	Global warming potential (kg CO ₂ eq)	Non-renewable energy (MJ)	Acidification potential (kg SO ₂ eq)
NC	0.342	520	3616	8.495
SC	0.324	425	3271	7.593
PC	0.364	439	3349	8.957
MLC	0.315	419	3307	7.513
VLC	0.283	389	2933	7.044
VC	0.288	392	2973	7.080

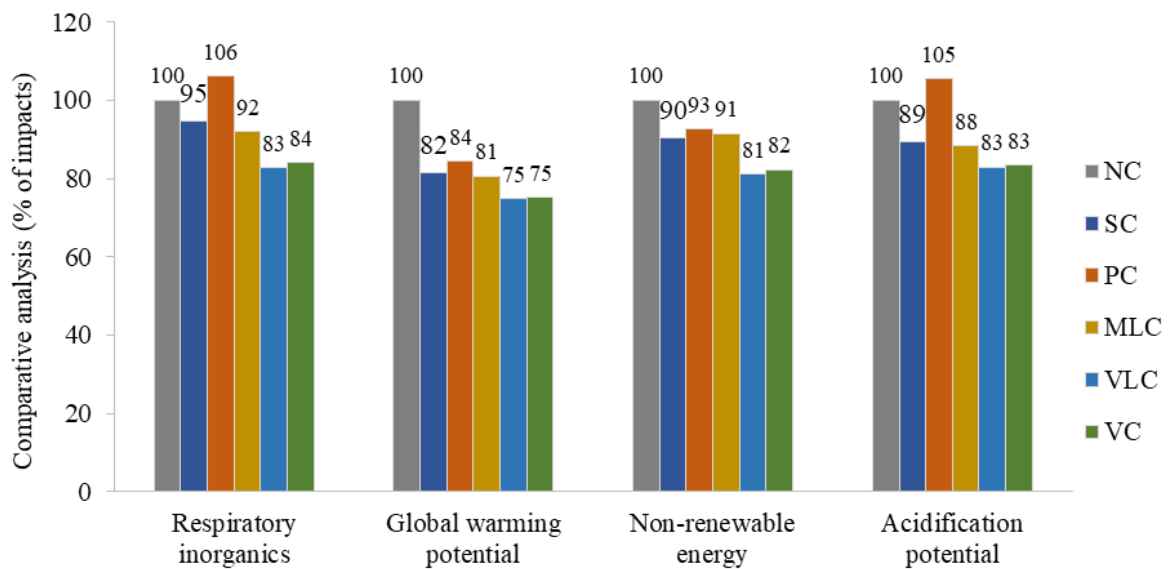


Fig. 2. Comparison of environmental impacts for the designed concretes.

4.2. Contribution analysis

The contribution analysis in the selected impact indicators for the studied concretes is shown in Fig. 3. For NC, OPC contributed to 71% of the total impacts, whereas 26% by

aggregates, 2% by superplasticizer and 1% by the production process. The use of alternative binders significantly reduces the impacts. For example, about 53%, 27%, 17%, 2% and 1% impact were contributed by OPC, aggregates, GGBS, superplasticizer, and the production process, respectively, for SC production. For PC production, the processing and transportation of PFA has contributed higher particle emission. As considerably lower impacts were found for VA-based concretes, about 90% of the total emission was responsible for OPC and aggregates. Irrespective of the type of binders, OPC shared highest percentages of PM_{2.5} emissions for all mixtures. Compared to VA, industrial SCMs have contributed much higher amount of PM_{2.5} eq emission. This is mainly due to the considered allocated impacts for industrial SCMs (Fig. 3).

About 88% of the total GHG emissions was shared by OPC for NC production, whereas 12% by other materials and the production process (Fig. 3). The use of alternative binders could reduce the carbon emissions significantly compared to OPC. About 75%, 14%, 10%, and 1% of the total emission was shared by OPC, aggregates, GGBS, and others, respectively for SC production. As considerably lower impacts were found for VA-based concretes, about 95% of the total GHG emission was responsible for OPC and aggregates (Fig. 3). Similar contributions were also observed for other impact categories such as non-renewable energy consumption and acidification potential (Fig. 3).

Compared to MK and LS, PFA and GGBS contributed much higher amounts considering their upstream allocated impacts. Much lower impacts were associated with VA than all other alternative binders, as less energy requirements for processing and the absent of allocated impacts.

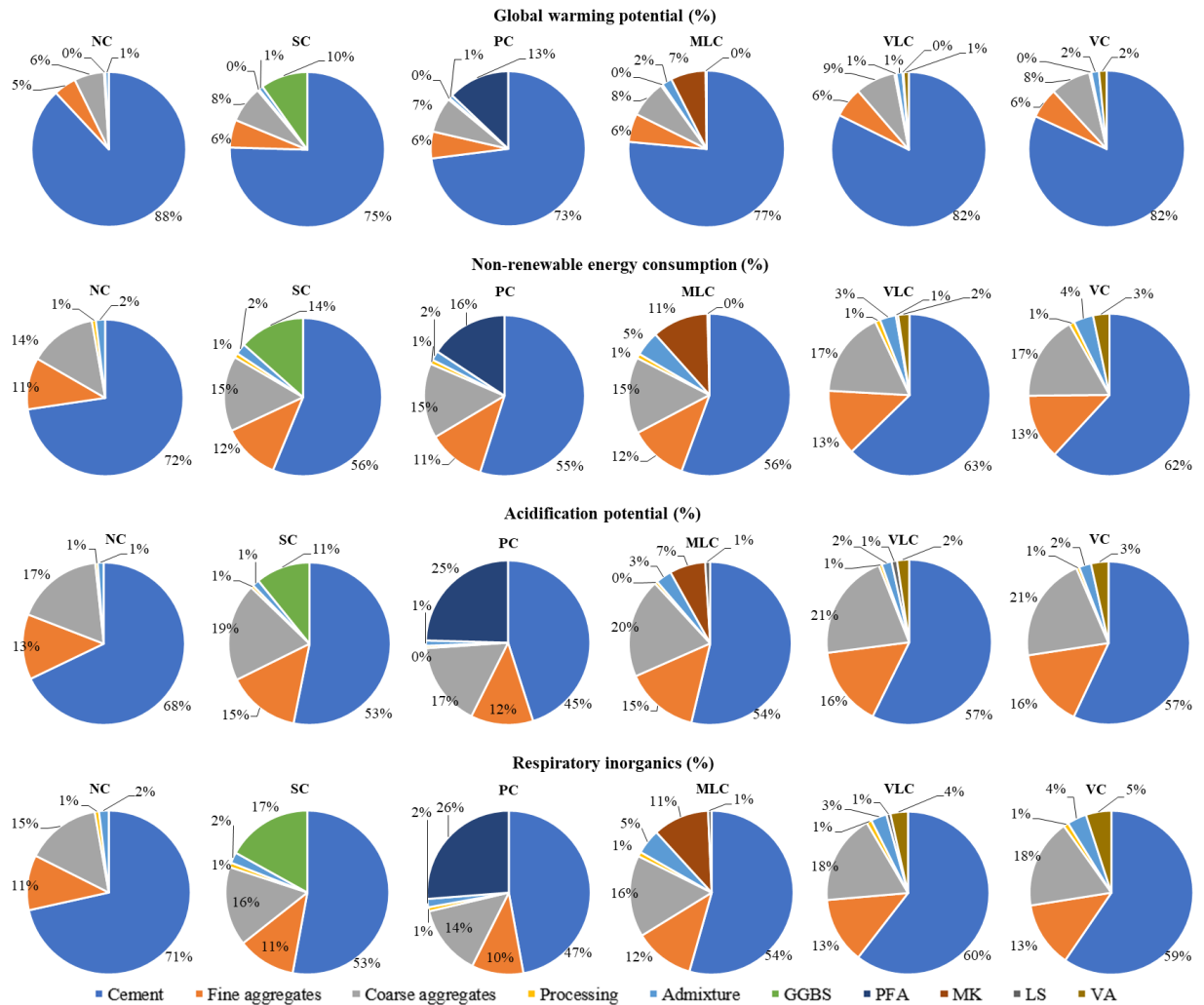


Fig. 3. Contributions analysis of the designed concretes to the global warming potential.

4.3. Cost analysis of the studied concretes

The comparative cost for the production of the designed concretes with different alternatives is given in Fig. 4. It can be seen that the highest cost is associated with the MK-based LC3 concrete production, and the lowest is for PFA-based concrete. The production costs for MLC is about 691 HK\$/m³, which is about 22% higher than the normal concrete (538 HK\$/m³). Similarly, about 26%, 30%, 30% and 34% higher costs are associated with the MLC, compared to that of VA, VLC, SC and PC, respectively. Except MLC, the production of NC has the higher costs than the concretes produced with other alternatives. It is about 5%, 10%, 10% and 15% than VA, VLC, SC and PC, respectively. In MLC, about 25% and 19% of the total costs were contributed by the MK and superplasticizer, whereas the contribution of limestone was very low (0.46%). In VA, only 7% of the total costs was contributed by VA, whereas 18% by superplasticizer. About 5%, 15% and less than 1% of

the total costs were contributed by VA, superplasticizer and limestone in VLC. Similarly, GGBS and superplasticizer has contributed to 9% and 12% of the total costs in SC, whereas the contribution of PFA and superplasticizer was 5% and 11%, respectively in PC.

Although 20% MK (together with 10% limestone) was used in LC3 concrete (MLC), the costs is much higher due to the high cost of MK. In addition, high doses of superplasticizer lead to the high production costs of MLC. In addition, superplasticizer cause to increase the production of VA-based concretes (for both VA and VLC), although the cost of VA is comparable to PFA and GGBS. It can be seen that superplasticizer is an influential factor for all types of concretes, and thus, the use of optimal doses is important for minimizing the total costs. The higher cost of MLC production may be one of the practical limitations.

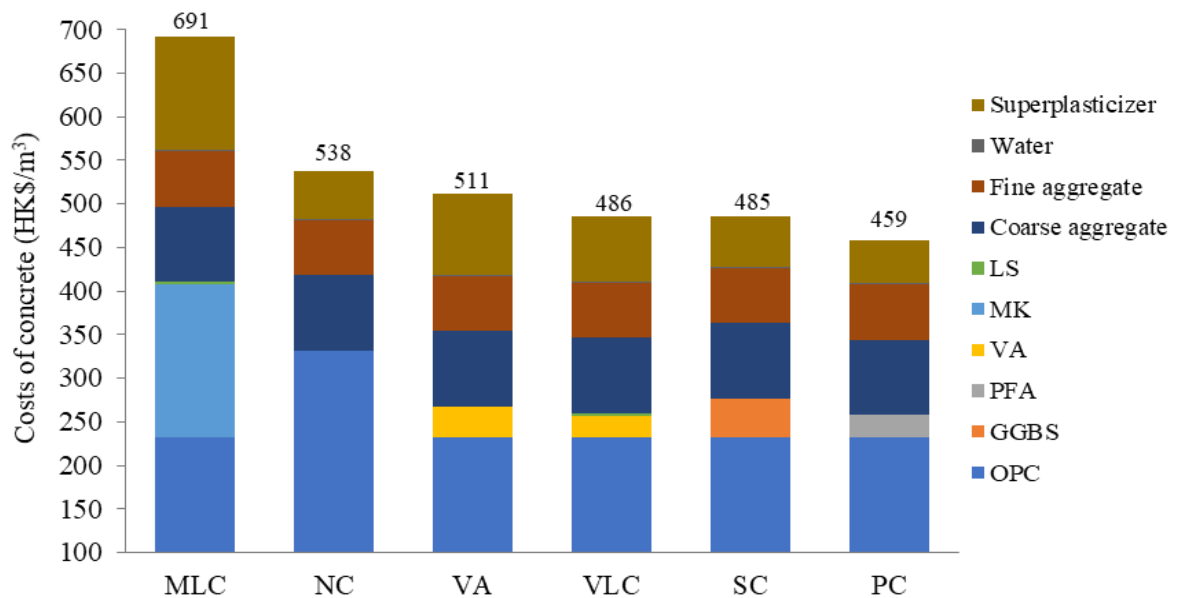


Fig. 4. Comparative cost analysis of the studied concretes.

4.4. Overall performance and ranking

It can be seen from the above results that the deciding parameters such as compressive strength, environmental impact indicators, and the associated production costs are significantly varied among the design concretes. Thus, multicriteria analysis was performed through normalizing the corresponding results to evaluate the performance score for selecting the optimized ones. Using the Equations 1-3, the normalized matrix was calculated from the corresponding value of each indicator selected in this study (Section 3.5), and the results is shown in Fig. 5. The results show that the lowest scores for all environmental

indicators were associated with VLC, while PC for lowest score for costs, and MLC for the highest score for compressive strength.

Based on the results of the selected indicators, the performance score for the designed concretes was calculated to analyze their corresponding ranks using Equation 1. The weighted performance score for the studied concretes is given in Table 5. The results demonstrated that VLC is ranked first among other designs because of the lowest environmental impacts (in all four indicators). While SC is ranked second because of its higher strength, lower environmental impacts and costs, and VC is the third for its lower impacts and cost. Though highest strength and lower impacts, MLC is ranked fourth because of its very high costs. Considering the moderate strength and impacts, PC is ranked fifth although the cost was lowest. Among all design, normal concrete achieved lowest score because of its higher environmental impact and moderate costs and strengths.

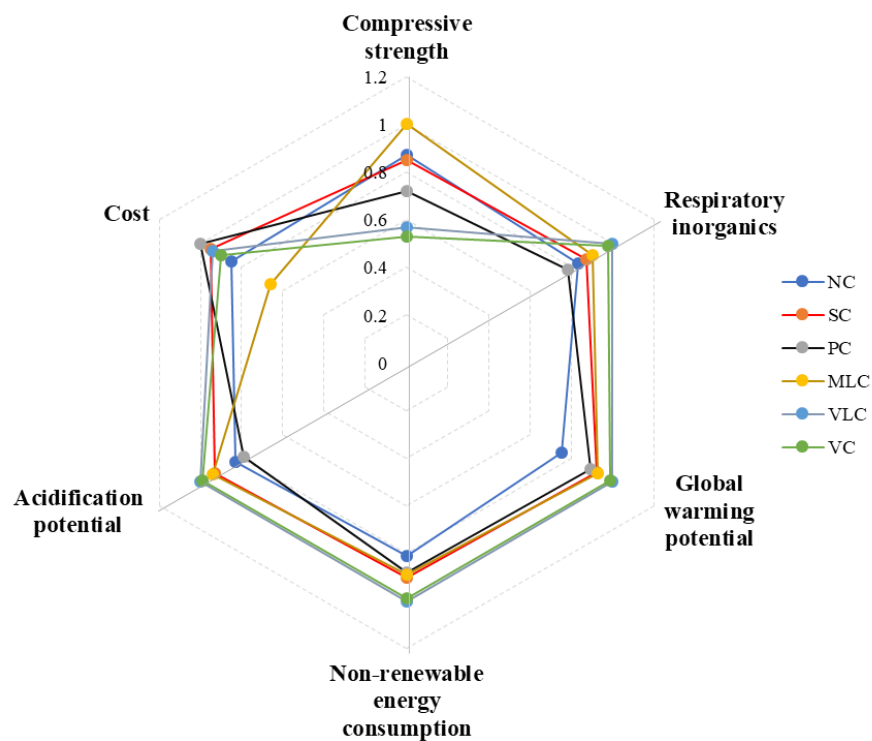


Fig. 5. Normalized matrix score of concretes with different alternatives.

Table 5. Performance (score) and overall ranking of the studied concretes.

Selection criteria	Concretes					
	NC	SC	PC	MLC	VLC	VC
Performance score (P_s)	0.825	0.905	0.845	0.888	0.920	0.898
Overall rank	6 th	2 nd	5 th	4 th	1 st	3 rd

5. Summary

In this study, the mixtures of six types of concrete produced with different types of commonly used binders such as OPC, PFA and GGBS, and potentially alternative ones such as MK and VA, were designed. Summary of this study are highlighted as follows:

- The highest 28 days compressive strength was found for metakaolin-based concrete (30% OPC replaced with 20% MK and 10% LS), as the strength was 12%, 15%, 28%, 42% and 47% higher compared to NC, SC, PC, VLC and VC.
- VA-based concretes showed the lowest strength, even compared to industrial SCMs. For instance, it is about 32% and 20%, and 38% and 26% for VLC and VC, respectively, compared to SC and PC.
- However, VA-based concretes are associated with much lower environmental impacts than all other alternative binders. For example, about 8%, 11% and 7% lower GHG emission was associated with VLC production compared to SC, PC and MLC, respectively. Compared to NC, the saving was even much higher (about 25%).
- The production costs for LC3 concrete (MLC) is about 22%, 26%, 30%, 30% and 34% higher than NC, VA, VLC, SC and PC, respectively.
- Based on the multicriteria analysis, the designed concretes can be ranked as following order: VLC, SC, VC, MLC, PC and NC, indicating that all alternative binders showed the better performance than the OPC concrete, and the VA-based concretes were the best among other supplementary materials. Because the use of 20% VA together with 10% limestone in VLC can enhance the mechanical and environmental performances and reduce the production costs.
- This indicates that all the six concrete mixtures containing granite aggregates are not vulnerable to ASR.
- The apparent chloride diffusion coefficient of VC is only about half of the counterpart of NC, but comparatively higher than that of SC, PC and MLC.

Part III: Comparative Performance of High-volume VA in Concrete

1. Mixtures of concrete

The following mixtures has been designed for using high volume VA in concrete to test the mechanical performance, carbon emission and associated materials costs of concrete.

Table 1. Mixtures of concrete

Materials (kg/m ³)	Control (OPC only)	VA30-0.47	VA30-0.4030	VA30-0.33	VA40-0.33	VA50-0.33
Ordinary Portland cement (OPC)	450.0	315.0	343.0	380.0	326.5	274.2
Ground granulated blast furnace slag (GGBS)	0.00	0.0	0.0	0.0	0.0	0.0
Fly ash (FA)	0.00	0.0	0.0	0.0	0.0	0.0
Volcanic ash (VA)	0.00	109.0	118.6	131.4	179.6	226.3
Metakaolin (MK)	0.00	0.0	0.0	0.0	0.0	0.0
Limestone (LM)	0.00	0.0	0.0	0.0	0.0	0.0
Coarse aggregates	1015	1015.0	1015.0	1015.0	1015.0	1015.0
Fine aggregates	745	745.0	745.0	745.0	745.0	745.0
Water	198	198.0	184.7	168.8	167.0	165.2
W/B ratio	0.45	0.47	0.40	0.33	0.33	0.33
Superplasticizer	2.2	3.7	5.0	8.2	9.0	10.8

2. Initial setting time of cementitious paste

The initial setting time of cementitious paste is shown in Fig. 1. The initial setting time of VA30-0.47 is slightly higher than control, but comparable to other SCMs. With reducing w/b ratio, the initial setting time of VA-based concrete is significantly reduced.

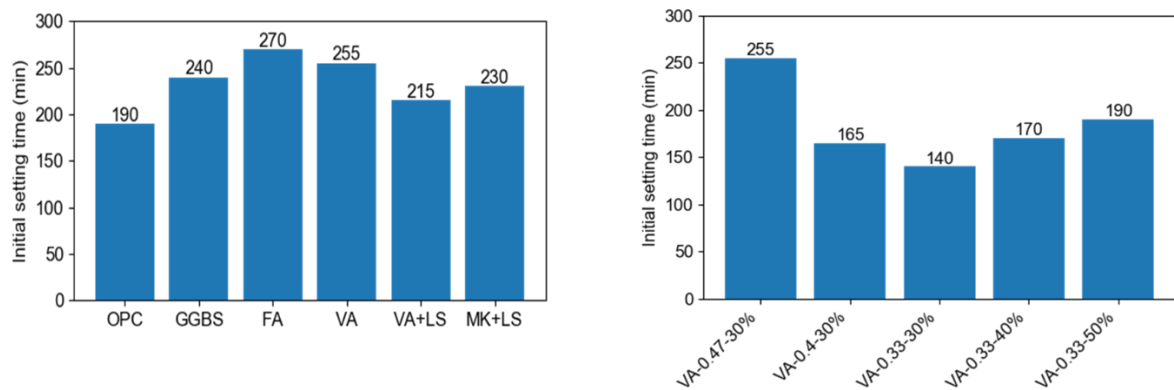


Fig. 1. Initial setting time of cement paste.

3. Final setting time of cementitious paste

The final setting time of cementitious paste is shown in Fig. 2. Similar to initial setting, the final setting time of VA30-0.47 is slightly higher than control, but comparable to other SCMs (even lower than PFA and GGBS-based concretes). With reducing w/b ratio, the final setting time of VA-based concrete is significantly reduced.

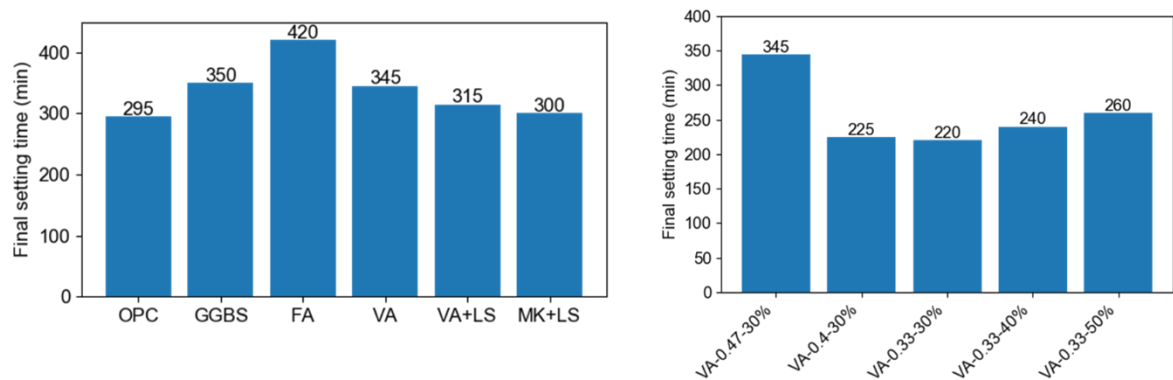


Fig. 2. Final setting time of cement paste.

4. Compressive strength development

The comparative compressive strengths at 3 days, 28 days and 56 days of the design concrete mixes is given in Fig. 3. It can be seen that lowest strength is found for w/b ratio 0.47 for the replacement of 30% of OPC by VA. For example, about 40% lower 28 days strength is found for VA30-0.47 compared to control one. However, considering the use of lower w/b ratio, the strengths at all experimental days are significantly increased. For instance, VA30-0.40 is associated with only 22% lower 28 days strength compared to control (which is almost half than those for w/b 0.47). Almost similar 28 days strength was for VA30-0.33 compared to control (only 4% lower). Even, comparable strengths were found for very high replacement ratios (e.g. 40% and 50%). About 68.7 MPa and 64.0 MPa compressive strength at 28 days was found for replacing 40% and 50% OPC by VA with the use of 0.33 w/b ratio, which are only 9% and 15% lower than the control. The results demonstrated that high volume of VA can be used for the desired strength with the adjustment of w/b ratio, but would be very effective for substituting 30-40%.

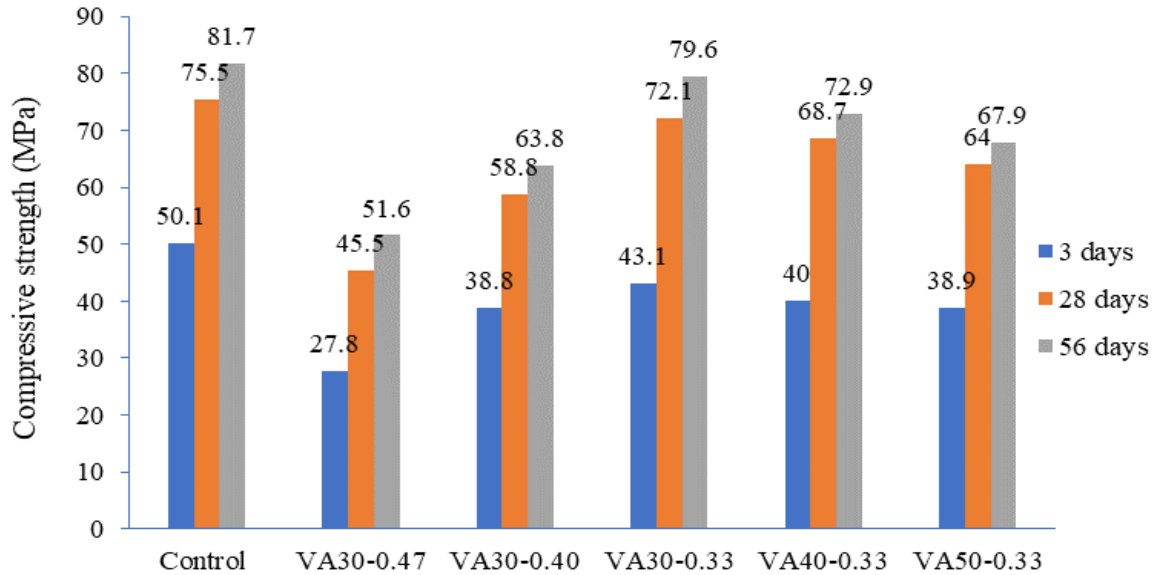


Fig. 3. Comparative compressive strength of concrete.

5. Comparative carbon emission

The comparative carbon emission of the design concrete mixes is shown in Fig. 4. It can be seen that lower carbon emission was found for using 30% VA with the w/b ratio of 0.47, which is 25% lower than the OPC concrete. However, the emission for different replacement rates (e.g. for 30% with w/b 0.40, 30% with w/b 0.33, and 40% with w/b 0.33) were increased mainly for using higher amount of superplasticizer. Though the emission was increased, it is still significantly lower than that of OPC concrete, for instance, 19%, 10%, and 20%, respectively. The lowest carbon emission was found for replacing 50% OPC by VA, and it is about 29%. The saving is mainly due to replacing higher amount of highly emitted OPC, and it could even higher as the considerable saving was off set by high doses of admixture. Overall, the benefits of using higher amount of VA substitution is diminished due to the use of higher amount of superplasticizer.

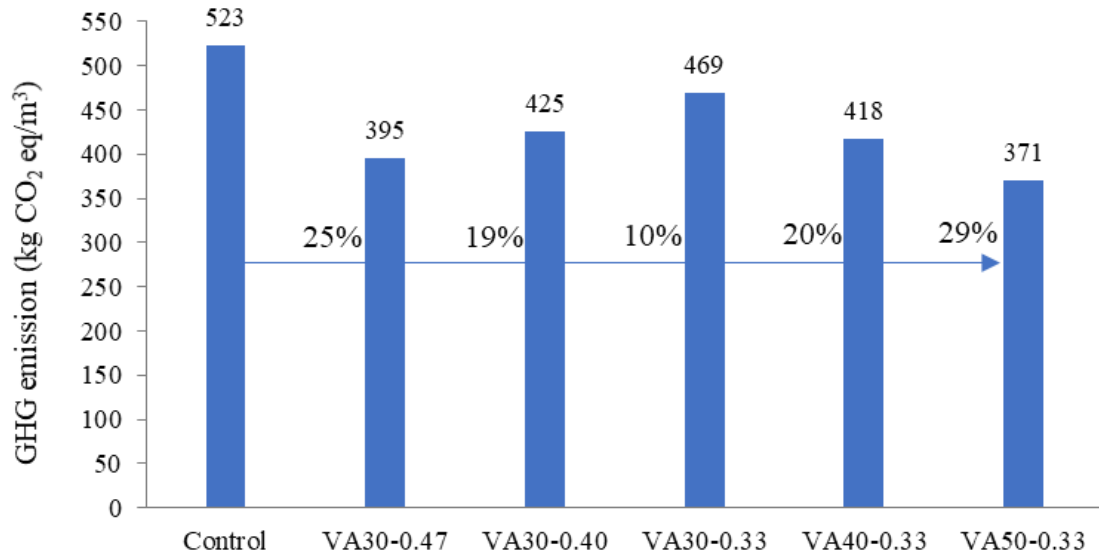


Fig. 4. Comparative carbon emission of the designed concrete.

6. Comparative costs of materials

The comparative costs for the materials of the design concrete mixes is shown in Fig. 5. It can be seen that about 9% lower costs was found for using 30% VA with the w/b ratio of 0.47, which is almost similar for 30% VA with w/b 0.40 to the OPC concrete. The highest cost was found for the same replacement ratio with w/b ratio 0.33 due to the use of higher amount of total binders and the higher amount of superplasticizer. The cost is about 12% higher than the control one. Although the higher replacement ratios (e.g. 40% and 50%) with the w/b ratio 0.33 reduces about 2% of the total cost compared to VA30-0.33 one, such high replacements do not necessarily mean to reduce the costs compared to the control one. About 9% higher cost is associated with VA40-0.33 and VA50-0.33 concrete compared to the normal concrete, because the use of very high amount of admixture.

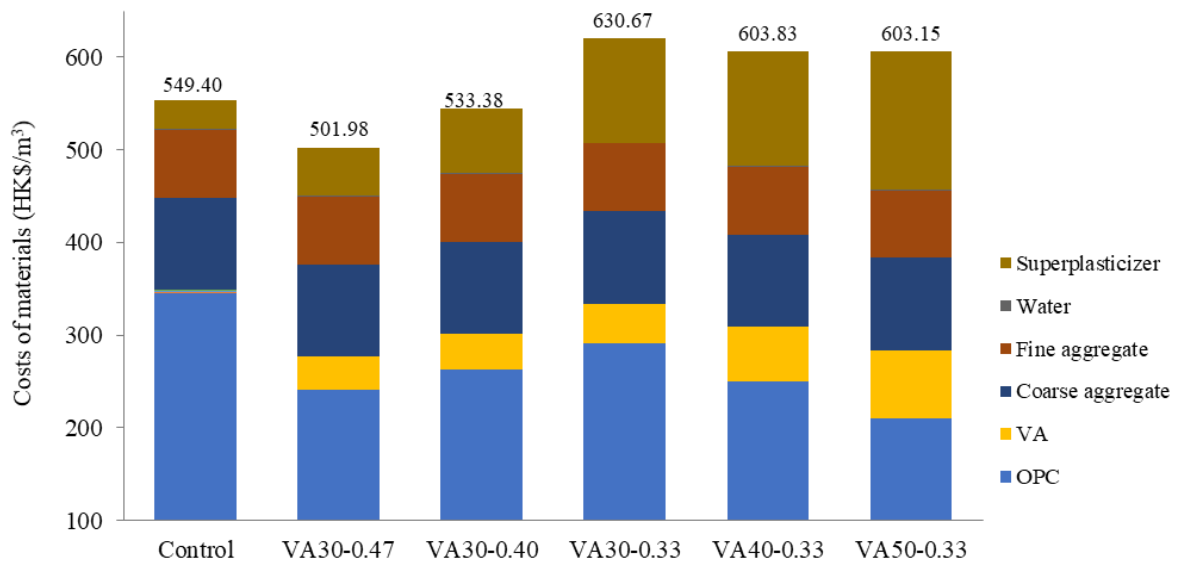


Fig. 5. Comparative costs analysis of the designed concrete.

7. Summary

- High volume of VA can be used for the desired strength with the adjustment of w/b ratio, but would be very effective for substituting 30-40% OPC by VA.
- Up to 29% carbon emission was associated with VA-based concretes for replacing 30-50% OPC by VA, compared to the control one.
- Both positive and negative cost implications were found for using different w/b ratios and replacement levels. The costs are slightly lower for replacing 30% OPC by VA with w/b ratios 0.47 and 0.40 than the normal concrete (with w/b 0.45). However, the cost is significantly (about 12%) increased for the same replacement level with w/b 0.33. For higher replacement ratios (40% and 50%), the materials costs are only 9% higher than the control one.
- Overall results in terms of mechanical, environmental and cost performance, it can be concluded that VA can be effectively used as a substitute of OPC by a range between 30-50% in the concrete, depending on the targeted concrete in terms of strength, costs and carbon emission.

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