

# THE DEVELOPMENT OF MIXED RECYCLED AGGREGATE POWDER BASED GEOPOLYMER WITH THE ADDITION OF METAKAOLIN

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*In this paper, the feasibility of making geopolymers with mixed recycled aggregate powder (MRAP) was investigated. Metakaolin (MK) was introduced into the matrix to improve the chemical activity of MRAP. The influence of MK replacement ratio and alkali concentration were investigated in the experiments. With proper addition of MK, the compressive strength of MRAP based geopolymer can be as high as 36.7 MPa. It was also found that the compressive strength of MRAP based geopolymer increased with the increase of MK replacement ratio when the alkali concentration was higher than 10 mol/L. By contrast, the MK replacement ratio was recommended to be lower than 0.3 when the alkali concentration was lower than 8 mol/L. Meanwhile, the correlation between flexural and compressive strength of MRAP based geopolymer was also proposed in this paper.*

**Keywords:** Alkali; Metakaolin; Waste treatment; Compressive strength

## 1. Introduction

Construction and demolition waste (CDW) is one of the heaviest and most voluminous waste streams generated in the EU, including concrete, bricks, gypsum, wood, glass, metals, plastic, solvents, asbestos and excavated soil. CDW arises from activities such as the construction of buildings and civil infrastructure, total or partial demolition of buildings and civil infrastructure, road pavement and maintenance. While many of these materials are not necessarily dangerous to handle, they can impact the environment and water if improperly disposed.

Most of CDW materials, e.g., concrete, bricks and glass, are rich in aluminosilicate, thus it may be feasible to make geopolymer by using CDW. Lampris et.al [1] investigated the feasibility of making geopolymer with silt filter cake. The results showed that the compressive strength of silt filter cake based geopolymer could be as high as 39.7 MPa after being cured with a temperature of 105°C for 24 h. Sun et.al [2] used waste ceramic to make geopolymers and the 28-day compressive strength of the synthesized geopolymer pastes could be 71.1 MPa. Robayo-Salazar [3] tried to make geopolymers with clay brick waste, glass waste and concrete waste, it was found that the compressive strength of geopolymer specimens could be as high as 57 MPa. Some other types of CDW materials have also been applied to make geopolymers by other researchers [4-7].

Generally, it was found that the chemical

activity of the CDW or CDW powder is relatively low due to its semi-crystalline property. In order to increase the early compressive strength of geopolymer made with CDW powder, the high temperature curing method with the temperature ranging from 70° to 120° was commonly provided. However, high temperature curing may be inconvenient for the practical application of geopolymer specimens. Additionally, the environmental friendliness of geopolymer is also decreased since high temperature curing method is also accompanied with high consumption of energy and emission of carbon dioxide. Turner et.al [8] compared the carbon footprint of geopolymer and conventional concrete considering their whole life stages from manufacturing to application and demolition. Even though geopolymer have long been considered as energy-saving and eco-friendly constructional material, it was found that the carbon emission of geopolymer is only about 9% lower than that of conventional concrete. One of the reasons is that the high temperature curing method for geopolymer will also consume large amount of energy with the emissions of CO<sub>2</sub>.

In order to avoid high temperature curing, cement or high-calcium slag was normally applied to increase the chemical activity of CDW powder with ambient curing method. Vásquez [9] found that the addition of Portland cement would increase the compressive strength of geopolymer made with CDW powder. Hwang [10] found that the compressive strength of waste brick powder based geopolymer could be as high as 93 MPa with the

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addition of high-calcium slag. However, it was also noticed that the flowability and workability of CDW based geopolymer decreased significantly with the addition of slag and cement [11]. Meanwhile, even though the addition of slag or cement could effectively increase the early strength of CDW based geopolymer, the long-term durability is still questionable since the reaction process is too rapid and the micro-structure may not be dense enough[12-14].

This paper was intended to manufacture mixed recycled aggregate powder (MRAP) based geopolymer with ambient curing method. As a typical CDW, it was found that the chemical activity of MRAP is relatively low. Thus, different dosage of metakaolin (MK) was added in the solid precursor to increase the content of silicon-aluminum material. The influences of MK replacement ratio and alkali concentration on the compressive strength of MRAP based geopolymer were investigated. The relationship between compressive strength and flexural strength was also investigated based on the experimental data.

## 2. Materials and methods

The mixed recycled aggregate used in this study was provided by CDW recycling company in Belgium, which contains red bricks, waste concrete and ceramics particles, as shown in Fig. 1. The received mixed aggregate was firstly washed before natural drying. Then, the mixed recycled aggregates was milled into powder with an electric milling machine (48-D0500/G, Controls Group) for four hours. The as received MRAP is shown in Fig.2(a) and the microtopography of MRAP is shown in Fig.3(a), which contains numerous plate-like crystals with a length about 30  $\mu\text{m}$ . The chemical composition of MRAP is shown in Table 1, it can be seen that the total content of silicoaluminates material is only 53%, indicating the chemical activity of MRAP is relatively low[15]. In order to increase the content of silicoaluminates, the metakaolin (MK) was added in the solid precursor. The macro and micro morphology of MK are shown in Fig.2(b) and Fig.3(b), respectively. It can be seen that MK was mainly composed with numerous needle-like crystals with a length about 5  $\mu\text{m}$ . The particle size distribution curves for both MK and MRAP are shown in Fig.4. It can be seen that the MK particles were mainly distributed in the range of 0.5  $\mu\text{m}$  to 10  $\mu\text{m}$ , while MRAP particles were mainly distributed in the range of 5  $\mu\text{m}$  to 100  $\mu\text{m}$ , once again demonstrate that the MK has a much higher fineness and specific surface area than MRAP. Also, as can be seen in Table 1, the total content of silicoaluminates material for MK is as high as 93.9%, thus it is expected that the addition of MK is effective to increase the early strength of MRAP based geopolymer with ambient curing method.

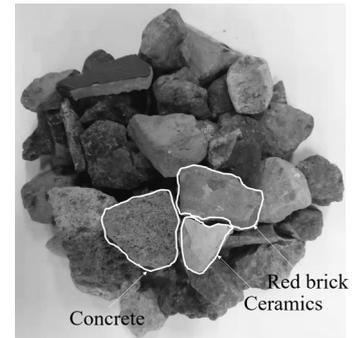


Fig.1 Typical mixed aggregate.

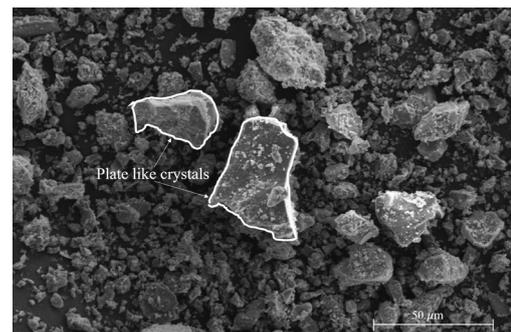


(a) MRAP

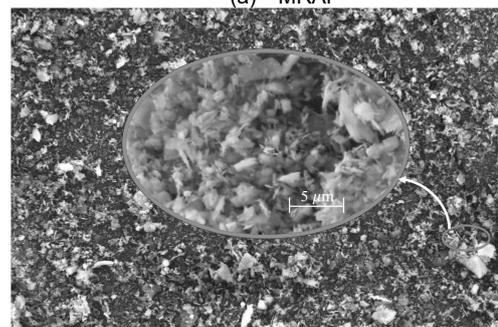


(b) Metakaolin (MK)

Fig.2 The raw materials.



(a) MRAP



(b) Metakaolin (MK)

Fig.3 - The microtopography of raw materials.

Table 1

Chemical composition of raw materials(Wt.%)												
Oxide	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO	Fe <sub>2</sub> O <sub>3</sub>	MgO	Na <sub>2</sub> O	TiO <sub>2</sub>	MnO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>	SO <sub>3</sub>	K <sub>2</sub> O	
MRAP	37.8	15.2	12.5	13.7	9.7	0.9	0.7	0.1	0.2	5.7	3.1	
MK	61.4	32.5	0.2	0.8	2.1	0.7	0.2	0.2	0.1	0.2	0.8	

The cube and prism specimens were designed for the compression and three-point bending test, respectively. Thirty series of geopolymer specimens were designed which is listed in Table 2. As the nomenclature of each specimen in Table 2, the suffix number was designed to indicate the alkali concentration and MK replacement ratio. For example, the first number “4” in specimen G-4-0.3 indicates the alkali concentration was 4 mol/L and the second number “0.3” indicates the MK replacement ratio was 30%. The alkali activator was made with KOH

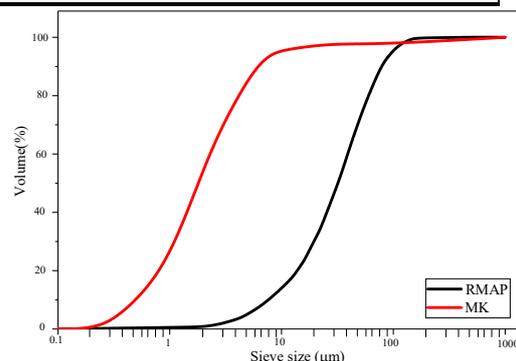


Fig.4 - Particle size distribution curves of raw materials.

Table 2

Geopolymer specimens					
Series	Specimen	Compressive strength (MPa)	Standard deviation	Flexural strength (MPa)	Standard deviation
A	G-4-0.0	4.55	0.81	2.01	0.01
	G-4-0.1	11.46	0.38	3.43	0.42
	G-4-0.2	12.24	0.45	3.39	0.04
	G-4-0.3	14.35	0.73	3.68	0.07
	G-4-0.4	9.47	1.29	3.23	0.03
B	G-6-0.0	5.10	0.77	2.55	0.03
	G-6-0.1	13.97	0.68	3.75	0.01
	G-6-0.2	16.09	1.51	3.71	0.23
	G-6-0.3	19.14	1.87	3.90	0.08
	G-6-0.4	16.34	0.17	3.78	0.07
C	G-8-0.0	15.14	1.91	4.06	0.01
	G-8-0.1	6.09	0.13	2.31	0.02
	G-8-0.2	15.14	0.79	3.70	0.09
	G-8-0.3	20.92	1.21	4.17	0.08
	G-8-0.4	22.05	1.19	4.93	0.03
D	G-10-0.0	21.97	2.16	4.35	0.12
	G-10-0.1	21.11	0.90	4.18	0.18
	G-10-0.2	6.72	0.23	2.38	0.36
	G-10-0.3	19.12	1.02	3.99	0.54
	G-10-0.4	24.95	1.84	5.30	0.13
E	G-12-0.0	26.72	2.45	5.19	0.08
	G-12-0.1	28.92	1.58	5.55	0.13
	G-12-0.2	31.59	2.49	5.21	0.13
	G-12-0.3	7.43	0.31	2.51	0.17
	G-12-0.4	22.36	1.40	4.39	0.09
	G-12-0.5	28.51	1.44	5.30	0.14
	G-12-0.6	30.43	1.62	5.18	0.18
	G-12-0.7	34.04	1.00	6.05	0.13
	G-12-0.8	36.78	1.58	6.12	0.14
	G-12-0.9				

(98% purity, density 2.1g/cm<sup>3</sup> ) and Na<sub>2</sub>SiO<sub>3</sub> solution (39.8 wt.% Na<sub>2</sub>SiO<sub>3</sub>, relative density 1.42g/cm<sup>3</sup> ). The weighted KOH flake was dissolved in pure water and then mixed with Na<sub>2</sub>SiO<sub>3</sub> solution with a cooling process for 24 hours. Before pouring alkali activator to the solid precursor, the solid precursor was mixed with an electric blender for 5 minutes. After the alkali activator was added in the solid precursor, the electric blender was mixed for another 10 minutes to get homogeneous geopolymer mixture. The mixture was then casted in the mould and covered with plastic film. The specimens were cured in

ambient environment (20±5°C with 60% humidity) for 24 hours before demolding. After demolding, the specimens were cured in ambient environment for another 6 days before testing. A semi-automatic concrete testing machine (MATEST, Italy) was applied to get the compressive and flexural strength of geopolymer specimens, which is shown in Fig.5. Three cubic specimens and three prism specimens were casted and tested for each geopolymer mix design. The average compressive and flexural strength of geopolymers are also shown in Table.2.

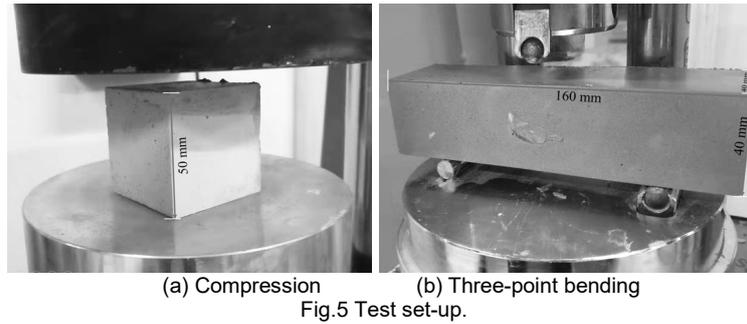


Fig.5 Test set-up.

### 3.Results

#### 3.1 The influences of MK replacement ratio

The MK replacement ratio was defined as the mass ratio between MK and solid precursor including MK and MRAP. The MK replacement ratio was increased from 0.0 to 0.5 in this paper and the influence of MK replacement ratio on the 7-day compressive strength of geopolymer is shown in Fig.6.

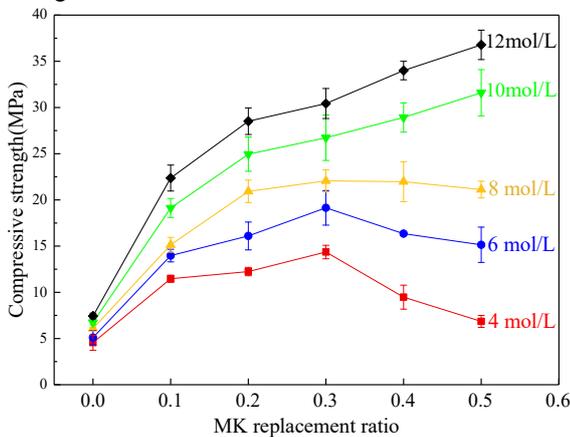
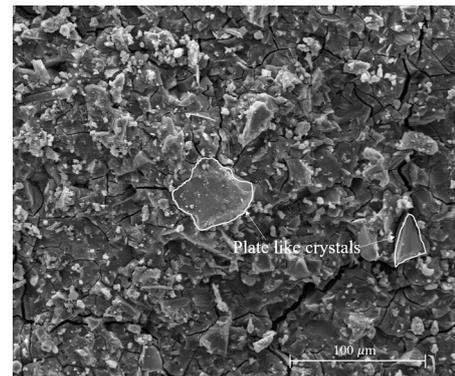


Fig.6 - The influences of MK replacement ratio.

When the alkali concentration was 4 mol/L, which was plotted as the red line in Fig.6, the 7-day compressive strength for pure MRAP based geopolymer (G-4-0.0) was only 4.55 MPa. It is attributed to the reason that the chemical activity of MRAP was relatively low and the silicoaluminates particles cannot be dissolved when the alkali concentration was only 4 mol/L, thus less geopolymer gel was formed in the matrix. The typical failure mode and microtopography of specimen G-4-0.0 is shown in Fig.7. An obvious splitting failure mode with numerous unreacted particles were noticed for specimen G-4-0.0, as shown in Fig.7(a). The typical microtopography of specimen G-4-0.0 is shown in Fig.7(b), the plate-like crystals with a length over 50  $\mu\text{m}$  were still visible, indicating that the chemical activity of MRAP was relatively low and the geopolymerization process was not sufficient. As the MK replacement ratio increased from 0.0 to 0.3, the compressive strength increased gradually even though the alkali concentration was only 4 mol/L. However, the compressive strength decreased with the further increase of MK



(a) Failure mode

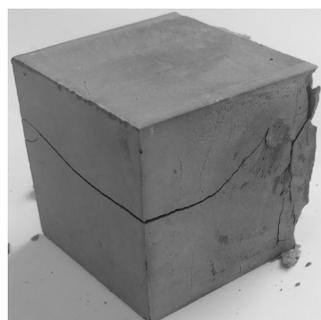


(b) Microtopography

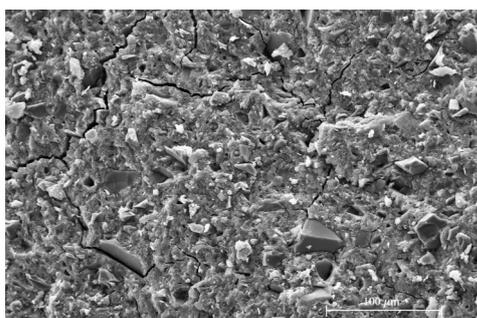
Fig.7 - Specimen G-4-0.0.

replacement ratio. For example, the compressive strength for specimen G-4-0.5 is much lower than G-4-0.3. The similar phenomenon was noticed when the alkali concentration was 6 mol/L or 8 mol/L. By contrast, when the alkali concentration was 10 mol/L or 12 mol/L, the compressive strength increased sustainably with the increase of MK replacement ratio. The 7-day compressive strength for specimen G-12-0.5 was 36.7 MPa, which was the highest compressive strength achieved in this test. The typical failure mode and microtopography of specimen G-12-0.5 are shown in Fig.8. It can be seen that the specimen G-12-0.5 was more brittle under compressive load and the microstructure was much denser. Even though there are still some unreacted crystal in the matrix, but the size seems to be much smaller.

It can be concluded that the increase of MK replacement ratio was not necessarily beneficial



(a) Failure mode



(b) Microtopography

Fig.8 - Specimen G-12-0.5.

for the compressive strength of MRAP based geopolymer, especially when the alkali concentration was relatively low. In this paper, the MK replacement ratio should be controlled within 0.3 when the alkali concentration was lower than 8 mol/L. It may be attributed to the reason that MK has a much higher fineness and specific surface area than MRAP, thus MK would react with alkali activator firstly with the consumption of OH<sup>-</sup>. When the alkali concentration was relatively low and almost all OH<sup>-</sup> have been consumed by MK, the remaining MRAP particles would have a negative effect on the compressive strength of geopolymer specimen. By contrast, when the alkali concentration was higher than 8 mol/L, the alkali activator could provide sufficient OH<sup>-</sup> to react with both MK and MRAP thus the compressive strength would be increased as the MK replacement ratio increased from 0.0 to 0.5.

### 3.2 The influences of alkali concentration

The influence of alkali concentration on the compressive strength of MRAP based geopolymer is shown in Fig.9.

For all geopolymer specimens with different MK replacement ratio, the compressive strength increased gradually with the increase of alkali concentration, which is consistent with the conclusions made by other researchers [16,17]. As the alkali concentration increased, the aluminum and silicon atoms would be more easily dissolved from the solid precursor, thus more geopolymer gel was formed and the micro-structure would be much denser. It was also noticed that when the MK

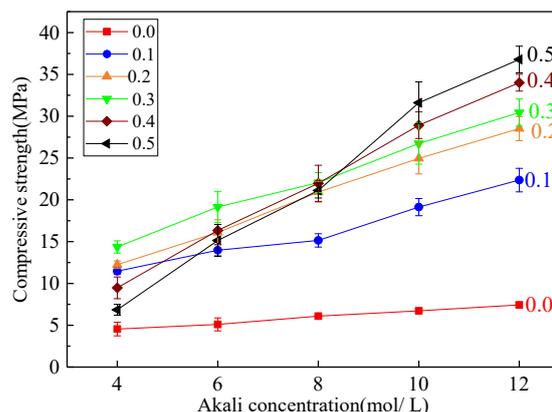


Fig.9 – The influences of alkali concentration.

replacement ratio was relatively low, the alkali concentration has limited influence on the compressive strength of geopolymer specimen. For example, the compressive strength for G-12-0.5 is 5.36 times higher than G-4-0.5, while G-12-0.0 is only 1.63 times higher than G-4-0.0. It is also due to the low chemical activity of MRAP and the single increase of alkali concentration may be less effective if the matrix was in lack of active silicoaluminate material. With the increase of MK replacement ratio, the compressive strength increased rapidly with the increase of alkali concentration since the matrix could provide enough active silicoaluminate by MK.

### 3.3 The relationship between flexural and compressive strength

In this paper, the three-point bending test was also conducted for geopolymer prism specimens. It was found that the MK replacement ratio and alkali concentration have similar influences on the flexural strength of geopolymer specimens. It is reasonable since the flexural stress can be decomposed into compressive and tensile stress.

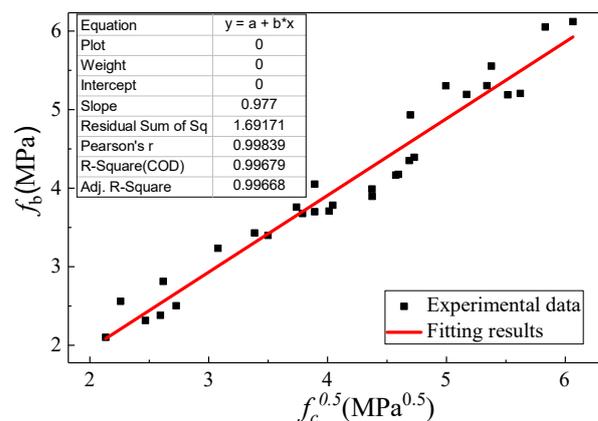


Fig.10 - The fitted curve for the flexural strength.

For concrete specimens, it has been reported that the flexural strength is about 15% of its compressive strength. The equations between flexural strength and compressive strength are

normally expressed as  $f_b = mf_c^{0.5}$  [18-21], where  $f_b$  and  $f_c$  are the flexural and compressive strength,  $m$  is the reduction factor derived from regression analysis based on experimental data.

The similar method was applied to investigate the correlation of flexural and compressive for MRAP based geopolymer, which is shown in Fig.10. According to the regression analysis, the flexural strength of geopolymer can be expressed as  $f_b = 0.977f_c^{0.5}$ .

#### 4. Conclusions

In this paper, the feasibility of making geopolymers with mixed recycled aggregate powder (MRAP) was investigated. The main conclusions are listed as follows.

1. It is feasible to make MRAP based geopolymer with the addition of MK, the average compressive strength of specimen G-12-0.5 can be as high as 36.7 MPa.

2. It was found that the compressive strength of geopolymer increased with the increase of MK replacement ratio when the alkali concentration was 10 mol/L or 12 mol/L. By contrast, the MK replacement ratio was recommended to be lower than 0.3 when the alkali concentration was lower than 8 mol/L.

3. Based on the experimental data, the correlation between flexural and compressive strength of MRAP based geopolymer was expressed as  $f_b = 0.977f_c^{0.5}$  in this paper.

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#### REFERENCES

- [1] Lampris C, Lupo R, Cheeseman C R. Geopolymerisation of silt generated from construction and demolition waste washing plants. *Waste Management*, 2009, **29**(1), 368-373.
- [2] Sun Z, Cui H, An H, et al. Synthesis and thermal behavior of geopolymer-type material from waste ceramic. *Construction and Building Materials*, 2013, **49**, 281-287.
- [3] Robayo-Salazar R A, Rivera J F, de Gutiérrez R M. Alkali-activated building materials made with recycled construction and demolition wastes. *Construction and Building Materials*, 2017, **149**, 130-138.
- [4] Al-Saadi T H A, Mahdi Z H, Abdullah I T. Foaming geopolymers preparation by alkali activation of glass waste. *Revista Romana de Materiale*, 2019, **49**(3), 352-360.
- [5] Panizza M, Natali M, Garbin E, et al. Assessment of geopolymers with Construction and Demolition Waste (CDW) aggregates as a building material. *Construction and Building Materials*, 2018, **181**, 119-133.
- [6] Komnitsas K, Zaharaki D, Vlachou A, et al. Effect of synthesis parameters on the quality of construction and demolition wastes (CDW) geopolymers. *Advanced Powder Technology*, 2015, **26**(2), 368-376.
- [7] Rakhimova N R, Rakhimov R Z. Alkali-activated cements and mortars based on blast furnace slag and red clay brick waste. *Materials & Design*, 2015, **85**, 324-331.
- [8] Turner L K, Collins F G. Carbon dioxide equivalent (CO<sub>2</sub>-e) emissions: A comparison between geopolymer and OPC cement concrete. *Construction and Building Materials*, 2013, **43**, 125-130.
- [9] Vásquez A, Cárdenas V, Robayo R A, et al. Geopolymer based on concrete demolition waste. *Advanced Powder Technology*, 2016, **27**(4), 1173-1179.
- [10] Hwang C L, Yehualaw M D, Vo D H, et al. Performance evaluation of alkali activated mortar containing high volume of waste brick powder blended with ground granulated blast furnace slag cured at ambient temperature. *Construction and Building Materials*, 2019, **223**, 657-667.
- [11] Suwan T, Fan M. Influence of OPC replacement and manufacturing procedures on the properties of self-cured geopolymer. *Construction and Building Materials*, 2014, **73**, 551-561.
- [12] Chindaprasit P, Phoo-ngernkham T, Hanjitsuwan S, et al. Effect of calcium-rich compounds on setting time and strength development of alkali-activated fly ash cured at ambient temperature. *Case Studies in Construction Materials*, 2018, **9**, 187-198.
- [13] Lavanya G, Jegan J. Durability study on high calcium fly ash based geopolymer concrete. *Advances in Materials Science and Engineering*, 2015, (2015), 1-7
- [14] Wardhono A, Gunasekara C, Law D W, et al. Comparison of long term performance between alkali activated slag and fly ash geopolymer concretes. *Construction and Building materials*, 2017, **143**, 272-279.
- [15] De Silva P, Sagoe-Crenstil K, Sirivivatnanon V. Kinetics of geopolymerization: role of Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub>. *Cement and Concrete Research*, 2007, **37**(4), 512-518.
- [16] Nath S K, Kumar S. Role of alkali concentration on reaction kinetics of fly ash geopolymerization[J]. *Journal of Non-Crystalline Solids*, 2019, **505**, 241-251.
- [17] Wang W C, Wang H Y, Tsai H C. Study on engineering properties of alkali-activated ladle furnace slag geopolymer. *Construction and Building Materials*, 2016, **123**, 800-805.
- [18] ACI Committee 318 (2005) Building Code Requirements for Structural Concrete (ACI 318-05) and Commentary (318R-05), p.430, ACI, Farmington Hills, MI.
- [19] ACI Committee 363 (1992) Report on High-Strength Concrete (ACI 363R-92), p.56, American Concrete Institute, Farmington Hills, MI.
- [20] Standards Association of Australia (2001) Concrete Structures – AS 3600, Standards Australia, Strathfield (NSW).
- [21] CEB-FIB, Model code 90 (1993) Final draft, Lausanne.; Bulletin 203, CH, Switzerland.

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