Journal of Applied Research and Technology

www.jart.icat.unam.mx

Journal of Applied Research and Technology 22 (2024) 189-196

Original

Fly ash and ceramic residue-based geopolymer mortars

S. A. Muttaqin[a] • S. Subari[b] • B. D. Erlangga[c]* • H. Aliyah[a]

[a]*Department of Physics, Faculty of Science and Technology, UIN Sunan Gunung Djati, Bandung 40614, Indonesia*
[b]*Research Center for Advanced Materials, National Research and Innovation Agency (BRIN), Tangerang Selatan 15314, Indonesia*
[c]*Research Center for Geological Resources, National Research and Innovation Agency (BRIN), Bandung 40135, Indonesia*

Received 05 11 2023; accepted 12 05 2023
Available 04 30 2024

Abstract: A binary geopolymer based on fly ash and ceramic residue was developed under ambient water curing. The alkaline medium comprises commercial Na_2SiO_3 and NaOH with concentration of 10 M and 12 M. Analyses were carried out to characterize the raw materials and their geopolymer properties. Characterization of materials was conducted through x-ray diffraction (XRD), x-ray fluorescence (XRF) and scanning electron microscope (SEM). The compressive strength, porosity and abrasion rates were measured to determine geopolymer performance. The ceramic residue consists mainly of SiO_2 and Al_2O_3, which is similar to fly ash. Among the specimens, the highest compressive strength produced from the specimens was 25.2 MPa at the age of 28 days. This highest strength was followed by the lowest water absorption and abrasion rate, which are 1.99% and 0.007 mm/min, respectively. The microstructure showed geopolymer matrix with some unreacted particles from ceramic residue powder. There were no signs of efflorescence observed on the geopolymer specimens. Each mix demonstrated a slight increment in density and reduced the porosity during the curing period.

Keywords: geopolymer, fly ash, ceramic residue, water curing

*Corresponding author.
E-mail address: bagu010@brin.go.id (B. D. Erlangga).
Peer Review under the responsibility of Universidad Nacional Autónoma de México.

1. Introduction

Concrete is one of the most used materials on Earth. The reliance on ordinary Portland cement (OPC) in producing concrete materials is prominent. However, the production of OPC cement generated a significant amount of CO_2 into the atmosphere (Barcelo et al., 2014), which accelerated climate change issues to the environment. This has drawn the attention of researchers to develop such sustainable building materials. In the late 1970s, Joseph Davidovits firstly coined the term "geopolymer" (Davidovits, 1989), which can act as a geological glue or binder for producing new materials. This geopolymer system can completely replace cement binder in producing mortars or concretes.

The development of the geopolymer materials derived from the reaction between solid materials rich in alumina-silicate and alkaline solution, yielding Si-O-Al bonds (Davidovits, 1991). This alumina-silicate content can be found in many types of minerals, such as metakaolin. The utilization of waste materials for the source of alumina silicate in geopolymer encourages more sustainable development, but the performance of the geopolymer materials should be thoroughly addressed (Mohajerani et al., 2019). Those waste materials used in geopolymer include fly ash (Hardjito et al., 2005), foundry slags (Shi & Day, 1999) and other building demolitions (Vásquez et al., 2016).

Fly ash-based geopolymer has been progressively developed since it is a by-product from coal combustion process, which is still the main source for generating electricity globally. Therefore, the amount of fly ash is abundant near the power plant areas. However, fly ash showed slow gaining strength in geopolymer at ambient temperature due to a lack of calcium (Ekaputri et al., 2015) and the presence of crystalline, chemically inert phases (Luhar & Luhar, 2022). Ground granulated blast furnace slag (GGBFS) has been used to accelerate the early strength of such geopolymer, providing calcium silicate hydrate (C-S-H) and (A-S-H) at the early stage (Kumar et al., 2010; Shi & Day, 1999) or calcium alumina silicate hydrate (C-A-S-H) at the later stage formation (Puligilla & Mondal, 2013).

A similar residue, rich in alumina silicate, was also generated by ceramic industries. This ceramic waste usually ended up not being used in a landfill or a dumping area. A previous study showed ceramic powder waste (CPW) has pozzolanic properties, which are suitable for concrete material, maintaining strength and durability (Samadi et al., 2015). Another research has effectively utilized ceramic waste for fine aggregates, replacing conventional sand in concrete mortars (Pacheco-Torgal & Jalali, 2010).

Mahmoodi et al. thoroughly reported geopolymer using ceramic tile waste with other supplementary cementitious materials (SCM) (Mahmoodi et al., 2020). The study revealed that the optimum compressive strength at ambient temperature curing was the combination of ceramic waste 45% and fly ash 55%, resulting in around 40 and 60 MPa for age 7 and 28 days, respectively. Wardhono et al. experimented with ambient water curing for fly ash and GGBFS mixture, obtaining optimum compressive strength at 62 MPa with the same percentage of both precursors (Wardhono et al., 2015). The geopolymer mortar based on fly ash and rice husk ash presented slightly higher compressive strength for water immersion than the room temperature treatment method (Rangan et al., 2020). Water-submerged curing was also found to be an effective means of preventing efflorescence in geopolymer specimens (Simão et al., 2021).

In this study, ceramic waste residue and fly ash were used for geopolymer mortar and cured in water until the designated age for testing. The larger proportion of ceramic waste in this binary geopolymer system was applied to complement the previous research gap.

2. Materials and method

2.1. Materials

Fly ash was derived from a coal combustion power plant in Banten province. The ceramic residue was collected from the Center for Ceramics in Bandung, which is in the form of debris from testing ceramic materials. The ceramic waste was firstly ground into powder prior to the mixing process. The photographs of those solid ingredients can be seen in Figure 1. Meanwhile, the alkaline solution medium is a combination between sodium hydroxide (NaOH) and sodium silicate Na_2SiO_3. The NaOH pellets and fresh water were used to prepare alkaline liquid of 10 M and 12 M, while sodium silicate granules were then added to the solution.

Figure 1. A photograph of fly ash (a), ceramic debris (b), and ceramic waste powder (c).

2.2. Method

The research design was carried out as shown in Table 1 All three mixes, denoted as code A, B and C, used sodium solution with concentrations 10 M and 12 M, each having a ratio with sodium silicate of 1:2 and 1:1 alkali activator. As a result, the variation of the binder (alkaline medium and fly ash) with ceramic waste was 1:1. This formulation was used for the high ceramic waste powder (CPW) incorporation in fly ash-based geopolymer, ranging between 60-71% CPW. Meanwhile, this also resulted in different liquid and solid ratios ranging from 0.20 to 0.43.

The geopolymer mixtures were manually cast in cube molds with the size of 50x50x50 mm^3. Then, the specimens were left at ambient condition for 4 h prior to demolding. The specimens were kept in water until the time for testing at 7, 14 and 28 days. The mechanical performance of the specimens was measured by compressive strength, water absorption and abrasion rate. A scanning electron microscope (SEM) was used to analyze the microstructure of the best-performance sample.

3. Results and discussion

3.1. Materials characterization

The ceramic waste used in this study was firstly ground to obtain powder particle size. The ceramic waste powder was then measured for particle size distribution, as shown in Figure 2. The process of grinding seized the particle size ranging from 8 mesh to 200 mesh, which can be categorized as fine aggregate based on ASTM C-125-07 (America society for testing and material (ASTM) C125, 2007). Most of the particle size was at 70 mesh, accounted for approximately 30% of the total.

Table 1. Mix design of geopolymer mortars.

Mix codes	Molarity	NaOH solution (g)	Na_2SiO_3 (g)	FA (g)	Binder (g)	CWP (g)	FA/CWP	Liquid/ solid
A	10 M	100	200	200	500	500	29:71	0.43
	12 M	100	100	200	400	400	33:67	0.33
B	10 M	100	200	300	600	600	33:67	0.33
	12 M	100	100	300	500	500	38:62	0.25
C	10 M	100	200	400	700	700	36:64	0.27
	12 M	100	100	400	600	600	40:60	0.20

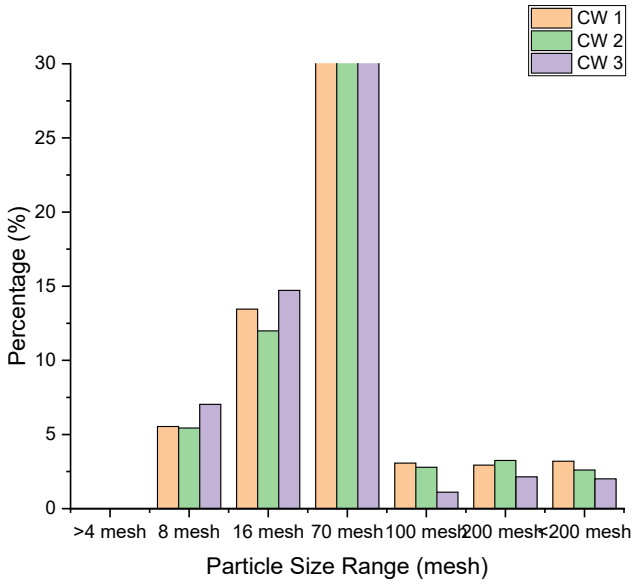

Figure 2. Particle size distribution of ceramic waste powder (CWP).

The oxides content of fly ash and ceramic waste powder can be seen in Table 2. Fly ash in this study comprises mainly SiO_2, Al_2O_3, CaO and Fe_2O_3. The significant amount of SiO_2 and Al_2O_3 in the fly ash makes it widely used in the development of geopolymer (Amran et al., 2021). Meanwhile, the ceramic waste has relatively the same proportion of SiO_2 and Al_2O_3, with significant magnesium oxide. Taking into account this chemical composition, the ceramic residue may have the same potential as the alumina silicate in fly ash for manufacturing geopolymer binder.

The x-ray diffraction analysis showed that the fly ash has mullite, quartz and sillimanite phases (Figure 3). Mullite appears with an orthorhombic crystal structure (quartz with a hexagonal crystal structure) and sillimanite with an orthorhombic crystal structure. It can be seen that the maximum peaks in each phase of mullite, quartz, and sillimanite are at 26.26°, 26.63° and 26.67°, sequentially. The mullite phase is a transformation mineral consisting of SiO_2 and Al_2O_3 under high-temperature exposure. However, mullite phase was also found in as-received coal fly ash in companion with quartz, corundum, and glassy phases (Ma et al., 2019). This may be due to the high temperature in the coal combustion proses, which generated the fly ash.

Table 2. Oxides content of fly ash and ceramic waste obtained by XRF.

Oxides	Conc. (%)	
	Fly ash	Ceramic residue
SiO_2	44.97	46.26
Al_2O_3	20.77	40.16
CaO	13.43	0.794
Fe_2O_3	10.1	1.64
MgO	3.91	8.86
Na_2O	1.479	0.348
SO_3	0.912	0.022
TiO_2	0.997	0.515
K_2O	0.651	0.85
P_2O_5	0.155	0.050
MnO	0.187	0.024
LOI	2.05	0.28

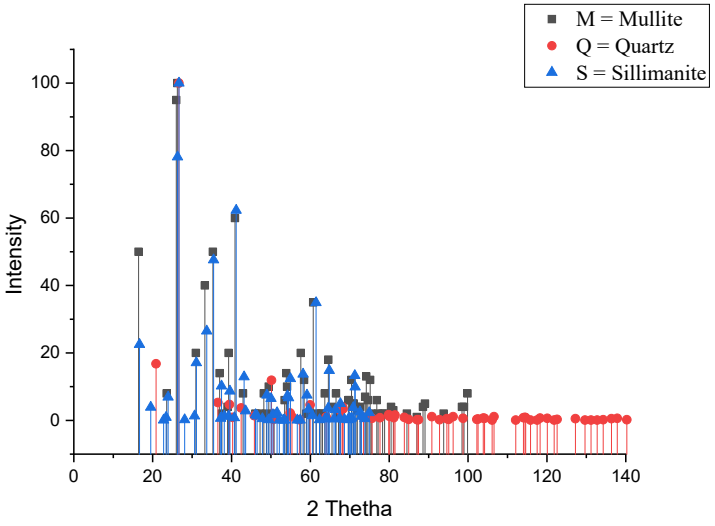

Figure 3. XRD phase match analysis of fly ash.

3.2. Materials performance

The cube geopolymer specimens in the water-submerged curing can be seen in Figure 4. There was no sign of efflorescence (crusty white sodium release) on the specimens after the curing. Each variation experienced a gradual increase in compressive strength during the curing until 28 days as shown in Figure 5. The highest compressive strength belonged to the 28-day code C specimen at 25.2 MPa, which incorporated the smallest CWP than the other mixes. Specimen C28 can withstand the most compressive force, even within a low liquid-to-solid ratio. This may be attributed to applying higher molarity of alkaline liquid at 12 M in turns of reducing the liquid to solid ratio.

Figure 4. Geopolymer specimens under water curing.

The particle size of CWP may also play an important role in the compressive strength as, in this case, they are used in the fine aggregate range, mostly at 210 µm and above (+70 mesh), as shown in Figure 2. It was reported that the reduction in particle size of tile ceramic from -470 and -140 µm (d50 decreases from 76 to 14 µm) for geopolymer demonstrated a significant increase in compressive strength (Komnitsas et al., 2015). Ceramic tile waste passing through 76 µm demonstrated higher strength with 55% proportion in fly ash-based geopolymer (Mahmoodi et al., 2020). This particle size may be an indication that applying more of this CWP would lower the strength.

The result of water absorption measurement showed a pattern where it gradually declined in accordance with longer curing time until 28 days for all mixes (Figure 6). The data revealed the lowest water absorption at 1.99% for specimen C28. In addition, the abrasion rate, porosity, and density, as shown inTable 3, also showed a similar pattern, which can be related to their compressive strength. The abrasion rate reduced along with increasing curing time, resulted in lower rate for the most compressive strength. Moreover, the porosity and density data indicate the densification process during the water curing time. This can be clearly seen when the porosity declined, the density experienced an increase. Meanwhile, in a dry ambient curing temperature, density of the geopolymer would decrease during the curing time due to excess water evaporation (Albidah et al., 2021). In this case, the slight increment of density during water immersion was in agreement with normal concrete cured in water (Raheem et al., 2013). This may be due to the hydration reaction involved during the curing. Further, this densification phenomenon can also lead to better mechanical performance, as shown by each mix during the curing period.

Table 3. Abrasion rate, porosity, and density of geopolymer specimens.

Code	Abrasion rate (%)		Porosity (%)		Density (g/cm^2)	
	10 M	12 M	10 M	12 M	10 M	12 M
A;7	0.51	0.62	7.53	17.43	1.69	1.87
A;14	0.53	0.34	6.1	9.61	1.7	1.87
A;21	0.19	0.13	3.77	5.57	1.71	1.89
A;28	0.11	0.06	3.57	5.49	1.86	2.12
B;7	0.15	0.11	8.98	7.66	1.83	1.58
B;14	0.08	0.64	7.85	6.76	1.85	1.59
B;21	0.07	0.03	5.49	5.37	1.89	1.67
B;28	0.04	0.02	4.84	4.99	2.06	1.69
C;7	0.01	0.01	15.42	4.47	1.78	1.8
C;14	0.08	0.04	11.03	4.45	1.93	1.82
C;21	0.01	0.02	9.83	3.62	1.93	1.93
C;28	0.02	0.007	9.83	3.46	2	1.93

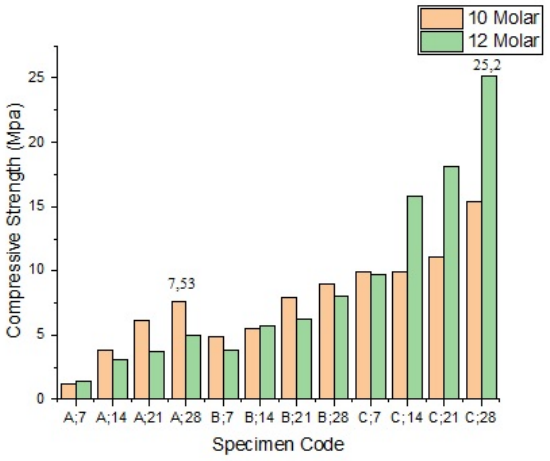

Figure 5. Compressive strength of geopolymer specimens.

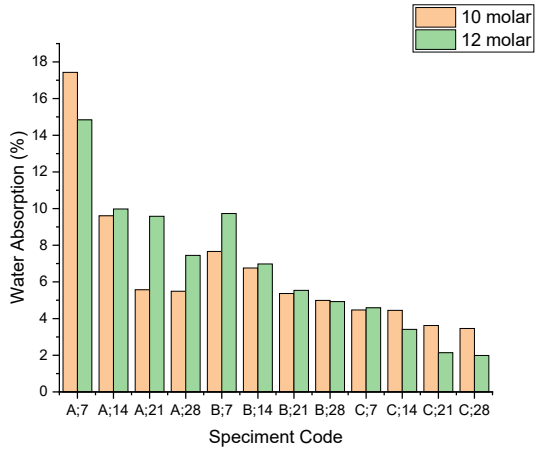

Figure 6. Water absorption of geopolymer specimens.

The morphological image of fly ash and specimen C28 performed by SEM in 500x magnification can be seen in Figure 7 . Fly ash particles were indicated by spherical shape with a size under 50 µm, which can be effective in promoting geopolymer reaction. Furthermore, the microstructure of C28 showed geopolymer matrix produced through the bonding within dissolved/reacted fly ash and a smaller size of CWP in an alkaline medium. The fine aggregate particles seemed to incorporate in the matrix where the gap between particles was connected by the binder, supporting denser geopolymer mortar. This denser microstructure would induce the geopolymer performance. However, microcracks markedly appeared on the surface. This may be the effect of sorptivity in the specimen during the water immersion, which leads to microcracks (Giasuddin et al., 2013). The interconnected microcracks on the surface can also be in relation to the high uptake of water (Collins & Sanjayan, 2001). In this case, water exposure might lead to water uptake in the specimens during hardening time so that microcracks propagate. This can create channeling network through the weak points of the bonding between the geopolymer matrix and the larger particle size from CWP. Some voids also appeared on the surface, which was more likely due to unreacted sodium leached during the water curing.

The SEM image at higher magnification (Figure 7c) represented some incomplete geopolymer reaction of CWP. This may occur due to the exposure of water on the surface, which hinders geopolymerization. Meanwhile, fly ash seems completely dissolved and thus contributing to the geopolymer matrix of the mortar specimen.

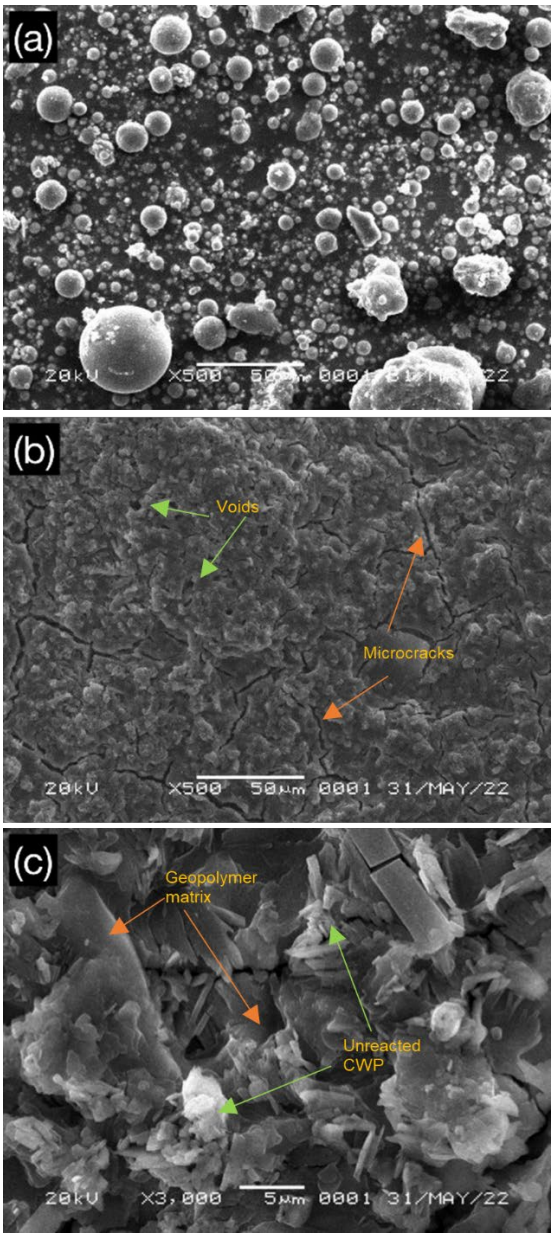

Figure 7. Microstructure of fly ash (a), geopolymer specimen C28 with 500x-, (b) and 3000x magnification (c).

4. Conclusion

Taking into account all of the data provided, it can be concluded that ceramic waste material possesses significant alumina silicate, which is essential in geopolymer reaction. In combination with fly ash, this ceramic waste powder (CWP) can contribute to the production of geopolymer matrix. The microcracks propagation on the surface may be associated with the weak interaction within particles in the matrix and the influence of water exposure. This must be noted that the CWP used in this study is within the category of fine aggregate particle size. Therefore, the mechanical properties decreased with the further addition of the CWP in the mix. There is also a linear correlation between compressive strength, water absorption and abrasion rate. The compressive strength experienced an increase while the water absorption and abrasion rate decreased. In addition, densification occurred with the indication of a slight increment in density followed by a decrease in porosity.

The best mechanical properties are shown by specimen C28, owing compressive strength at 25.2 MPa, water absorption at 1.99%, abrasion rate at 0.007 mm/min. Therefore, the ceramic waste can be of potential value for utilization in fly ash-based geopolymer mortars. It was confirmed that water curing can be applied to develop this geopolymer and potentially reduce the risk of efflorescence. Further research is required to optimize this binary geopolymer system and thus can be put in place in the near future.

Conflict of interest

The authors have no conflict of interest to declare.

Acknowledgements

The authors would like to express gratitude to those who provided invaluable assistance in this study. This includes Dr. Yudha Satya Perkasa from UIN Sunan Gunung Djati. Dr. Gunawan, head of Center for Ceramics in Bandung where this experiment was conducted. We extend our thanks to Mokhamad Syai from PLTU Banten 2 Labuan OMU (PT. Indonesia Power) for generously providing the fly ash for this study.

Funding

The authors received no specific funding for this work.

References

Albidah, A., Alghannam, M., Abbas, H., Almusallam, T., & Al-Salloum, Y. (2021). Characteristics of metakaolin-based geopolymer concrete for different mix design parameters. *Journal of Materials Research and Technology*, *10*, 84–98. https://doi.org/10.1016/j.jmrt.2020.11.104

America society for testing and material (ASTM) C125. (2007). *Standard Terminology Relating to Concrete and Concrete Aggregates* (pp. 1–5).

Amran, M., Debbarma, S., & Ozbakkaloglu, T. (2021). Fly ash-based eco-friendly geopolymer concrete: A critical review of the long-term durability properties. *Construction and Building Materials*, *270*, 121857. https://doi.org/10.1016/j.conbuildmat.2020.121857

Barcelo, L., Kline, J., Walenta, G., & Gartner, E. (2014). Cement and carbon emissions. *Materials and Structures/Materiaux et Constructions*, *47*(6), 1055–1065. https://doi.org/10.1617/s11527-013-0114-5

Collins, F., & Sanjayan, J. G. (2001). Microcracking and strength development of alkali activated slag concrete. *Cement and Concrete Composites*, *23*(4–5), 345–352. https://doi.org/10.1016/S0958-9465(01)00003-8

Davidovits, J. (1989). Geopolymers and geopolymeric materials. *Journal of Thermal Analysis*, *35*, 429–441. https://doi.org/10.1007/BF01904446

Davidovits, J. (1991). Geopolymers - Inorganic polymeric new materials. *Journal of Thermal Analysis*, *37*(8), 1633–1656. https://doi.org/10.1007/BF01912193

Ekaputri, J. J., Ulum, M. B., Triwulan, Bayuaji, R., Susanto, T. E., & Al Bakri Abdullah, M. M. (2015). A Comprehensive Characterization and Determination of Fly Ashes in Indonesia Using Different Methods. *Applied Mechanics and Materials*, *754–755*, 320–325. https://doi.org/10.4028/www.scientific.net/amm.754-755.320

Giasuddin, H. M., Sanjayan, J. G., & Ranjith, P. G. (2013). Strength of geopolymer cured in saline water in ambient conditions. *Fuel*, *107*, 34–39. https://doi.org/10.1016/j.fuel.2013.01.035

Hardjito, D., Wallah, S. E., Sumajouw, D. M. J., & Rangan, B. V. (2005). Fly ash based geopolymer concrete. *Australian Journal of Structural Engineering*, *6*(1), 1–10.

Komnitsas, K., Zaharaki, D., Vlachou, A., Bartzas, G., & Galetakis, M. (2015). Effect of synthesis parameters on the quality of construction and demolition wastes (CDW) geopolymers. *Advanced Powder Technology*, *26*(2), 368–376. https://doi.org/10.1016/j.apt.2014.11.012

Kumar, S., Kumar, R., & Mehrotra, S. P. (2010). Influence of granulated blast furnace slag on the reaction, structure and properties of fly ash based geopolymer. *Journal of Materials Science*, *45*(3), 607–615. https://doi.org/10.1007/s10853-009-3934-5

Luhar, I., & Luhar, S. (2022). A Comprehensive Review on Fly Ash-Based Geopolymer. *Journal of Composites Science*, *6*(8), 1–59. https://doi.org/10.3390/jcs6080219

Ma, B., Su, C., Ren, X., Gao, Z., Qian, F., Yang, W., Liu, G., Li, H., Yu, J., & Zhu, Q. (2019). Preparation and properties of porous mullite ceramics with high-closed porosity and high strength from fly ash via reaction synthesis process. *Journal of Alloys and Compounds*, *803*, 981–991. https://doi.org/10.1016/j.jallcom.2019.06.272

Mahmoodi, O., Siad, H., Lachemi, M., Dadsetan, S., & Sahmaran, M. (2020). Development of ceramic tile waste geopolymer binders based on pre-targeted chemical ratios and ambient curing. *Construction and Building Materials*, *258*, 120297. https://doi.org/10.1016/j.conbuildmat.2020.120297

Mohajerani, A., Suter, D., Jeffrey-Bailey, T., Song, T., Arulrajah, A., Horpibulsuk, S., & Law, D. (2019). Recycling waste materials in geopolymer concrete. *Clean Technologies and Environmental Policy*, *21*(3), 493–515. https://doi.org/10.1007/s10098-018-01660-2

Pacheco-Torgal, F., & Jalali, S. (2010). Reusing ceramic wastes in concrete. *Construction and Building Materials*, *24*(5), 832–838. https://doi.org/10.1016/j.conbuildmat.2009.10.023

Puligilla, S., & Mondal, P. (2013). Role of slag in microstructural development and hardening of fly ash-slag geopolymer. *Cement and Concrete Research*, *43*(1), 70–80. https://doi.org/10.1016/j.cemconres.2012.10.004

Raheem, A. A., Soyingbe, A. A., & Emenike, A. J. (2013). Effect of Curing Methods on Density and Compressive Strength of Concrete. *International Journal of Applied Science and Technology*, *3*(4), 55–64. https://doi.org/10.1088/1757-899X/471/3/032059

Rangan, P. R., Dendo, E. A., Bokko, J., & Mantirri, P. A. (2020). Mortar Geopolimer Abu Sekam Padi Berbahan Dasar Limbah Abu Batu Bara Hasil Pembakaran Asphalt Mixing Plant. Journal Dynamic Saint, 5(1), 927-938.

Samadi, M., Hussin, M. W., Lee, H. S., Sam, A. R. M., Ismail, M. A., Lim, N. H. A. S., ... & Khalid, N. H. A. (2015). Properties of mortar containing ceramic powder waste as cement replacement. J. Teknol, 77(12), 93-97.

Shi, C., & Day, R. L. (1999). Early strength development and hydration of alkali-activated blast furnace slag/fly ash blends. *Advances in Cement Research*, *11*(4), 189–196. https://doi.org/10.1680/adcr.1999.11.4.189

Simão, L., Fernandes, E., Hotza, D., Ribeiro, M. J., Montedo, O. R. K., & Raupp-Pereira, F. (2021). Controlling efflorescence in geopolymers: A new approach. *Case Studies in Construction Materials*, *15*. https://doi.org/10.1016/j.cscm.2021.e00740

Vásquez, A., Cárdenas, V., Robayo, R. A., & de Gutiérrez, R. M. (2016). Geopolymer based on concrete demolition waste. *Advanced Powder Technology*, *27*(4), 1173–1179. https://doi.org/10.1016/j.apt.2016.03.029

Wardhono, A., Law, D. W., & Strano, A. (2015). The strength of alkali-activated slag/fly ash mortar blends at ambient temperature. *Procedia Engineering*, *125*, 650–656. https://doi.org/10.1016/j.proeng.2015.11.095