Influence of drying temperature on compressive strength of geopolymer binder

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Abstract. Environmentally friendly materials with low CO₂ emissions have become an inevitable trend of the construction industry. This requires research establishments to develop new construction material products and manufacturing enterprises to take the chance and change production directions to meet the needs of the market. Among those new materials, Geopolymer materials promise to contribute to the development of increasingly sustainable construction. This paper aims to study the effect of drying temperature and drying time on compressive strength of geopolymer binders made from type F fly ash, ground granulated blast furnace slag combined with activators solution, including 10M NaOH caustic solution, liquid glass. Research results show that when the drying temperature increases from 60°C to 120°C and the drying time increases from 6 to 24 hours, the compressive strength of geopolymer binders could be enhanced from 2.5 to 3.0 times, compressive strength can reach over 50MPa, fully meet the requirements of the strength of binders used for manufacturing building materials products in civil engineering and industrial construction.

1. Introduction

Using environmentally friendly green materials is becoming an indispensable trend and is the goal of the building materials manufacturing industry. In order to achieve this goal, building materials products must meet two requirements: Consume less energy for its production and save energy consumption when put into use. Each year the demand for the production and the use of Portland cement is increasing due to the development speed of the construction industry [1]. According to statistics, Portland cement production worldwide increases by 3% annually [2]. Therefore, the amount of CO₂ emissions into the atmosphere during the production of Portland cement is also increasing. In addition, Portland cement is one of the most energy-consuming building materials, after aluminum and steel [3]. Therefore, the production of new environmentally friendly construction materials products, using industrial waste, as raw materials for production, will help both protecting the environment and saving natural resources in production. Fly ash, a fine solid waste generated by burning coal in thermal power plants, is one of the most common worldwide wastes.

In Vietnam, as reported by the Ministry of Industry and Trade: Fly ash, slag, gypsum currently mainly generated from coal-fired power plants. These plants consume about 47.8 million tons of coal/year with ash and gypsum ash discharged more than 16.4 million tons annually. It is expected that by 2020, there will be more coal thermal power projects being put into operation, consuming about 60 million tons of coal / year and generating about 20.5 million tons of ash and gypsum. However, only about 30% Of these industrial waste is used in concrete, as a leveling or stabilizing material, the remaining 70% must be discharged outside the surrounding environment of the factory, occupying a lot of valuable land area, causing enormous pressure on dumping and environmental protection.
Currently, a number of establishments producing construction materials have researched and invested in technology so that they can use ash, slag, gypsum and other industrial waste to replace raw materials in the production of unburnt construction materials and cement production, concrete, landfilling materials in construction works [5].

Fly ash has been used as a mineral additive to produce Geopolymer binder in construction. In 1978, Davidovits introduced the term geopolymer, which is an inorganic polymer, formed by the reaction between an alkaline solution, silica and alumina in the material. The solid material has an amorphous 3-dimensional structure similar to alumina silicate glass. However, unlike glass, these materials are formed at low temperatures, as a result, can be combined with aggregates and other reinforced materials such as fiber reinforcement and steel reinforcement during hardening. The most common activators are a mixture of water, a sodium hydroxide solution and a sodium silicate solution (liquid glass). This binder has received more and more attention from countries around the world because it can replace Portland cement, save natural mineral resources such as limestone, clay and emit less CO\(_2\) [6]. The main reason and also the driving force of the development of this Geopolymer is that concrete using this type of binder has high strength at early-age, low shrinkage, good fire resistance and is durable against the aggressive environment when compared to concrete using conventional Portland cement [7,8]. The strength and microstructure of geopolymer binder as well as geopolymer concrete have been studied with many types of raw mineral materials [9-11]. The researches confirm the potential of this binder through the results on mortar, concrete, brick, composite materials, heat-resistant coatings, etc. in the construction industry [12, 13]. Some research areas on geopolymer including mix design, aggregate alkali reaction, early and long-term strength, bonding ability with reinforcement, durability in different environments and even high strength have also been studied [14-16]. In Vietnam there have been a number of researches on geopolymer concrete [17-21]. These studies also have confirmed the ability to make geopolymer entirely from materials in Vietnam, the mechanical properties of geopolymer binder are as good as that of Portland cement but geopolymer binder has superior durability under the aggressive condition and high temperatures.

In some studies on geopolymer using fly ash, the initial hardening process at room temperature usually takes place very slowly; it requires higher temperature (drying process) to decrease the hardening time [22, 23]. In this study, in order to increase the practicality of producing geopolymer binder with high amount of fly ash, the article has combined fly ash and ground granulated blast furnace slag to improve the initial strength of binder, helping to demould without heat curing as Portland cement binder after 24h. The binder will then be dried to promote strength development according to the drying temperature and drying time.

2. Materials and experimental methods

2.1. Materials

To make geopolymer binder, the research uses the following materials:

2.1.1. NaOH solution 10M

A NaOH solution 10M with 1.33 kg/liter is used as an alkaline activator mixed with liquid glass solution of sodium silicate (\(Na_2SiO_3\)). In this paper, caustic soda in the form of flakes is mixed with distilled water to form a solution of 10M concentration as desired.

2.1.2. Liquid glass solution (\(Na_2SiO_3\))

Liquid glass has a density of 1.296 kg/liter. The chemical composition of liquid glass is as follows: \(Na_2O\) (11.1%), \(SiO_2\) (30.7%), \(H_2O\) (58.2%), silicate module 2.766.

2.1.3. Fly ash

This paper uses fly ash from Pha Lai Thermal Power Plant. This is fly ash with low calcium content, type F according to ASTM C618, total oxide content (\(SiO_2 + Al_2O_3 + Fe_2O_3 = 87.7 > 70\%\)). The chemical composition and some properties of fly ash are shown in Table 1 and Table 2:
Table 1. Chemical composition of Pha Lai fly ash

| Content of oxides, (% by mass) | SiO$_2$ | TiO$_2$ | Al$_2$O$_3$ | Fe$_2$O$_3$ | MnO | MgO | CaO | Na$_2$O | K$_2$O | P$_2$O$_5$ | L.O.I (%) |
|------------------------------|---------|---------|-------------|-------------|------|------|------|---------|-------|-----------|----------|
| SiO$_2$                      | 54.5    | 0.86    | 27.3        | 5.9         | 0.05 | 1.1  | 1.02 | 0.2     | 6.38  | 0.2       | 2.23     |

Table 2. Properties of Pha Lai fly ash

| N°  | Characteristic                        | Unit       | Result | Test method          |
|-----|---------------------------------------|------------|--------|----------------------|
| 1   | Density                               | g/cm$^3$   | 2.45   | TCVN 4030:2003       |
| 2   | Activity Index                        | %          | 82.5   | TCVN 6882:2001       |
| 3   | Particle analysis by Laser            |            |        |                      |
|     | Content of particle pass through the  | %          | 10     |                      |
|     | Particle size diameter                | (µm)       | 1.462  |                      |
|     | Average particle diameter             | (µm)       | 7.630  |                      |

2.1.4. Ground granulated blast furnace slag (GGBFS)

Hoa Phat blast furnace slag is finely ground in a vibrating ball mill for 90 minutes. The purpose of using blast furnace slag is to shorten the demolding time of the Geopolymer binder. The properties of GGBFS are shown in Table 3 and Table 4:

Table 3. Chemical composition of Hoa Phat GGBFS

| Content of oxides, (% by mass) | SiO$_2$ | TiO$_2$ | Al$_2$O$_3$ | Fe$_2$O$_3$ | MnO | MgO | CaO | Na$_2$O | K$_2$O | P$_2$O$_5$ | L.O.I (%) |
|------------------------------|---------|---------|-------------|-------------|------|------|------|---------|-------|-----------|----------|
| SiO$_2$                      | 34.52   | -       | 12.38       | 0.66        | 0.03 | 0.85 | 41.54| 0.43    | 0.24  | 0.23      | 0.96     |

Table 4. Properties of Hoa Phat GGBFS

| N°  | Characteristic                        | Unit   | Value  | Test method         |
|-----|---------------------------------------|--------|--------|---------------------|
| 1   | Density                               | g/cm$^3$ | 2.94   | TCVN 4030:2003      |
| 2   | Activity Index                        | %      | 104.3  | TCVN 4315:2007      |
| 3   | Average particle diameter             | µm     | 10.4   |                      |
| 4   | L.O.I                                 | %      | 0.96   | TCVN 141:2008       |

2.1.5. Standard sand

The standard sand according to the Vietnamese standard of TCVN 6016: 2011 and ISO 679: 2009 is used to determine the strength of geopolymer binder.

2.1.6. Mixing water

The mixing water meets all technical requirements according to TCVN 4506: 2012 "Water for concrete and mortar".

2.2. Experimental methods

The mechanical and physical properties of raw materials are determined according to Vietnamese standards. In addition, to determine the strength of geopolymer binder, the paper is based on TCVN 6016:2011 in which the mortar for determining binder strength is cast into the mold 40 × 40 × 160 mm and hardening process take places at laboratory temperature (20°C - 30°C). After some experimental investigations, the proportion of materials used to determine the strength of geopolymer binder was calculated based on the fixed parameters as follows:

- Ratio by weight of \[rac{\text{liquid glass}}{\text{NaOH 10M solution}}\] = is 2.5.
- Ratio by weight of \( \frac{GGBFS}{FA} \) is 10/90.
- Ratio by weight of \( \frac{\text{Liquid glass} + \text{NaOH 10 M Solution}}{FA+GGBFS} \) is 0.5.
- The proportions of materials according to TCVN 6016: 2011 are as follows:
  + Ratio by weight of standard sand/binder: \( \frac{\text{Sand}}{\text{Binder}} \) = 3.0. The Geopolymer binder in this paper is the total amount of solid material (Na\(_2\)O oxide + SiO\(_2\) oxide in liquid glass + Na\(_2\)O oxide in 10M NaOH solution + FA + GGBFS).
  + Ratio by weight of Mixing water/binder: \( \frac{\text{Water}_{\text{total}}}{\text{Binder}} \) = 0.5. (The amount of mixing water is the total amount of water in the liquid glass solution, the caustic soda solution, and the amount of free water added).

The composition of materials used for determining geopolymer binder strength according to TCVN 6016: 2011 is shown in Table 5 as follows:

| Table 5. Composition of materials used for determining geopolymer binder strength (kg/m\(^3\)) |
|---------------------------------------------------------------|
| Raw materials                                      | FA, (kg) | GGBFS, (kg) | Liquid glass, (kg) | NaOH 10M, (kg) | Free water, (kg) | Standard sand (kg) |
| Quantity, (kg/m\(^3\))                | 293.3    | 125.8       | 149.7            | 59.9           | 115.3           | 1488              |

The raw materials (GGBFS, FA, standard sand) were mixed about 2 minutes in the mixer. A mixture of activator solutions, including liquid glass, 10M NaOH solutions and free water, then was poured into the homogeneous mineral mixture above. After mixing for 3 more minutes, the mortar samples were cast into 4x4x16 cm molds and cured at room temperature for 24 hours. After that, all specimens were demoulded, wrapped in sealed aluminum bags to allow the curing process to take place under endogenous conditions. There were 2 groups of specimens:

  + Group 1: Specimens in endogenous condition at room temperature were determined the compressive strength according to standards TCVN 6016: 2011 at 3, 7 and 28 days.
  + Group 2: After 3 days, the specimens (always in sealed aluminum bags) were dried from 6 - 48 hours in an oven with the temperature range of 60-120°C. After cooling to room temperature, these specimens were determined compressive strength according to standards TCVN 6016: 2011.

The results of compressive strength of group 2 were compared with that of group 1 to clarify the effect of drying process.

3. Results and discussion

3.1. Compressive strength development of Geopolymer binder in endogenous conditions

The mortar mixture was mixed according to materials proportion shown in Table 4, the results of compressive strength in endogenous conditions over time were presented in Table 6 and Figure 1 as follows:

| Table 6. Compressive strength of Geopolymer binder in endogenous conditions |
|--------------------------------------------------------------------------|
| Nº | Name of sample | Compressive strength with time, MPa |
|----|----------------|-----------------------------------|
|    |                | 3 days  | 7 days  | 28 days |
| 1  | Control sample | 15.3    | 25.2    | 41.5    |
The results showed that the compressive strength of mortar samples in the autogenous condition (without the effect of temperature) increased over time. The compressive strength reached 15.3 MPa at 3 days and reached nearly 42 MPa after 28 days of hardening. The addition of ground granulated blast furnace slag has helped promote the hardening process of the Geopolymer binder, so that the demolding process can be done after 1 day of casting which is similar to that of ordinary Portland cement. Compared with other studies [22, 23], if using 100% fly ash, the curing process of geopolymer will take place very slowly. Therefore, blast furnace slag plays an important role to increase practicality in the application of geopolymer binders containing high amount of fly ash in construction and building material industry.

### 3.2. Influence of drying temperature and drying duration on compressive strength of Geopolymer binder

The purpose of the drying process is to accelerate the polymerization process occurring in geopolymer binders. The effect of the drying process on the compressive strength of Geopolymer adhesive is shown in Table 7:

**Table 7. Influence of drying process on compressive strength of Geopolymer binder**

| Nº | Drying duration, hour | Temperature, °C | Compressive strength, MPa |
|----|-----------------------|-----------------|---------------------------|
| 1  |                       | 60°C            | 37.1                      |
| 2  | 6h                    | 90°C            | 41.8                      |
| 3  |                       | 120°C           | 46.8                      |
| 4  |                       | 60°C            | 43.9                      |
| 5  | 24h                   | 90°C            | 48.2                      |
| 6  |                       | 120°C           | 49.6                      |
| 7  |                       | 60°C            | 45.4                      |
| 8  | 48h                   | 90°C            | 50.7                      |
| 9  |                       | 120°C           | 52.2                      |
3.2.1. Influence of drying duration on compressive strength

Through results of the compressive strength in Table 7, the effect of the drying process on the compressive strength of geopolymer binder is shown in Figure 2 as follows:

![Graph a) 60°C](image)

Values of 28-day compressive strength of control sample (without drying)
Rn = 41.5MPa

![Graph b) 90°C](image)

Values of 28-day compressive strength of control sample (without drying)
Rn = 41.5MPa
3.2. Influence of drying duration on compressive strength

The research results show that, under the influence of temperature, the geopolymer strength grows very fast. At all 3 levels of drying temperature of 60°C, 90°C and 120°C, the development of compressive strength increases with drying time, mortar samples of 48-hour heat curing have the highest strength. However, the difference between the 24-hour heat curing and 48-hour heat curing samples was negligible, only approximately 10%. Thus, an optimal drying time of about 24 hours for geopolymer binder can be recommended to save energy and production costs. For samples dried at 60°C for a period of 6 hours, the compressive strength of the binder was the lowest, only about 37MPa. This could be explained by the fact that the effect of temperature on the polymerization process is not strong enough when compared to the remaining samples. Therefore, it should require a drying time of at least 24 hours to promote the chemical reaction and achieve the strength value like that of control samples in endogenous condition at 28 days.

3.2.2. Influence of drying temperature on compressive strength

The analytical results of drying temperature on compressive strength of geopolymer mortar are shown in Figure 3:
Figure 3. Effect of drying temperature on compressive strength of geopolymer binder
a) Compressive strength at different temperature when drying samples for 6 h
b) Compressive strength at different temperature when drying samples for 24 h
c) Compressive strength at different temperature when drying samples for 48 h

The results in Figure 3 show that the strength development speed of all geopolymer binder samples at different drying temperature ranges are relatively similar. When the drying temperature increased from 60-120°C, the compressive strength at the age of 3 days increased by 2.5-3.0 times compared to non-
heat curing samples, and the compressive strength of the mortar samples dried at 120°C is the highest. It means that the activation reaction in geopolymer binders takes place more strongly when drying temperature increases higher. However, the geopolymer's hardening process will slow down gradually over time, particularly the samples after 24 hours seem to have reached their maximum strength values of 40-50Mpa. When the drying time is only 6 hours, the difference in compressive strength at drying temperatures of 60°C, 90°C and 120°C is quite obvious, the strength difference depending to temperature is about 10-20%. Whereas, if the drying time is longer than 24 h, the compressive strength at 90°C compared to samples dried at 120°C is almost negligible. Therefore, drying temperature of 90°C can be considered as the optimal choice for making geopolymer binders with high fly ash content combined with blast furnace slag in the composition.

4. Conclusion

The paper presented the effect of drying temperature and drying duration on the compressive strength development of geopolymer binders. Through the research results, some important conclusions are drawn as follows:

- The use of a small content of ground granulated blast furnace slag allows to shorten the geopolymer hardening time at early age, allow to demoulding after 24 hours of mixing and casting. This technological time is similar to that of Portland cement.
- Drying temperature and drying duration play an important role in activating the polymerization process in geopolymer binder. When drying from 6 to 48 hours, the compressive strength will increase. The speed of hardening and polymerization is very slow at 60°C when compared to samples dried at 90°C and 120°C. The difference in compressive strength of geopolymer binder dried at 24h and 48 hours is negligible. Therefore, it is recommended that the drying time of about 24 hours and the drying temperature of about 90°C is optimum to save energy and production cost.
- The compressive strength of geopolymer binder samples can reach over 40Mpa after only 24 hours of curing; this further clarifies the feasibility in manufacturing of products using geopolymer in construction.

References

[1] Ross S and Dru 2010 *Green Building Materials: A Guide to Product Selection and Specification* (3rd Edition, John Wiley & Sons, Inc)
[2] Habert G 2014 *Eco-efficient Construction and Building Materials, Life Cycle Assessment (LCA), Eco-Labelling and Case Studies*, Woodhead Publishing Series in Civil and Structural Engineering.
[3] Davidovits J 1994 Global Warming Impact on the Cement and Aggregate Industries, *World Resource review*, (6) 2: 263-278.
[4] Hung L V 2015 Report: Research on using fly ash with L.O.I greater than 6% as an additive for the production of concrete and mortar in construction-Code-RD 122-13 Vietnam Institute of Construction Materials.
[5] Tuan N V 2016 Research on using fly ash and bottom ash (An Khanh - Thai Nguyen) and grit waste to make concrete bricks, Ministry of Construction project, code RD113-16TX.
[6] Davidovits J 2015 *Geopolymer Chemistry and Applications*, Institut Géopolymère.
[7] Provis J L and Deventer J S 2009 *Geopolymers-Structure, Processing, Properties and Industrial Applications* CRC Press, Woodhead Publishing Ltd. UK.
[8] Duxon P J A, Provis J L, Luckey G C, Palomo A and Veventer J V 2007 Geopolymer Technology: The Current State of the Art *Journal of Material Science* (42):2917-2933.
[9] Palomo A, Grutzeck M W and Blanco M T 1999 Alkali-Activated Fly Ashes: A Cement for the Future Cement and Concrete Composites (29): 1323-1329.
[10] Steveson M and Sagoe-Crentsil K 2005 Relationships between Composition, Structure and Strength of Inorganic Polymers Journal of Material Science (40):4247-4259.
[11] Latella B A, Perera D S, Durce D, Mehrtens E.G. and Davis J 2008 Mechanical Properties of Metakaolinite-based geopolymers with Molar Ratios of Si / Al ~ 2 and Na / A ~ 1 Journal of Material Science (43):2693-2699.
[12] Yang K H, Song J K, Lee K S and Ashour A F 2009 Flow and Compressive Strength of Alkali-Activated Mortars ACI Materials Journal (106): 50-58.
[13] Daniel L Y K and Sanjayan J G 2010 Effect of Elevated Temperatures on Geopolymer Paste, Mortar and Concrete Cement and Concrete Research (40): 334-339.
[14] Khadka S D, Priyantha W J and Senadheera S 2005 Relationships between Composition, Structure and Strength of Inorganic Polymers Journal of Material Science (40): 4247-4259.
[15] Hardjito D, Steenie E W, Dody M J S and Rangan B V 2004 On the Development of Fly Ash Based Geopolymer Concrete ACI Materials Journal (101): 467-472.
[16] Fernandez-Jimenez A M, Palomo A and Lopez-Hombrados C 2006 Engineering Properties of Alkali-Activated Fly Ash Concrete ACI Materials Journal (103): 106-112.
[17] Chanh N V, Trung B D and Tuan D V 2008, “Recent Research Geopolymer Concrete”, the 3rd ACF International Conference-ACF/VCA, Ho Chi Minh City.
[18] Dong D V 2009 Research on some mechanical properties of inorganic polymer concrete, Vietnam Bridge and Road Magazine (12): 22-26.
[19] Dong D V 2010 Research on compressive and tensile behavior of inorganic polymer mortar Journal of Transport (3):28-34.
[20] Dong D V 2011 Research on destructive behavior of inorganic polymer mortar at high temperature Transport Magazine (3):30-33.
[21] Dong D V 2011 Research on some properties of inorganic polymer mortar in seawater corrosive environment Transport and Communications Magazine (6).
[22] Khoa T N, Tuan-Anh L and Lee K 2016 Theoretical and experimental study on mechanical properties and flexural strength of fly ash-geopolymer concrete Construction and Building Materials (106): 65-77.
[23] Hung P D 2016 Mechanical properties of geopolymer concrete using fly ash reinforced with polypropylene fibers Journal of Construction Science and Technology (1): 60-67.