HIGH CALCIUM FLY ASH GEOPOLYMER CONTAINING NATURAL RUBBER LATEX AS ADDITIVE

Prinya Chindaprasirt\textsuperscript{1} and * Charoenchai Ridtitru\textsuperscript{2}

\textsuperscript{1}Khon Kaen University, Faculty of Engineering, Department of Civil Engineering, Sustainable Infrastructure Research and Development Center, Khon Kaen, Thailand; \textsuperscript{2}Rajamangala University of Technology Isaan, Faculty of Engineering, Department of Civil Engineering, Thailand

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ABSTRACT: This research studied the use of natural rubber latex as an additive in high calcium fly ash geopolymer. The high calcium fly ash geopolymer with natural rubber latex (medium ammonia concentrated latex type) content of 0, 1, 2, 3, 5 and 10 percent by weight of fly ash was incorporated in the mixtures. In this study, geopolymer mortars were prepared. The ratios of sand to fly ash of 2.75, liquid to fly ash ratio of 0.6 and Na\textsubscript{2}SiO\textsubscript{3} to NaOH ratio of 0.33 were used for all mixtures. Setting time, workability, absorption, apparent porosity, compressive strength, flexural strength, and surface abrasion resistance were tested. The test resulted shows that the optimum natural rubber latex content was 1.0 percent by weight of fly ash to obtain geopolymer mortar with improved mechanical properties and a slight reduction in workability, and an increase in setting time. The addition of more than 1 percent by weight of fly ash latex started to reduce the compressive strength due to increases in rubber particle, interface, and gap between rubber particle and paste.

Keywords: Geopolymer, Fly ash, Rubber, Setting time, Medium ammonia concentrated latex

1. INTRODUCTION

Geopolymer is an aluminosilicate material consisting of a structure of amorphous and semi-crystalline phases. The substrate used in geopolymer production can be liberated from the particle surface with the use of base solution at both room temperature and moderate temperatures of 40-80 °C. Consequently, the geopolymerization leads to the reaction, which forms aluminosilicate chain. The material sets and endures compressive strength well [1].

The high calcium fly ash consists of a high CaO content and is categorized as type C fly ash. The high CaO content gives a higher self-cementitious property for high calcium fly ash [2]. The high calcium content also leads to the shortening of the setting time [3] and comparable strength development to the class F fly ash. The main advantages of using high calcium fly ash are increase in the degree of hydration at the early age and the hardening and gain in strength at ambient curing temperature without the use temperature heat curing [4, 5].

The mechanical property of geopolymer mortar and concrete is similar to the mechanical property of those from Ordinary Portland cement [6]. One of the outstanding properties of geopolymer is the high strength gain in a short period when the heat is used to accelerate the reaction [7, 8]. This is suitable for precast concrete, which is in high demand in the market. The fly ash geopolymer can resist acidic and alkaline environments better than Portland cement products because it has a strong structure of aluminosilicate [9, 10] with excellent fire resistance [7, 11]. However, there are reports that geopolymer concrete exhibits higher brittleness than OPC concrete [12, 13] and to improve brittleness, elastomeric latexes can be applied to the geopolymer system [14].

Elastomeric latex is a natural or synthetic polymer having elastic properties. The use of elastomeric latex improves the durability in concrete [15, 16], and increases the flexural strength [17]. It also decreases absorption, improves workability, resists chemical aggressions, and reduces brittleness [14]. In general, there are two basic elastomers for cement mixes viz., natural rubber latex (NRL) and synthetic rubber latexes (SRL). These elastomers affected the property of concrete in a similar way. The advantage of NRL is that it is readily usable and consumes less processing and energy in the production than that of synthetic rubber latex.

Moreover, the synthetic rubber latex cost is high as it required additional processing and other natural resources [18]. Very few researchers have studied the use of NRL with geopolymer. Lee (2016) used Styrene-butadiene with geopolymer [19] and found that it significantly increased the flexural strength in the order of 50%. Generally, the use of NRL with OPC needs to be mixed, with non-ionic surfactant to avoid agglutination when it is mixed with water and cement [20, 21]. However, geopolymer has high alkalinity, and the anion is similar to that of NRL.
Therefore, it should distribute itself well in the geopolymer system. The basically sound properties of geopolymer could thus be improved with the incorporation of rubber latex. This research is thus aimed to study the properties of geopolymer mortar incorporating NRL. The setting time, water absorption, abrasion resistance, compressive strength, direct tensile strength and bending strength were tested.

2. EXPERIMENTAL DETAILS

2.1 Materials

The high calcium fly ash was from Mae Moh power station in the north of Thailand. The oxide composition of fly ash is summarized in Table 1 and its X-ray diffraction (XRD) is shown in Fig. 1. Scanning Electron Microscopy (SEM) results as shown in Fig. 2 showed that the fly ash particles were spherical. The fly ash was relatively fine with 24% retained on sieve No. 325 (45 µm), the median particle size of 15.6 µm, and a specific gravity of 2.64. The particle size distribution also revealed that 76% of fly ash particles were smaller than 50 µm as shown in Fig. 3.

The NRL (medium ammonia concentrated latex type) was used as an additive to improve the properties of geopolymer mortar. The properties of concentrated latex are presented in Table 2. Sodium silicate solution (Na2SiO3) and sodium hydroxide solution (NaOH) with Na2SiO3 to NaOH ratio of 0.33 were used as alkali activators. The low Na2SiO3 to NaOH ratio was selected for reason of environmental concern. Sodium hydroxide solutions with 6, 8, 10, and 12 molar (M) concentrations were prepared using sodium hydroxide flakes of 97% purity and distilled water. Sodium silicate with 15.32% Na2O, 32.87% SiO2 and 51.80% H2O by weight was used. The fine aggregate was local river sand in saturated surface dry condition with a specific gravity of 2.62 and a fineness modulus of 2.95.

Table 1 Chemical composition and properties of Fly ash

| Chemical composition (wt%) | Fly ash |
|---------------------------|---------|
| SiO2                      | 35.86   |
| CaO                       | 23.31   |
| Al2O3                     | 16.27   |
| Fe2O3                     | 14.06   |
| SO3                       | 8.00    |
| K2O                       | 1.62    |
| TiO2                      | 0.44    |
| MnO2                      | 0.16    |
| SrO                       | 0.14    |
| As2O3                     | 0.05    |
| Rb2O                      | 0.02    |
| ZrO2                      | 0.03    |
| Cr2O3                     | 0.02    |
| LOI*                      | 0.50    |

Fig. 1 X-ray diffraction of Fly ash.

Fig. 2 Scanning Electron Microscopy of Fly ash.

Table 2 Properties of Concentrated Latex

| Count           | 2Theta(Coupled Two Theta/Theta) |
|-----------------|---------------------------------|
| 0               | 1200                            |
| 20              | 1000                            |
| 40              | 800                             |
| 60              | 600                             |
| 80              | 400                             |
| 100             | 200                             |

Fig. 3 Particle size distribution of Fly ash.
Table 2 Properties of natural rubber latex

| Property                  | Value   |
|---------------------------|---------|
| Total solid content       | 62.05   |
| Dry rubber content        | 60.35   |
| Non-rubber content        | 1.70    |
| pH                        | 10.10   |
| Volatile fatty acid number |VFA(    | 0.041   |
| Mechanical stability time | sec(    | 685     |
| Ammonia content NH3(%)    | 0.50    |
| Color                     | Milky white |

2.2 Mix Proportion and Specimen Preparation

In this study, geopolymer mortars were prepared. The ratios of sand to fly ash of 2.75, liquid to fly ash ratio of 0.6 and Na₂SiO₃ to NaOH ratio of 0.33 were used for all mixtures with the NRL addition of 0, 1, 2, 3, 5 and 10 % by weight of fly ash.

For mixing, fly ash and sodium hydroxide were mixed for 5 minutes. Sand was added at this stage and the mixing was done for another 2 minutes. The premixed rubber latex and sodium silicate were then added, and the mixing was continued for three more minutes. For mortar, the fresh geopolymer mortar was tested for flow in accordance with ASTM C1437-15 (2015) and setting time in accordance with ASTM C807-13 (2013). The fresh mortar was then cast in 50x50x50 mm cubic moulds as described in ASTM C109/C109M-16a (2016). The mortar was also cast in 100x100x100 mm cube mold for abrasion test and 40x40x160 mm prism for the flexural strength test. All specimens were covered with plastic sheets and left in a controlled 25 °C and 50 % R.H. room for 24 hours. They were then demolded and wrapped with plastic sheets to prevent moisture loss and stored in a 25 °C and 50 % R.H controlled room until the testing age.

2.3 Testing Procedure

The workability of the mortar mix was tested using the flow of mortar following the ASTM C109 /C109M - 16a (2016) The setting time of the mortar was tested in accordance with the ASTM C807-99 (1999) The compressive strength and Flexural strength of mortar were tested at the ages of 7 and 28 days in accordance with ASTM C109 /C109M - 16a (2016) and ASTM C348 - 12 (2018). The reported strengths were the average of three tests.

The abrasion resistance of mortars was determined according to ASTM C944 /C944M – 12 (2012) with a normal load on the specimen surface of 98 N for 2 minutes. The absorption of mortar was also tested using 50 mm cube specimen oven dried at 85 °C for 24 hours with immersion time of 48 hours. The reason for applying 85 °C was to avoid the effect of high temperature on geopolymer structure [19].

3.RESULT AND DISCUSSION

3.1 Setting Time and Flow

The results of setting time and flow of mortar are shown in Table 3. The increase in NRL content resulted in longer initial and final setting times due primarily to the reduced increased water content of system as rubber latex contained some water. Without rubber latex additive, the initial and final setting times of geopolymer mortar were 60 and 105 mins and conformed with the previously reported results [22]. The addition of 10% rubber latex slightly increased these values to 87 and 135 mins.

With regards to the flow of mortar, the increase in the rubber latex content decreased the flow of fresh mortar. The flow of mortar without rubber latex additive was 20.6 cm and reduced to 11.4 cm with 10% NRL. The NRL was a high viscosity liquid, and thus its presence reduced the workability of mixture.

Table 3 Setting time and flow of geopolymer mortars

| Rubber Latex (%) | Initial(min.) | Final(min.) | Flow(cm) |
|-----------------|--------------|-------------|---------|
| 0               | 60           | 105         | 20.6    |
| 1               | 62           | 120         | 18.1    |
| 2               | 65           | 122         | 17.2    |
| 3               | 67           | 125         | 17.0    |
| 5               | 80           | 130         | 14.5    |
| 10              | 87           | 135         | 11.4    |

3.2 Absorption and Apparent Porosity

The results of absorption and apparent porosity of mortar are shown in Fig. 4. The incorporation of 1.0 % rubber latex resulted in the reduced apparent porosity and absorption of mortar. The reduced porosity indicated the filling of pore with rubber latex. As the particles of rubber latex were between 50-100 nanometers. The cavity of paste is slightly larger at between 100 picometers to several hundred nanometers [23].

However, when the rubber latex additive was increased, the absorption and apparent porosity of mortar started to decrease. The optimum amount of rubber latex was 1.0% and the increase in the additive beyond this level resulted in the surplus
rubber latex in the matrix. The increased latex particles resulted in the increases in non-homogeneity, interface, and the gap between rubber latex and paste as shown in Fig. 5. The reduced absorption was one of the important factors that improved chemical absorption resistance ability of specimen[24]. The addition of 1% rubber latex decreased the absorption and porosity of specimen at the age of 28 days by 0.7% and 3%, respectively. The geopolymer mortar containing appropriate amount of rubber latex could enhance the absorption characteristics of mortar. In addition, the experiment also showed that the absorption improved with age due to the continued reaction of geopolymer and the forming of additional calcium silicate hydrate (C-S-H) and calcium aluminosilicate hydrate (C-A-S-H) and sodium aluminosilicate hydrate (N-A-S-H) in the geopolymer matrix[25]. When the age of specimen increased, the matrix of the sample became denser and the apparent porosity decreased by 9-16%.

3.3 Compressive Strength

The results of the compressive strength of geopolymer mortars are shown in Fig. 6. The optimum amount of latex was 1% and this produced mortar with increased compressive strength. The addition of latex beyond this level resulted in a decrease in compressive strength. A small amount of latex could fill the pore of matrix and make the mortar denser. The filling of pore at this level reduced the void in the matrix and slightly increased the compressive strength. The addition of more latex started to reduce the compressive strength due to increases in rubber particle, interface, and gap between rubber particle and paste. The compressive strength results agreed with the porosity which showed that the lowest porosity (optimum) was with the incorporation of 1% NRL. The reduction in the mechanical properties with increased NRL content complies with other reported results on the use of elastomer[16, 19]. Under the compression loading, the additional rubber latex and interface resulted in additional internal tensile strain resulting in a decrease in the compressive strength of mortar. The 7-day compressive strength of 1% latex sample was optimum at 30.0 MPa and this reduced to 7.0 MPa with the incorporation of 10% latex.

3.4 Flexural Strength

The results of flexural strength of geopolymer mortars are shown in Fig. 7. At the age of 7 days, the flexural strengths of specimens containing 1-3% NRL were approximately the same with that without NRL. The good performance of samples with a small amount of NRL followed similar trend to that of the compressive strength. However, at the age of 28 days, the increase in the amount of NRL reduced the flexural strength of sample. The incorporation of rubber latex seemed to have an adverse effect on the flexural strength of the mortar particularly for the samples with high strength normally obtained with advanced age. In this test,
there is an indication here that the benefit of incorporation of a small amount of NRL on the flexural strength was more pronounced at the low strength level, i.e. at the early age of sample of 7 days.

Fig. 7 Flexural strength of NRL modified geopolymer mortar at 7 and 28 days

3.5 Surface Abrasion Resistance

The result of abrasion resistance as shown in Fig. 8 follows the same trend as that of compressive strength. The surface abrasion resistance has been shown to relate to the compressive strength of samples [26]. In this research, the optimum NRL content of 1% resulted in the lowest surface abrasion weight loss which corresponded to the highest compressive strength. The optimum amount of NRL addition resulted in the filling of pore of geopolymer paste and thus enhanced the properties of the mixture. The presence of NRL has also been shown to increase the absorption of impact and the impact resistance of samples [27]. The better characteristics of absorbing energy rendered the sample with increased resistance to the abrasion action of the rotation and impact of rotating cutters and decreased the surface abrasion weight loss as shown in Fig. 8. As already pointed out that the increase in the NRL resulted in the non-homogeneity, interface, and gap between rubber latex and paste. In addition, the incorporation of more than 1% NRL resulted in surplus NRL and reduced amount of paste for the basic strength unit as it reduced the amount of fly ash in the system. In some cases, the surplus NRL was prone to adhere to each other and this larger particle caused the weakness in the matrix. The results thus confirm that resistance to the surface abrasion is related to the strength of materials as indicated by the direct relation to the compressive and tensile strength of the samples.

4. CONCLUSIONS

Based on experiment result, the following conclusion can be made:

(1) The incorporation of natural rubber latex (NRL) increased the setting time of geopolymer mortar due to the increase in the water content of the system. The flow of mortar decreased with increasing NRL content as the NRL was a high viscosity fluid and thus increased the viscosity of system.

Fig. 8 Surface abrasion weight loss of rubber modified geopolymer mortar at 28 days

(2) The incorporation of 1% of NRL was optimum for the geopolymer mortar system to obtained reductions in apparent porosity and absorption of specimens. This would have a positive effect on the durability of the samples as the decreased apparent porosity and low absorption would lower the chemical absorption of sample.

(3) The compressive, and flexural strength tests confirmed that the optimum amount of NRL addition was 1%. This produced mortar with increased strengths due to the filling of pore and associated improved mechanical properties. The incorporation of NRL beyond this level resulted in the surplus NRL and reduction the strengths of mortar was observed. The surplus NRL resulted in increases in non-homogeneity, interface, and gap between rubber latex and paste. In addition, the amount of geopolymer gel was also reduced as the amount of rubber was increased.

(4) The results also confirmed that the resistance to surface abrasion was highest with the incorporation of optimum amount of NRL of 1%, which resulted in the lowest surface abrasion loss.

Overall, the results showed that NRL could be used as additive in the high-calcium geopolymer with optimum content of 1% by weight of fly ash. The geopolymer mortar with 1% NRL showed improved properties with increased in strengths (compressive, and flexural strengths), increased resistance to surface abrasion, and reduced porosity and absorption.
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