Effect of utilizing geopolymer fly ash on potential and corrosion rate of reinforcement in high-strength concrete

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Abstract. With the increasing use of high-strength reinforced concretes as a structural element of construction, their resistance to corrosion in environmental conditions needs to be studied. It is well known that cement manufacturing at plants is un-environmentally friendly due to plenty of fuel consumption and \(\text{CO}_2\) emission to the atmosphere leading to greenhouse effect. Therefore, substituting cement with environmentally friendly natural materials and natural wastes become a smart solution. This study aims to measure the potential level and corrosion rate of reinforcing steel in high-strength concrete with variations in cement replacement by geopolymer fly ash, environmental conditions, and concrete covering thickness. Corrosion potential measurement was carried out with the half-cell potential mapping method, while the corrosion rate used the linear polarization resistance method. Data collection was executed every two weeks during eight weeks of immersion. High-strength concrete beams with compressive strength target 70 MPa having dimension 15cm x 20cm x 80cm were substituted with 15% coal fly ash (CFA), 15% palm oil blast furnace slag fly ash (POSFA), and 10% pozzolanic fly ash (PFA) from cement weight. High-strength concrete specimens without fly ash substitution were made as a comparison (HSCWFA). It was installed 4 longitudinal reinforcement Ø10 mm and 7 stirrups Ø6 mm with a distance of 10 cm. The concrete covering thickness was 2 cm and 4 cm. Immersion medium uses artificial seawater (3.5% \(\text{NaCl}\)) and shallow well water. The results showed that specimens resulting in the lowest corrosion rate were PFA, CFA, POSFA, and HSCWFA respectively. Corrosion potential was obtained at low to medium levels, while corrosion rates were at low corrosion level criteria. It can be clearly concluded that the utilizing of a geopolymer fly ash could increase the permeability of high-strength concretes through their higher resistance to the penetration of chloride ions.

1. Introduction

High-strength concrete is made with a low cement water ratio requiring a large amount of cement besides needing additives and admixtures with the best quality aggregates. As a result, a dense structure with low porosity and permeability as well as high durability is physically obtained. Mechanically, a structure having high carrying capacity and serviceability is gained which is very suitable for use in advanced and complex infrastructures. It is well known that the cement production process in the factory is not environmentally friendly because it requires many raw natural materials in the form of limestone and clay containing a lot of silica oxide, alumina oxide, and iron oxide. These raw materials must be refined and heated to a temperature of 1500\(^\circ\)C until a new mixture is formed called clinker, so it will need high fuel. To produce 1 ton of cement, 1.7 tons of raw material are needed and 1 ton of CO and CO\(_2\) gas is released into the atmosphere which can cause the greenhouse effect as a trigger for global warming [1].
Based on the fact, efforts to reduce the high amount of cement in high-strength concrete production are necessary. One of them is using geopolymer fly ash from volcanic eruption material such as pozzolanic ash and industrial waste materials like coal fly ash and palm oil blast furnace slag ash. Such a geopolymer can be used as a cement replacement because it also contains cementitious materials such as SiO$_2$, Al$_2$O$_3$, Fe$_2$O$_3$, and CaO.

Aulia et al. [1] and Aulia et al. [2] state that the substitution of coal fly ash 15%, palm oil blast furnace slag fly ash 15%, and pozzolanic fly ash 10% of cement weight can still produce good shear capacity and bending capacity comparing with high-strength reinforced concrete beams without fly ash substitution. Further, the use of a combination of natural nano fly ash and nano lime with strength target $f'_c$ 35 MPa and w/c ratio 0.49 generated higher compressive strength at 7 days compared to plain specimens without nano fly ash and nano lime [3].

Nowadays, investigation of the corrosion resistance and mechanism of reinforcement in reinforced concrete subjected to corrosive environments has been widely investigated. Several factors are affecting the corrosion potential of reinforcement in reinforced concrete, namely concrete moisture content, water to cement ratio, concrete cover thickness, as well as chloride content [4]. The effect of water to cement ratio variations and the type of cement on the electrical resistance of unsaturated normal concrete has been reported in [5] and the results showed that the water to cement ratio did not affect the concrete electrical resistivity value when using the same type of cement, but, on the other hand, the cement type had a significant effect, where the type of cement having additional blast furnace slag and pozzolan produced higher electrical resistance.

The measurement of corrosion potential of reinforcement and its possibility in reinforced concrete using half-cell potential mapping method has also been widely carried out as reported in [6], and it was concluded that the method can be used as an indicator to detect rebar corrosion initiation [7]. Based on the measurement of corrosion potential using half-cell potential method in concrete incorporating geopolymer fly ash, it was concluded that geopolymer concrete generated higher electrical resistance and yielded, therefore, better corrosion performance than corresponding concrete without fly ash [8]. Furthermore, measuring the corrosion rate using the linear polarization resistance (LPR) method has been conducted in [9], whose results showed that LPR could identify the initial phases of concrete structures degradation subjected to aggressive environments containing chloride ions. The reinforcement corrosion rate of geopolymer concrete was reported lower than that of cement-based concrete. High alkalinity in geopolymer concrete and strong silicate membranes found in geopolymer concrete could provide a passive layer on reinforcement [10]. According to Zainal et al. [11], better corrosion resistance and low corrosion rate were produced by geopolymer fly ash concrete added with slag (fly ash-slag geopolymer).

High-strength concretes are characterized by a dense structure leading to low porosity and high durability. These characteristics contribute obviously to better reinforcement corrosion resistance. However, the behavior of high-strength concretes against the corrosion resistance with the use of geopolymer fly ash as cement replacement needs to be thoroughly investigated. Therefore, this research aims to obtain the potential level and corrosion rate of reinforcement in high-strength concretes containing geopolymer fly ash, i.e., 15% coal fly ash, 15% palm oil blast furnace slag fly ash, and 10% pozzolanic fly ash from cement weight with variations in environmental conditions and concrete covering thickness. The corrosion potential level was measured with the half-cell potential mapping method, while the corrosion rate was measured with the linear polarization resistance method. Data collection was taken by soaking high-strength concrete beams with compressive strength target 70 MPa having dimension 15cm x 20cm x 80cm every two weeks during eight weeks of immersion using artificial seawater (3.5% NaCl) and shallow well water. High-strength concrete specimens without fly ash substitution were made as a comparison. Four longitudinal reinforcement Ø10 mm and seven stirrups Ø6 mm with a distance of 10 cm was embedded in the beams, with concrete covering thickness 2 cm and 4 cm.
2. Methodology

2.1. Materials

The used coal flyash emanated from electric steam power plant Nagan Raya, Aceh Province, with the chemical composition content as follows: \( \text{SiO}_2 = 26.65\% \), \( \text{Al}_2\text{O}_3 = 9.6\% \), \( \text{Fe}_2\text{O}_3 = 17.56\% \), \( \text{SO}_3 = 2.51\% \) and \( \text{MgO} = 1.74\% \) [1-2]. The amount of \( \text{SiO}_2 \), \( \text{Al}_2\text{O}_3 \), and \( \text{Fe}_2\text{O}_3 \) of coal flyash is 53.81\%, which according to ASTM C618 [12], it is classified into Class C flyash with a minimum content about 50\% [12]. The used palm oil blast furnace slag possessed the composition of chemical content as follows: \( \text{SiO}_2 = 34.11\% \), \( \text{Al}_2\text{O}_3 = 3.57\% \), \( \text{Fe}_2\text{O}_3 = 2.06\% \), \( \text{SO}_2 = 0.2\% \), and \( \text{MgO} = 1.95\% \) [1]. Furthermore, the pozzolanic flyash used in this research originated from Beurandeh Village, Regency of Aceh Besar which have composition of chemical content as follows: \( \text{SiO}_2 = 42.96\% \), \( \text{Fe}_2\text{O}_3 = 1.92\% \), \( \text{CaO} = 0.42\% \), \( \text{MgO} = 0.28\% \), \( \text{Na}_2\text{O} = 0.13\% \), \( \text{K}_2\text{O} = 0.36\% \), and \( \text{TiO}_2 = 0.12\% \) [1].

These fly ashes were utilized as cement replacement in the amount of 15\% for coal fly ash, 15\% for palm oil blast furnace slag ash, as well as 10\% for pozzolanic fly ash, all from cement weight, to produce high-strength reinforced concrete (HSRC) beams. The replacement percentage was taken based on the optimum value from previous research [2]. Plain beams without fly ash were cast and tested as a comparison.

HSRC beams having a dimension of 15 cm x 20 cm x 80 cm were cast with a w/c-ratio 0.30 consisting of 600 kg/m\(^3\) Portland cement type I using polycarboxylate ether-based superplasticizer ViscoCrete-10 with an amount of 1.5\% from cement weight. Crushing stones with a maximum diameter of 4.76 mm (2-5 mm) were used as fine aggregates and with a maximum diameter of 12 mm (5-8 and 8-12mm) were used as coarse aggregates. Fine aggregate with a maximum diameter of 2 mm (0-2 mm) used was river sand. The concrete mixture was designed to have a compressive strength of 70 MPa.

Testing of potential and corrosion rate of reinforcement was carried out with a variation of the concrete covering thickness, i.e., 2 cm and 4 cm, and immersion medium, i.e., artificial seawater (using 3.5\% NaCl) and shallow well water. The total amount of specimens was 16 with each treatment had one specimen. Reinforcements were embedded in the beams comprising of 4 D 10 mm as longitudinal reinforcement as well as 7 pieces Ø6 mm – 100 mm for stirrups. The specimen dimension and reinforcement arrangement were shown in figure 1. Prior to testing of potential and corrosion rate of reinforcement embedded in HSRC beams, the specimens were beforehand cured with immersion in water for 28 days to achieve the designed compressive strength.

![Figure 1. Specimen dimension and setup.](image)

2.2. Instrument and testing

The main equipments used in the testing of potential and corrosion rate of reinforcement in high-strength reinforced concrete beams include: profometer to map the location of reinforcement and concrete covering thickness; digital half-cell meter to measure potential corrosion value of reinforcement in concrete with a non-destructive test method; potentiostat/galvanostat; digital multimeter; reference electrode; as well as counter electrode.
Testing of resistance and corrosion behavior of the reinforcement was preceded by mapping the reinforcement in concrete beams with drawing grids on the specimen surface, which was carried out on two sides, namely side A (top) and side B (bottom). Each meeting point of the longitudinal reinforcement and stirrups of the mapping was numbered, point 1 to 14 on side A, and point 15 to 28 on side B, as shown in figure 2.

Data of reinforcement corrosion potential values were taken on the concrete surface using Digital Half-Cell Potential Meter (PC1018). The method used was according to ASTM C 876 standards. Firstly, the excessed reinforcement tip was cleaned with sandpaper. Then the connecting cable from the reference electrode was linked to the reinforcement. After the number on the Digital Half-Cell device was stabilized (close to zero), this instrument was ready for use by attaching the tip of the reference electrode to the concrete surface vertically. The number that came out from the instrument was recorded as corrosion potential value data up to three times, to then be estimated so that the data obtained was more accurate. The set-up of the corrosion potential value measurement is shown in figure 2. The criteria for reinforcement corrosion potential values using the half-cell potential mapping technique for several half-cell standards can be seen in table 1 [13].

| No | Cu/CuSO₄ (CSE) | Ag/AgCl | Hydrogen Standard (SHE) | Calomel (SCE) | Corrosion Condition |
|----|----------------|---------|-------------------------|---------------|---------------------|
| 1  | >-200 mV       | >-106 mV| >+ 116 mV               | >-126 mV      | Low (10% Corrosion Risk) |
| 2  | -200 to -350 mV| -106 to -256 mV | +116 to -34 mV | -126 to -276 mV | Medium Corrosion Risk |
| 3  | < -35 mV       | < -256 mV| < -34 mV                | < -276 mV     | High (<90% Corrosion Risk) |
| 4  | < -500 mV      | < -406 mV| < -184 mV               | < -426 mV     | Very High Corrosion Risk |

The measurement of the corrosion rate by LPR (linear polarization resistance) method was executed by placing the measuring instrument on the outer surface of the reinforced concrete beam specimen. The required data were the potential value ($E$) and current ($I$) which was controlled using potentiostat/galvanostat, as well as the value of the anodic and cathodic corrosion current density ($i$) as the result of the calculation between the current and the cross-sectional area of measurement. During
measuring, all sets of equipment must be in a state of quiet and balance. Measurement with a swaying circuit will produce incorrect measurement data.

The measurement mechanism using potentiostat/galvanostat was initiated by the entry of the current \((I)\) given by the potentiostat/galvanostat to the concrete through the counter electrode. The current that entered into the concrete will then be polarized. The potential of resulting polarization of reinforcement will be measured by the reference electrode to produce potential data \((E)\) in the form of anodic potential and cathodic potential. The device used to measure the corrosion rate consisted of a series of instruments, which can be seen in figure 3.

After reinforcement mapping on the concrete surface, the potential value and its corrosion rate were measured at initial conditions (Week 0). Measurements were made at each numbering point. The next step was to immerse the specimens in the medium of artificial seawater (NaCl 3.5%) and shallow well water. The subsequent measurement of the potential value and corrosion rate was carried out every two weeks for eight weeks of immersion. In the second week onwards, the test specimens were removed from the immersion tub and aerated for 24 hours before measurement. This was carried out so that the reading of potential values was more stable. The concentration of NaCl solution in the immersion media of artificial seawater was examined every 2 weeks using buemeter. If the concentration changes due to the effect of evaporation, the water or NaCl was added until the concentration returned 3.5%.

The potential data of the resulting measurement, the supplied current, and density of the resulting calculation were written in tabular form for each column point of the resulting measurement. Then a polarization curve was created based on data from the table of resulting measurement. This curve was plotted with the x-axis as the current density value \((i)\) and the y-axis as the measured potential value. Further, the polarization curve was lined up to get a slope pattern and determine the straight line. By using the straight-line equation of the curve, it can be determined the corrosion potential value in the anodic region by entering the desired current density value, as well as the corrosion potential value and the current density in the cathodic region. For the accuracy of the corrosion rate calculation, the measured reinforcement corrosion potential value was in the range of 10 to 20 mV in the polarization curve.

The amount of polarization resistance \((Rp)\) was calculated using the following equation [14]:

\[
Rp = \frac{\Delta E}{\Delta i_{app}}
\]  

(1)

\(Rp\) = polarization resistance \((\Omega \cdot \text{m}^2)\),
\(\Delta E\) = potential change \((\text{Volt})\), and
\(\Delta i_{app}\) = difference in measured current \((\text{A/m}^2)\).

Calculating the corrosion current density \((i_{corr})\) can use the following equation:

\[
i_{corr} = \frac{B}{R_p}
\]

(2)

where \(B\) is a constant value determined from anodic and cathodic table slope with a value of 0.074 V.

The value of the corrosion rate \((CR)\) can be determined based on Faraday’s law.

\[
CR = i_{corr} \times K \frac{a}{nD} \text{ (mpy)}
\]

(3)

where \(CR\) is corrosion rate \((\text{mpy})\), \(i_{corr}\) is corrosion current density \((\mu\text{A/cm}^2)\), \(K\) is a constant \((0.129 \text{ mpy})\), \(a\) is reinforcing steel atomic weight \((55.8)\), \(i\) is density of corroded iron current \((1 \mu\text{A/cm}^2)\), \(n\) is number of electrons lost during the corrosion process for iron \((2)\), and \(D\) is density \((7.86 \text{ g/cm}^3)\).
3. Results and discussion

3.1. Measurement of corrosion potential
Test results on deep well and shallow well water sources at the Structural and Material Laboratory of Syiah Kuala University, Banda Aceh, showed that both water sources contained negligible corrosive materials, as shown in table 2. The pH value of water was also within the limit that could not damage the passive layer of reinforcement, which was around 7.00 so that the two well water could be used as water for making NaCl solution by adding a 3.5% chloride compound.

Table 2. The examination results of the water chemical content.

| No. | Testing Parameters | Testing Method | Standard Value | Deep Well (mg/L) | Shallow Well (mg/L) |
|-----|-------------------|----------------|----------------|------------------|-------------------|
| 1   | Dissolved Oxygen  | Electrical Conductivity | Probe In-situ | -                | 8.5 | 8.3 |
| 2   |                   |                |                | 1017             | 661 |
| 3   | Sulfate (SO4)     | Gravimetric    | 400            | 1.922            | 3.517 |
| 4   | Chloride (Cl)     | Titrimetric    | 500            | 99.4             | 49.98 |
| 5   | pH                | Probe          | 4.5-8.5        | 7                | 7.03 |

Based on the measurement results of corrosion potential, locations with the most negative potential values on the specimen surface are obtained. This location is the area with the highest corrosion possibility (top and bottom) as shown in table 3. The corrosion potential values for both beam surfaces are given in table 4.

According to table 3, it can be seen that the highest (negative) corrosion potential value was found in HSCWFA, followed then by POSFA, CFA, and PFA. This value was obtained for corrosion potential at the end of immersion, i.e., week 8 for all variations and treatments. In accordance with table 1, the corrosion risk criteria for all high-strength concrete beams using geopolymer fly ash with the variation of concrete covering thickness and immersion medium are at a medium corrosion risk (-126 to -276 mV).

Table 3. The results of mapping the highest potential location (negative) on each surface of the specimen.

| Concrete Covering Thickness | HSCWFA | CFA | PFA | POSFA |
|----------------------------|--------|-----|-----|-------|
|                            | Well Water | NaCl 3.5% | Well Water | NaCl 3.5% | Well Water | NaCl 3.5% | Well Water | NaCl 3.5% |
| 2 Cm                       | Point | A.3 | A.1 | A.2 | A.4 | A.13 | A.14 | A.5 | A.1 |
|                            | Point | B.15 | B.15 | B.17 | B.22 | B.28 | B.24 | B.28 | B.15 |
| 4 Cm                       | Point | A.1 | A.3 | A.8 | A.2 | A.2 | A.8 | A.9 | A.8 |
|                            | Point | B.20 | B.16 | B.21 | B.19 | B.21 | B.28 | B.16 | B.22 |

Table 4. The results of the measurement of the highest potential value (negative) on each surface of the specimen.

| Concrete Covering Thickness | HSCWFA | CFA | PFA | POSFA |
|-----------------------------|--------|-----|-----|-------|
|                             | Well Water | NaCl 3.5% | Well Water | NaCl 3.5% | Well Water | NaCl 3.5% | Well Water | NaCl 3.5% |
| 2 Cm                        | -110 to -230 | -106 to -279 | -111 to -183 | -109 to -230 | -89 to -147 | -105 to -220 | -106 to -208 | -102 to -245 |
|                             | -108 to -257 | -79 to -295 | -83 to -181 | -88 to -234 | -77 to -146 | -82 to -248 | -95 to -203 | -64 to -245 |
| 4 Cm                        | -111 to -176 | -111 to -190 | -89 to -166 | -99 to -196 | -94 to -121 | -97 to -163 | -115 to -151 | -108 to -172 |
|                             | -92 to -151 | -83 to 192  | -77 to -161 | -79 to -195 | -75 to -121 | -81 to -168 | -76 to -156 | -84 to -177 |
The difference in concrete covering thickness of 2 cm and 4 cm affects the measurement results of the reinforcement potential values at all test specimens. From table 3 it can be seen that the corrosion potential values of each beam tend to be more positive (lower) on the concrete covering thickness of 4 cm. This shows the ability of concrete as a conductor is becoming less good for thicker layers, where the resistance or barriers to the electric current will be even greater. In addition, the diffusion of chloride ions in the immersion medium using 3.5% NaCl became smaller on larger concrete covering thickness.

Corrosion potential values for immersion treatment in well water and artificial seawater (3.5% NaCl) in general showed the same trend, but the high-strength concrete beams immersed in water with 3.5% NaCl produced a more negative value (high) as shown in figure 4. High-strength concrete substituted with pozzolanic fly ash with 2 cm concrete cover immersed in water with 3.5% NaCl resulted in a fluctuating corrosion potential values at each measurement point, but with 4 cm concrete cover it was returned almost uniform. Cement replacement with coal fly ash, pozzolanic fly ash, as well as palm oil blast furnace slag fly ash could reduce the corrosion risk at high-strength concrete. This was proved by the corrosion potential values which were increasingly positive (decrease) compared with high-strength concretes without using additives.

![Figure 4. Comparison of corrosion potential values at week 8 immersed in well water (left); and using artificial sea water (NaCl 3.5%) (right).](image)

3.2. Measurement of corrosion rate
As the results of corrosion potential measurements on high-strength concrete beams, corrosion rate measurement results also showed the same tendency, as shown in table 5. The same high-strength concrete with the same concrete covering thickness, but using different immersion media, produced different effects on the corrosion rate of reinforcement. Immersion in artificial seawater (3.5% NaCl) resulted in higher corrosion rate values when compared to the immersion in well water. Concrete covering thickness of 2 cm also yielded a greater corrosion rate than a 4 cm concrete cover. The thicker the concrete covering thickness, the smaller the permeability of the concrete, so that penetration of corroding elements such as oxygen, carbon dioxide, water, and chloride ions entering the concrete through the pores contained in the concrete cover will be less.

| Table 5. Calculation of the corrosion rate for all specimens with all variations in week 0 to week 8. |
|---------------------------------------------------------------|
| Corrosion Rate Values of Concrete Having Covering Thickness 2 cm with Immersion in Well Water (µm/y) |
| No. | Specimen | Week-0 | Week-2 | Week-4 | Week-6 | Week-8 |
|-----|----------|--------|--------|--------|--------|--------|
| 1   | HSCWFA   | 2.20935| 2.44881| 2.64384| 2.66942| 2.76232|
This is consistent with the findings by Law et al. [14] who found that the electrical resistivity of concrete improved with increasing concrete strength and concrete covering thickness. Concrete electrical resistivity served to reduce the corrosion rate by inhibiting the electric current in the concrete due to electrochemical corrosion.

According to Neville [15], one of the causes of corrosion in concrete is caused by the bond between Mg and SO$_4$ forming Magnesium Sulfate (MgSO$_4$) which reacts with Calcium Hydroxide Ca(OH)$_2$ in concrete. These sulfate compounds attack free Ca(OH)$_2$ resulting from the hydration process in cement. According to the results of the chemical examination of the pozzolanic fly ash, coal fly ash and palm oil blast furnace slag fly ash, as explained in Section 2.1, it appeared that Mg compound was more contained in the palm oil blast furnace slag fly ash than in coal fly ash and pozzolanic fly ash, this is assumed to cause high-strength concretes with coal fly ash and especially with pozzolanic fly ash having lower corrosion rate than high-strength concrete using palm oil blast furnace slag fly ash.

4. Conclusion
Substituting cement with environmentally friendly geopolymer materials and natural wastes, i.e., coal fly ash, palm oil blast furnace slag fly ash, and pozzolanic fly ash, could increase the corrosion resistance of high-strength reinforced concrete beams in the form of corrosion potential level and corrosion rate. The lowest corrosion potential values and corrosion rate were given by PFA, CFA, POSFA, and HSCWFA respectively. Corrosion potential level was obtained at low to medium levels, while corrosion rates were at low corrosion level criteria. In terms of concrete covering thickness, the thicker the covering thickness, the smaller the permeability of the concrete, so that penetration of corroding elements in the form of oxygen, carbon dioxide, water, and chloride ions entering the concrete through the pores contained in the concrete cover will be less. The electrical resistivity of concrete improved
with applying high-strength concretes and increasing concrete covering thickness. Concrete electrical resistivity served to reduce the corrosion rate by inhibiting the electric current in the concrete due to electrochemical corrosion.

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