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A literature review on the durability of ferrous and non-ferrous geopolymer concrete for sustainable developments

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Abstract. The usage of cement in concrete caused the release of CO_2 emission which can cause global warming. Moreover, cement as main ingredient can result in depletion or even loss of limestone which serves as a water supplier for farmers. Geopolymer concrete does not use cement but industrial byproducts containing aluminate (Al) and silicate (Si) such as slag, fly ash, and metakaolin. Moreover, these materials are from industrial solid waste (slag) so that it becomes an added value for construction. This paper discusses the durability of geopolymer concrete with material derived from the ferrous and non-ferrous melting processes because of the lack of study in durability. In construction, the concrete durability is the main thing that must be considered. Durability analysis aims to make the concrete last a long time, especially in aggressive environments. It refers to high concentrated CO_2 level in an urban area or marine environment that has sodium chloride and sodium sulfate levels. Increasing of non-ferrous content can lower total pore volume in geopolymer binder while the macropores are formed. High-calcium ferrous content addition has pore-enhancing effect. The lower the porosity, the lower the permeability in the concrete. It lowers the water content and compounds such as chloride absorbed in concrete. Chloride and CO2 diffusion can cause carbonation in concrete leading to corrosion in steel reinforcement and shrinkage. This literature study aims to enhance knowledge in concrete durability so building construction can last as the planned period. Hence, the utilization of natural resources as construction material can be more efficient.

1. Introduction

Infrastructure development aimed to increase investment in efforts to increase the productivity of a country's economy. This has led to an increase in the amount of infrastructure in Indonesia. However, infrastructure development has several negative impacts on the environment. Building construction increased demand for concrete production. Concrete is the most commonly used construction material and is the second most used after water [1]. One of the materials in concrete that has negative effect for the environment is cement. Cement production using excessive limestone can cause damage to an environmental ecosystem in an area. Depletion or even disappearance of limestone mountains in an area has a big influence. Limestone mountain has been a supplier of water to farmers. The quantity of water will be decreased along with the thinning of limestone. Also, liquid waste from cement factories can affect water quality even worse resulting in the decreasing of soil fertility. The declining quality and quantity of water are detrimental to residents, especially farmers. Another impact of the cement industry is air pollution which caused around 5% of carbon dioxide (CO_2) emissions [2]. As a result, damage to the ozone layer in the atmosphere can cause a greenhouse effect.

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One solution to these impacts is the use of geopolymer concrete which does not use cement. Cement replacement materials in geopolymer concrete are industrial byproducts containing aluminate (Al) and silicate (Si) such as slag, fly ash, and metakaolin which can be added value for construction. This literature study discusses the durability of geopolymer concrete with industrial byproducts in the form of slag. Slag is the residual product of the metal (ferrous) and non-metal (non-ferrous) smelting. Slag waste that accumulates around the smelter (directly or indirectly) can pollute and damage the environment. The Peraturan Pemerintah (PP) Nomor 101 Tahun 2014 also stated that B3 (Bahan Berbahaya dan Beracun) waste can be reused and recycled as a material substitution for human health. This is due to the B3 waste management process aimed to reduce or eliminate hazardous and toxic nature. Peraturan Pemerintah (PP) No. 101 Tahun 2014 also stated that slag waste including B3 waste can be reused and recycled with the aimed of turning it into a product that can be used as a substitute for raw materials, auxiliary materials, and fuels that are safe for human health and the environment. The purpose of this paper is to be able to design concrete so that it can last for a long time especially in aggressive environments using environmentally friendly materials. Durability is reviewed through porosity, permeability, and carbonation in geopolymer concrete in this paper. The method used in this paper is a literature review. The data obtained are the conclusions from the analysis of 37 papers. In this paper, the literature discusses the durability of geopolymer concrete.

2. Geopolymer concrete

Joseph Davidovits, a French scientist, introduced geopolymer concrete first in 1978. Geopolymer concrete is an environmentally friendly concrete because it does not use cement. Davidovits [3] stated that the chemical reaction of alumino-silicate oxides with alkali polysilates in geopolymer concrete produces a Si-O-Al polymer bond. The main components in geopolymer concrete are alkali activators and source binder materials. Alkaline activator is a combination of sodium silicate and sodium hydroxide which are important in the polymerization process. Then, the binder source material in geopolymer concrete contains high aluminate (Al) and silicate (Si). Aluminosilicate material can be in the form of industrial byproducts such as slag and fly ash. The binder is produced by activating the solid aluminosilicate source material with a highly alkaline activating solution and is assisted by a curing process [4-6]. The composition of the binder material is very important to consider in the design of geopolymer concrete.

In several studies, it was explained that the mixture of fly ash and high magnesium nickel slag (HMNS) that classified as non-ferrous and ground granulated blast furnace slag (GGBFS) classified as ferrous can improve mechanical properties and perfect pore structure in geopolymer concrete. This is caused by the combination of Ca^{++} and Mg^{++} into the geopolymer gel which produces a new crystalline phase. The combination of fly ash, HMNS, and GGBFS will produce C-S-H, A-S-H, and Na-Al-(Mg)-Si-H gel formations which leads to improved mechanical properties. Also, the activation of slag which forms C-A-S-H gel can improve pore structure with better filling properties [1,7,8]. The mixture between ferrous and non-ferrous is expected to be used as a new alternative source of cement material, thereby reducing the impact of air pollution.

Several factors influenced the characteristics of geopolymer concrete are the ration of alkaline solution with binders, molarity level of geopolymer concrete, and curing temperature. A ratio of alkaline solutions with binders by mass was suggested by Davidovits [9] between 0.30 and 0.45. Also, research Wongpa et al. [10] stated that the S / A ratio was the most important factor controlling the compressive strength rate of concrete. The S/A ratio is the ratio between the content of the solution (S) and the content of the binder used (A). Research Bernal et al. [11] stated that concrete formulated with an overall (S/A) ratio (activator + solids) of 0.44 shows the lowest pore volume. A lower S/A ratio causing an increase in porosity in the test specimen. In addition, the increase in activator modulus also showed a slight change in absorption values. A test was conducted in a research Huseien et al. [12] with varying molarity levels and it was found that the compressive strength results increased with the decreasing of water absorption. Test specimens were prepared with molarity levels of 6, 8, 10, 12, 14, and 16 M with two curing methods, then showed that the higher the molarity level, the lower the water absorption. Furthermore, it was reported that the reaction of complete polymerization reaction occurred in the temperature range between 40°C - 85°C [13]. Research Palomo et al. [14] stated that the mechanical strength of fly ash-based geopolymer concrete at curing temperature 85°C for 2 to 5 hours was higher than curing temperature at 65°C.

3. The durability of geopolymer concrete

The durability of concrete is the main issue that must be considered in construction. Durability is how the concrete can endure all circumstances and is designed so that the concrete experiences less damage during its lifetime. Concrete durability affected serviceability, design life, and safety [15]. It aimed to ensure that concrete in construction can last for 30, 50, or even 70 years. However, durability depends on the interaction between the concrete material and the surrounding environment. The aggressive environment greatly influences the durability of concrete. An example of an aggressive environment is an urban environment with high pollution containing high CO_2 concentrations. The reaction of carbon dioxide (CO_2) in concrete caused damage slowly starting from the outside then into the inside of the concrete. The entry of CO_2 into the concrete pore causes diffusion which will result in carbonation in the concrete. Another example of an aggressive environment is the marine environment or near the coast because seawater contains sodium chloride and sodium sulfate. Hydrochloric acid is a solution that can cause corrosion and porous concrete. Meanwhile, sulfuric acid is a mineral acid (inorganic) which can damage the structure of the paste in concrete, causing cracking on the concrete surface, and strength of concrete [16]. The diffusion of acid in the concrete pore triggers chloride penetration which is the main cause of corrosion in concrete structures in the marine environment. Research Sufian et al. [17] stated that geopolymer concrete is better than conventional concrete in terms of resistance to acid attack, has good ability when facing sulfate settlement, and is very stable when there is a high increase in the temperatures.

3.1. Porosity

Porosity is the pore content in concrete containing air, water, or a liquid which is interconnected and is called a concrete capillary. Concrete capillary will still exist even though the water used has evaporated. It will reduce the density of concrete produced [18]. One factor that can affect porosity is the diversity of grain size. Small granules can fill the space between larger grains in concrete pores. If there were more space in the concrete pore, the porosity value in the concrete will increase. It can cause the entry of air, water, or a solution into the concrete that takes place through the pores of the concrete. Air, water, and the solution that enters the pore will reduce the compressive strength of concrete. Therefore, porosity is the main thing that must be considered in the durability analysis of geopolymer concrete.

The pore size for ferrous and non-ferrous mixed geopolymers and cement concrete (OPC) as in research Yang et al. [8] was determined by the Mercury Intrusion Porosimetry (MIP) test using the Poremaster GT-60 to test the pore structure of geopolymer and cement samples. There are 2 types of pore size classification, mesopores and macropores. Mesopores are pore size less than 50 nm (<50 nm); macropores are pore size of 50 to 200 nm (50-200 nm). The mixture of materials in the concrete binder also affects the pore structure. In several studies, it was found that a mixture of ferrous and non-ferrous in geopolymer concrete can provide a pore improvement effect. Figure 1 shows the reduction in total porosity along with the addition of high magnesium nickel slag (HMNS) content in the mixture of fly ash-HMNS concrete. The smallest porosity level was found with the addition of HMNS by 60%, but more macropores (> 50 nm) were formed. Figure 2 shows the reduction in total porosity along with the addition of ground granulated blast furnace slag (GGBFS) content in the fly ash-GGBFS mixture concrete. There is a difference between the addition of HMNS and GGBFS. The more addition of GGBFS, the total pore volume in the geopolymer binder becomes lower and the less formed macropores (> 50 nm) [7,19]. Several studies explained that the addition of GGBFS which contains high calcium reduction in the effect of pore refinement. It occurred due to the presence of more bound water in the dominant C-(A)-S-H gel formed in the slag-rich geopolymer system that will cause more pore filling capacity for the binding pore structure. Capillary pores did not appear to exist in alkali-activated fly ash geopolymers that react well with slag addition [7,8,11,19]. Research Bouaissi et al. and Yang et al. [7,19]. formulated with an S/A ratio of 0.42 contributed to reduce the water content and improve the microstructure of the specimens.

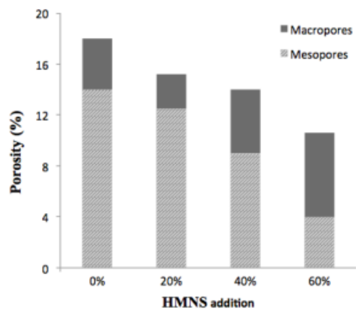

Figure 1. High Magnesium Nickel Slag (HMNS) addition [19].

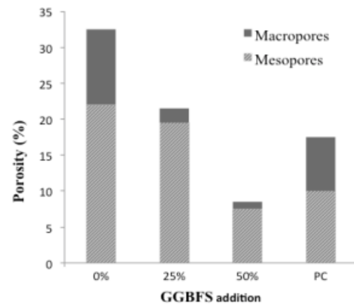

Figure 2. Ground Granulated Blast Furnace Slag (GGBFS) addition [8].

Total porosity and absorption levels were calculated according to ASTM C642-06 [13,15-17]. The relationship between total porosity and absorbency is linear. The smaller the total porosity, the less the absorbent. Figure 3, shows the addition of GGBFS by 50% that resulted in the optimum results in the absorption rate. Besides, the results were better than cement concrete (OPC). The absorption of geopolymer concrete decreases with the addition of GGBFS which varies according to the reaction of the improvement of the pore structure [8](Yang, Yao, and Zhang 2014b). In several studies, the determination of water absorption and porosity is determined through the volume of the permeable cavity (VPV) [17,19,20].

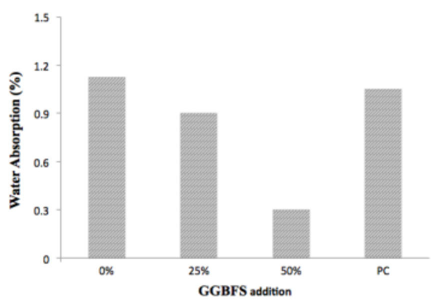

Figure 3. Geopolymer water absorption based on fly ash-slag mixture [8].

3.2. Permeability

Permeability is the presence of an empty space in concrete that is able to flow water/solution so that they are interconnected and have permeability properties. Permeability is an important component because in addition to its function to withstand the compressive stress, the tensile area of the concrete also serves to protect reinforcing steel from direct contact with outside air which can cause oxidation reactions and corrosion [18]. Permeability has a relationship with porosity. If the porosity value is small, then the permeability value and the amount of pore content in the concrete will be small as well. This circumstance allowed less water/chemical solution to enter into the concrete resulting a low absorption rate so that the quality of the concrete is relatively well. In addition, porosity also has a relationship with compressive strength. The research Wardhono at al. and Bradley [21,22] stated that the relationship between porosity and compressive strength of concrete is inversely proportional and if the value of porosity is low, the value of compressive strength is high. The water permeability coefficient tends to decrease continuously when the compressive strength increases. A possible reason is that the lower compressive strength of the geopolymer concrete mix contains a higher S/A ratio which results in fresh polymer paste being sticky and more difficult to print. The study Bernal et al. [11], showed that an increase in the S/A ratio in a 90-day sample gives a decrease in the value of k, which is related to total water absorption and total porosity of the test specimen. Figure 4 shows a water permeability tests. Concrete permeability can also be referred to as the coefficient of permeability (k), which is evaluated based on Darcy's law [18]:

$$\frac{1}{A}\frac{dq}{dt} = k\,\frac{dH}{L}$$

Remarks:

$\frac{dq}{dt}$ = The rate of water flow
A = Sectional area of the concrete sample
dH = High waterfall
L = Thickness of concrete samples
k = Coefficient of permeability

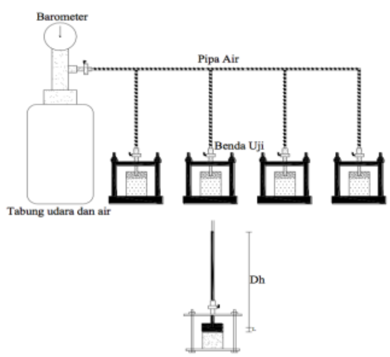

Figure 4. Water permeability testing.

In water permeability tests, both capillary absorption and applied pressure contribute to the water flow rate. Figure 5 showed a comparison between the permeability coefficient values of Alkali Activated Slag (AAS) and low calcium Fly Ash Geopolymer (FAGP) [21]. Concrete with slag material (AAS) showed an increase in the value of water permeability with age but is classified as water-permeable concrete at all ages because it does not exceed the maximum permeability value. Based on the ACI 301-729 standard, the maximum permeability value is 1.5xE-11 m/sec (4.8x10-11 ft/sec). Concrete with Fly Ash (FAGP) material has a water permeability value that is much higher than AAS for up to 90 days but decreases at 180 days and decreases with age until it has lower values than AAS concrete at 360 and 540 days. Therefore, it is expected that non-ferrous mixed geopolymer concrete (for example Fly Ash) and ferrous (GGBFS) can have good and stable permeability values with age.

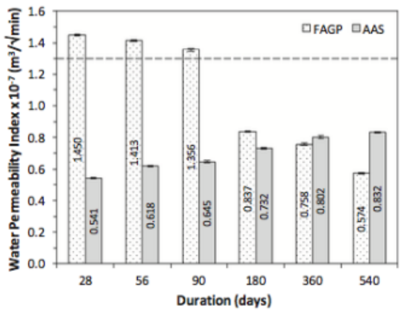

Figure 5. Water permeability index between FAGP and AAS [21].

3.3. Chloride migration

Chloride migration followed Nord Test Build 492 to find out the element of chloride that is soluble in water, the levels of chloride that can dissolve in acids, and the depth of the chloride that enters the concrete sample. Several studies were conducted to determine the relationship between Nord Test Build 492 and RCPT of concrete resistance to the entry of chloride ions. The procedures between Nord Test Build 492 and RCPT are different, but both test are designed to isolate and accelerate the transportation of chloride ion migration through concrete through diffusion [22]. Research Ismail et al. [23] stated that the ongoing microstructure reaction in a base binder slows chloride penetration. The Nord Test Build 492 method is considered to be more reliable as an effective way to assess the durability of an alkali-activated material. Table 1 showed that increasing levels of fly ash can increase chloride penetration at 90 days. It showed that the performance of geopolymer concrete is better in alkali-activated slags because the C-A-S-H binding gel dominates compared to fly ash geopolymer binders. The specimen is sprayed by AgNO3 to determine the depth of chloride penetration. Figure 6 shows the boundary of chloride penetration in concrete samples using the Nord Test Build 492. Figure. 6(A) is 100% granulated blast furnace slag (GBFS), 6(B) is a combination of 75% GBFS/25% fly ash, 6(C) is a combination of 50% GBFS/50% fly ash, and 6(D) is OPC. The correlation between the chloride diffusion coefficient with depth in the test object is directly proportional. Higher chloride penetrastion values can be seen by the number of the depth in the specimen that is getting bigger. There is no significant difference in parts A and B, but it has a smaller height than C and D. Geopolymer concrete mixes of fly ash and slag (A, B, and C) have higher values than OPC concrete (D) [23]. It indicated that geopolymer concrete has better resistance in chloride compared to OPC concrete.

Table 1. Chloride migration coefficients of 4 samples based on Nord Test Build 492 [23].

Sample (wt% slag/wt% fly ash)$_o$	Chloride migration coefficient (*Dp*) (x 10^{-12} m^2/s)	
	28 days	90 days
100/0	1.02 (0.75)	0.51 (0.18)
75/25	0.24 (0.21)	0.65 (0.21)
50/50	1.24 (0.29)	2.01 (1.20)
OPC	18.6 (4.1)	16.8 (0.1)

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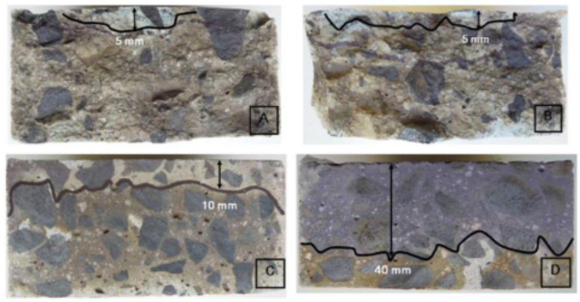

Figure 6. Geopolymer concrete chloride penetration in 28 curing days based on Nord Test Build 492 [23].

The research van Deventer et al. [24] also showed that the higher the compressive strength, the lower the chloride diffusion coefficient measured by the Nord Test Build 492 method. Geopolymer concrete will have better durability in seawater because the chloride diffusion coefficient is very low. Factors influencing the rate of chloride penetration are pore geometry, chemical reactions (long and short term), environmental conditions (temperature and humidity fluctuations), and repairs such as cracks [25]. The colorimetric method was used to identify the depth of the chloride ion in the concrete obtained by spraying silver nitrate on a cross-section of the split concrete. The chloride diffusion coefficient obtained from the ponding test and the colorimetric method is linearly correlated after the ponding test. The colorimetric method can provide a cheaper and easier way to get the diffusion coefficient [26].

3.4. Rapid Chloride Penetration Test (RCPT)

Dangerous conditions for concrete in seaside areas are the low tides conditions. Because they are exposed to high oxygen, chloride, and water. RCPT is a test to measure chloride content in concrete. Concrete that is exposed to chloride salts will experience diffusion which results in the concrete experiencing a decrease in strength. One mechanism of damage (deterioration) is the pressure of crystallization. At a high tide (rising water), seawater containing sodium chloride and sodium sulfate will enter the concrete pore. Then, at a low tide (low water) evaporation occurs and the chloride settles in the pore. The chloride penetration test aims to determine the chloride content in the concrete. It is due to chloride can damage the concrete structure slowly. The research Kushartomo et al. [27]. stated that the level of resistance of concrete is influenced by the ability to bind chloride because it is relatively powerless against chloride in the pore. The Rapid Chloride Permeability Test (RCPT) is based on the ASTM C1202 standard as shown in Figure 7.

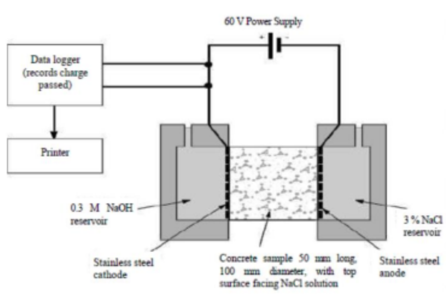

Figure 7. Rapid Chloride Permeability Test (RCPT) based on ASTM C1202.

Table 2. Criteria based on RCPT based on ASTM C1202.

Charge Passed (coulombs)	Chloride Ion Penetrability
>4000	High
2000-4000	Moderate
1000-2000	Low
100-1000	Very Low
<100	Negligible

The results of accelerated chloride testing can be categorized according to Table 2 which is reviewed through missed ions and chloride penetration in concrete. The charge that passes through reinforced concrete specimens is used to estimate the permeability of chloride ions according to ASTM C1202 [25,26,28,29]. Research Tennakoon et al. [29] stated that the chloride content of Ordinary Portland cement (OPC) concrete is higher than geopolymer concrete at all depths. Chloride penetration has little value if the porosity level in concrete is also small. Research Ma et al. [30], showed a lower chloride diffusion occurs when the pore structure is very dense. Figure 8 shows that the smaller the sorptivity value, Lesser charge passing through the specimen. The addition of 15% of RHA (Rice Husk Ash) can provide density that makes the pore perfect. However, The addition of 20% RHA causing the presence of high silica that made the polymerization process was inhibited and produced little pore in the microstructure [31]. Research Bernal et al. [11], stated that the addition of metakaolin to the specimen with an S/A ratio of 3.6 resulted in a reduction in the total charge passing through the specimen for 28 days by 40%. The addition of metakaolin is related to the reduced capillarity and characteristics of the binding microstructure formed by sodium-calcium (alumino) silicate hydrate (N-C-(A)-S-H). The presence of Na^+ in the pore will lead to counter-diffusion when the Cl ions are electrically driven through the pore network leading to an increase in the value of the charge passed.

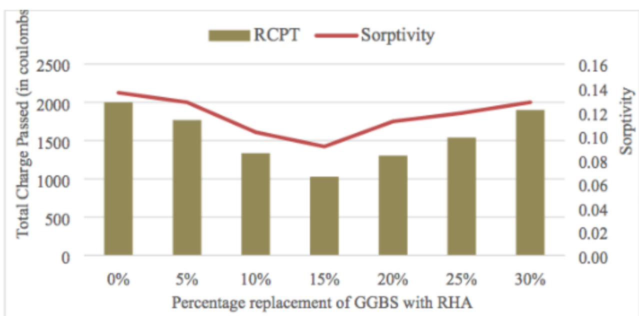

Figure 8. Chloride permeability and absorption of geopolymer concrete with RHA as a partial replacement for GGBS at 28 days [31].

The presence of a high concentrated pore chloride solution in the porous aluminosilicate gel system leaves salt deposits during drying [8]. Geopolymer concrete based on fly ash shows a smaller penetration value of chloride ions compared to OPC specimens. Research Kupwade-Patil and Allouche [15] showed that geopolymer concrete with higher calcium content can make it easier to react to forming calcium chloride ($CaCl_2$). The reaction of chloride with flying ash particles in the geopolymer matrix combined with high calcium causes the formation of Friedel salts. Furthermore, it causes dissolution of the chloride complex which contributes to damage to the passive layer at the rebar/binder interface and corrosion initiation. The reaction from the presence of chloride ions in the concrete structure can cause loss of the passive layer. The passive layer is a layer that protects the steel in concrete. The reinforcing steel in the concrete is corroded if the passive shield is lost i.e. the environmental pH in the concrete-steel contact area drops to <9.5. Based on accelerated corrosion test

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[13], geopolymer concrete has lower level of corrosion activity and longer time span than portland cement (OPC) concrete.

3.5. Carbonation

High pollution environments containing carbon dioxide in the atmosphere at certain humidity react with concrete binding materials. In the natural environment, concrete is exposed to an area with some pores, therefore it is easier for CO_2 to penetrate the concrete [32]. In the geopolymer system, low calcium fly ash can create the formation of sodium carbonate because of the exposure of CO_2. It can also cause extraction of Na^+ that can be found in geopolymer gels, alkali aluminosilicate gels, and formation of carbonate salts [17]. Research Pasupathy et al. [33], stated that on the surface of the geopolymer concrete, especially from the section of the culvert foot which is often in contact with saline lake water, that made it disappears and the aggregate is exposed on the surface. It happened because of the salt scaling when experiencing wet and dry cycles causing crystallization of salt on the concrete surface. The salt scaling process of geopolymer concrete is higher than OPC concrete, especially if geopolymer concrete contains excessive Na^+. Figure 9 shows that geopolymer concrete with Fly Ash material has higher ingress sulfate than cement concrete (OPC).

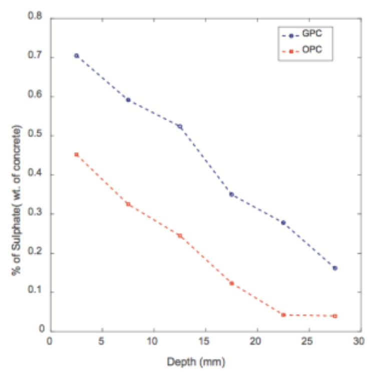

Figure 9. Sulphate content in GPC (fly-ash based) and OPC [33].

Research Bernal et al. [11], stated that porosity is not the only parameter that controls the loss of carbonated binder strength. The water-geopolymer solid ratio is the most influential parameter that affects the properties of geopolymer concrete [34]. It was explained in the study Zhuguo and Sha [35], that the carbonation resistance of geopolymer concrete is influenced by not only the level of density, but also the type of alkali activator, and pore characteristics. If the number and the size of the distribution pores have the same value, then the concrete will have a lower carbonation resistance.

The depth of carbonation in concrete will be seen by spraying phenolphthalein [13,23,33,34,36]. In addition, a change in color will occur and indicate the pH of the concrete. If the pH becomes more alkaline, the color will be more red. It can be more acidic or neutral but it will not cause a discoloration. The colorless zone (pH <9) is fully carbonated while the pink zone (pH> 9) is not carbonated. The transition between active and passive regions of iron is very close to the range where the phenolphthalein indicator begins to fade to colorless, and consequently, the results obtained using this indicator cannot be convincing about the alkalinity conditions of the system, and the potential for corrosion susceptibility to metal reinforcement [17]. Figures 10 shows the changes of color that occur after being sprayed with phenolphthalein.

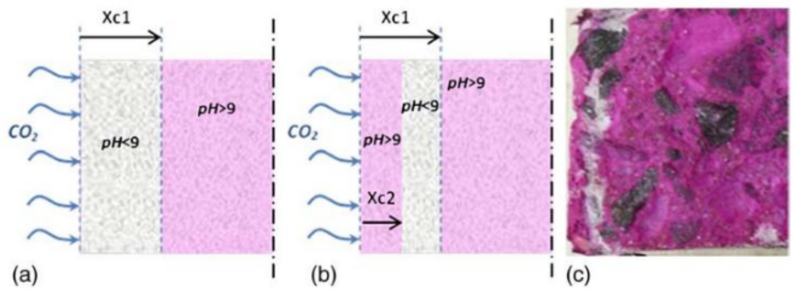

Figure 10. Schematic representation of carbonated parts [37].

4. Conclusion

This paper aims to determine the durability of geopolymer concrete in ferrous and non-ferrous mixtures. Durability analysis is reviewed through porosity, permeability, migration chloride, rapid chloride permeability test (RCPT), And accelerated carbonation testing, where all of it are interconnected. If the porosity value is small, then the permeability value and the amount of gas, water, or liquid such as chloride that enter the concrete will be small as well. The diffusion of chloride and CO_2 can cause carbonation in concrete which causes corrosion of steel reinforcement. Carbonation can eliminate the protective layer due to a decreased of the pH in the concrete. In result, the steel will be susceptible to corrosion which will slowly damage the concrete. Damage to the concrete is very detrimental to the structure of the building. Some studies stated that geopolymer concrete has comparable durability and mechanical properties even better than OPC concrete. Geopolymer concrete mixture that can be selected is the one with a composition of 50% non-ferrous, consisting of GGBFS with high CA + content and 20% -50% non-ferrous consisting of HMNS, Fly Ash, or RHA. The mixture of non-ferrous and ferrous materials is expected to have good resistance and as desired based on the data obtained from porosity, permeability, chloride migration, RCPT, and carbonation tests. Therefore, by doing an analysis of geopolymer concrete it is expected to be able to design a concrete that is suitable for a construction where it can withstand the design period. The damage of the concrete is expected to be less, so the maintenance cost of the building structures can be reduced.

Geopolymer concrete is environmentally friendly. Besides from using industrial waste materials, the process of geopolymer concrete does not require high energy as the process of cement concrete. In the process of cement concrete, heating is needed in the furnace to ± 1450°C to get clinker. The process of geopolymer concrete does not harm the environment. Geopolymer concrete is more efficient in terms of natural resources proven by its abundant amount. Therefore, the use of materials that utilize natural resources as building materials can be more efficient and not cause pollution to the surrounding environment.

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