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Strength and Durability of Concrete incorporating Natural Pozzolan in Aggressive Solomon Sea Environment

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ABSTRACT— *This paper reports the results of a comparative study on the performance of natural pozzolana (volcanic ash) based blended cement concrete in marine environment. Variables were volcanic ash (VA) content and curing environment. VA contents as cement replacement was varied from 0 to 35% and the samples were cured in the actual sea in two different conditions and in normal water. Volcanic ash concrete (VAC) showed better performance in terms of strength and durability (related to seawater ion diffusivity) compared to control normal concrete (NC) due to pozzolanic action of VA in marine condition.*

Keywords— Natural pozzolan, concrete, marine environment, strength, diffusivity, porosity

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1. INTRODUCTION

The search for alternative binders or cement replacement materials had been continued for the last decades. Research had been carried out on the use of volcanic ash (VA), fly ash (FA), blast furnace slag, rice husk ash, silica fume etc. as cement replacement material [1-8]. The VA and FA are pozzolanic materials, because of their reaction with lime (Calcium hydroxide) liberated during the hydration of cement. Amorphous silica present in the pozzolanic materials combines with lime and forms cementitious materials. These materials can also improve the durability of concrete and the rate of gain in strength and can also reduce the rate of liberation of heat, which is beneficial for mass concrete. ASTM Standards [9-11] exist for the use of natural pozzolans, fly ash, and silica fume and blast furnace slag in concrete. The optimum use of VA can improve the workability of concrete and can provide low cost concrete of satisfactory strength [12-14].

This paper presents the results of investigation on the performance of volcanic ash concrete (VAC) in marine environment where VA is used as replacement of cement. Durability of concrete is one of its most important properties and the lack of durability can be caused by external agents arising from the environment or by internal agents within the concrete. It is essential that the VAC should be capable of preserving its durability throughout the life of structures.

Although numerous researches had been conducted on the durability properties of blended cement concrete in the normal and marine environment. Little research has been done on the degradation of VAC subjected to aggressive environment especially in actual marine condition [12,13,15]. The corrosive action of seawater has been attributed mainly due to sulphate and chloride attack [16-17]. Concrete incorporating fly ash, blast furnace slag, lime stone filler and silica fume exhibited better chloride penetration resistance than ordinary Portland cement (OPC). In addition, such concretes also showed better performance in sulphate environment with significantly lower expansion and better strength retention than OPC concrete [16-21]. The use of chloride ion diffusivity in combination with compressive strength has been trialed in various projects for specifying concrete in marine environment. Australian Standard AS 3600 suggests compressive strength and corresponding concrete cover as the requirements and key properties as recent methods of specifying concrete for coastal and marine structures [16].

In this paper, the performance of VAC is compared with that of normal concrete (NC) based on compressive strength and durability (in terms of ion permeability and diffusivity) at different curing ages under two different seawater curing conditions in addition to normal water curing. The results of this investigation will be helpful for construction and manufacturing industries involved in the use volcanic material based concrete in structures subjected to extreme marine environment.

2. EXPERIMENTAL INVESTIGATION

The concrete mixes cast and tested in this experimental program were divided into VAC and control normal concrete (NC) mixes (with 0% VA). Table 1 summarizes the details of concrete mixes used in this study. Percentage of volcanic ash used as cement replacement in the mixes was varied from 0 to 35% by weight. The NC mix was designed for a 28-day targeted strength of 30 MPa. Water to cementitious material ratio was kept constant at 0.45 in all the mixes. The air content of the mixes varies from 2.3 to 3.1%.

Table 1: Details of concrete mixes

Mix	**Cement**	**Volcanic ash (VA)**	**Fine aggregate**	**Coarse aggregate**		**Water**
	kg/m^3		**Sand**	**10mm**	**20mm**	**kg/m^3**
			kg/m^3			
0VAC (NC)	320	0	800	270	645	144
5VAC*	16	304				
10VAC	32	288	* numeric represents the percentages of VA by weight in the VAC mixes			
15VAC	64	256				
20VAC	96	224				
25VAC	128	192				
30VAC	160	160				
35VAC	192	128				

2.1 Materials and properties

The volcanic ash (VA) used in this investigation were collected from the Rafael area in the East New Britain province of Papua New Guinea (PNG) and the source is a volcano called Mount Tavurvur. The cement used was locally manufactured Portland cement (PC) similar to ASTM type I.

Table 2: Chemical and physical properties of volcanic ash and Cement

	Chemical composition	
	Volcanic ash (VA)	**Portland cement (PC)**
Chemical Compound	**Percent**	**Percent**
Calcium oxide (CaO)	6.10	60-67
Silica (SiO_2)	59.32	17-25
Alumina ($Al2O_3$)	17.54	3-8
Iron oxide (Fe_2O_3)	7.06	0.5-6.0
Sulphur trioxide (SO_3)	0.71	1-3
Magnesia (MgO)	2.55	0.1-4.0
Sodium oxide (Na_2O)	3.80	0.5-1.3
Potassium oxide (K_2O)	2.03	0.5-1.3
Loss on ignition	1.03	1.22
Chloride	<0.01	--
Sulphate	0.08- 0.68	--
pH	5.4	--
Physical properties		
Fineness, m^2/kg	242	320

Chemical and physical properties of VA are compared with those from Portland cement in Table 2. Chemical analysis indicated that the VA has almost similar composition and are principally composed of silica (about 60%) while the main components of cement are calcium oxide (maximum 70%). However, VA has cementitious compounds like calcium oxide, alumina and iron oxide (total about 31%). The amount of oxides of sodium and potassium known as 'alkalis' is found to be higher in VA (5.83%) than that in cement (2.6% maximum). Higher alkali presence in the VA may have deleterious effects leading to disintegration of concrete due to reaction with some aggregate and affect the rate of gain in strength of cement. Quantitative XRD analysis of PC and blended cements with 20% VA (PVAC) as cement replacement, provided valuable information and the phase composition of these materials is presented in Table 3.

Table 3: Potential phase composition of the cementing materials from X-Ray diffraction

Phase	Portland cement (PC)	PVAC (PC blended with 20%VA)
C_3S	68.1	46.3
C_2S	14.1	9.5
C_3A	5.9	5.1
C_4AF	9.2	6.4
Other	2.4	5.2
Total	99.7	72.5
Glassy fraction*	0.3	27.5

* obtained by difference

Natural VA (fineness of 242 m^2/kg) is found to be much coarser than cement (fineness of 320 m^2/kg) which may lead to the increase of setting time. The coarse aggregate was 20 mm and 10 mm maximum size crushed stones having a bulk density of 2.5 and absorption of 0.5 percent. River sand having a specific gravity of 2.65 and absorption of 0.6 percent was used as fine aggregate.

2.2 Specimen casting, curing and testing

Standard concrete cylinders (150 x 300 mm) were cast. The concrete constituents were mixed in a revolving-drum type mixer for ten minutes. The specimens were demoulded after 24 hours of casting and cured in three different curing conditions: normal potable and seawater curing.

The normal water curing of the specimens was performed in laboratory curing tanks. For seawater curing, the specimens were subjected to actual sea conditions. The selected site was the shoreline of the Solomon Sea and situated within the protected area of PNG Halla cement factory in the city of Lae (Fig.1).

Figure 1: Location of the site (City of Lae, Papua New Guinea)

The authority of the factory helped to ensure the security of the specimens and looked after the specimens. Seawater curing of the specimens was performed in two conditions:

Sea water curing 1: These specimens were submerged alternatively during high tides while during low tide they remain unsubmerged. The period of submergence varied between 10-12 hours.

Sea water curing 2: These specimens were fully submerged for the whole period of curing irrespective of high or low tide condition of the sea.

For each mix, 16 cylinders had been cast to provide four cylinders for different curing ages. It was planned to cure the specimens for 7, 14, 28 and 91days under seawater water curing conditions 1 and 2. The specimens were examined for any physical changes after the specific curing period and then tested for compressive strength.

After curing period of 91 days in seawater, powder XRD (X-ray diffraction) analysis was conducted on samples taken from the fully submerged (seawater curing 2) cylinder specimens. Samples were also collected from the 91-day cured cylinders (under seawater curing 2) at various depths from the surface using electrical drills. These samples were then ground into fine powder passing the 150µm sieve. The total chloride contents were then measured as per JCI [22]. A 2-M HNO_3 solution was added to the powder, and the mixture was titrated with 0.005M $AgNO_3$ solution to determine the total chloride content of the mortar powder. The results were used to plot graphs of the chloride content versus depth from the surface of the specimen.

The mercury intrusion porosimetry (MIP) was used to measure the total pore volume (TPV) and pore radius of concrete at 7, 14, 21, 56 and 91 days of seawater curing 2 of the specimens.

3. RESULTS

3.1 Influence of volcanic ash and curing condition on the compressive strength

The typical variation of compressive strength (7 and 91 day) for different curing condition with different VA content is presented in Fig. 2. Generally the strength is found to decrease with the increase of VA content for different curing conditions (as shown in Fig. 2; 7 day strength development) with some interesting exception reflecting the benefit of VA in marine environment. With the increase in age of concrete (as can be seen from 91 day strength), the addition of VA as cement replacement is found to increase the strength of VAC compared to NC for all three curing conditions especially for 5 and 10% VA. Between 28 and 91 day, the strength of 10% VAC exceeds the strength of 5% VAC as can be seen from the ascending branch of the curves within this range.

At 91 day, the strength of 10% VAC in seawater curing 2 (Fig. 2) exceeds the strength from other two curing conditions which suggests the better performance of VAC concrete with long exposure in the marine environment. On the other hand, the 91-day strength of NC in both seawater curing conditions is found to be less than that of normal water curing. This indicates the long-term poor performance of NC in marine environment compared to VAC and hence justifies the use of VA in marine structures.

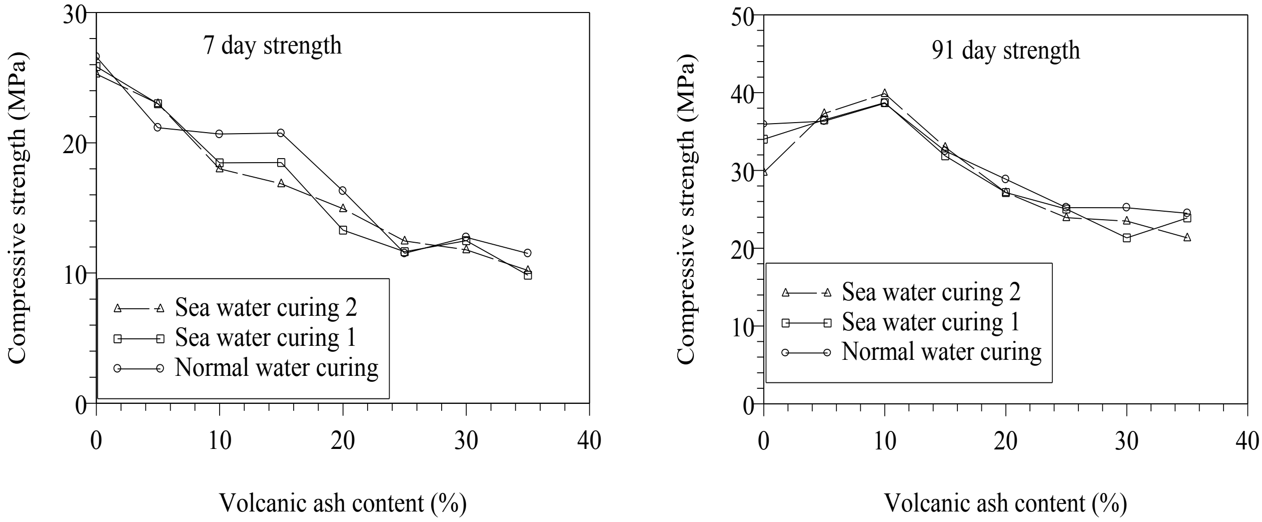

Figure 2: Variation of compressive strength with % of VA

3.2 Influence of curing age and curing condition on the compressive strength

The performance of NC and VAC with different dosages (up to 35%) of VA is compared in Fig. 3. NC shows poor performance in seawater curing. The strength from both seawater curing is less than normal water curing (Fig. 3). More exposure to the sea condition results in more reduction in strength as can be seen from the lower strength of the specimens in seawater curing condition 2 compared to specimens in seawater curing condition 1. Extension of curing age also leads to more strength reduction in NC and gives an indication of long-term poor performance of NC in marine condition.

Fig. 3 also compares the performance of VAC and NC under different curing conditions. The better performance of VAC with curing in marine condition compared to NC for VA dosages ranging from 5 to 15% is observed. Within the curing age of 91 day, the strength of VAC in seawater curing conditions exceeds the strength of normal water curing. It is interesting to note that more severe seawater curing (condition 2) produces higher strength within 91 day.

The performance of VAC with VA dosages ranging from 20 to 35% is also examined. Seawater curing seems to produce lower strength compared to that of NC. At VA dosages of 25%, the 28-day strength in all three curing conditions seems to produce similar strength and on the other hand 91-day strength is reduced in seawater curing. The lower strength development of VAC with VA dosages ranging from 20 to 35% particularly in the long term, suggests the identification of an optimum dose of VA. Current results suggest an optimum dose of VA ranges between 5 and 15%.

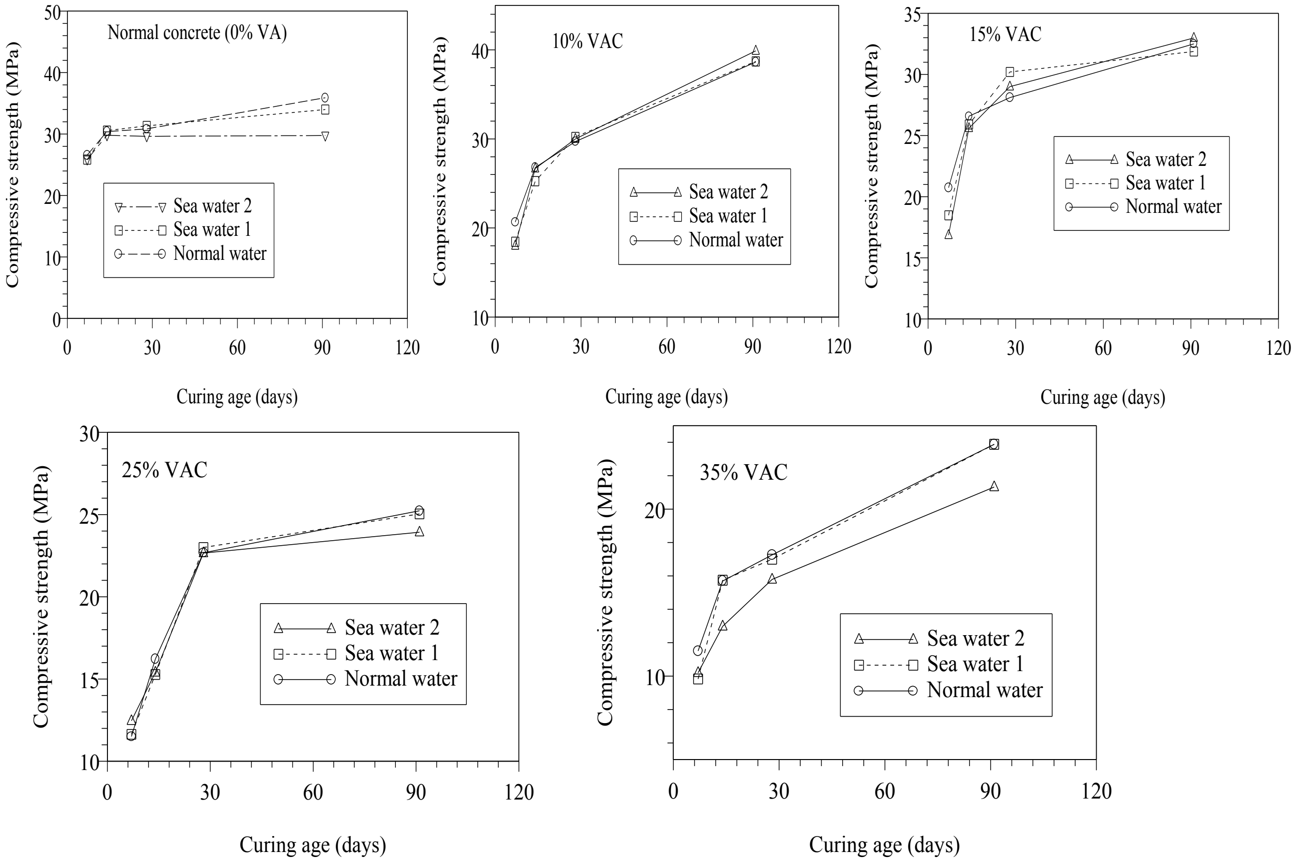

Figure 3: Performance of VAC with curing age and conditions

3.2 Effect of curing age/condition and volcanic ash content on strength gain or loss

Fig. 4 shows the strength gain (+ve value) or loss (-ve value) expressed as % in VAC compared to control normal concrete under different curing condition and with varying VA replacements. At the age of 7 day, all VAC mixes loss their strength compared to normal concrete and the loss of strength is found to be higher in sea water curing conditions 1 and 2 compared to normal water curing. But at the age of 28 day, 5% VAC starts to gain strength (+ve values) compared to NC. It records 14 to 15% strength enhancement in seawater curing condition 2 compared to about 4 to 5% in seawater curing condition 1 and normal water curing. Beyond 5%, the loss of strength can be observed but strength loss is found to be less in seawater curing condition 2 compared to other two conditions. It reflects the overall better performance of VAC in seawater curing.

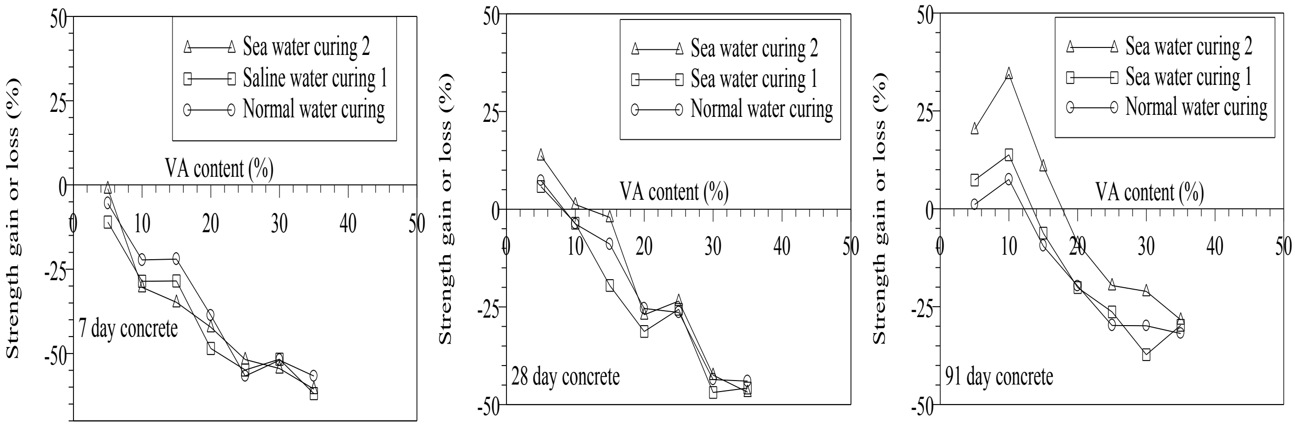

Figure 4: Comparative study of strength loss or gain

The benefit of marine environment on the performance VAC is clearly revealed at the curing age of 91 day, which can be seen from Fig. 3. The seawater curing condition 2 exhibits better performance with about 20%, 35% and 10% strength gain compared to NC for VAC with 5%, 10% and 15% VA content, respectively. Even the seawater curing condition 1 exhibits better performance compared to normal water curing with strength gain of about 5% and 12% for VA content of 5% and 10%, respectively. The range of VA content for strength gain extends from 5 to 15% in the case of seawater curing 2 compared to 5 to 10% in curing condition 1 and normal water.

4. DISCUSSION ON POZZOLANIC ACTIVITY, STRENGTH, DURABILITY AND MICROSTRUCTURAL CHRACTERISTICS

The chemical deterioration of concrete in marine environments has been a topic of interest to concrete technologists in the last few decades. Seawater contains up to 35000 ppm of dissolved salts - about 78% of the salt is sodium chloride, and 15% is chloride and sulphate of magnesium. The concomitant presence of sulfate and chloride ions in marine environments causes deterioration of reinforced concrete structures and reinforcement corrosion. The reaction of the concrete with the sulfate ions in marine environments is similar to that of sulfate ions in non-marine environments, but the effects are different due to the presence of chloride ions in the former [23-24]. The effect of the conjoint presence of chlorides and sulfates on the sulfate resistance of hydrated Portland cements is inconclusive and highly debated [25]. The sulfate attack in marine environment gives rise to expansive ettringite, gypsum, and brucite and sometimes is associated with calcite formation [26].

The corrosive action of seawater has been attributed to the reaction of MgSO4 with Ca $(OH)_2$ liberated, forming gypsum and $Mg(OH)_2$ according to the following equation:

$$MgSO_4 + Ca(OH)_2 \rightarrow CaSO_4 + Mg(OH)_2$$

The gypsum formed reacts with calcium hydroxide liberated during hydrolysis of calcium silicates and forms calcium hydrosulphoaluminate.

$$3\ CaSO_4 + C_3A + 3\ H_20 = C_3A.\ 3\ CaSO_{4.}\ 3\ H_20 \qquad \text{(ettringite)}$$

When ettringate and partly $CaSO_4$ are liberated, they enlarge the volume, resulting in concrete expansion affecting the durability of concrete. Expansion caused by ettringite formation is the most widely recognised mechanism of sulphate attack. In addition, the chlorides presents in seawater reacts with $Ca(OH)_2$ liberated as well as C_3A to form calcium hydochloroaluminate and possibly thaumastic simultaneously according to:

$$MgCl_2 + Ca(OH)_2 \rightarrow Ca\ Cl_2 + Mg\ (OH)_2$$

$$CaCl_2 + C_3A + 10H_2O \rightarrow C_3A.CaCl_2.10H_2O$$

The formation of calcium hydrochloroaluminate could be one of the factors leading to reduction of concrete strength. The principal methods available to prevent sulfate attack using sulfate-resisting construction materials are changing Type I to Type II or Type V cement and introducing pozzolana such as fly ash, blast furnace slag in concrete [25, 27-33]. Researchers have shown that limitation on C_3A content is not the ultimate answer to the problem of sulfate attack [25, 30, 34, 35]. The use of blended cement made with fly ash, silica fume, and blast furnace slag is therefore recommended in sulfate environments [27].

The long-term improvement of strength of VAC in marine environment is confirmed from the current test results. Despite corrosive action of seawater, the strength of VAC having 5 to 15% VA is increased compared to NC and normal water curing conditions within a period of 91 day. This is an indication of the development of resistance against corrosive action of seawater due to the presence of VA in VAC. The presence of VA in VAC improves the strength due to their pozzolanic reaction with $Ca(OH)_2$ to produce a greater solid volume of cementitious calcium silicate gel leading to an additional reduction in capillary porosity during hydration.

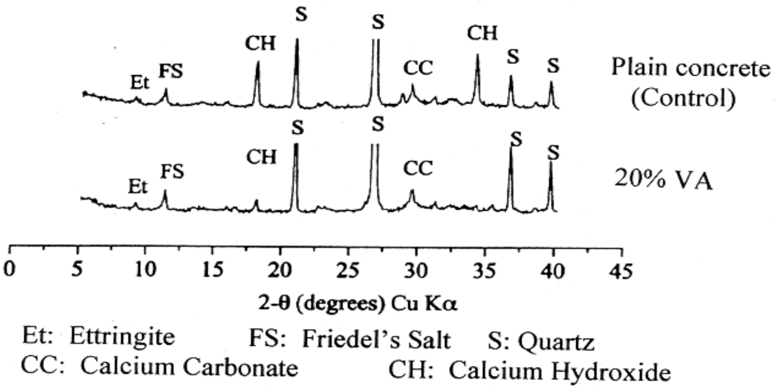

Figure 5: XRD Spectra of concrete samples

The XRD curves (Fig. 5) of the powder samples taken from the fully submerged specimens (seawater curing 2) reveal that the calcium hydroxide content of the VA–blended specimens is lower than that of the plain concrete specimens. This indicates the pozzolanic reactivity of VA that consumes calcium hydroxide resulting from hydration of the cement.

Comparatively higher Friedel's salt formation in VAC mixes compared to control mix is also confirmed from the XRD spectra in Fig. 5. Similar phenomena were also observed in fly ash concrete [36]. The VA certainly does not have C_3A which can adsorb more chloride ions to form Friedel's salts. The presence of lower C_3A content in PVAC compared to PC (Table 2) confirms the fact. It is more likely that VA has aluminium in the glass that is available for the chemical reaction resulting in Friedel's salt production.

Friedel's salt formation consequently lowers the levels of free chloride and hence, can reduce the chloride ion diffusivity of concrete. This can be evident from Fig. 6 which shows the chloride concentration profiles of the cylinder specimens fully submerged for 91 days under seawater curing 2. The chloride content at different depths decreased with the increase of VA content. The partial replacement of cement by VA results in a reduction in chloride ingress significantly beyond the depth of 25 mm compared with the control concrete with 0% VA. 35% cement replacement results in the lowest chloride ingress at deeper depths in all sets of specimens. This can also be attributed to the capability of VA to partially obstruct voids and pores leading to a decrease of pore size with refinement of pore structure and to a smaller effective diffusivity for chloride or other species.

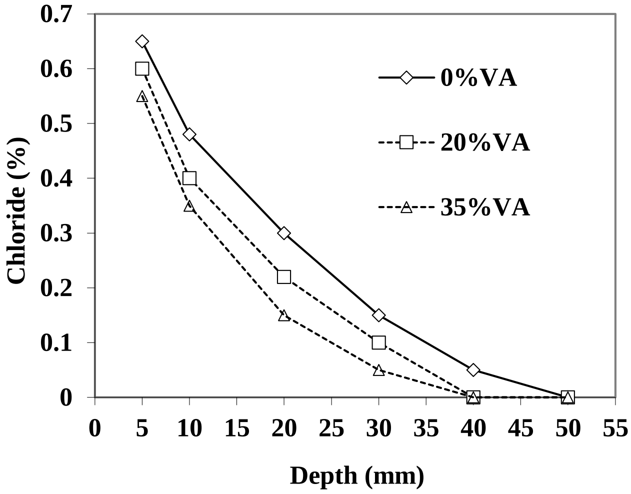

Figure 6: Chloride concentrations at various depth from the surface for VAC

The effect of VA on the pore size distribution within the total pore volume (TPV), for pore sizes less than 20nm (micropores) and pore sizes greater than 20nm (macropores) at different curing times (seawater curing 2) is presented in Fig. 7. It is noted that increasing levels of replacement of cement with VA (up to 35%) produce a refinement of pore structure. Such refinement of pore structure is attributed to the lowering of permeability/diffusivity of VAC and hence, leads to the improved long-term seawater ion resistance and durability.

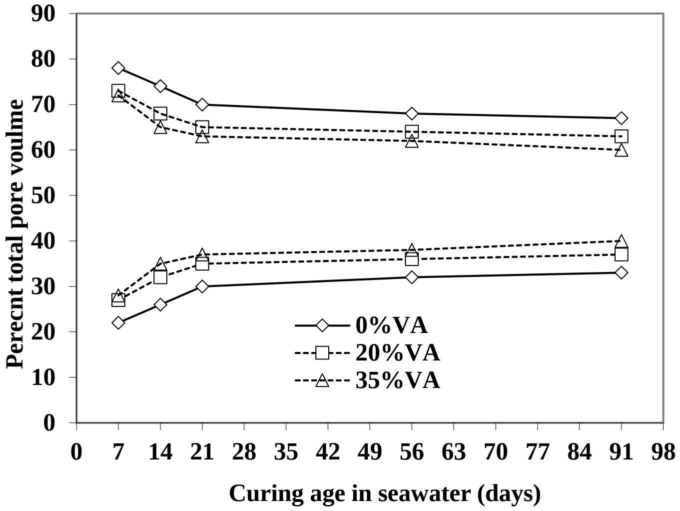

Figure 7: Pore size distribution with seawater curing time

The consumption of $Ca(OH)_2$ by VA prevents the formation of ettringite and calcium hydrochloroaluminate, thus alleviating the corrosive attack of seawater. However, the reaction of VA with $Ca(OH)_2$ could be dependent on the amount of VA in the mix and curing age. As the pozzolanic action of VA is a slow process, its benefit will be reflected in the long-term improvement of strength or durability (as confirmed from this research) of VAC compared to normal concrete. Investigations are in progress to study the long term (10-year period) strength and durability performance of VAC under marine environment.

5. CONCLUSIONS

The performance of volcanic ash concrete (VAC) incorporating different dosages of volcanic ash (VA) as cement replacement (0 to 35% by weight) is presented and compared with that of control normal concrete (NC) in sea and normal water curing conditions. The following conclusions are drawn from the study:

- The seawater curing showed better performance of VAC (having 5%, 10% and 15% VA content) with about 20%, 35% and 10% strength gain, respectively compared to NC. The strength gain range may further increase when curing age is further extended.
- Seawater curing condition 2 (fully submerged for whole duration) exhibits better performance with a maximum strength gain of 35% at 91 day compared to seawater curing condition 1 (10 to 12 hours of submerged per day). The better performance of VAC depends on the use of optimum amount of VA in the mix and curing age. The optimum amount of VA identified from this study ranges between 5 to 15%. VA can enhance the long-term strength of VAC compared to NC due to their pozzolanic action. In this study benefit is clearly revealed at the curing age of 91 day.
- X-ray diffraction (XRD) analysis confirms the presence of lower calcium hydroxide content and formation of comparatively higher Friedel's salt (reduces the levels of free chloride) in the VA– blended specimens compared with the control concrete specimens. In addition, the incorporation of VA leads to refinement of the pore structure. The proportion of pores with radii smaller than 20nm is increased as the replacement level of cement by VA increases. These processes reduce the diffusion of seawater ions in the concrete and improve long-term durability.
- VA blended specimens showed better resistance to chloride ingress exhibiting the lowest chloride ingress at deeper depths. This study suggests the application of VA blended cement concrete in marine structures to provide better durability.

6. ACKNOWLEDGEMENT

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