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Effect of Geopolymer Aggregate on Strength and Microstructure of Concrete

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The properties and microstructure of a novel manufactured geopolymer coarse aggregate have been investigated. The analysis has included compressive and tensile strengths of concretes made with the manufactured geopolymer coarse aggregate and a comparative natural crushed coarse aggregate. In addition, the microstructure and pore structure development of both concretes at the interfacial transition zone (ITZ) and bulk cement matrix were studied though scanning electron microscopy and X-ray computed tomography. The data showed that the novel geopolymer coarse aggregate satisfied the requirements of Australian Standard AS 2758.1 and is comparable to the natural aggregate. The dry density of the geopolymer aggregate concrete was less than that of the natural aggregate being just over 2000 kg/m³ (124.9 lb/ft³), with a mean 7-day strength in excess of 30 MPa (4.44 ksi) and a mean 28-day compressive strength in excess of 40 MPa (5.8 ksi). Moreover, it showed a 60% reduction in porosity between 7 and 28 days with a wellcompacted and dense ITZ observed at 28 days. In addition, the flexural strength demonstrated a good correlation with compressive strength, comparable to that of the natural aggregate concrete. Overall, the geopolymer investigated in this research shows potential as a lightweight coarse aggregate for concrete, with the additional benefit of reducing the environmental impact of fly ash from coal-fired power generation.

Keywords: fly ash; geopolymer aggregate; microstructure; pore structure; strength; X-ray tomography.

INTRODUCTION

It has been estimated that the annual fly ash generation from coal power plants in Australia had reached 14 million tonnes (15.4 million tons) in 2015. Worldwide fly ash production is much higher and it is anticipated to increase up to approximately 2000 million tonnes (2204 million tons) in 2020.¹ Out of this production, between 35 and 45% of fly ash is being used for various purposes, including cement and concrete production, and the balance is disposed of in landfills and storage lagoons at a significant cost, posing a potential risk to the water supply from local aquifers due to the possible leaching of heavy metals.^{2,3} As the fly ash waste continues to accumulate, there is pressure on the coal power industries to find a solution for its disposal.

Meanwhile, natural aggregate reserves are depleting fast, particularly in some desert regions of the world. Worldwide natural aggregate production is approximately 4.5 billion tonnes (5.0 billion tons),⁴ and Australia alone consumes approximately 130 million tonnes (143.3 million tons) of aggregates annually.⁵ The production of 1 tonne of natural crushed aggregate emits 7.4 to 8.0 kg (16.3 to 17.6 lb) of CO₂. In addition, emissions from trucks as well as the use of crushers, which result in large dust and particulate emissions, contribute to the increased global warming. Further-

more, the quarrying removes rock and impacts the natural drainage pattern, adversely affects the aesthetics of the environment, the stability of slopes, due to the removal of trees and affects fauna due to blasting and human activity. These can result in changes to rainfall patterns and more extreme impacts due to climate change, such as floods and droughts. Aggregate demand is also increasing with the expansion of construction. Thus, developing concrete with nonconventional aggregates is an essential sustainable approach in terms of the environmental and economic aspects.

A review of the literature illustrates that a range of waste products are currently used in concrete production as partial or full replacement for the cement. While the inclusion of recycled aggregates from demolition and building waste in concrete has been widely investigated by previous researchers, major problems in the use of these materials is their purity together with higher water absorption. This in turn can affect concrete durability such as alkali aggregate reaction and the ingress of chloride and carbon dioxide.⁶⁻¹⁰ The lower quality of recycled aggregate concrete, due to higher water absorption, higher porosity, and a weaker ITZ hinders the application of this concrete in higher-grade concrete.¹¹ Other alternatives include lightweight fly ash aggregate which can be manufactured^{12,13}; however, this consumes large amounts of energy in the production stage. Ramamurthy and Harikrishnan¹⁴ also studied concrete produced with sintered fly ash aggregate. Their study was focused on the effect of different binders on the properties of the aggregate and concluded that sintered fly ash aggregate has higher porosity and water absorption. The microstructure of sintered fly ash aggregate indicates that the shapes of the pores are primarily irregular, spherical, and discrete, while others are elongated and interconnected. The lack of binding among the grains results in lower strength being observed in this type of manufactured aggregate.

Manufactured geopolymer coarse aggregate using low-calcium fly ash has recently been developed.¹⁵ The technology for the manufacture of this geopolymer coarse aggregate uses novel techniques employing high pressure and reduced temperature production methods. The reaction mechanism of the geopolymer coarse aggregate is similar to that of fly-ash-based geopolymer concrete. The silicates and aluminates in low-calcium fly ash react with highly alka-

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	By weight, %											
Material	SiO_2	Al ₂ O ₃	Fe ₂ O ₃	CaO	P ₂ O ₅	TiO ₂	MgO	K ₂ O	SO ₃	MnO	Na ₂ O	LOI*
Cement	22.5	4.5	0.4	66.3	0.67	0.20	0.51	0.15	2.8	0.10	0.17	1.7

*Loss on ignition (unburnt carbon content).

Table 2—Mixture design used for concrete, kg/m³

Comont	Aggregates		Watar		
Cement	Fine	Coarse	water	TIX W KA	
350	662	885	122.5	1.75	
350	662	1305	122.5	1.75	
	Cement 350 350	Cement Fine 350 662 350 662	Aggregates Fine Coarse 350 662 885 350 662 1305	Cement Fine Coarse Water 350 662 885 122.5 350 662 1305 122.5	

*HRWRA (high-range water-reducing admixture) is calculated as a proportion of cement weight (0.5%).

Table 3—Sieve analysis of coarse aggregates

	Cumulative sie	% retained on	Cumulative % passing on sieve		
Sieve size, mm	Natural aggregate	GPA	Natural aggregate	GPA	
9.50	6.5	6.6	93.5	93.4	
6.70	86.1	87.0	13.9	13.0	
4.75	98.1	99.1	1.9	0.9	
2.36	98.9	99.9	1.1	0.1	
1.18	99.0	100.0	1.0	0.0	
<1.18	100.0	100.0	1.6	0.0	

Note: 1 mm = 0.0393 in.

line activators and produce a sodium-aluminosilicate gel. This geopolymeric gel consists of a three-dimensional (3-D) network of silicon and aluminum atoms linked by oxygen atoms in a fourfold coordination.¹⁶⁻²⁰ If viable, use of manufactured geopolymer coarse aggregate from industrial waste fly ash could lead to improved sustainable concrete design and a greener environment while reducing use of landfill and storage lagoons. This paper presents an investigation of the impact of this manufactured novel geopolymer coarse aggregate on the compressive and flexural strengths of concrete. The paper also discusses the microstructure of the geopolymer aggregate concrete. Moreover, the observed test results are compared with a similar concrete made with natural crushed granite coarse aggregate.

SIGNIFICANCE OF RESEARCH

This research reports the replacement of natural crushed aggregate with manufactured geopolymer coarse aggregates in concrete. The use of this novel aggregate in concrete has the potential to reduce the reliance on conventional aggregate quarries as well as using a waste material, hence reducing the environmental impacts. The microstructure and pore development of the geopolymer aggregate concrete presented will contribute to the understanding of the impact of manufactured geopolymer coarse aggregate on the properties of the concrete produced.

EXPERIMENTAL PROCEDURE Materials and mixture design

Commercially available Type 1 portland cement (PC) conforming to ASTM C150²¹ was used to produce geopolymer coarse aggregate (GPA) and natural (crushed granite) aggregate concrete specimens. The chemical composition of PC, determined by X-ray fluorescence analysis, is shown in Table 1. The specific surface area of PC, determined by Brunauer Emmett Teller (BET) method by N2 absorption, is 460 m²/kg. The mixture proportions used for concretes made with GPAs and natural aggregates is summarized in Table 2. The ingredients of the concrete mixture, such as cement, fine aggregate, coarse aggregate, and water, was calculated based on the absolute volume method.²² The fine aggregate used was river sand in uncrushed form with a specific gravity of 2.5 and a fineness modulus of 3.0. The sieve analysis of GPA and natural aggregate is tabulated in Table 3. Both types of coarse aggregates used were in saturated surface-dry condition to prevent water absorption from the concrete mixture. The quantity of total aggregates in both concretes was kept to 60% of the entire mixture by volume, and the water-cement ratio (w/c) was fixed to 0.35. Sulfonated naphthalene-formaldehyde condensate type high-range water-reducing admixture in liquid form was used, together with demineralized water, throughout the experiment.

Mixing, casting, and curing of concrete

Concrete production was carried out using a 120 L (4.238 ft³) planetary concrete mixer. The dry materials (cement, sand, and coarse aggregates) were first mixed for 4 minutes. Both water and high-range water-reducing admixture were then added to the dry mix and mixed continuously for another 8 minutes. This produced a glossy and well-combined mixture. A slump test was conducted in accordance with AS 1012.3.123 to ensure concrete achieved the required slump range of 65 to 75 mm (2.56 to 2.95 in.). The concrete mixture was then poured into 100 x 200 mm (0.328 x 0.656 ft) cylindrical specimens for compressive strength testing and 100 x 100 x 350 mm (0.328 x 0.328 x 1.148 ft) beams for flexural strength testing. All molds were filled with concrete in two stages while vibrated using a vibration table for 1 minute to remove air bubbles. All concrete specimens were cured in potable water, $23 \pm 2^{\circ}C$ $(73.4 \pm 3.6^{\circ}F)$, until tested.

Testing

Manufactured geopolymer coarse aggregate—A series of tests were conducted for manufactured geopolymer and natural aggregates in accordance with the relevant standards. In each specific coarse aggregate test, three samples were tested and the mean value was reported. The aggregate crushing value and the aggregate impact value were examined in accordance with AS 1141.21²⁴ and RC 371.03,²⁵ respectively. Moreover, the Los Angeles abrasion value, the Particle Shape-Flakiness Index, and the aggregate soundness were examined in accordance with AS 1141.23,²⁶ AS 1141.15,27 and AS 1141.24,28 respectively. According to AS 1141.23, Los Angeles abrasion testing equipment was used to measure the mass loss of aggregates due to abrasion. Conversely, aggregates having a nominal size not greater than 63 mm (2.48 in.) and having at least 80% by mass retained on a 4.75 mm (0.187 in.) sieve were used to test the flakiness index. During the soundness test, aggregates were exposed to sodium sulfate solutions over repeated cycles of wetting and drying and measured the mass loss of aggregates. The specific gravity, the unit weight, the loose bulk density, and the compacted (rodded) bulk density of both coarse aggregates were examined in accordance with ASTM C127,²⁹ AS 1141.4,³⁰ and ASTM C29,³¹ respectively.

The water absorption and porosity (voids in aggregates) were examined in accordance with ASTM C127 and ASTM C29/C29M. Further, alkali-silica reactivity and the soluble chloride and sulfate contents of coarse aggregates were examined in accordance with RC 376.03³² and AS 1012.20.³³ The degradation temperature and specific heat capacity of geopolymer and natural aggregates were determined using thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). Aggregates were ground into fine particles and filtered using a 75-micron sieve to obtain the required powder samples for testing.

Geopolymer aggregate concrete—The initial and final setting time of geopolymer aggregate concrete and natural aggregate concrete mixtures were examined using a Vicat apparatus in accordance with AS 1012.18. The compressive strength tests were performed on GPA and conventional concrete samples using an MTS machine with a loading rate of 20 MPa/min (2.9 ksi/min) according to AS 1012.9.34 The flexural strength test was conducted to determine the tensile strength of GPA concrete in accordance with AS 1012.11.35 This test was carried out on an MTS machine under a four-point bending test with a loading rate of 1 MPa/min (0.145 ksi/min). The dry density test was conducted in accordance with AS 1012.12.2.36 All tests were performed at 7 and 28 days from casting. In each specific concrete test, three samples were tested, and mean value and standard deviation were reported. The microstructure was examined using scanning electron microscopy (SEM) imaging employing backscatter electron detector with 15eV of energy. Specimens were cut using a diamond saw to a size of 2 to 4 mm (0.079 to 0.158 in.) in height and 5 to 10 mm (0.197 to 0.394 in.) in diameter. The samples were subsequently carbon coated and then mounted on the SEM sample stage with conductive, double-sided carbon tape.

The pore structure was observed using X-ray computed tomography (CT). Specimens were cut using a diamond saw to a size of 10 mm (0.394 in.) length x 10 mm (0.394 in.) width x 50 mm (1.968 in.) height. The scans at 20 μ m (0.000787 in.) resolution were done at 180 kV and 100 μ A, and 1000 images were recorded during one full rotation of the sample. A background detector region of interest was selected, which corrects for potential X-ray flux variations.

All scans were performed with a copper filter of 0.6 mm to reduce beam hardening artefacts.

Data was analyzed with a commercial software package using the defect analysis module. The data is smoothed using a median filter before any analysis to remove noise. The procedure for obtaining porosity information involves selecting the object using an adaptive rectangle around the object, and an average void fraction was calculated using the volume analysis tool. A surface fit function is then applied using the threshold value as the central value between the peak of the material and air, in the data histogram. This 3-D surface indicates the transition between material and pore. An automated defect analysis is then done using the defect analysis module of a high-end software, limited to a pore size of at least eight voxels in total. This function generates the pore volume of individual pores.

EXPERIMENTAL RESULTS AND DISCUSSION Properties of geopolymer coarse aggregate

The characteristics of the manufactured geopolymer coarse aggregate determined are summarized in Table 4. Test results were compared with natural aggregate used for concrete, and also with the criteria recommended in AS 2758.1³⁷ for coarse aggregate. The permissible limits for the Los Angeles abrasion value and sodium sulfate soundness are 35% and 9%, respectively, to satisfy the exposure classification of B1 and B2 (AS 2758.1). Moreover, a flakiness index less than 35% (AS 2758.1) and alkali-silica reactivity less than 0.1 (RC 376.03) are recommended for coarse aggregates.

It was noted that the abrasion value of manufactured geopolymer coarse aggregates is almost half of the value observed for natural aggregate. According to AS 1141.23, a similar mass of both aggregates was tested for abrasion. Due to lighter density, geopolymer coarse aggregate occupies a larger volume in the Los Angeles abrasion machine. Hence, it is recommended that testing of abrasion using the same volume of geopolymer and natural aggregates would provide a better comparison.

On the other hand, geopolymer aggregate shows slightly higher water absorption and porosity compared to natural aggregate, although there is a considerable difference in the aggregate density. The lower specific gravity of fly ash would be expected to lower the density of the manufactured geopolymer aggregates. However, the increase in the packing density and compactness during the aggregate manufacturing process (that is, in pressure application and heat curing) is hypothesized as to lower the porosity and water absorption of the coarse aggregate. Overall, it is noted that the manufactured geopolymer coarse aggregate satisfied all these conditions, and the values obtained are comparable to those of the natural coarse aggregate.

Setting time of geopolymer aggregate concrete

Figure 1 shows the variation of penetration resistance of the geopolymer aggregate and the natural aggregate concrete mixtures as a function of time. The initial and final setting times are defined as the time at which the penetration resistance reach values of 3.5 and 27.6 MPa (0.508 and 4.0 ksi), respectively. The calculated initial and final setting times

Table 4—Properties of	i geopolymer	and natural	aggregates
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Characteristic of	aggregate	Geopolymer aggregate	Natural aggregate	
Aggregate crushin	g value, %	29.5	23.2	
Aggregate impact	t value, %	28.4	23.6	
Los Angeles abrasio	on value, %	17.4	32.7	
Sodium sulfate sou	indness, %	6.6	0.4	
Flakiness ind	ex, %	23	14	
Specific gravity (oven-dry)	1.812	2.671	
Unit weight, I	kg/m ³	1709	2550	
Loose bulk densi	ty, kg/m ³	894	1345	
Compacted (rodded) bul	k density, kg/m ³	958	1460	
Water absorpt	ion, %	1.60	0.92	
Porosity (voids in ag	gregates), %	2.63	1.84	
Alkali-silica read	ctivity, %	0.089	0.077	
Soluble salts-chl	orides, %	0.021	0.004	
Soluble salts-su	lfate, %	0.039	0.016	
	at 100°C	1.012	1.039	
Specific heat capacity, J/gC	at 200°C	1.124	1.149	
Decredation temperature %C	at 5% weight loss	569	>900	
Degradation temperature, 'C	at 10% weight loss	>900	>900	

Notes: $1 \text{ kg/m}^3 = 0.062 \text{ lb/ft}^3$; $T_{(^\circ\text{F})} = [T_{(^\circ\text{C})} \times 9/5] + 32$.

Table 5—Setting times of concretes

	Setting time, hours	
Concrete type	Initial	Final
Natural aggregate concrete	4.85 ± 0.05	8.05 ± 0.10
Geopolymer aggregate concrete	4.90 ± 0.10	8.05 ± 0.05

for both concretes are tabulated in Table 5. It is noted that concretes made with geopolymer aggregate and natural aggregate showed a similar initial and final setting time. From this it can be concluded that replacing natural aggregate with manufactured geopolymer coarse aggregate in the portland cement concrete does not affect the concrete setting times.

Strength of geopolymer aggregate concrete

The dry density of the geopolymer aggregate concrete increased from 2055 to 2140 kg/m³ (128.29 to 133.60 lb/ft³) from 7 to 28 days, while dry density of natural aggregate concrete increased from 2585 to 2645 kg/m³ (161.38 to 165.12 lb/ft³). The density increase for the geopolymer aggregate was similar to that of the natural aggregate concrete, but with a density that is approximately 20% lower than the natural aggregate concrete has a density in the range of 300 to 1850 kg/m³ (18.73 to 115.5 lb/ft³). While density of the geopolymer coarse aggregate is approximately 35% lower than the density of the crushed granite aggregate, the geopolymer aggregate concrete demonstrated only 20% reduction in density, and well above the upper density limit of lightweight concrete.

Figure 2 shows the compressive strength development and compressive strength activity index (percentage strength



Fig. 1—Influence of coarse aggregate on penetration resistance of concrete. (Note: 1 MPa = 0.145 ksi.)



Fig. 2—(a) Compressive strength development; and (b) compressive strength activity index of different aggregate concretes. (Note: 1 MPa = 0.145 ksi.)

ratio of geopolymer aggregate to natural aggregate concrete) of geopolymer and natural coarse aggregate concretes at

7 and 28 days. The geopolymer concrete achieved 30.6 MPa (4.44 ksi) at 7 days compared to 43.0 MPa (6.24 ksi) for natural aggregate concrete. The compressive strength of the geopolymer aggregate concrete increased to 39.4 MPa (5.71 ksi) at 28 days, while the natural aggregate concrete achieved 48.3 MPa (7.0 ksi). It is noted that the geopolymer aggregate concrete gave a 29% compressive strength increase compared to a 12% strength increase in the natural aggregate concrete that more than 80% of the compressive strength is achieved at 28 days using geopolymer coarse aggregates as a 100% replacement for natural crushed aggregates in concrete.

The 95% characteristic compressive strength of geopolymer aggregate concrete at 28 days is 37.5 MPa (5.44 ksi). This is well in excess of the specified minimum strength for the design of concrete structures in exposure category B1 (32 MPa [4.64 ksi]) in accordance with AS $3600.^{38}$ Thus, this concrete could be used for a range



Fig. 3—(a) Flexural strength development; and (b) flexural strength activity index of different aggregate concretes (Note: 1 MPa = 0.145 ksi.)

of applications including industrial buildings, ground slabs, and near coastal structures.

The flexural strength of the geopolymer and natural aggregate concretes increased from 4.51 to 4.81 MPa (0.654 to 0.698 ksi) and from 6.25 to 6.38 MPa (0.906 to 0.925 ksi) at 7 and 28 days, respectively (Fig. 3). Flexural strength generally determines the failure tensile stress of a concrete beam without steel reinforcement. The flexural strength increased with time in both concretes, and were approximately 12 to 14% of the compressive strength. This is slightly higher than the range of 9 to 12% typically cited for conventional aggregate concrete in literature.²²

Microstructure of geopolymer aggregate concrete

A microstructural investigation was conducted to understand the fracture surfaces of the concretes. As the bulk cement matrixes were the same due to the use of the 100% portland cement as binder with a constant w/c, the microscopy examinations were conducted on the cement-aggregate interface.

In PC concrete, coarse aggregate has a two-layer interfacial transition zone (ITZ) of approximately 100 μ m depth. The outer layer is a duplex film which is normally formed at the surface of the aggregate, and the second layer is formed internally, which is a porous transition zone of 20 to 100 μ m deep.³⁹ The ITZ is critical because it is known to have a different microstructure from the bulk of the hardened PC paste and the cement-aggregate interface is also considered as the specific location of early cracking. Scrivener et al.⁴⁰ reported that ITZ in PC concrete is caused by the disorder in the packing of the anhydrous cement grains in the transition zone.

Figure 4 compares the microstructure of ITZ in the geopolymer concrete and the natural aggregate concrete



Fig. 4—*Microstructure of cement-aggregate interface at 7 days: (a) to (c) geopolymer aggregate (GPA) concrete; and (d) to (f) natural aggregate (NA) concrete.*

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Fig. 5—*Microstructure of cement-aggregate interface at 28 days: (a) to (c) geopolymer aggregate (GPA) concrete; and (d) to (f) natural aggregate (NA) concrete.*

at 7 days. More microcracks were evident in the cementgeopolymer aggregate transition zone than in the cementnatural aggregate interface. It was noted that the cracks created in different ITZs have propagated through the bulk cement gel matrix and combined creating a crack network in the geopolymer aggregate concrete. The thickness of the geopolymer aggregate-cement interface varied along the geopolymer particle surface. This further illustrates that interfacial zone appeared to consist of loose particles, and the width of the loose zone was approximately 40 to 60 µm. Figure 4(c) shows, at a higher magnification, that the interfacial zone of the geopolymer aggregate concrete consists of larger microcracks. In contrast, Fig. 4(f) shows that the interface between the natural aggregate and the cement matrix was much denser, and the interfacial transition zone cannot be easily distinguished. Overall, continuing microcracking through the cement-aggregate interface and formation of a crack network would be expected to create a weaker bond between bulk gel matrix and ITZ in geopolymer aggregate concrete at early stage.

Figure 5 compares the microstructure of ITZ in geopolymer concrete and natural aggregate concrete at 28 days. It is noted that geopolymer aggregate concrete had an almost identical interfacial zone density to the natural aggregate concrete at 28 days. In geopolymer aggregate concrete, the interfacial zone primarily consisted of a relatively dense layer of hydrates, which was substantially different from that observed at 7 days in the geopolymer aggregate-cement interface. In particular, ITZs in both

concretes showed less microcracks than at 7 days and that these were of reduced width compared to the earlier-age specimens. Granular whisker-like hydrates and flake-like crystals could be found in both concretes in the ITZs, and the energy-dispersive X-ray (EDX) analysis confirmed that hydrates were principally C-S-H gel.

In the natural aggregate concrete, a water film will normally form around the coarse aggregates due to bleeding and wetting effects. The local w/c at the interfacial transition region can be twice as that in the bulk cement paste.²² As hydration progresses, the water-filled space near the coarse aggregate is gradually replaced by an increasing amount of C-S-H gel products. The less condensed, cracked cementaggregate interface at 7 days is hypothesized as being partly due to the moisture content of the geopolymer coarse aggregate. The increase in density of the ITZ could be accounted for by geopolymer aggregate, having a high water absorption capacity (Table 4), absorbing a certain amount of the free water, thus lowering the initial w/c in the ITZ during early hydration. However, as hydration progresses, newly formed hydrates gradually fill in the porous cement-aggregate interface while healing the microcracks, and thus effectively improved the interfacial bond between the aggregate and cement matrix. The nature and density of the ITZ is one of the main factors governing both compressive strength and flexural strength of concrete. Hence, increasing the density of ITZ in geopolymer aggregate concrete strengthens the aggregate-matrix bond resulting in the strength development observed in GPA concrete over time.



Fig. 6—3-D view with color-coded voids according to size from 3-D analysis.

Pore structure of geopolymer aggregate concrete

Figure 6 uses a color-coded pore distribution to illustrate the 3-D structure of geopolymer aggregate concrete and natural aggregate concrete at 7 and 28 days. The 3-D analysis allows void size distribution, visual inspection of selected voids, as well as categorization of voids. The pores detected through CT analysis include partial capillary pores, ITZs. The air voids and the total voxel count is the main indicator used to acquire the total porosity. The analysis shows 4.74%, 4.25%, 1.87%, and 1.72% for the GPA-7d, NA-7d, GPA-28d, and NA-28d concrete samples, respectively. It is noted that both concretes displayed a 60% reduction in porosity between 7 and 28 days. This is attributed to the ongoing hydration process filling the gaps/voids. Neville²² reported that the porosity of natural aggregate concrete generally varies between 10 and 40%, based on mercury intrusion porosimetry (MIP) analysis. The difference between the X-ray CT measurement and MIP values may be attributed to the gel pores, which occupies approximately 28% of the total volume of gel.²²

Figure 6 shows a higher heterogeneity of the porosity distribution along the Z-axis. In the bulk cement matrix, the pores determined at different heights revealed that there were some larger void spaces, such as air voids and ITZs. During mixing and casting, air bubbles are entrained or entrapped, and anhydrous grains can produce a loose arrangement in the vicinity of aggregates due to the wall effect. Sofie⁴¹ mentioned that porosity distributions along the Z-axis may be closely linked to the casting direction, and the random distributions of hydration products of cement, aggregates, formed pores, and ITZs. The 3-D images of both concretes represent the complete 3-D pore network, showing the pore connectivity and tortuosity. Geopolymer aggregate concrete displays a long pore network penetration with higher pore connectivity at an early age. However, this decreased with time and showed significantly lower pore connectivity in the 3-D pore network at 28 days. Indeed at 28 days, both geopolymer aggregate and natural aggregate concretes showed similar pore distributions and pore connectivity.



Fig. 7—2-D view with color-coded void distribution of XY, YZ, and ZX planes.

This is due to the densification of the ITZ and a decrease in voids at the gel-aggregate interface.

Figure 7 presents the two-dimensional (2-D) view with color-coded void distribution in three different planes. Most of the voids were distributed within or close to the ITZ. This cement-aggregate interface has a different microstructure, consistent with the microcracks observed in the SEM images at 7 days. These cracks would allow the generation of more macro-pores and voids in the weakest areas of the ITZ. However, the CT scan images of XY, YZ, and DX planes at 28 days show a decrease in the number of voids in this region. This again can be attributed to the hydration mechanism that fills the microcracks, pores, and voids. Hence,

the increase in compressive and flexural strength observed at 28 days.

Figure 8 shows the changing total pore volume distribution of geopolymer aggregate and natural aggregate concretes between 7 and 28 days. The histograms show the range of voids. The largest detected void is 3,503,380 μ m³ (1.237 × 10⁻¹⁰ ft³) and the smallest is 399,322 μ m³ (1.41 × 10⁻¹¹ ft³). The trend indicated that most pores are of the smallest size interval. It is noted that total pore volume decreased between 7 and 28 days. This is again well correlated with the SEM analysis. The bulk cement matrix is similar in both concretes; thus, the hydration and gel formation increase the compactness of the ITZ.



Fig. 8—Histogram of total pore volume distribution of GPA and NA concretes at: (a) 7; and (b) 28 days. (Note: $1 \ \mu m = 3.937 \times 10^{-5}$ in.)

SUMMARY AND CONCLUSIONS

The following conclusions can be drawn based on the systematic experimental study:

1. Concrete with mean compressive strengths up to 40 MPa (5.8 ksi) can be produced using the novel geopolymer coarse aggregate.

2. Geopolymer aggregate concrete showed similar initial and final setting time to the natural aggregate concrete.

3. The dry density of the geopolymer aggregate concrete was being 2140 kg/m³ (133.60 lb/ft³) at 28 days compared to 2645 kg/m³ (165.12 lb/ft³) for the natural aggregate concrete.

4. At 7 days, a greater number of microcracks were evident in the cement-geopolymer aggregate transition zone than in the cement-natural aggregate interface, creating a crack network in the geopolymer aggregate concrete. However, at 28 days, the geopolymer aggregate concrete displayed a well-compacted and denser ITZ, similar to the natural aggregate concrete.

5. The rough surface of GPA and different fracture planes obtained due to aggregate production process are believed to enhance the cement paste-aggregate bond in GPA concrete.

6. The increased density of the ITZ at 28 days strengthens the aggregate-matrix bond, which results in an increase in the compressive and flexural strength of the geopolymer aggregate concrete at 28 days.

7. Geopolymer aggregate concrete showed 60% reduction in porosity compared to 7 days. Overall, the geopolymer aggregate concrete showed a similar pore distribution and pore connectivity to natural aggregate concrete at 28 days.

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