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Effects of micro-structure characteristics of interfacial transition zone on the compressive strength of self-compacting geopolymer concrete

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HIGHLIGHTS

- The goepolymer concrete is allowed to compact in its own weight.
- Different test methods are used to assess the workability of geopolymer concrete.
- The functional relationship between compressive strength and ITZ thickness is determined.

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ABSTRACT

This paper presents an experimental study of the influence of different superplasticizer dosage on compressive strength and micro-structure characteristics of interfacial transition zone (ITZ) prepared with fly ash based self-compacting geopolymer concrete (SCGC). The correlations between compressive strength development and microstructure of interfacial transition zone were also investigated. Concrete specimens were prepared with different superplasticizer (SP) dosage namely 3%, 4%, 5%, 6% and, 7% and cured at 70 °C for duration of 48 h. Field emission scanning electron microscope (FESEM) observations revealed that improved performance of concrete was found when the compressive strength increased through formation of dense ITZ between the aggregate and binder matrix at higher SP dosage. There are good correlations between compressive strength and micro-structure characteristics of interfacial transition zone. The FESEM analysis revealed that relatively a loose and porous interfacial zone was found between the binder and aggregate for low SP dosage and theses loose and porous ITZ decreased the performance of concrete by lowering the compressive strength; however, a dense ITZ was found between the aggregate and binder matrix for higher SP dosage that enhanced the concrete performance by increasing the compressive strength.

1. Introduction

Since last few years there is a great pressure on industries contributing to greenhouse gas emission to improve and upgrade the level of environmental impacts, global cement industry is one of them. The global annual consumption of cement is on a continuous increase that causes the significant increase in the carbon dioxide (CO_2) emission to the atmosphere and it also consume enormous amount of rock extraction for quarried and minerals that may lead to deplete at one point of time. Manufacture of one ton of Portland cement (PC) generates nearly one ton of CO_2 to the environment depending on the production process used which represents 5% global CO_2 emission [1,2]. It is mentioned that the annual global manufacture of PC contributes about 1.35 billion tons of greenhouse gas emissions [3,4]. As a result of the manufacturing of PC, the CO_2 emission is likely to increase by about 50% from the current levels in 2020 [5]. To keep the global environment safe from the consequence of cement production, it is essential to explore the alternative materials that can completely or partially eliminate the use of cement in concrete and cause no environmental destruction. Until now, extensive efforts have been made to partially replace the use of cement in concrete. These include incorporation of waste by-products as cement replacement in concrete production and development of binder alternative to PC [6]. In recent times, to lower the environmental impact due to cement manufacturing, a new binding material is made from an alumino-silicate precursor activated in high alkali solution. This binding agent is known as geopolymer cement.

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Geopolymer concrete (GC) is one of the recently developed construction material [2], which in next future could partially replace cement from concrete industry. Geopolymer comprises of two main components, namely the source materials and the alkaline liquids. The source material must be rich in silica (Si) and alumina (Al) content to have a full potential to be used for alumino-silicate based geopolymer concrete. The by-product materials, such as fly ash [7], rice husk ash or silica fume [8] and GGBS [9] or natural minerals such as kaolinite and clay [10] could be used as source materials. The alkaline liquids could be from soluble alkali metals that are usually sodium or potassium based solution and the most common alkaline liquid used in GC is a combination of sodium hydroxide (NaOH) or potassium hydroxide (KOH) and sodium silicate (Na_2SiO_3) or potassium silicate (K_2SiO_3) [11]. Self-compacting geopolymer concrete (SCGC) can be considered as an advanced and innovative construction material in the concrete technology. As the name implies, it does not need any compacting efforts to achieve full compaction and utilizes fly ash together with alkaline solution and superplasticizer as a binder for matrix formation and strength [12]. The geopolymer paste is used to bind the loose coarse aggregates, fine aggregates and other unblended materials together in the presence of superplasticizer to attain the required workability for SCGC. This improved concreting operation which could offer many benefits and advantages over conventional concrete. These include; ease of filling in restricted and narrow sections, enhanced consolidation around reinforcement and bond with the reinforcement, better quality of concrete, reduction of on-site maintenance, faster rate of construction, lower overall costs, and improved concreting operation [13–15]. A significant progress of health and safety could also be achieved through reduction of CO_2 emission due to elimination of Portland cement production, suspending the use of vibrators and considerable minimization of environmental noise loading on and around a construction site [13,14].

The sustainable production of self compacting geopolymer binder hinges on controlling the mix proportion, determining the right quantities of NaOH and Na_2SiO_3 solution required to activate the source material Fly Ash and optimizing the superplasticizer dosage. The composition of SCGC mixes includes substantial proportions of fine-grained inorganic materials and this gives possibilities for utilization of mineral admixtures such as fly ash, which are currently waste products with no practical use and are costly to dispose-off. The particles fraction of size less than 0.125 mm is considered as the fines content of the geopolymer paste and should also be taken into consideration in calculating the water to geopolymer solids ratio because the water to geopolymer solids ratio is an important parameter to control the workability as well as the compressive strength of SCGC. Moreover, the fine aggregates significantly affect the fresh properties of SCGC than the coarse aggregate. In self-compacting concrete (SCC) mixes, it is believed that the high volume paste helps to minimize the friction between the sand particles and enhance the workability and use of improved particle size distribution is also very important. Self-compacting concrete mix design methods mainly use blended sands to achieve an optimized aggregate grading curve. The major role of superplasticizer in SCGC is to get adsorbed on to binder grains, impart a negative charge to them, which repel each other and get deflocculated and dispersed [15]. This gives better workability and superior performance for the SCGC by improving the plastic and hardened properties, enhances the microstructure and leading to higher compressive strength [1]. Similarly in cement based SCC, superplasticizer is used to produce flowable concrete in cases where placing in hard to reach areas or locations. Interfacial transition zone (ITZ) is the interface between mineral aggregates and cement binder in a cementitous system. Sun et al. [16] reported that the coarse aggregate surface has an ITZ of about 100 μm depth with two layers; one is a duplex film at the surface of the coarse aggregate which is about 1 μm deep with $Ca\ (OH)_2$, CH crystals and C–S–H gels as its components. The other layer is the porous transition zone which is about 20–100 μm deep, which contains more CH crystals, some C–S–H gels and little ettringite (needle like hydration product). In geopolymer matrix, these nanoporous sponges like amorphous product is known as geopolymer micells. These two layers combined to form the ITZ. Interfacial transition zone in fly ash based geopolymers can be defined as the interface between mineral aggregates such as sand and/or natural rocks, and geopolymer binder.

Many studies have been conducted on the microstructure, short-term and long-term behavior of normal geopolymer concrete incorporating waste materials [1,4,6,11,16]. However, this study is mainly focused on the assessment of workability and compressive strength of SCGC and the correlation of compressive strength and microstructure properties of ITZ thickness. Due to the relevance of ITZ in determining the enhancement of microstructure and compressive strength of SCGC, this study has analyzed the width of the transition zone using Field Emission Scanning Electron Microscope (FESEM). The FESEM technique was used to identify the changes occurred in the microstructure of the formed SCGC and Interfacial transition zone (ITZ). Investigation of ITZ is very crucial since the ITZ is known to have a different microstructure from the bulk of hardened geopolymer paste and the interface is also considered as the locus of early cracking.

The main objective of this study was to determine the effects of the microstructure characteristics of ITZ on the compressive strength of SCGC. The Field Emission Scanning Electron Microscopy (FESEM) analysis was applied to visualize the internal microstructure of the SCGC composite. The relationship between change in the compressive strength development and that in the microstructure was also investigated by studying the implications of the microstructure of the ITZ on the compressive strength development of SCGC.

2. Methodology

2.1. Materials and mix proportion

For this experimental work, materials were chosen according to the specifications that meet the requirements of British Standards and EFNARC guidelines [17]. Dry low-calcium fly ash obtained from thermoelectric power station was used as source material. The fly ash utilized in this study was ASTM C618 Class F with chemical composition, as analyzed by X-Ray Florescence (XRF) is shown in Table 1.

Coarse aggregate used in this study was crushed granite stone with maximum size of 14 mm [18] and specific gravity of 2.66 at saturated surface dry (SSD) condition while the fine aggregate used is dry clean natural Malaysian sand with the fineness modulus of 2.76, maximum size of 5 mm and a specific gravity of 2.61.

In geopolymer synthesis, alkaline solution plays a major role in the dissolution of silica and alumina from the source material as well as in the catalysis of polymerization reaction [19]. A combination of sodium silicate and sodium hydroxide was chosen as an alkaline liquid for this experiment [11].

Na_2SiO_3 (Grade A53) used with a composition of 55.52% water, 29.75% SiO_2 and 14.73% Na_2O. NaOH (99% purity, in the form of pellets) was mixed with distilled water to avoid the effect of unknown contaminants in the mixing water. The

Table 1
Chemical Composition of low calcium fly ash (LCFA) [22].

Compounds	Mass (%)	Requirement as per BS EN 450-1:2005
SiO_2	51.3	Min. 25%
Al_2O_3	30.1	–
Fe_2O_3	4.57	–
$SiO_2 + Al_2O_3 + Fe_2O_3$	85.9	Min. 70%
CaO	8.73	Max. 10%
P_2O_5	1.6	–
SO_3	1.4	Max. 3%
K_2O	1.56	–
TiO_2	–	–

activator alkaline solution was prepared at least one hour prior to its use. For all mixes, the concentration of solution was kept at 12 M and in order to make 1 kg of solution, 36.1% of pellets were added.

Superplasticizer (Sika Visco Crete-3430) was used to attain the required workability for fresh SCGC. The amount of SP used was in accordance with EFNARC 2002 [20]. The water added to the mix was tap water in accordance with BS EN 1008 [21].

The mix design proportion adopted in the research is shown in Table 2. A total of five mix samples S_1, S_2, S_3, S_4, and S_5 were prepared to investigate the effects of superplasticizer dosage on the fresh workability, hardened compressive strength and microstructure properties of SCGC. For all mixes, the designed SP dosages were 3%, 4%, 5%, 6% and 7% and all the other test parameters were held constant. For all mixtures, the ratio of sodium silicate to sodium hydroxide solution by mass and the mass ratio of fine aggregate to fly ash were 2.5 and 2.125 respectively.

2.2. Mixing, casting and curing

The concrete mixing procedure consists of dry and wet mixings. The solids components of SCGC, i.e. the fly ash, fine and coarse aggregates, were dry mixed in the pan mixer for about 2.5 min. Then the dry mixing is followed by wet mixing whereby the liquid part of the mixture, i.e. the sodium silicate solution, the sodium hydroxide solution, extra water and the superplasticizer, were premixed thoroughly before being added to the dry mixture [6] and the wet mixing continued for another 3 min.

After mixing, the fresh concrete was then filled into 100 mm × 100 mm × 100 mm steel moulds and allowed to fill all the spaces of the moulds by its own self weight without any compacting efforts. After casting the specimens were put in an oven at a temperature of 70 °C for curing with duration of 48 h. The oven curing was followed by post curing by putting the sample outside the room but protected from direct sunlight and rain. This is called post curing effect or stabilization period. The days of testing was counted starting from this stabilization period. The oven curing followed by post curing was adopted in this research to accelerate geopolymerization process at elevated temperature and to enhance the compressive strength performance as claimed by Nuruddin et al. [23]. The successful production of SCGC using oven curing facilitates further explorations, to apply in buildings and civil works and wider use. The reported compressive strength is the average strength of three specimens. The test specimens were then left at room temperature until the specified days of testing.

2.3. Testing procedure

2.3.1. Workability test

Filling ability, Passing ability and Resistance to segregation are the three fresh concrete characteristics mandatory for SCC. These three characteristics of workability should be satisfied to consider the concrete mix as SCC.

The three properties of SCC can be assessed using different test methods; however, so far there is no standard method to determine all the relevant workability aspects at a time, so each designed mix samples should be tested by more than one test method for the different workability characteristics. Filling ability and passing ability can be examined by the test methods shown in Table 3 and the resistance to segregation can be checked more or less in all tests based on observation through visual stability. The European Guidelines EFNARC has proposed different test methods to characterize an SCC mix. Test methods and workability properties along with their recommended values given by EFNARC are shown in Table 3.

In this research, slump flow, T-50, V-funnel, L-Box and J-Ring tests were carried out to fully characterize the workability properties [12] and these tests are performed in accordance with EFNARC guidelines.

2.3.2. Compressive strength and microstructure properties test

Compressive strength test on concrete determines its performance under compressive loads; the specimen is subjected to compressive load and the maximum sustained load is recorded.

In the present study, the hardened compressive strength test was performed on cube of 100 mm in accordance with BS EN 12390-3:2002 [24] using 2000 KN capacity digital compressive testing machine. A set of three cubes for each mix were tested at the ages of 1, 3, 7 and 28 days for compressive strength measurement and the average strength was presented.

The field emission scanning electron microscope analysis was carried out to identify the inner microstructure of SCGC samples containing different superplasticizer dosages namely 3%, 4%, 5%, 6%, and 7%. For microscopy analysis, sample preparation is very crucial to obtain reliable FESEM data on ITZ identification. Three samples were tested for each variable at ages of 28 days and test cubes of 100 mm were cored as cylindrical sample of 1 inch diameter and 100 mm thickness from the center of the cube in vertical direction using rock coring machine. Then using the rock cutting machine, the cylindrical sample were cut into slice of 5 mm thickness from different points in the vertical direction, one at the center, and the other two were taken randomly from the different points in the vertical direction, one near to the bottom and the other was taken near to the top of the cylindrical sample. The FESEM micrographs of prepared specimen from three different positions were taken to provide uniformity of analysis. The correlation between compressive strength development and ITZ thickness were determined for all samples, i.e. for the sample taken at the center, near to the top and bottom of cylindrical samples. The average of ITZ thickness measurement was taken in determining the relation between compressive strength development and ITZ thickness.

3. Result and discussion

3.1. Workability test results

In this section, the experimental results of different workability properties tested by slump flow test (slump flow diameter and $T_{50\ \mathrm{cm}}$), J-ring test (J-ring Blocking step (B_J)); L-box test (ratio of heights at the two edges of L-box (H_2/H_1)); V-funnel test (time taken by concrete to flow through V-funnel after 10 s T_{10S}); for various mix compositions are discussed. The results of the workability tests are shown in Table 4.

A total of five mixtures were made to study the influence of superplasticizer on the workability, compressive strength and microstructure properties of SCGC. High workability is the main criteria to make the geopolymer concrete self-compactable and to obtain the required compressive strength and microstructure properties. The test results of the quantitative analysis and visual

Table 2
Mix design proportion.

Mix sample	Fly ash	Coarse aggregate	Fine aggregate	NaOH		Sodium silicate	Extra water		Super		Curing	
	(kg/m^3)	(kg/m^3)	(kg/m^3)	(kg/m^3)	Molarity	(kg/m^3)	(kg/m^3)	(%)	(kg/m^3)	(%)	Time (h)	Temperature (°C)
S_1	400	950	850	57	12	143	48	12	12	3	48	70
S_2	400	950	850	57	12	143	48	12	16	4	48	70
S_3	400	950	850	57	12	143	48	12	20	5	48	70
S_4	400	950	850	57	12	143	48	12	24	6	48	70
S_5	400	950	850	57	12	143	48	12	28	7	48	70

Table 3
Test methods, property and recommended values as per EFNARC guidelines [20].

S. no.	Methods	Workability property	Acceptance values as per EFNARC Guide lines	
			Minimum	Maximum
1	Slump flow by Abrams cone	Filling ability	650 mm	800 mm
2	$T_{50\ \mathrm{cm}}$ slump flow	Filling ability	2 s	5 s
3	V-funnel	Filling ability	6 s	12 s
4	L-Box (H_2/H_1, ratio)	Passing ability	0.8	1.0
5	J-Ring	Passing ability	0 mm	10 mm

Table 4
Workability and compressive strength test results.

Mix sample	Slump flow (mm)	$T_{50\text{ cm}}$ Slump flow (s)	V-funnel flow time (s)	L-Box (H_2/H_1) ratio	J-Ring blocking step, B_J (mm)	Compressive strength			
						1-Day (MPa)	3-Days (MPa)	7-Days (MPa)	28-Days (MPa)
Workability test results									
S_1	625	6.5	15.5	0.84	13	40.85	41.77	42.84	44.69
S_2	640	6.0	14	0.88	10	42.02	42.68	44.17	46.86
S_3	665	5.0	12.5	0.9	8	44.74	45.28	46.19	48.90
S_4	690	4.5	10	0.94	7	47.83	48.52	49.44	51.52
S_5	710	4.0	7.0	0.96	5	51.03	51.98	52.26	53.08
Acceptance criteria for SCC as per EFNARC [20]									
Min.	650	2	6	0.8	0				
Max.	800	5	12	1.0	10				

observations showed that mix samples S_1, S_2 and S_3 with 3%, 4% and 5% respectively are failed to exhibit the required workability due to insufficient amount of superplasticizer that made the mixes less workable; however, mix samples S_4 and S_5 with 6% and 7% SP respectively had the desired fresh properties and the workability properties were within the EFNARC limits of SCC. As expected, it was observed that with the increase in SP dosage the workability increased. The mixes with 6% and 7% SP improved the workability to the extent required for SCC by coating themselves around the binder particles and transfer them a highly negative charge so that they repel each other. This in turn results deflocculation and dispersion of binder particles to enhance the workability to the extent required for self-compactability.

3.2. Compressive strength

Compressive strength is a major property used to assess the performance and quality of almost all types of concrete. From Fig. 1, it can be seen that mix sample S_5 with SP dosage of 7% shows highest compressive strength at all days of testing as compared to the other mixes that have lower SP dosage of 3%, 4%, 5%, and 6%. Fig. 1 shows that the maximum compressive strength performance was at 7% SP dosage for all days of testing. The maximum compressive strength was achieved at 28 days of age, which is, 53.80 MPa. SP is required in geopolymer concrete to improve not only workability but also enhances the hardened compressive strength of SCGC. Heat should be provided to facilitate the hardening process as the condensation polymerization that takes place during hardening process is endothermic in nature. This phenomenon is different from OPC based concrete as geopolymer concrete does not involve water in its geopolymerization reaction. Water is used to wet the solid components of the mixture and in the mix it plays a vital role in synthesis and acts as a medium for dissolution, condensation and polymerization of Al and Si precursors into polymeric structures [11]. This in turn helps the mixing and casting process to increase the workability of fresh concrete together with superplasticizer. From Fig. 1 it can be shown that as the SP dosage in the mix increases, the compressive strength also increases. This was due to the more effective action of the superplasticizer in increasing the workability of the mix. Microcracks and pores were observed at lower SP dosages which could cause the premature failure of concrete at lower stress level when the sample is subjected to compressive load. Fig. 1 shows that mix contained 7% SP produced the highest compressive strength of 53.80 MPa at 28 days. It was observed a slight increase in the compressive strength for mix contained 7% SP as compared to mix with 6% SP. SP dosage of 6% was taken as the optimum due to the fact that satisfactory performance was obtained in both fresh and hardened SCGC. Compressive strengths of 51.52 MPa and 53.80 MPa were obtained for 6% and 7% SP at 28 days of testing respectively and no significant difference in terms of compressive strength for 6% and 7% SP use. Reduction of SP dosage by 1% is very important for bulk quantity production in the construction industry as far as economy is concerned.

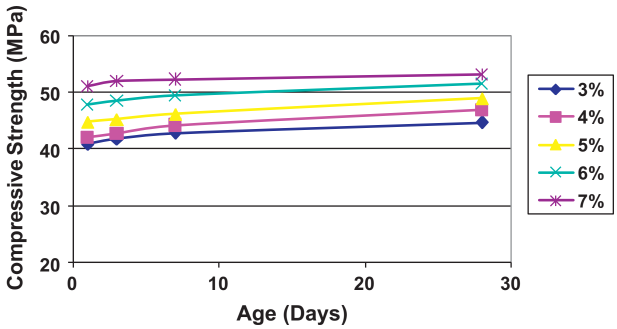


Fig. 1. Effect of SP dosage on compressive strength.

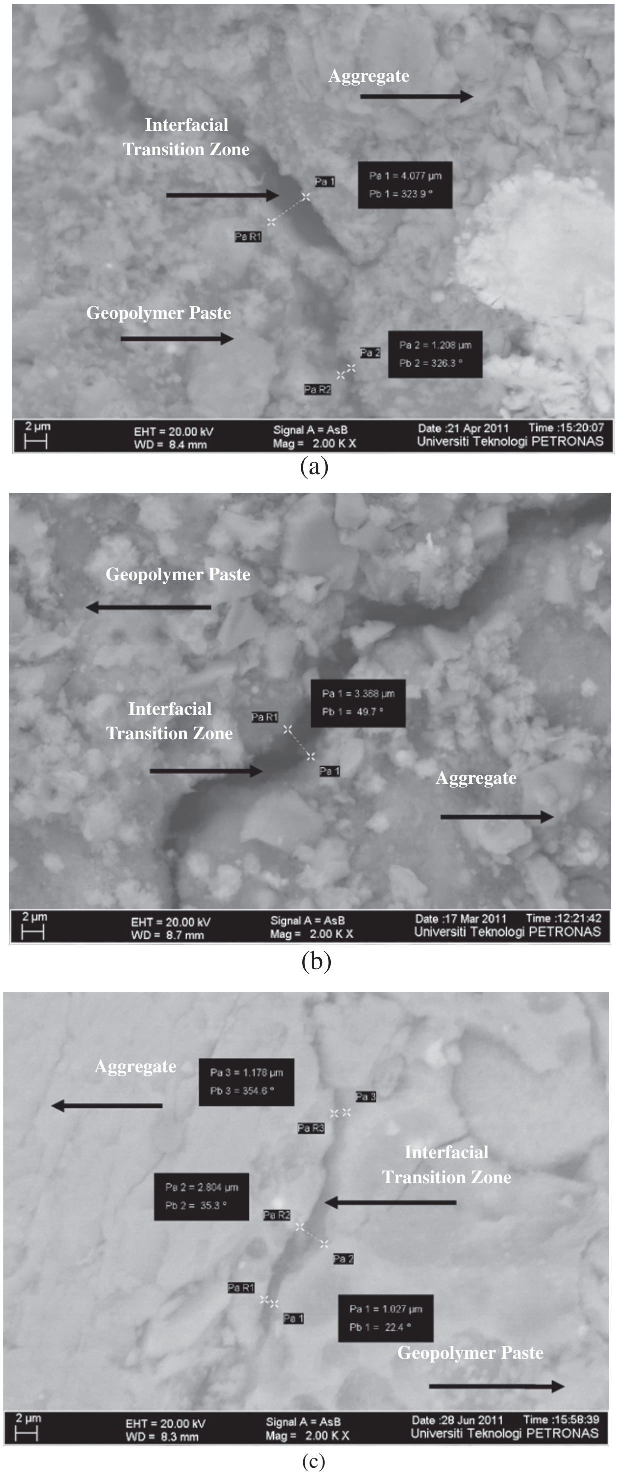
3.3. Microstructure analysis of SCGC

In the following sections, the effects of SP on interfacial transition zone were investigated. The different SP dosages were 3%, 4%, 5%, 6% and 7%. In the investigation of microstructure properties of SCGC, the effect of SP on microstructure properties was assessed due to the fact that superplasticizer played a major role in making geopolymer concrete self-compactable. Investigation of ITZ is very crucial since the ITZ is known to have a different microstructure from the bulk of hardened geopolymer paste and the interface is also considered as the specific location of early cracking. The ITZ measurement manifested that micropores was observed for the mix contained 3%, 4% and 5% SP. Nanopores was observed for 6% SP dosage and no apparent ITZ was observed for 7% SP dosage. The width of the ITZ to which the transition zone between the geopolymer paste and aggregate ranged from 2.8–4.08 μm for 3% SP dosage, 1–1.8 μm for 4% SP dosage, 1.1–1.27 μm for 5% SP dosage, 710ηm-960ηm (0.71–0.96 μm) for 6% SP dosage and no apparent ITZ was observed for 7% SP dosage.

Porosity in SCGC was the volume not filled by fly ash grains and/or geopolymerization products. Quantitative image analysis in cement based concrete clearly showed that ITZ is caused by the disorder of packing the anhydrous cement grains in the transition zone as claimed by Scrivener et al. [25]. Similarly in SCGC, ITZ was caused by incomplete packing of unreacted fly ash microsphere particles in the transition between the geopolymer paste and aggregates since the composition of alumino-silicate gel formed by the reaction between fly ash and alkaline solution was variable and highly dependent on the type, concentration and reactivity of alkaline solution. This phenomenon was due to incomplete dissolution of large proportion of fly ash and the existence of different composition and size of unreacted fly ash particles. The ITZ was a region of transition which is highly heterogeneous, not uniform, varies from point to point along each aggregate particle.

As shown in Fig. 2, the utilization of 3% SP showed lower strength and stiffness of the ITZ which resulted in less dense microstructure of SCGC since ITZ tends to act as "the weak link in the bond" when compared to the bulk geopolymer paste and the aggregate particles. The width of ITZ in SCGC mix that contains 3% SP is about 2.8–4.08 μm, considerably showed enhanced micrcostructure as compared to cement based concrete which has an ITZ thickness of about 20–100 μm.

It was observed that a small decrease in ITZ width as the SP dosage increased from 3% to 4%. The noticeable width of ITZ for 4% SP

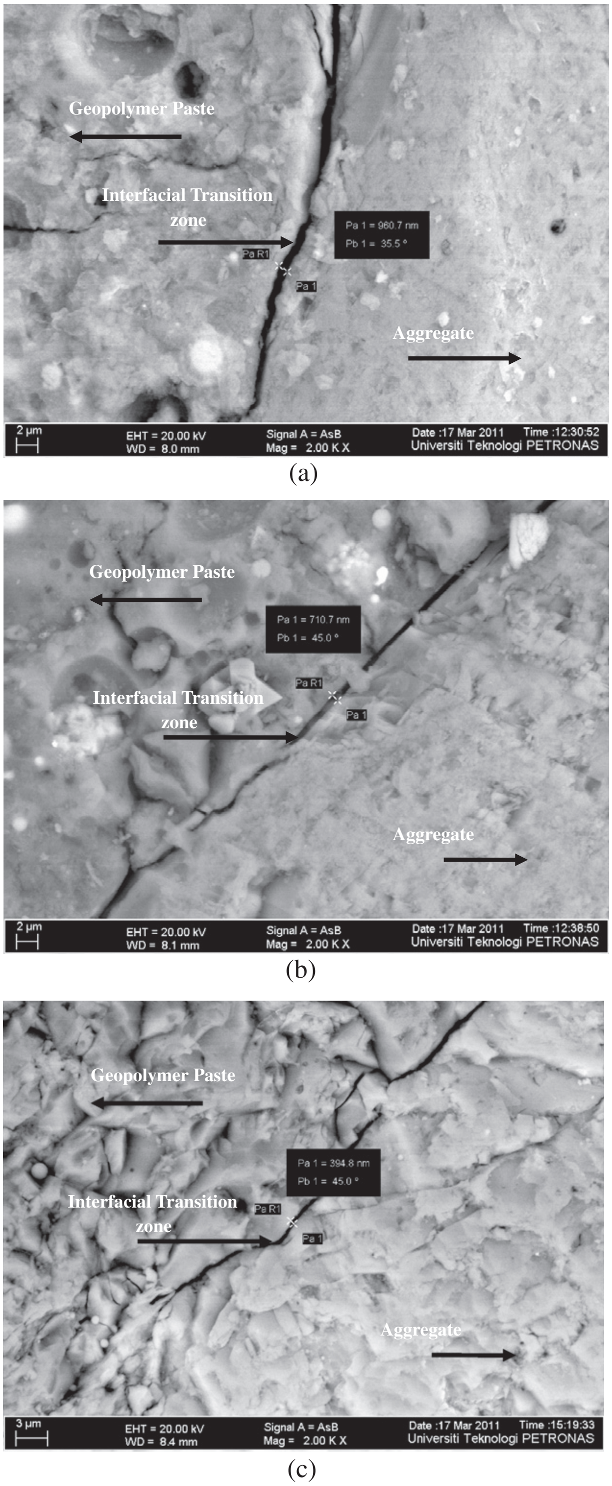


(a)

(b)

(c)

Fig. 2. FESEM micrographs of SCGC samples for 3% SP: (a) sample taken from the center of the cube, (b) sample taken near from top of the cube, (c) sample taken near from bottom of the cube.



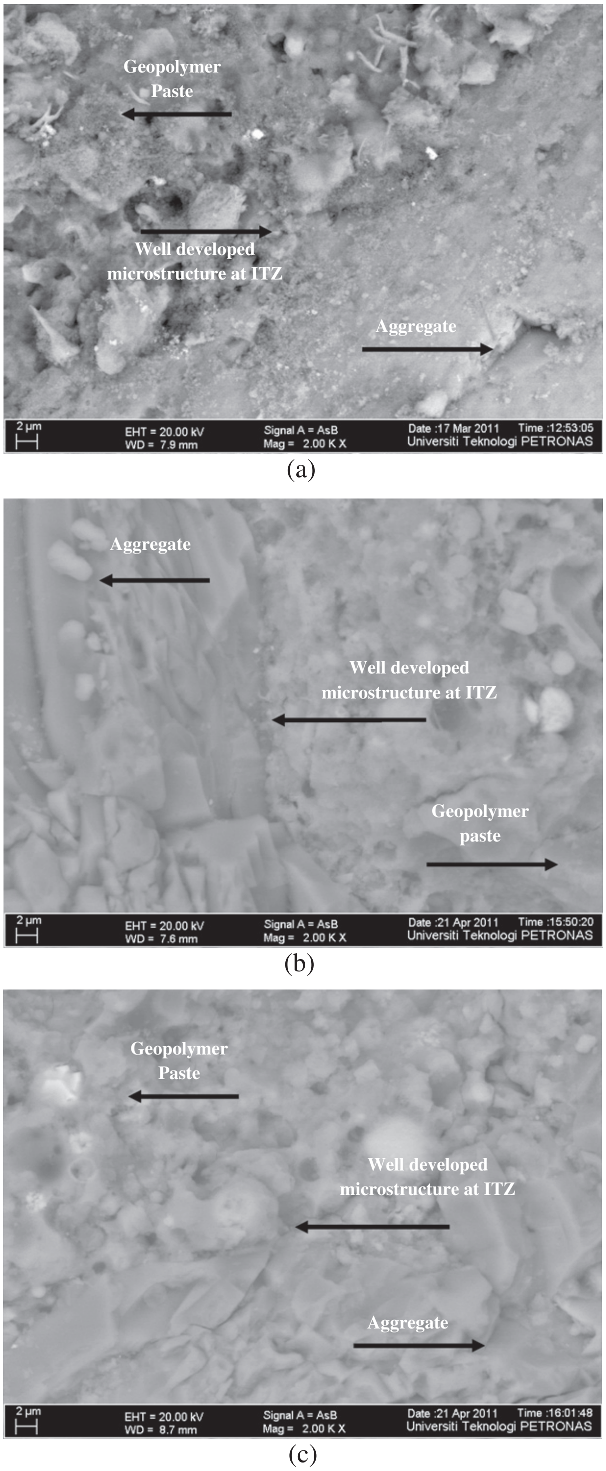
(a)

(b)

(c)

Fig. 3. FESEM micrographs of SCGC samples for 6% SP: (a) sample taken from the center of the cube, (b) sample taken near from top of the cube, (c) sample taken near from bottom of the cube.

dosage in which the transition between geopolymer paste and aggregate is clearly observed, which is 1–1.8 μm, appears to be slightly narrower than the concrete sample which contains 3% SP with the value of 2.8–4 μm. The presence of the gap between the aggregate and geopolymer paste was due to insufficient dosage of SP that made the paste less workable and decreased its bond with the aggregate. Studies have manifested that microcracks at the interface between coarse aggregate and cement paste exist even prior to the application of load on concrete due to unavoidable differences in mechanical properties between hydrated cement paste and the coarse aggregate along with thermal movement and shrinkage. Since cracking is the cause for failure of concrete, efforts were made to reduce this inevitable cracking from the concrete by increasing the SP content.



(a)

(b)

(c)

Fig. 4. FESEM micrographs of SCGC samples for 7% SP: (a) sample taken from the center of the cube, (b) sample taken near from top of the cube, (c) sample taken near from bottom of the cube.

It is generally deduced that, as the width of ITZ decreases, the SCGC compressive strength increases. If it is required to manufacture concrete with improved strength, it is necessary to refine the interface between the aggregate and geopolymer paste. An FESEM micrograph of the specimen demonstrates that a highly complex product morphology that consists of unreacted, partially reacted, and completely reacted fly ash spheres that are surrounded by geopolymer matrix. Increasing the superplasticizer content has much effect on microstructure enhancement. Due to this reason the main target is to reduce the ITZ width as crack usually occurs from this interface zone and it is believed that ITZ can be improved efficiently by utilizing relatively high amount of superplasticizer content in the mix.

The ITZ of mix sample with 6% SP dosage is shown in Fig. 3, which is much denser than the ITZ of mix samples with 3%, 4% and 5% SP dosages. It demonstrates that the geopolymerization reaction in 6% SP utilization significantly enhances the microstructure properties of SCGC. It was observed that a marked difference in ITZ width between 3% and 6% SP dosage. The porosity of the ITZ and its width considerably decreased as compared to 3%, 4% and 5% SP dosage. Therefore the utilization 6% SP is a more efficient way of densifying (refining) the ITZ, because it avoided many of the pores, disconnect the microcrack path and making its structure more homogenous in composition. Such method can provide more effective means of attaining better/enhanced microstructure. Therefore, for high performance concretes, such as self-compacting, high durability or high strength concretes, enhanced microstructure properties from refinement in the properties of ITZ seem to be more significant if it is required to achieve this new material with full potential. The weak ITZ almost vanished while the SP increased to 6% and the microstructure of the samples appeared as geopolymer matrix comprising of partially reacted spherical particles and gel phase. The amount of resulting alumino-silicate gel and good interface between aggregate and alumino-silicate gel influenced much on the development of compressive strength.

It was found that when the liquid component of SCGC that contained little SP dosage, the compressive strengths of the geopolymeric binders and concretes were significantly weaker than those mixed with high dosages of SP. No apparent interfacial transition zone (ITZ) could be identified near the aggregates if the SP dosage were higher.

As a result, stronger aggregate/binder interfaces as well as denser binders were formed with increasing SP dosage. This then led to stronger geopolymeric products including binders, mortars and concretes. If the interfacial bonding between aggregates and geopolymeric binder activated with alkaline solution and low dosages of SP was interrupted by, for example, SP dosage, the overall mortar and concrete strengths were significantly weaken as compared to the mix activated with alkaline solution and high SP dosage. Therefore, the interfacial bonding between aggregates and geopolymeric binders is the critical factor in determining the compressive strengths and mechanical strength of SCGC. At 7% SP dosage, the geopolymerization reaction product in ITZ developed more compactly and it is apparent that SCGC with 7% SP improved the ITZ microstructure properties at most. It showed that the existence of microcrystalline nearly amorphous, mainly as alumino-silicate gels from geopolymerization reaction. The utilization of 7% SP

Table 5
Compressive strength and ITZ thickness for different SP dosages for the sample taken from the center of the cube (S_c), near to the top of the cube (S_t) and near to the bottom of the cube (S_b).

SP dosage (%)	Compressive strength (MPa)	ITZ thickness for S_c (μm)[a]	ITZ thickness for S_t (μm)[b]	ITZ thickness for S_b (μm)[c]
3	44.69	2.643	3.368	1.67
4	46.86	1.55	1.727	1.36
5	48.9	1.091	1.151	1.117
6	51.52	0.9607	0.711	0.3948
7	53.8	0	0	0

[a] S_c sample taken from the center of the cube.
[b] S_t sample taken near to the top of the cube.
[c] S_b sample taken near to the bottom of the cube.

made the microstructure of ITZ denser and effectively filled the transition zone as it is shown by FESEM micrograph in Fig. 4.

3.4. Investigation of the relationship between compressive strength and microstructure

It was observed that the development of the interfacial bond varies with utilization of different amount of SP and the developed interfacial bond determines the compressive strength of SCGC. So far the effects of SP on workability, compressive strength and microstructure properties of SCGC have been explained. In this section the different microstructures of ITZ and their relationship with compressive strength development for the sample taken at the center, near to the bottom and top of cylindrical sample are discussed below.

The strength of a linear relationship between compressive strength and microstructure properties of ITZ can be estimated by using Pearson correlation coefficient. Pearson correlation coefficient is widely used in the sciences as a measure of the strength of linear dependence between two variables i.e. the measurement of linear dependence could be for the variables such as compressive strength and ITZ thickness. Correlation of concrete compressive strength with ITZ thickness is a measure of how ITZ and compressive strength move in a relation to each other. If the correlation coefficient is near to unity, it indicates a very strong functional relationship (see Table 5).

Fig. 5 shows that the compressive strength of concrete increases with a decrease in ITZ thickness. Considerable difference in microstructure properties were found for concrete containing different SP dosages and the relationship between compressive strength and ITZ thickness was highly influenced by the quantity of SP as improvement in microstructure properties was observed with the increase in SP use. Well developed microstructure in ITZ was formed at higher SP dosage. The correlation coefficient between ITZ thickness and compressive strength was 0.919 indicating good functional relationship. Fig. 5 indicates that if the ITZ thickness is 0.9 μm, the compressive strength of SCGC is 50 MPa. Similarly Fig. 5 also indicates that if the ITZ thickness is 1.5 μm, the compressive strength of SCGC is 45 MPa. Compared to concrete which has small ITZ thickness and concretes with large ITZ thickness, concrete containing small ITZ thickness has higher compressive strength, whereas, concrete containing large ITZ thickness has a lower compressive strength.

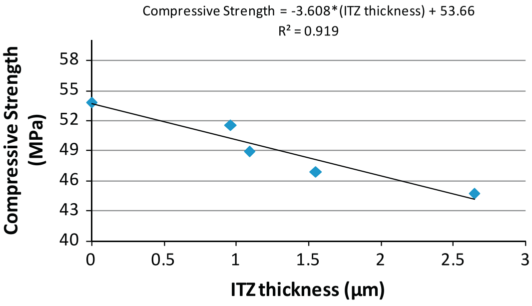


Fig. 5. Compressive strength vs. ITZ thickness for the sample taken at the center.

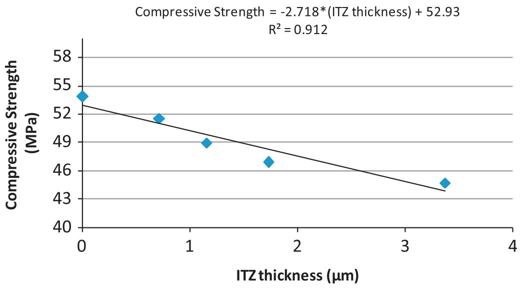


Fig. 6. Compressive strength vs. ITZ thickness for the sample taken near to top of the cube.

Fig. 6 shows that the correlation between compressive strength development and microstructure ITZ thickness for the sample taken near to the top of the cube.

Similarly for the sample taken near the top of the cube, concrete containing small ITZ thickness has high compressive strength. This is because of the formation of dense microstructure at ITZ with the increase in SP dosage and thus leading to less size ITZ which can result improved compressive strength. This is attributed to the refinement of the ITZ with the increase in SP use due to the elimination of many of the pores and disconnection of the microcrack that can make the microstructure more homogeneous in composition.

The ITZ and compressive strength relationship for the sample taken near the top of the cube shows that there is an inverse relationship between them i.e. as the ITZ thickness increases, the compressive strength decreases. If the ITZ thickness is less than 0.7 μm, the compressive strength of SCGC is greater than 50 MPa. Similarly Fig. 6 also indicates that if the ITZ thickness is less than 1.7 μm, the compressive strength of SCGC is greater than 45 MPa.

Fig. 7 shows that the correlation between compressive strength development and microstructure ITZ thickness for the sample taken near to the bottom of the cube.

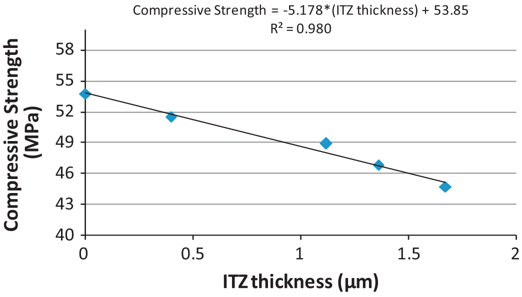


Fig. 7. Compressive strength vs. ITZ thickness for the sample taken near to bottom of the cube.

The ITZ-compressive strength relationship was obtained for the samples taken near the bottom of the cube. Fig. 7 shows that the Pearson correlation coefficient was 0.98 indicating a very strong relationship. The correlation equation for compressive strength based on ITZ thickness will most likely provide an estimate of compressive strength and if the ITZ thickness is less than 0.39 μm, the compressive strength of SCGC is greater than 51 MPa. Similarly Fig. 7 also indicates that if the ITZ thickness is less than 1.5 μm, the compressive strength of SCGC is greater than 44 MPa.

The comparison of SCGC sample containing different SP dosage shows the concrete with lower SP dosage are more porous due to incomplete packing of unreacted fly ash particles in the transition zone. The lowest SP use i.e. 3% SP was insufficient that made the mix less workable and decreased the bond strength between the geopolymer paste and the aggregate. As the geopolymerization progresses, the space between the aggregate and the geopolymer paste was gradually replaced by growing amount of geopolymerization product. Although it is anticipated that the geopolymerization process using 3% SP would create some interfacial bonding effects, the microstructure investigation revealed a relatively porous and loose interfaces. This phenomenon increases the width of ITZ between the geopolymer paste and the aggregate and decreased the compressive strength. The high porosity with low SP content made the aggregate to take up a large amount of water during the initial mixing period and hence the loose interfacial zone in the hardened concrete. A comparatively dense ITZ was obtained with the increase in SP dosage. The mechanism could be that the mix that had high SP dosage and moderate initial water content enhanced the workability of the mix and lowered the initial water to geopolymer solids ratio in the ITZ at early geopolymerization process. Newly formed geopolymer products effectively improved the interfacial bond between the aggregate and the geopolymer binder. Generally, the FESEM analysis revealed that relatively a loose and porous interfacial zone was found between the binder and aggregate for low SP dosage and theses loose and porous ITZ decreased the performance of concrete by lowering the compressive strength while a dense ITZ was found between the aggregate and binder matrix for higher SP dosage that could enhance the concrete performance by increasing the compressive strength.

4. Conclusion

From the experimental investigations, the following conclusions are drawn:

- The utilization of superplasticizer not only improved the workability and compressive strength of SCGC but also enhanced the development of microstructure at the ITZ of concrete.
- It can be seen that SP dosages of 3%, 4% and 5% were found insufficient to produce the required workability such as flowability and resistance to segregation. However, mixes with superplasticizer dosage of 6% and 7% provided the desired workability properties and were within the range of EFNARC limits of SCC.
- 6% SP dosage could result SCGC compressive strength up to 51.52 MPa tested at 28 days. Concrete specimen containing 7% of SP exhibited the highest compressive strength at all ages and enhanced the microstructure properties by refining the ITZ of SCGC at most.
- The microstructural alterations and the different ITZ thickness due to variation of SP dosage affected the compressive strength properties of the concrete. The increase in SP use improved the compressive strength development of SCGC, as well as formed better microstructure by lowering ITZ thickness.
- The compressive strength of concrete has increased with a decrease in the thickness of ITZ and this relationship depends on the dosage of SP use.

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