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Characterization of Spherical Waste CRT Glass as Aggregates in Concrete

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Abstract: *Hazardous cathode ray tube (CRT) glass waste has been used to partially or totally replace natural aggregates in concrete. It is an effective and environmentally friendly method of recycling the increasing number of discarded CRT in the electronic industry. However, little research has been made on other recycling methods, in particular, melting and annealing operations. The typical way is by crushing the CRT funnel glass and grading it to size less than 4.75 mm. The crushing operations have caused the formation of micro-cracks in the glass, led to high concentration of lead leaching. Therefore, this study aimed to investigate the properties of spherical CRT glass (GS) as coarse aggregate in concrete, which was shaped using the techniques melting and annealing. The results demonstrated that the GS is a stronger coarse aggregate than crushed CRT glass (GC) and natural aggregate. The annealing technique has managed to reduce the internal pressure of the glass and strengthens the glass product i.e. GS. Other than that, GS shows the lowest value of specific gravity, unit weight, and percentage absorption. In addition, the use of different morphological features of CRT glass as aggregates are found to be beneficial to concrete strength. The combination of 60% GS and 40% GC has made the coarse aggregates consisting of different shapes and sizes produce a more compacted concrete structure, which significantly lower the percentage reduction of compressive strength between the CRT concrete and the conventional concrete, at only 8.9% loss. The properties of GS are seen as an important step towards hazardous waste CRT glass recycling as a sustainable construction material.*

Keywords: Waste glass, Cathode ray tube, Coarse Aggregate, Concrete

1. Introduction

Over the last few years, the amount of electronic waste (e-waste), especially cathode ray tube (CRT), has been on the rise and it has become an emerging concern in many developing countries. Through a global perspective review, the increasing amount of e-waste in the incoming years is due to the rapid advances in information technology and the increase of market demand for electronic equipment (Singh, Wang, & Li, 2016) (Yoshida et al., 2016). Besides that, the growth of population and economy globally are also the reasons of this increasing amounts. In the year 2012, the world produced about 49 million per tons of e-waste and expected to increase up to 65.4 million per tons in 2017 (United Nations University (UNU), 2013). On a more specific scope, such as in the Southeast Asia in the year 2012, the e-waste generation in Malaysia, Thailand, Indonesia, Philippines, and Vietnam was estimated around 289 thousand tons, 379 thousand tons, 708 thousand tons, 243 thousand tons, and 187 thousand tons, respectively (Yoshida et al., 2016). Meanwhile, in the European Union, 7.5 million per tons of e-waste was generated by 2016 in which 80% of the total e-waste is CRT technology that comes from the old TV sets and computer monitors (Singh, Wang, et al., 2016). In the United States, the similar situation is observed, as the CRTs take the largest part of e-waste stream, about 43% in 2013 and was expected to increase up to 85% for the next 10 years. This is because of around 6.9 million tons of CRTs are still remained to be collected from homes and offices (Shaw Environmental, 2013).

Generally, CRT technology can be found in the old television sets, computer monitors, video cameras, video game machines, automated teller machines, radar displays, and oscilloscopes. But, the advances in the electronic technology led to the continuous replacement of CRT by the new technologies, that is Plasma Display Panels (PDPs) and Liquid Crystal Displays (LCDs) (Rashad, 2015). The replacement of CRT technology has caused an increase of CRTs wastes. In China year 2012, an estimated around 74 million old TV sets and 190 million computer monitors have been outdated, as it is using CRT technology and the amounts are most likely to increase in the coming years. The increment of CRTs wastes can also be seen in Thailand, as in 2004, 750 thousand tons of discarded CRTs was recorded, whilst in 2010, the amount increases about 1.05 million (Singh, Wang, et al., 2016). Other than that, Rocchetti and Beolchini (2014) estimated that in Europe, about 50 thousand to 150 thousand tons of CRT wastes were collected and then treated every year and expected to increase in the year ahead. Zhao, Poon, and Ling (2013) expecting the drastically increase of the amount of CRT disposed of in the year 2050, as it is expected to be 6 times higher than the year 2013. The increasing amount of CRT's waste has become a world concern as the mismanagement of this waste can create a serious impact on the environment. It is because of CRT glass consist of heavy metals especially the high concentration of lead (Pb) metals, up to 39%. The Pb metals contained in the CRT glass is used for protecting the audiences from the radioactivity of the electron gun, as the Pb metals help in eliminate the X-ray radiation (T.-C. Ling, Poon, Lam, Chan, & Fung, 2012). The high concentration of Pb in CRT glass has resulted in high Pb leaching rate, which exceeds the allowable limit. Hence, it should not be disposed in the landfill sites without proper treatment as it has a potential to cause damage in the aspects of acidification, abiotic depletion, and global warming. Besides, it can also bring harm to human body. For that reason, the recycling of CRT waste glass is necessary since it helps preserve the natural resources, lessen the use of landfill space, prevent environmental pollution and harm to human body (Yao, Xie, & Tang, 2016) (Rashad, 2015).

2. Recycling of waste CRT glass review

Generally, there are various recycling techniques which most of it aiming for removing the Pb elements from the CRT glass. It is due to the presence of Pb metals at the inner glass of CRT that hinder the recycling of this waste. The Pb metals that had been extracted from the CRT glass either through lead smelting, mechanical activation, chloride volatilization process, or etc. can be used as the raw material for crystal products and fluorescent tube, which offers an improvement in properties of the product and save a lot of Pb resources. Meanwhile, the CRT glass that frees from Pb metals can be used to make a high-grade product such as glass foams, beads, containers and glass windows. However, these recycling approach increase the treatment cost and may cause secondary pollution (Yu & Liu, 2016) (Singh, Li, & Zeng, 2016).

On the other hand, there are also some researchers that proposed to use the CRT glass, both treated (lead-free) and untreated as a fine aggregate in the production of mortar and concrete (Sua-Iam & Makul, 2012). The presence Pb in untreated CRT glass is very effective in an act as shielding materials for X- and gamma rays. Ling et al. (2012) have crushed the CRT funnel glass (treated and untreated) to a size less than 2.36 mm for use as fine aggregate in mortar, aim for shielding the surroundings from X-ray radiation. The results show the ability of hardened mortar with CRT glass aggregate as radiation-shielding since it can withstand the high-level of X-ray energy. Besides, several studies have proven that CRT waste glass that was crushed to a specific size are an effective replacement material of natural sand in either mortar or concrete production (Sua-Iam & Makul, 2012) (Zhao et al., 2013) (Rashad, 2015). However, the use of crushing techniques to recycle the CRT glass as aggregates has resulted in high Pb leaching rate due to the formation of micro-cracks in the glass. Ling and Poon (2012) reported the increase of Pb leaching values from 14.65 mg/L to 32.72 mg/L as the increase of replacement level of crushed CRT glass as fine aggregates from 50% to 100%. Therefore, this study aimed to investigate other techniques to recycle the waste CRT glass as aggregate, which are melting and annealing techniques. By using these techniques, a new CRT glass product, namely spherical CRT glass (GS) was produced. The characteristics of GS as a coarse aggregate were studied and its effect to the concrete compressive strength was analyzed.

3. Materials and Methods

Production of spherical and crushed CRT glass

The GS and GC were obtained from Nippon Electric Glass Malaysia (NEGM), Selangor, Malaysia. The production process of GS and GC was summarized in Figure 1.

Mix design

The concrete specimens were designed based on the absolute volume as specified in the ACI 211.4R-08. Ordinary Portland Cement was used in this study, which was obtained from Tasek Corporation Berhad, Malaysia. The GS and GC supplied by NEGM will be used as coarse aggregates, where its performance will be compared with the natural coarse aggregates (granite). Meanwhile, river sand was used as fine aggregates. Table 1 shows the mix design of concrete mixtures. The control mixture is referred to as NC, while mixtures GS1.0, GC1.0, and GS0.6GC0.4 are for the GS and GC types and percentage replacement as coarse aggregates.

Testing procedure

Experiments are performed to assess the properties of GS and GC as coarse aggregates, i.e. sieve analysis test, specific gravity, bulk density and absorption, aggregate crushing value (ACV) and aggregate impact value (AIV) test. In addition, the chemical composition and leaching of Pb of GS and GC were determined using x-ray fluorescence (XRF) and toxicity characteristics leaching procedure (TCLP). The strength of concrete mixes containing GS and GC as coarse aggregates was also investigated based on compressive strength at 7, 28, and 56 days.

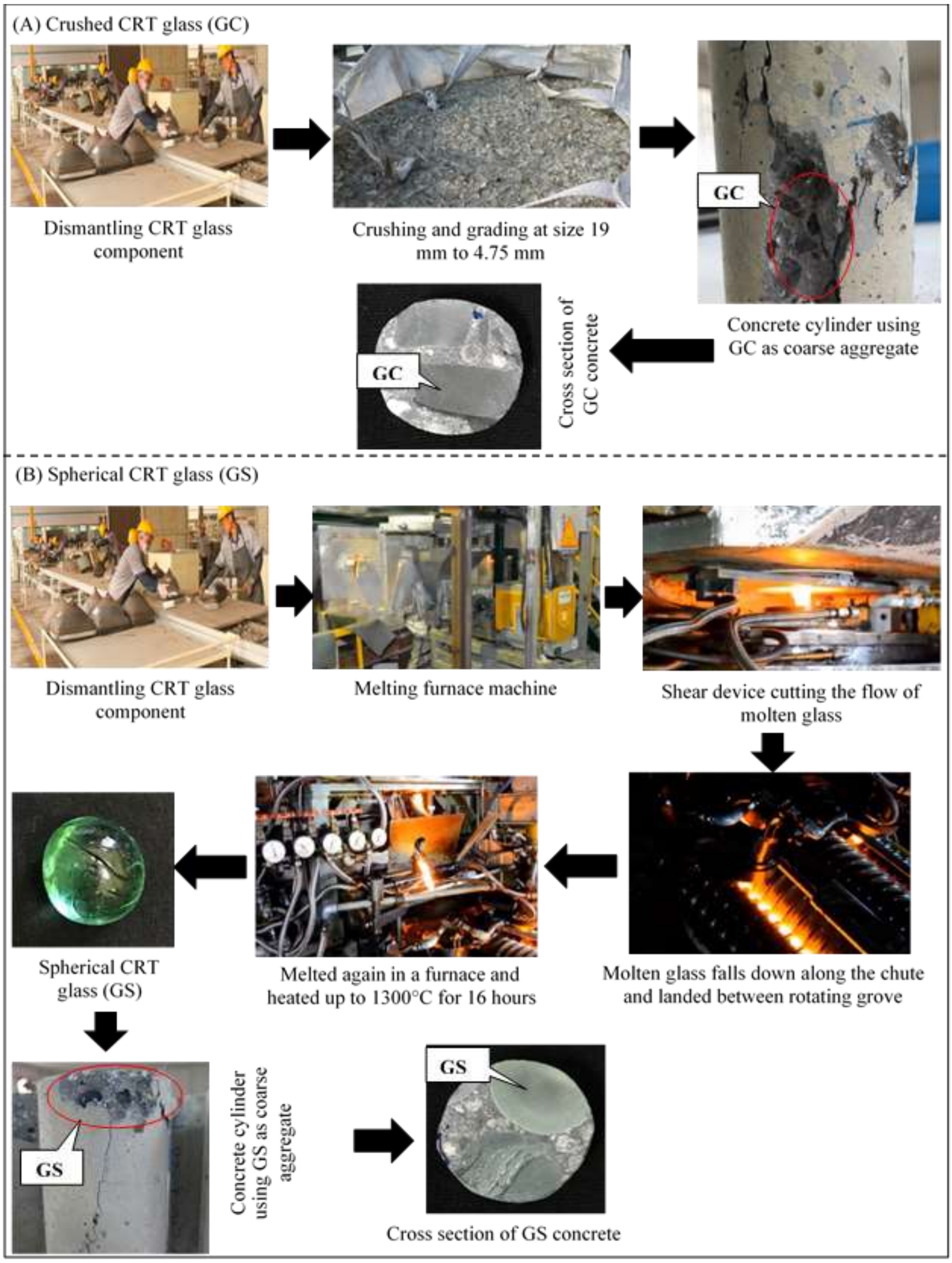

Figure 1: Production process of GS and GC

Table 1: Material proportions of concrete specimens

Mix notations	**Cement (kg/m³)**	**w/c ratio**	**Coarse aggregates (kg/m³)**			**Sand (kg/m³)**	**Water (kg/m³)**
			Granite	**GS**	**GC**		
NC	376.0	0.45	1049.5	0	0	814.4	169.2
GS1.0	376.0	0.45	0	913.2	0	854.0	169.2
GC1.0	376.0	0.45	0	0	1086.2	801.6	169.2
GS0.6/GC0.4	376.0	0.45	0	547.9	434.5	844.3	169.2

4. Results and Discussion

Properties of GS and GC

In normal strength concrete, the aggregates generally occupy 60% to 75% of the concrete volume. Thus, the recycling of CRT waste glass in the form of GS and GC as a substitution of natural coarse aggregates will definitely affect the concrete performances. Table 2 shows the physical and mechanical properties of GS, GC, and granite. GS that was produced using marble forming machines has made it in the form of spherical shape, while GC is in irregular shape because it was made using crusher machine.

Table 2: Physical and mechanical properties of GS, GC, and granite

Properties	Unit	GS	GC	Granite
3d shape	—	Sphere	Irregular (angular)	
Surface texture	—	Smooth	Smooth	Rough
Size	mm	19	4.75 to 19	4.75 to 19
Flakiness index	%	—	46.40	36.91
Elongation index	%	—	27.43	15.20
Specific gravity	—	2.43	2.70	2.64
Absorption	%	0.03	0.06	0.23
Unit weight	kg/m³	1409.25	1676.30	1619.65
Void	%	41.89	37.79	38.53
Aggregate crushing value (ACV)	%	14.83	26.08	22.75
Aggregate impact value (AIV)	%	—	21.90	17.03

Figure 2 shows the various shapes of GC. It can be seen that each GC particle is angular with minimum 8 corners. Besides that, the granite used as a control coarse aggregate in this study has similar shapes with the GC. Table 2 also indicated that the flakiness index of GC is higher than granite, which is 46% and 37%, respectively, meaning that GC has higher flat surfaces than granite. The lower flakiness index of granite also shows that the shape of granite is close to cube shape (ACI E1-99, 1999) (Miskovsky, Duarte, Kou, & Lindqvist, 2004). The low crushing level of GC that due to the intention of using it as a coarse aggregate (size > 4.75 mm) has resulted in the high flakiness index. In addition, the frequency of micro-cracks in GC is lower than granite. Past studies (Miskovsky et al., 2004) (Afeni & Ayiti, 2018) discovered that based on Spearman Correlation analysis, the flakiness index of aggregates is positively influenced by the frequency of micro-cracks. They claimed that the higher the flakiness index, the lower the frequency of micro-cracks. However, the use of GC as a coarse aggregate could adversely affect the concrete quality. This is because of high content of flaky aggregate that is GC are more likely to segregate the concrete mixes, decreased the compaction factors and concrete strength. The test results also show that the elongation index of GC is higher than granite, 27% and 15%, respectively, indicated that the length of GC particles is greater than 1.8 of its width. Castro and Brito (2013) prove that the glass sizes greater than 4 mm are considered as an elongated aggregate. Therefore, GS is expected to be more durable coarse aggregate than GC because of the flaky and elongated aggregate, i.e. GC has a higher tendency to cracks and crushed than aggregates in cubic and spherical form. Other than that, GS and GC have a smooth surface texture, whereas granite surface is rougher.

Table 2 shows that GS has the lowest value of specific gravity, unit weight, and percentage absorption. The melting and annealing processes in the production of GS have decreased its density. The GS specific gravity of 2.43 is approximately similar to other glass products that were produced using the same manner, e.g. glass marble (2.40), glass paperweight (2.44), etc (Afolabi, Johnson, & Abdulkareem, 2012) (Hawley, 2005) (Kepler International, 2010). Meanwhile, the specific gravity of GC at 2.70 are much lower compared to the CRT aggregates that was crushed to size less than 4.75 mm, as it generally between 2.99 to 3.10 (Lee, Yoo, Park, Cho, & Seo, 2015) (Zhao & Poon, 2017) (Zhao et al., 2013).

Figure 2: Shapes of GC

Besides that, the nature of glass as an impermeable material has explained the lowest percentage absorption by GS and GC, nearly 0%. GC has the highest unit weight at 1676.30 kg/m³, followed by granite and GS, at 1619.65 kg/m³ and 1409.25 kg/m³. These unit weights are within the ranges of coarse aggregate unit weight, 1280 kg/m³ to 1920 kg/m³ (ACI E1-99, 1999). In addition, Table 2 also shows that the percentage of voids for GS is the highest at 41.89%, followed by granite and GC, at 38.53% and 37.79%, respectively. Figure 3 shows the compactness of GS and GC particles in a 250 mL beaker. The size and shape of GS that is fixed to 19 mm and in sphere shape has resulted in poor compactness and less contact points (Moncrieff, 1953). Because of that, the use of GS as coarse aggregates led to the highest void percentage. Whereas, the irregular shape and sized of GC from 4.75 mm to 19 mm has made the GC particles fill in the space effectively compared to GS and granite, which explained the lowest percentage of voids of GC.

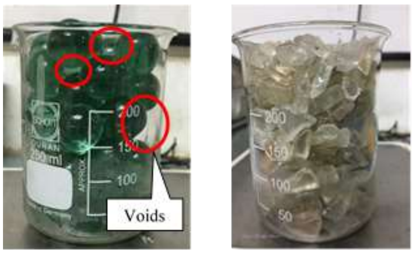

Figure 3: Compactness of GS and GC particles in beaker

Other than that, the mechanical properties of GS and GC as coarse aggregates have been identified through the ACV and AIV tests. Table 2 indicated that the ACV of GS was the lowest, followed by granite and GC, where at 14.83%, 22.75%, and 26.08%, respectively. It means that the total mass of GS after being crushed by the compression load and passing 2.36 mm size is the lowest, which it indicated GS is a stronger coarse aggregate compared to granite and GC. The annealing technique in producing GS has strengthened the glass product (Veer, Louter, & Bos, 2009) (Berenjian & Whittleston, 2017). On the other hand, granite has lower AIV value of 17.03%, while GC at 21.90%. The amount of GC that was crushed due to the impact loads and at size less than 2.36 mm is higher than the granite. Test results indicated that GC is a weaker coarse aggregate than granite, in terms of the resistance to compression and impact loads. This is because of the shape of GC that is flaky and elongated has decreased the resistance level of GC to compression and impact loads (Molugaram, Shanker, & Ramesh, 2014) (Benediktsson & Wigum, 2015). Besides that, based on the mechanical strength, it shows that GS and GC are able to resist the rollers and traffic loads as its crushing values are less than 35%, thus it is expected compatible to use as road construction materials (IS 383, 1997) (Puthussery, Kumar, & Garg, 2016).

Figure 4 shows the percentage finer versus sieve size graph of GC and granite as coarse aggregates. It can be seen that the size distribution of GC and granite are within the upper and lower limits of coarse aggregates as specified in ASTM C33. In addition, it is observed that the distribution of GC particles at size 4.75 mm and 14.00 mm are close to the lower limit of coarse aggregate grading. Meanwhile, at size 20 mm, the GC particle distribution is found approaching the upper limit grading. Nevertheless, the granite and GC are found to have a smooth grading curve, where it can significantly reduce the concrete voids (ACI E1-99, 1999) (Pawar, Sharma, & Titiksh, 2016). The reduction of concrete voids is expected could decrease significantly the cost of concrete production. It is because if the aggregates cannot fill the gaps between the particles adequately, thus that gaps will be filled with the cement paste. The higher cost of cement than the aggregates causes the increase of cement paste volume, which will increase significantly the cost of concrete production. Therefore, a good grading of coarse aggregates that comply to the ASTM C33 is important in minimizing the concrete production cost (Braga, Silvestre, & de Brito, 2017). The sieve analysis test was not conducted on GS particles because it's sized fixed to 19 mm.

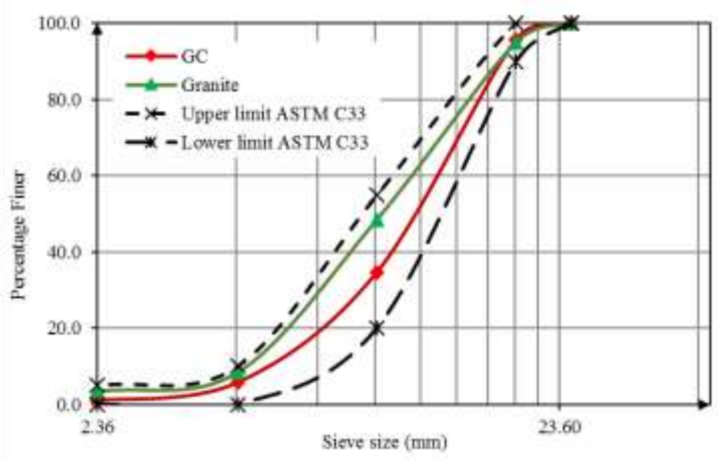

Figure 4: Grading curve of GC and NCA

Table 3 shows the chemical compositions of GS and GC. The table shows that the major chemical oxide present in GS and GC was silicon dioxide (SiO_2), PbO, sodium oxide (NaO), and potassium oxide (K_2O). Both GS and GC show the same types of chemical oxide composition and differ only in its percentage content. This is because, GS and GC are produced from the same material, that is funnel component of CRT glass. However, it should be noted that Pb is a hazardous chemical, and both GS and GC have shown higher concentration of PbO. Besides, PbO was the second highest percentage of chemical oxide present in the GS and GC. Thus, it should be noted that there might be a risk of Pb leaching from GS and GC, which will have harmful effects on the environment.

Table 3: Chemical compositions of GS and GC

Material	**Oxide compositions (wt. %)**									
	SiO_2	**PbO**	**K_2O**	**Na_2O**	**CaO**	**Al_2O_3**	**BaO**	**SrO**	**MgO**	**Fe_2O_3**
GS	43.64	22.47	7.44	6.05	3.21	3.41	2.44	2.14	1.34	0.22
GC	41.51	27.55	7.85	6.86	3.87	4.06	1.73	0.58	1.63	0.22

Compressive strength of concrete

The results of the 7-day, 28-day and 56-day compressive strength of control and CRT concrete mixes with different coarse aggregates contents are shown in Figure 5. It can be observed that for a given type of coarse aggregates, the compressive strength of CRT concrete mixes (GS1.0, GC1.0, GS0.6/GC0.4) were lower than that of control mix (NC). The 56-day compressive strength of NC mixes was at 39.6 MPa, while CRT concrete mixes were varied from 31.0 MPa to 36.2 MPa. The use of GS and/or GC as coarse aggregates has decreased the compressive strength, which due to the weak bonding between the glass aggregates and the cement paste. The smooth surface of the glass led to a weaker interface and reduced the strength of the concrete mixes. Besides that, it can be observed in Figure 5 that the use of GS and GC as coarse aggregates has delayed the strength development. The strength of CRT concrete specimens was drastically increased after 28-days curing, while only a slight increase of strength for NC specimens. Observing the influence of types of CRT glass product, the compressive strength became the lowest with the use of 100% GS as coarse aggregates, at 31 MPa. Whereas, test results indicated that the combination of GS and GC as coarse aggregates, at ratio 0.6GS:0.4GC, has resulted in the highest CRT concrete strength of 36.2 MPa. The combination of GS and GC at ratio 0.6:0.4 has made the coarse aggregates consisting of different shapes and sizes produce a more compacted concrete structure, which significantly lower the percentage reduction of compressive strength between the CRT concrete and the control specimens, as the strength loss only 8.9% (Li, Guo, Qiang, & Poon, 2017) (Wang, Guo, Dai, Si, & Ma, 2019).

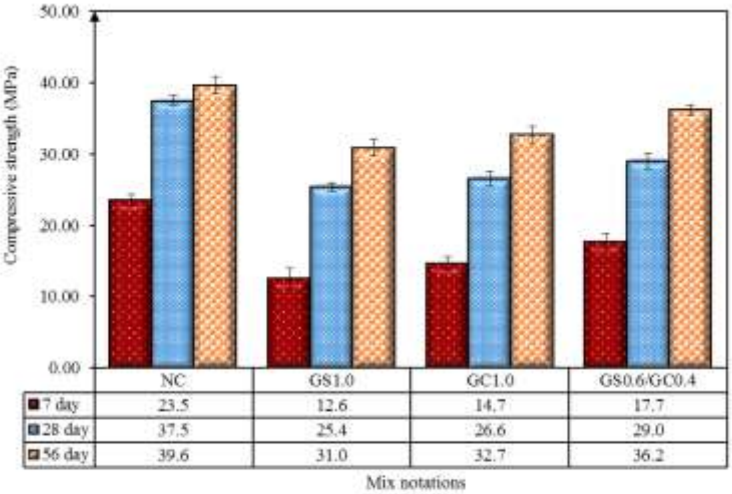

Figure 5: Compressive strength of control and CRT concrete

4. Conclusions

The paper compares the properties of two types of CRT glass products, namely spherical CRT glass (GS) and crushed CRT glass (GC), with the normal coarse aggregates (granite). Both of the glass was produced using a different techniques of CRT recycling. The melting and annealing techniques have decreased the GS density and increase its resistance to crushing. Meanwhile, the crushing techniques in the production of the GC have produced a flaky and elongated aggregates. In addition, this study uses GS and GC as an alternative replacement of natural coarse aggregate in production of concrete. The introduction of CRT concrete will not only convert a hazardous waste glass into a valuable product, but also could preserve the natural resources. However, replacing natural coarse aggregate with GS or/and GC has reduced the compressive strength of concrete. But this study also discovers that the use of different morphological features of CRT glass as aggregates are found to be beneficial to concrete strength. The combination of GS and GC at ratio 0.6:0.4 has made the coarse aggregates consisting of different shapes and sizes produce a more compacted concrete structure, which significantly lower the percentage reduction of compressive strength between the CRT concrete and the control specimens.

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