

BEHAVIOR OF CEMENT PASTE WITH DIFERENT PARTICLE SIZE OF RECYCLED GLASS ON ASR AND POZZOLANIC ACTIVITY

LUDMILA Soares Carneiro^{(1)*} and Takeshi IYODA⁽²⁾

^{(1)*} Master Student, Dep. Civil Engineering, Shibaura Institute of Technology, Tokyo/JP

⁽²⁾ Professor, Department Civil Engineering, Shibaura Institute of Technology, Tokyo/JP

*Corresponding author: me19103@shibaura-it.ac.jp

ABSTRACT

The harmful effects for the environment due to the high use of ordinary Portland cement (OPC) products claims for actions to decrease the use of this material by the society. The sustainable development goals (SDG) are 17 initiatives to promote global actions, focusing on the future of the planet and society. The use of recycled materials as supplementary cementitious materials can decrease the use of OPC and consequently the carbon dioxide emissions in the atmosphere, promoting the 13th SDG that regards on climate changes. In this research, glass with different particle sizes were investigated on pozzolanic activity in order to co-relate the results with the occurrence of alkali-silica reaction. Both reactions have been closely related and the ASR presents a huge issue when glass is used with cement. For this goal, glass with sand particle size was tested for ASR and also glass powder, so it could be verified the existence of deleterious expansion. Also, TG-DTA was performed to measure the amount of Calcium Hydroxide in the samples with different particles sizes. The results show that, as the particle size decreases, the ASR expansion also decrease and more calcium hydroxide is consumed. Consequently, it can be concluded that pozzolanic reaction is happening more strongly and ASR can be suppressed for recycled glass of small particle sizes.

Keywords: *Cement paste, recycled glass, sustainable goals, pozzolanic activity, Alkali-silica reaction, durability.*

1. INTRODUCTION

1.1 Environmental Issue

The use of recycled material in the construction industry is an alternative to decrease the exploitation of natural resources. The concrete, is one of the most used material in the world (Metha & Monteiro, 2006) so consequently, the use of its component materials have huge influence in the consumption of natural resources globally.

The high production volume of concrete presents a possibility of huge destination amount of recycled and industrial waste material. This alternative destination for unwanted materials can help to decrease landfills disposal sites and the intense exploration of natural



Fig. 1. Sustainable development goals (UN, 2020)

aggregates. Furthermore, the environmental issues related to the use of concrete are not only associated to the natural materials exploitation but also connected to the greenhouse gases emissions. The entire life cycle of concrete involves CO₂ emission and around of 9 to 10% of the CO₂ emitted in the world is produced by concrete life cycle (Taylor et. al, 2006). The major cause of high carbon dioxide emission is the production of cement, the main component of concrete.

The harmful behavior of cement claims for actions that helps to decrease the environment impacts related to this material. Many international cooperation initiatives are aiming to decrease the environmental impacts of human consumption and one of this actions from United Nations is the Sustainable Development Goals (SDG's) (UN, 2015). This goals are 17 integrated necessary actions in order to ensure balance of social, economic and environmental sustainability in 20 years from now (by 2030). According to UN, this actions could promote sustainable development and together they can help to decrease the harmful effect of human consumption in the earth.

The Goal 13, take urgent action to combat climate change and its impacts, is also connect with the Paris Agreement that states that the greenhouse gas emissions must be falling by 7.6% each year starting in 2020. This goal is related to the global warming and the changes in temperature of the earth that causes wildfires, hurricanes, droughts, floods and other climate disaster across the continents, affecting millions of people's lives. The construction industry, mostly because of the cement production, contributes very much for the increase in carbon dioxide emission, one of the main causes of greenhouse effect. The substitution of cement with other materials could decrease the CO₂ emissions and help to meet the Paris Agreement target, promoting balance to a

sustainable development for among all nations.

Aiming to decrease the harmful consequences of high use of cement and promote the SDG's, the use of waste or/and recycled materials such as glass is promissory option. Glass materials are used in large scale by the modern society and it is considered a non-biodegradable material because of the deterioration time in natural environment. It could take more than 4 thousands years to deteriorate in natural conditions. So, also the disposal of this material on landfills could not present the most appropriate option.

In order to be applied by the construction industry, the behavior of the structures with recycled glass has to meet some requirements of performance. Among durability considerations, pozzolanic reaction and alkali-silica reaction are the most studied topics for glass when used in concrete or mortar.

1.2 Pozzolanic Reaction and Alkali-Silica Reaction

The glass most notorious characteristic is the huge amount of silicon, so the silica dioxide can be about 70% of its composition. This component makes glass a promising pozzolanic material however, it can also react strongly with alkalis from cement resulting in a hydrophilic gel that expands causing cracks, and consequent damages called alkali-silica reaction.

Taylor (1990) also considered ASR as an pozzolanic reaction on chemical terms, with the different effects in concrete related to the particle size and siliceous material (in this study, recycled glass). In his book, he explains that the hydroxyl ions attack the Si-O-Si bridges replacing it for SiO^- groups. It fragments the silica framework into separate silicate anions, they are balanced by available cations, K^+ and Na^+ . As the framework is damaged and deformable, it can imbibe water and expand. This situation differs from C-S-H that have a rigid structure based on Ca-O layers. It is unstable in the presence of Ca^{2+} that reacts to form C-S-H. This regenerate OH^- in the solution. Alkali silicate gel can accommodate Ca^{2+} but a point is reach were the C-S-H begin to form as a different constituent. So, in pozzolanic reaction the gel is formed in an environment rich on Ca^{2+} , so it is quickly converted in C-S-H. On the other hand, ASR is formed in a poor environment of Ca^2 because the cement paste cannot supply it fast enough and a huge amount of gel overflow is formed.

So, the particle size is a very interesting topic related to this issue because is often associated to the occurrence of ASR or Pozzolanic activity of the material, so it provides a complex dual behavior. Following that, it could be observed over the bibliographic reviews (Omran, 2018/Du and Tan, 2014) that, the ASR become weaker as de particle size decrease for most of the color, content, system or uses of recycled glass in cement structures. However, there is no agreement about which particle size provide the maximum expansion or can mitigate the effects caused by ASR (Rashad 2014 Jani et. al 2014).

When it is used in an appropriated dosage and size,

the recycled glass can mitigate the reaction in concretes susceptible to ASR. The complexity of the recycled material also gives more variants to be considered in the process of the reaction to happen. The color, type, proportion and use as binder or aggregate can also play a role in the reaction. The glass chemical composition is directly correlated with the color and type (such as soda-lime glass), so it also is an important consideration factor to describe the reaction possibilities.

The glass used in this study is a rich calcium material, with low amount of sodium and mixed colors (however the predominant color is green). With that said, this research has as an overall objective to promote and contribute for the use of recycled materials in the construction industry. If the behavior of the recycled glass can be evaluated when used in cement structures, it can also provide a destination to this recycled material, presenting a prospective substitute binder for ordinary Portland cement. This measurement can help to decrease the amount of carbon dioxide emission in the atmosphere from clinker production by the cement industry.

As specific objective, this paper aims to verify the reactivity of the recycled glass, and how it can decrease the expansion of ASR due to the variation of particle size and pozzolanic activity. For this goal, the research prepared samples for ASR expansion of the critical sizes (sand and powder) and TG-DTA experiment for specific particle sizes (0.075mm, 0.150mm, 0.300mm, 0.600mm, 1.18mm, 2.36mm). From this, the profile of the reaction can be traced from the big particle sizes (sand) to the smaller (powder) and the expansion of ASR could be implied by the residual calcium hydroxide amount in the samples measured by TG-DTA. So, the information of the recycled glass as pozzolanic material in order to decrease ASR can be found. This can increase our knowledge about the material and promote its use in construction as supplementary cementitious material (SCM).

2. EXPERIMENTAL OUTLINE

2.1 Materials and Mix Proportions

The recycled crystal sand glass is from Osaka region at Japan and it is considered as mixed colors with predominant green color (Figure 1).

The recycled glass chemical composition can be seen at Table 1.

In order to verify the reactivity of the recycled glass, JIS A 1146 ASR mix proportion was performed and 0, 25, 50, 75 and 100% of natural sand was substituted by recycled glass crystal sand.

For ASR the samples with glass powder, the samples were casted with the specified proportions from standard ASTM C1567. The mix used up to 30% of cement substitution. It was also casted a reference sample with 100% of OPC. As aggregate it was used glass crystal sand because of the high reactivity.

The TG-DTA samples was prepared by separating the material particle size as distributed in the standard ASTM C 1567 and JIS A1146. For these mixes design it



Fig. 2. Recycle glass: color and size distribution

Table 1. Recycled glass chemical composition.

Recycled Glass		
	Content	wt%
1	SiO ₂	69.2740
2	CaO	20.9849
3	Na ₂ O	4.6117
4	K ₂ O	1.8740
5	Al ₂ O ₃	1.8376
6	Fe ₂ O ₃	0.4682
7	MgO	0.2504
8	BaO	0.1364
9	Cr ₂ O ₃	0.1340
10	TiO ₂	0.1079

Table 2. Nomenclature.

	Particle size in mm	Nomenclature
1	OPC	0.020
2	0.075 (Powder)	0.075
3	0 – 0.150 (Pan)	0.150
4	0.150 – 0.300	0.300
5	0.300 – 0.600	0.600
6	0.600 – 1.180	1.180
7	1.180 – 2.360	2.360

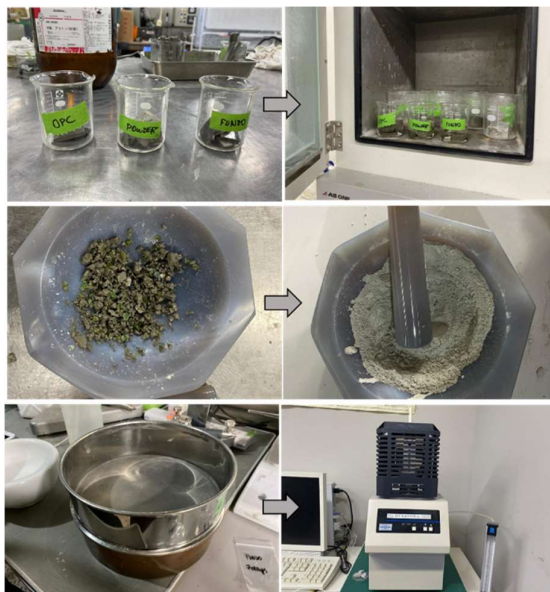


Fig. 3. Cement paste experimental method

was casted 7 proportion, 6 with 75% of ordinary Portland cement (OPC) and 25% of recycled glass with different particle sizes (in volume) and the reference mix (100% OPC). The water cement ratio as fixed as 0.50. The grading sizes can be seen at Table 2.

The glass crystal sand and Powder (75 μ m) were provide from Fujino Kogyo Ltd. and Ordinary Portland cement from Taiheiyo Cement Corporation (NaOeq = 0.62%). The alkali used for the mix design and soaking solution and JIS standard adjustments was Na produced by Kanto Chemical Co. as extra pure (95.0%) solid NaOH.

2.2 Testing Methods

In order to evaluate the reactivity of the recycled glass for ASR, mortar samples were prepared according to the Japanese Industrial Standard A 1146 (JIS). The JIS that established one part of cement for 2.25 parts of graded aggregate and water-binder ratio of 0.50. The amount of cement is fixed as 600 grams. The standard also determines that the amount of alkalis in each mix has to be fixed as 1.2% of Na₂Oeq. As the Taiheiyo cement content was Na₂Oeq = 0.62%, all the alkalis amounts were adjusted according to the standard request. After cast process, the specimens were submitted to curing in a controlled chamber under the temperature of 40°C and relative humidity (RH) of 95%. The samples expansion was read at 1, 2, 3, 4, 6, 8, 10, 13, 15, 18 and 26 weeks of age.

The recycled glass powder (0.075mm) influence in the expansion of ASR was measured by performing the accelerated mortar bar test (AMBT) from ASTM. For this, the glass powder was added in the mortar mix, substituting OPC in different amounts. The source of material mix proportions was the ASTM C1567 that established one part of binder for 2.25 parts of graded aggregate and water-binder ratio of 0.47, with the total amount of binder of 440g. The readings were made on 1, 2, 5, 7, 10 and 16 days of curing inside a NaOH solution at 80 degrees Celsius.

The residual calcium hydroxide amount was measured by casting cement paste of a mix of OPC and glass with different particle sizes. The curing was performed inside a chamber of controlled temperature and humidity (20°C and 60% RH). The samples were test on 7 and 21 days of age. At the specific day, the sample was immersed in acetone and put on vacuum condition for 3 hours. The samples were crushed and sieved in 0.150mm. So, the material was tested using a TG-DTA – MTC 21000SA Bruker on Nitrogen atmosphere. The temperature was increased until 1000 degrees Celsius. The process can be seen at figure 3. As the samples were sieved in the 0.150 mm, the material of 0.300, 0.600, 1.18 and 2.36 mm was retained in the sieve. The 0.075mm and 0.150 mm was incorporated in the material analyzed in the TG-DTA.

3. RESULTS AND DISCUSSION

3.1 Alkali-Silica Reaction

3.1.1 Glass crystal sand ASR reactivity

As introduced before, the glass amount of silica dioxide makes it a promisor material to be used as pozzolans, however, these same characteristics can cause a pathologic reaction of ASR and cause damages to the structure. This events have been related to the particle size of the siliceous material. At figure 4 it can be observed that when glass is used as sand, with large particle size (around 0.150mm to 4.75mm), the results present a huge expansion. It indicates that the ASR expansion is happening strongly and is caused a deleterious expansive behavior in the samples.

In the figure 4 can also be seen that the use of 100% of glass as natural aggregate presents the highest expansion, decreasing as the amount of glass also decreases.

3.1.2 Glass powder ASR reactivity

The experiment of ASR expansion using glass powder (0.075mm) can be seen at figure 5. In this result it can be observed that, when a small particle size is used, the expansion decrease as the percentage of substitution increase.

This behavior of the glass powder can confirm the possible pozzolanic activity of the glass, when used in small particle sizes. The powder used as 30% of substitution of OPC presented a reduction in expansion when compared to the 100% OPC, both used with high reactive aggregates. This reduction of expansion is one of the characteristic of ASR reduction by the use of pozzolanic materials.

3.2 TG-DTA

The Figure 6 shows the results obtained by TG-DTA on Calcium Hydroxide (CH) amount in the cement paste of recycled glass and OPC according to the particle size. As an overall tendency, as the particle size decrease, the amount of CH decrease. This behavior is due to the propensity of the pozzolanic material to reacts and consumes CH, forming cementitious products. The samples with 0.075 and 0.150 mm showed a small amount of CH at 7 and 21 days when compared with the ordinary Portland cement.

The samples of 0.150mm presented a slightly smaller results when compared with 0.075 because this material contains also smaller particles. The 0.150 particle size includes 0 to 0.150mm, the named "pan" because is the material that pass in the last sieve. The 0.075 result is from the powder material. This material has in an average particle size of 0.075mm, with more consistent particle sizes. This condition favors that the consumption of CH by the pozzolanic reaction will happen strongly with the samples 0.150.

The particle size from 0.300 to 2.360 showed higher amount of CH in the samples. It can be concluded that the reaction is happens less intensely because of the particle size that makes the glass reactive only in the surface of the material. The small particle sizes, silica can be consumed fully and quickly because of the dissolution process in the high alkaline environment, however the big particles take more time to dissolute and

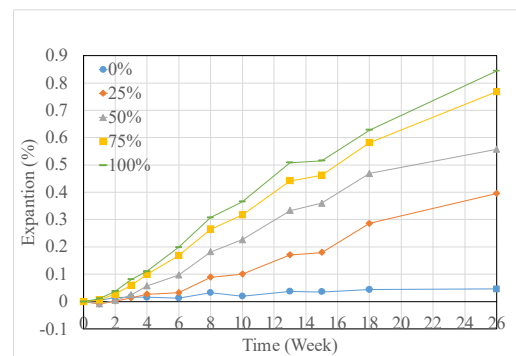


Fig. 4. ASR Expansion glass sand

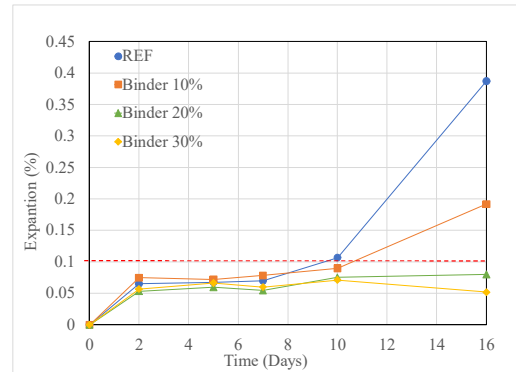


Fig. 5. ASR expansion for glass powder

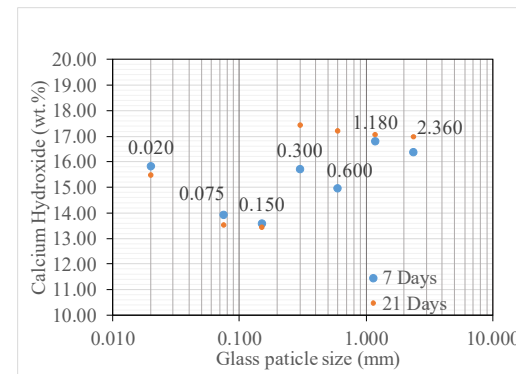


Fig. 6. Calcium hydroxide amount per particle size

also create a reaction rim around the particle that, over the time can decrease the reaction speed.

For 0.300 and 0.600, the consumption of CH was similar to the OPC cement paste for the initial age of 7 days. The next reading shows that the amount of calcium hydroxide increase to a similar value of 1.18 and 2.36. It can be implied that the pozzolanic reaction was happening in the 7 days however it decreased over the time, leaving more free CH from cement hydration in the samples. It can be explained by the formation of the reaction rim around the glass particles that makes the pozzolanic reaction decrease over the time because the particle cannot be reached by the alkalis from cement and so, participate in the pozzolanic reaction.

The OPC (0.020), 1.18 and 2.36 results are also very similar over the time. It can be attributed to the fact that the big particles do not react very much with the calcium hydroxide what is characteristic of the pozzolanic reaction. The amount of CH is high and do not vary too

much from 7 to 21 days of age. The same pattern can be seen at OPC cement paste and on the samples where the glass had strong pozzolanic behavior.

From this results, it can be implied that as the pozzolanic reaction increase, the effects of ASR decrease for the recycled glass material. The particle sizes of 0.075 and 0.150 are pozzolanic and can present a good prospective material to be used in order to obtain a smaller ASR expansion results.

4. CONCLUSIONS

The results of this study follow below.

- 1) Glass sand is high reactive for ASR;
- 2) When used in small particles, such as powder, the recycled glass can reduce ASR expansion, probably due to the pozzolanic activity of the material;
- 3) The consumption of CH increases as the particle sizes of recycled glass decreases;
- 4) The highest consumption of CH was found for recycled glass powder (0.075mm) and 0.150 (pan);
- 5) The 0.300 and 0.600 shows that there is more CH from the hydration of cement free at late ages (21 days), it can be concluded that the pozzolanic reactivity decreased because of the formation of a reaction rim around the particle that made difficult from the Si of the glass to react forming cementitious products (pozzolanic reaction);
- 6) The 1.18 and 2.36 did not have a strong pozzolanic reaction;
- 7) Recycled glass with particle size smaller than 0.150 can behave as a pozzolanic material and has potential to decrease the ASR effects on concrete or mortar.

As the material is recycled, the investigation of the properties for potential use are very relevant. This specify recycled glass presented good results when used in smaller sizes such as 0.150mm. As this material presents a high alkali-silica reactivity due to the amorphous behavior of glass, it can be concluded that the ASR harmful expansion can decrease for the particle size smaller than 0.150 because of the pozzolanic behavior. The directly investigation of ASR experiment is necessary in all particle sizes for confirmation of this behavior, so it will be suggested as future experimental plan.

ACKNOWLEDGMENT

The author acknowledges the support of co-author Professor Takeshi Iyoda, Fujino Kogyo CO. Ltda., Shibaura Institute of Technology and Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

REFERENCES

Du, Hongjian, and Kiang Hwee Tan. "Effect of particle size on alkali-silica reaction in recycled glass mortars." *Construction and Building Materials* 66 (2014): 275-285.

Jani, Yahya, and William Hoagland. "Waste glass in the production of cement and concrete—A review." *Journal of environmental chemical engineering* 2.3 (2014): 1767-1775.

Mehta, P. Kumar, and Paulo JM Monteiro. "Concrete: microstructure, properties, and materials." (2006).

Omran, Ahmed, et al. "Performance of ground-glass pozzolan as a cementitious material—a review." *Advances in Civil Engineering Materials* 7.1 (2018): 237-270.

Rashad, Alaa M. "Recycled waste glass as a fine aggregate replacement in cementitious materials based on Portland cement." *Construction and Building Materials* 72 (2014): 340-357.

Taylor, Harry FW. *Cement chemistry*. London: Thomas Telford, 1990.

Taylor, Michael, Cecilia Tam, and Dolf Gielen. "Energy efficiency and CO2 emissions from the global cement industry." *Korea* 50.2.2 (2006): 61-7.

U.N. - United Nations. (2015). Sustainable development goals. Retrieved from <https://www.un.org/sustainabledevelopment/development-agenda/> - accessed on December, 2020.

PHOTOS AND INFORMATION



Ludmila Soares Carneiro received the B.E. in Civil Engineering from University of Mato Grosso do Sul/Brazil (2016). Master student - Shibaura Institute of Technology (Tokyo/Japan).



Takeshi Iyoda received the B.E. (1997) and M.E. (1999) in Civil Engineering from Shibaura Institute of Technology, PhD. (2003) degree from University of Tokyo. Professor of the Department of Civil Engineering - Shibaura Institute of Technology (Tokyo/Japan).