

Performance of Green Concrete Comprising Waste Ceramic as a Fine Aggregate Replacement: Effect of Sulfuric Acid on Degradation Process

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ABSTRACT

This study investigated the durability performance of concrete exposed to Sulfuric acid, as well as assists to reduce the landfill and environmental pollution problems, by incorporating 30% ceramic waste powder as a replacement of fine aggregate. Concrete specimens of 100 × 100 × 100 mm in dimensions were cast with a 0.5 water/cement ratio, and 31.4 MPa compressive strength at age of 28-day. Durability of concrete cubes was periodically examined by means of measuring the change in mass, residual compressive strength, and visual inspection for a period of 14 weeks of exposure to 5% Sulfuric acid attack (H₂SO₄). The results showed that incorporated 30% of waste ceramic powder contributed to enhancing the resistance of concrete by improving the pore structure of concrete under sulphate attacks due to formation of extra C-S-H gel through the chemical reaction between calcium hydroxide and silica. It can be concluded that the ceramic waste powder restricted the ability of sulphate ions to penetrate the concrete, which led to better performance in acidic solution.

Key Words: Waste ceramic powder, Sulfuric acid attack, Concrete, Fine aggregate, Compressive strength.

1. INTRODUCTION

Urbanisation and globalisation are responsible for running down the world's natural resources, leading to a rise in CO₂ emissions and environmental damage. High quality materials, particularly sand, are increasingly being sought for sustainable infrastructure development projects. Concrete is an important element of the drive towards sustainability, and features at the production and demolition points [1]. Some 60-75 per cent of the total volume of concrete is made up of aggregate, and of this 35 per cent is fine aggregate. Quarry mining of sand is environmentally inequitable and makes sand into an over-valued material [2, 3]. As a result, the construction industry is looking for alternatives which are both sustainable and reasonably priced. In addition, construction solid wastes, including clay bricks, ceramic tiles, old concrete, rubbers, and waste glass, have been assessed for their potential to be used as an additional cementitious material, or as aggregates [4-7]. Ceramic products are made of clay, feldspar, and quartz, raw materials which are subsequently processed through mixing, drying, and burning [8]. The manufacture of ceramic products and the demolition of buildings leads to a substantial quantity of ceramic waste [9], and estimates have suggested that 10-30 per cent of ceramic production ends up as waste [10]. Although ceramics are both functional and highly used in our everyday lives, their waste results in significant environmental stresses, and invariably ends up being taken to landfill. Ceramic waste is extremely resistant to chemicals and other forms of degradation. Conversely, no technology has been proposed to be recycled [11]. The ceramic industry clearly needs to address this issue and find a new way of disposing of its waste. Lim et al. [12] suggest that ceramic waste could be a sustainable material in creating mortar and concrete for construction projects, since this will reduce costs and ecological damage as well as minimise the need for raw materials. The high compressive strength of ceramic waste was tested by Rashad and Esaa [13] who exposed this waste to temperatures as high as 1000 degrees centigrade, by partly replacing slag with 5-50 per cent by weight. Binici, [14] substituted waste fine ceramic particles for 100 per cent of fine aggregates, in order to determine the long-term compressive strength of concrete, and found a drop of 10 per cent, after the substitution. Senthamarai et al. [15] experimented with ceramic waste powder as a replacement for cement and discovered that it has the potential to be used as a supplementary cementing material, since it has sufficient strength and durability to be used in concrete. Torkittikul, and Chaipanich [16] determined that the concrete which incorporated up to 50 per cent of fine ceramic aggregates as natural aggregates met all the benchmarks for strength and durability performance. One issue which needs

examination is sulphate attack, namely the reaction which occurs between sulphate ions and the hydration products of cement and causes damage, posing an actual and major threat to the durability of concrete in an aggressive environment. Sulphate ions are found in ground water, sea water, soils, and waste waters [17], and when they react with hydration products of cement, they form gypsum, ettringite and/or thaumasite. The result is cracking, spalling, softening, expansion, loss of strength and other types of damage [18, 19]. A large number of methods for reducing the effects of an acid attack on concrete have been proposed to date, including fly ash, silica fume, inhibitor, recycled concrete, limestone concrete, blast-furnace slag and glass powder [20-24]. Gruyaert et al, [23] evaluated how replacing OPC with blast-furnace slag led to a sizable fall in acid deterioration in BFS concrete, a result which was for the most part ascribed to the binder's different chemical composition. Said et al, [25] stated that incorporating glass powder as a replacement for cement in mortar made it more able to withstand Sulfuric acid attack. Makhloufi et al, [26] found that using admixtures of minerals such as silica fume and fly ash in concrete has made it more resistant to Sulfuric acid attack. This is because of the reduction in the presence of calcium hydroxide, which becomes vulnerable to acid attack over time. In contrast, the effect green concrete, which comprises of waste ceramic tile against Sulfuric acid attack is very limited. This being the case, the time has come to assess the effect of using 30 per cent of ceramic powder as a replacement for fine aggregate, to stave off Sulfuric acid attack, as well as reduce the demand for natural resources, send less waste to landfill and preserve the environment from pollution.

2. METHODS

2.1 Materials

Ordinary Portland cement (OPC – type II) was utilised in this investigation. Waste ceramic particles were collected from the local tile industries, which are broken pieces of tiles. In order to remove unwanted materials such as eliminate dregs and sediment, the broken ceramic pieces were washed under running tap water and then left to dry under sunlight for 2 hours. After drying, the large particles of ceramics waste were crushed into small pieces with dimensions not exceed 5×7 cm by using pestle. Thereafter, about 10 kg of ceramics were ground into powder utilising Los Angeles abrasion machine which is consisted of 12 steel balls with 40 mm-diameter for 6 hours. Next, the resulting ceramic powder was sieved with $150 \mu\text{m}$ for farther use it as a fine aggregate replacement. The preparation process of waste ceramic powder was depicted in Figure 1. The specific gravity was found to be 2.63 and 2.60 for sand and ceramic powder respectively. Finally, the powder was subjected to X-ray fluorescence (XRF) to characterise the chemical compositions. The results obtained were listed in Table 1.



Figure. 1: Preparation of waste ceramic powder in the lab.

Table 1: Chemical compositions of waste ceramic powder

Chemical composition (% by mass)	Waste ceramic powder
SiO ₂	71.50
Al ₂ O ₃	13.80
Fe ₂ O ₃	0.59
CaO	0.02
K ₂ O	0.03
TiO ₂	0.08
Na ₂ O	12.91
MgO	0.93
SO ₃	0.01
LOI	0.14

2.2 Mix Proportioning and Specimen Preparation

A total of 30 concrete specimens have dimensions of 100 × 100 × 100 mm were cast using a concrete mix design of 1:1.79:2.76 and 0.5 of w/c ratio to reach the required strength and appropriate workability. In this investigation, two sets of concrete mixes were prepared based on the percentages of fine aggregates. The concrete specimens with 100% fine aggregate are called control while the specimens with 30% of ceramic waste powder are called ceramic concrete. The 30% of ceramic powder was partially replaced based on the mass of fine aggregate. Moreover, 30% of waste ceramic powder was selected as the optimum value from a variety of percentages that have been tested, which showed a maximum compressive strength at the age of 28 days. Following 24 hours of casting, the concrete cubes were demoulded and cured in fresh water for a period of 28 days. Next, the specimens were immersed in 5% H₂SO₄ solution with concerted of 98% for 14 weeks. The resistance of concrete specimens to Sulfuric acid (H₂SO₄) attack was assessed based on mass change, compressive strength loss, and visual inspection in accordance with ASTM C267 [27].

The pH of acidic environment was calibrated (pH ≈ 1) and kept by weekly refreshing the solution for a period of 14 weeks at room temperature ± 2 °C and relative humidity about 68%.

Triplicate of specimens for each set of concretes were selected and mean of strength loss were recorded after 7-day, 28-day, and 90-day of exposure to acidic solution, while the visual inspection of specimens was left for 14-week. In addition, the mass change in specimens were determined every 7 days, the specimens were taken out from the acidic solution, cleaned with fresh water, and left to dry for 1 hour before being weighed. The following equations were utilised to calculate the mass change and compressive strength loss for both control and ceramic concretes [28];

$$\text{Mass change} = \left(\frac{w_i - w_t}{w_i} \right) \times 100$$

Where;

w_i is the average value of initial mass in grams, for three cubes before immersing in Sulfuric acid. While w_t is weekly changed in mass for three cubes after exposed to 5% Sulfuric acid in grams.

$$\text{Compressive strength loss} = \left(\frac{c_t - c_s}{c_t} \right) \times 100$$

Where;

C_t is the compressive strength of three cubes (average) immersed in normal water (MPa). While C_s is the compressive strength of three cubes (average) immersed in 5% H₂SO₄ (MPa).

3. RESULTS AND DISCUSSION

3.1 Mass change

Figure 2 illustrates the mass change of control and ceramic concrete specimens, which have been exposed to a 5 per cent Sulfuric acid solution over a period of 14 weeks. Concrete which has been exposed to sulphate solutions undergoes a step by step degeneration, beginning with expansion, then cracking and a reduction in strength. The expansion is linked to the amount of sulphate which has penetrated the interior of the specimen whereupon it expands mass. In this experiment, both control concrete and ceramic concrete specimens increased in mass over the first six weeks of being exposed to sulphate solution - as a result of the formation of ettringite. Sulphate ions react with portlandite, and this creates gypsum, weakening the portlandite, destabilising the matrix bond and lowering the sulphate resistance of the concrete surface. The penetration of the sulphate inside the concrete increases, as it enters through the weak points, and impacts negatively on the internal bonding inside the concrete matrix.

Qi et al, [29] note that while the sulphate attack can lead to a minor expansion, it more commonly shows itself through a loss of stiffness and strength and adhesion. These findings explain why, from week seven onwards, it was the control specimen which showed greater mass loss, as its edges began to spall and, indirectly, resulted in mass reduction - caused by the formation of gypsum and decalcification of C-S-H gel which weakened the concrete. As exposure continued, the concrete softened, lost cementitious structure and began to degrade. In contrast, the ceramic concrete performed well, and this is partly due to the higher water absorption of the ceramic powder, which lowered the water/binder ratio. Since the positive effect of water absorption of ceramic powder raising specimen density is larger than the negative effects of the defects caused by control concrete, it becomes clear that waste ceramic concrete performs better than concrete with 100 per cent natural aggregate.

Figure 2 shows that the maximum mass loss of specimens made with 100 per cent natural aggregate (the control specimen) is 9.6 per cent - after 14 weeks of exposure - whereas the specimen made with 30 per cent ceramic had a maximum mass loss of 3.7 per cent. Concrete which is attacked by sulphates benefits, and is more resistant, if it has ceramic content, due to the pore structure of ceramic concrete and the greater density of its microstructure. In addition, the pozzolanic reaction of ceramic concrete could lead to the consumption of calcium hydroxide, and reduce the amount of formed gypsum in the mixtures with ceramic concrete, when compared to the control concrete.

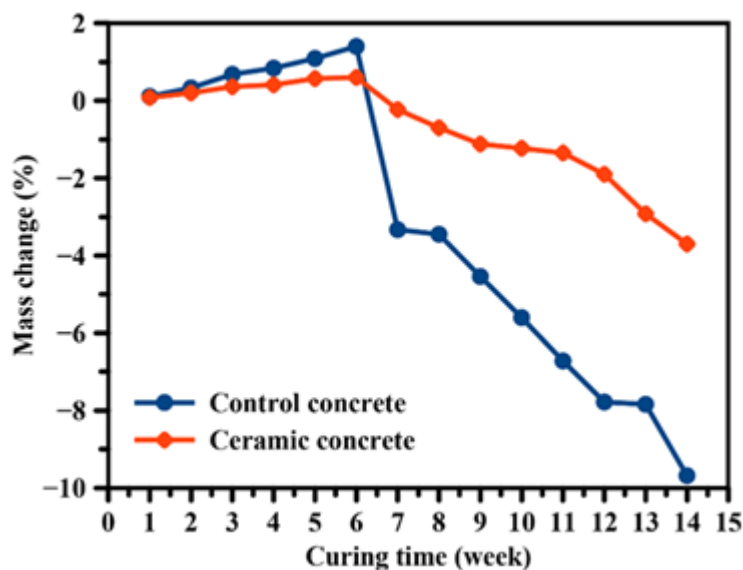


Figure 2: Mass change of concrete specimens after 14 weeks of exposure to 5% Sulfuric acid.

3.2 Compressive strength loss

Figure 3 illustrates the results of compressive strength of the control and ceramic concretes at 7, 28, and 90 days of exposure to 5% H₂SO₄. It shows that, in the first seven day period, strength values of 23.07 MPa and 27.3 MPa were found for control and the ceramic concretes, respectively. As the curing period continued, the control specimens lost more compressive strength than the ceramic concrete. Thus, for example, at the end period of exposure (90 days), the control concrete loss in compressive strength was measured at 34.3 MPa, some 18.53 per cent higher than that recorded for ceramic concrete, which was 46.3 MPa with loss in compressive strength of 3.87 per cent. This difference in the loss of the strengths can be ascribed to pozzolanic reaction which

took place among the reactive silicon oxide (SiO_2) where there was a high percentage of ceramic powder, and the OPC-Portland cement hydration products, for example, calcium hydroxide (Ca(OH)_2). This finding demonstrates that the chemical reactions between SiO_2 and Ca(OH)_2 led to the formation of extra C-S-H gels in the ceramic concrete matrix, thereby reducing both porosity and the loss in compressive strength over longer curing periods. In summary, using waste ceramic particles as partial fine aggregates substantially improves the compressive strength of concrete exposed to a harsh environment.

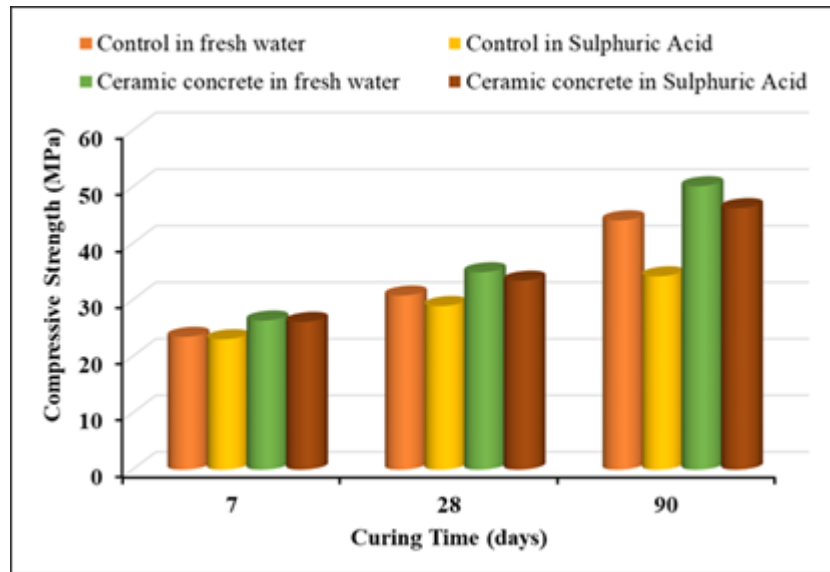


Figure 2: Compressive strength loss of concrete specimens after exposure to 5% Sulphuric acid at different ages.

3.3 Visual inspection

Figure 3 illustrates the visual inspection of control and of ceramic concrete specimens after they had been immersed in 5% H_2SO_4 solution for 14 weeks, and reveals changes in dimension and condition in the specimens. The control specimen (Fig. 3(a)) has major surface deterioration with deterioration depth of 14 mm-which is less in the case for the ceramic concrete specimen (Fig. 3(b)) with deterioration depth of 5 mm. In addition, the control specimen demonstrates that gypsum has formed as the result of a sulphate ions attack, where the sulphate solution and Ca(OH)_2 in the OPC had a chemical reaction. Hence, gypsum could react with other cement particles - for example, calcium aluminate - which resulted in the formation of ettringite, which increased the volume of the specimen. This, in turn, led to spalling at the corner of concrete cubes and the creation of cracks. In contrast, the ceramic concrete mix had less Ca(OH)_2 , the gypsum formation was lower. Additionally, in the ceramic mix, the consumption of calcium hydroxide during the hydration process led to the formation of additional C-S-H gels which were stronger in repelling an acid attack. Thus, less degradation was observed in the ceramic concrete as well as no visible cracks and spalling upon the surface.

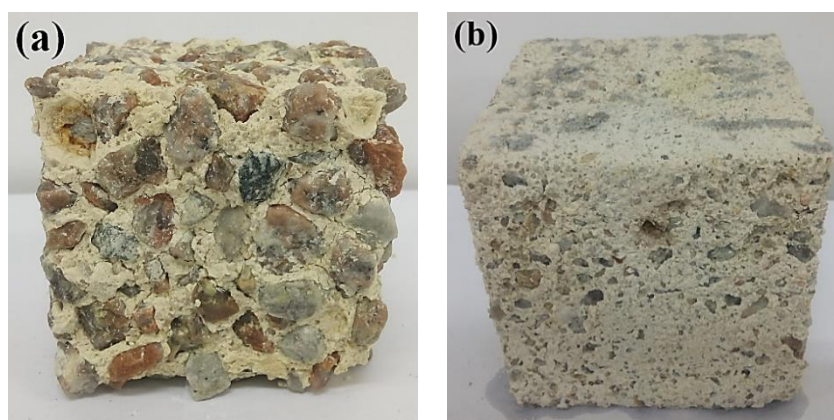


Fig 3: Visual inspection for concrete specimens exposed to the Sulphuric acid solution for 14 weeks, (a) control concrete, and (b) with 30% of waste ceramic powder.

4. CONCLUSIONS

The experimental analysis on the durability performance of concrete specimens exposed to 5% Sulfuric acid solution was presented in this investigation, where the 30% of waste ceramic powder was partially utilised as a fine aggregate replacement. The following conclusions can be drawn:

- i. Incorporation of waste ceramic powder significantly enhanced the resistance of concrete against sulphate ions attack, by improvement of pore structure and creation a denser microstructure of concrete specimens.
- ii. The maximum mass loss of control specimens was 9.6% in comparison with 3.7% mass loss for ceramic concrete after 14 weeks of exposure.
- iii. Compressive strength for control concrete was measured after 90 days of exposure to be 34.3 MPa, with strength loss of about 21%. In contrast, the ceramic concrete specimens recorded 46.3 MPa with only loss in compressive strength of 3.87% at the same period of exposure to Sulfuric acid.
- iv. Visual inspection shows less degradation of ceramic concrete, the specimens present sharp edges, no cracks, and no spalling as well. While control specimens display high degradation with 14 mm loss at the corner. Moreover, cracks, spalling, and gypsum were also observed.
- v. Replacing of waste ceramic powder with a fine aggregate exhibit technical, environmental as well as economic benefits that are significant in the existing context of sustainability in the civil engineering field especially as repairing materials.

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REFERENCES

- [1] S.K. Kirthika, S.K. Singh, A. Chourasia, Alternative fine aggregates in production of sustainable concrete- A review, *Journal of Cleaner Production* (2020) 122089.
- [2] M. Gonzalez, I. Navarrete, P. Arroyo, G. Azúa, J. Mena, M. Contreras, Sustainable decision-making through stochastic simulation: Transporting vs. recycling aggregates for Portland cement concrete in underground mining projects, *Journal of Cleaner Production* 159 (2017) 1-10.
- [3] S. Mundra, P.R. Sindhi, V. Chandwani, R. Nagar, V. Agrawal, Crushed rock sand – An economical and ecological alternative to natural sand to optimize concrete mix, *Perspectives in Science* 8 (2016) 345-347.
- [4] G.F. Huseien, A.R.M. Sam, J. Mirza, M.M. Tahir, M.A. Asaad, M. Ismail, K.W. Shah, Waste ceramic powder incorporated alkali acti-vated mortars exposed to elevated Temperatures: Performance evaluation, *Construction and Building Materials* 187 (2018) 307-317.
- [5] M. Saberian, J. Li, B. Nguyen, G. Wang, Permanent deformation behaviour of pavement base and subbase containing recycle concrete aggregate, coarse and fine crumb rubber, *Construction and Building Materials* 178 (2018) 51-58.
- [6] R. Mejía de Gutiérrez, M.A. Villaquirán-Caicedo, L.A. Guzmán-Aponte, Alkali-activated metakaolin mortars using glass waste as fine aggregate: Mechanical and photocatalytic properties, *Construction and Building Materials* 235 (2020) 117510.
- [7] G.F. Huseien, M.M. Tahir, J. Mirza, M. Ismail, K.W. Shah, M.A. Asaad, Effects of POFA replaced with FA on durability properties of GBFS included alkali activated mortars, *Construction and Building Materials* 175 (2018) 174-186.
- [8] S. Subaşı, H. Öztürk, M. Emiroğlu, Utilizing of waste ceramic powders as filler material in self-consolidating concrete, *Construction and Building Materials* 149 (2017) 567-574.
- [9] L.G. Li, Z.Y. Zhuo, J. Zhu, J.J. Chen, A.K.H. Kwan, Reutilizing ceramic polishing waste as powder filler in mortar to reduce cement content by 33% and increase strength by 85%, *Powder Technology* 355 (2019) 119-126.
- [10] H. Mohammadhosseini, N.H.A.S. Lim, M.M. Tahir, R. Alyousef, H. Alabduljabbar, M. Samadi, Enhanced performance of green mortar comprising high volume of ceramic waste in aggressive environments, *Construction and Building Materials* 212 (2019) 607-617.
- [11] L. Li, W. Liu, Q. You, M. Chen, Q. Zeng, Waste ceramic powder as a pozzolanic supplementary filler of cement for developing sus-tainable building materials, *Journal of Cleaner Production* 259 (2020) 120853.
- [12] N.H.A.S. Lim, H. Mohammadhosseini, M.M. Tahir, M. Samadi, A.R.M. Sam, Microstructure and Strength Properties of Mortar Con-taining Waste Ceramic Nanoparticles, *Arabian Journal for Science and Engineering* 43(10) (2018) 5305-5313.
- [13] A.M. Rashad, G.M.F. Essa, Effect of ceramic waste powder on alkali-activated slag pastes cured in hot weather after exposure to ele-vated temperature, *Cement and Concrete Composites* 111 (2020) 103617.

- [14] H. Binici, Effect of crushed ceramic and basaltic pumice as fine aggregates on concrete mortars properties, *Construction and Building Materials* 21(6) (2007) 1191-1197.
- [15] R.M. Senthamarai, P.D. Manoharan, D. Gobinath, Concrete made from ceramic industry waste: Durability properties, *Construction and Building Materials* 25(5) (2011) 2413-2419.
- [16] P. Torkittikul, A. Chaipanich, Utilization of ceramic waste as fine aggregate within Portland cement and fly ash concretes, *Cement and Concrete Composites* 32(6) (2010) 440-449.
- [17] R. El-Hachem, E. Rozière, F. Grondin, A. Loukili, New procedure to investigate external sulphate attack on cementitious materials, *Cement and Concrete Composites* 34(3) (2012) 357-364.
- [18] H.N. Atahan, D. Dikme, Use of mineral admixtures for enhanced resistance against sulfate attack, *Construction and Building Materials* 25(8) (2011) 3450-3457.
- [19] G. Indu Siva Ranjani, K. Ramamurthy, Behaviour of foam concrete under sulphate environments, *Cement and Concrete Composites* 34(7) (2012) 825-834.
- [20] S. Boudali, D.E. Kerdal, K. Ayed, B. Abdulsalam, A.M. Soliman, Performance of self-compacting concrete incorporating recycled concrete fines and aggregate exposed to sulphate attack, *Construction and Building Materials* 124 (2016) 705-713.
- [21] P.K. Acharya, S.K. Patro, Acid resistance, sulphate resistance and strength properties of concrete containing ferrochrome ash (FA) and lime, *Construction and Building Materials* 120 (2016) 241-250.
- [22] Z.-T. Chang, X.-J. Song, R. Munn, M. Marosszeky, Using limestone aggregates and different cements for enhancing resistance of concrete to Sulfuric acid attack, *Cement and Concrete Research* 35(8) (2005) 1486-1494.
- [23] E. Gruyaert, P. Van den Heede, M. Maes, N. De Belie, Investigation of the influence of blast-furnace slag on the resistance of concrete against organic acid or sulphate attack by means of accelerated degradation tests, *Cement and Concrete Research* 42(1) (2012) 173-185.
- [24] M.A. Asaad, N.N. Sarbini, A. Sulaiman, M. Ismail, G.F. Huseien, Z. Abdul Majid, P. Bothi Raja, Improved corrosion resistance of mild steel against acid activation: Impact of novel *Elaeis guineensis* and silver nanoparticles, *Journal of Industrial and Engineering Chemistry* 63 (2018) 139-148.
- [25] H. Siad, M. Lachemi, M. Sahmaran, K.M.A. Hossain, Effect of glass powder on sulfuric acid resistance of cementitious materials, *Construction and Building Materials* 113 (2016) 163-173.
- [26] Z. Makhloufi, T. Bouziani, M. Hadjoudja, M. Bederina, Durability of limestone mortars based on quaternary binders subjected to sulfuric acid using drying-immersion cycles, *Construction and Building Materials* 71 (2014) 579-588.
- [27] American Society for Testing and Materials (ASTM), Standard Test Methods for Chemical Resistance of Mortars, Grouts, and Mono-lithic Surfacing and Polymer Concretes, C39/C39M, West Conshohocken, 2012.
- [28] K. Muthusamy, N.W. Kamaruzzaman, M.A. Zubir, M.W. Hussin, A.R.M. Sam, A. Budiea, Long Term Investigation on Sulphate Resistance of Concrete Containing Laterite Aggregate, *Procedia Engineering* 125 (2015) 811-817.
- [29] B. Qi, J. Gao, F. Chen, D. Shen, Evaluation of the damage process of recycled aggregate concrete under sulfate attack and wetting-drying cycles, *Construction and Building Materials* 138 (2017) 254-262.