

Bauxite Residue as Raw Material for Manufacture of Synthetic Aggregate

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Abstract

The amount of solid waste generated in mining and metallurgy is a relevant socio-environmental issue. The construction industry can be a potential consumer of most of this material, mainly to supply the shortage of raw material. In this context, this work presents the studies carried out for the recycling of bauxite residue as raw material for the manufacture of synthetic aggregate for construction. Two different types of synthetic aggregates were produced, varying the proportions of raw materials (bauxite residue, silica and clay) and sintered at 1200 °C. The aggregates were characterized by X-ray fluorescence, X-ray diffraction, scanning electron microscopy and compressive strength. It was observed that the properties of these materials depend on the control of parameters such as free silica and clay content, particle size distribution and sintering temperature. The control of these variables allowed an understanding of the glassy phase formation, which is responsible for the properties of ceramic materials such as porosity, compressive strength and density. The results indicated that the different types of aggregates produced are in accordance with the technical criteria for the construction industry.

Keywords: Bauxite residue, Aggregate, Construction industry.

1. Introduction

Synthetic aggregate is known in the literature as lightweight expanded clay aggregate and has been used in Brazil since the 1980s in civil engineering projects including geotechnical applications, due to its diversity in density, high shear strength and favorable drainage characteristics [1], providing various applications in the construction industry.

The synthetic aggregate is formed from silicoaluminates submitted to a sintering process at high temperatures (1100 - 1250 °C), and may also present expansion due to the inclusion of gases,

normally processed in rotary furnace, resulting in an increase in the porosity of the grains and unique characteristics, such as: lightweight, water absorption capacity and insulation, both thermal and acoustic. Its low density makes this material suitable for use in construction as embankments on soils with low rigidity and as a filling material in retaining structures, aiming at reducing active pressures [2].

Because it is a manufactured product, many of its characteristics, such as porosity and density, can be properly modified in the manufacturing process in order to obtain the desired grain characteristics. Regarding its lightweight, the most important parameter is the porosity of the core. Pores can be divided into accessible and closed pores, which cannot be filled with water, even if the grain is submerged for long periods [1, 2].

Research shows that the compressive strength of the aggregate is conditioned by the characteristics of each grain; therefore, there are several studies on the compressive strength of grains under uniaxial compression in different materials. In addition to the material that forms the grain, breakage is influenced by other factors such as water content, grain size, shape and chemical composition, which depends on the composition of the original clay and other materials added during the process [2].

Studies have shown several successful applications of synthetic aggregate as a geotechnical infill material in the 1980s, including the rehabilitation of a river port terminal and the construction of a bridge. Due to its grain size and lightweight nature, synthetic aggregate is easy to transport, fill and handle. Developed during the sintering process, the pores can be fully closed within a given particle or exposed to its external surface, with the surface of the synthetic aggregate particle being more resistant than the interior due to the sintering of the expanded clay into a material of the ceramic type [1].

Synthetic aggregate is a versatile material used in various applications, such as in the construction industry, where it can be used in the production of lightweight blocks, precast concrete, as well as in structural backfill of foundations. Research has also shown that the aggregate can be used for water treatment, removing fluoranthene, phenanthrene and pyrene. Within horticulture or agricultural field, it can be used for wastewater treatment, due to its high capacity to remove numerous pollutants such as total suspended solids, polyphenols and nitrogen, pesticides and pharmaceuticals [3].

There are many publications related to the use of synthetic aggregate in the construction industry. These publications focus on the use of synthetic aggregate as a partial or complete substitute for normal weight aggregates, which provides greater workability to the resulting material [3]. The literature shows that the aggregate has been used in studies for the production of lightweight concrete, since the use of synthetic aggregate brings benefits by making construction safer, more economical and with less impact on the environment. Other authors have also studied the influence of aggregate sizes on the workability and compressive strength of concrete [4].

Due to different oxides in its composition, the bauxite residue can be an important raw material for the production of synthetic aggregate, since the amount of these oxides can influence the phase transformations that occur during the sintering of the aggregate [5-8]. In this context, the present work aims to carry out the production of synthetic aggregate with bauxite residue in a mixture with silica and kaolinite clay.

2. Materials and Methods

The experiments were carried out for two different types of aggregates, whose amount of bauxite residue varied from 70 to 75 % by weight (AGG-70) and from 80 to 85 % by weight (AGG-80). In both samples, the amount of clay used was 5 % by weight, while the silica varied from 10 to 25 % by weight. Figure 1 presents the flowchart of the synthetic aggregate production process, from the preparation of raw materials to obtaining the final product.

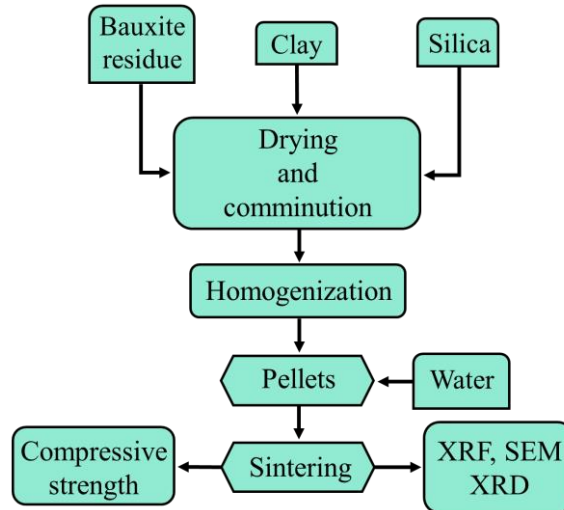


Figure 1. Flowchart of the synthetic aggregate production process.

Before starting the synthetic aggregate production process, the raw materials (bauxite residue, clay, and silica) were prepared through drying and comminution processes. After the treatment of the raw materials, they were mixed and homogenized in a ball mill for a period of 30 minutes. Then, the pellets were produced in a mixture with water for the formation of aggregates. The pellets were sintered at 1200 °C for 3 h.

For the characterization of the aggregate, analyzes of X-ray fluorescence (XRF), X-ray diffraction (XRD), scanning electron microscopy (SEM) and compressive strength were performed.

In the XRF analyses, the samples were mixed with a binding agent in the proportion 10 g of sample to 2 g of binding agent until complete homogenization. The next step was pressing using a manual press spectrometric sample compactor, pressure 20 ton, to make the sample pellets.

For XRD analyses, the aggregates were broken and ground using a hydraulic press. After spraying, there was a granulometry control in accordance with the specifications recommended for the powder method. Subsequently, the samples were exposed to a heat treatment at 105 °C for a period of approximately 2 hours to remove excess moisture.

The samples were analyzed using a scanning electron microscope operating with 5 kV beams. The sample preparation procedure consisted of mechanically breaking the aggregate and polishing its surface. Then, the samples were exposed to an acid attack to remove the glass phase, or part of it, in order to allow a better visualization of the mullite phase, in case it was formed during the sintering process. The acid used was hydrofluoric acid in a 10 % concentration solution, in which the samples were immersed for 60 seconds followed by washing in running water. After the acid attack, the samples were dried in an oven with air circulation for a period of 24 hours at 105 °C.

For the analysis of compressive strength, initially, tests were carried out to obtain the best mortar content, in order to obtain a concrete that meets the requirements of workability, mechanical strength and durability. The specimens were molded in a cylindrical shape, 100x200 mm, and demolded after 24 hours to be cured in a humid environment. The specimens were removed from the cure 4 hours before the compressive strength analysis, which took place at three different ages: 3, 7 and 28 days.

The sintering process of the aggregate represents the closing of the pores, which may be associated with the compressive strength and with the efficiency of the solid-state reactions, which are responsible for the formation of mullite and the glassy phase [5, 9, 10].

The secondary mullite (responsible for improving the mechanical properties of the ceramic material) is formed in an exothermic reaction which, for kaolinite, occurs at 1250 °C. However, as the bauxite residue and clay present impurities, such as Na, K and Fe, the formation of mullite can occur between 1050 and 1250 °C [5]. These transformations can be observed by X-ray diffraction analysis and complemented by scanning electron microscopy, as will be presented in this work.

3. Results and Discussion

3.1 Chemical Analysis

Table 1 shows the chemical compositions in terms of the oxides present in the aggregates AGG-70 and AGG-80.

Table 1. Main chemical composition of AGG-70 and AGG-80 (wt. %).

Component	AGG-70	AGG-80
Na ₂ O	7.11	7.68
MgO	0.578	0.559
Al ₂ O ₃	14.5	16.9
SiO ₂	39.2	29.3
SO ₃	0.224	0.201
K ₂ O	0.299	0.323
CaO	1.12	1.02
TiO ₂	4.53	5.22
Cr ₂ O ₃	-	0.104
MnO	0.148	0.153
Fe ₂ O ₃	30.6	37
ZrO ₂	0.975	1.05
Pr ₆ O ₁₁	0.118	0.106
Nd ₂ O ₃	0.24	0.252

It can be seen in Table 1 that the aggregate samples have a high content of fluxing material such as iron and sodium, from the bauxite residue, which significantly reduces the sintering temperature.

The sintering process of the bauxite residue, mixed with clay and silica, generates the stability of the sodium contained in the residue in the form of glass. Thus, all soluble sodium before sintering becomes stable and chemically inert. The small differences between the chemical composition of the two samples may be related to the amount of bauxite residue used in the aggregate composition.

3.2 X Ray Diffraction

Figure 2 shows the X-ray diffraction analysis performed on the AGG-80 sample.

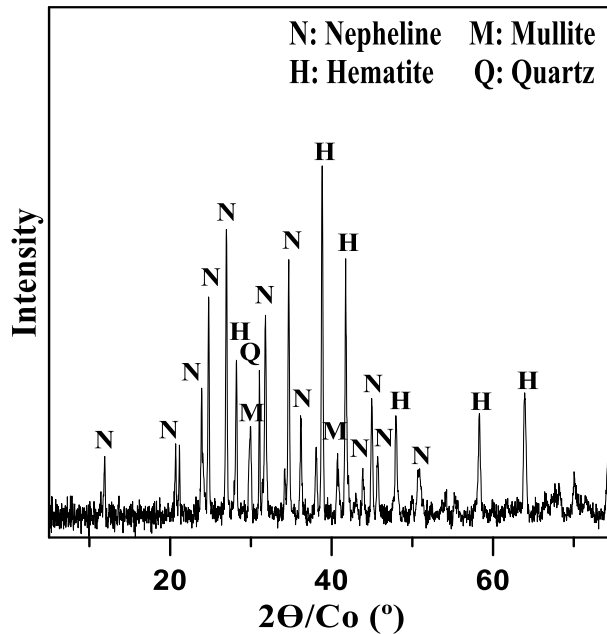


Figure 2. Diffractogram of AGG-80.

The results of the X-ray diffraction analysis of Figure 2 indicated the presence of quartz, hematite, mullite, and nepheline. The presence of mullite, formed during the sintering process, is associated with an increase in the compressive strength of ceramic materials. Studies have shown that the reduction in quartz content is related to the increase in mullite formation [5].

The AGG-80 diffractogram also shows the presence of nepheline, whose formation is due to the thermal treatment of the bauxite residue and the presence of considerable amounts of oxides of sodium and silicon, as shown in Table 1, resulting in the formation of species of the type $\text{Na}_5\text{Al}_3\text{Si}_3\text{O}_{15}$, which are precursors of nepheline [7, 11].

3.3 Scanning Electron Microscopy

Scanning electron microscopy is important to verify the phase formations of the aggregate after the sintering process. Figures 3 and 4 show the scanning electron microscopy analysis for samples AGG-70 and AGG-80, respectively, whose objective was to identify the presence of secondary mullite.

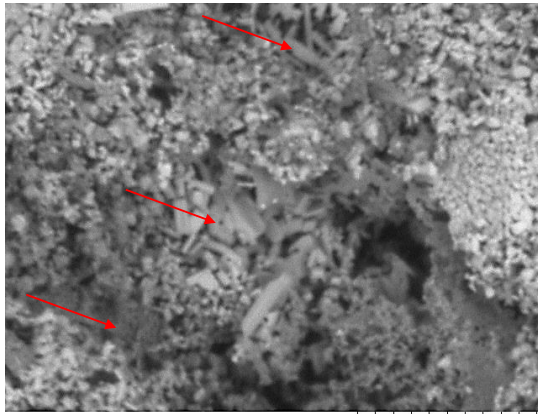


Figure 3. Scanning electron microscopy of AGG-70. Red arrow: secondary mullite.

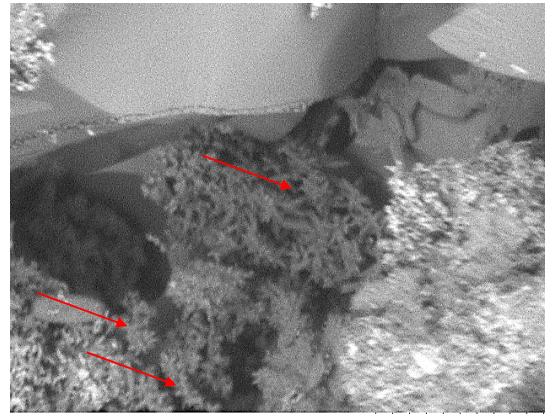


Figure 4. Scanning electron microscopy of AGG-80. Red arrow: secondary mullite.

In the AGG-70 and AGG-80 samples it is possible to observe the presence of the secondary mullite (red arrows) which presents a circular morphology. It should be noted that for its formation a combination of factors is necessary, such as a 3/2 ratio between Al_2O_3 and SiO_2 , adequate temperature, which can vary according to the presence of fluxing agents contained in the material, as in the case of bauxite residue that contain large amounts, which considerably reduces the temperature of mullite formation.

3.4 Compressive Strength

Table 2 presents the results of compressive strength of the samples analyzed at different ages.

Table 2. Compressive strength at different ages (MPa).

Sample	3 days	7 days	28 days
AGG-70	17.96	22.94	35.57
AGG-80	14.34	16.87	26.30

The results in Table 2 showed a satisfactory experimental behavior in relation to compressive strength over time. The specimens presented compressive strength of 35.57 and 26.30 MPa for samples AGG-70 and AGG-80 in 28 days, respectively.

The difference between the compressive strengths for the two samples may be associated with the amount of silica, which was higher for AGG-70 in relation to AGG-80. In the case of sintering carried out at 1200 °C, the increase in the silica content will result in a greater formation of mullite, which is directly related to the compressive strength of the material.

4. Conclusions

Clays with high silica content, or which do not present satisfactory properties to be applied in the ceramic industry, can have their characteristics improved by mixing with bauxite residue, which cannot be used alone as a raw material due to low plasticity, being essential to mix it with clay to allow the extrusion of the material.

The use of bauxite residue as a raw material for the production of synthetic aggregate proved to be adequate, since it has considerable levels of Al_2O_3 and glass-forming compounds, such as Na and Fe. These components allowed the formation of amorphous material (glass) at lower temperatures than the conventional ones, thus facilitating the nucleation of secondary mullite at these temperatures, which was observed in the characterization analyzes.

The application of bauxite residue, in mixtures with clay and silica, offered a great alternative for the manufacture of synthetic aggregate, mainly because they are low-cost raw materials that are generated in large quantities. The two samples, AGG-70 and AGG-80, presented satisfactory results of compressive strength, 35.57 and 26.30 MPa in 28 days, respectively, which should be better evaluated in experiments with full-scale elements (beams, slabs, and columns).

5. References

1. Hongmei Gao, Dynamic shear modulus and damping of cemented and uncemented lightweight expanded clay aggregate (LECA) at low strains, *Soil Dynamics and Earthquake Engineering*, Vol. 142, (2021), 106555.
2. Elias A. Roces-Alonso et al., Experimental study on grain failure of lightweight expanded clay aggregate under uniaxial and biaxial load conditions, *Powder Technology*, Vol. 383, (2021), 542-553.
3. Alaa M. Rashad, Lightweight expanded clay aggregate as a building material - An overview, *Construction and Building Materials*, Vol. 170, (2018), 757-775.
4. Ali H. Nahhab, and Ali K. Ketab, Influence of content and maximum size of light expanded clay aggregate on the fresh, strength, and durability properties of self-compacting lightweight concrete reinforced with micro steel fibers, *Construction and Building Materials*, Vol. 233, (2019), 117922.
5. José Antônio da Silva Souza, *Use of residues from the Bayer process as raw material in the production of synthetic aggregates for the civil construction industry*, PhD Thesis, Federal University of Pará, Belém, Brazil, 2010.
6. D.S. Quaresma et al., Sintering study in vertical fixed bed reactor for synthetic aggregate production, *Cerâmica*, Vol. 63, (2017), 169-177.
7. Bruno Marques Viegas et al., The influence of temperature on phase transformations of the minerals present in the red mud: reduction of hematite to magnetite, *Matéria (Rio J)*, Vol. 25, No. 1, (2020), e-12594.
8. Bruno Marques Viegas et al., Experimental study and mathematical modelling of red mud leaching: application of Bayesian techniques, *International Journal of Environmental Science and Technology*, Article in Press, (2022).
9. H.J. Oel, Sintering of crystalline ceramics, *Fortschritte der Mineralogie*, Vol. 63, Supplement 1, (1985), 167-167.
10. H. Mörtel, and K. Heimstädt, Mikrobielle Werkstoffzerstörung - Simulation, Schadensfälle und Gegenmaßnahmen für anorganische nichtmetallische Werkstoffe: Keramik, *Werkstoffe und Korrosion*, Vol. 45, No. 2, (1994), 128-136.
11. J.M. Rivas Mercury et al., Thermal behavior and physical-mechanical properties of red mud, *Matéria (Rio J)*, Vol. 15, No. 3, (2010), 445-460.