EVALUATION OF MECHANICAL PROPERTIES, RHEOLOGY, AND ENVIRONMENTAL IMPACT OF ALKALI-ACTIVATED TERNARY PASTES USING WASTE GLASS AS A PRECURSOR

Avaliação de propriedades mecânicas, reologia e impacto ambiental de pastas ternárias álcali-ativadas utilizando resíduo de vidro como precursor

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Resumo

O estudo de materiais álcali-ativados como substituto do cimento Portland tem crescido nos últimos anos devido a natureza sustentável desse tipo de material. Apesar disso, pastas álcali-ativadas ternárias utilizando resíduo de vidro como precursor foram insuficientemente estudadas em relação as propriedades reológicas, mecânicas e microestruturais e em relação ao impacto ambiental desse tipo de mistura. Esse artigo busca estudar o efeito do teor do resíduo de vidro (GP) como precursor e o efeito do modulo de sílica (Ms) em pastas ternárias álcali-ativadas utilizando metacaulim, Basic Oxygen Furnace slag e GP. As soluções ativadoras de NaOH e Na_2SiO_3 foram utilizados na produção das pastas. A razão água/ligante foi mantida fixa em 0,30 e dosagem de álcalis foi fixada em 12% Na_2O. Os parâmetros de reologia, resistência a compressão para 1 e 28 dias, impacto ambiental e microestrutura foram analisados.

Palavras-chave: Pasta álcali-ativada; Resíduo de vidro; Módulo de sílica; Reologia; Impacto ambiental.

ABSTRACT

The study of alkali-activated materials as replacements for Portland cement has been growing in recent years due to the sustainable nature of these materials. Nevertheless, ternary alkali-activated pastes using waste glass as a precursor, have been insufficiently studied concerning their rheological, mechanical, and microstructural properties and the environmental impact of this type of mixture. This paper aims to study the effect of glass powder (GP) content as a precursor and the effect of silica modulus (Ms) in alkali-activated ternary pastes of Basic Oxygen Furnace (BOF) slag, metakaolin (MK), and GP. Activator solutions of NaOH and Na_2SiO_3 were used in the production of the pastes. The water/binder ratio was kept fixed at 0.3, and the alkali dosage was fixed at 12% Na2O. The parameters of rheology, compressive strength for 1 and 28 days, environmental impact, and microstructure were analyzed.

Keywords: Alkali-activated paste; Waste glass; Silica modulus; Rheology; Environmental impact.

1 INTRODUCTION

Portland cement is the most used binder in the construction materials industry and its manufacturing process generates environmental liabilities, mainly due to the high release of carbon dioxide (CO_2) into the atmosphere, amounting to 800 kg of CO_2 for every ton of cement produced (LU et al., 2021; DELGADO-PLANA et al., 2021). Alkali-activated materials (AAMs), occasionally called geopolymers, are one of the most studied types of materials today and are seen as possible substitutes for OPC binders since they promote the use of different types of waste and generate a 50-80% decrease in CO_2 emission compared to OPC production process (LONGHI et al., 2019; DUXSON et al., 2007).

The use of glass waste powder (GP) as a precursor in AAMs has been growing because of its high silica content, which meets the requirement for aluminosilicate sources in AAMs (KASTIUKAS and ZHOU 2017). However, the impact of GP on the rheological properties and the study of the silica modulus (Ms = SiO_2/Na_2O of activators) as a variable in GP-based ternary AAMs remains poorly explored. Therefore, this study aims to investigate the influence of the silica modulus (Ms) and GP content on the mechanical properties, rheology, microstructure, and environmental impact of alkali-activated ternary pastes of Basic Oxygen Furnace (BOF) slag, metakaolin, and GP.

2 EXPERIMENTAL PROGRAM

2.1 MATERIALS AND MIX DESIGN

The materials used in this research as precursors were BOF slag, metakaolin (MK) and glass powder (GP) obtained through bottle processing. The materials were milled and then sieved in a 150 µm mesh sieve. The alkaline activators used in the mixtures were NaOH (97,9% purity) and Na_2SiO_3 (SiO_2/Na_2O = 2.21) solutions. The NaOH solution was produced by dissolving solid pellets in water and was mixed with the sodium silicate. The mixing process was carried out using a planetary mortar mixer at 140± 5 rpm for 5 minutes. This paste was then cast into 40-mm side cubic specimens, placed in thermal curing at 60 °C for 24 hours, and stored at room temperature until the required age for hardened state tests.

The particle size analysis of the materials generated results of D_{50} = 52.49 µm and D_{90} = 140.91 µm (GP), D_{50} = 58.79 µm and D_{90} = 148.17 µm (BOF slag), and D_{50} = 23.24 µm and D_{90} = 58.55 µm (MK). In addition, the XRF test allowed the identification of the main oxides present in the chemical composition of the materials, where GP is predominantly composed of SiO_2 (65.34%), CaO (10. 71%) and Na_2O (15.51), BOF slag has a composition based on Fe_2O_3 (25.57%), SiO_2 (12.88%), CaO (37.65%) and Al_2O_3 (7.48%) and MK is mostly composed of SiO_2 (65.44%) and Al_2O_3 (26.66%). The mix design details are shown in Table 1. A total of 9 mixtures were produced, with 3 GP content variations and 3 Ms variations.

Table 1: Material proportions for the alkali-activated mixtures.

Mix	MK (%)	BOF slag (%)	GP (%)	SiO_2/Al_2O_3	Ms	w/b	%Na_2O
T1, T4, T7	45%	45%	10%	2.7	Ms = 1.1 (T1, T2, T3) Ms = 0.85 (T4, T5, T6) Ms = 0.65 (T7, T8, T9)	0.3	12
T2, T5, T8	45%	35%	20%	3.1			
T3, T6, T9	45%	30%	25%	3.4			

2.2 TEST METHODS

The compressive strength of the mixtures was determined at the ages of 1 and 28 days, following the ASTM C109/C109M (2016) and ABNT NBR 13279 (2005) standards, using a DL 30000 Universal Testing Machine of the EMIC brand was used for the tests with a load application speed of 500 N/s. Microstructural analysis of the alkali-activated pastes was conducted using a Quanta 450-FEG scanning electron microscope (SEM) at 28 days of age. Also, an environmental impact analysis was performed to understand the impact of GP content and Ms on CO_2 emissions and energy consumption, comparing the impact of AAMs with OPC-based blends.

The rheology of the alkali-activated pastes was assessed by performing a flow test using plate-to-plate geometry with a 40 mm diameter and a gap set at 1 mm in an AR2000 rotational rheometer (TA Instruments). The test was conducted at 25°C and consisted of a pre-shear procedure at a shear rate of 100 s^{-1} for two minutes, followed by 3 cycles of positive and negative variation of the shear rate with a rate range of 0 to 100 s^{-1}. Each cycle lasted two minutes, totaling a 14-minute test duration. This test was performed 10 minutes after producing each paste. The Bingham model ($\tau = \tau_0 + \mu_p \cdot \dot{\gamma}$) was selected as a representative rheological model for the alkali-activated pastes.

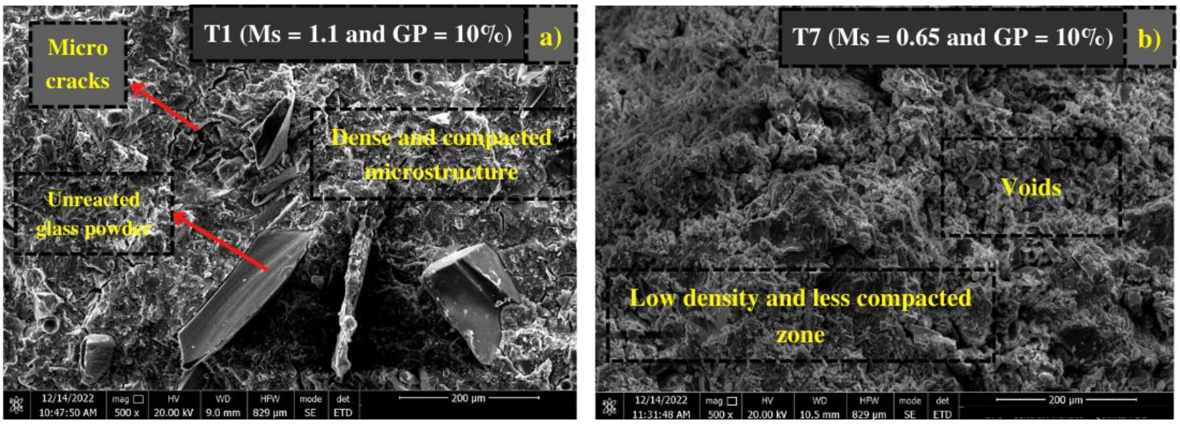
3 TEST RESULTS AND DISCUSSION

3.1 COMPRESSIVE STRENGTH AND MICROSTRUCTURE

The compressive strength results are exposed in Figure 2. The silica modulus has a significant impact on the compressive strength because of the increase in the amount of soluble silica in the matrix for higher values of Ms. As a result, this leads to an increase in alkali-activation processes and reactions, resulting in a greater formation of N-A-S-H and C-A-S-H gels, linked to higher strengths and denser microstructures, as shown in Figures 1 and 2 (MOUSAVINEJAD *et al.*, 2022; DELGADO-PLANA *et al.*, 2021).

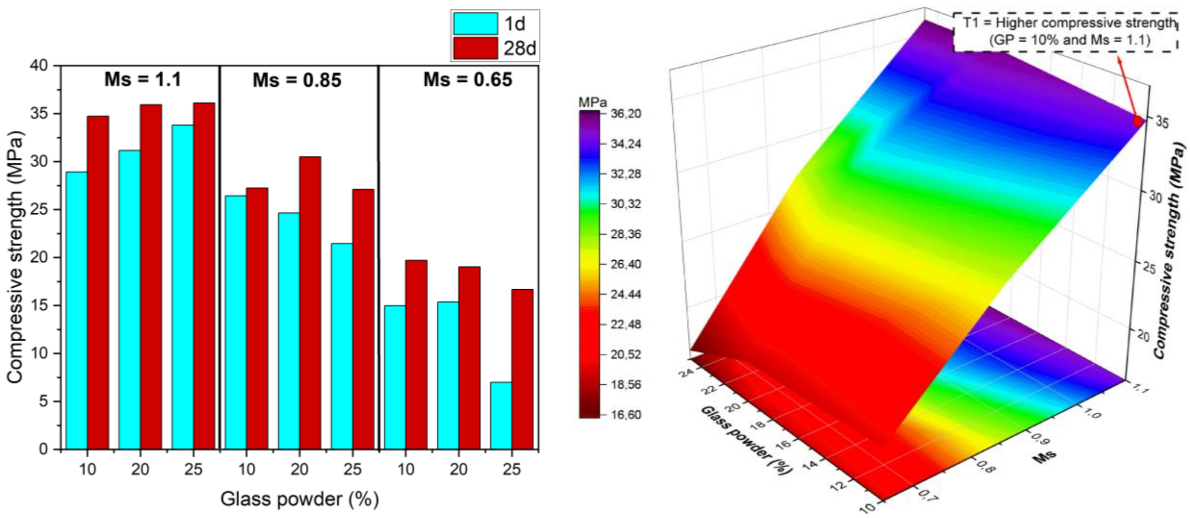
A decrease in compressive strength was observed at 28 days for mixtures containing higher contents of glass powder. This negative impact on mechanical properties can be attributed to the increase in the Si/Al ratio in the matrix from the incorporation of glass as a precursor, leading to the formation of weaker bonds in the alkali-activation process. (LU and POON, 2018; MANIKANDAN *et al.*, 2022). The impact of Ms and GP content on the compressive strength of the mixtures is shown in the response surface graph in Figure 2(b).

Figure 1: SEM images with 500x magnification of the alkali-activated mixtures T1 (a) and T7 (b).



Source: Author (2023)

Figure 2: Compressive strength results for 1 and 28 days (a) and response surface graph of compressive strength considering GP content and Ms as variables (b)



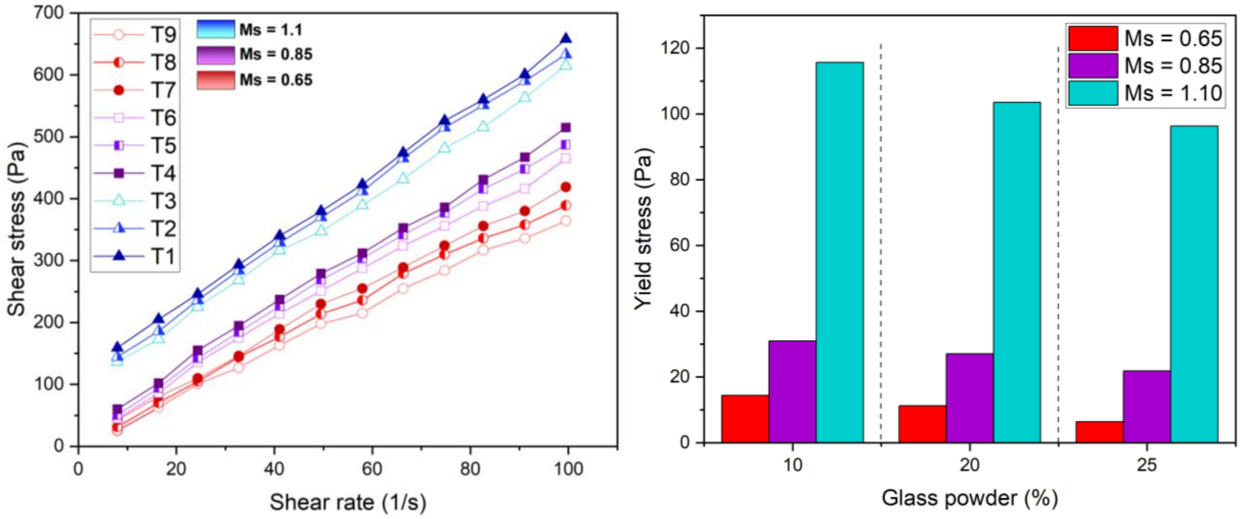
(a) (b)

Source: Author (2023)

3.2 RHEOLOGY

By adopting the Bingham model, Figure 3(a) represents the rheological behavior of the analyzed alkali-activated pastes and shows that the pastes with higher Ms generated higher shear stresses for the same shear rate. Furthermore, an increase in the glass powder content in the mixture resulted in a decrease in shear stress for the flow application at a given shear rate, which is related to the non-absorptive and low reactivity nature of the glass particles. The increase in Ms increases the yield stress (Figure 3(b)), indicating greater resistance to flow. Previous research has reported similar findings for the alkali-activated slag + metakaolin matrix, where an increase in the silica modulus (SiO_2/Na_2O) resulted in higher yield stress and plastic viscosity of AAM (LU *et al.*, 2021; HASNAOUI *et al.*, 2019).

Figure 3: Flow test curves of the alkali-activated mixtures (a) and yield stress values obtained by Bingham model equation (b).



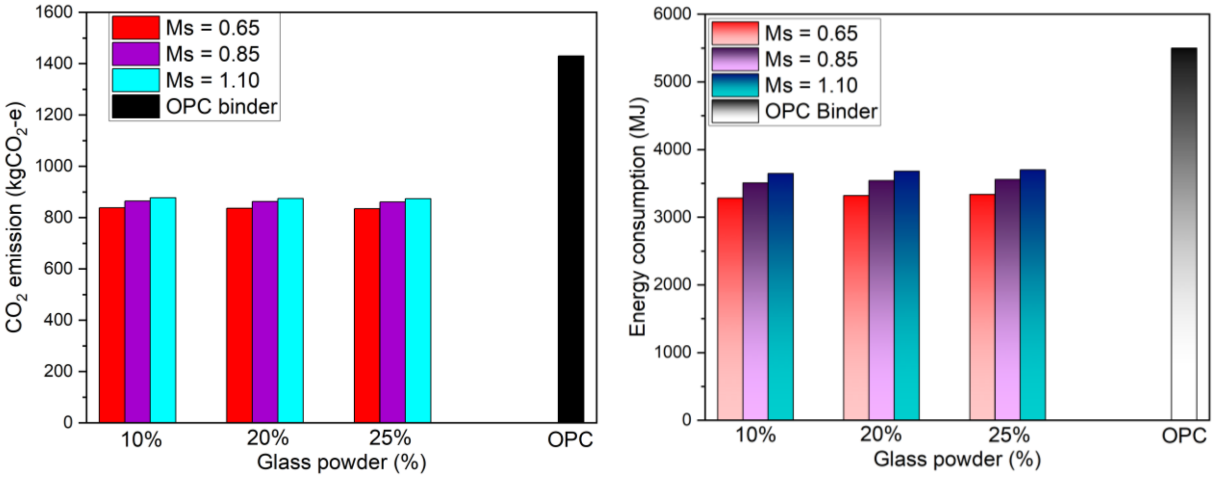
(a) (b)

Source: Author (2023)

3.3 ENVIRONMENTAL IMPACT

The environmental impact of the alkali-activated mixtures was determined based on the literature values (MOCHARLA *et al.*, 2022; XIAO *et al.*, 2020; MAHMOODI *et al.*, 2022). These values made it possible to calculate the equivalent amount of CO_2 emitted and the energy consumed for each of the mixtures studied, considering the production of 1 m³ of paste. These results are shown in Figure 4 and make it possible to identify the impact of Ms on both properties because for higher Ms values, greater quantities of sodium silicate are used, and this activator is highly linked to larger environmental impacts. Furthermore, there was a significant reduction in environmental impacts for the alkali-activated mixtures compared with the OPC-based mixtures.

Figure 4: CO_2 emission and energy consumption for 1 m³ of the 9 alkali-activated ternary pastes and the OPC paste.



Source: Author (2023)

4 CONCLUSIONS

Based on the findings, the following conclusions were drawn:

- The increase in Ms increased the rheological properties of the yield stress (τ_0). It is also possible to observe a modification in this rheological property when the GP content increases, however, decreasing then.
- The compressive strength showed an increase of approximately 90% when Ms was increased from 0.65 to 1.1. GP content has no significant effect on compressive strength.
- Silica modulus (Ms) had a positive influence on microstructural density, resulting in denser microstructures and fewer voids for Ms =1.1 and the opposite for Ms = 0.65
- The increase in the Ms values generated more equivalent CO_2 emissions and energy consumption for the alkali-activated mixtures. However, the 9 studied pastes generated a lower environmental impact than the OPC-based pastes. These results indicate the environmental benefit of the alkali-activated mixtures studied.

REFERENCES

ABNT NBR 13279: Argamassa para assentamento e revestimento de paredes e tetos–Determinação da resistência à tração na flexão e à compressão. **Associação Brasileira de Normas Técnicas: São Paulo**.

ASTM, A. C109/C109M: Standard Test Method for Compressive Strength of Hydraulic Cement Mortars (Using 2-in. or [50-mm] Cube Specimens). **ASTM International, West Conshohocken**.

DELGADO-PLANA, P.; RODRÍGUEZ-EXPÓSITO, A.; BUENO-RODRÍGUEZ, S.; et al. Effect of Activating Solution Modulus on the Synthesis of Sustainable Geopolymer Binders Using Spent Oil Bleaching Earths as Precursor. **Sustainability: Science Practice and Policy**, v. 13, n. 13, p. 7501, 2021. Multidisciplinary Digital Publishing Institute.

DUXSON, P.; FERNÁNDEZ-JIMÉNEZ, A.; PROVIS, J. L.; et al. Geopolymer technology: the current state of the art. **Journal of Materials Science**, v. 42, n. 9, p. 2917–2933, 2007.DOI: 10.1007/s10853-006-0637-z

GHASEMZADEH MOUSAVINEJAD, S. H.; SAMMAK, M. An assessment of the effect of Na2SiO3/NaOH ratio, NaOH solution concentration, and aging on the fracture properties of ultra-high-performance geopolymer concrete: The application of the work of fracture and size effect methods. **Structures**, v. 39, p. 434–443, 2022.

HASNAOUI, A.; GHORBEL, E.; WARDEH, G. Optimization approach of granulated blast furnace slag and metakaolin based geopolymer mortars. **Construction and Building Materials**, v. 198, p. 10–26, 2019.

KASTIUKAS, G.; ZHOU, X. Effects of waste glass on alkali-activated tungsten mining waste: composition and mechanical properties. **Materials and Structures**, v. 50, n. 4, p. 194, 2017.

LONGHI, M. A.; ZHANG, Z.; RODRÍGUEZ, E. D.; KIRCHHEIM, A. P.; WANG, H. Efflorescence of alkali-activated cements (geopolymers) and the impacts on material structures: A critical analysis. **Frontiers of Materials Science**, v. 6, 2019. doi: 10.3389/fmats.2019.00089

LU, C.; ZHANG, Z.; SHI, C.; et al. Rheology of alkali-activated materials: A review. **Cement and Concrete Composites**, v. 121, p. 104061, 2021. https://doi.org/10.1016/j.cemconcomp.2021.104061

LU, J.; POON, C. Use of waste glass in alkali activated cement mortar. **Construction and Building Materials**, 2018.

MAHMOODI, O.; SIAD, H.; LACHEMI, M.; DADSETAN, S.; ŞAHMARAN, M. Optimized application of ternary brick, ceramic and concrete wastes in sustainable high strength geopolymers. **Journal of Cleaner Production**, v. 338, p. 130650, 2022. https://doi.org/10.1016/j.jclepro.2022.130650

MANIKANDAN, P.; NATRAYAN, L.; DURAIMURUGAN, S.; VASUGI, V. Influence of Waste Glass Powder as an Aluminosilicate Precursor in Synthesizing Ternary Blended Alkali-Activated Binder. **Silicon Chemistry**, v. 14, n. 13, p. 7799–7808, 2022. https://doi.org/10.1007/s12633-021-01533-2

MOCHARLA, I. R.; SELVAM, R.; GOVINDARAJ, V.; MUTHU, M. Performance and life-cycle assessment of high-volume fly ash concrete mixes containing steel slag sand. **Construction and Building Materials**, v. 341, p. 127814, 2022.

XIAO, R.; MA, Y.; JIANG, X.; et al. Strength, microstructure, efflorescence behavior and environmental impacts of waste glass geopolymers cured at ambient temperature. **Journal of Cleaner Production**, v. 252, p. 119610, 2020.