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Characterisation of the Interfacial Transition Zone Between the Cement Matrix and Glass Powder Particles

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Abstract: The article deals with determining of the width of the interfacial transition zone (ITZ) between the waste glass powder (WGP) and the cement matrix. The transition zone negatively affects the resulting mechanical properties of the composite. The article deals with surface treatments using plasma treatments under different gases to reduce the size of the ITZ, namely oxygen (O_2) and hydrogen (H_2) plasma. ITZ is measured using a scanning electron microscope (SEM) in combination with the determination of the percentage representation of individual elements using energy dispersive spectroscopy (EDS). The results are compared with mechanical tests on samples of 20 × 20 × 100 mm size. As part of the article, a reference sample without surface treatment was also measured. The results show that by using oxygen plasma it is possible to reduce the ITZ and thereby also improve the mechanical properties of the resulting cement composites.

Keywords: interfacial transition zone; waste glass particles; cement matrix; scanning electron microscope; mechanical properties.

1 Introduction

Cement composites are made up of three main components: matrix, aggregate, and interfacial transition zones (ITZ) [1]. Furthermore, the cement matrix consists of C-S-H gel, unhydrated clinkers, and portlandite (CH). The ITZ is acknowledged as the most vulnerable segment of cement composites, resulting in a reduced compressive strength compared to aggregate and hardened cement paste [2]. Furthermore, the presence of micro cracks within the ITZ contributes to changing the response of cement-based materials from linear to non-linear under loading conditions [3]. Mechanically, the extent of micro cracks within the ITZ is dictated by the degree of strain mismatch between the aggregate and the paste [4]. When stress is evenly distributed, differences in strain arise due to variations in the elastic modulus, the shear modulus of the aggregate, the matrix, ITZ, and the stress concentration. Serving as a bridge between the matrix and the aggregate, if the ITZ does not transmit stress effectively between phases, the compressive strength of the cement composite is compromised, even if one phase exhibits greater stiffness [5].

To understand the mechanism of formation of ITZ and its impact on the properties of cement composites, a detailed microstructural analysis was performed. It has been observed that the presence of CH, along with relatively high porosity, respectively, low-density C-S-H gel (LD CSH) and large capillary pores, significantly influences mechanical properties. Furthermore, the presence of large CH particles weakens van der Waals forces and has adverse effects on mechanical properties [6]. Apart from CH, micro cracks and high porosity are prone to forming within the ITZ, further diminishing mechanical properties. Another indicative factor for ITZ formation is the calcium-to-silicon (Ca/Si) ratio in the cement matrix. A ratio of 0.5-1.5 results in the formation of high-density C-S-H gel (HD CSH), while a ratio of 1.5 – 2.0 yields LD CSH [7]. Another significant effect is the high content of amorphous SiO_2 in WGP, exceeding 70 %, which, when exposed to an alkaline environment, can dissolve and generate a secondary C-S-H gel [8]. This pozzolanic reaction occurs gradually, enhancing the strength of the samples at later stages [9].

The focus of this article lies in the utilisation of WGP within cement paste, with the aim of enhancing its activation through surface plasma treatments. The objective is to decrease the thickness of the ITZ between WGP and the cement matrix, thereby leveraging the potential for the secondary formation of C-S-H gels. This approach aims to create a compact structure with a higher strength value compared to the reference sample.

2 Materials and Samples

The samples were composed of 80 wt. % Portland cement CEM I 42.5R (Radotín) and 20 wt. % of WGP, which comes from the waste material of the Recifa Packaging Industry, as. The water-to-binder ratio (w/b) was maintained consistently at 0.35. Furthermore, the glass powder underwent two different plasma treatments commonly employed to alter surface-wetting properties. The radiofrequency (RF) plasma treatment was conducted at low temperatures using a large-area, low-pressure system (AK 400, Roth & Rau). This process was carried out for 5 minutes in either oxygen or hydrogen atmospheres at a pressure of 15 Pa, with RF power set to 600 W and a substrate stage bias between 10 and 15 V, while keeping the temperature below 50 °C. This process, termed “room temperature” treatment, was repeated four times with periodic mixing of the glass powder to ensure uniformity.

From each batch, 5 samples containing various surface-treated WGP were created. These included a reference sample without surface-treated WGP (REF), WGP treated in an oxygen atmosphere (O_2), and WGP treated in a hydrogen atmosphere (H_2). The samples measured 20 × 20 × 100 mm and were destructively tested at the age of 3 months. Fragments from the destructive testing were embedded in epoxy resin under vacuum. They were then ground and polished in several steps to achieve a smooth surface. Grinding was done using SiC foils with grain sizes of 500 grains/cm², 1000 grains/cm², 2000 grains/cm², and 4000 grains/cm². For polishing, a suspension containing diamonds of 3 and 1 microns in size was used. Finally, the surface of the samples was coated with a conductive layer necessary for electron microscopy.

3 Experimental Methods

The samples were first tested for flexural strength using a three-point bending test. The support span during the test was 80 mm, and the loading was displacement-controlled at a rate of 0.5 mm/min. Subsequently, the broken beams were tested for compressive strength using a uniaxial compression test, which was also displacement-controlled at a rate of 3 mm/min. The effective loaded area was 2 × 2 cm. In both tests, the force at the time of sample failure was measured, which, in combination with the cross-sectional modulus or the cross-sectional area, provides information about the material's strength.

Another experimental method used was microscopic analysis. For this purpose, a ZEISS Merlin scanning electron microscope with a Schottky cathode was employed. The microscope is capable of detecting both secondary and backscattered electrons. In this study, backscattered electrons were analyzed as they provide information about phase contrast. The brighter the phase in the gray scale, the higher the atomic weight of the constituent elements. Additionally, an energy-dispersive spectrometer (EDS) from Oxford Instruments was used. Depending on the emitted X-ray energy, can directly determine the chemical composition of the examined area. To observe the transition zone, line scans were performed to monitor the mass content of calcium, silicon, and oxygen. The line scans ranged from 20 to 35 microns in length, with each scan consisting of 512 measurement points.

4 Results and Discussion

Results of the destructive tests indicate that oxygen plasma treatment improved both compressive strength and flexural tensile strength. The reference sample (REF) had a flexural tensile strength of 9.61 ± 0.30 MPa and a compressive strength of 84.9 ± 5.6 MPa. In comparison, the sample with a surface treated with oxygen plasma (O_2) exhibited a flexural tensile strength of 10.6 ± 0.59 MPa and a compressive strength of 90.0 ± 3.20 MPa. The samples treated with hydrogen plasma (H_2) showed the poorest performance, with a flexural tensile strength of 9.55 ± 0.12 MPa and a compressive strength of 78.3 ± 10.1 MPa. The results suggest that oxygen plasma positively affects the ITZ between the cement matrix and the WGP.

To confirm these findings, electron microscopy was conducted. In Fig. 1, line scans tested using EDS can be seen. According to the available literature, more than 20 different phases can form in the cement matrix, each with distinct structural and micromechanical properties. For simplification, the phases in the matrix can be categorized as follows: unhydrated clinker, which has a high atomic weight and appears white in the structure; portlandite ($Ca(OH)_2$), which appears light gray in images; C-S-H gel, which is gray; and pores and cracks, which are black. C-S-H gel is the most prevalent phase in the structure, and depending on the calcium-to-silicon ratio, it can be identified as low-density C-S-H gel (LD C-S-H) with poor mechanical properties or high-density C-S-H gel (HD C-S-H) with good mechanical properties. According to the literature, the Ca/Si ratio for LD C-S-H ranges from 1.5 to 2.0, and for HD C-S-H from 0.5 to 1.5 [7]. The line scan of the investigated area was always chosen in the region of C-S-H gel. The images in Fig. 1 show that for the reference sample (REF) and the H_2 sample, the size of the ITZ, where the Ca/Si ratio is higher, is 10 microns. In the case of the O_2 sample, the region is smaller, approximately 5 microns and a gradual increase in Ca/Si can be seen, indicating that there is no sudden change in mechanical properties at the interface between the cement matrix and the WGP. These results are consistent with the findings of the destructive tests.

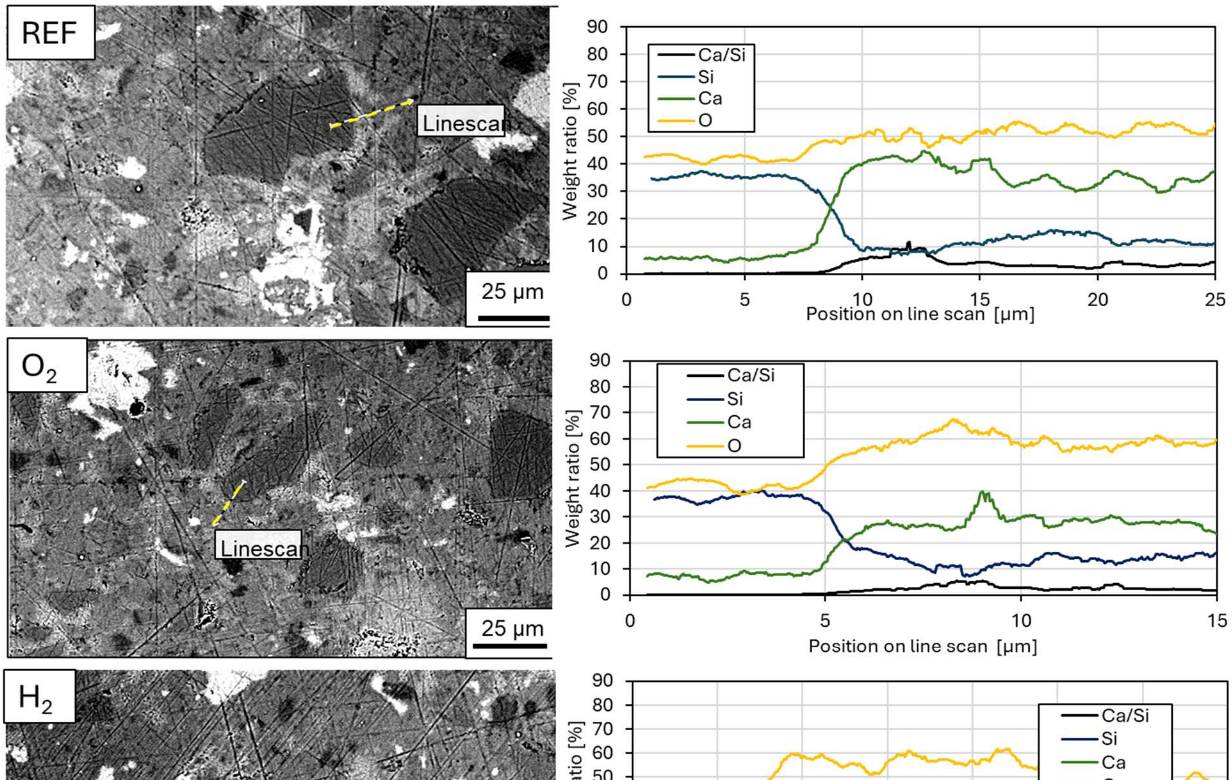

Fig. 1: SEM images of the backscattered electron detector magnified 500 × with the indicated area of line scan, evolution of the weight percentages of oxygen calcium and silicon in line scan.

5 Conclusion

In conclusion, oxygen plasma treatment significantly enhances the mechanical properties of cement composites with WGP. Destructive testing revealed that samples with the content of WGP treated with oxygen plasma showed improved flexural tensile strength and compressive strength compared to the reference and hydrogen-treated WGP. This improvement indicates a beneficial effect of oxygen plasma on the ITZ between the cement matrix and WGP.

Microscopic analysis supports these findings by showing a refined microstructure in the sample with oxygen-treated WGP. Scanning electron microscopy and energy-dispersive spectroscopy revealed a more gradual and smaller ITZ in samples with oxygen-treated WGP, approximately 5 microns, compared to 10 microns in the reference and hydrogen-treated WGP. The smoother transition zone correlates with enhanced mechanical properties, confirming that oxygen plasma treatment effectively improves the interface quality and material performance in cement composites with treated WGP.

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