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STUDY OF WETTABILITY OF CLAYEY CERAMIC AND FLUORESCENT LAMP GLASS WASTE POWDERS

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Abstract

The glass tube of spent fluorescent lamps is contaminated with mercury, which might be a serious hazard in the case of conventional recycling by melting with other glasses. A possible solution could be its incorporation into a clay body to fabricate common fired ceramics such as bricks and tiles. The objective of this work is to characterize a type of fluorescent lamp glass waste to be incorporated into a clayey ceramic. The characterization was performed in terms of wettability tests to evaluate the interaction between the surface of the clayey ceramic and glass waste as a function of the firing temperature. The results showed that the contact angle decreased with increasing temperature, reaching a value of 79°, at a temperature of 1100°C, but not sufficient to completely wet the ceramic. However, compatible chemical composition and reduction of porosity by the flow of soft glass waste between the clay particles favor the consolidation of the ceramics structure above 900°C.

Introduction

Fluorescent lamps use mercury (Hg) as a vital component for the emission of light. This toxic metal is associated with contamination of soils and waters as well as absorption by animals and plants causing serious diseases. As part of the food chain, these Hg-containing animals and plants could be responsible for human health problems. The environmental contamination by Hg from spent fluorescent lamps is a worldwide situation, which is aggravated in Brazil due to the accelerated substitution of incandescent lamps [1]. Indeed, the Brazilian government is promoting a saving in energy program, "Plano Nacional de Eficiência Energética" (National Plan for Energy Efficiency) [2] to replace all incandescent lamps by florescent ones. This program determines that, by July 2016, incandesce lamps shall no longer be produced or commercialized in Brazil. On the other hand, the production of lower energy consuming fluorescent lamps is now being encouraged in the country [3].

Despite the advantages of energy saving, the glass tubes of spent fluorescent lamps will pose an increasing environmental contamination and health problems in Brazil, owing to the Hg release to the ambient. A solution, which is already being conducted, is the Hg decontamination of the spent fluorescent lamps glass tubes [1]. However, the glass waste resulting from his decontamination process still contains traces of Hg and, therefore, should not be directly discarded or recycled in conventional smelting with other glasses. A more secure solution is the incorporation of limited amounts of fluorescent lamp glass waste (FLGW) into clayey ceramics for building construction [1]. The clayey ceramic production is currently one of the industrial sectors that have traditionally been used for incorporation of several types of wastes [4]. This has the advantage of not only mitigate the environmental impact but also recycle the waste to improve the clayey ceramic properties. In particular, glass wastes are being investigated as possible incorporation to clayey ceramics [4-10]. One aspect that has not yet been fully studied in the incorporation of glass wastes is its surface interaction with the clayey ceramic. This study is based on wettability tests that are essential to determine the efficiency of bonding formation between a solid substrate, the ceramic, and a fluid, the molten glass waste. The wettability test measures the contact angle (θ) between the tangent line separating the fluid/solid interface [11] from a drop, spontaneously spread onto the substrate, as schematically shown in Fig. 1.

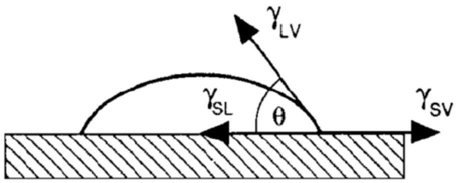

Figure 1. Schematic of a fluid drop spontaneously spread onto a solid substrate. Interface energies: solid/liquid = γ_{SL}; liquid/vapor = γ_{LV}; solid/vapor = γ_{SV}. [11]

The results of wettability tests, Fig. 1, allow the comprehension of the surface interaction mechanism and contribute to select the adequate formulation of phases as well as the convenient firing temperature. In the present work, the change with temperature of the wettability behavior of a FLGW with respect to a clayey ceramic was investigated.

Materials and Methods

The basic materials used in the present work were a fluorescent lamp glass waste (FLGW) and a clayey ceramic. The FLGW was supplied by Brazilian decontamination firm "Brazil – Comércio e Serviços", which uses a "Bulb Eater" equipment, Fig. 2(a), of the Air Cycle Corporation. The decontaminated FLGW was crushed, Fig. 2(b), and still contains a small amount of Hg that is not considered harmful. According to the supplier, the FLGW is a type of soda-lime glass and classified by Brazilian norm [12] as Class II-A; non-dangerous but not inert. In a previous work [1], it was found by optical dilatometry that the same FLGW exhibits a range of working temperature in its pasty condition from 868 to 1049°C and complete melting at 1117°C.

The clayey ceramic used as substrate, Fig. 1, was a typical mixture of kaolinitic clays kindly provided by the "Rodolfo Gama Azevedo – Sardinha" ceramic industry located at the municipal area of Campos dos Goytacazes, state of Rio de Janeiro, Brazil.

To perform the wettability tests, in which the contact angle θ in Fig. 1 is established by the balance between the surface tensions, both the clayey ceramic substrate and the FLGW had to be especially prepared. Press-molded 25 x 25 x 10 mm ceramic plates were first dried in a stove at 110°C for 24 h and then sintered in an electrical laboratory furnace at 1100°C for 120 min.

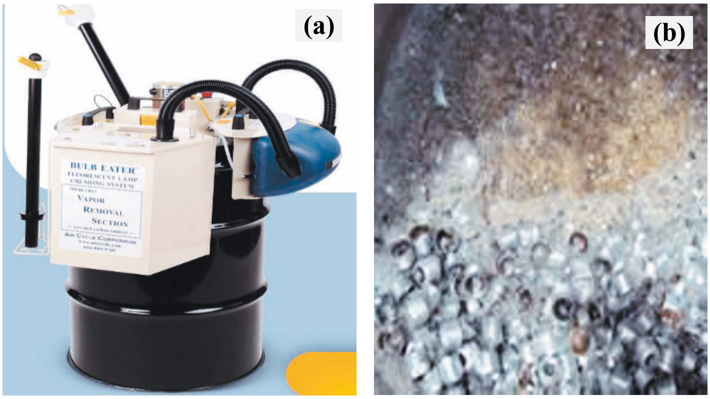

Figure 2. Hg decontamination of glass tubes of fluorescent lamps: (a) “Bulb eater” equipment, and (b) glass fragments produced in the crushing system of the equipment.

The as-received crushed FLGW, Fig. 2(b), was first sieved to 100 mesh (0.149 mm) and then mixed with about 10% of liquid styrene for a pasty consistency to facilitate handling. The mixture was press-molded at 50 MPa in a model MA 098/C Marconi hydraulic press to produce cylindrical samples with 4.0 mm in diameter and 4.5 mm in height.

Wettability tests were conducted with the cylindrical samples (FLGW powder mixed with styrene) placed onto she sintered clayey substrate, Fig. 3, and introduced inside an alumina tubular Lindberg/Blue electrical furnace. Separated samples were heated under air at different temperatures of 850, 900, 950, 1000, 1050 and 1100°C with a heating rate of 10°C/min and dwell time of 10 min. The image of the geometrical transformation of each heated sample was registered in a model Meteor 2 Matrox capture plate operating with a digital JVC-color video camera coupled with a 4/50 objective lens and controlled by a Leica image software. At the end of each experiment, the sample was rapidly cooled to preserve the geometrical change and a picture of the sample was taken with a digital Sony camera. The contact angle was measured in the picture using the Image J computer program.

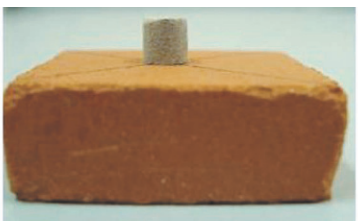

Figure 3. Wettability test sample of a cylindrical glass waste (FLGW) placed onto a clayey ceramic substrate.

The FLGW particles and the sintered clayey ceramic substrate were analyzed by scanning electron microscopy (SEM), after gold sputtering, in a model JSM 6460 Jeol microscope operating with 20 kV.

Results and Discussion

Figure 4 shows SEM images of FLGW particles (a) and the surface of a 1100°C sintered clayey substrate (b). In Fig 4(a) it is observed a broad range of particle sizes from about 1 to 100 µm. Most of the small particles are forming agglomerations, probably due to electrostatic forces. This condition did not apparently affect the high temperature softening to form the molten drop of glass for the wettability tests.

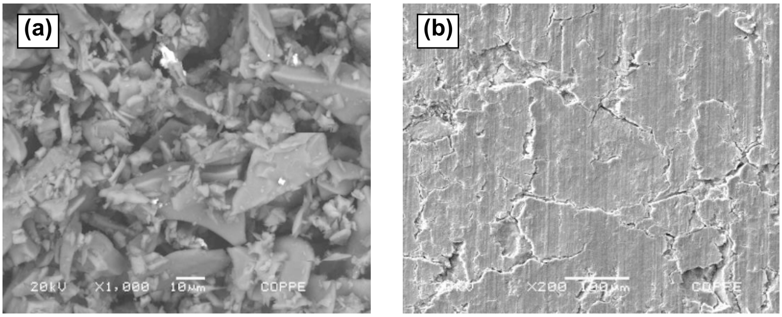

Figure 4. SEM images of (a) fluorescent lamp glass waste particles and (b) surface of the 1100°C sintered clayey substrate.

The relatively flat surface of the clayey ceramic substrate sintered at 1100°C, Fig. 4 (b) reveals a smooth appearance with cracked contours of possible regions associated with crystalline domains. This flat and smooth surface is convenient for the spread of the molten glass, which allows the contact angle in Fig. 1 to be measured.

Figure 5 shows a sequence of images illustrating the FLGW cylindrical sample, Fig. 3, behavior with increasing sintering temperature. In Fig. 5(a), at 850°C, the bulging of the cylindrical lateral surface indicates that the FLGW is beginning to soften, as expected for a soda-lime type of glass [1]. At 900°C, Fig. 5(b), an almost spherical shape of the sample, associated with a θ = 180°, indicates that the glass waste is still unable to wet the substrate. This is interpreted as a non-adhesion condition between the two materials [14].

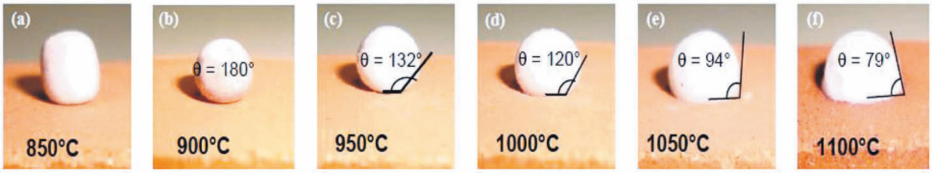

Figure 5. Fluorescent lamp glass waste behavior during wettability tests onto a clayey ceramic substrate at different temperatures.

At 1050°C, Fig. 5(e), the sample assumes an almost semi-spherical shape with $\theta = 64°$, which is related to the melting point [15]. Actually, this temperature is lower than the melting point of a soda-lime glass. However, as a waste, the FLGW is certainly contaminated with impurities that contribute to lower the melting point. At the limit temperature investigated of 1100°C, Fig. 5(f), the contact angle of $\theta = 79°$, is an indication that the FLGW has not yet presented a good wettability with the clayey ceramic. Indeed, a perfect wetting would occur for a contact angle close to zero, i.e., $\theta \approx 0°$. Therefore one may infer that the FLGW at 1100°C has not decreased its viscosity to permit and efficient infiltration inside the open pores of the clayey ceramic structure. In spite of the low wettability, the FLGW may still contribute to consolidate the clayey structure at temperatures above 900°C. The soft glass waste, even with low viscosity, might be squeezed in between the hard particles of the clayey ceramic during industrial firing operation. This process partially contributes to close the clayey particles open interspaces. Moreover, a typical soda-lime glass composition (74% SiO_2; 16% NaO; 5% CaO; 4% MgO; and 1% Al_2O_3) [16] is compatible with the clayey ceramic composition [5]. This also helps to form effective bonding between the soft/molten FLGW and the clay particles during high temperature sintering.

As a final remark, it should be emphasized that the wettability behavior shown in Fig. 5 indicates that, above 900°C, a sensible adherence begins to take place between the FLGW vitreous phase and the clayey ceramic substrate. Although the area of contact was slightly greater than $\boldsymbol{\pi r^2}$ (r = sphere radius) for a contact angle of 79° at 1100°C, the wettability might be enough to promote penetration of the vitreous phase between the clay particles. This results in porosity reduction at temperatures lower than those required for the contribution of solid state diffusion in the neat clayey ceramic. As a consequence, densification should primarily occur due to the flow of the FLGW soft/molten vitreous phase into the open spaces between clay particles. One might then expect that above 900°C the addition of the FLGW should improve the technical properties, mainly water absorption and mechanical strength, of clayey ceramics.

Conclusions

- Wettability tests of fluorescent lamp glass waste, as a fluid phase, onto a clayey ceramic substrate revealed a melting point at 1050°C and a contact angle of 79° at 1100°C.
- Despite the relatively higher contact angle, which is associated with a lower degree of wettability, the glass waste could effectively contribute to the consolidation of the clayey ceramic structure above 900°C, in which $\theta < 180°$.
- The compatible composition of the soda-lime type glass waste and the clay particles contributes to an efficient atomic bonding as the fluid vitreous phase penetrates between the clay particles and promotes densification of the ceramic structure.

Acknowledgments

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