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Verres borosilicatés à séparation de phase pour applications dentaires. Formulation de la composition en relation avec la dissolution des ions en milieu acide et la microstructure.

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Phase separated borosilicate glasses for dental applications. Mixture design in relation with the ion release in acid condition and microstructure.

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ABSTRACT

The research for efficient dental materials has been a constant throughout the history of dentistry. As better materials emerged, the development of ever more innovative materials has been pushed forward. Moreover the combination of different sciences such as chemistry, biology, physics and engineering has provided better knowledge to the demanding requirements of the dental restorations. The BIODENSOL project is a European commission funded mobility research project managed by the University of Lyon and LUCIDEON Ltd, Stoke-on-Trent, UK. The project is designed to help building relations between academic and commercial research to enhance medical innovations and associates solid state chemists and hospital practicioners in direct contact with the patients with dental problems. This project proposes three PhD studies to address the problems of caries and enamel erosion by acidic foods (leading to sensitive teeth) encouraging re-mineralization to help avoid these problems. The researches independently evolved in three different routes investigating three different materials that could provide potential solutions. The main objective of this thesis concerns the study of Borosilicate phase separated glass powders for glass ionomer cements. As silicate bioglasses developed by Hench in 1969, regarding their bioactivity and antimicrobial properties, borosilicates glasses are promising materials and have been widely studied for biomedical applications for scaffolds in soft tissues and for bone repair and could be of interest in dentistry. The glass system studied here is a borosilicate glass (SiO_2-K_2O-B_2O_3-CaO-Al_2O_3) with a strong tendency to phase separate.

The objective of this work is to characterize a series of the novel borosilicate formulations and to understand the mechanism and kinetic of dissolution related to their microstructure and composition. The use of thermal treatment to promote phase separation as a means of influencing the rate of ion leaching was especially studied. The dissolution process of borosilicates in neutral body solution (simulating body fluid) or in acid solution (simulating scenarios where bacteria or acid foods are present) provides information regarding the type and concentration of species released by the glass. Different compositions have been investigated in which SiO_2 and K_2O are fixed while the other elements are varied in order to assess the effect on the phase separation. As the novel borosilicates produced with the melt-quenching technique is immersed in an aqueous environment, B-, Si-, Ca-, K- and Al- species are released to different degrees as a function of the time. Ion leaching trends can be related to the glass chemistry and microstructure.

It has been shown that the amorphous phase separation (APS) causes an initially homogeneous single phase to separate into two or more phases of different compositions. The degree of interconnectivity of the two glass phases depend on the nature of the phase separation mechanism. This process can occur by a nucleation and growth process which gives isolated spherical particles or by spinodal decomposition where an interconnected structure is obtained. It is significant that before a heat-treatment the glasses are optically clear, but turn opalescent to different degrees following a heat treatment depending on the wt% of the elements in the composition. The pattern of the ion release implies that one of the phases is more reactive and susceptible to acid attack and will be leached out from the glass earlier. The other phase will remain in the cement improving the mechanical properties of the dental

restorative material. Moreover, the mixing of the glass powder with a commercial poly(acrylic acid) has shown interesting mechanical and bioactive properties.

This work showed how the ions leaching are influenced by the glass composition and the heat-treatment. The study of the behaviour of the borosilicate in different acid solutions and the elements leached out could be a promising investigation for the analysis of the properties of the final dental restoration.

Key words: Borosilicates, Glass ionomer cements, Ions release, Phase separation, Mechanical properties, Biological properties.