Water dynamics in glass ionomer cements

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Received 20 October 2015 / Received in final form 2 May 2016
Published online 15 July 2016

Abstract. Glass ionomer cements (GIC) are an alternative for preventive dentistry. However, these dental cements are complex systems where important motions related to the different states of the hydrogen atoms evolve in a confined porous structure. In this paper, we studied the water dynamics of two different liquids used to prepare either conventional or resin-modified glass ionomer cement. By combining thermal analysis with neutron scattering data we were able to relate the water structure in the liquids to the materials properties.

1 Introduction

The incorporation of a resin component to conventional glass ionomer cements (CGIC) is one strategy used to strengthen such material. Resin-modified glass ionomer cements (RMGIC) have longer working time, improved aesthetics, a rapid initial setting and enhanced initial strength when compared to CGIC \cite{1–3}. However, in RMGIC the inclusion of the resin component makes the setting reaction more complex. In this material both (i) an acid-base reaction between the glass particles and the polyacid solution and (ii) a polymerization reaction of the resin monomers (i.e. hydrophobic methacrylates) induced by photo-initiation occur. Since a percentage of the water in RMGIC is replaced by resin monomers (in variable amounts depending on the commercial product), the initial setting is due to the rapidly occurring polymer chain propagation as a result of the polymerization reaction \cite{1}. Consequently, a significant improvement in the strength of RMGIC during the critical first hours of setting is observed when compared to CGIC \cite{2}. It is generally accepted that during the early RMGIC setting process, the acid-base and polymerization reactions compete and inhibit each other \cite{4–6}. Thus, proper polymerization of the resin component is vital to avoid RMGIC setting too slowly and consequently becoming weaker than a CGIC.

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To fully understand these limitations, there is a clear need to understand proton mobility within CGIC and RMGIC.

In this work we aimed to better understand the dynamics of the water contained in the different liquids used to prepare the GIC. Neutron scattering was deemed desirable since the neutron is not only supremely sensitive to proton mobility but also is able to penetrate deeply into the cement material without perturbing its chemical make up. In addition, the high incoherent scattering cross section of the hydrogen atom makes incoherent inelastic neutron scattering ideal for measuring the dynamics, and consequently hydrogen bond formation, of liquid mixtures. Indeed, when combined with thermal analysis, proton dynamics can be directly linked to liquid crystallisation [7].

We compared the thermal behaviour of two commercial liquids provided by Voco GmbH (Germany). One contains an aqueous solution of polyacrylic acid (hereafter referred to as Liquid Poly) and the other a polyacrylic acid aqueous solution containing different methacrylates, mainly 2-hydroxyethyl methacrylate (hereafter referred to as Liquid Hema). In the later case the methacrylates partly replace the water. The first solution is used to prepare a CGIC (Ionofil Molar AC, hereafter referred to as Poly), while the second is used to prepare a RMGIC (Ionolux AC, hereafter referred to as Hema). Both GIC powders contain irregular particles of fluoroaluminosilicate glass. In the present study, we have also related the thermal behaviour of the liquids to the formation of hydrogen bonds in both cements during the first 24 hours of setting.

2 Experimental details

2.1 Differential scanning calorimetry

The phase transitions of water in Liquid Poly and Liquid Hema were measured by means of differential scanning calorimetry (DSC) using a DSC 214 Polyma from Netzsch. The experimental conditions were: N\textsubscript{2}-atmosphere (40 ml/min), heating (cooling) rates of 10, 5 and 2 K/min. The samples weights were approx. 20 mg. These measurements were performed between 40°C (313 K) and −180°C (93 K) in an aluminium closed crucible. An empty crucible was also measured and used for instrument correction.

After first cooling at 10 K/min, the sample was kept at the lowest temperature for 5 minutes and then heated up to 40°C. Reproducibility of the process was verified after having kept the sample at 40°C for 5 minutes and then cooling it again using the same rate. For each different temperature rate, the same procedure was followed. The data was processed using the software provided by Netzsch.

2.2 Quasi-elastic neutron scattering

For all samples proton mobility was investigated using quasi-elastic neutron scattering (QENS) by means of the elastic fixed window (EFW) method. Here we evaluate the evolution of only those neutrons scattered elastically by the sample as a function of time or temperature. Monitoring the elastic signal as a function of temperature allows us to determine the onset of proton mobility by noting points of inflexion in the elastic scattering response. In contrast, by evaluating the elastic intensity as a function of time it is possible to follow the hydration process, and thus access the rate of hydrogen bond formation [8].

In this paper the first 24 hours of setting for two freshly mixed cements, namely Poly and Hema, were evaluated. The investigated cements were prepared by mixing