Secondary relaxations in ultrastable glasses and their connection with structural relaxation

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Abstract

Secondary relaxations persistent in the glassy state below Tg are remarkably relevant due to their connection to several properties of the glass. There are different types of secondary relaxations and not all of them are important for the same reason. One of the main concerns is to identify which of those secondary relaxations are relevant for their connection to the structural relaxation. Glass aging is often studied to understand this issue, but it is often insufficient, since it is a slow and inefficient process to explore the energy landscape. The so-called ultrastable glasses, formed by physical vapour deposition at proper growth conditions, exhibit density and enthalpy levels comparable to those ordinary glasses aged for thousands or millions of years, and permit to analyse the evolution of secondary processes in cases where aging does not provide any information. Here, we present a recent study on the secondary relaxations of several ultrastable glassy materials to identify different types of relaxations and its relationship with the structural relaxation. We show the existence of two clearly differentiated groups of relaxations: those which are slower in the ultrastable state and those which are faster, with respect to the ordinary unaged glass. For the first group, we find an unprecedented connection between secondary and structural relaxations in the ultrastable glass in exactly the same manner as in the ordinary glass, manifested through different properties, such as thermodynamic state or devitrification temperature at variable pressure. Furthermore, we propose a model based on density heterogeneities to distinguish between those relaxation which are connected to the structural relaxation and those which are originated from the particular microstructure of the material.

Keywords: Secondary relaxation, ultrastable glasses, physical vapour deposition, dielectric spectroscopy, organic glasses.

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