Combined Effects of Compression and Sub-$T_g$ Annealing on Structure and Properties of Aluminosilicate Glass

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The structure and properties of glasses can be modified through changes in their volume (density). Densification has previously been found to increase the glass hardness1,2, refractive index1, heat capacity overshoot during glass transition1,3, and elastic moduli2 across a variety of oxide compositions. However, the densification mechanism and the structural origin of the changes in properties are not well understood. Here, we present results on the combined effects of isostatic compression at elevated temperature (hot compression), sub-$T_g$ annealing, and K$^+$-for-Na$^+$ ion exchange on the mechanical properties of a commercial sodium-magnesium aluminosilicate glass. Considered separately, each of these three post-treatments are known to increase the glass hardness, but the combined effects are not well understood. Here we use $^{23}$Na 3Q (triple quantum) MAS (magic angle spinning) NMR spectroscopy to study the relation between changes in local atomic environments, density, and mechanical properties. The effect of ion exchange on hardness is found to be sensitive to the isostatic compression temperature, which in turn governs the local structure around Na$^+$. When pre-compressed glasses are sub-$T_g$ annealed at ambient pressure, the pressure-induced densified structure relaxes, i.e., density decreases. On the other hand, when pre-annealed samples are hot compressed at $T_g$, the structural modifications induced by sub-$T_g$ annealing will also relax. To combine the effects of sub-$T_g$ annealing and hot compression, we therefore perform hot compression followed by in situ sub-$T_g$ annealing under 1 GPa pressure. We find that this combined treatment increases the hardness to a greater extent than hot compression alone. We also study the hardness and density relaxation during subsequent ambient pressure sub-$T_g$ annealing, along with changes in the heat capacity overshoot during glass transition.