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Pressure-Promoted Relaxation: Access to Forbidden Glassy States

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Abstract: The structure and properties of glass can be modified through compression near the glass transition temperature (T_g), but once the compressed glass undergoes annealing near T_g at ambient pressure, the modified structure and properties will relax. First, we show how the property relaxation is correlated with both the local and the medium-range structural relaxation in a sodium borate glass that has first been compressed at its T_g at 1 GPa, and then annealed at ambient pressure under different temperature-time conditions. The pressure-induced structural conversions are reversible during ambient pressure annealing near T_g , but exhibit a dependence on the annealing temperature. However, the conversions between structural units cannot account for the pressure-induced densification, and instead we suggest the packing of structural units as the main densification mechanism. Second, we also show that by first compressing an aluminosilicate glass at 1 GPa at T_g , followed by sub- T_g annealing *in situ* at 1 GPa, it is possible to combine the effects of hot compression and ambient pressure annealing. Through density, hardness, and heat capacity measurements, we demonstrate that the effects of hot compression and sub- T_g annealing can be combined to access a “forbidden glass” regime of high density and hardness that is inaccessible through thermal history or pressure history variation alone.