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Publication date: 2018

Link to publication from Aalborg University

Citation for published version (APA):

Smedskjær, M. M. (2018). Densification and Cracking Behavior of Modifier-Free Mixed Network Glasses: Invited Talk. Abstract from ICG Annual Meeting 2018, Yokohama, Japan.

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### **Densification and Cracking Behavior of Modifier-Free Mixed Network Glasses**

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Obtaining a combination of high crack resistance and hardness is important for many glass applications, but unfortunately these tend to be mutually exclusive. Focusing here on mixed network glasses without the presence of network modifiers, we propose a new strategy for overcoming this conflict by identifying new structural motifs for designing hard and crack resistant glasses. Specifically, we report an unusual phenomenon of increase in the densification contribution to deformation of mixed network Al<sub>2</sub>O<sub>3</sub>-B<sub>2</sub>O<sub>3</sub>-P<sub>2</sub>O<sub>5</sub>-SiO<sub>2</sub> bulk glasses following pre-densification of the glasses at elevated temperature [1]. These glasses feature the unusual AlPO<sub>4</sub> and BPO<sub>4</sub> units with four bridging oxygens in PO<sub>4</sub> tetrahedra, and their relatively low atomic packing density favors network densification when subjected to high local stress [2]. However, hitherto unique to the studied glass composition, the high-temperature densification treatment reduces the residual stress during subsequent sharp contact loading, which in turn leads to a simultaneous increase in hardness and crack resistance. Based on structural characterization using micro-Raman and NMR spectroscopy, we show that the more densified medium-range order of the hot compressed glass results in formation of certain structural states, which could not be reached through any composition or thermal path. Accessing such "forbidden" structural states through the identified densification mechanism thus pushes the boundaries for structural design of mechanically resistant glasses.

[1] Kapoor S., Januchta K., Youngman R. E., Guo X. J., Mauro J. C., Bauchy M., Rzoska S. J., Bockowski M., Jensen L. R., Smedskjaer M. M. Combining high hardness and crack resistance in mixed network glasses through high temperature densification. *Physical Review Materials* 2, 063603 (2018).

[2] Kapoor S., Guo X. J., Youngman R. E., Hogue C. L., Mauro J. C., Rzoska S. J., Bockowski M., Jensen L.
R., Smedskjaer M. M. Network Glasses Under Pressure: Permanent Densification in Modifier-Free Al<sub>2</sub>O<sub>3</sub>B<sub>2</sub>O<sub>3</sub>-P<sub>2</sub>O<sub>5</sub>-SiO<sub>2</sub> Glasses. *Physical Review Applied* 7, 054011 (2017).