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Publication date:
2023

Document Version
Publisher's PDF, also known as Version of record

Structural Fingerprint of Crystallization in Mixed-Alkali Bioactive Glasses

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Abstract: Hench-type bioactive glasses such as 45S5 exhibit excellent biological and therapeutic performance, including osteogenesis, angiogenesis, bactericidal activity, and anti-inflammation properties [1, 2]. However, the pronounced devitrification tendency significantly reduces the processing window, which limits their clinical use [3]. In this study, we aim to decipher the underlying structural fingerprint correlated with the crystallization propensity of such glasses. To this end, the atomic-scale arrangements of mixed-alkali bioactive (MAB, 46.1SiO_2-2.6P_2O_5-26.0CaO-(24.2-x)Na_2O-xLi_2O) glasses were determined using high energy synchrotron X-ray diffraction, reverse Monte Carlo simulation, Raman and solid-state nuclear magnetic resonance spectroscopy. The glasses were prepared by two quenching protocols with different cooling rates. The MAB glasses formed through rapid cooling (containerless aerodynamic levitation quenching) show much better stability (higher glass transition T_g and crystallization T_c temperatures) and processability ($\Delta T = T_c$-T_g) compared to the slowly cooled glasses (conventional melt quenching). Moreover, these thermal properties exhibit significant composition dependence with the Li:Na ratio. Overall, T_g shows a nonlinear negative deviation, while ΔT displays a parabolic-like tendency consistent with the mixing entropy. Variations of T_g and ΔT are intricately correlated with the hierarchical-scale network connectivity prompted by rapid cooling and mixed alkali effects, including but not limited to the flexible Si-O-P linkages that were validated in the latest simulations [4]. A physics-based structural fingerprint is then developed, where the contours of topological constraints and local configurational entropy projected on individual network-formers (Si, P) are associated with the barriers of potential nucleation. We show that the enhancement of crystalline resistance is linked to the decrease of possible nucleation sites.

Key words: Bioactive glasses; high-energy synchrotron X-ray diffraction; topological structure

References:

[1] Montazerian M, Zanotto ED, Mauro JC, Model-driven design of bioactive glasses: from molecular dynamics through machine learning, Int Mater Rev, 2019, 65(5): 297-321.

[2] Brauer DS, Bioactive glasses-structure and properties, Angew Chem Int Ed Engl, 2015, 54(14): 4160-4181.

[3] Fu Q, Mauro JC, Rahaman MN, Bioactive Glass Innovations Through Academia-Industry Collaboration, Int J Appl Glass Sci, 2016, 7(2): 139-146.

[4] Bhaskar P, Maurya Y, Kumar R, et al. Cooling rate effects on the structure of 45S5 Bioglass: computational and experimental evidence of Si–P avoidance. Cond Mat Mtrl Sci. arXiv:1906.10111v1.