We have derived models for the structure of Zr-Cu-Al bulk metallic glasses (BMGs) from hybrid Reverse Monte Carlo (HRMC) simulations combining fluctuation electron microscopy (FEM) data and an empirical interatomic potential [1]. The FEM data constrains the nanoscale, medium-range order of the models, and the potential constrains the physical and chemical short-range order. Figure 1 shows the structures formed in a model of Zr$_{50}$Cu$_{45}$Al$_5$ by the inclusion of the FEM data: chains of icosahedral nearest-neighbor clusters and more spherical clusters with crystallographically-allowed four- and six-fold rotational symmetry. A model based only on the potential does not have these structures and does not agree with the FEM data.

Figure 2(a) shows FEM data V(k) for Zr$_{50}$Cu$_{35}$Al$_{15}$, which is a poorer glass former than Zr$_{50}$Cu$_{45}$Al$_5$. It contains peaks at 0.37 Å$^{-1}$ and 0.41 Å$^{-1}$, identified as arising from icosahedral-like and crystal-like medium-range order respectively from HRMC simulations. Compared to Zr$_{50}$Cu$_{45}$Al$_5$, the as-quenched state has stronger crystal-like order and weaker icosahedral-like order. The icosahedral-like order increases with minimal annealing (10 minutes at 0.83$T_g$), consistent with the behavior of Zr$_{50}$Cu$_{45}$Al$_5$, but the crystal-like order is comparatively more stable. Figure 2(b) shows the medium-range correlation length $\Lambda$ derived from variable resolution (VR) FEM experiments [2], in which the probe size is systematically varied from 1.3 to 11 nm. The decay in V(k = 0.37 or 0.41 Å$^{-1}$) as a function of probe size gives $\Lambda$ for the icosahedral- and crystal-like structure types respectively. Minimal annealing significantly increases $\Lambda$ for the icosahedral-like order and decreases it for the crystal-like order.

The correlations between structure and glass-forming ability for the two alloys suggest that icosahedral medium-range order as well as icosahedral short-range order favors glass formation, and that more stable crystal-like order disfavors it. Detailed HRMC modeling of Zr$_{50}$Cu$_{35}$Al$_{15}$ will also be discussed.

References

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Fig. 1: Zr₅₀Cu₄₅Al₅ HRMC model: (a) Crystal-like cluster, with the atoms colored blue to red by their local five-fold symmetry (b) 3D reciprocal space of the crystal-like cluster showing 4- and 6-fold rotational symmetry, (c) edge-on view of an icosahedral chain showing 2-fold symmetry, (d) an icosahedral chain.

Fig. 2: Zr₅₀Cu₃₅Al₁₅ FEM data as a function of annealing at 0.83Tᵣ (673 K). (a) V(k, R = 2 nm) showing peaks associated identified as icosahedral- and crystal-like order. (b) Variable-resolution FEM medium-range order correlation lengths Λ for the two structure types as a function of annealing.