
Kinetics and spatial correlations in phase separated soda-lime-silica glasses

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Abstract

Amorphous phase separation in glasses is a well-known phenomenon and has been the subject of intense investigations since the 70s of the last century. The two mechanisms leading to phase separation are binodal separation in the metastable region of the miscibility gap, and spinodal decomposition in the unstable region. Although their thermodynamical origins are drastically different, which leads to distinct morphologies at the beginning of the separation process, both structures eventually ripen into spherical droplets to reduce the interfacial energy of the system. Thus, experimentally, it has proven difficult to determine whether an observed microstructure was formed by a binodal or spinodal mechanism.

In the present work, we quantified the evolution and kinetics of the microstructures formed in a soda-lime-silica glass with a composition that intersects both the binodal and spinodal curves of the miscibility gap. From the nearest neighbor distances between particles, the spatial correlations in the microstructures were evaluated by an aggregation index, that was originally developed to quantify the relations between tree populations (1), recently tested for glass ceramics (2), and here applied to phase-separated glasses for the first time. The observed nearest neighbor distances deviate from a uniform, random distribution and indicate a more distanced particle spacing. A binodal process should lead to a uniform, random distribution of nuclei, while a spinodal structure forms under wavelike, periodic fluctuations. Based on the calculated aggregation indices, it is suggested that spinodal microstructures keep some of their periodic character with increased particle spacing during spherization and ripening. The aggregation index could therefore serve as an easy way to determine whether an observed microstructure had a binodal or spinodal origin.

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Keywords: Phase separation, microstructure, silica glass

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