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RESEARCH ARTICLE

Effect of Al_2O_3 addition on the thermal expansion of sodium alkaline-earth silicate glasses: A molecular dynamics study

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Abstract

Coefficient of thermal expansion (CTE) is an important property to consider when utilizing oxide glasses in thermal treatment processes to avoid thermal damage at the interfaces of the glasses with heterogeneous materials. It is thus important to know the effect of additives on CTE for designing glasses. The use of alumina efficiently improves chemical and mechanical durability of oxide glasses while maintaining the functionality and productivity; however, alumina-doping often induces nonlinear variation of CTE. In this work, we therefore tried to investigate the relationship between CTE and the microstructure of sodium alkaline-earth aluminosilicate glasses using classical molecular dynamics (MD) simulations. To accurately model the glasses, we extended a force-matching potential by optimizing the parameter sets for Ca–O, Mg–O, and Na–O pair interactions using Bayesian optimization. The MD simulations reproduced the nonlinear variation of CTE as a function of alumina content, and detailed structural analyses identified inhomogeneous expansion in the glasses. It was found that the nonuniform CTE change at the nanoscale was related to the formation of an alumina-rich region, in which more fivefold-coordinated aluminum exist, when alumina content exceeded Na_2O content. Accordingly, the microstructural change by alumina-doping was identified as the origin of the nonlinear variation in the CTE of the glasses.

KEYWORDS
aluminosilicates, molecular dynamics, thermal expansion

1 | INTRODUCTION

Oxide glasses are used for a variety of applications because their mechanical, optical, chemical, thermal, and electrical properties can be widely tailored by varying the ratio of their components, such as network formers and modifiers. Indeed, more than several hundreds thousands of glasses have already been investigated and their properties are stored in the international glass database system, INTERGLAD.[1] Among these properties, the coefficient of thermal expansion (CTE) is an important factor when applying glasses in a thermal treatment process.

Glassy materials often come in contact with different types of materials, such as silicon, copper, and polyimide,[2,3] for instance, in electric devices (including semiconductors) and in the packaging of computer systems. At these interfaces, CTE mismatch should be minimized to avoid interfacial damages as the tensile stress caused by such mismatch initiates and accelerates crack propagation in brittle materials.[4] To precisely fabricate thin-film transistor (TFT) arrays, glass substrates with

