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Structural origin of intrinsic ductility in binary aluminosilicate glasses

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ABSTRACT

We evaluate the fracture mechanism in the binary aluminosilicate glasses using molecular dynamics simulations. The simulations using two independent force fields reveal that increasing the alumina content promotes shear and suppresses fracture, thereby increasing the intrinsic ductility, in agreement with experimental observations. In indentation simulations, it is directly demonstrated that the deformation mechanism shifts from densification to shear flow with the increase in alumina content. The origin of this intrinsic ductility is that the Al atoms are more amenable to plastic flow and can reduce the creation of lower coordinated weak spots during deformation.

1. Introduction

Apart from surface strengthening techniques, intrinsic damage resistance by composition design is one important method to improve the performance of oxide glasses against contact cracking. Currently, the type of intrinsic damage resistance that has been widely observed in experiment and employed in commercial applications is predominantly correlated with the increase in compaction during indentation [1,2]. Compaction is achieved by increasing the free volume, which usually decreases the Young's modulus, hardness, and fracture toughness.

Another source of intrinsic ductility is through extensive shear [3], as demonstrated through molecular dynamics (MD) simulations. It is observed in three distinct families of non-crystalline solids that reducing the angular constraint in bonding (covalency) and increasing the bond strength is the key for promoting shear and increasing the intrinsic ductility [3], which is in agreement with the bond constraint theory [4,5]. Such trend is also supported by the experimental observation that compares oxide glasses with metallic glasses [6,7]. Even in crystalline materials, angular constraints were proposed as one of the controlling factors for ductility [8,9].

Intrinsic ductility via shear is usually accompanied by decreasing free volume and increasing modulus and fracture toughness considering a wide range of amorphous solids including oxide glasses and metallic glasses [3,7,10]. Therefore, one potential advantage of intrinsic ductility via shear is the simultaneous attainment of high hardness and high toughness. Most recently, high modulus, high hardness, and high crack resistance have been achieved in the glass $40SiO_2 60Al_2O_3$ made experimentally by aerodynamic levitation [11]. In this study, we use MD simulations to directly examine the fracture mechanism of aluminosilicate glasses and find that increasing alumina content increases the intrinsic ductility via shear using two independent force fields. The origin of the intrinsic ductility is found to be that the Al atoms are more amenable to plastic flow and can reduce the creation of low coordinated weak spots during deformation.

2. Methods

We study the same series of binary glasses in experiment [11] with the mol% composition of $(Al_2O_3)_x(SiO_2)_{100-x}$, x = 30, 40, 45, 50, 55, 60. For completeness, we also include pure SiO_2 and Al_2O_3. These compositions are referred to as Al0, Al30, Al40, Al45, Al50, Al55, Al60 and Al100 for short. While glassy Al_2O_3 is difficult to generate in experiment, it is possible in MD simulation due to the accessible fast quenching rate. The quenching process starts with a slab sample made of approximately 20,000 atoms with random coordinates equilibrated at 4000 K for 1 ns. Then the high temperature melt is quenched to 300 K continuously over a period of 8 ns in an NPT (constant number of atoms, constant pressure, and constant temperature) ensemble under ambient pressure, or in an NVT (constant number of atoms, constant volume, and constant temperature) ensemble [12] with the initial density as an input. Finally, the glass is relaxed at 300 K under atmospheric pressure for 1 ns. Periodic boundary conditions are applied in all directions. We employ two widely used force fields developed by Pedone et al. [13], and by Teter et al. [14] In the Pedone force field, we observed crystallization in Al_2O_3 using a quenching rate of 0.5 K/ps. Therefore, for Al_2O_3, we shortened the quenching time to 0.8 ns for both force fields. In the NVT quenching process, the initial densities of the melts are set to be the experimental values [11]. Since the Pedone force field is known to overestimate the density, if the exact experimental values are imposed in NVT quenching, large holes can be resulted in the quenched samples. Therefore in the Pedone force field, the initial density is set to be 5% higher than the experimental values [11]. The final density is shown in Fig. 2.

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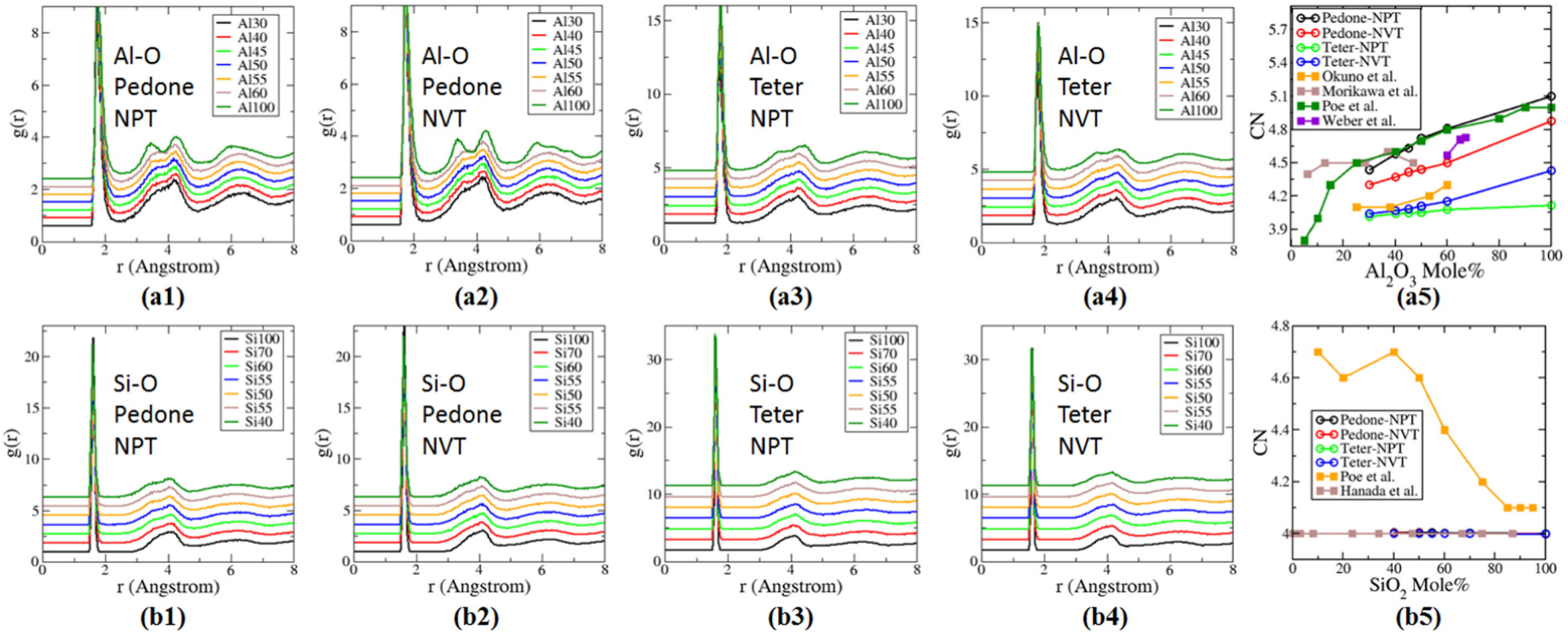

Fig. 1. Radial distribution functions (RDF) for Al-O pair in (a1) to (a4) and Si-O pair in (b1) to (b4) and coordination number as a function of composition for Al in (a5) and for Si in (b5), using different force fields and quenching ensembles. In (a5) and (b5), experimental results by Okuno et al. [25], Morikawa et al. [26], Poe et al. [27], Hanada et al. [28], and Weber et al. [29] are also shown for comparison. To compute the CN, the cutoff distance is set to be 0.25 nm for Al and 0.2 nm for Si.