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Study on crystallization process of SiO_2 based SiO_2-Li_2O nano-wire glass ceramic: A molecular dynamics simulation based on SCC-DFTB calculations

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ABSTRACT

The aim of this study was to investigate the crystallization behavior of nano-wire SiO_2-Li_2O glass ceramic (GC) during the slow cooling process by using density functional theory (DFT). For this purpose, the extended tight-binding with self-consistent charge (SCC-DFTB) was used to investigate the geometric optimization and molecular dynamics (MD) process for model system. The structural development was analysed by radial distribution function (RDF) at determined temperatures. The results show that the system tends to crystallization at lower temperatures and transforms from liquid phase to crystal phase with a slow cooling rate.

1. Introduction

Today, glass-ceramics (GC) are applied in many advanced technology areas due to their superior corrosion resistance and toughness properties compared to traditional glasses and metals [1, 2]. Some of them include biomedical applications, superconducting materials, materials with high dielectric constant and applications to the field of electronics [3]. In addition, they are produced in many lithium-based materials and have great advantages in terms of their physical and mechanical properties [4]. Among these, lithium dioxide (Li_2O) is mild compared to other compounds, has a low viscous temperature, high conductivity, good formability and high optical properties [5-7].

Being used as materials in different technological areas, lithium-silicate glass-ceramics are well known for their good mechanical properties [7]. Lithium silicates are particularly interesting materials due to their tritium-forming and similar properties and their compatibility with building materials [4-7]. Lithium silicate glass-ceramics have superior mechanical and optical properties when used in multi-component systems. Due to these properties, they have many studies in theory and many applications in practice. Lithium silicate GCs are mostly used in ceramic-metal sealing joints and dental applications [8]. However, most of their characteristic behaviours are still unexplained. In particular, there are deficiencies in the nanostructure related to processes based on a number of experimental measurements, such as crystallization mechanism and kinetics. The use of computational methods to eliminate such deficiencies is increasing day by day. Especially, molecular dynamics (MD) simulations based on DFT and classical interactions are useful way to investigate many physical properties of molecules as a function of time and temperature [9-11]. This method used to calculate, for example, the total energy during physical and chemical processes can be determine because it provides thermal treatment for a period of time [12].

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2. Material and Method

2.1. DFT and MD details

The extended tight-binding with self-consistent charge (SCC-DFTB) approach based on quantum mechanical methodology is a DFT-based method regarding to the electron density fluctuations [13, 14]. The set of parameters QUASINANO2013.1 [15] available in the Amsterdam Density Functional (ADF) library is utilized for single point calculations of molecules [16-18]. More details about the SCC-DFTB method can be found in Refs. [13, 14]. The MD simulations are performed using the ADF software (version 2020.104) [18]. In our study, SCC-DFTB is used for MD simulations. The Berendsen [19] thermostat and barostat are adopted to control temperature and pressure of the system and time-step is set as 1.25 fs during the simulation. Firstly, the system is equilibrated for 12.5 ps at 2500 K and secondly, the temperature of system decreases from 2500 K to 300 K within 625 ps with a cooling rate of 3.52×10^{12} K/s. The main parameters and atomic configurations are recorded at every 12.5 ps to investigate the structural and thermal changes.

3. Results and Discussions

We have built a SiO_2-Li_2O glass ceramic structure to investigate the geometric optimization and MD process using SCC-DFTB. For this purpose, SiO_2 crystal lattice has been set up according to Fd3m space group by using builder option in SCIGRESS software [20]. Li_2O atoms are distributed to upper and top side of SiO_2 in the simulation box which contains 120 atoms. As a start, the bulk GC system is transformed to a nanowire structure with 39 Å lattice parameter at one dimension. Then, this nano-wire GC is optimized to determine most stable structure. The nanowire SiO_2-Li_2O GC model system is shown in Fig. 1.

Fig. 2 shows the geometric optimization process and the final structure of SiO_2-Li_2O GC at the end of optimization with respect to the frame number or optimization iteration steps. The energy of the GC system has the maximum value at the beginning of optimization. The energy reaches its minimum value at the final optimization because the system has high stability compared with the beginning structure.

Fig. 3 and Fig. 4 show the variation of some physical parameters with frame number or MD steps during the MD process for SiO_2-Li_2O GC. As seen from figure that the temperature of system is linearly decrease with MD time after relaxation. The total energy of system decreases with increasing MD time during slow cooling process. The bond lengths between some selected atoms exhibit the fluctuations at liquid phase due to increase of atomic mobility. These fluctuations dramatically decrease after 400 ps because the system transforms a stable crystal phase.

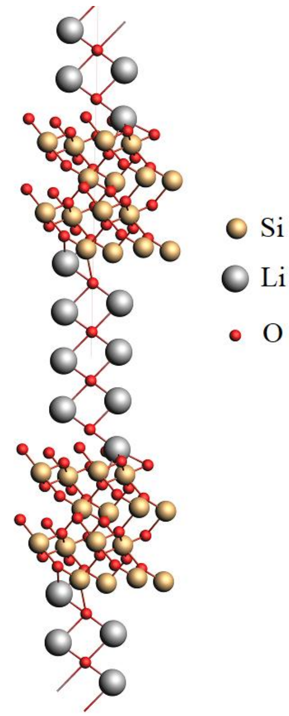

Figure 1. The nanowire SiO_2-Li_2O GC model (Gray atoms, yellow atoms and red atoms represent Li, Si and O, respectively).

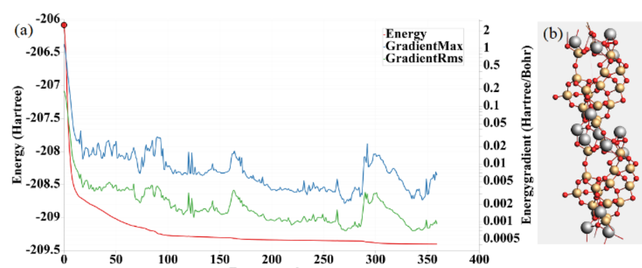

Figure 2. Optimization process of model system (a) variation of energy with optimization iteration (b) final structure of SiO_2-Li_2O nano-wire GC at the end of optimization.

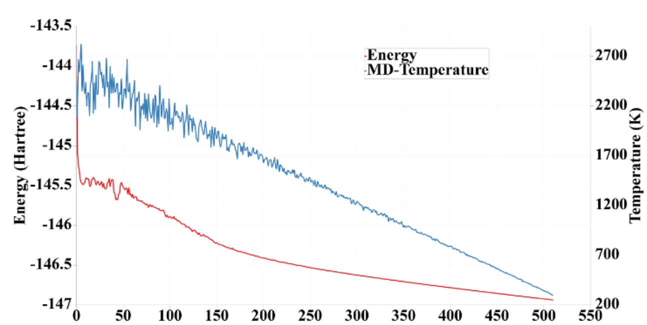

Figure 3. The variation of total energy and temperature of system during MD process.

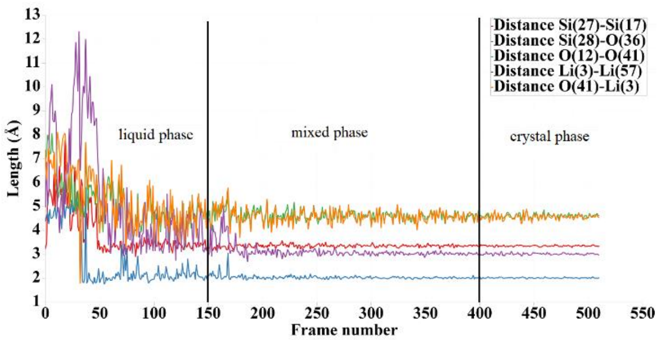

Figure 4. Variation of distance between some selected atoms during MD process.

Fig. 5 shows a snapshot from MD simulations corresponding to nanowire model system at beginning structure, the structure at 2500 K and final structure at 300 K, respectively. We can say from figure that the atomic distribution in the system at 2500 K more disordered than beginning structure due to increase of atomic mobility as seen Fig. 4. Hence, the volume of system is seen broadly at 2500 K. But, the atomic mobility increases with the decreasing of temperature. It can be said that the system has a more ordered structure. These results can be support with radial distribution function (RDF) analysis.

The structural characteristics of a system can be analysed by radial distribution function (RDF, g(r)) which provides the probability of finding neighbor atoms at a distance of r from an atom [21]. Fig. 6 shows the partial RDFs or g(r) of bond pairs (Si-Si, Si-O, O-O and Si-Li) for system at 2500 K, respectively. The first peak in the RDFs represents the short-range order, and the second peak becomes more broadened compared to the first peak of the RDF curve because the periodic crystal order is not dominant at the liquid phase. Fig. 7 shows the partial RDFs of bond pairs (Si-Si, Si-O, O-O and Si-Li) for system at 300 K, respectively. From figure, the much higher order peaks at long atomic distances in the RDFs represent the long-range order because of formation of stable crystal structure. We observe that slow cooling rate causes in better-defined crystal peaks with higher values of the first and second maximum of gSi-O(r).

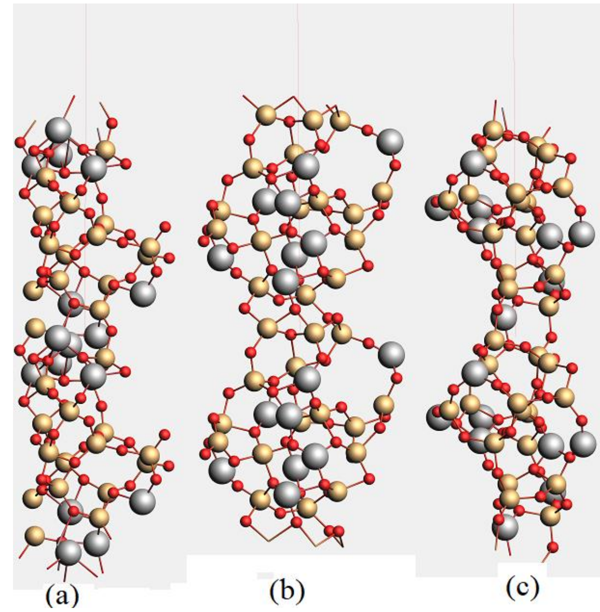

Figure 5. A snapshot corresponding to nanowire model (a) beginning structure b) structure at 2500 K c) final structure at 300 K.

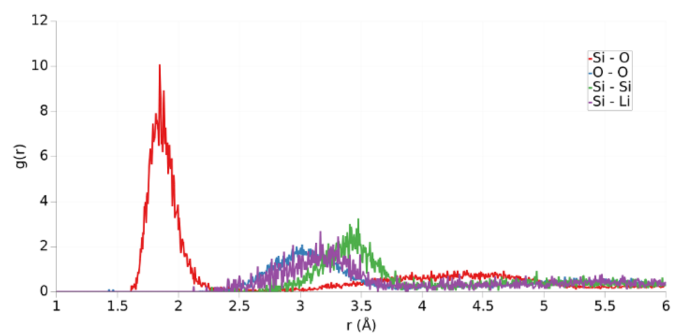

Figure 6. The partial RDF curves at 2500 K.

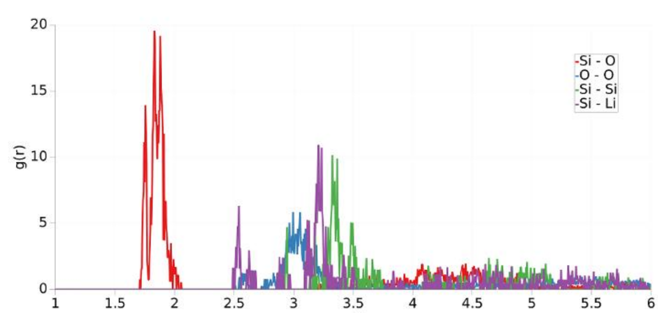

Figure 7. The partial RDF curves at 300 K.

4. Conclusions

In summary, we performed SCC-DFTB based on density functional theory calculations on the crystallization process of nano-wire SiO_2-Li_2O glass ceramic with molecular dynamics simulation method. The total energy was minimum value at the final of optimization. With the DFTB calculations, MD simulations for the slow cooling of system have been conducted. The atomic mobility's decreased during solidification and the position of the RDFs peaks emerged at longer atomic distances because the crystallization occurs at lower temperatures with a slow cooling rate.

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