Usage Report for Fiscal Year 2018

Project Title:

Structure and properties of oxide melts

Name: ○Nguyen Van Hong(1), Toshiaki Iitaka(1)

Laboratory at RIKEN:
(1) Computational Engineering Applications Unit, RIKEN

1. Background and purpose of the project, relationship of the project with other projects

The multicomponent oxide glass system based on B_2O_3-SiO_2 is a group of materials with many good properties such as corrosion resistance, good mechanical properties, low thermal expansion coefficient, low melting temperature.... So, this materials group has been widely applied in many fields such as: microelectronics, medicine (bio-material), high-technology materials, and immobilization of toxic metals. The glasses based on B_2O_3-SiO_2 are interesting from both fundamental and application points of view since their flexible network structure, their structure and properties can be easily adjusted by changing the oxide contents. With flexible network structure and low melting temperature, B_2O_3-SiO_2-based glasses have been widely applied in immobilization of toxic metals (toxic metals in industrial-, hospital-, and nuclear-waste). The network structure of B_2O_3-SiO_2-based glasses allows it to incorporate and immobilize many different elements (a wide range of elements) including: alkali and alkaline-earth metals, heavy metals, transition metals, rare earth elements, radioactive elements. Adding Na_2O to borosilicate will lead to increasing the $[NBO]^-$ (Non-Bridging Oxygen) concentration, reducing the melting temperature and viscosity. Adding Al_2O_3 to sodium-borosilicate will lead to increasing the BO (Bridging Oxygen) concentration and generating $[AlO_4]^-$, $[BO_4]^-$ units. As BO concentration increases, mechanical properties of the glass system will be improved. The increase of $[NBO]^-$, $[AlO_4]^-$ and $[BO_4]^-$ concentration will improve the immobilization capacity of toxic metal cations via mechanism of the charge balance. The concentration of $[NBO]^-$, $[BO_4]^-$ and $[AlO_4]^-$ in B_2O_3-SiO_2-based glasses can also be adjusted by changing the pressure or cooling rate. Thus, investigating the influence of chemical composition and technological conditions on the structure and properties of B_2O_3-SiO_2-based glasses will help optimized structure to improve desirable properties, and contribute to improving the efficiency of immobilization of toxic metal cations in the glass network structure at the atomic level. Information on these issues is difficult to trace directly by experiment and ones usually apply **molecular dynamics simulations.** Therefore, present project is devoted to giving some new insight into the above mentioned problems.

2. Specific usage status of the system and calculation method

Molecular dynamics simulation, Topology analysis methods, Recognition and Visualization methods are applied to clarify the structure and dynamics of MO_xSiO_2, MO_x-B_2O_3-SiO_2 oxides

3. Results

The structure and dynamics of SiO_2 and Al_2O_3-SiO_2 have been clarified. The investigation results have been published on 03 papers

4. Conclusion

The calculation also reveals that the diffusion in SiO_2 liquid is realized via bond-breaking mechanism. The reactions are non-uniformly distributed in the space and the liquid exhibits DH the degree of which reduces with increasing temperature. Furthermore, the simulation shows a strong correlation between mobility of atoms and init-bond function. We find that DH is accompanied

with structure heterogeneity and cooperative movement of atoms via like-molecules Si_xO_y.

The structure of silica glass consists of three phases: SiO_4-, SiO_5- and SiO_6-phases. At pressure beyond 20 GPa, most of basic structural units are SiO_6 forming SiO_6-cluster (SiO_6-phases) and they tend to arrange orderly, forming stishovite crystalline. The silica glass will tend to transform to crystalline under high pressure. This is supported by the fact that the materials in the inner-core of the earth (where the pressure is very high) have the crystalline structure.

The structure of $3Al_2O_3$-$2SiO_2$ system mainly comprises a mixture of basic structural units TOx(x = 3-7; T is Si, Al) forming a continuous random network in three-dimensional space. The short range order structure is not very sensitive to compression while the The intermediate range order structure is signicantly changed under compression, especially in the pressure range 6-60 GPa.

Most of the adjacent SiOx units are linked together by the corner-sharing bonds (CSBs) and the number of CSBs increases with pressure. The adjacent SiO_x units do not exist the edge-sharing bonds (ESBs) and face-sharing bonds (FSBs) at low pressure. However, at high pressure, the number of ESB and FSB exists but is not signi_cant. The links between two adjacent AlOx and between AlO_x and SiO_x units have all three bond types: CSBs, ESBs and FSBs, with CSBs and ESBs are dominant. At all pressure, we always have $N_{corner} > N_{edge} > N_{face}$ and the number of all types of links increases under compression.

Under compression, the Al-O bonds are broken lead to incorporation of Al atoms into Si-O subnet through both NBO and BO forming -Al-O-Si-network (Al-O-Si, Si-O-Al_2, Si_2-O-Al, Si-O-Al_3, Si_2-O-Al_2 and Si_3-O-Al). The degree of polymerization (DOP) of the silica network and alumina network increase with pressure and the DOP of SiOx-network is lower than that of AlOx-network under compression.

The TO_x-clusters tend to incorporate each other to form larger networks instead of isolated subnets. The AlO_x-clusters are surrounded by the SiO_x-clusters and coversely the SiO_x-clusters are surrounded by the AlOx-clusters.This result reveals the compositional heterogeneity in $3Al2O_3$-$2SiO_2$ system.

The SiO4 units tend to link each other forming SiO_4 cluster. Similarly, SiO_5 and SiO_6 units tend to form SiO5- and SiO6-clusters. For AlOx units, AlO_3, AlO_4, AlO_5, AlO_6 and AlO_7 units tend to link to each other forming AlO_3-,AlO_4-, AlO_5-, AlO_6- and AlO_7-clusters, respectively. This reveals the structural heterogeneities in $3Al_2O_3$ -$2SiO_2$ system.

5. Schedule and prospect for the future

In next time, we will focus on the structural investigation of multicomponent oxide glass (MO_x-B_2O_3-SiO_2). This is a group of materials with many good properties such as corrosion resistance, good mechanical properties, low thermal expansion coefficient, low melting temperature.... So, this materials group has been widely applied in many fields such as: microelectronics, medicine (bio-material), high-technology materials, and immobilization of toxic metals. The Mox-B_2O_3-SiO_2 systems are interesting from both fundamental and application points of view since their flexible network structure, their structure and properties can be easily adjusted by changing the oxide contents. With flexible network structure and low melting temperature, B_2O_3-SiO_2-based glasses have been widely applied in immobilization of toxic metals (toxic metals in industrial-, hospital-, and nuclear-waste)

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Fiscal Year 2018 List of Publications Resulting from the Use of the supercomputer

[Paper accepted by a journal]

[1] Nguyen Thu Nhan, Giap Thi Thuy Trang, Toshiaki Iitaka and Nguyen Van Hong, Crystallization of amorphous silica under compression, Canadian Journal of Physics (accepted Feb, 2019), DOI: 10.1139/cjp-2018-0432.

[2] P.K. Hung, L.T. Vinh, N.V. Hong, N.T. Thu Ha, Toshiaki Iitaka, Two-domain structure and dynamics heterogeneity in a liquid SiO2, Journal of Non-Crystalline Solids 484 (2018) 124–131.

[3] Mai Thi Lan, Toshiaki Iitaka and Nguyen Van Hong, Simulation of structural characteristics of Mullite melt at high pressure, Journal of Modern Physics B Vol. 32, No. 24, 1850271 (2018)