Usage Report for Fiscal Year 2017

Project Title: Materials properties under extreme conditions: Understanding planets in depth

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

MO_xSiO_2 oxides (here M is Pb, Al, Mg, Ca or Na) have received a great attention from materials science communities for long times, because they have certain specific properties such as high mechanical strength, special optical absorptions, corrosion and abrasion resistance... These oxides are also known as the glass-forming material due to that their structure is built by basic units which randomly distribute in the space and link to each other by common oxygen (see Fig.1). Further, the majority of basic units are SiO_4 tetrahedra and a number of other types like SiO_m and MO_n also present in the system. The structure as well as many properties of MO_xSiO_2 oxides strongly depends on concentrations of MO_x. For example, the absorption of x-rays of $PbOSiO_2$ significantly increased when the concentration of PbO is greater than 30 mol%. In the case of $Al_2O_3SiO_2$ the material has high melting temperature (2000K) and its mechanical property is significantly improved in comparison with the pure SiO_2. The physics-chemical properties of $CaSiO_3$ demonstrated a good likeness with human bone, so it is widely used in the treatment of bone-related diseases. Thus, detail knowledge about the network structure when MO_x is added into the oxide system is very important to understanding the glass-forming oxide. One among interesting directions of recent studies is to clarify how the units SiO_4 spatially distribute when the concentration MO_x varies, and what role other type units (defect-units) plays. These issues are very difficult to trace directly by experiment and ones usually apply the simulation at the atomic level. Recently, the spatial distribution of defect-units as well as their equilibrated concentration has been studied intensively; however, the opinions about this problem are still controversial. Therefore, more studies need to treat mention issues. Especially, the coexistence of microscopic regions with different network structure is so far understood and represents a challenge for physicists. The present project is devoted to give some new insight into the above-mentioned problem. In particular, we focus on the thermodynamics of defect-units and the technique to detect them. Besides the techniques such as the topology and cluster analysis, simplexes approach... the recognition and visualization methods are developed in order to identify different types of network structure.

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 oxides

3. Results

The structure and dynamics of SiO_2, GeO_2, $MgSiO_3$, $CaSiO_3$ have been clarified. The investigation results have been published on 04 papers

4. Conclusion

The Ge-Ge pairs with edge-sharing bonds tend to cluster with each other, forming edge-sharing clusters. Similarly, the Ge-Ge ion pairs with face-sharing bonds also tend to form face-sharing clusters. The size of edge- and face sharing clusters is strongly dependent on pressure. The Ge-Ge bond length in the edge- and face-sharing bonds is much shorter than that in corner-sharing bonds. This is the origin of the first peak splitting of the Ge-Ge pair.

The calculation also reveals that the diffusion in liquid silica 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 SixOy
An addition of CaO (or MgO) into silica glass makes the Si-O network broken and a lot of NBO in the network is formed. The Ca^{2+} ions tend to incorporate in Si-O network via NBO. This mechanism can be applied in processing hazardous waste by immobilization of heavy metal as radioactive elements in glass form. However, NBOs make the Si -O network broken. Under compression, the Si-O network will be more polymerized and it creates the SiOx polyhedra with a negative charge. These negative charged SiOx polyhedral will attract Ca^{2+}. Replacing Si^{4+} in SiO_4 by an element with valence of 3+, we will have $(AlO4)^-$ polyhedra in the network that will attract Ca^{2+}, Mg^{+2} (in general it is M^+, here M^+ is metal ions). $CaSiO_3$, $MgSiO_3$ glass also has Si-rich regions and Ca-rich (Mg-rich) regions that reveals the compositional heterogeneity. However, the Si-rich region has the form of a chain, not cluster, meanwhile the Ca-rich (Mg-rich) region has the form of clusters.

5. Schedule and prospect for the future

In next time, we will focus on the structural investigation of multicomponent oxide glass system based on 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 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)

Fiscal Year 2017 List of Publications Resulting from the Use of the supercomputer

[Publication]

[1] Mai Thi Lan, Tran Thuy Duong, **Toshiaki Iitaka** and **Nguyen Van Hong**, Computer simulation of CaSiO3 glass under compression: correlation between Si–Si pair radial distribution function and intermediate range order structure, Mater. Res. Express 4 (2017) 065201.
http://dx.doi.org/10.1080/00319104.2016.1166362

[2] Luyen Thi San, **Nguyen Van Hong**, **Toshiaki Iitaka**, and Pham Khac Hung, Structural organization, micro-phase separation and polyamorphism of liquid MgSiO3 under compression, Eur. Phys. J. B (2016) 89: 73.
http://dx.doi.org/10.1140/epjb/e2016-60740-4

[3] Tran Thuy Duong, **Toshiaki Iitaka**, Pham Khac Hung, **Nguyen Van Hong**, The first peak splitting of the Ge-Ge pair RDF in the correlation to network structure of GeO2 under compression, Journal of Non-Crystalline Solids 459 (2017) 103–110.
https://doi.org/10.1016/j.jnoncrysol.2017.01.003

[4] 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.
https://doi.org/10.1016/j.jnoncrysol.2018.01.023