JASMAC Conference of the Japan Society of Microgravity Application

OS2-11

融液の熱物性と原子配列をベースとした機能性高充填密度ガラスの設計指針の確立

Materials design of functional densely packed glasses using thermophysical properties and atomic arrangements of melts

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1. Introduction

Oxide glasses are generally considered to be materials in which the so-called network former oxides, such as SiO_2, B_2O_3, and P_2O_5, serve as the main components, forming corner-sharing three-dimensional random network structures (Figure1(a)). However, we have recently succeeded in synthesizing a variety of highly densely packed glasses without corner-shared networks by using a levitation technique[1,2] (Figure 1(b)). These glasses exhibit fascinating physical properties compared to conventional glasses, including a high refractive index with low wavelength dispersion, strong luminescence, high elastic modulus, high cracking resistance, high dielectric constant, and a large magneto-optical effect[3–5]. It is suggested that these remarkable functionalities arise from the high packing density; however, the structure-property relationship remains unclear. Additionally, the structural and thermophysical origin of glass formation in the absence of network formation, despite high packing density, are still not well understood. Given that glasses are obtained by cooling melts, it is crucial not only to clarify the structural features responsible for functionality of these glasses but also to understand the glass formation process by which such characteristic atomic arrangements are formed. To achieve this, it is first necessary to investigate the correlation between the structure of melts and supercooled liquids and their thermophysical properties such as viscosity.

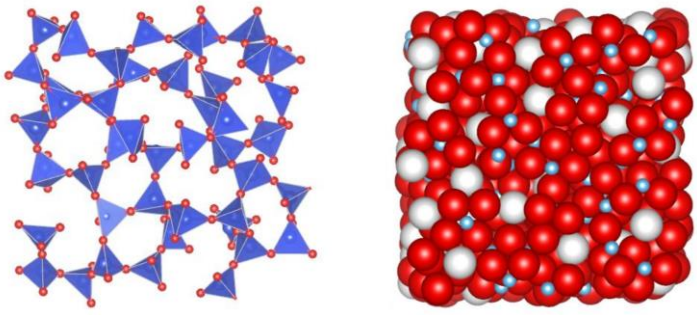

Figure 1 Atomic arrangements of (a) conventional network glass and (b) densely packed glass.

2. Purpose of the project

This research project builds on the FY2022 project titled "Physical property measurements of functional densely packed oxide glass melts by using ELF for understanding the atomistic origin of the functionality". The current project's objective is to develop guidelines of materials design of functional densely packed glasses by examining the thermophysical properties and atomic arrangements of melts. This represents a more in-depth investigation compared to the previous study, which primarily aimed to elucidate mechanism behind the functionalities of these glasses. In the FY2022 project, we focused on understanding the mechanisms underlying the functionalities of densely packed glasses, shown in Table 1. Thermophysical properties (density and viscosity) and diffraction data of the glass melts will be obtained. Building on these results, this study will explore a wider composition range to examine the composition dependence of these functionalities and their glass forming abilities. By adjusting the chemical compositions, specifically by altering the size of cations in modifier oxides and the concentration of modifier oxides, we aim to identify characteristic atomic arrangements that contribute to functionalities and structural factors that enhance glass formation.

Table 1. Chemical compositions of samples for FY2022 and FY2023 projects.

	Refractive index	**Thermal expansion**	**Transparency**
FY2022	$50La_2O_3$–$50B_2O_3$	$25La_2O_3$–$75MoO_3$	$30La_2O_3$–$70SiO_2$
FY2023	$50Y_2O_3$–$50B_2O_3$	$25Er_2O_3$–$75MoO_3$	$30Y_2O_3$–$70SiO_2$
	$30La_2O_3$–$70B_2O_3$	$20La_2O_3$–$80WO_3$	

To evaluate glass formation and calculate the Fragility Index—a parameter that quantifies glass-forming ability of melts—we will measure the temperature dependence of density and viscosity in space. The use of the electrostatic levitation furnace (ELF) on the ISS Kibo is crucial for accurately measuring the density and viscosity across a wide temperature range, from above the melting point to significantly supercooled temperatures[6]. On the ground, Raman scattering spectroscopy, along with X-ray and neutron diffraction experiments, will be conducted to analyze the structure of the melts. Structural models that reproduce the experimental data will be prepared using molecular dynamics simulations. The reliability of these models will be significantly enhanced by incorporating the densities obtained from space experiments. The structural models will be analyzed using various geometrical methods to extract characteristic atomic arrangements responsible for the functionality[7–10]. Combining thermophysical data with structural information of these unconventional oxide glass melts, we aim to uncover important insights into the atomistic origin of functionality and the glass formation process.

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