Structure and Thermophysical Properties of Molten BaGe by Using Electrostatic Levitation Technique

M. Watanabe[C,S], A. Ishikura and A. Mizuno
Department of Physics, Gakushuin University, Tokyo, Japan
masahito.watanabe@gakushuin.ac.jp

T. Masaki and T. Ishikawa
Institute of Space and Astronautical Science, Japan Aerospace Exploration Agency, Tsukuba, Japan

J. Kohara
Japan Synchrotron Radiation Research Institute, Sayo-gun, Hyogo, Japan

BaGe alloys with two compositions (Ba_6Ge_{25} and Ba_8Ge_{43}) around their eutectic point form open framework structures called clathrate structure. The semiconductor materials with the clathrate structure have been interested in the new thermoelectric materials as the concept of "phonon-glass and electron-crystal" (PGEC) proposed by Slack [1], in which PGEC materials should possess the low thermal conductivity of a glass, but maintain the high electronic mobilities of crystalline semiconductors. The PGEC behavior could be seen in clathrate structure with heavy cations "rattling" inside oversized lattice cavities, which could create ideal conditions for extensive thermal phonon scattering that leads to very low thermal conductivity. For Ba_8Ge_{43} clathrate, Ge forms open tetrahedral networks having pentagonal dodecahedra and tetrakaidecahedra. Two different cages are condensed in three dimensions by face sharing. On the other hand, for Ba_6Ge_{25}, distorted pentagonal dodecahedra are the only forming blocks. These dodecahedra are helically condensed by sharing three pentagonal faces and are interconnected through external bonds. Ba atoms are found in the center of these polyhedras. However, the formation mechanism of these clathrate structures has not been clarified due to any information of their liquid state structure and properties. This is because molten BaGe is highly reactive to several elements. Therefore, in order to clarify the formation mechanism of the clathrate structure, thermophysical properties of molten BaGe around eutectic composition were measured by using the electrostatic levitation (ESL) technique [2] and also the structure of them was observed by using the high-energy X-ray (113.4keV) diffraction method combined with the ESL in the BL04B2 beamline of SPring-8 in Japan. The ESL technique is most suitable to studying of molten BaGe properties, since the ESL technique is containerless method. We succeeded using ESL technique to measure the density, surface tension and viscosity of molten BaGe with various compositions. We found from the thermophysical properties and the structure analysis that the short range order based on the clathrate structure would exist even in the liquid state at the clathrate forming compositions.

[1] G.A. Slack, in *CRC Handbook of Thermoelectrics*, ed. by D. M. Rowe (CRC Press, 1995), p. 407.
[2] T.Ishikawa, P.-F.Paradis, T.Itami and S.Yoda, *Meas. Sci. Technol*, **16**, 443 (2005).