Undercooling Limits and Thermophysical Properties in Glass Forming Alloys

Dr. Won-Kyu Rhim, Principal Investigator
Jet Propulsion Laboratory
California Institute of Technology
Mail Stop 183-401
4800 Oak Grove Drive
Pasadena, CA 91109
Phone: (818) 354-2925
Fax: (818) 393-5039
E-mail: won-kyu.rhim@jpl.nasa.gov

Co-I's: Dr. Kenichi Ohsaka, JPL; Phone: (818) 354-3111
Prof. R. Erik Spjut , Harvey Mudd College; Phone: (909) 607-3890

Objectives of the Research

The primary objective of this program is to produce deeply undercooled metallic liquids and to identify factors that limit the undercooling and the glass formation. The main research objectives are:

(i) Investigation of undercooling limits in glass-forming alloys and identify those factors that affect undercooling.

(ii) Thermophysical property measurements and investigation of the validity of the classical nucleation theory and other existing theories in the extreme undercooled states.

(iii) To investigate the limits of electrostatic levitation technology in the ground base, and to identify thermophysical parameters that might require reduced-g environment.

Relation to the Gravitational Field

When experiments are conducted in 1-g environment, all gravity-caused effects (such as the convective flows, the sedimentation, and the buoyancy) still remain in the levitated melts. In addition, large forces that are needed to levitate the sample against the gravity can cause flows in the melts. The High Temperature Electrostatic Levitator (HTESL) is a relatively recent development. Its limitation in 1-g condition has not been fully established. Although considerably less flows are expected the HTESL, very little is known about the flows induced by the electrostatic forces. The most seriously affected properties might be those transport properties such as the atomic diffusion, the viscosity, and the thermal conductivity. In this ground-based program, we will identify the thermophysical properties and their temperature ranges which require reduced-g condition for their measurements.

Progress of the Research

Thermophysical properties of Ni-Zr alloys (NiZr, $NiZr_2$, $Ni_{36}Zr_{64}$ and $Ni_{24}Zr_{76}$) have been extensively measured in order to investigate the glass formability of these alloys. Properties measured were the specific volume, the surface tension, the viscosity, and the ratio between the specific heat to the hemispherical total emissivity. The specific volume and viscosity measurements of the Ni-Zr liquid alloys show that the associated species in the form of NiZr and $NiZr_2$ exist in the liquid alloys. The presence of the NiZr species was previously known from the

mixing enthalpy study. On the other hand, the $NiZr_2$ species are identified for the first time in the present study. The specific volume data show that the $NiZr_2$ species are densely packed. As a result, the $NiZr_2$ liquid alloy experiences the deviation from the thermodynamic equilibrium specific volume above the melting point under a moderate cooling rate. The $NiZr_2$ liquid alloy shows the relatively small undercooling level which is attributed to the small interfacial energy between the liquid and the solid because of their similar local structures. On the other hand, the NiZr species are more openly packed according to the specific volume data; however, they are still responsible for the high viscosity. The viscosity data show that the NiZr and $NiZr_2$ species are stable and exist at temperatures well above their melting points. The high glass formability of the Ni-Zr alloys can be attributed to the presence of the associated species which have been known as a facilitator for the glass formation. The presence of the associated species may not be advantageous for suppressing the formation of the solid nuclei but they greatly reduce the solid phase growth rate, which give a chance for the glass to form and grow once the liquid reaches the temperature where the atomic relaxation time exceeds the experimental time scale.

The JPL High Temperature Electrostatic Levitator (HTESL) was upgraded. The spectral emissivity measurement capability was added so that the true temperature can be determined. A 100 W YAG laser was installed so that the sample temperature can be measured without blinding the pyrometer during the heat-up cycle. This high power laser will be able to heat the sample higher than 2000 °C.

A new non-contact technique was developed and implemented in the HTESL. Using this technique, a levitated conducting sample can be rotated in a controlled manner. Such sample rotation capability, together with the already existing sample oscillation capability, opened up a new possibility of conducting various drop dynamics experiments using molten metals in high vacuum condition. The sample rotation capability added the following new possibilities to the JPL HTESL:

(i) Shapes and stability of rotating molten metal drops carrying net surface charges were experimentally investigated in a high vacuum, and the prediction by Brown and Scriven for rotating drops have been confirmed. However, rotating tin drops showed a marked deviation from the prediction. At the present time, we speculate that the anomaly is caused by the nuclear isotope separation in the rotating tin drops under the influence of ~ 100-g centrifugal force.

(ii) A new method of measuring surface tension of molten metals, especially of those which are highly viscous, was developed. This will also open up a possibility of developing a new non-invasive method of measuring viscosities of highly viscous liquids.

(iii) The oscillation frequency of a rotating aluminum drop was measured as a function of rotation rate. This is important for the surface tension measurement techniques which utilize drop oscillation. Without accurate knowledge on drop rotation rate at the time of the oscillation measurement, the surface tension determined by it will be erroneous.

(iv) A non-contact technique of measuring the electrical conductivity (or resistivity) of conducting liquids was developed. This technique measures relative changes in torque as a function of temperature when a rotating magnetic field was applied.

(v) Naturally derived from (iv) above is the indirect determination of the thermal conductivity from the experimentally determined electric conductivity using the Wiedemann-Franz-Lorenz relation. Thermal conductivity measurement is known to be more susceptible to the convective flows than the electrical conductivity. It is hoped that indirect measurement increases the accuracy of the thermal conductivity over the direct measurement. Much more work needs be done on this subject.