

Talk 1: Current challenges in liquid and glass science at high pressure

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In the past decade it has become increasingly apparent that from a fundamental point of view the liquid state is not simply a blank, randomly disordered void as is so often implied by a glance at the phase diagram. Rather a considerable number of transitions, both gradual and abrupt, occur in the liquid and fluid states, which has researchers now penciling in transitions and bands along with the well-documented crystalline transitions. As may be expected, structural rearrangements with pressure and temperature are normally associated with corresponding kinks in thermodynamic and physical properties such as density, viscosity, and conductivity. Such structural transitions are known to be associated with changes in electronic structure, coordination number changes, variations in topology and polymerization to name a few. Of particular importance to the applied sciences are transitions leading to Pressure Induced Amorphization, especially those processes which can lead to the synthesis of new families of materials with different properties and characteristics than those possessed at ambient conditions.

Talk 2: Structure, density and viscosity measurements in the PE press

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The simultaneous collection of high quality X-ray diffraction data, density and viscosity at high pressure and high temperatures is very important in various domains of research such as geosciences or materials science. However, the practical realization of such multi-purpose device is very challenging. We present such a development at beamline ID27 which is based on the newly developed VX5 Paris-Edinburgh press. The press is interfaced to newly developed Soller slits and imaging systems to measure high quality diffraction patterns and high resolution X-ray images of the sample assembly. The press is mounted on an optimized 2 circle diffractometer which allows to move the press upside down in order to perform viscosity measurements using the falling sphere method. The potential of this new development will be illustrated on some school case examples.

Talk 3: Properties of silicate melts at high pressure

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In situ studies of silicate liquids at high pressure, aim at answering questions relevant to the presence of magmas at depth, whether that be in the present Earth or in its earliest times, during differentiation of the planet. Understanding the macroscopical physical properties of magmas requires an accurate microscopic structural description. Complementary structural, density and viscosity measurements at high P-T conditions generated by Paris-Edinburgh cells will be presented. Data will include results obtained on a simple end-member composition, i.e. molten fayalite, by means of energy-dispersive X-ray diffraction on ID16-BM-B (APS, HPCAT), and on a complex natural composition, i.e. a basalt, by means of angle-dispersive X-ray diffraction on ID27 (ESRF). Both energy-dispersive and angle-disperse techniques have pros and cons to measure the properties of melts, that will be discussed along with the geophysical implications of the obtained results.

Talk 4: Combined structure, elastic wave velocity, and viscosity measurements of liquid and amorphous materials in the Paris-Edinburgh cell

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Correlation between structure and physical properties is fundamental for understanding the behavior of materials. However, those of liquids and amorphous solids at high pressure and high temperature conditions have not been well studied due to experimental difficulties. The sector 16-BM-B, HPCAT at the APS is capable of amorphous and liquid structure measurement by using multi-angle energy dispersive x-ray diffraction measurement at high pressure and high temperature conditions in a Paris-Edinburgh (PE) cell (e.g., Yamada et al., 2011). The PE cell is capable to compress large volume sample ($>1 \text{ mm}^3$) up to 7 GPa and to increase the sample temperature to 2000 °C by graphite heater. In addition to the liquid and amorphous structure measurement capability, we recently developed ultrasonic elastic wave velocity and falling sphere viscosity measurement techniques in the PE cell. The elastic wave velocity is measured by ultrasonic pulse echo overlap method in conjunction with white x-ray radiography measurement for determining the wave travel distance (Kono et al., 2012). The falling sphere viscosity measurements are made with high-speed white x-ray radiography ($>1000 \text{ frame/second}$) (Kono et al., 2013). The integration of liquid structure measurement with elastic wave velocity measurement and viscosity measurement in a PE cell provides a unique opportunity to investigate *in situ* correlation between the microscopic structure and the macroscopic properties of liquids and amorphous solids at high pressure and high temperature conditions.

References

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Talk 5: Measurements on physical properties of liquid alloys under high pressure

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Physical properties of liquid Fe-alloys, such as density, sound velocity, interfacial tension and interconnectivity, under high pressure are important for understanding the properties and dynamics of terrestrial liquid outer core. Density and sound velocity are key parameters to clarify the composition of the core by comparing these properties with the seismic data obtained from the core. Interfacial tension and interconnectivity of liquid Fe-alloys in the solid/liquid silicate are closely related to a separation mechanism between core and mantle in the early terrestrial planets. We have studied these physical properties of liquid Fe-alloys under high pressure based on 2D/3D X-ray imaging combined with Paris-Edinburgh and multi-anvil apparatuses. In this presentation, I will introduce some techniques for obtaining these properties and their results.

For the simultaneous measurements of density and sound velocity, X-ray absorption method (e.g., Terasaki et al. 2010) and ultrasonic pulse-echo overlap method (e.g., Higo et al. 2008; Nishida et al. 2013) were applied. High pressure experiments were performed using 80-ton uniaxial press (Urakawa et al., 2010) installed at X-ray computed micro-tomography (CT) beamline (BL20B2), SPring-8 synchrotron radiation facility. Wave signals of 37 MHz was produced and detected by the transducer and P wave travel time in the liquid was measured. Sound velocity can be obtained from the travel time and a sample thickness measured from radiography image. X-ray absorption profile of the sample was obtained from the X-ray radiography image and an X-ray path length of the sample was directly measured from the CT slice image. Density of the liquid can be deduced from these data based on the Beer-Lambert's law.

For the measurements of interfacial tension, the sessile drop method was used (Terasaki et al. 2009, 2013). In this method, since a liquid shape is determined by force equilibrium between gravity and interfacial tension, the interfacial tension can be estimated from its shape observed as X-ray radiography image.

Talk 6: Sound velocity of Fe and Fe-rich liquids at high pressures with implications to the lunar core

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Geophysical observations suggest that the Moon possesses a molten or partially molten iron core. Similar to Earth and other terrestrial planets, light elements are likely present in the lunar core. The comparison of laboratory data on sound velocities of pure Fe and Fe-rich liquids under the deep Moon conditions with seismic models will provide tight constraints on the composition, density, and thermal state of the lunar core, hence facilitating a better understanding of dynamics and chemical evolution of the Moon. However, previously sound velocity measurements on Fe-rich liquids were mostly performed at ambient pressure. In this study, we have developed techniques to directly measure the ultrasonic sound velocities of Fe-rich liquids at high pressures using both a Paris-Edinburgh cell and a Kawai-type multi-anvil apparatus. The sound velocity was determined by measuring the travel time difference between the sample echo and the buffer rod echo using a waveform generator and a digital oscilloscope and by measuring the sample thickness using X-ray radiographic images. X-ray diffraction was also used to determine the pressure of the experiments and to confirm the melting of the samples. Using this technique, we have successfully obtained sound velocities of three Fe-S liquid compositions (Fe-10wt%S, Fe-20wt%S, and Fe-27wt%S), two Fe-Si liquid compositions (Fe-17wt%Si and Fe-25wt%Si), and pure Fe liquid at high pressure and temperature conditions up to 8 GPa and 2073 K. Results show significant differences between Fe-S and Fe-Si liquids: (1) The velocity of liquid Fe decreases with increasing sulfur content, but increases with silicon content; (2) Velocity is nearly independent of temperature for Fe-S liquids, but decreases with increasing temperature for Fe-Si liquids. Using lunar seismic data, our experimental results tightly constrain the sulfur content, density, and temperature of the lunar outer core. The temperature insensitivity of velocity suggests a top-down solidification scenario for the lunar core, i.e., an “iron snow” that may have resulted in the growth of the inner core

Talk 7: Melts at high pressure: linking structure to physical properties

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Structures of polymerized (ex. jadeite) and depolymerized (ex. diopside) silicate melts were investigated as a function of pressure to 5 – 6 GPa. The two types of liquids show distinct responses to pressure. For jadeite melt, T-O (T denotes tetrahedrally coordinated Al and Si) bond length, T-T bond length, and T-O-T angle all exhibit rapid and non-linear decrease between 0 and ~2 GPa. Above 2 GPa, the decrease becomes much more subtle and linear. For diopside melt, these parameters show very little change. A large viscosity dataset from the literature shows that, with increasing pressure, viscosities of polymerized liquids first decrease with pressure, and reach a minimum before turning over. On the other hand, viscosities of depolymerized melts increase monotonically with pressure. Here we present our recent progress on structural evolution of a number of melts and glasses at high pressures and its relation with the measured physical properties. Using high pressure synchrotron x-ray diffraction and molecular dynamics (MD) modeling, we find that the viscosity turnover pressure corresponds to the tetrahedral packing limit, below which density increases rapidly by reducing free volume through tightening of the intra-tetrahedral T-O-T angle, and above which T-O coordination increases to allow further densification. Our final goal is to understand the physical connection between structure and the material's physical properties.

Talk 8: Power-law Scaling of Density in Metallic Glass

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Glass lacks the long-range translational periodic order that characterizes a crystal. Thus, it is very difficult to get detailed information about the atomic structure of glasses by diffraction, which remains a long-standing scientific problem. In a crystalline material, atoms are arranged periodically on a lattice and the first Bragg peak in the powder diffraction pattern corresponds to the largest interplanar distance (d_1 is proportional to $1/q_1$, where q_1 is the first peak position in momentum transfer). When scaling this linear length ($1/q_1$) with the atomic volume (V_a), a power of 3 is obtained as a direct consequence of the periodic crystalline order. On the other hand, a signature of 'disordered' structure is the emergence of a few diffuse scattering haloes rather than sharp Bragg peaks. In a molecular gas, where only short-range order (SRO) is present, $1/q_1$ is related to the interatomic distance following the well-known Ehrenfest relationship [1] and obeys the 3 power-law scaling with the molecular volume. In amorphous solids such as a metallic glass, atomic correlations extend well beyond SRO,[2,3] but still lack long-range periodicity. However, it is believed that the first maximum halo in its diffraction pattern still carries significant information about order in metallic glasses. On the basis of neutron and X-ray diffraction experiments of metallic glasses with different compositions at ambient conditions, D. Ma *et al.* proposed a new fractal network packing scheme, which surprisingly shows a universal power-law correlation function with a power of 2.31, significantly different from 3.[4] Pressure can also be used to dramatically shift the position of the diffraction peaks. However, the accurate pressure-density relationship is currently limited to well-crystallized solids where X-ray or neutron diffraction can yield accurate unit cell data at high pressures, and whether the same power-law correlation function holds is still unresolved. By combining the *in-situ* high pressure ultrasonic measurements and tomography with x-ray diffraction, we established the relationship between the first maximum halo peak position ($1/q_1$) and density of metallic glasses. Our results provide a critical check of the reliability of fractal network packing scheme and help to uncover the real atomic structure of metallic glass.

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