

SCG009-01

会場:301A

時間:5月23日 14:15-14:45

Speciation of P in and partitioning between aqueous fluids and silicate melts to upper mantle temperatures and pressures Speciation of P in and partitioning between aqueous fluids and silicate melts to upper mantle temperatures and pressures

Bjorn Mysen^{1*}

Bjorn Mysen^{1*}

¹Geophysical Lab, CIW

¹Geophysical Lab, CIW

Evidence from high-grade metamorphic terranes suggest that phosphorus solubility and solution mechanisms in silicate melts and aqueous fluids depend on composition resulting from a range of possible P-bearing structural complexes. The structure of phosphorus-bearing, H₂O-saturated silicate melts, silicate-saturated aqueous fluids, and silicate-rich single phase (supercritical) liquids has been characterized, therefore, via in-situ experiments from ambient temperatures and pressures to 800°C/1486 MPa. The solution mechanisms and partitioning of structural species between fluids and melts were determined with the aid of confocal microRaman and with FTIR spectroscopy backed up with published phosphorus-31 MAS NMR data [1]. The experiments were conducted in an Ir-gasketed hydrothermal diamond anvil cell. Temperature and pressure were recorded with thermocouples (1°C uncertainty) and pressure- and temperature-dependent Raman shift of ¹³C diamonds (40 MPa uncertainty). Starting materials were aluminum-free Na₂O·4SiO₂ (NS4) and with 10 mol % Al₂O₃ (NA10) substituting for SiO₂, both with 5 mol % P₂O₅. These compositions enabled characterization of phosphorus behavior with coexisting haploandesite melt and aluminosilicate-saturated aqueous fluids with variable Al-content.

Aluminosilicate species of Q₀, Q₁, Q₂, and Q₃ type exist in coexisting fluid and melt and in single phase liquid together with phosphate species, PO₄, P₂O₇, and Q_nP. In the Q_nP notation, the n-value denotes the number of oxygen in the structural species shared with P and Si. Al substitutes for Si predominantly in the Q_nP species. In melts, the abundance of the most depolymerized silicate species, Q₀, is positively correlated with temperature and pressure, whereas that of the most polymerized species, Q₃, decreases with temperature and pressure. In the silicate solute in aqueous fluids, the opposite relationship exists with Q₀ abundance decreasing and Q₃ (and Q₁ and Q₂) abundance increasing with increasing temperature and pressure. The silicate melts, therefore, become increasingly depolymerized and the silicate solute in aqueous fluids decreasingly depolymerized. The P₂O₇ and Q_nP are the dominant phosphate species in fluid, melt, and single phase liquid with orthophosphate, PO₄, playing a subordinate role. The fluid/melt partition coefficients for P₂O₇ and Q_nP species are in the 0.15-0.7 range with that of Q_nP being greater than that of P₂O₇. The PO₄ fluid/melt partition coefficients are <0.2. In all cases, the partition coefficients increase with increasing temperature and pressure. There is no clear influence of Al₂O₃. Hence, it appears that P-bearing complexes in fluids and melts are associated with Na⁺. Mobility of phosphorus during metamorphic processes likely is principally governed by availability of alkali metals (and perhaps alkaline earths), whereas Al/Si-ratio may be a less important composition parameter. The P-partitioning between fluids and melts likely are significantly pressure-dependent because fluid and melt speciation is sensitive to pressure.

[1] Cody, B. O., Mysen, B. O., Saggi-Szabo, G., and Tossell, J. A., 2001. *Geochim. Cosmochim. Acta* 65, 2395.

キーワード: melt structure, aqueous fluid, phosphorus species, species partitioning, spectroscopy

Keywords: melt structure, aqueous fluid, phosphorus species, species partitioning, spectroscopy

SCG009-02

会場:301A

時間:5月23日 14:45-15:00

高温高圧下のアルミノ珪酸塩メルトのX線構造解析 X-ray diffraction studies on the structure of aluminosilicate melt under pressure

浦川 啓^{1*}, 中塚 明日美¹, 渡辺 了², 亀掛川卓美³

Satoru Urakawa^{1*}, Asumi Nakatsuka¹, Tohru Watanabe², Takumi Kikegawa³

¹ 岡山大学大学院自然科学研究科, ² 富山大学理学部, ³ 高工ネ研・PF

¹Dept. Earth Sci., Okayama Univ., ²Dept. Earth Sci., Toyama Univ., ³Photon Factory, KEK

The physical properties of silicate magma, such as density and viscosity, are important to understand migration of magma within the planetary interior as well as evolution of magma ocean. As these properties are related to the structure of magma, the structural studies of silicate magma at high pressures are fundamental to understand the magma related processes within the planetary interior. Therefore, we studied the structure of aluminosilicate melts at high pressures. Here we report the results of X-ray diffraction analysis on $\text{Ca}_3\text{Al}_2\text{Si}_6\text{O}_{18}$, $\text{Mg}_3\text{Al}_2\text{Si}_6\text{O}_{18}$, and $\text{NaAlSi}_2\text{O}_6$ composition melts up to 7 GPa.

Static structure of aluminosilicate melts under pressure has been studied by in situ x-ray diffraction experiments using synchrotron radiation at Photon Factory, KEK, Japan. X-ray diffraction patterns were acquired just above the melting temperature to about 7 GPa by energy-dispersive x-ray diffraction method and were analyzed by Fourier method.

Radial distribution functions of aluminosilicate melts show the increase of T-O distance as pressure increases. The T-O distance is an indicator of the coordination number of the network-forming cation, Si and Al. The higher the coordination number is, the longer the T-O distance is. In these melts, it is expected that the coordination number of Al increases in these pressure range. The first sharp diffraction peak (FSDP) of interference function shifts higher Q-side with increasing pressure, indicating the shrinkage of intermediate range structure in these melts.

The changes in the structure are related strongly to density and viscosity in these melts. These results are important to understand how these properties changes with pressure.

SCG009-03

会場:301A

時間:5月23日 15:00-15:15

Viscosity of CaMgSi₂O₆ liquid at high pressure revisited Viscosity of CaMgSi₂O₆ liquid at high pressure revisited

鈴木 昭夫^{1*}

Akio Suzuki^{1*}

¹Tohoku University

¹Tohoku University

The knowledge of the viscosity of silicate melts under high pressure is of importance to understand magmatic processes in the Earth's interior. It is known that the pressure dependence of viscosity is strongly related to the structure of melt. The diopside (CaMgSi₂O₆) composition melt is characterized as a depolymerized melt, and a positive pressure dependence of viscosity has been reported. However, there is a discrepancy in a curvature in previous studies. Scarfe et al. (1979) and Brearley et al. (1986) reported three times increase from 1 atm to 1.5 GPa. In contrast, Taniguchi (1992) showed that the positive pressure dependence was half of the previous studies. The viscosity of CaMgSi₂O₆ liquid was measured by Reid, Suzuki et al. (2003) up to 13 GPa. However, the data at the low-pressure range between 3.5 and 7.0 GPa were scarce. In the present study, high-pressure viscosities of the diopside (CaMgSi₂O₆) composition melt were measured between 1 and 4 GPa. X-ray radiography technique was used to observe falling spheres in situ. We conducted experiments using the Kawai type multianvil apparatus loaded in the MAX-III press on the PF-AR NE7A station at the High Energy Acceleration Research Organization (KEK). The measured viscosities between 1 and 2 GPa were consistent with Taniguchi (1992) and inconsistent with Scarfe et al. (1979) and Brearley et al. (1986). A positive correlation to pressure was observed up to 4 GPa.

キーワード: 粘度, マグマ, マントル, 高圧, X線ラジオグラフィ

Keywords: viscosity, magma, mantle, high pressure, X-ray radiography

Japan Geoscience Union Meeting 2011

(May 22-27 2011 at Makuhari, Chiba, Japan)

©2011. Japan Geoscience Union. All Rights Reserved.



SCG009-04

会場:301A

時間:5月23日 15:15-15:30

非晶質物質の圧力下における弾性波速度と体積の同時測定

Simultaneous measurements of the elastic wave velocities and the volume for amorphous materials under pressures

山田 明寛^{1*}, 河野義生¹, ワン ヤンビン², 井上徹¹

Akihiro Yamada^{1*}, Yoshio Kono¹, Yanbin Wang², Toru Inoue¹

¹ 愛媛大学地球深部ダイナミクス研究センター, ² シカゴ大

¹GRC, Ehime Univ., ²GSECars, Univ. of Chicago

なし

キーワード: X線トモグラフィー, 超音波速度, 高圧, ガラス

Keywords: x-ray tomography, ultrasonic, high pressure, glass

SCG009-05

会場:301A

時間:5月23日 15:30-16:00

Universal behavior in pressure-induced melt-polymerization in primordial magmatic reservoirs Universal behavior in pressure-induced melt-polymerization in primordial magmatic reservoirs

Sung Keun Lee^{1*}

Sung Keun Lee^{1*}

¹Seoul National University

¹Seoul National University

Pressure-induced changes in melt-composition, viscosity, entropy, and solubility of elements in silicate melts in magma oceans provided a unique situation in which significant chemical differentiation of the silicate earth could have occurred and formed unrecycled partial by forming a hidden reservoir in Earth's mantle. Although the structures of silicate melts have been linked to these key properties, the melt structures at high pressure remained largely unknown. Even more challenging is to unveil the structure of natural silicate magmas in the Earth's mantle because any experimental effort to reveal the complex structure tends to be hampered by inhomogeneous broadening in experimental data associated with such complexity. Therefore, chemical constraints such as the non-bridging oxygen (NBO) content at 1 atm, rather than the real structural parameters for melt polymerization at high pressure, are commonly used to account for pressure-induced changes in the melt properties in Earth's interior. Here, we show that the pressure-induced NBO fraction in diverse silicate melts show a universal behavior where all the reported experimental NBO fractions at high pressure can be simplified into a single decaying function, regardless of melt composition. This simplicity in the pressure-induced changes in melt polymerization enables us to account for the non-linear variations in thermodynamic and the transport properties of multi-component and thus natural silicate melts at high pressure. The current results with universality in melt polymerization thus provide atomistic insights into the density crossover between melts and crystals and a formation of hidden reservoir with distinct chemical composition.

キーワード: silicate melts, high pressure, melt polymerization, mantle reservoirs

Keywords: silicate melts, high pressure, melt polymerization, mantle reservoirs

SCG009-06

会場:301A

時間:5月23日 16:00-16:15

Fe-S 融体の過剰モル体積の圧力依存性 Pressure effect on excess molar volume of liquid Fe-S

西田 圭佑^{1*}, 大谷 栄治¹, 寺崎 英紀¹, 立山 隆二¹, 柴崎 裕樹¹, 鈴木 昭夫¹, 亀卦川 卓美²

Keisuke Nishida^{1*}, Eiji Ohtani¹, Hidenori Terasaki¹, Ryuji Tateyama¹, Yuki Shibazaki¹, Akio Suzuki¹, Takumi kikegawa²

¹ 東北大学大学院理学研究科地学専攻, ² 物質構造科学研究所

¹Department of Earth and Planetary Mater, ²Institute of Materials Structure Science

The excess molar volume of liquid Fe-light element alloys at high pressure represents non-ideality for mixing of end-member components and, therefore, it is very important to estimate the light element contents in the outer core based on the density deficit in the core. Sulfur is considered to be a major candidate of light elements, because it can dissolve into liquid Fe even at low pressure and it is depleted in the crust and mantle relative to the other volatile elements. Previous works (e.g. Poirier, 1994) estimated the light element contents in the core assuming ideal-mixing behavior between iron and light elements, i.e., neglecting their excess volumes. The excess molar volume of liquid Fe-S at 4 GPa was reported to be large and to have a negative value (Nishida et al., 2008). Therefore, if this excess molar volume can be applied at the core condition, the outer core may contain more light elements than the previous estimates. However, pressure effect on the excess molar volume of liquid Fe-S has never been reported.

In this study, we measured the density of liquid Fe-S at 0.5 GPa and 1650 °C using sink/float method. We fitted the present molar volume assuming Fe-S liquid can be treated as the regular solution. Derived negative excess molar volume of Fe-S at 0.5 GPa and 1650 °C is larger than that at 4 GPa. The negative excess molar volume of liquid Fe-S decreases with increasing pressure. This result may suggest the excess molar volume of liquid Fe-S at the pressure of the Earth's outer core might be small and negligible.

Keywords: Fe-S, liquid, density, excess molar volume, high pressure

SCG009-07

会場:301A

時間:5月23日 16:30-17:00

Alkali effect in silicate melts ? A new vision on an old hypothesis Alkali effect in silicate melts ? A new vision on an old hypothesis

Daniel Neuville^{1*}, Charles Lelosh¹, Pierre Florian², Alain Baronnet³, Dominique Massiot³
Daniel Neuville^{1*}, Charles Lelosh¹, Pierre Florian², Alain Baronnet³, Dominique Massiot³

¹CNRS-IPGP, ²CEMHTI-CNRS, ³CINaM-CNRS

¹CNRS-IPGP, ²CEMHTI-CNRS, ³CINaM-CNRS

Rheological properties of silicate melts govern both magma ascension from the mantle to the surface of the earth and volcanological eruptions styles and behaviors. In this mind, it is very important to understand what parameters influence these properties. Up to now, we know for example that viscosity of silicate melts is dependent of temperature, pressure and chemical composition (Bottinga and Weill, 1972; Urbain et al, 1982). In this work, we will focus on the Na₂O-K₂O-Al₂O₃-SiO₂ system, which is of a prime importance because it deals with a non-negligible part of natural melts, like for instance Vesuvius (Italy) or Erebus (Antartica) magmas. We will first present our viscosity data, and then the Adam and Gibbs theory that allows theoretically modeling Na-K mixing in aluminosilicate melts using the so-called mixed alkali effect. On the basis of these rheological results, the Na-K mixing cannot be explained with this mixed alkali effect. To go further and as rheological properties are directly linked with structural properties, we will present our first results obtained by Raman and NMR spectroscopy. These last ones provide important structural informations on the polymerization of glasses and melts, and also on the environment of tetrahedral coordinated cations. These structural results are directly linked with viscosity measurements and shown that substituting Na by K in aluminosilicate melts induces structural changes in both alkali environment and aluminosilicate network. This implies that Na and K atoms are non-randomly distributed in the aluminosilicate network. Na melts present a network with some channels and a random distribution of Al and Si. K networks are different, they present a non-random distribution of Al and Si, with two sub-networks: one rich in Si and fully polymerized, the other containing Al and K. On this view, mixing Na and K melts returns to change these configurations and induces complicated and non-linear effects.

キーワード: glass, melt, aluminosilicate

Keywords: glass, melt, aluminosilicate

SCG009-08

会場:301A

時間:5月23日 17:00-17:15

Probing the effect of composition on structural disorder of basaltic and slab-driven melts using solid state NMR

Probing the effect of composition on structural disorder of basaltic and slab-driven melts using solid state NMR

Sun Young Park^{1*}, Sung Keun Lee¹
Sun Young Park^{1*}, Sung Keun Lee¹

¹Seoul National University

¹Seoul National University

Whereas the structure of multi-component silicate melts has strong implication for the properties of natural silicate melts and relevant magmatic processes in mantle and crust of the Earth, little is known about their atomic structures due to lack of suitable experimental probes of multi-component amorphous oxides. Whereas most of the progress in melt structure has been made for relatively simple binary and ternary silicate glasses, recent advances in high-resolution solid-state NMR unveil previously unknown structural details of multi-component silicate melts. Here, we report the experimental results of the effect of composition on the atomic structure and disorder in quaternary [CaO-MgO-Al₂O₃-SiO₂ (CMAS)] using multi-nuclear high-resolution 1D and 2D solid-state NMR. We also report the first NMR results for the diverse glasses with compositions of natural silicate melts. The Al-27 NMR results for diopside- Ca-Tschermakite pseudobinary join, suggest a increases in topological and configurational disorder with increasing diopside content. While the glasses with basaltic compositions show that ⁴Al is dominant, non-negligible fraction of ⁵Al were observed for basaltic composition melts while negligible fraction of ⁵Al was observed for the slab-driven melts . The high-resolution O-17 3QMAS NMR spectra of diopside-Ca-Tschermakite pseudobinary join show that three types of bridging oxygens (BO; Si-O-Si, Al-O-Al, and Si-O-Al) and two types of NBO (Ca-NBO, and mixed?NBO) are partially resolved. Previously unknown structural details in Ca-Mg aluminosilicate glasses include nonrandom distributions of Ca²⁺ and Mg²⁺ around NBO and BO and significant fraction of Al-O-Al in natural basaltic magmas. The preferential partitioning of Ca²⁺ and Mg²⁺ between NBO and BO may results in a variation of activity coefficient of CaO and MgO, thus controlling composition of melts generated at the mid-ocean ridge and subduction zone.

キーワード: basaltic melt, multi-component glass, NMR, atomic structure

Keywords: basaltic melt, multi-component glass, NMR, atomic structure

SCG009-09

会場:301A

時間:5月23日 17:15-17:30

Oxidation state and coordination structure of Fe in silicate glasses and melts Oxidation state and coordination structure of Fe in silicate glasses and melts

Benjamin Cochain^{1*}

Benjamin Cochain^{1*}

¹ISEL, Okayama University

¹ISEL, Okayama University

In both magmatic and industrial systems, Fe is the most abundant transition element. Due to its heterovalent nature and the different crystal- chemical behavior of the reduced and oxidized species, Fe affects a wide number of physical and chemical properties of magmas or final glass products, such as density, viscosity, stability of phases, and nucleation during crystallization. Particularly, the viscosity of Fe-bearing silicate melts decreases with decreasing Fe³⁺ content of the melt, which provides clear but indirect evidence for differences in the structural role of Fe³⁺ and Fe²⁺. Many studies have addressed the structural role of Fe in melts using glasses as structural analog (Calas and Petiau, 1983). In most cases, Fe³⁺ in silicate melts was assigned to tetrahedral site geometry, although evidence for higher coordination was also found. On the other hand Fe²⁺ was found distributed over sixfold-, fivefold- and fourfold-coordinated sites in melts, with the last two dominating (Rossano et al., 2000).

In the work presented here, in-situ X-ray absorption spectroscopy at the Fe K-edge was used to characterize the local structural environment of Fe³⁺ and Fe²⁺ in alumino-boro-silicate melts at high temperature (up to 2000K) in comparison to their quenched glassy analog at room temperature. Changes in the structural environment of Fe were evaluated by analyzing the pre-edge feature of the EXAFS spectra. The most useful characteristics of the Fe-K pre-edge for determining Fe oxidation state and coordination number are the position of its centroid and its integrated intensity. To plot these pre-edge parameters in the variogram after Wilke et al. (2001) allow determining the oxidation state and the coordination of iron. In an effort to complete this variogram, a series of Fe-bearing minerals, with Fe³⁺ and Fe²⁺ coordination ranging from 4 to 6 O atoms, has also been analysed by X-ray diffraction and Fe K-edge EXAFS techniques. The coordination structure of Fe and Fe-O distances in minerals were thus determined. The characteristics of the pre-edge features of the EXAFS spectra are now related with oxidation state, local coordination environment of Fe atoms but also with Fe-O distances.

In this study, new information about incorporation of Fe³⁺ and Fe²⁺ into a variety of alumino-boro-silicate glasses and melts will be presented. These results on the coordination structure of iron in silicate glasses and melts will be also discussed in regards to the kinetics of iron redox reactions in silicate melts (Magnien et al., 2008; Cochain et al., 2010).

Calas G. et Petiau J. [1983] Sol. Stat. Com., 48, 625-629.

Rossano S., Ramos A. and Delaye J-M [2000] J Non Cryst Solids, 273, 48-52.

Wilke M., Farges F., Petit P.E., Brown Jr G. E., and Martin F. [2001] Amer. Mineral., 86, 714-730.

Magnien V., Neuville D.R., Cormier L., Roux J., Hazemann J.-L., De Ligny D., Pascarelli S., Pinet O. et Richet P. [2008] Geochim. Cosmochim. Acta., 72, 2157-2168.

Cochain B., Neuville D. R., de Ligny D., Roux J., Baudalet F., Struklejev E. and Richet P, [2009] Journal of Physics IV, 190, 012182.

キーワード: XAS (XANES, EXAFS), Fe- K-edge, Fe in silicate melts, Fe in minerals, Fe coordination and redox state

Keywords: XAS (XANES, EXAFS), Fe- K-edge, Fe in silicate melts, Fe in minerals, Fe coordination and redox state

SCG009-10

会場:301A

時間:5月23日 17:30-17:45

Billouin 散乱と Raman 散乱による高圧下での含水ナトリウム珪酸塩ガラスの構造変化

Structural change of hydrous sodium silicate glass under high pressure using Brillouin and Raman spectroscopies

西本 太郎^{1*}

Taro Nishimoto^{1*}

¹ 西本 太郎, ² 村上 元彦, ³ 大谷 栄治

¹Taro Nishimoto, ²Motohiko Murakami, ³Eiji Ohtani

The structure and physical properties of silicate melt are important to understand the Earth's mantle. However, it is technically difficult to conduct experiments of the melt at high pressure and high temperature. The glass is important for analog materials of the melt and one of the plausible approaches to understand its structure and density changes. It is important for melt to understand the glass which contains SiO₂ as a basic component of melt. Elastic velocities of glass consist of the bulk modulus, shear modulus and density. Elastic velocities enable us to discuss the structure and density changes of melt. Brillouin scattering with DAC enables us to discuss about the glass structure and density changes under high pressure indirectly based on pressure effect on trend elastic velocities. Additionally, water owes its importance to the dramatic influence which it exerts even at very low concentrations on variety of physical properties. In this study, we measured the elastic velocity of Na₂Si₄O₉ glass, which is a binary system of SiO₂-Na₂O glass, under high pressures up to 50 GPa based on Brillouin scattering together with diamond anvil cell. The other starting material is hydrous Na₂Si₄O₉ glass. I synthesized it based on hydrothermal experiment. I observed the sample using a polarization microscope and analyzed it using FT-IR to estimate the content of water in the sample. I observed the peak derived from Si-O bond of the glass using Raman spectroscopy. To expect the structure and density changes of the hydrous glass based on the elastic velocities, I measured hydrous Na₂Si₄O₉ glass elastic velocity up to 50 GPa based on Brillouin scattering together with a diamond anvil cell at SPring-8. We observed the apparent elastic velocity profile change around 35 GPa. Below 35 GPa, the relatively steeper gradient (V_p ; $dV/dP=0.11$, V_s ; $dV/dP=0.043$) of the elastic velocity profile was observed. Above 35 GPa, the relatively gentle gradient (V_p ; $dV/dP=0.05$, V_s ; $dV/dP=0.024$) was observed. Based on these results, the relatively steeper gradient suggests that the structure of Na₂Si₄O₉ glass changes from 10 to 35 GPa. This tendency is consistent with the result of previous Raman spectroscopy (Wolf et al., 1990) which shows that the coordination number of silica changes from 4 to 6 between 20 and 33 GPa and above 33 GPa, the coordination number of silica is constant of 6 coordination. Density vs. pressure relationship was also calculated from 35 to 50 GPa based on the observed values of V_p and V_s . The relationship possibly helps to construct the precise equation of state of sodium silicate glass under high pressure, although it requires quantitative values of glass density of ambient conditions. The measurement of elastic sound velocities using Brillouin scattering could be one of the most favorable approaches to understand the structure and density changes of glass. Combined measurement with the other spectroscopic methods like Raman scattering or X-ray diffraction and absorption would leads farther understanding of density and structure change of glass. We will present about details of the result and discussion of hydrous glass experiments.

キーワード: ガラス, メルト, 構造変化, Brillouin 散乱法, Raman 散乱法

Keywords: glass, melt, structural change, Brillouin scattering, Raman spectroscopy

SCG009-P01

会場:コンベンションホール

時間:5月23日 10:30-13:00

EXEFS 法による高圧急冷珪酸塩ガラスの構造研究 Application of the EXEFS to the structure of the high-pressure aluminosilicate glasses

横田 育美^{1*}, 浦川 啓¹, 芳野 極²

Ikumi Yokota^{1*}, Satoru Urakawa¹, Takashi Yoshino²

¹ 岡山大学自然科学研究科, ² 岡山大学地球物質科学研究センター

¹Dept Earth Sci, Okayama Univ, ²ISEI, Okayama Univ

Silicate melts play an important role in the chemical evolution of the planetary mantle during the early magma ocean stage as well as the subsequent long history. Movement of silicate melt in the mantle is controlled by its viscosity and density that are related to the melt structure. Therefore, the structural study of silicate melts under pressure is fundamental to understand the magma-related phenomena within the planets. Pressure-induced structure change in silicate melts have been studied with the quenched glass by using spectroscopic methods such as the NMR and the XAFS. Here we report the first results of application of the extended x-ray emission fine structure (EXEFS) to the structure analysis of the quenched silicate glass. The EXEFS arises from the radiative Auger effect and has the same structure as the XANES. The EXEFS can be measured using a wavelength-dispersive electron microprobe.

We measured the EXEFS spectra of the $\text{Ca}_3\text{Al}_2\text{Si}_6\text{O}_{18}$ (CAS) composition glasses quenched at 0.1MPa and 8 GPa. High-pressure glass was prepared by using a KAWAI type multi-anvil apparatus. The Si EXEFS spectra show that the silicon in the CAS glasses takes four fold coordination up to 8 GPa. On the other hand, the coordination change of aluminum from four to five is detected by the EXEFS, which is consistent with the results of the Al NMR study on the same composition quenched glasses (Allward et al. 2005).

SCG009-P02

会場:コンベンションホール

時間:5月23日 10:30-13:00

X線吸収法による Fe-Si 合金融体の密度測定 Density measurement of liquid Fe-Si using X-ray absorption method

立山 隆二^{1*}, 西田 圭佑¹, 寺崎 英紀¹, 浦川 啓², 中塚 明日美², 大谷 栄治¹, 片山 芳則³
Ryuji Tateyama^{1*}, Keisuke Nishida¹, Hidenori Terasaki¹, Satoru Urakawa², Asumi Nakatsuka², Eiji Ohtani¹, yoshinori katayama³

¹ 東北大学大学院理学研究科地学専攻, ² 岡山大学大学院自然科学研究科, ³ 日本原子力研究開発機構

¹Tohoku University, ²Okayama University, ³Japan Atomic Energy Agency

The density of liquid Fe alloys under high pressure is important for estimating the amount of light elements in the Earth's outer core. Here, we performed the density measurement on the liquid Fe-10wt%Si using X-ray absorption method in order to clarify the effect of pressure on the density and to determine the equation of state of the liquid.

X-ray absorption method provided the density of the liquid Fe-10wt%Si at pressures and temperatures up to 3.6 GPa and 2173 K, respectively. The density of the solid Fe-10wt%Si decreased with increasing temperature (1073-1373 K). However, the density of liquid Fe-10wt%Si did not show a clear tendency to the temperature. The thermal derivative of the density of this study was $-0.00055 \text{ gcm}^{-3}\text{K}^{-1}$ at 3.5 GPa, whereas that of ambient pressure was $-0.001 \text{ gcm}^{-3}\text{K}^{-1}$. Therefore, the effect of temperature on the density of the liquid under high pressure is much weaker than that of the ambient pressure. Vinet equation of state yielded isothermal bulk modulus $K_{0T} = 59(5) \text{ GPa}$ with its pressure derivative $K' = 4$ at 1873 K. The present results revealed that the substitution of Si into Fe decreases not only the density of liquid Fe but also the bulk modulus of that. Based on the obtained density and bulk modulus of liquid Fe-Si, the bulk sound velocity (V_P) of liquid Fe-Si is lower than that of pure liquid Fe in the range of our experimental condition.

キーワード: 高圧, 密度, 軽元素, X線吸収法, 状態方程式

Keywords: high pressure, density, light element, X-ray absorption method, equation of state

SCG009-P03

会場:コンベンションホール

時間:5月23日 10:30-13:00

Structure of jadeite-diopside melts at high pressure by in situ x-ray diffraction Structure of jadeite-diopside melts at high pressure by in situ x-ray diffraction

坂巻 竜也^{1*}, Yanbin Wang¹, Tony Yu¹, Changyong Park², Guoyin Shen²
Tatsuya Sakamaki^{1*}, Yanbin Wang¹, Tony Yu¹, Changyong Park², Guoyin Shen²

¹GSECARS, The University of Chicago, ²HPCAT, Geophysical Laboratory

¹GSECARS, The University of Chicago, ²HPCAT, Geophysical Laboratory

Properties of silicate melts control magma-related processes such as volcanic activity and evolution of the Earth. Since these processes take place in Earth's deep interior, there is considerable interest in documenting experimentally how pressure affects properties of silicate melts. Macroscopic physical properties are largely determined by the microscopic structure. The bond length and strength between tetrahedrally coordinated cation (T=Si⁴⁺, Al³⁺) and oxygen (T-O length) are especially important in the relationship between structure and physical properties of silicate melts. For silicates, T-O lengths are the shortest among a large variety of melts, therefore we need XRD data with large Q coverage in order to obtain accurate T-O length experimentally. In this study, we tried out XRD experiments by Paris-Edinburgh press, which enables us to get XRD patterns to 2theta angles as high as 40 degree, and photon energies in excess of 100 keV. On the basis of the structural investigation at ambient pressure, the jadeite melt is a typical polymerized melt, while the diopside melt is depolymerized. Considering the structural parameters under ambient condition the ratio between non-bridging and tetrahedrally bonded oxygen (NBO/T), jadeite melt is 0 and diopside melt is 2. Therefore, these two compositions would allow us to examine the relationship between structure and composition of silicate melts.

High-pressure and high-temperature XRD experiments were carried out in the Paris-Edinburgh press, which was developed by GSECARS and installed at the HPCAT beamline 16-BM-B of APS. The compositions of starting materials were synthetic jadeite (Jd), diopside (Di), and Jd₅₀Di₅₀. The sample container was graphite. The encapsulated samples were enclosed in an hBN cylinder, which served both as an electric insulator and a pressure marker. High-temperature was generated by resistive heating of graphite heater outside the BN cylinder. Pressure medium consisted of ZrO₂, MgO and boron-epoxy. The center of the pressure medium was boron-epoxy and MgO, because of their low absorption to X-ray. The incident X-ray was collimated by a vertical slit (0.5 mm) and a horizontal slit (0.1 mm) to irradiate the sample. The diffracted X-ray was detected by a Ge solid state detector with a 4000 multi-channel analyzer, through vertical (0.5 mm) and horizontal (0.1 mm) receiving slits as well as a collimator. The diffraction patterns were collected for 12 fixed diffraction angles (2theta=3, 4, 5, 7, 9, 11, 15, 20, 25, 30, 35, 39.5 degrees).

The structure measurements of jadeite-diopside melt were carried out in the pressure range from 1 to 5 GPa and at 1600 to 2000 K. Results on structure factors S(Q) and radial distribution functions G(r) of these melts at high pressures and high temperatures will be discussed.

Keywords: melt, high pressure, high temperature, structure, X-ray diffraction

SCG009-P04

会場:コンベンションホール

時間:5月23日 10:30-13:00

Na₂Si₂O₅ メルトの分子動力学シミュレーション Molecular dynamics simulations of sodium silicate melts

則竹 史哉^{1*}, 河村 雄行¹

Fumiya Noritake^{1*}, Katsuyuki KAWAMURA¹

¹ 東京工業大学大学院地球惑星科学専攻

¹Earth and Planetary Sci., Tokyo Tech

Inter-atomic potentials are essential for precise reproduction and prediction of physical property of the system using molecular dynamics simulation. In the silicate system, many pair potential models have been proposed. Some of these can reproduce crystal structures and its elastic properties (e.g. van Beest et al. 1990) and were used for silicate melts (Lacks et al. 2005). However, these previous models which can reproduce qualitative pressure dependence of viscosity seem not to reproduce temperature dependence of Q-species (Maehara et al. 2005). Understanding the physical properties and their characteristic behavior of silicate melts, nano-structure and its temperature, pressure and composition dependence are needed to know.

To investigate the nano-structure of silicate melts, we employed newly developed inter-atomic potential model for silicate systems. Our potential model definitely includes coulombic interaction, short range repulsion, van der Waals interaction and radial covalent interaction terms. Molecular dynamics simulations were performed for Na₂Si₂O₅ system using the MXDORTO. The number of atoms, pressure and temperature are maintained constant (NPT ensemble, Natom=5994, P=0.1MPa). Physical property and Q-species were obtained at every 300K during the cooling from 3000K. The equilibrium data were obtained after 2-3ns (2-3,000,000steps) relaxation at each temperature. From this simulation, thermal expansion and temperature dependence of Q-species were investigated. The temperature dependence of Q-species was qualitatively reproduced, however it was not enough yet at quantitative aspect.

To improve the inter-atomic potential parameters, we are performing MO calculation using Gaussian09. To focus on Si-O-Si bond, we calculated energy surfaces of silicate clusters contain Si-O-Si bond (e.g. dimer, rings) for structural changes. By fitting inter-atomic potential parameters to these energy surfaces, improvement of reproducibility of physical properties is expected and the relation between physical properties and atomic structure might be discussed.

Keywords: MD, molecular dynamics, silicate melt