In-situ visualization of experimentally reproduced chondrule textures from crystallizing silicate melts, Atul Srivastava[1], Y. Inatomi[2], K. Tsukamoto[1], and H. Miura[1], [1]Department of Earth and Planetary Materials Science, Graduate School of Science, Tohoku University, Sendai 980-8578, Japan (atuldotcom@gmail.com), [2]Institute of Space and Astronautical Science, Japan Aerospace Exploration Agency, Sagamihara, Kanagawa 229-8510, Japan.

Introduction: The present work is concerned with the real time *in-situ* visualization of the crystallization process of strongly supercooled silicate melts using optical imaging techniques. The crystallization experiments are carried out for forsterite (Mg_2SiO_4) composition under container-less conditions. A gas-jet levitation arrangement is employed to hold the silicate spherules during the experimental run time. The spherules are heated up to their liquidus temperature using a high power CO_2 laser and allowed to cool down under black body radiation conditions to initiate the crystallization process. A combination of two refractive index-based imaging techniques, namely schlieren and shadowgraph [1-2], is employed to visualize melt convection and subsequent crystallization process inside the silicate spherules *in-situ*. The primary objective of *in-situ* visualization is to explore the potential of optical techniques for imaging melt convection at near liquidus temperatures and prediction of possible chondrule textures [3] in real time non-destructively. The crystallization experiments reveal suppression of homogeneous nucleation under container-less conditions. Formation of porphyritic textures is observed for a supercooling of about 580 K below the liquidus temperature while a rim-bar structure is reproduced for ΔT ~ 400 K. The results of the present experiments also reveal that for very large values of supercooling, it is possible to have the nucleation centre inside the melt droplet. The *in-situ* predictions of experimentally reproduced chondrule textures are compared with the textures revealed by photomicrographs of the corresponding thin-sections and a good agreement is seen between the two observations. The overall assessment to emerge from the present study is that the refractive index-based optical techniques present a novel approach for real time observation of the crystallization processes and provide a pathway for quantitatively determine the experimental conditions responsible for the formation of various textures as observed in natural chondrules.

The specific objectives of the present study are:

1. Imaging of convection inside the melt droplet at near liquidus temperature using white-light schlieren technique;
2. Reproduction and *in-situ* visualization of the possible chondrule textures in real time during the crystallization process from supercooled silicate melts using shadowgraph technique.

The on-line prediction of possible chondrule textures during the crystallization process itself in real time is of significant importance as it is capable of providing *a priori* information of final internal structures in a non-destructive sense (without cutting the final crystallized sample). To demonstrate this, the *in-situ* predictions of the experimentally reproduced chondrule textures using shadowgraph technique have been compared with the final textures revealed by the photomicrographs of the corresponding thin sections.

Optical arrangement: The primary instrument used in the present work for the visualization of convection and the whole process of crystallization inside the silicate melt droplets is a combination of schlieren and shadowgraph imaging techniques. A high power white light source (Xenon lamp, 75 W power output) has been employed as the light source. It provides luminescence that is comparable to the intensity of radiation from the sample heated up to ~2000^{o}C. Strong divergent light beam coming out of the Xenon lamp has been collimated using a pair of lenses. A decollimating lens tightly focuses the optical beam onto the levitated silicate melt droplet. Irrespective of the imaging technique (schlieren or shadowgraph), the light beam passing through the melt droplet contains path-integrated information about the distribution of refractive index and the image formation is due to the deflection of the light beam because of variable refractive index field within the spherule. The diverging light beam is finally made parallel using a collimator. In order to get the desired schlieren effect, this parallel beam of light is focused onto a knife edge (KE) using a decollimator. The optical components and path of the light beam to generate schlieren effect have been shown in the form of dashed lines in the figure. The knife edge is kept horizontal and is positioned to cut off a part of light focused on it, so that the initial illumination is uniformly reduced. After the initiation of the crystallization process, the *in-situ* visualization of the possible chondrule textures has been performed using the shadowgraph optical arrangement in real time.

Materials and methods: Magnesium silicate spherules (diameter: 1-2 mm, made of MgO and SiO_2 powder reagents mixed in a molar ratio of 2.0) of forsterite composition (Mg_2SiO_4) have been employed in the present experiments as the starting material. The experiments have been performed in air (oxidizing atmosphere) at normal pressure conditions. In order to simulate container-less conditions, the silicate spherules were levitated using a gas-jet levitator. The spherules of the starting material were transformed into complete melt using focused radiation from a 100 Watt continuous-wave carbon dioxide laser (DEOS, LC-100NV, wavelength: 10.6 µm, spot size: 1 mm). Temperature of the melt droplet was controlled by adjusting the output power of the laser. With this arrangement, the sample could be heated up to a temperature of ~2000^{o}C within 10 sec, and a cooling rate of 700 K/s at 2000^{o}C could be achieved by turning off the laser power. The surface temperature of the melt droplet was monitored using a monochromatic pyrometer (Impac, IN140/5, effective wavelength: 5.14 µm, spot size: 0.9 mm, and response time: 10 ms).

After the completion of the crystallization experiments, thin sections with thickness 0.03 mm (30 microns) of experimentally reproduced chondrules were prepared for microscopic examination of the internal textures using transmitted polarized light. Finally, the chondrule textures as observed using polarized microscopy have been compared with the corresponding in-situ real time shadowgraph images of the crystallization process. The major objective of the present work is to demonstrate the importance of in-situ visualization as a tool to predict possible chondrule textures in real time without damaging the samples.

Results: Figure 1 shows a time sequence of schlieren images of convection inside the melt droplet. A schlieren image carries field information related to the local temperature in the form of an intensity distribution as a function of position in a plane normal to the direction of propagation of the light beam. Therefore, the images shown in the figure also qualitatively reveal the degree of uniformity of temperature distribution during this stage of molten droplet. It is to be seen that the temperature gradients are too strong during the initial stages of heating of the silicate spherule resulting in completely non-uniform temperature distribution in spatial coordinates (as evident by colors of different shades in Figure 1(a-b)).

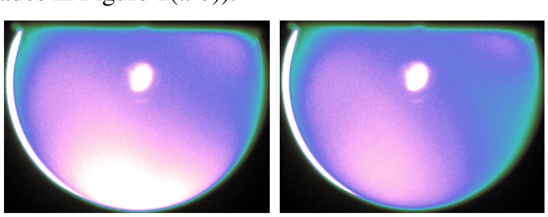

Figure 1: Sequence of schlieren images (false color) showing the transient evolution of melt convection inside the silicate melt droplet.

The time evolution of the crystallization process of subcooled molten silicate droplets as recorded using shadowgraph optical arrangement has been presented in Figure 2. As evident from the first image of the time sequence (Figure 2(a)), nucleation appears to have taken place at a number of sites simultaneously resulting in the appearance of a large number of small crystals inside the subcooled molten droplet. The amount of supercooling (ΔT) employed before the onset of nucleation process was ~ 580K. With the passage of time, these small crystals tend to grow in size and also the number density of such crystals increases, which in turn indicates that the nucleation process takes place at several locations and over a finite period of time. The levitation arrangement rotates/spins the molten droplets at high rotational speeds which in turn impart strong centrifugal forces on the crystals formed inside the silicate spherules. Therefore, these crystals tend to move towards the surface of the spherule (as can be seen from the successive images of Figure 2(b-c)) and finally give rise to a structure that closely resembles the porphyritic textures. The porphyritic nature was confirmed by comparing the observed textures with those observed using polarized microscopic images of the thin section of the crystallized spherule at the end of the experiment. A photomicrograph of the thin section is shown in Figure 2(d). A close agreement between the textures exhibited by the two images can be seen.

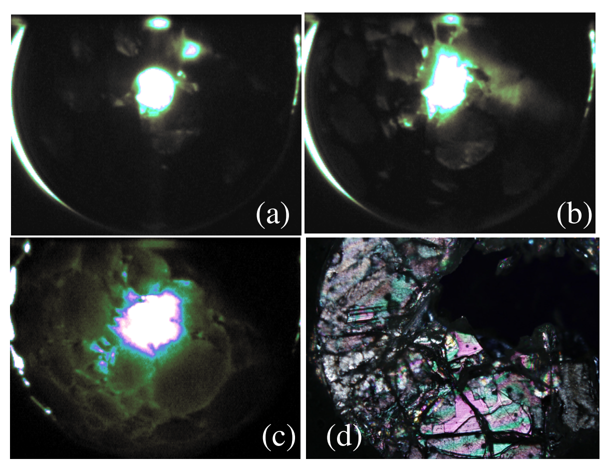

Figure 2: (a-c) Time sequence of instantaneous shadowgraph images showing the transient evolution of the crystallization process inside the silicate melt droplet; (d) Photomicrograph of thin section of the silicate spherule showing the porphyritic texture at the end of the crystallization process.

Details of experimental results would be presented.

References: [1] Goldstein R.J., Kuehn T.H., in: R.J. Goldstein (Ed.) (1996) Fluid Mechanics Measurements, Taylor & Francis, New York, 451-508. [2] Srivastava A., Muralidhar K. and Panigrahi P.K. (2005) *App. Opt. 44*, 5381-5392. [3] Hewins R.H. and Radomsky P.M. (1990) *Meteorites*, *25*, 309-318.