DEVELOPMENT OF SAMPLE PREPARATION TECHNIQUE FOR ISOTOPE ANALYSIS OF FLUID INCLUSIONS BY SIMS

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Introduction: Fluid inclusions in halite crystals from meteorites provide direct information of extraterrestrial liquid water [1]. In-situ isotope analysis of the fluid inclusions was realized by SIMS equipped with a cold-stage [2]. However, the analytical precision is limited to ±14 ‰ (1σ) and ±7 ‰ for $\delta^{18}O$ and $\delta^{17}O$, respectively, due to electrostatic charge up by deep sputter crater to excavate fluid inclusions. One of the methods to improve the analytical precision would be to expose fluid inclusions on the sample surface before sputtering. In order to realize the condition, we developed cryogenic sample preparation techniques including cryo-polishing, cryo-coating and cryo-transfer system for SIMS instruments.

Cryogenic Sample Preparation for SIMS: We froze the fluid inclusions and polished under cryogenic condition using a cryo-polishing system consists of a customized polisher (JEOL, HLA-2) in a glove box filled with nitrogen gas. The temperature of the polisher was controlled by liquid nitrogen flow through internal trench to -35 °C when the dew point in the glove box was about -45 °C. The sample surface was observed by an optical microscope (KEYENCE, VHX) integrated into the polisher to check whether the fluid inclusion is exposed or not in between polishing. The exposed fluid inclusion on the sample surface was coated with conductive Au thin film using a cryo-coating system consists of an Au ion-coater (SANYU DENSHI, SC-701AT) in a glove box. The polished sample was settled on the bottom of deep well of a cryo-coating stage for the coater. The sample and the cryo-coating stage were cooled by liquid nitrogen before coating to prevent sublimation of the frozen fluid in the vacuum coating chamber. The temperature of the sample maintained under -120 °C after the coating process. The Au coated sample was set into a sample holder of Cameca ims-1270/1280 milled from a stainless-steel block. The machined sample holder is useful to suppress deformation with temperature change until liquid nitrogen temperature. The sample holder was put into a cryo-transfer vessel filled with liquid nitrogen and carried to the Cameca ims-1270/1280. The vessel has a connection port compatible to the loadlock chamber of the Cameca ims-1270/1280 for sample introduction. We attached a valve to the loadlock chamber in order to keep the frozen sample in nitrogen gas atmosphere. The sample holder was picked up from the vessel to the loadlock chamber and transferred to the cryo-stage of SIMS instrument cooled to -190 °C with liquid nitrogen. The liquid nitrogen cooled the cryo-stage supplied by a roots pump which was adopted to reduce the sample stage vibration.

Isotope Analysis: Oxygen isotopic compositions of synthesized fluid inclusions in halite crystal were measured with the Camaca ims-1280 SIMS in Hokkaido University using the cryogenic sample preparation techniques developed in this study. A Cs^+ primary ion beam was focused on the frozen fluid inclusion. A normal incidence electron flood gun was applied to compensate positive charging. Entrance slit and exit slit was 60 μm and 180 μm, respectively. We obtained mass spectrum of oxygen isotopes from the H_2O-ice. Intensity of $^{16}O^-$ peak was about 2.8×10^5 cps and a tale was observed symmetrically at both side of the peak below 3 cps. Three oxygen isotopes were collected using peak jumping mode: $^{16}O^-$ was measured on a Faraday cup, $^{17}O^-$ and $^{18}O^-$ were measured with an axial electron multiplier. Total count rate of $^{16}O^-$ was ~5–6×10^6 cps. We obtained the reproducibility of ±2.8 ‰ and ±5.2 ‰ for $\delta^{18}O$ and $\delta^{17}O$ when the calculated statistical errors were ±1.8 ‰ and ±3.8 ‰, respectively. The $\delta^{18}O$ deviation was suppressed successfully in this study. The interference of tail from $^{16}OH^-$ to $^{17}O^-$ was estimated to be about 10 ‰ because the intensity of $^{16}OH^-$ was comparable to $^{16}O^-$ peak. Oxygen isotope analysis using $^{16}OH^-$ and $^{17,18}OH^-$ signal would be effective for further high precision oxygen isotope analysis for H_2O ice as well as the tail correction method used in [2].

References: [1] Zolensky M. et al. (1999) *Science*, 285:1377−1379. [2] Yurimoto H. et al. (2014) *Geochemical Journal*, 48:549−56.