HALOGEN EVOLUTION IN THE EARLY SOLAR SYSTEM: DIFFUSION OF HALOGENS DURING CHONDRULE FORMATION

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Introduction: In the nascent solar nebula, halogens were likely present as H-[F,Cl,Br,I] gas. As the nebula cooled, halogen bearing phases condensed, with less refractory phases found further from the Sun. Estimates of 50 % condensation temperature (Tc) for the halogens at 10^{-4} bar range from 390 °C for K-Iodide to 674 °C for F-apatite [1,2]. Models for chondrule formation call for a later period of intense but transient heating: it is this later heating and cooling step that determines the concentrations of halogens found in chondrule mesostasis. We present new SIMS and EPMA data on halogens in mesostasis glass in chondrules from enstatite chondrites, as well as complementary experiments to understand halogen behavior during chondrule melting. We are investigating the behaviour of halogens in chondrule melts, so that we can assess whether halogen concentrations observed in chondrules are representative of the halogen content of chondrule precursor solids. Following initial melting, halogens will simultaneously be concentrated into the melt during crystallization, as halogens are incompatible in most silicate phases; and halogens will diffuse out of the chondrule as well as potentially condensing into the chondrule as it cools.

Methods: We analysed 14 chondrules from Sahara (SAH) 97072 (EH3), Larkman Nunatak (LAR) 12156 (EH3), LAR 12252 (EH3), Elephant Moraine (EET) 90012 (L4), Dominion Range (DOM) 14021 (EH3) and Qingzhen (EH3). Glass compositions, including Cl, were measured using a Cameca SX5 FEG electron probe at the University of Oxford. Halogen (F, Br, Cl) and sulfur concentrations were measured using a Cameca IMS 1280-HR SIMS at CRPG, Nancy, France. We experimentally examine halogen behaviour in silicate melt at 1×10^{-4} and 0.5 GPa, with initial total halogen concentrations of 0.5-5 wt. %. We chose a bulk composition to approximate EC mesostasis, doped with B and Zn to help retain Cl [3]. We will use the experimental glass as standards.

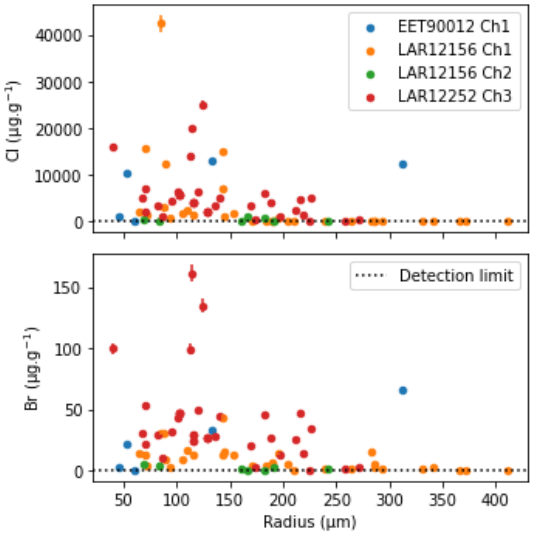

Figure 1: Concentration of Cl and Br vs radius in glass from four EC chondrules. Cl and Br show decreases towards the edge of chondrules.

Results: We looked at 14 chondrules, with typical glass abundance of 5 vol.%. The maximum concentration of Cl found by EPMA was 5.3 wt.%. Four of the chondrules are regulary shaped, with mesostasis distributed at all radial distances: these were analysed in detail to get radial data. For two of the four, our SIMS data show a concentration gradient with increasing Cl and Br contents from the surface to the core of chondrules (Figure 1). The maximum and minimum concentrations of halogens in all chondrules are: F, 1000 - 152 µg.g^{-1}; Cl, 42000 – 11 µg.g^{-1}; Br, 161 – 0 µg.g^{-1}; S 20 – 18 0000 µg.g^{-1}, with some points up to 80 0000 µg.g^{-1}. Fluorine shows no trend with radius. Br and Cl are well correlated for all chondrules. Neither Br nor Cl are correlated with F or S. In the 1 atm experiments (30 minutes at 1400 °C) preliminary results show that the concentrations of all halogens in run products were below detection limit of SEM EDS.

Discussion: Halogen concentrations in the centres of chondrules represent minimum initial concentrations. The Cl and Br concentration gradients in some chondrules indicate Cl and Br loss. We can evaluate the possibility that this gradient represents by diffusion-controlled evaporative loss during chondrule melting. The characteristic timescales of diffusion of halogens in glass are rapid (~2 h for Cl in a 2 mm diameter chondrule at 1400 °C [4]). For a concentration gradient to be 'frozen in', the timescale of cooling must have been similarly rapid. Given the difficulty in retaining halogens in the low-pressure analogue experiment, we suggest an elevated halogen partial pressure, analogous to the high partial pressure suggested for Na [4] was needed to retain halogens in the EC chondrule-forming region.

[1] B.J. Wood, D.J. Smythe, T. Harrison, *American Mineralogist*, 2019, **104**, 844. [2] H. Palme, K. Lodders, A. Jones, in *Treatise on Geochemistry: Second Edition*, 2013, **2**, 15. [3] B.J. Riley *et al.*, *Journal of Nuclear Materials*, 2014, **452**, 178. [4] C.M.O.D. Alexander *et al.*, *Science*, 2008, **320**, 1617.