Quantitative Halogen Analysis in Carbonaceous Chondrites and Exploration of their Thermal History by I-Xe Dating

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Quantifying halogen abundances in carbonaceous chondrites, crucial for understanding processes such as water-rock interactions and thermal metamorphism, is challenging due to their low halogen concentrations (ppb-ppm levels). Recent analytical methods, including Radiochemical Neutron Activation Analysis (RNAA) and Neutron Irradiation and Noble Gas Mass Spectrometry (NI-NGMS), have shown discrepancies in determining halogen concentrations in bulk meteorites [1, 2]. Possible causes for this discrepancy include i) differences in methodology, ii) variations in sample preparation, including contamination, loss, or the sample form (powder or small chip), and iii) sample heterogeneity. Clay et al [2] reported extremely low halogen concentrations of Allende using NI-NGMS. Because their sample volume is very small (0.7 mg of powder), the discrepancy in results is likely due to iii) sample heterogeneity. To verify this, it is important to analyze the halogen concentrations of several larger samples using NI-NGMS and compare the results. Here, we determined halogen concentrations in small chips and powder samples from Allende using NI-NGMS and compared the results with previous studies to investigate potential causes (i-iii). In addition, attempts were made to determine halogen (Cl, Br, I) concentrations and I-Xe ages in small chips from several carbonaceous chondrites to gain insight into the distribution of extracted halogens at different temperature steps, and the timing of thermal events on their parent bodies.

For halogen analysis, 3 g of Allende whole rock samples were crushed and sieved to produce four powder fractions (>150 μm in diameter) ranging from 1.4 to 17.1 mg. In addition, two small chip samples of 15.9 and 33.1 mg were used for analysis. For halogen analysis and I-Xe dating, additional small chips (< 3mm) were prepared for carbonaceous chondrites (NWA 801 34.5 mg, Aguas Zarcas 30.4 mg, Jbilet Winselwan 23 mg, Tagish Lake 29.9 mg). All samples were irradiated with neutrons at the Kyoto University Research Reactor. Noble gas isotope analyses (Ar, Kr, Xe) were performed using a modified VG3600 mass spectrometer at the University of Tokyo [3]. The noble gases were extracted by stepwise heating the samples between 500 and 1800 ℃.

The halogen concentrations obtained from Allende were consistent with each other regardless of the sample form (powder or chip): Cl = 217–357 ppm, Br = 1119–1883 ppb, I = 73–105 ppb, which were in agreement with those in some previous studies regardless of the method of analysis [1, 4]. However, the small chip sample of 33.1 mg showed significantly higher concentrations for all halogens. Extraction of this sample's high-temperature fraction (above 1200 degrees) to distinguish meteorite-intrinsic halogens from terrestrial contamination resulted in Br/Cl and I/Cl ratios that deviated even more from the literature values [2]. This suggests that the discrepancy is not due to i) methodological differences or ii) contamination but is due to iii) sample-derived heterogeneity.

All I-Xe ages yielded upper limits around 4.5 Ga, except for Allende showing an age of 4558 ±4 Ma, NWA801 at 4491 ±19 Ma, and Jbilet Winselwan at 4517 ±15 Ma. The age obtained in Allende reflects thermal conditions around 4558 Ma, consistent with thermochronological modeling indicating that temperatures of about 400 ℃ persisted around this age [5]. The 4517 ±15 Ma obtained in Jbilet Winselwan is younger than the timing of aqueous alteration 4564.7 ±1 Ma [6], suggesting that a post-aqueous alteration thermal disturbance may have occurred around this age.

References: [1] Ozaki and Ebihara (2007), [2] Clay et al. (2017), [3] Kobayashi et al. (2019), [4] Lodders and Fegley Jr. (2023), [5] Carporzen et al. (2010), [6] Fujiya et al. (2022)

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