RUTHENIUM ISOTOPE FRACTIONATION DURING CRYSTALLIZATION OF PLANETESIMAL CORES
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Introduction: Magmatic iron meteorites are thought to be fragments of the metallic cores of differentiated planetesimals because their chemical compositions can largely be accounted for by crystal-liquid partitioning during fractional crystallization of metallic magma [e.g., 1]. Fractional crystallization may have induced stable isotope fractionation between early- and late-crystallized irons of a given core. However, such systematic mass-dependent isotope fractionations for magmatic irons have not been investigated in detail yet. Here we report Ru stable isotope data for a comprehensive set of magmatic iron meteorites. These data are used to examine the effect of core crystallization on the isotopic composition of iron meteorites, and to further constrain the formation and crystallization history of diverse groups of magmatic iron meteorites.

Samples and Methods: We developed a $^{98}$Ru-$^{101}$Ru double spike for the precise measurement of mass-dependent Ru isotope variations [2] and obtained Ru isotope data for a set of 36 magmatic iron meteorites from the five major groups (IIAB, IID, IIIAB, IVA, IVB). The samples were selected to cover most of the crystallization sequence within each group. We also analyzed four ordinary chondrites to assess the Ru isotopic composition of undifferentiated material. Iron meteorite samples (0.05 to 0.25 g) were spiked with the $^{98}$Ru-$^{101}$Ru double spike before digestion using 20 ml of reverse aqua regia in closed Teflon beakers at 120°C for 24 hours. Ordinary chondrites (0.5 g) were spiked prior to Carius tube digestion using reverse aqua regia at 220°C for 48 hours. After digestion, Ru was separated from the sample matrix using a combination of cation exchange chromatography and a Savillex™ PFA distillation unit [2]. The Ru isotope measurements were conducted using a Neptune Plus MC-ICPMS at Münster. The data are reported as $\delta^{102/99}$Ru values relative to an Alfa Aesar standard. The external reproducibility (2 s.d.) of the Ru isotope measurement is 0.05 % for $\delta^{102/99}$Ru, as determined by repeated measurements of terrestrial rock standards doped with Ru.

Results and Discussion: The investigated iron meteorites exhibit large Ru stable isotope variations of up to almost ~1 %. For samples of groups IIAB, IIIAB and IVB, the Ru isotopic composition becomes increasingly heavier with decreasing Ru concentrations, consistent with isotope fractionation during fractional crystallization of metallic magma. Our data show that solid metal is isotopically light compared to the melt they crystallize from, leading to a progressive enrichment of heavy Ru isotopes in the residual melt. The magnitude of the Ru isotope fractionations observed within each group is inversely correlated with the initial S content inferred for each core (IIAB > IIIAB > IVB) [3], indicating that the isotope fractionation factor between solid and liquid metal becomes smaller at higher liquidus temperatures of the core. For group IID and IVA irons Ru isotope fractionations are not as clearly correlated with Ru concentrations as for the other groups. Several IVA iron meteorites display increasingly heavy Ru isotope compositions with decreasing Ru content, but this correlation shows considerable more scatter than is observed for the IIAB, IIIAB and IVB groups. The IID irons do not show any correlation between Ru isotope composition and Ru concentration, although the entire spectrum from early- to late-crystallized samples was analyzed. Thus, both the IVA and IID data cannot be explained by simple fractional crystallization of one homogeneous metallic magma, indicating that the crystallization of these cores was more complex than those of the IIAB, IIIAB and IVB irons. Of note, elemental trends in group IVA and IID irons are also less consistent with simple fractional crystallization than trends in the other groups [4, 5]. The Ru isotopic composition of early-crystallized IIAB and IIIAB irons is similar to that of ordinary chondrites, indicating that the IIAB and IIIAB parent bodies have an ordinary chondrite-like stable Ru isotope composition. However, the early-formed IVB irons already exhibit a heavier Ru isotopic signature than ordinary chondrites. Consequently, the IVB parent body seems to have a non-chondritic Ru stable isotope composition. This fractionated composition probably results from solar nebula processes acting on the precursor material of the IVB parent body. This is consistent with non-chondritic relative abundances of highly siderophile elements inferred for the IVB core, which has also been attributed to fractionation by solar nebula processes [6].

Conclusion: Ruthenium stable isotopic signatures of magmatic iron meteorites are generally consistent with formation by fractional crystallization of metallic magma, but also reveal differences in the solidification history of iron meteorite parent bodies. Whereas the data for IIAB, IIIAB and IVB irons are consistent with simple fractional crystallization of their parental cores, the formational histories of IID and IVA irons were more complex.