

ENDEMIC Ru ISOTOPIC ANOMALIES IN IRON METEORITES AND IN ALLENDE. J. H. Chen^{1,2}, D. A. Papanastassiou^{2,1}, and G. J. Wasserburg¹. ¹The Lunatic Asylum, Division of Geological and Planetary Sciences, M/C 170-25, Caltech, Pasadena, CA 91125. ²Earth and Space Sciences Division, M/S 183-335, Jet Propulsion Laboratory, Caltech, 4800 Oak Grove Drive, Pasadena, CA 91109-8099 (jchen@gps.caltech.edu).

Small variations for Mo isotopes have been observed recently in the Allende meteorite and in iron meteorites, mesosiderites, and pallasites, using ICP-MS [1]. Large effects for Mo have been reported for leaches of Orgueil [2] and in SiC and graphite from Murchison [3-5]. Variations for Mo in bulk Allende and in Murchison have also been presented by NTIMS [6]. Effects in Ru isotopes can define further the preserved exotic r, s, and p contributions in this mass region, and possible effects in ⁹⁸Ru and ⁹⁹Ru from ⁹⁸Tc (4.2 Ma half-life) and ⁹⁹Tc (0.21 Ma half-life). Previous attempts at determination of Ru isotopes yielded no resolved effects [7-10]. The present work represents a substantial improvement in precision over the earlier work.

We have developed chemical and mass spectrometric analytical techniques to determine the Ru isotope compositions in terrestrial standards and in meteorites. Because the Mo isotope interferences can affect ⁹⁶Ru and ⁹⁸Ru, we have used ⁹⁹Ru/¹⁰¹Ru for the mass fractionation correction and applied the exponential law. Through improved chemical separations, we have reduced the Mo interference to ⁹⁸Mo/⁹⁸Ru < 0.00002. All measurements were obtained in the new mass spectrometry laboratory at JPL, using the ThermoFinnigan Triton, in static mode. The results on terrestrial standards of different chemical composition show external precision (reproducibility) of isotope ratio measurements to better than 0.3ε (2σ) for ¹⁰⁰Ru/¹⁰¹Ru, 0.6ε for ¹⁰²Ru/¹⁰¹Ru and ¹⁰⁴Ru/¹⁰¹Ru, and 1ε to 2ε for ⁹⁶Ru/¹⁰¹Ru and ⁹⁸Ru/¹⁰¹Ru. The range of reproducibility for normal Ru is shown as dashed lines in Fig. 1.

The results are shown in Fig. 1. The y-axis lists the sample sequence number. Ordinary chondrites measured are: Olivenza (LL5, #1 & #2), Portales Valley (H4, #3), Ransom (H4, #4), Garnet (H4, #5). These ordinary chondrites plot within the error envelopes of the terrestrial standards for all Ru isotopes. The minor isotopes ⁹⁶Ru and ⁹⁸Ru, show slightly larger uncertainties and scatter, but no anomalies. In contrast, analyses of two whole rock samples of the Allende carbonaceous chondrite (#6-7) show hints of enrichments in ⁹⁶Ru, ⁹⁸Ru and ¹⁰⁴Ru and well-resolved depletions in ¹⁰⁰Ru of 1.2 and 1.6 ε. The measurements on Allende were obtained after the ordinary chondrite and iron meteorite analyses. Iron meteorite metal from Canyon Diablo (IA, #8) and Pitts (IB, #9) show normal values. For three IIAB irons, Bennett County (#10), Negrillos (#11) and Coahuila (#12), all Ru isotopes except ¹⁰⁰Ru, show normal abundances. These samples show well-

resolved depletions in ¹⁰⁰Ru/¹⁰¹Ru, from 0.5±0.14 to 0.75±0.09 ε. Similar results were obtained for IIIAB irons, Cape York (#13), Grant (#14), Acuna (#15), Bella Roca (#16) and Tres Castillos (#17). They show ε₁₀₀ ranges also from -0.5±0.08 to -0.76±0.21. Gibeon (IVA, #18) yields ε₁₀₀ = -0.40±0.06. Multiple analyses of Hoba (IVB, #19-21) confirm the presence of a larger ε₁₀₀ effect (-1.08±0.11 ε). Tlacotepec (IVB, #22) and Pinon (An, #23) also show ε₁₀₀ effects close to -1 ε. Analyses of 3 pallasites, Salta (#24), Springwater (#25) and Thiel Mountain (#26) yield ε₁₀₀ ranges from -0.4±0.08 to -0.58±0.05. Only Thiel Mountain plots outside of the error envelopes and shows a resolvable effect. In summary, we have found endemic isotope anomalies in Ru. For the normalization used, we observe up to 1 ε depletion in the pure s-process isotope, ¹⁰⁰Ru in groups IIAB, IIIAB, IVAB and An irons and possibly in a pallasite. Group IAB irons and ordinary chondrites show a normal Ru composition. The Allende WR samples show larger ε₁₀₀ effects and hints of added effects in ⁹⁶Ru, ⁹⁸Ru and ¹⁰⁴Ru. The presence of Ru isotopic effects is clearly resolved, but the attribution of the isotope anomalies to specific isotopes depends on the choice of normalization for isotope fractionation.

In Fig. 2, we use the Hoba data to demonstrate the effect of different mass fractionation normalizations:

Case 0. For the normalization to ⁹⁹Ru/¹⁰¹Ru (diamonds), we obtain normal Ru isotope ratios except for well-resolved deficits in ¹⁰⁰Ru/¹⁰¹Ru (ε₁₀₀ = -1.08±0.11).

Case 1. Rotating the isotope pattern, by normalization to ¹⁰⁰Ru/¹⁰¹Ru (triangles) yields large enrichments (5.9 and 3.2 ε) in the two p-process isotopes (⁹⁶Ru and ⁹⁸Ru) and a +2.16 ε effect in r- & s-process ⁹⁹Ru. It also yields large depletions in the r- & s-process ¹⁰²Ru (-1.3 ε) and in the pure r-process ¹⁰⁴Ru (-2.7ε). This normalization would require the presence of both p- and s-process excesses for Ru.

Case 2. Normalization to ⁹⁹Ru/¹⁰⁰Ru (squares) yields depletions in ⁹⁶Ru and ⁹⁸Ru and enrichments in ¹⁰²Ru and ¹⁰⁴Ru.

Case 3. Finally, the Hoba Ru data (in the ⁹⁹Ru/¹⁰¹Ru normalization) were re-calculated by adding Ru with an isotope composition obtained from s-process calculations [11], with the aim to reduce the observed deficit for the s-process only ¹⁰⁰Ru to zero. In this Case, after addition of s-process Ru and renormalization using ⁹⁹Ru/¹⁰¹Ru, other Ru isotopes also show close to normal values (circles, Fig. 2).

The Ru data in iron meteorites can be explained more simply by a small ($\sim 10^{-4}$) depletion in s-process nuclei (Case 3). Case 1 suggests that short-lived nuclei ^{98}Tc and ^{99}Tc could have been alive when Hobas (and other irons) was formed. The relative abundances of $^{99}\text{Tc}/^{101}\text{Ru}$ in AGB stars predicted by the instant injection and mixing model is $\sim 2.9 \times 10^{-5}$ [12]. The predicted value is about a factor of 3 lower than that in Hobas. However, any intervening time interval for the formation of the differentiated planetary bodies (iron meteorites) in which the Ru effects are found would make the presence of live ^{99}Tc effects highly unlikely. In comparison, Mo isotopes ($^{96}\text{Mo}/^{98}\text{Mo}$ normalization) in iron meteorites show mainly effects in two pure p-process nuclei, ^{92}Mo (up to 2.9 ϵ) and ^{94}Mo (up to 1.8 ϵ) [1]. In irons, there is no resolvable effect in ^{100}Mo [1], which is a pure r-process nuclide. Large p- & r-process excesses in Mo isotopes were reported for Allende and Murchison [1, 6]. This is in sharp contrast with the p- & r-process depletion pattern (or an s-process enrichment pattern) in a mainstream SiC grain from Murchison [3].

In meteorites, the Re/Os ratios range from 0.06 to 0.13. If Tc behaves like Re in meteorites, then we expect only a factor of ~ 2 fractionation in Tc/Ru in meteorites, which would not explain the observed range in ϵ_{99} . However, the chemical behavior of Tc and Ru in the early solar nebula needs to be carefully addressed. Our current Ru results on iron meteorites did not show any correlation with ^{107}Pd as inferred from excess in ^{107}Ag in iron meteorites and pallasites [13]. This is consistent with the short half-life of ^{99}Tc and the much smaller expected chemical fractionation for Tc/Ru relative to the volatility-controlled, large Pd/Ag fractionation.

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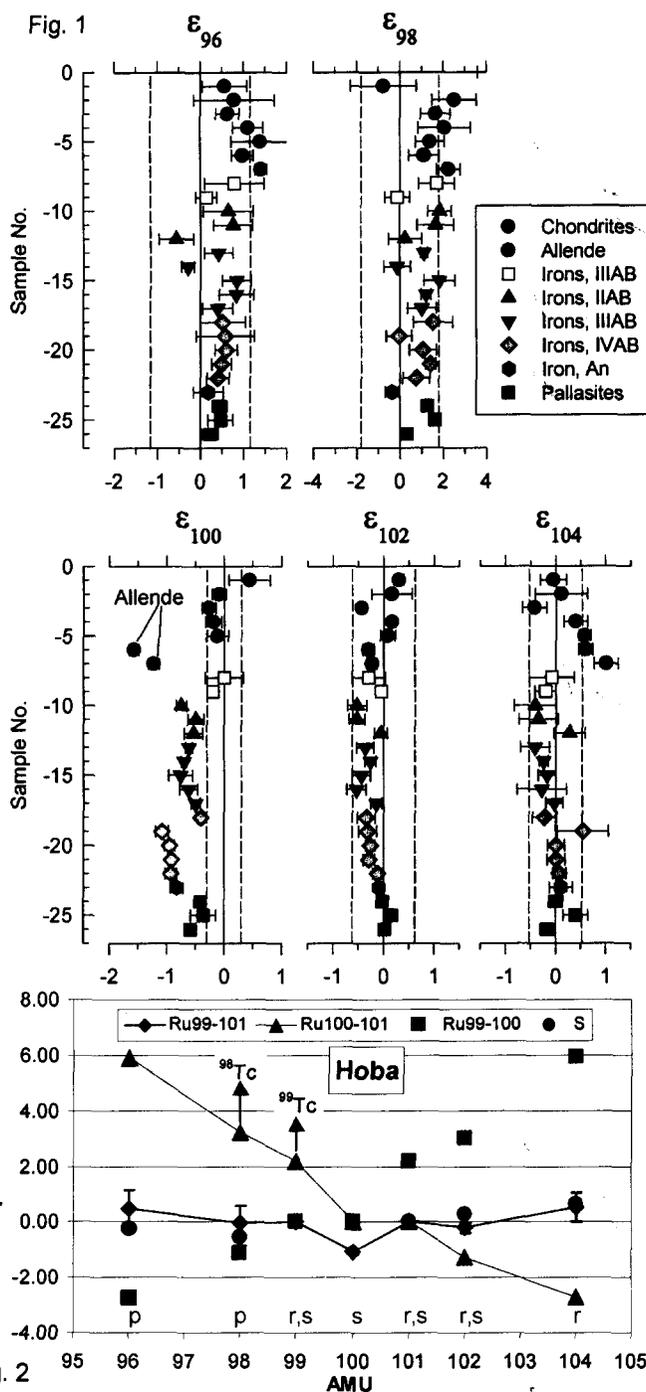


Fig. 2

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