ENDEMIC Ru ISOTOPIC ANOMALIES IN IRON METEORITES AND IN ALLENDE. J. H. Chen[1, 2], D. A. Papanastassiou[2, 1], and G. J. Wasserburg[1]. [1]The Lunatic Asylum, Division of Geological and Planetary Sciences, M/C 170-25, Caltech, Pasadena, CA 91125. [2]Earth 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 ^{98}Ru and ^{99}Ru from ^{98}Tc (4.2 Ma half-life) and ^{99}Tc (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 ^{96}Ru and ^{98}Ru, we have used ^{99}Ru/^{101}Ru for the mass fractionation correction and applied the exponential law. Through improved chemical separations, we have reduced the Mo interference to ^{98}Mo/^{98}Ru<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 0.3εu (2σ) for ^{100}Ru/^{101}Ru, 0.6εu for ^{102}Ru/^{101}Ru and ^{104}Ru/^{101}Ru, and 1εu to 2εu for ^{96}Ru/^{101}Ru and ^{98}Ru/^{101}Ru. 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 ^{96}Ru and ^{98}Ru, 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 ^{96}Ru, ^{98}Ru and ^{104}Ru and well-resolved depletions in ^{100}Ru of 1.2 and 1.6 εu. 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 ^{100}Ru, show normal abundances. These samples show well-resolved depletions in ^{100}Ru/^{101}Ru, from 0.5±0.14 to 0.75±0.09 εu . Similar results were obtained for IIIAB irons, Cape York (#13), Grant (#14), Acuna (#15), Bella Roca (#16) and Tres Castillos (#17). They show ε_{100} ranges also from -0.5±0.08 to -0.76±0.21. Gibeon (IVA, #18) yields ε_{100} = -0.40±0.06. Multiple analyses of Hoba (IVB, #19-21) confirm the presence of a larger ε_{100} effect (-1.08±0.11 εu). Tlacotepec (IVB,#22) and Pinon (An, #23) also show ε_{100} effects close to –1 εu. Analyses of 3 pallasites, Salta (#24), Springwater (#25) and Thiel Mountain (#26) yield ε_{100} 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 εu depletion in the pure s-process isotope, ^{100}Ru 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 ε_{100} effects and hints of added effects in ^{96}Ru, ^{98}Ru and ^{104}Ru. 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 ^{99}Ru/^{101}Ru (diamonds), we obtain normal Ru isotope ratios except for well-resolved deficits in ^{100}Ru/^{101}Ru (ε_{100}=-1.08±0.11).

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

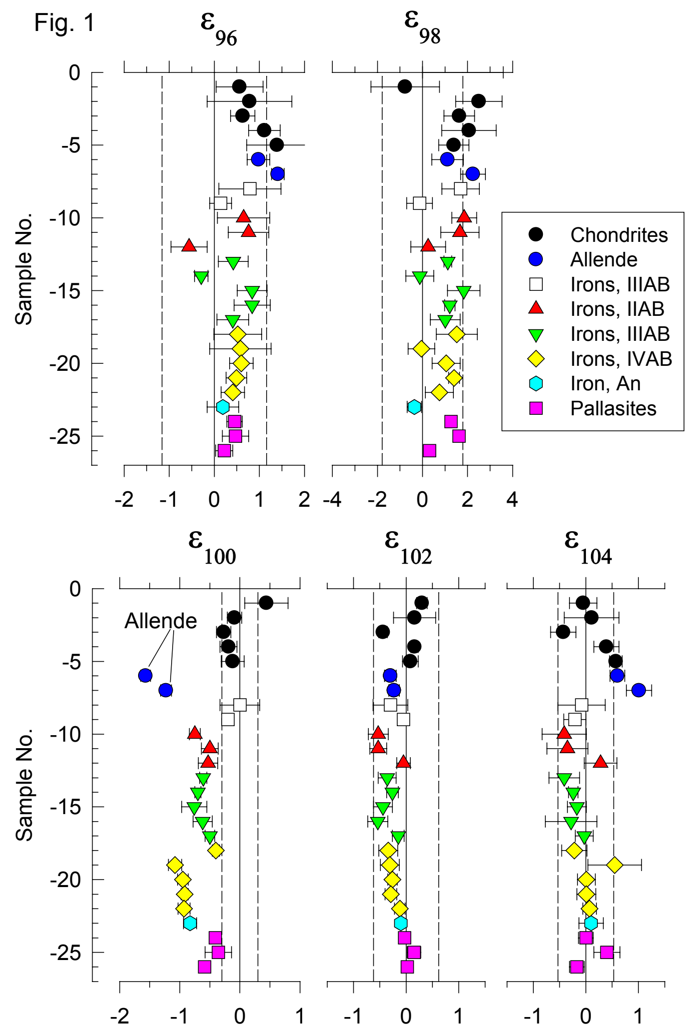
Case 2. Normalization to ^{99}Ru/^{100}Ru (squares) yields depletions in ^{96}Ru and ^{98}Ru and enrichments in ^{102}Ru and ^{104}Ru.

Case 3. Finally, the Hoba Ru data (in the ^{99}Ru/^{101}Ru 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 ^{100}Ru to zero. In this Case, after addition of s-process Ru and renormalization using ^{99}Ru/^{101}Ru, 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 (~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 Hoba (and other irons) was formed. The relative abundances of ^{99}Tc/^{101}Ru in AGB stars predicted by the instant injection and mixing model is ~2.9×10^{-5} [12]. The predicted value is about a factor of 3 lower than that in Hoba. 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}Mo/^{98}Mo normalization) in iron meteorites show mainly effects in two pure p-process nuclei, ^{92}Mo (up to 2.9 εu) and ^{94}Mo (up to 1.8 εu) [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.

Fig. 1



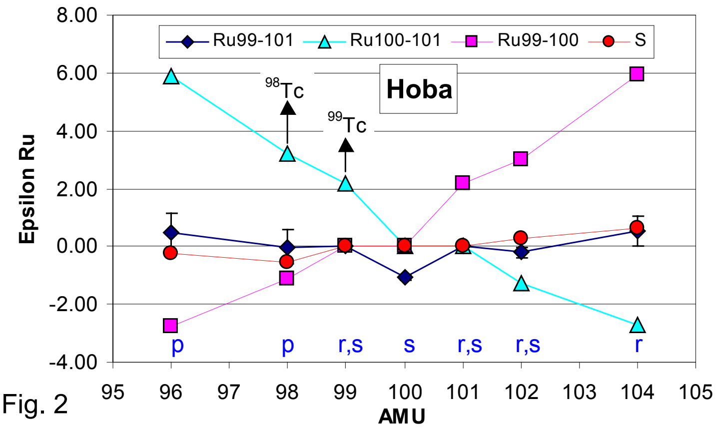


Fig. 2

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