EXTINCT TECHNETIUM IN PRESOLAR GRAINS.

M. R. Savina[1], A. M. Davis[2,3], C. E. Tripa[1,2], M. J. Pellin[1], R. Gallino[4], R. S. Lewis[2], and S. Amari[5], [1]Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, [2]Enrico Fermi Institute and [3]Department of the Geophysical Sciences, University of Chicago, Chicago, IL 60637, [4]Dipartimento di Fisica Generale, Università di Torino and Sezione INFN di Torino, I-10125 Torino, Italy, [5]Laboratory for Space Sciences and the Physics Department, Washington University, St. Louis, MO 63130,

Introduction: Presolar grains carry isotopic records of the nuclear processing in their parent stars. Their analysis, combined with appropriate stellar models, yields insights into stellar evolution and nucleosynthesis. The majority of SiC grains (mainstream) were produced in outflows from low-mass (~1.5 to 3 $M_\odot$) asymptotic giant branch (AGB) stars [1]. In AGB stars, elements heavier than Fe are synthesized by slow neutron capture (the *s*-process). The isotopic compositions of Zr, Mo, Sr, and Ba in mainstream SiC grains show the *s*-process signatures predicted by models of low-mass AGB stars from which SiC condenses [2].

Merrill's spectroscopic observation of Tc in certain red giants (later recognized as AGB stars) more than 50 years ago [3] proved that *s*-process nucleosynthesis is ongoing in those stars. Since then, Tc has been detected in giants of types MS, S, and C (stars at progressively later stages along the AGB) [4, 5]. The ^{99}Tc/^{99}Ru decay couple ($T_{1/2} = 2.1\times10^5$ y) has been investigated in non-presolar meteoritic materials as a means of discovering the timescales of processes such as planetary differentiation and the injection of *s*-process material into the protosolar nebula, however only very small (of order one ε-unit) anomalies have been found [6-9]. Deviations of ~0.5 to 1.5ε in Ru isotopes in several classes of meteorites have been interpreted as depletions in *s*-process Ru due to incomplete mixing of SiC with other protosolar material [9].

The *s*-process in AGB stars produces Ru and Tc isotopes with a distinct isotopic signature that should be preserved in SiC. Because the half-life of ^{99}Tc is comparable to the AGB stage of stellar evolution, live ^{99}Tc should have been present in the grains when they formed, and should be detectable as excess ^{99}Ru when compared with *s*-process models.

Experimental: Ruthenium isotopic measurements were made by resonant ionization mass spectrometry (RIMS) [10] on 19 grains from the Murchison meteorite (fraction KJG, mount CHRL108, average grain size 3 μm) [11]. Two of the grains were known from previous C, N, and Si isotopic analysis to be mainstream SiC. The RIMS scheme was $[\mathrm{Kr}]4d^75s\ (^5\mathrm{F}) \rightarrow [\mathrm{Kr}]4d^65s5p\ (^5\mathrm{F})$ at 287.583 nm, followed by a 403.983 nm transition to an autoionizing state. Isobaric interferences from non-resonant ionization of molecules were present at masses 96 and 98 in most grains, but the other Ru isotopes were unaffected.

Results and Discussion: Fig. 1 shows the Ru isotopic composition for a SiC grain that has the *s*-process pattern expected from low-mass AGB stars. The *p*-process isotopes ^{96,98}Ru (which had little or no isobaric interference in this particular grain) and the mostly *r*-process ^{104}Ru are strongly depleted compared to the *s*-process-only ^{100}Ru, while the mixed *s*- and *r*-process isotopes show intermediate depletions. This pattern is similar to that seen in Mo in presolar SiC [2].

Heavy element (>Fe) isotopic compositions in mainstream SiC grains have been successfully reproduced by models of low-mass thermally pulsing AGB stars [2]. In these stars, heavy nuclides are built up from lighter ones by slow neutron capture. The main source of neutrons is the $^{13}\mathrm{C}(\alpha,n)^{16}\mathrm{O}$ reaction occurring periodically in a thin He-rich shell between the C/O-rich core and the H-rich envelope of the star, specifically in a small region known as the ^{13}C pocket. Neutrons provided by brief ignitions of the $^{22}\mathrm{Ne}(\alpha,n)^{25}\mathrm{Mg}$ reaction during thermal pulses also play a role. Mo, Tc, and Ru nuclides in the He shell capture neutrons, then mix with envelope material to convert the initial Ru isotopic composition to the pattern seen in Fig. 1. The apparent depletions in the *p*-, *r*-, and mixed *s*- and *r*-process Ru isotopes are actually the result of strong production of the *s*-process-only ^{100}Ru (overproduction factor = 12.7 for a 1.5 $M_\odot$ star) compared to the weaker production of ^{99}Ru, ^{101}Ru, ^{102}Ru, and ^{104}Ru, and destruction of ^{96}Ru and ^{98}Ru (~2-4% of the amount initially present in the He shell and envelope). At the end of the AGB phase, all or nearly all of the envelope is lost to the interstellar medium (ISM) in the form of gas and dust.

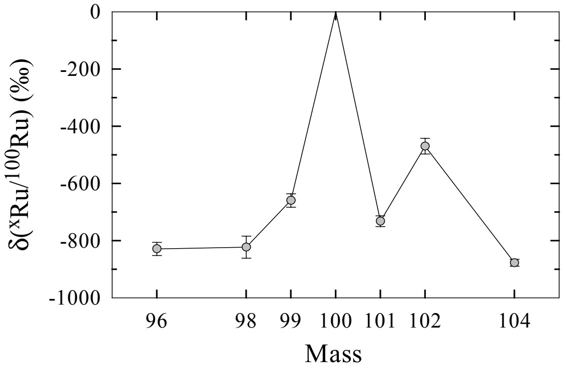

Figure 1: Ru isotopic composition in SiC grain L2 from Murchison indexed to the *s*-only isotope ^{100}Ru and normalized to a Ru terrestrial standard. Error bars are 2σ.

Data for all 19 grains is plotted in Fig. 2, along with the predictions of a 1.5 M$_\odot$ AGB model with initially solar elemental and isotopic composition. There is excellent agreement for ^{101}Ru, but the predictions for ^{99}Ru and ^{102}Ru differ from the measurements. In the case of ^{102}Ru, the difference can be accounted for by the error induced in the calculated values by uncertainties in neutron capture cross-section of ^{102}Ru [12]. In contrast, the measured δ^{99}Ru values lie far outside the model uncertainties, which change the predictions by less than 20‰ when included. Data and predictions for ^{99}Ru agree, however, assuming that ^{99}Tc condensed into the grains at the same Tc:Ru ratio as in the envelopes of the parent stars (~1:3). This implies that the grains formed from a gas whose Tc:Ru ratio matched that of the stellar envelope, and that the Tc and Ru did not fractionate from one another during the condensation process.

Because the half-life of ^{99}Tc is long compared to the timescale over which the ^{13}C pocket is active (2.1×10^5 years vs. a few tens of thousands of years for any given cycle), ^{99}Tc can be regarded as a stable *s*-process element while it is in the pocket. It is produced by β-decay of ^{99}Mo (which is in turn produced by neutron capture on ^{98}Mo). Some ^{99}Tc captures a neutron to form ^{100}Tc, which decays rapidly to ^{100}Ru. In contrast, all of the ^{99}Ru that passes through the ^{13}C pocket is consumed by neutron capture. Thus, all of the ^{99}Ru produced by AGB stars results from decay of ^{99}Tc either in the stellar envelope or in the ISM.

Assuming that grain formation times are on the order of a year or so after the mass loss event [13], the Tc:Ru ratio of the gas from which SiC condenses must match that of the stellar envelope. Thus, the points labeled ^{99}Ru on Fig. 2 represent unprocessed ^{99}Ru (^{99}Ru that never passed through the ^{13}C pocket) plus daughter ^{99}Ru from ^{99}Tc decay in the envelope, while the points labeled ^{99}Ru + ^{99}Tc represent this same material plus daughter ^{99}Ru from ^{99}Tc decay in the grains.

Our measurements show that ^{99}Tc was alive in the grains when they condensed, but not necessarily when the solar system formed. Evidence of live ^{99}Tc in the early solar system (ESS) must come from ^{99}Ru excesses in differentiated meteoritic materials. If the our models are correct, then mixing freshly-minted AGB material with the protosolar nebula just prior to (and perhaps triggering) its collapse would produce a ^{99}Ru excess of $< \sim 3\times10^{-5}$ [14], which is comparable to the precision of current measurements [8, 9]. Our data show that the models accurately predict the amount of ^{99}Tc ejected from AGB stars. Therefore, given that Tc/Ru fractionation in the ESS is expected to be minimal [8], it seems unlikely that evidence for live AGB-produced ^{99}Tc in the early solar system will be found.

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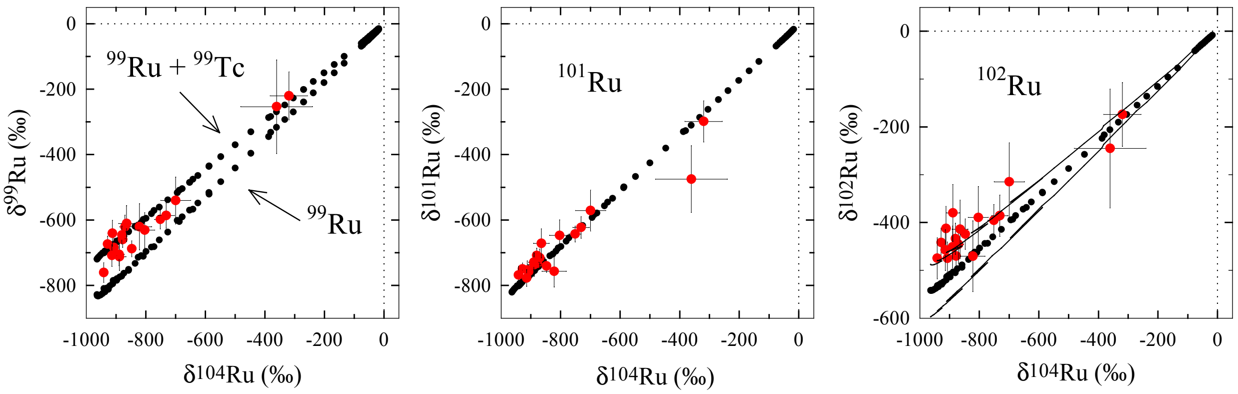

Figure 2: Three-isotope plots comparing presolar SiC grains (red circles) with predictions for a 1.5 M$_\odot$ AGB star (black circles). Error bars are 2σ. The δ^{99}Ru plot shows the values predicted by including *in situ* decay of ^{99}Tc in grains assuming a Tc:Ru condensation ratio of 1:1. The δ^{102}Ru plot shows the 2σ errors in the calculation (solid lines) caused by uncertainties in the neutron capture cross section of ^{102}Ru. Errors in the δ^{99}Ru predicted values (not shown) are approximately the width of the symbols.