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SUPERNOVA SOURCES AND THE ^{92}Nb-^{92}Zr *p*-PROCESS CHRONOMETER

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ABSTRACT

We report new Zr isotope evidence for live ^{92}Nb (mean life: $\bar{\tau}_{^{92}\mathrm{Nb}} = 52$ Myr) within the early solar system resulting in $(^{92}\mathrm{Nb}/^{93}\mathrm{Nb})_{\mathrm{initial}} \approx 10^{-3}$. The meteoritic minerals rutile and zircon have, respectively, very high and very low Nb/Zr ratios and are ideal for exploring the ^{92}Nb-^{92}Zr chronometer. Rutiles exhibit high positive $\epsilon_{^{92}\mathrm{Zr}}$ (~14–36) while a zircon has a negative $\epsilon_{^{92}\mathrm{Zr}}$ (~−4), as would be expected if ^{92}Nb was live in the early solar system. The meteoritic rutiles appear to be young, with apparent times of formation of ~80–220 Myr subsequent to the origin of the solar system. The initial ^{92}Nb/^{92}Mo for the solar system is broadly compatible with a model of uniform production if the ^{92}Nb/^{92}Mo production ratio for Type II supernova (SNII) sources with neutrino-driven winds is used. Data for all the now extinct *p*-process nuclides (^{92}Nb, ^{97}Tc, and ^{146}Sm) are consistent with these isotopes being derived by uniform production from SNII sources and a free decay interval of ~10 Myr. Consideration of a range of models indicates that the average *p*-process production ratio of ^{92}Nb/^{92}Mo needs to be at least in the range of 0.06–0.25.

Subject headings: ISM: abundances — nuclear reactions, nucleosynthesis, abundances — solar system: formation — supernovae: general

1. INTRODUCTION

Based on radiogenic excesses of their decay products in a variety of meteorites, it has been discovered that a number of now extinct short-lived nuclides were present in the early solar system. They provide important information both about chronometry and the nucleosynthetic sources of solar system material and are useful for constraining the timing of early solar system chemical differentiation processes. Recently, Busso, Gallino, & Wasserburg (1999) have shown that some short-lived nuclides (^{26}Al, ^{41}Ca, ^{60}Fe, and ^{107}Pd) are compatible with the contamination of the nebular cloud from which the Sun formed with a nearby asymptotic giant branch (AGB) star source, while the nuclides ^{146}Sm, ^{244}Pu, ^{235}U, ^{238}U, ^{232}Th, ^{182}Hf, ^{129}I, and ^{53}Mn may be understood in terms of uniform production (UP) from supernova sources.

Most model calculations of the *p*-process in supernovae predict too low abundance of the now extinct ^{146}Sm as well as stable ^{92}Mo. As a result of our recent advances in analytical techniques for isotopic measurements of Zr, Mo, and Ru, we have found the first evidence for extinct ^{97}Tc in the early solar system at the level of $^{97}\mathrm{Tc}/^{92}\mathrm{Mo} \sim 5 \times 10^{-5}$ (Yin & Jacobsen 1998). The presence of ^{146}Sm at a level of $^{146}\mathrm{Sm}/^{144}\mathrm{Sm} = 0.01$ is now well established (see Jacobsen & Wasserburg 1984 and many later studies). Like ^{97}Tc and ^{146}Sm, ^{92}Nb is a shielded "*p*-only" nuclide. It decays by electron capture to ^{92}Zr. Its potential as a cosmochronometer has long been recognized (Minster & Allègre 1982). Recently, a ^{92}Zr anomaly was reported in rutile from the Toluca iron meteorite and presented as the first evidence for live ^{92}Nb in the early solar system (Harper 1996). Here we report new Nb-Zr isotopic results (a preliminary report was given by Yin et al. 1999) on meteoritic mineral grains with Nb/Zr ratios that are both much higher and lower than the average solar Nb/Zr ratio.

2. SAMPLES, ANALYTICAL TECHNIQUES, AND DATA REPRESENTATION

Zircon ($ZrSiO_4$) is a rare mineral in meteorites. Since this mineral has a very low Nb/Zr ratio, it is ideal for Nb-Zr isotopic studies in that it may potentially record the initial solar system value of $^{92}\mathrm{Zr}/^{91}\mathrm{Zr}$. For this study, we analyzed zircon from the mesosiderite Chaunskij. Rutile (TiO_2) is another rare mineral in iron meteorites. The sample used for this study (Zagora) is an IAB iron meteorite with troilite-rich silicate inclusions. Three rutile grains (40–150 μm) found in the inclusion have concentrations of Nb ~ 9000 parts per million (ppm) and Zr ~ 5000 ppm and yield Nb/Zr ratios up to about 30 times the chondritic value.

A laser ablation system interfaced with a Micromass IsoProbe multicollector magnetic sector ICPMS (Inductively Coupled Plasma Mass Spectrometer) was used to determine the Zr isotopic composition in situ of mineral grains. The laser ablation system is the 193 nm light source that was jointly developed by Harvard (Horn, Rudnick, & McDonough 2000) and Merchantek. The array of Faraday cups allowed for the simultaneous static mode collection and integration of the masses of ^{90}Zr, ^{91}Zr, ^{92}Zr, ^{93}Nb, ^{94}Zr, ^{95}Mo, ^{96}Zr, and ^{97}Mo; ^{93}Nb was measured so that we could obtain simultaneously the Nb/Zr ratio. Both ^{95}Mo and ^{97}Mo were measured in order for us to monitor the potential Mo isobaric interferences to ^{92}Zr, ^{94}Zr, and ^{96}Zr. Such contributions were found to be negligible. Ion beam intensities for ^{90}Zr ranged from 0.7 to 8×10^{-11}, both for meteoritic and terrestrial rutiles and zircons, and depended on Zr content and laser spot size. For meteoritic rutiles, the signal lasted only 40–80 s because of the limited thickness of the thin section (~30 μm). The system was optimized and tuned with the terrestrial rutile and zircon standards. These standards were measured for their isotopic composition using a time-

TABLE 1
ISOTOPIC RESULTS

Sample	$f^{Nb/Zr}$	$\epsilon_{^{92}Zr}$	$^{92}Nb/^{93}Nb$	Time (Myr)
Zircon (Chaunskij)	−0.99968	−4.0 ± 2.1	(1.09 ± 0.57) × 10^{-3}	0
Rutile 1 (Zagora)	29.8 ± 0.3	24.1 ± 5.8	(2.20 ± 0.52) × 10^{-4}	83 ± 15
Rutile 2 (Zagora)	33.2 ± 0.2	20.3 ± 6.0	(1.67 ± 0.49) × 10^{-4}	97 ± 20
Rutile 3 (Zagora)	22.2 ± 0.7	26.1 ± 9.9	(3.20 ± 1.21) × 10^{-4}	64 ± 20
Rutile (Toluca)	158 ± 8	9.1 ± 1.7	(1.57 ± 0.29) × 10^{-5}	220 ± 16
Terrestrial rutile (LB-17)	...	0.0 ± 0.6	...	...
Terrestrial zircon (91,500)	−1	0.0 ± 0.4	...	...
Terrestrial baddeleyite	...	0.0 ± 0.4	...	...

NOTE.—An exponential mass fractionation correction was applied assuming $(^{90}Zr/^{91}Zr)_N \equiv 4.5845$. We obtained the following reference values for normal or average terrestrial Zr by our laser ablation ICPMS methods: $^{92}Zr/^{91}Zr = 1.53020$, $^{94}Zr/^{91}Zr = 1.55365$, and $^{96}Zr/^{91}Zr = 0.250156$. The Toluca rutile data are from Harper 1996.

resolved procedure; the gas blank was measured, then the laser was turned on, and the Zr, Nb, and Mo isotopes were measured by multicollection.

Variations in $^{92}Zr/^{91}Zr$ ratio are reported in ϵ-values, i.e., the deviation in parts per 10,000 of the isotopic ratio measured in the sample with respect to the chondritic uniform reservoir (CHUR). The CHUR reference value $(^{92}Zr/^{91}Zr)_{CHUR} = 1.53020$ (Table 1) is believed to represent both bulk Earth and average chondritic meteorite values. The reference isotope ^{91}Zr is chosen for its relatively large abundance and for not being affected by isobaric interferences:

$$\epsilon_{^{92}Zr} = \left[\frac{(^{92}Zr/^{91}Zr)_{sample}}{(^{92}Zr/^{91}Zr)_{CHUR}} - 1\right] \times 10^4.$$

Nb/Zr ratios ($^{93}Nb/^{91}Zr$) are reported as fractionation factors ($f^{Nb/Zr}$) relative to the chondritic (CHUR) value:

$$f^{Nb/Zr} = \frac{(^{93}Nb/^{91}Zr)_{sample}}{(^{93}Nb/^{91}Zr)_{CHUR}} - 1.$$

The ratio $(^{93}Nb/^{91}Zr)_{CHUR} = 0.56131$ was derived from the chondrite data of Jochum et al. (1986) and the Zr isotope composition in Table 1. Results are given in Table 1 and shown in the fossil isochron diagram ($\epsilon_{^{92}Zr}$ vs. $f^{Nb/Zr}$; Fig. 1). The $^{92}Zr/^{91}Zr$ ratio measured in the terrestrial rutiles, zircons, and baddeleyites are identical to our terrestrial average values (Table 1) to within ~0.5 ϵ-units.

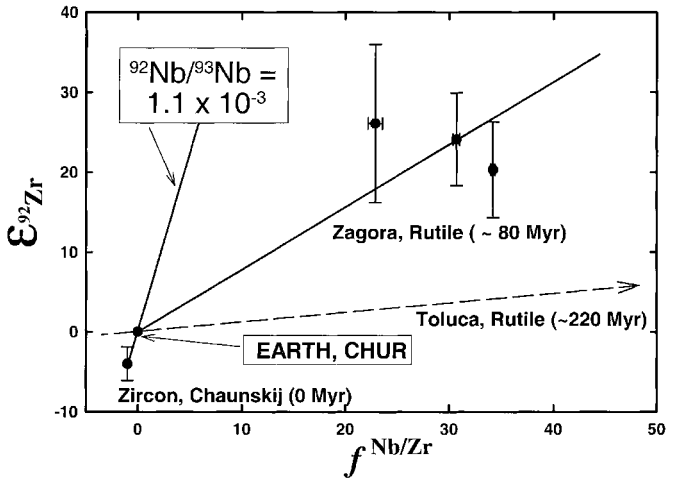

FIG. 1.—Fossil isochron diagram for the ^{92}Nb-^{92}Zr chronometer shown as a graph of $\epsilon_{^{92}Zr}$ vs. $f^{Nb/Zr}$.

3. APPARENT INITIAL $^{92}Nb/^{93}Nb$ ABUNDANCE AND AGES

Utilizing the systematics outlined by Jacobsen & Wasserburg (1984) for the ^{146}Sm-^{142}Nd chronometer and the measured $\epsilon_{^{92}Zr}$ and $f^{Nb/Zr}$ for the meteorite samples, we estimate the initial $^{92}Nb/^{93}Nb$ ratio at the time of crystallization directly:

$$\epsilon_{^{92}Zr} = Q_{^{92}Zr} f^{Nb/Zr} \left(\frac{^{92}Nb}{^{93}Nb}\right)_{initial}.$$

Here $Q_{^{92}Zr} = 10^4(^{93}Nb/^{92}Zr)_{CHUR} = 3668$ is calculated from the present Nb/Zr ratio and Zr isotope composition of CHUR given above.

Thus, for the Chaunskij mesosiderite zircon $f^{Nb/Zr} \sim -1$, $(^{92}Nb/^{93}Nb)_{initial} = -\epsilon_{^{92}Zr}/Q_{^{92}Zr}$. Insofar as zircons yield $\epsilon_{^{92}Zr}$ close to the initial solar value, it is not important to know their exact age in order to obtain a relatively reliable initial $^{92}Nb/^{93}Nb$ value for the solar system. The Chaunskij zircon data indicate an initial Zr isotope composition for the solar system that is at least as low as $\epsilon_{^{92}Zr} = -4 \pm 2$. This results in a relatively high initial $^{92}Nb/^{93}Nb$ value of (1.09 ± 0.57) × 10^{-3} for the solar system. A similar argument was used by Harper & Jacobsen (1996) to discuss the possible range of initial $^{182}Hf/^{180}Hf$ values for the solar system based on measurements of the initial solar $^{182}W/^{183}W$ value in meteorites free of Hf.

Three Zagora rutile grains reveal 20–26ϵ excesses relative to the terrestrial rutile in the $^{92}Zr/^{91}Zr$ ratio with an uncertainty of 6–10ϵ as shown in Figure 1. Here $(^{92}Nb/^{93}Nb)_{initial} = -\epsilon_{^{92}Zr}/f^{Nb/Zr}Q_{^{92}Zr}$ ranges from 1.67 to 3.20 × 10^{-4} for this meteorite. Also shown in Figure 1 is an arrow pointing toward the direction of the published Toluca iron meteorite rutile result (Harper 1996). For the Toluca rutile, $f^{Nb/Zr} = 158 \pm 8$. This value, together with $\epsilon_{^{92}Zr} = 9.1 \pm 1.7$, yielded $(^{92}Nb/^{93}Nb)_{initial} = (1.57 \pm 0.29) \times 10^{-5}$ for this meteorite.

If we take the estimated initial solar $^{92}Nb/^{93}Nb$ value of 1.09 × 10^{-3} for the Chaunskij zircon, then we obtain young ages for the meteoritic rutiles: formation times of 60–100 Myr after the formation of the solar system for Zagora rutiles and 220 Myr for Toluca rutile. The most reasonable interpretation of these young ages is that they are cooling ages and that they represent roughly the time at which these minerals became closed systems in their meteorite parent bodies. Two alabandite (MnS) grains, analyzed in the same section of Zagora, show no excess of ^{53}Cr from the decay of ^{53}Mn ($\bar{\tau}_{^{53}Mn} = 5.3$ Myr; Hutcheon et al. 1992), consistent with a late time of the for-

TABLE 2

p-PROCESS PRODUCTION RATIOS OF SUPERNOVA SOURCES

Production Ratio	SNII[a]	SNII[b]	SNIa[c]	SNIa[d]	SNII[e]	SNII[f]
$^{92}Nb/^{93}Nb$	0.0026	...	0.0021	0.0054	0.0092 ± 0.0070	0.35
$^{92}Nb/^{92}Mo$	0.0048	...	0.0039	0.01	0.017 ± 0.013	0.65
$^{97}Tc/^{92}Mo$	...	...	0.02	0.03	0.017 ± 0.008	...
$^{146}Sm/^{144}Sm$	0.024	<0.48	0.05	0.04	0.95 ± 0.55	...
$^{146}Sm/^{92}Mo$	5.1×10^{-4}	<0.01	0.00106	8.5×10^{-4}	0.020 ± 0.012	...

NOTE.—$(^{92}Mo/^{93}Nb)_{\odot} = 0.542$; $(^{92}Mo/^{144}Sm)_{\odot} = 47.25$ (Anders & Grevesse 1989).

[a] Woosley & Howard 1978.

[b] Woosley & Howard 1990.

[c] Howard, Meyer, & Woosley 1991.

[d] Howard & Meyer 1993.

[e] Rayet et al. 1995.

[f] Hoffman et al. 1996.

mation/equilibration of rutile and alabandite grains in this meteorite. Very young ages of sulfide inclusions in iron meteorites, reflecting similar open system behavior, have also been found using the ^{187}Re-^{187}Os chronometer (Shen, Papanastassiou, & Wasserburg 1996).

4. THE INITIAL $^{92}Nb/^{92}Mo$ RATIO

While ^{93}Nb is 85% *s*-process and 15% *r*-process (Arlandini et al. 1999), ^{92}Nb is a shielded *p*-only nuclide; ^{92}Mo is also a *p*-only nuclide, and we choose this nuclide as a stable reference isotope for further discussion. By using the solar value of $(^{92}Mo/^{93}Nb)_{\odot} = 0.542$ (Table 2), we obtain an initial value of $(^{92}Nb/^{92}Mo)_{ESS} = 2.01 \times 10^{-3}$ for the early solar system (ESS).

Following the principles outlined by Schramm & Wasserburg (1970), we estimate the $(^{92}Nb/^{92}Mo)$ ratio in the interstellar medium (ISM) at the time of the origin of the solar system:

$$\left(\frac{^{92}\mathrm{Nb}}{^{92}\mathrm{Mo}}\right)_{\mathrm{ISM}} = \frac{P_{^{92}\mathrm{Nb}}}{P_{^{92}\mathrm{Mo}}}\frac{\bar{\tau}_{^{92}\mathrm{Nb}}}{T^*},$$

where $T^* = T\langle P\rangle/P(T)$, $\langle P\rangle$ is the mean production rate, and $P(T)$ the final production rate. Thus, for UP with $T^* = 10^{10}$ yr, and since $(^{92}Nb/^{92}Mo)_{ISM} \geq (^{92}Nb/^{92}Mo)_{ESS} = 2.01 \times 10^{-3}$, we must have the following constraint on the production ratio: $(P_{^{92}Nb}/P_{^{92}Mo}) \geq 0.39$.

Early studies of the *p*-process were reviewed extensively by Lambert (1992). Various calculations of the relative *p*-process yields of ^{92}Nb and ^{92}Mo, and thus the $(P_{^{92}Nb}/P_{^{92}Mo})$ ratio, are summarized in Table 2 for calculations of both Type Ia (SNIa) and Type II (SNII) supernova sources. Most of these ratios are too low and would not explain the meteorite values. Only the ratio of 0.65 from Hoffman et al. (1996) for SNII sources with neutrino-driven winds can explain the high $(^{92}Nb/^{92}Mo)_{ESS}$ value we observe. This latter calculation is also the only *p*-process calculation that can explain the high abundance of stable ^{92}Mo in the solar system. It therefore seems reasonable to use this value. However, we emphasize that there are no consistent models yet to explain all *p*-only nuclides.

If we use $(P_{^{92}Nb}/P_{^{92}Mo}) = 0.65$, then we obtain $(^{92}Nb/^{92}Mo)_{ISM} = 3.38 \times 10^{-3}$. If there is a free decay interval (Δ), then $(^{92}Nb/^{92}Mo)_{ESS} = (^{92}Nb/^{92}Mo)_{ISM} e^{-\Delta/\bar{\tau}_{^{92}Nb}}$. For $\Delta = 10$ Myr, as for supernova actinide (SNAC) sources (Wasserburg, Busso, & Gallino 1996), we obtain $(^{92}Nb/^{92}Mo)_{ESS} = 2.78 \times 10^{-3}$, and for $\Delta = 100$ Myr, which seems to be required for ^{129}I (Wasserburg et al. 1996), we obtain $(^{92}Nb/^{92}Mo)_{ESS} = 0.49 \times 10^{-3}$; both are within error of the initial solar estimate.

5. OTHER *p*-PROCESS CHRONOMETERS

We compare our ^{92}Nb results with other *p*-process chronometers (Table 2). Table 3 gives the mean life values for short-lived radioactive (*R*) isotopes, solar abundances $(N_S^{\odot})$ of the stable (*S*) reference isotopes used, as well as estimates of initial solar ratios of radioactive to stable species $(N_R/N_S)_{ESS}$ determined from meteorite data. The production ratio $P_{^{146}Sm}/P_{^{144}Sm}$ used in our calculations was based on the highest value of 0.95 in Table 2, similar to the value of 1 used by Wasserburg et al. (1996). For $P_{^{97}Tc}/P_{^{92}Mo}$, we used a value ≤ 1; this must be an upper limit since ^{92}Mo is at the *p*-process abundance peak in this mass region. Due to the uncertainty in this production ratio, and since the $^{97}Tc/^{92}Mo$ value used depends on a Tc/Mo fractionation factor inferred from Re/Mo fractionation factors in the same meteorites (Yin & Jacobsen 1998), we added a question mark next to ^{97}Tc in Figure 2.

For a pair of radioactive (*R*) and stable (*S*) reference isotopes, we define $\alpha_{R/S} \equiv (N_R/N_S)_{ESS}/(P_R/P_S)$. Then, following Schramm & Wasserburg (1970), we have $\alpha_{R/S} = (\bar{\tau}_R/T^*)e^{-\Delta/\tau_R}$, where $\bar{\tau}_R$ is the mean life of *R*, and Δ and T^* are as above. Estimates of $\alpha_{R/S}$ for the *p*-process chronometers are given in Table 3 and shown in Figure 2*a*. While we do not know the uncertainty in $\alpha_{R/S}$, we show error bars in Figure 2 that correspond to a $\pm 50\%$

TABLE 3

OBSERVED (ESS) AND ESTIMATED (UP-ISM) ABUNDANCES OF *p*-PROCESS NUCLIDES

R/*S*	$\bar{\tau}_R$ (Myr)	$N_S^{\odot}$	$(N_R/N_S)_{ESS}$[a]	P_R/P_S[b]	$\alpha_{R/S}$	$(N_R/N_S)_{UP\text{-}ISM}$
$^{92}Nb/^{93}Nb$	52	0.698	1.09×10^{-3}	0.35	3.1×10^{-3}	1.8×10^{-3}
$^{92}Nb/^{92}Mo$	52	0.378	2.01×10^{-3}	0.65	3.1×10^{-3}	3.4×10^{-3}
$^{97}Tc/^{92}Mo$	3.8	0.378	5×10^{-5}	≤ 1	$\geq 5 \times 10^{-5}$	$\leq 3.8 \times 10^{-4}$
$^{146}Sm/^{144}Sm$	149	0.008	0.01	0.95	1.05×10^{-2}	0.014
$^{146}Sm/^{92}Mo$	149	0.378	2.1×10^{-4}	0.02	1.05×10^{-2}	3.0×10^{-4}

[a] Meteorite values: $^{92}Nb/^{93}Nb$ (this work), $^{97}Tc/^{92}Mo$ (from Yin & Jacobsen 1998), and $^{146}Sm/^{144}Sm$ (from Jacobsen & Wasserburg 1984).

[b] The production ratios are the high values from Table 2, except for $P_{^{97}Tc}/P_{^{92}Mo} \leq 1$ (see text).

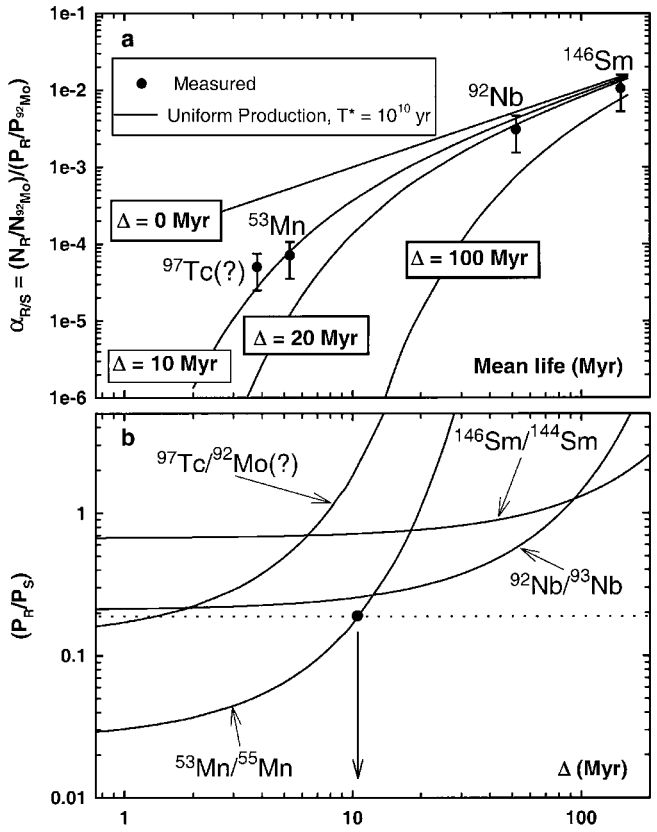

FIG. 2.—(*a*) Graph of $\alpha_{R/S} = (N_R/N_S)_{ESS}/(P_R/P_S)$ vs. $\bar{\tau}_R$ (Myr) for *p*-process chronometers as well as the ^{53}Mn-^{53}Cr chronometer for comparison ($P_{^{53}Mn}/P_{^{55}Mn} = 0.19$ [dotted line in (*b*)] and $^{53}Mn/^{55}Mn = 1.34 \times 10^{-5}$; see Busso et al. 1999 and references therein). (*b*) Graph of production ratios P_R/P_S vs. Δ (Myr) as constrained by meteorite data and the UP model (Δ = 10 Myr for ^{53}Mn).

uncertainty in $\alpha_{R/S}$. For comparison, in Figure 2*a*, we also show curves calculated for UP using the equation above for $T^* = 10^{10}$ yr and Δ-values in the range of 0–100 Myr. It is clear from Figure 2*a* that the data for the *p*-process chronometers and ^{53}Mn all agree with a UP model with a free decay interval of about 10 Myr. For comparison, the last column of Table 3 gives N_R/N_S ratios in the ISM estimated from the UP model. Wasserburg et al. (1995, 1996) and Busso et al. (1999) concluded that some of the short-lived nuclides (^{26}Al, ^{41}Ca and ^{60}Fe, ^{107}Pd) could be accounted for by a nearby polluting AGB star source, while the actinides (^{244}Pu, ^{235} U, ^{238}U, and ^{232}Th), ^{182}Hf, and ^{146}Sm could be produced by a UP model and a 10 Myr free decay interval. The exception was ^{129}I, which appears to require a longer decay interval of Δ ~ 100 Myr. Cameron, Vanhala, & Höflich (1997) suggested that the SNAC source is SNII while ^{129}I is produced by SNIa. The results of Figure 2*a* show that ^{92}Nb, ^{146}Sm, and ^{97}Tc(?) can be produced in the SNAC source by uniform production with Δ = 10 Myr, which is the same as for the SNAC source, and that they are thus most likely produced by SNII, consistent with the Hoffman et al. (1996) scenario.

Because the production ratios are so uncertain, it is fruitful to see what constraints are obtained on the production ratios by using the meteorite data (the fourth column of Table 3) and a simple UP model. The results of such calculations are shown in Figure 2*b*, where the calculated production ratios are shown as a function of the free decay interval. The resulting production ratios for *p*-process chronometers and Δ in the range of 1–100 Myr are all greater than ≈0.2. In particular, for Δ = 10 Myr, we obtain $P_{^{92}Nb}/P_{^{92}Mo} = 0.25$, $P_{^{97}Tc}/P_{^{92}Mo} = 1.4$, and $P_{^{146}Sm}/P_{^{144}Sm} = 0.7$.

It is useful to consider other, more complex open-system models such as the ISM-mixing model of Clayton (1983). In this model, the mixing time between different reservoirs in the ISM and molecular clouds takes the place of the free decay interval. However, it is clear that this model does not help in reducing the required production ratios. On the other hand, a Galactic chemical evolution model with infall (Clayton 1985) will affect all the curves in Figure 2*b* by a constant factor. Using Clayton's (1985) estimate for the *k*-parameter in his model, we obtain production ratios that are about a factor of 4 lower than those obtained for the UP model: $P_{^{92}Nb}/P_{^{92}Mo} = 0.06$, $P_{^{97}Tc}/P_{^{92}Mo} = 0.35$, and $P_{^{146}Sm}/P_{^{144}Sm} = 0.18$. Even these lower ratios are, in general, higher than those summarized for a variety of *p*-process calculations in Table 2, with the exception of the calculations of Hoffman et al. (1996), who consider *p*-process nucleosynthesis in neutrino-driven winds from a nascent neutron star. We conclude that there is a great need for further theoretical and experimental constraints on *p*-process nucleosynthesis.

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