

First $^{94}\text{Nb}(n, \gamma)$ Measurement: Constraining the Nucleosynthetic Origin of ^{94}Mo in Presolar Grains

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Isotopic measurements of presolar silicon carbide grains from dying stars have revealed a puzzling overabundance of ^{94}Mo that stellar nucleosynthesis models have failed to reproduce for two decades. This discrepancy challenged our understanding of the slow neutron-capture process (s -process) that forges approximately half of the elements heavier than iron. The key uncertainty lies at ^{94}Nb , a radioactive branching point where competition between neutron capture and beta decay governs the ^{94}Mo production, yet the neutron-capture cross section had never been measured. Here, we report the first experimental determination of the $^{94}\text{Nb}(n, \gamma)^{95}\text{Nb}$ cross section important for Mo isotopic abundances. The measurement was enabled by a coordinated effort involving high-purity target preparation at Institute of Solid State and Materials Research Dresden, radioactive sample production at the Institut Laue-Langevin Grenoble, radiochemical characterization at Paul Scherrer Institut Villigen, and the time-of-flight CERN n_TOF facility using for the first time segmented total-energy detectors. Incorporation of the resulting Maxwellian-averaged cross section into fully coupled nucleosynthesis models of low-mass asymptotic giant-branch stars brings them into agreement with the presolar grain data. These results remove a major nuclear-physics input uncertainty at the ^{94}Nb branching point and provide a firmer foundation for understanding the origin of ^{94}Mo in the Solar System.

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Introduction—Primitive meteorites preserve a record of the nucleosynthesis processes that produced the heavy elements in our Galaxy. Among their most remarkable constituents are presolar grains, microscopic mineral condensates that formed in the outflows of evolved stars and got incorporation into the Solar System. These grains carry

isotopic signatures that provide direct, quantitative tests of stellar nucleosynthesis models, particularly for the slow neutron-capture process (s -process) responsible for approximately half of the elements beyond iron. The majority of SiC grains recovered from primitive meteorites carry distinctive s -process isotopic signatures of heavy elements such as Mo, firmly establishing their origin in low-mass ($\sim 1.5\text{--}3M_{\odot}$) asymptotic giant-branch (AGB) stars, the main stellar site for the s -process [1–3].

In the bulk solar inventory, ^{94}Mo is regarded as a p -only isotope: The s -process flow mostly bypasses ^{94}Mo because the long-lived ^{93}Zr nucleus, with a half-life of about 1.5 Myr, behaves effectively as stable under s -process conditions and redirects neutron capture toward ^{94}Zr . Thus, its solar abundance is dominantly attributed to the p -

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process. The s -process production of ^{94}Mo in individual AGB stars, however, is not negligible—it is recorded directly in the isotopic signatures of AGB-origin SiC grains, where the ^{94}Mo excesses relative to the initial stellar composition constitute a local fingerprint of s -process nucleosynthesis internal to their parent stars. It is this stellar s -process signature that the present work addresses.

Because Mo is efficiently synthesized by the s -process and repeatedly dredged up into the envelope, the surface Mo isotopic composition—recorded by SiC grains condensing from the surface outflow—directly reflects the interior s -process conditions with minimal sensitivity to the initial stellar composition. Dust condensation produces at most small, mass-dependent isotopic fractionation [4], which is well below the permil level and entirely negligible compared to the several-hundred-permil Mo anomalies recorded in these grains. As a result, the Mo isotope ratios preserved in SiC grains provide a direct and sensitive probe of AGB s -process nucleosynthesis.

Isotopic analyses of AGB-origin SiC grains over the past two decades consistently indicate that the s -process significantly increases the initial ^{94}Mo abundance with respect to ^{92}Mo [5–7]. However, most postprocessing calculations coupled to AGB stellar models predict insufficient productions of ^{94}Mo by the s -process and, therefore, fall short of explaining the grain data [5,6,8]. Along the s -process path (Fig. 1), the production of ^{94}Mo is sensitive to the branching point at ^{94}Nb , whose fate depends on the competition between neutron capture and β^- decay [$T_{1/2} = 2.03(16) \times 10^4$ yr at terrestrial conditions [9]] and, thus, on the $^{94}\text{Nb}(n, \gamma)^{95}\text{Nb}$ reaction rate. At typical s -process temperatures ($\sim 3 \times 10^8$ K), the β^- -decay half-life of ^{94}Nb is predicted to decrease to less than a year [10], while the $^{94}\text{Nb}(n, \gamma)^{95}\text{Nb}$ rate remains purely theoretically estimated and unverified experimentally [11,12]. Consequently, accurate measurements of both the $^{94}\text{Nb}(n, \gamma)^{95}\text{Nb}$ cross section and the temperature-dependent β^- -decay rate of ^{94}Nb are essential for constraining the s -process contribution from this branching point isotope to ^{94}Mo and improving the reliability of AGB nucleosynthesis models.

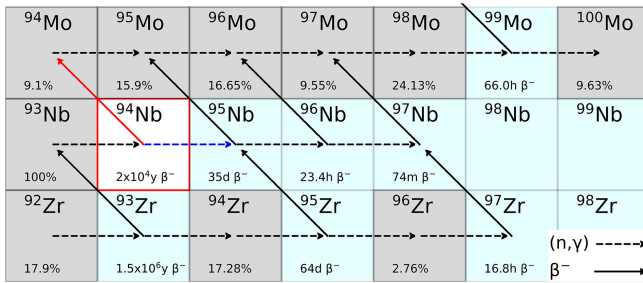


FIG. 1. The s -process path along the Zr-Nb-Mo region, i.e., ^{93}Zr and ^{94}Nb . In the figure, the ^{94}Nb branching point isotope is highlighted.

Only a very limited number of (n, γ) cross sections on radioactive isotopes have been measured at n_TOF, including ^{151}Sm [13], ^{93}Zr [14,15], ^{63}Ni [16], ^{171}Tm [17], ^{204}Tl [18], ^{79}Se [19], and the present ^{94}Nb study. This scarcity reflects the significant experimental challenges associated with such measurements [20].

In this Letter, we report the first experimental determination of the $^{94}\text{Nb}(n, \gamma)^{95}\text{Nb}$ cross section beyond the thermal point [21–23], measured using a radioactive sample of ^{94}Nb at the high-flux EAR2 station of the CERN n_TOF facility [24–27]. The production and characterization of the ^{94}Nb sample represented one of the most challenging aspects of this measurement, accomplished only through a collaborative effort involving the Institut Laue-Langevin (ILL) high-flux reactor for activation of a high-purity ^{93}Nb sample, which was produced at Institute of Solid State and Materials Research (IFW) Dresden. Radiochemical characterization of the activated Nb sample was accomplished at Paul Scherrer Institut (PSI). We present the experimentally determined Maxwellian-averaged cross sections (MACS) and assess their astrophysical implications by incorporating the new reaction rates into AGB stellar models computed with the FRUITY code [28]. These models employ a full nuclear network extending up to bismuth, coupled to the physical evolution of the stellar structure. In this framework, the formation of the ^{13}C pocket, where the $^{13}\text{C}(\alpha, n)$ reaction takes place as the major neutron source in AGB stars, is attributed to mixing induced by buoyant magnetic flux tubes.

Experiment—The synthesis of ultrahigh-purity ^{93}Nb material for the sample used in this experiment was originally conducted at IFW Dresden utilizing refined molten-salt electrolysis, zone melting, and high- and ultra-high-vacuum heat treatments [29]. An amount of 304 mg of ^{93}Nb material with less than 1 ppm of Ta was obtained, consisting of two thin wires, 45 and 47 mm in length and 0.8 mm in diameter. This material was subsequently neutron activated in the V4 irradiation tube at the high-flux ILL reactor in Grenoble for 51 days, corresponding to a thermal neutron fluence of $\approx 4.3 \times 10^{21} \text{ cm}^{-2}$.

After irradiation, the Nb wires were cut into small pieces, distributed in a circle of 16 mm diameter, and sealed from both sides using 25 μm Kapton foil. A careful characterization of the activated sample was performed at PSI Villigen using a customized HPGe setup. The γ -ray spectroscopy analysis yielded an activity of 10.01 MBq of ^{94}Nb with no trace of radioactive contaminants. This activity corresponds to $9.2(7) \times 10^{18}$ atoms using evaluated nuclear structure data file [30] value, approximately 1% of the total number of atoms in the ^{93}Nb - ^{94}Nb bulk sample.

For the data analysis (see below), the rather unconventional geometry of the sample could be approximated as multiple thin cylinders irradiated perpendicular to their long axis. Additionally, the high β^- radioactivity

($Q_\beta = 2045.0$ keV followed by a two-step γ -ray cascade of 702 and 871 keV) and the low amount of ^{94}Nb introduce important challenges for a time-of-flight (n,γ) measurement. These difficulties were mitigated by using the high instantaneous neutron flux station EAR2 at CERN n_TOF [24–27] in combination with a highly segmented detection apparatus called sTED. The latter is based on an array of relatively small cells of C_6D_6 , which were designed for rapid response and high count-rate conditions [31,32]. This system allows one to handle the high instantaneous count rates at EAR2 during each neutron pulse, as well as the effects of the γ -ray flash at the short flight-path of EAR2, thus enabling a wide neutron energy range where the detectors can operate with stable and well-controlled performance. Nine sTED detection units were set up in an unconventional compact-ring configuration, which permitted us to maximize the signal-to-background ratio [33,34].

The neutron-capture yield was measured by detecting the prompt γ rays emitted by the (n,γ) capture reactions. The pulse height weighting technique (PHWT) [35] was employed during data processing to ensure that the efficiency of detecting the γ -ray cascade is proportional to the total energy of the cascade, E_C , thus rendering it independent of the specific deexcitation path of each individual cascade. After applying the PHWT, the experimental yield is calculated as

$$Y_{\text{exp}}(E_n) = f_N \frac{C_W(E_n) - B_W(E_n)}{E_C \phi(E_n)}. \quad (1)$$

Here, $C_W(E_n)$ and $B_W(E_n)$ represent the number of counts after application of the PHWT from the sample and background, respectively. Furthermore, $\phi(E_n)$ denotes the n_TOF EAR2 neutron fluence [25–27], while f_N is the absolute normalization factor for the yield. The latter was determined using the well-established $^{197}\text{Au}(n,\gamma)$ 4.9 eV saturated resonance method [13,36]. For this purpose, high-purity Au and ^{93}Nb samples with well-defined disk geometries were utilized, having diameters of 16.34 mm and thicknesses of 50 μm for Au and 2.1 mm for ^{93}Nb , respectively. The methodology experimentally validated at n_TOF [13,37] was applied to account for the loss of part of the γ -ray cascade due to experimental thresholds. To this aim, detailed Monte Carlo simulations for $^{197}\text{Au}(n,\gamma)$ and $^{93}\text{Nb}(n,\gamma)$ were carried out [33,38]. In addition, corrections for dead time and pileup effects were rigorously implemented following the procedure described in Ref. [39]. The absolute normalization of the yield calculated by this method was applied to the ^{94}Nb sample by scaling the two first ^{93}Nb resonances (35.9 and 42.3 eV) present in the measured yield. The uncertainty associated with this normalization, which includes cascade corrections, dead time, and the translation to the final sample yields, amounts to 8%.

The resonance analysis presented in this Letter was performed using the Bayesian R -matrix code SAMMY [40]. However, the standard yield calculation in this code had to be replaced by a custom implementation, which incorporates multiple-scattering and self-shielding corrections tailored to the nonstandard sample geometry. This implementation follows the methodology described in Ref. [41] and is essential for determining accurate ^{94}Nb resonance parameters. Additional details on the analysis and the results for the ^{94}Nb and ^{93}Nb samples measured in this work will be presented in a forthcoming publication [42].

In total, we have observed and analyzed ten $^{94}\text{Nb}(n,\gamma)$ resonances up to a neutron energy of about 800 eV. For the first resonance, we were able to determine a radiation width of $\Gamma_\gamma = 145(40)$ meV. Where necessary, this value was fixed for the other resonances during the determination of other parameters. A summary of the resonance parameters determined in this work is available in Supplemental Material [43].

Taking into account the observed resonances and the detection threshold, the fraction of missing resonances was estimated, yielding an s -wave resonance spacing of $D_0 = 32(7)$ eV and a neutron strength function of $S_0 = 0.19(8) \times 10^{-4}$. The MACS at different stellar temperatures were calculated using the average resonance parameters, following the approach of Refs. [17,44]. In practice, p -wave neutrons contribute significantly to the MACS of $^{94}\text{Nb}(n,\gamma)^{95}\text{Nb}$. This contribution was evaluated by assuming the same average radiative width Γ_γ as for s -wave neutrons—an approximation fully supported by statistical-model calculations [45,46]—and by adopting a neutron strength function S_1 based on values from neighboring isotopes. D_1 was estimated under the assumption of parity-independent level densities, using the spin cutoff parameter described in Ref. [47].

Results—Figure 2 compares the MACS as a function of stellar thermal energy (kT) obtained in this work with the values recommended in KADoNiS v0.3 and other theoretical predictions using the Hauser-Feshbach formalism with different parameterizations [11,12,48–52]. Because all previous estimates were purely theoretical, no uncertainty is displayed for them. Uncertainties presented for n_TOF as a red band correspond to the systematic uncertainty of 8%, arising from the different experimental corrections discussed above. The gray-shaded band indicates the $\pm\sigma$ statistical uncertainty for the n_TOF result inferred from the uncertainties in the average resonance parameters used in the calculations. A more detailed description of this procedure and $\pm 2\sigma$ exclusion limits can be found in Supplemental Material [43], which also provides the numerical values of the MACS.

The new determined MACS values at $kT = 5$ and 30 keV are 1167(93) and 460(37) mb, respectively. They correspond to a 26% higher value at 5 keV, while at 30 keV the experimental result is within 5% of the KADoNiS

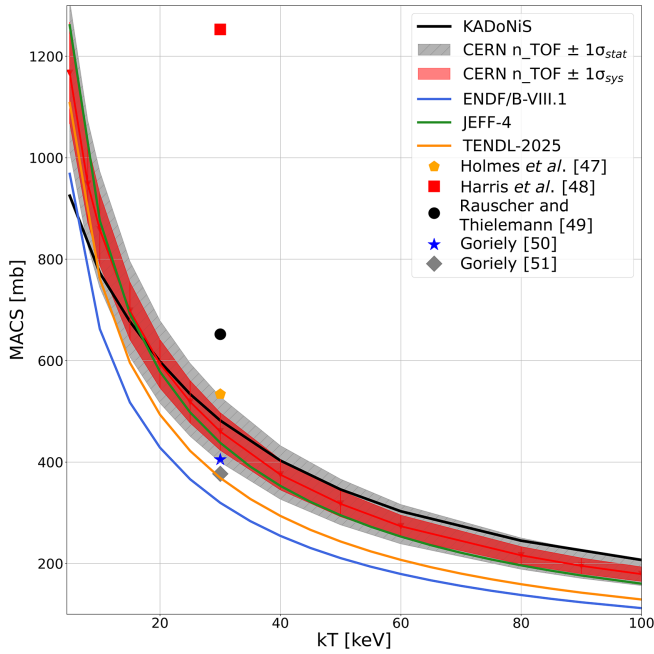


FIG. 2. MACS as a function of kT , compared with KADoNiS [12] and five theoretical predictions. See the text for details.

value. At higher kT values, the new MACS are slightly lower than those of KADoNiS, by up to 13%.

Discussion—The new $^{94}\text{Nb}(n, \gamma)^{95}\text{Nb}$ MACS reported here enables a more reliable constraint on one of the key branching points along the Zr-Nb-Mo s -process path, when compared to the pure theoretical calculations available thus far. Over the astrophysically relevant range of thermal energies, our MACS values are comparable to or slightly higher than previous theoretical estimates [11], ruling out a significantly slower $^{94}\text{Nb}(n, \gamma)^{95}\text{Nb}$ rate as a mechanism to enhance the s -process production of ^{94}Mo in low-mass AGB stars. Grains with inferred AGB origins comprise the mainstream (MS) ($\sim 90\%$), Y ($\sim 1\%$ – 5%), and Z ($\sim 1\%$ – 5%) groups, differing in their Si isotopic compositions but exhibiting indistinguishable Mo isotopic ratios [6]. These shared s -process Mo patterns show that all these three groups formed in near-solar-metallicity AGB stars [53]. When incorporated into fully coupled magnetic FRUITY AGB models [28], the overproduction of ^{94}Mo relative to the initial abundance remains at the level inferred from MS, Y , and Z SiC grains [6,7,54], and the revised model predictions overlap well with the grain data shown in Fig. 3.

The FRUITY models also allow assessing the sensitivity of the ^{94}Mo yield to the ^{94}Nb branching in a fully coupled stellar framework. Across the range of low-mass, near-solar-metallicity AGB stars relevant for MS, Y , and Z grains, adopting our experimentally determined $^{94}\text{Nb}(n, \gamma)^{95}\text{Nb}$ rate in place of the previous theoretical estimate [11,12] leads to only modest changes in the final $^{94}\text{Mo}/^{96}\text{Mo}$ ratios. This limited response indicates that

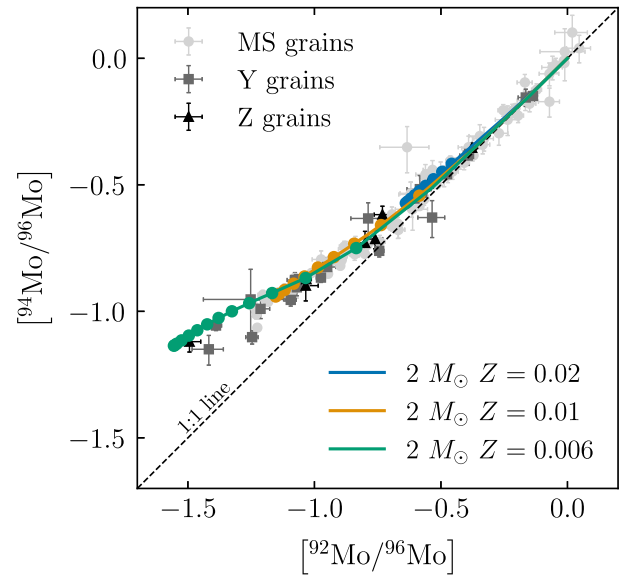


FIG. 3. Molybdenum three-isotope plot comparing MS, Y , and Z grains to magnetic FRUITY models of $2M_{\odot}$ AGB stars. The grain data are plotted with 1σ uncertainties [6,7,54]. The lines with symbols trace the trajectory of envelope isotopic compositions at two different metallicities (Z), with symbols indicating phases where the envelope becomes C-rich ($C/O > 1$), a necessary condition for SiC dust condensation. The $Z_{\odot}$ adopted in the models is 0.0185.

earlier theoretical rates were already close to the MACS determined experimentally. Importantly, with the new rate, the FRUITY models reproduce the s -process contribution to the ^{94}Mo abundance inferred from SiC grains (Fig. 3), consistent with the conclusions of a recent postprocessing study [55]. Given that the nuclear inputs in most of previous postprocessing calculations [6,8] are similar to those adopted in this work, we infer that the long-standing ^{94}Mo data-model discrepancy likely arose from limitations in their postprocessing treatments—particularly their approximate handling of the temperature- and density-dependent β^- -decay rate of ^{94}Nb —rather than from unrealistic neutron-capture rates. As the stellar β^- -decay rate of ^{94}Nb remains based solely on theoretical calculations, experimental constraints are essential to fully validate the ^{94}Mo production pathway in AGB stars. The PANDORA project [56] is specifically designed to address this by measuring β -decay rates of radioactive isotopes in laboratory plasmas that reproduce stellar temperature and density conditions. The β^- decay of ^{94}Nb is one of its primary targets, and it will provide the missing experimental anchor for this branching point.

However, the s -process contribution to the solar ^{94}Mo inventory remains marginal, as ^{94}Mo is traditionally a p -only isotope. The ^{94}Mo excess in SiC grains reflects local AGB nucleosynthesis rather than bulk solar composition. By using net stellar yields to isolate the actual s -process production, our magnetic FRUITY models consistently

predict a fraction of $\sim 4.6\%$ across all metallicities. This matches the $\sim 4\%$ value extrapolated from grain data at the pure s -process end member. In agreement with galactic chemical evolution model decompositions [57,58], this confirms that p -process nucleosynthesis remains responsible for the vast majority of the solar $^{92}\text{Mo}/^{94}\text{Mo}$ ratio.

To assess the robustness of our results, we tested the sensitivity of our stellar predictions by varying the newly measured ^{94}Nb neutron-capture cross section by its 2σ uncertainty. We found that the resulting $^{94}\text{Mo}/^{96}\text{Mo}$ isotopic ratio varies by less than 3%. Such a narrow theoretical uncertainty band is negligible compared to the wide spread of the presolar SiC grain data, confirming that our predictions and the resulting s -process contribution estimates are robust against current nuclear-physics uncertainties.

By providing the first experimental determination of the $^{94}\text{Nb}(n, \gamma)^{95}\text{Nb}$ rate, this work removes a significant source of uncertainty in the nuclear input and sets a firmer foundation for future efforts to disentangle the stellar and galactic processes that shaped the ^{94}Mo inventory in the Solar System. For completeness, measurements on the stable Mo isotopes have recently been conducted at n_TOF [59,60] to confirm or update the remaining cross sections involved in the s -process production of ^{94}Mo network with constrained uncertainties.

Conclusions—We have performed the first experimental determination of the (n, γ) cross section on the radioactive isotope ^{94}Nb , a key branching point isotope in the s -process flow toward ^{94}Mo . This challenging ^{94}Nb experiment required a comprehensive multifacility collaboration: production of ultrapure ^{93}Nb material at IFW, activation at the ILL high-flux reactor, radiochemical characterization at PSI, and high-resolution time-of-flight measurements at the CERN n_TOF EAR2 facility. The use of the newly developed sTED detection system was essential to achieve the required detection sensitivity. The resulting MACS values represent the first experimental constraints on this critical reaction, with values comparable to, or slightly higher than, previous theoretical estimates.

To assess the astrophysical implications of our results, we incorporated the measured rates into fully coupled AGB stellar model calculations based on the magnetic-FRUITY framework. The calculations demonstrate that fully coupled modern AGB models employing magnetic buoyancy-induced mixing, when combined with our $^{94}\text{Nb}(n, \gamma)^{95}\text{Nb}$ reaction rates, successfully reproduce the $^{94}\text{Mo}/^{96}\text{Mo}$ ratios observed in AGB-origin presolar SiC grains. Our results confirm that the previously problematic ^{94}Mo abundances can be solved through the combination of accurate treatment of thermally sensitive s -process branchings and experimentally constrained reaction rates, without requiring exotic astrophysical scenarios.

While our measurement rules out uncertainties in the $^{94}\text{Nb}(n, \gamma)^{95}\text{Nb}$ reaction as a source of model-data discrepancies, the complete picture of ^{94}Mo production in AGB

stars still requires better constraints on the temperature-dependent ^{94}Nb β -decay rate under stellar conditions. Combined with continued refinements in AGB stellar modeling, these efforts will provide a comprehensive understanding of s -process branching-point nucleosynthesis and its signatures in presolar stardust.

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Data availability—The data that support the findings of this article are available from the corresponding author upon reasonable request.

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End Matter

Appendix: Time evolution of the key Zr, Nb, and Mo isotopes during thermal pulses—During interpulse periods—when the $^{13}\text{C}(\alpha, n)^{16}\text{O}$ source provides a low neutron density ($\sim 10^7\text{--}10^8\text{ cm}^{-3}$) [61] and the ^{93}Zr and ^{94}Nb branching points remain inactive—the s -process flow converts the initial amount of ^{93}Nb directly to ^{94}Mo , but it is then almost completely destroyed through neutron captures. Conversely, the long-lived ^{93}Zr is largely produced by neutron captures on ^{92}Zr . At the onset of the following thermal pulse (TP), partial activation of the $^{22}\text{Ne}(\alpha, n)^{25}\text{Mg}$ neutron source generates a brief episode of high neutron densities ($10^9\text{--}10^{11}\text{ cm}^{-3}$) [61], thereby activating the branching point at ^{94}Nb and causing neutron capture and β^- decay to compete. Under these conditions, the abundance of ^{94}Mo increases during the TP [5]: The large amount of ^{93}Nb produced by the decay of ^{93}Zr during the previous interpulse phase is converted into ^{94}Nb , a significant fraction of which undergoes β^- decay to ^{94}Mo (see Fig. 4). The alternative production channel through successive neutron captures on ^{92}Mo and ^{93}Mo remains negligible, given the low initial ^{92}Mo abundance. At the same time, most of the s -process flow from ^{93}Zr goes to ^{94}Zr rather than β decaying to ^{93}Nb . The newly formed ^{94}Zr continues to capture neutrons to form ^{95}Zr , which subsequently decays to ^{95}Nb and then to ^{95}Mo , thus bypassing ^{94}Mo entirely (Fig. 1). As a result, the $^{94}\text{Nb}(n, \gamma)^{95}\text{Nb}$ rate determined in this work confirms

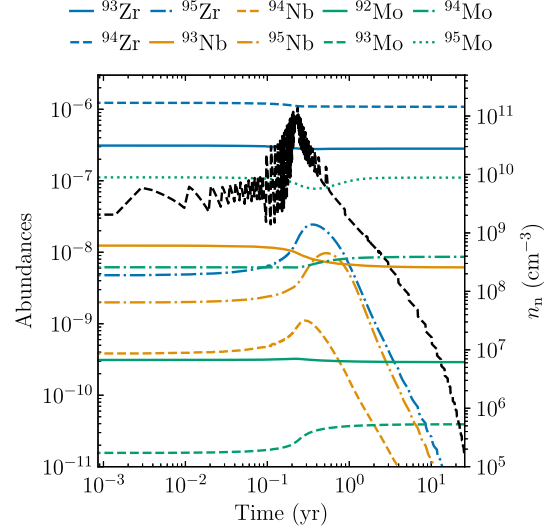


FIG. 4. Time evolution of the key Zr, Nb, and Mo isotopes and the neutron density (dashed gray line) during a representative thermal pulse for the $2M_{\odot}$ obtained from magnetic FRUITY models. The timescale starts when the temperature at the bottom of the convective TP reaches 200 MK.

that the synthesis of ^{94}Mo in AGB stars is tightly linked to the short high-neutron-density episodes that occur during thermal pulses. During the long interpulse phases, its production remains inefficient and ^{94}Mo is largely destroyed.