

Neutron capture reaction rates for silicon and their impact on the origin of presolar mainstream SiC grains

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We have made new, improved measurements of the $^{28-30}\text{Si}(n, \gamma)$ cross sections and have done a resonance analysis of these data including previous total cross sections. Together with the calculated contributions due to direct capture, we calculated the astrophysical (n, γ) reaction rates and investigated the s -process abundances of the Si isotopes. Measured isotopic anomalies of intermediate and heavy elements in SiC grains from meteorites appear to be attributable to the s -process in asymptotic giant branch (AGB) stars. But the Si isotopic ratios in these grains are substantially different than s -process models predict. Therefore, recent papers have invoked galactic chemical evolution or other effects to explain the Si isotope ratios in these grains. Our new reaction rates are significantly different than previous rates, and s -process calculations using these rates lead to much larger isotopic shifts in ^{30}Si . However, these exploratory calculations demonstrate that even with these substantially different rates the large observed variation in SiC grain from AGB stars cannot be explained by standard s -process models.

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About half of the abundances of elements having $A > 90$ are thought to be produced in asymptotic giant branch (AGB) stars via a chain of neutron capture reactions and β decays called the s process [1]. Because the neutron density during the s process is relatively low, the nucleosynthesis is constrained to follow the valley of β stability; hence, the result of s -process synthesis for particular environments can be computed in detail using mostly experimentally accessible neutron capture reaction and β -decay rates.

Grains of SiC recovered from primitive meteorites [2–4] appear to preserve the signature of the s process [5]. These grains contain small amounts of intermediate and heavy nuclides having very nonsolar isotopic compositions. Most of these isotopic ratios follow the pattern expected from the s process—compared to solar they are enriched in isotopes produced solely or predominantly by the s process. Based in part on the, for the most part, good match between predicted and observed isotopic ratios for these heavy elements, AGB stars inside of which the s process was occurring have been identified as originators of these grains [6–9]. The Si is about 50% of the total number of atoms in the grains, but the measured Si isotopic abundances in the meteoric SiC grains do not match the predictions from AGB stellar models, assuming that the Si abundances initially were solar and were modified only by the s process in the star from which the grain was formed. Instead, Si isotope ratios in these grains appear to offer a new window to galactic chemical evolution. Detailed analysis of the SiC grains show that $\sim 90\%$ are so-called mainstream grains [10–13]. As shown in Fig. 1, mainstream grains lie along a correlation line with a slope of 1.31 in a three-isotope plot for Si. This form of expressing deviations in per thousand (permil) from the solar isotopic ratio $(^i\text{Si}/^{28}\text{Si})_{\odot}$ is defined as $\delta(^{29}\text{Si}/^{28}\text{Si}) = [(^{29}\text{Si}/^{28}\text{Si}) / (^{29}\text{Si}/^{28}\text{Si})_{\odot} - 1] \times 1000$ and $\delta(^{30}\text{Si}/^{28}\text{Si}) = [(^{30}\text{Si}/^{28}\text{Si}) / (^{30}\text{Si}/^{28}\text{Si})_{\odot} - 1] \times 1000$ with $(^{29}\text{Si}/^{28}\text{Si})_{\odot}$

$= 0.0506331$ and $(^{30}\text{Si}/^{28}\text{Si})_{\odot} = 0.0334744$ [henceforth, we call $\delta(^{29}\text{Si}/^{28}\text{Si})$ $\delta^{29}\text{Si}$ and $\delta(^{30}\text{Si}/^{28}\text{Si})$ $\delta^{30}\text{Si}$]. There are at least three problems with ascribing the origin of the Si isotopic ratios in these grains to the s process using standard models. First, the range of $\delta^{29}\text{Si}$ and $\delta^{30}\text{Si}$ values observed in mainstream grains is much larger than predicted by s -process models for AGB stars. Second, the slope of 1.31 in the three-isotope plot is also much larger than predicted. Third, standard homogeneous galactic chemical evolution models predict that both $\delta^{29}\text{Si}$ and $\delta^{30}\text{Si}$ increase with time over the age of the galaxy, so that the fact that most mainstream grains have positive $\delta^{29}\text{Si}$ and $\delta^{30}\text{Si}$ values would lead to the conclusion that the Si isotopes found in these grains are younger than the Sun.

The inability of the s process to produce large enough $\delta^{29}\text{Si}$ and $\delta^{30}\text{Si}$ values is, in part, due to the fact that the neutron capture reaction rates for the light nuclides are generally very small. The failure of s -process models to produce a large enough slope in the three-isotope plot is also related to the neutron reaction rates. In these models, the abundance of ^{30}Si is modified much more than that of ^{29}Si because of a feeding from the $^{33}\text{S}(n, \alpha)^{30}\text{Si}$ reaction, which has a comparatively large rate. Several explanations have been proposed to resolve the puzzle that the heavy elements in mainstream SiC grains appear to come from the s process, while the Si isotopic ratios cannot be explained in this way. For example, in Ref. [14], it was shown that a renormalization of the three-isotope plot for Si from the interstellar medium (ISM) to the solar frame of reference could result in a slope in agreement with the mainstream grains, but with a displacement from the observed values indicative of the uniqueness of the Sun with respect to the ISM. Alternatively, in Ref. [13], it was proposed that the puzzle of the Si isotope ratios in mainstream grains could be explained as being due to heterogeneities from a limited number of supernova sources.

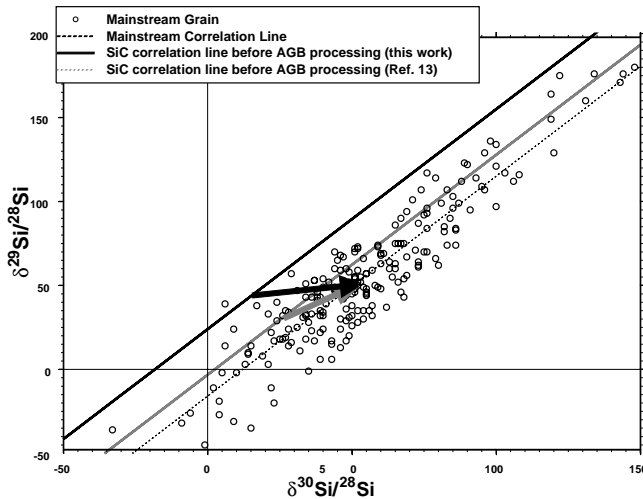


FIG. 1. Three-isotope plot for Si. Si having solar composition lies at the origin. Isotope ratios for mainstream SiC grains (for clarity, not all are plotted) are shown as open circles, and the dashed line represents the fitted correlation line with slope 1.31 for the mainstream grains. The gray dashed-dotted line represents the AGB-corrected mainstream grain correlation line from Ref. [13]. The solid line depicts the AGB-corrected mainstream grain correlation line using our new rates for the AGB model ST of Ref. [13]. The black and gray arrows depict the *s*-process shift from the AGB stellar calculation using our rate and from Ref. [13], respectively. The vectors illustrate the corresponding *s*-process shifts for each calculation.

Both of these solutions rely on the accurate prediction of the Si isotope ratios resulting from *s*-processing in AGB stars. However, the reaction rates for the Si isotopes are based on old cross section measurements that are suspected to be in error. For example, criticality benchmark calculations undertaken for the nuclear criticality safety program (NCSP) (for systems containing large amounts of Si mixed with ^{235}U and having a significant fraction of the neutron flux in the epithermal region) predicted that known critical systems would be subcritical. However, calculations run with substantial *ad hoc* reductions of neutron capture cross sections for Si agreed with the benchmark [15]. Furthermore, it is likely that the previous neutron cross section data [16] are too large because the correction for the prompt neutron sensitivity of the apparatus had been underestimated. This correction can be particularly important for these lighter nuclides that have small neutron capture, but large neutron scattering, cross sections. Several recent measurements (e.g., Ref. [17]) have demonstrated that this correction had been underestimated in other similar measurements using the same apparatus. Hence, we made new measurements of the Si neutron capture cross sections at the Oak Ridge Electron Linear Accelerator (ORELA). These new experiments were performed utilizing the C_6D_6 detector system at the 40-m flight station of the ORELA. With this new apparatus, we have reduced the prompt neutron sensitivity of the system to the point where it has been shown to be negligible. Details of the experimental technique as well as this improvement and others can be found in Refs. [17,18].

Because measurements were undertaken to support the

NCSP, only the major isotope ^{28}Si (92.23% abundance), was of interest. Therefore the sample used in this experiment was a silicon crystal of natural isotopic composition, with dimensions of 2.54×5.2 cm, and a thickness of 1.57 cm. It was located at a distance of 40.12 m from the ORELA neutron target in a well-collimated neutron beam. The energy dependence of the neutron flux was measured with a thin ^6Li loaded glass scintillator located 43 cm ahead of the sample. Individual runs to measure sample-out and average neutron-scattering backgrounds were made with an empty sample holder and a carbon sample, respectively. We used the saturated resonance technique [19], employing the 4.906-eV resonance in the $^{197}\text{Au}(n, \gamma)$ cross section, to convert the measured counts to absolute cross sections.

The determination of the astrophysical reaction rates was improved in two other ways compared to previous work. First, the resonance analysis of our new capture data included newer total cross section measurements on isotopically enriched silicon samples [20]. With these total cross section data, the reliability of isotopic assignments was much improved. In addition, these total cross section data, coupled with the fact that the excellent ORELA time-of-flight resolution resolves neutron resonances below ~ 700 keV, made possible precise corrections for resonance self-shielding and multiple scattering across the entire range of interest to astrophysics. Second, we calculated the component of the cross section due to direct capture (DC). This DC component cannot be determined using our experimental technique and was neglected in previous determinations of the reaction rate. Far from being negligible, we found that DC constitutes a substantial fraction of the reaction rate (see Table I). The data were analyzed in the resolved resonance region using the multilevel *R*-matrix code SAMMY [21]. The silicon capture data were fitted in the energy range from 100 eV to 700 keV, where we obtained resonance parameters for 18, 16, and 8 resonances for ^{28}Si , ^{29}Si , and ^{30}Si , respectively. We observed two resonances for ^{28}Si , which had not been previously reported [22]. In addition, we determined that one resonance previously assigned to ^{28}Si [22] is actually in ^{30}Si . Furthermore, a resonance in ^{30}Si at 2.235 keV that was added [22] to account for the measured resonance integral was not visible in our new capture data, nor in the transmission measurements using an enriched ^{30}Si sample [20]. This latter fact and the reduced capture kernel for the 4.977-keV resonance lead to a large reduction of the ^{30}Si neutron capture cross section at lower neutron energies.

We performed the direct capture calculations using the code TEDCA [23,24], which takes *E1*, *M1*, and *E2* transitions into account. In the calculation *E1* transitions were found to be dominant, therefore only these were used for the parametrizations of the cross sections. The uncertainty in the DC cross section was estimated by combining the uncertainty in the scattering length [25]—which is used to determine the optical potential—and an estimated uncertainty in the spectroscopic factors [26,27]. The resulting uncertainty in the DC component is rather large and is, in some cases, the dominant uncertainty in the reaction rate. Also, the comparatively large contribution of DC to the thermal cross section had to be taken into account in the *R*-matrix analysis by

TABLE I. Maxwellian averaged cross sections for the Si isotopes compared to the most recent evaluation.

kT (keV)	^{28}Si				^{29}Si				^{30}Si			
	Expt.	DC (mb)	Expt.+DC.	Ref. [30]	Expt.	DC (mb)	Expt.+DC	Ref. [30]	Expt.	DC (mb)	Expt.+DC.	Ref. [30]
5	0.19 ± 0.01	0.22 ± 0.09	0.41 ± 0.09	0.29	7.78 ± 0.78	0.51 ± 0.32	8.29 ± 0.84	10.3	14.68 ± 1.47	0.32 ± 0.16	15.00 ± 1.48	124.0
8	0.24 ± 0.01	0.20 ± 0.09	0.44 ± 0.09	0.60	10.64 ± 1.06	0.58 ± 0.38	11.22 ± 1.13	12.9	8.81 ± 0.88	0.33 ± 0.18	9.15 ± 0.90	62.5
10	0.33 ± 0.02	0.20 ± 0.09	0.53 ± 0.09	0.86	11.14 ± 1.11	0.63 ± 0.42	11.77 ± 1.19	14.4	6.59 ± 0.66	0.35 ± 0.20	6.94 ± 0.69	43.0
15	0.65 ± 0.03	0.20 ± 0.10	0.86 ± 0.10	1.9	10.57 ± 1.06	0.73 ± 0.50	11.30 ± 1.17	13.3	3.67 ± 0.37	0.38 ± 0.23	4.05 ± 0.43	22.0
20	0.94 ± 0.05	0.21 ± 0.10	1.15 ± 0.11	2.5	9.17 ± 0.92	0.82 ± 0.57	9.98 ± 1.08	11.3	2.35 ± 0.24	0.42 ± 0.25	2.77 ± 0.34	13.0
25	1.11 ± 0.06	0.22 ± 0.11	1.33 ± 0.12	2.8	7.77 ± 0.78	0.90 ± 0.63	8.67 ± 1.00	9.5	1.69 ± 0.17	0.45 ± 0.28	2.13 ± 0.32	8.8
30	1.19 ± 0.06	0.23 ± 0.11	1.42 ± 0.13	2.9	6.58 ± 0.66	0.98 ± 0.69	7.56 ± 0.95	7.9	1.34 ± 0.13	0.48 ± 0.30	1.82 ± 0.33	6.5
40	1.23 ± 0.06	0.25 ± 0.13	1.47 ± 0.14	2.8	4.88 ± 0.49	1.11 ± 0.79	5.99 ± 0.93	5.8	1.08 ± 0.11	0.53 ± 0.34	1.61 ± 0.35	3.8
50	1.18 ± 0.06	0.26 ± 0.14	1.45 ± 0.15	2.7	3.78 ± 0.38	1.23 ± 0.88	5.02 ± 0.96	4.4	1.04 ± 0.10	0.59 ± 0.37	1.63 ± 0.39	2.6

adjusting the parameters of the external levels as well as those of some of the resonances. Details about the resonance analysis, the final resonance parameters, and the DC calculations can be found in Ref. [28]. Cross sections calculated with these final parameters were found to give good agreement [15] with criticality benchmarks in the epithermal region which included silicon and ^{235}U . We calculated astrophysical $^{28-30}\text{Si}(n, \gamma)$ rates due to our resonance parameters [28] using standard techniques [21]. To this, we added the DC contributions to obtain the total reaction rates. Because we used a sample having natural isotopic abundance for our (n, γ) measurements, it contained much less $^{29,30}\text{Si}$ than ^{28}Si . Therefore, the statistical precision of the $^{29,30}\text{Si}(n, \gamma)$ rates (10%) is worse than for $^{28}\text{Si}(n, \gamma)$ (5%). This has to be combined with the uncertainty of DC calculations. However, because of the improvements in our measurements and in our resonance analysis, and because we included the DC component in the calculation of the reaction rates, our results should be much more reliable than the previously accepted rates (which also are based on measurements made with samples having natural isotopic composition). Our new rates with the combined uncertainties are compared to the most recent evaluation in Table I, and the astrophysical reaction rates are plotted in Fig. 2. Overall, our $^{28-30}\text{Si}(n, \gamma)$ reaction rates are much smaller than the previously accepted ones, especially $^{30}\text{Si}(n, \gamma)$. At $kT=30$ keV, our rates are approximately 50%, 4%, and 70% lower for $^{28-30}\text{Si}$, respectively, compared to the latest evaluation [30]. In current AGB stellar models of the s process, most neutron exposure occurs at $kT=8$ keV. At this temperature, our new rates are 25%, 13%, and 85% lower than Ref. [30] for $^{28-30}\text{Si}(n, \gamma)$, respectively.

Our reaction rates for $^{30}\text{Si}(n, \gamma)$ are significantly different from those obtained from a recent activation measurement [31]. At the two reported temperatures, $kT=25$ and 52 keV, the rates are 1.65 and 0.44 times our rates, respectively. Although the activation technique can, in principle, determine the rate due to both resonance and direct capture, the accuracy of the technique is problematical for cases like $^{30}\text{Si}(n, \gamma)$ where narrow, widely separated resonances dominate (96% and 80% of the rates at $kT=8$ and 25 keV, respectively, according to our data) the reaction rate. As the

authors of Ref. [31] point out themselves, in such cases the rates obtained by the activation technique are subject to unknown and possibly large systematic uncertainties due to deviations between the incident neutron spectrum of the measurements and a true Maxwell-Boltzmann distribution. In contrast, our time-of-flight technique allows a very accurate determination of the strengths, and hence the reaction rate due to these resonances. Even though the sample used for this experiment was rather thick, the corrections for multiple scattering as calculated by the well tested analyzing code

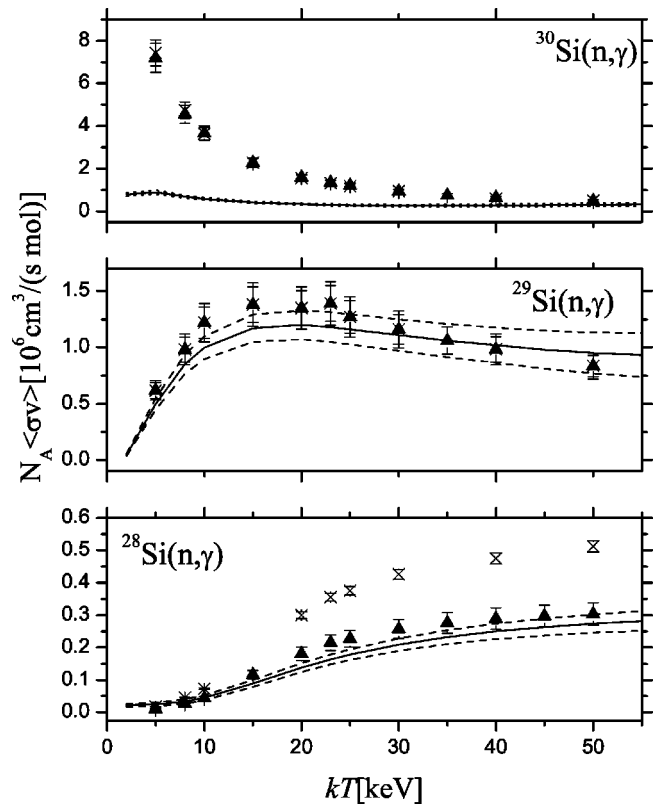


FIG. 2. Astrophysical reaction rates for the $^{28-30}\text{Si}(n, \gamma)$ reaction calculated from the cross sections of the present work (solid curves, with the dashed curves depicting the uncertainties), Ref. [29] (\blacktriangle), and Ref. [30] (X).

TABLE II. Comparison of the calculated δ -value ranges (in %) for the ratios of $^{29}\text{Si}/^{28}\text{Si}$ and $^{30}\text{Si}/^{28}\text{Si}$ using our new reaction rates and the values from Ref. [30]. The calculations were performed for the case ST with different stellar mass models [32]. Shown are the δ -value ranges for different pulses under C>O conditions in the stellar envelope where SiC grains form.

	1.5 M_{\odot}		3 M_{\odot}	
	This work	Using the reaction rates from Ref. [30]	This work	Ref. [30]
	(%)		(%)	
$\delta(^{29}\text{Si}/^{28}\text{Si})$	4.4–8.4	11.2–21.1	5.7–8.4	16.0–21.4
$\delta(^{30}\text{Si}/^{28}\text{Si})$	25.0–46.2	13.1–26.8	27.9–41.0	18.6–26.1

SAMMY were at most 2% for some of the ^{30}Si resonances. For the self-shielding of individual resonances, SAMMY applied a maximum correction of about 20%. These relatively small corrections, the well reproduced criticality benchmark calculations, and the fact that SAMMY has been tested extensively, give us confidence that our resonance parameters are reliable. The authors of Ref. [32] attempted to extrapolate the data from their measurements to obtain a reaction rate at the important s process temperature of $kT=8$ keV by adjusting the capture kernels of the two most important resonances at $E_n=4.98$ and 15.14 keV to arrive at a rate in agreement with their measurement at $kT=25$ keV. Because it is not possible to obtain a unique solution for each of two capture kernels by adjusting them both to agree with the rate at a single temperature, the temperature dependence of their extrapolated rate is highly uncertain. For all these reasons, the rates determined in the present work should be much more reliable than those of Ref. [31].

To assess the impact of our new, lower rates, the s -process abundances were computed for an AGB stellar model [32], starting with a solar isotopic composition (case standard in Ref. [8]) that has been found to best describe the meteoric isotopic anomalies for the heavy elements in SiC grains. The calculated Si abundances in the stellar envelope under conditions in which SiC grains are expected to form (C>O) were used to calculate the δ values for the Si isotopes. The calculations were performed for 1.5 and 3 M_{\odot} stellar models using both our rates as well as those recommended in the most recent compilation, Ref. [30], and are summarized in Table II. There are substantial differences in the Si isotopic abundances calculated using our rates compared to those calculated using Ref. [30]. For example, our rates lead to a factor of about $\frac{1}{3}$ smaller $\delta^{29}\text{Si}$ and twice as large a $\delta^{30}\text{Si}$ compared to the calculations using the rates of Ref. [30]. As a result, the δ values calculated using our rates lie on an AGB evolution line with a slope of 0.19, much smaller than the slope of 0.74 resulting from the old rates. This situation is depicted by the arrows in Fig. 1. Also, with our new rates, the 3 M_{\odot} stellar model is 10% more efficient at producing ^{30}Si than the 1.5 M_{\odot} mass model, whereas calculations made using the old rates lead to essentially the same production in both models. The main effects of our new rates appear to be that even less ^{29}Si is produced via the $^{28}\text{Si}(n, \gamma)$ reaction, and that very little ^{30}Si (produced via a feeding from the $^{33}\text{S}(n, \alpha)$ reaction) is destroyed.

Our interpretation of these exploratory calculations sug-

gest some intriguing implications for the solution to the puzzle of the Si isotope ratios in mainstream grains. For example, the larger shift in the $\delta^{30}\text{Si}$ and smaller shift in $\delta^{29}\text{Si}$ from the ST model calculations run with our rates indicate that the solution proposed in Ref. [13] will have to be modified. In Ref. [13], average shifts in $\delta^{29}\text{Si}$ and $\delta^{30}\text{Si}$ of 20 per thousand and 25 per thousand, respectively, were calculated by averaging over the 1.5 and 3 M_{\odot} AGB s -process models. In contrast, the model ST AGB calculations using our rates yield average shifts of 6.4 per thousand in $\delta^{29}\text{Si}$ and 35.4 per thousand in $\delta^{30}\text{Si}$. As a consequence, when these averaged AGB contributions are subtracted from the $\delta^{29}\text{Si}$ and $\delta^{30}\text{Si}$ observed for mainstream grains, the resultant AGB-corrected mainstream line that results from using our new reaction rates is significantly different than obtained in Ref. [13]. This is illustrated in Fig. 1, where the corresponding s -process shifts for the different calculations is shown. The vectors depicting the corresponding s -process shifts for each calculation are starting at the corresponding mean SiC grain composition of the parent star and point to the mean composition of the observed SiC grains. Furthermore, it is expected that the shift in $\delta^{30}\text{Si}$ would be even larger if the average shift from the complete range of stellar models used in Ref. [13] was calculated using our new rates. As a result, the AGB-corrected mainstream line may be shifted even further than illustrated herein. At the very least, the AGB-corrected correlation line will no longer pass very close to solar one, as was found in Ref. [13] (suggesting that sun was formed from materials having a composition different from the mean ISM at the time of solar birth), and a different mixture of supernova sources will be needed to fit the mainstream data. The much lower slope in the three-isotope plot for the AGB s -process modification of the Si isotope ratios indicated by these exploratory calculations suggest that if the solar material had a special composition relative to the mean ISM, at solar birth, as proposed in Ref. [14], this composition is substantially different than suggested in Ref. [14].

However, the main problem remains that current standard models of the s process in AGB stars do not produce large enough shifts in $\delta^{29}\text{Si}$ and $\delta^{30}\text{Si}$ to account for the observed slope of 1.31 for the mainstream SiC grains correlation line. Therefore, the observed Si correlation in these grains must be explained by contributions of massive stars to the ISM and in some fashion reflect galactic evolution. From our data it appears that the parent AGB star cannot account for the observed spread in the Si isotopes in the mainstream SiC

grains. Nevertheless, we would like to point out two other possible means for obtaining larger shift in $\delta^{29}\text{Si}$ and $\delta^{30}\text{Si}$. First, as already discussed by Ref. [13], AGB models having larger mass or lower metallicity are able to achieve larger shifts than the ST model used herein. Using our new reaction rates may result in even larger shifts for ^{30}Si . This is already the case for the $3 M_{\odot}$ mass model, which is 10% more efficient at producing ^{30}Si than the $1.5 M_{\odot}$ mass model. Second, a larger $^{32}\text{S}(n,\gamma)^{33}\text{S}$ reaction rate could yield a higher ^{30}Si abundance in the AGB star, via feeding from the $^{33}\text{S}(n,\alpha)^{30}\text{Si}$ reaction, which has a much larger rate than the destruction reaction $^{30}\text{Si}(n,\gamma)$. It is possible that the rate for

the $^{32}\text{S}(n,\gamma)^{33}\text{S}$ reaction has been underestimated because possible DC contributions have not been taken into account.

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