

The role of Fe and Ni
for *s*-process nucleosynthesis in the early Universe
and for innovative nuclear technologies

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Abstract

The early universe was enriched in heavy elements by massive stars via their s - and r -process contributions. Ultra metal-poor stars were found to show abundance patterns that scale exactly with the solar r component. While this holds exactly for elements heavier than barium, there is still confusion about significant discrepancies in the mass region below $A \leq 120$. It is known that massive stars contribute significantly to the abundances between Fe and Zr. This so-called weak s -process component was found to exhibit large uncertainties due to the poorly known cross sections, especially in the Fe-Ni region. In view of this problem it is proposed to perform accurate state-of-the art measurements on highly enriched samples of the stable Fe and Ni isotopes at the n_TOF facility. Transformation of these results into significantly improved stellar cross section rates will allow to disentangle the s and r contributions observed in the oldest stars for a reliable comparison with galactic chemical evolution models. These results are also very important for the design of advanced reactor concepts.

I. MOTIVATIONS

A. Nucleosynthesis in the early Universe

Neutron capture nucleosynthesis in massive stars has attracted great interest since accurate abundance observations in very rare, ultra metal-poor (UMP) stars in the galactic halo became recently available through high sensitivity UV spectroscopy with the Hubble Space Telescope (HST) [1]. The present discussion of the surface composition of these UMP stars [2, 3], which represent the oldest stars in the universe, is based on the fascinating discovery, that their abundance patterns were found to scale exactly with the r -process abundance distribution of the solar system.

The solar r distribution is obtained by subtraction of the s -process component from the solar composition,

$$N_r = N_{\odot} - N_s.$$

The isotopic s abundances are well established by the fact that they are determined by their respective (n, γ) cross sections, which can be determined in laboratory experiments. Moreover, also the corresponding stellar He burning scenarios can be reliably modeled.

The perfect agreement between the scaled solar r residuals with the abundance patterns of the very metal poor halo stars was interpreted as evidence for a very robust, primary r process, independent of the metallicity of the precursor star. However, it turned out that this agreement was limited to the mass region above barium, but that the observed abundances showed systematic deficiencies of about 20% compared to the solar values as shown in Fig. 1. This discrepancy was assumed to indicate the existence of at least two different s and/or r process mechanisms, one effective in the mass region below barium and one for heavier elements [6]. There are at least two important aspects to be considered in this discussion though, (i) the role of cross section uncertainties and (ii) the fact that the s -only isotopes exhibit exactly the same deficiency as the r residuals below barium. If the missing abundances below barium are produced in a separate process, the second point is important since it strongly suggests that this must be related to an s -process type scenario. Whether an additional process is indeed required is related to the first aspect and is discussed below.

The influence of cross section uncertainties could either be directly due to the correlation with the s abundances of the main component or due to a propagation effect in the weak s -process component produced in massive stars (see below). In fact there is evidence from new

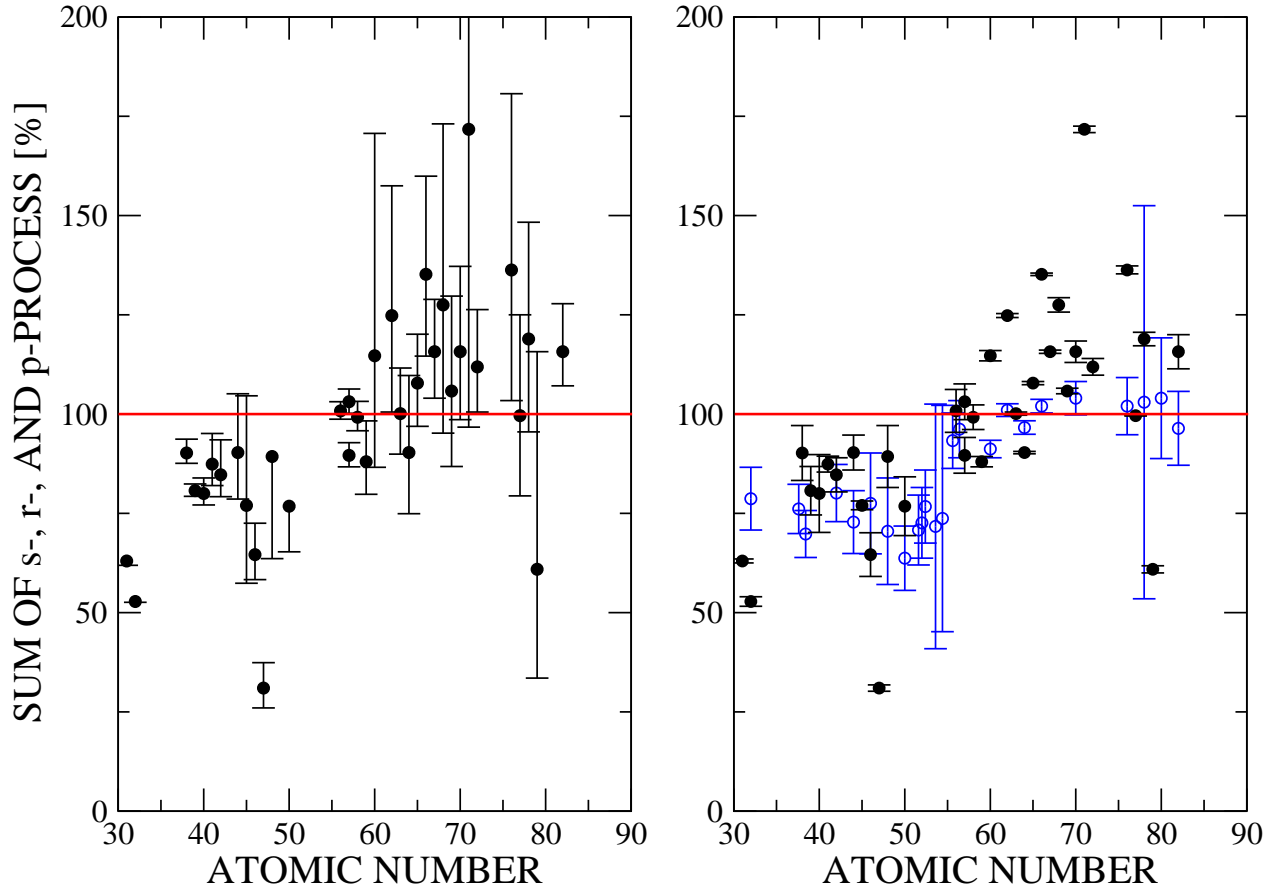


FIG. 1: Sum of the abundance contributions from the r , s , and p process, where the r -process part corresponds to the element pattern in the UMP star CS-22892-052 normalized at Eu, the s -process part has been adopted from Refs. [4, 5], and the p contribution is considered only for Mo and Ru. Uncertainties refer to the UMP observations (left panel) and to the stellar (n, γ) cross sections reflected in the s -process subtraction (right panel). The error bars in the two panels are similar in size below mass number 120, where the sum rule exhibits a 20% deficiency. Note, that the abundances of the s -only isotopes (right panel, open symbols) correlate with the overall distribution.

experimental resonance data on Zr [7] that a number of neutron capture cross sections in the mass region $A = 120$ may be systematically too high because of underestimated corrections for scattered neutrons. If confirmed by improved measurements, this would imply larger s abundances and, hence lower r residuals. In addition, the propagation effect of cross section changes in combination with a reduced effect from neutron poisons may well cause enhanced abundance contributions from massive stars up to the mass 120 region. The latter effect is

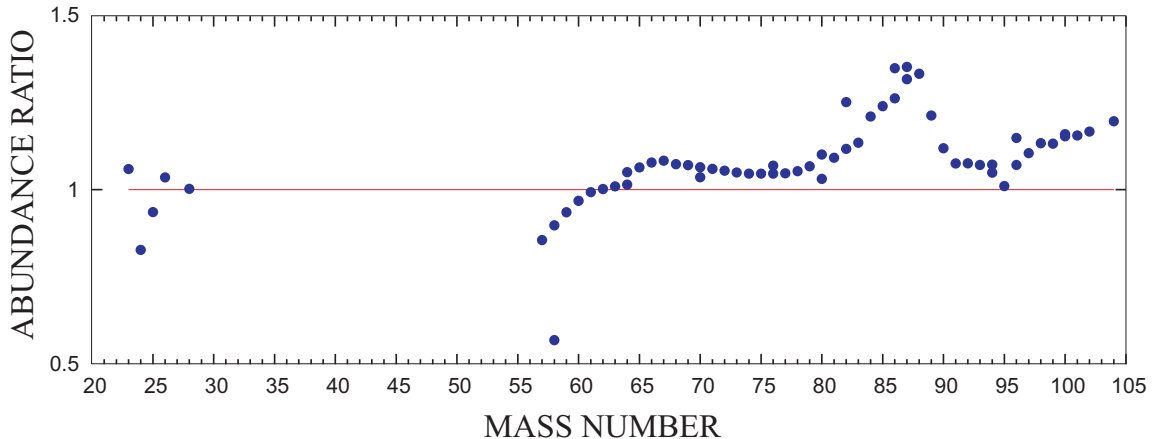


FIG. 2: Illustration of the enhanced isotope production by the weak s -process in massive stars resulting from a single neutron poison. In this case, the stellar (n, γ) cross section of ^{23}Na was arbitrarily reduced by a factor of two.

illustrated in Fig. 2, where only the reduction of the ^{23}Na cross section by a factor of two results in the strong enhancement in the abundances around the neutron magic isotopes with $N = 50$, reaching well beyond into the Mo/Ru region.

Inspection of the available stellar (n, γ) cross sections [8] shows that the data in the mass region $A \leq 120$ suffer from unacceptably large uncertainties. Moreover, the data sets from different experiments exhibit discrepancies by far larger than the quoted uncertainties. For a thermal energy of $kT = 25$ keV these discrepancies have been confirmed by the results of a series of recent activation measurements [9]. Differences of up to a factor two were found compared with the recommended values from the compilation of Ref. [8]. In this situation new accurate TOF measurements are urgently called for. While the results of activation experiments are useful complements, they are limited in two respects, i.e. to capture reactions leading to an unstable product nucleus, and to only three thermal energies, for which a quasi-stellar neutron spectrum can be simulated [10]. The complete cross section information for covering the full temperature range in massive stars can only be provided by accurate TOF data ranging from about 100 eV to 1 MeV.

As indicated above, TOF measurements in the mass region of interest are difficult because of the pronounced resonance structures, which led to severe systematic uncertainties in previous experiments due to the effect of scattered neutrons. In measurements at the n_TOF facility, this problem is successfully eliminated by unique combination of the optimized setup

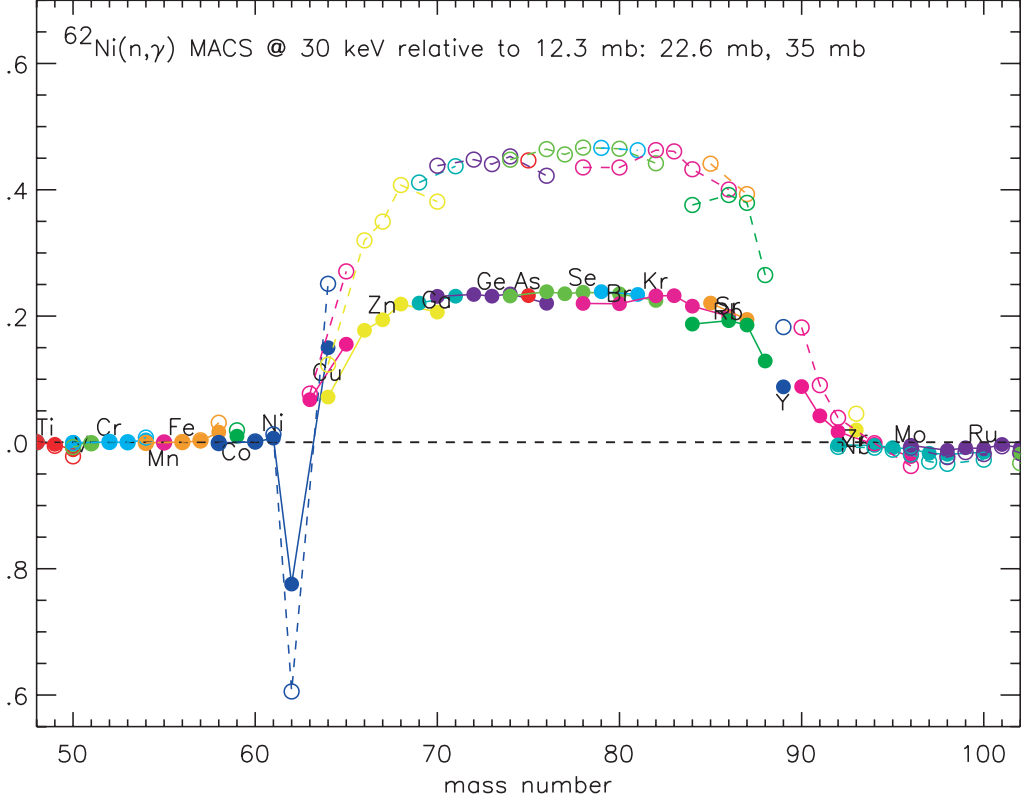


FIG. 3: The effect of cross section uncertainties on the s -process efficiency in massive stars illustrated at the example of ^{62}Ni [13] (see text).

with C_6D_6 detectors [11], the total absorption calorimeter (TAC) [12], and the excellent performance of the neutron source in terms of resolution in neutron energy, superior flux, and low repetition rate. For these reasons, CERN is ideally suited for an accurate assessment of the basic Fe and Ni cross sections required by the s -process models of massive stars.

The effect of the Fe/Ni cross sections on the s -process efficiency in massive stars is illustrated in Fig. 3 at the example of the stellar ^{62}Ni cross section. The abundances in the mass region of the weak s component produced in massive stars exhibit a pronounced propagation effect if only the previously evaluated cross section of 12 mbarn [8] of this single isotope was replaced by the 28 mbarn obtained in a recent activation measurement [13]. Since similar effects are expected from other Fe and Ni isotopes as well, the motivation for the proposed, basic measurements is obvious. The cross sections of the stable isotopes of Fe and Ni data are extremely important for the quantitative description of the s process in massive stars, i.e. for the s abundances between Fe and Y. From an experimental point of view, the TOF method is the only way (i) to provide the stellar cross sections for the full temperature

range of the s process zones in massive stars, and (ii) to determine these data, wherever the activation technique is not applicable.

B. Innovative Nuclear Technology Systems

Iron and nickel are ubiquitous as constituents of structural components in ADS (Accelerator Driven Systems) and EA (Energy Amplifier) systems. As an example, all stable isotopes of iron and nickel are considered in the reference ADS geometry used in the framework of an OECD-NEA benchmark [14], both in the composition of the fuel (cladding material) as well as in the reflector. Furthermore, one of the crucial components of the ADS system is the window separating the proton accelerator from the spallation target. According to the most recent calculations, the severe radiation damage in this window requires its periodic replacement every 3 to 6 months. Several steels are considered as candidates for constituent materials of the window and other components of the system with the aim to stabilise the mechanical and thermal properties in high radiation environments. In this context, accurate neutron cross sections are required for the most abundant elements for limiting the possible choices of structural materials and for assessing the related uncertainties.

The impact of nuclear data uncertainties on the transmutation of actinides in ADS has been recently discussed by Aliberti *et al.* [15]. The relative uncertainties in the neutron capture cross sections of the iron isotopes range from 15% (for the highest energy group from 19.6 MeV to 6.07 MeV) down to 8% in the region between 2 keV and 100 keV, and very similar uncertainties are quoted for ^{58}Ni . However, discrepancies as large as 50% for ^{54}Fe and close to 15% for ^{57}Fe and ^{58}Fe are found by comparison of the corresponding neutron capture data listed in different nuclear data libraries [16].

Finally, one should consider that the (n, γ) reactions on ^{58}Ni and ^{62}Ni leading to ^{59}Ni ($t_{1/2} = 7.5 \times 10^4$ yr) and ^{63}Ni ($t_{1/2} = 100$ yr) determine the level of short and medium term activation. For both reactions, the capture cross sections are poorly known, particularly for ^{62}Ni , where two recent measurements reported data with discrepancies of almost 50% at keV neutron energies [13, 17].

Therefore, the significant improvement of the neutron capture cross sections for the stable iron and nickel isotopes to uncertainties $\leq 5\%$ would be of great value for the accurate assessment of the system parameters and the activation of structural components in advanced

TABLE I: Isotopic enrichment, chemical form, and loan fee for the stable Fe and Ni isotopes of this proposal.^a

Isotope	Chemical form	Enrichment (%)	Loan fee/g/yr (USD)
⁵⁴ Fe	metal	99.9	48
⁵⁶ Fe	metal	99.7	23
	oxide	99.7	23
⁵⁷ Fe	metal	95.9	120
	oxide	96.6	120
⁵⁸ Fe	metal	93.2	2000
⁵⁸ Ni	metal	99.8	16
⁶⁰ Ni	metal	94.7	50
	oxide	99.6	60
⁶¹ Ni	metal	91.4	760
⁶² Ni	metal	97.7	540
	oxide	98.5	550
⁶⁴ Ni	metal	96.1	700
	oxide	88.6	592

^a Information provided by Leonid Kazakov from Obninsk.

reactor concepts as well as for resolving the discrepancies between different nuclear data libraries.

II. CROSS SECTION MEASUREMENTS

Accurate measurements are suggested at the n_TOF facility to determine the (n, γ) cross sections of the isotopes ^{54,56,57,58}Fe and ^{58,60,61,62,64}Ni with accuracies of less than 3% on average. The measurements will be performed using the C₆D₆ detector setup [11] in combination with the 4 π BaF₂ TAC array [12] to cover the relevant neutron energy range from 0.1 to 500 keV.

The measurements will be performed on metal samples with very high isotopic enrich-

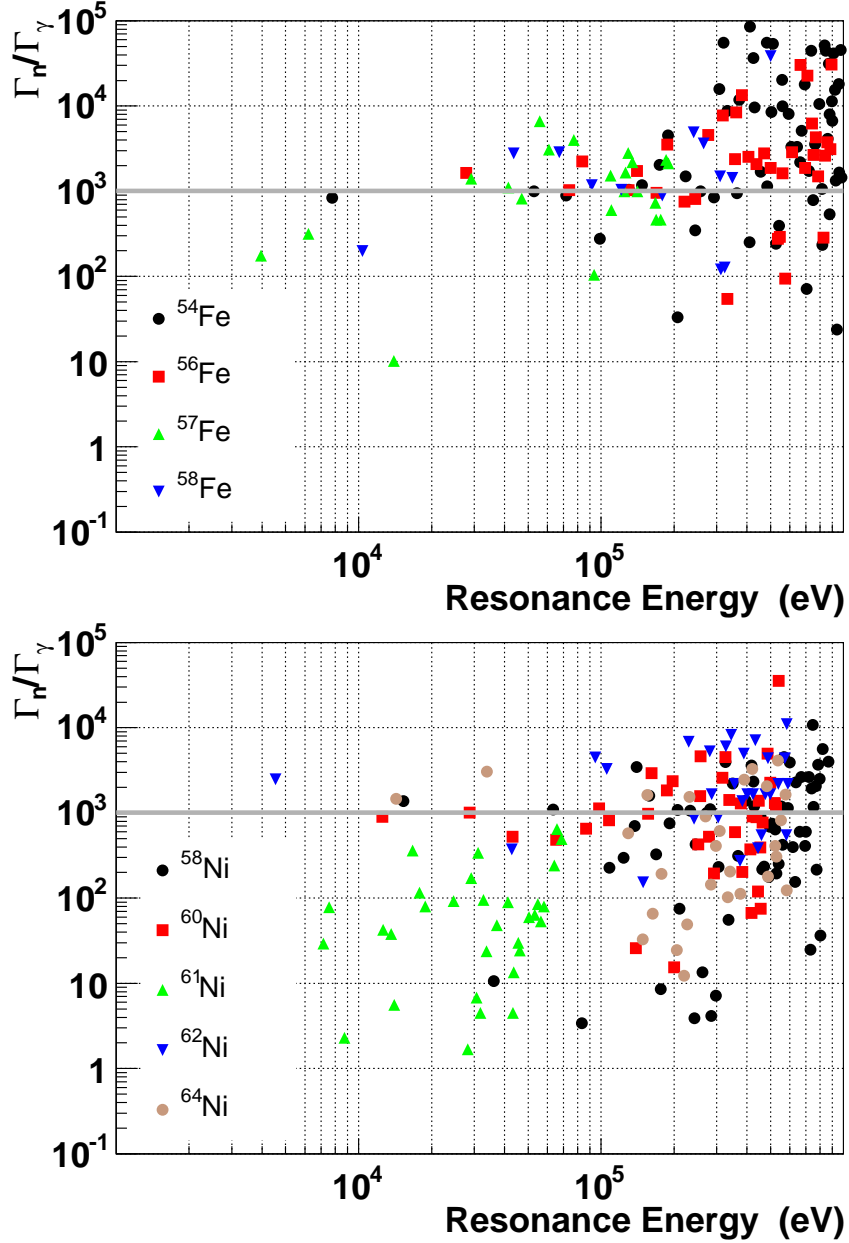


FIG. 4: Ratio of the neutron width to the gamma width for the Fe and Ni isotopes considered in this proposal. The line drawn at a ratio of 10^3 represents the lower limit where corrections due to scattered neutrons with less neutron sensitive set-ups employed in the past are important.

ment (see Tab. I). According to previous experience with n_TOF measurements on natural iron [18] samples of 20 mm in diameter and 2 g in mass can be used. More massive samples would imply large corrections for neutron multiple scattering and self-absorption and, hence, would lead to large systematic uncertainties.

In view of the different efficiency for neutron capture events of the C_6D_6 detectors ($\epsilon_c = 10$

to 15%) and the TAC ($\epsilon_c \geq 95\%$) the count rate estimates are essentially based on the performance of the C_6D_6 detectors with respect to the broad s-wave resonances. The s-wave resonances, which exhibit large scattering to capture ratios (see Fig. 4), can best be studied with C_6D_6 detectors because of their very small sensitivity for scattered neutrons [11]. The higher efficiency of the TAC and its capability to reduce gamma rays from inelastic scattering may give complementary information, in particular for weak p-wave resonances.

The calculated reaction rates are presented in Fig. 5 for a few isotopes and the choice made of sample size. Based on this calculation and on the use of C_6D_6 detectors the resonance count rates are calculated and shown in the figure as well. In order to collect sufficient statistics for accurate resonance analyses, we request a total of 2×10^{18} protons per isotope. Only in this way we can be sure to reach the 3% uncertainty, which is needed for a thorough discussion of the astrophysical consequences related to the *s*-process efficiency in massive stars and for settling the effects of structural materials in advanced concepts in nuclear technology.

III. STELLAR MODELS AND GALACTIC CHEMICAL EVOLUTION

With the improved experimental cross sections, the astrophysical consequences for the weak *s*-process component related to element production in massive stars of different mass and metallicity will be worked out. These results will then be used for a detailed discussion of the abundance patterns in UMP stars and for investigating the consequences for galactic chemical evolution.

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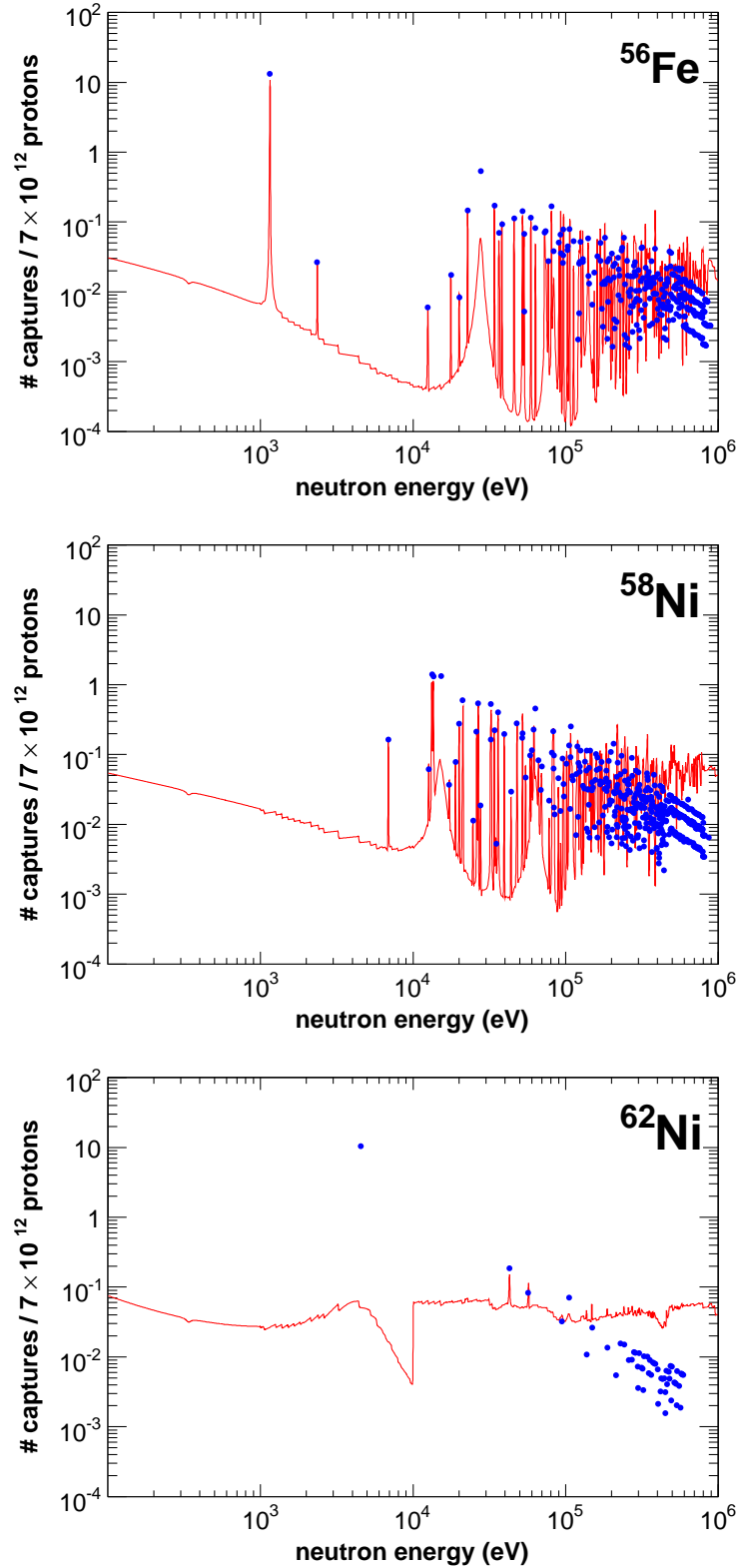


FIG. 5: The reaction-rate estimate for three typical isotopes considered in the present proposal based on the cross sections available from the evaluated nuclear data library ENDF/B-VI. Assuming a sample mass of 2 g, the estimated rates refer to a standard bunch of 7×10^{12} protons. The solid lines refer to reaction rate values calculated with 200 bins/neutron energy decade.