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Neutron capture data for unstable nuclei – Astrophysical quests and experimental challenges

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Abstract The abundances of the chemical elements heavier than iron can be attributed in about equal parts to the *r* and *s* processes, which are taking place in supernova explosions and during the He and C burning phases of stellar evolution, respectively. So far, quantitative studies of the *r*-process are out of reach, because it involves reactions on extremely short-lived neutron-rich nuclei. On the contrary, the situation for the *s*-process is far advanced, thanks to a comprehensive database of experimental (*n*, γ) cross sections for most isotopes along the reaction path from ¹²C to the Pb/Bi region. For the stable isotopes last gaps in the data are presently closed, but further studies are clearly needed to reach the required accuracy and to resolve remaining discrepancies. The quest for cross sections of unstable isotopes remains a persisting challenge though. In particular, nuclei which act as branching points are of prime interest, because they provide key information on the deep stellar interior. While the activation method is limited to a few exceptional branch-point nuclei, successful measurements via the time-of-flight technique are depending on intense pulsed neutron sources and elaborate methods for sample production. Current developments in Europe are providing promising perspectives in both areas.

Keywords Radiative neutron capture, stellar cross section data, *s*-process nucleosynthesis unstable isotopes, laboratory approaches

INTRODUCTION: ABUNDANCES AND PRODUCTION SCENARIOS

By the pioneering work of Burbidge, Burbidge, Fowler and Hoyle (B2FH) [1] and Cameron [2] the origin of the heavy elements from the Fe group to the Pb/Bi region and up to the actinides has been attributed to the slow (*s*) and rapid (*r*) neutron capture processes, which are termed according to their typical neutron capture times being slow or rapid compared to average β -decay half-lives. Meanwhile, astrophysical scenarios are clearly identified for the *s* process, which takes place during the He and C burning phases of stellar evolution [3, 4]. For the *r* process, supernova explosions appeared to be the most promising sites, but as a conclusive model is still missing, neutron star mergers are being considered as alternative scenarios [5].

While the *s* and *r* process are holding about equal shares of the observed solar-system abundances between Fe and U [6], a third process, the so-called *p* (proton capture or photodissociation) process, has been invoked for explaining the origin of about 32 rare, proton-rich nuclei, which, however, contribute less than 1% to the observed abundances in general.

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Among these processes, only the *s* process has been established quantitatively, yielding rather detailed information concerning the *s* component in the solar system abundances as well as on the role of the *s* process in galactic chemical

Fig. 1. The main neutron-capture processes responsible for the observed abundances between iron and the actinides. The reaction path of the *s* process follows the stability valley, while the *r*-process path is shifted to very neutron-rich nuclei, which later decay (dashed arrows) and are mixing with the *s* component. Note also the occurrence of local branchings in the *s*-process path (circles) at unstable isotopes with similar neutron capture and β decay rates.

evolution. In fact, two different *s* components have been identified, the main and the weak *s* process, which are ascribed to specific phases of stellar evolution in stars of different mass.

The main *s* process occurs in the He-burning layers of asymptotic giant branch (AGB) stars with $M \leq 3M_{\odot}$ [3, 7] and contributes predominantly to the mass range between ^{90}Zr and ^{209}Bi . Neutrons are alternately produced by (α, n) reactions on ^{13}C and ^{22}Ne at average neutron densities of about 10^7 and 10^8 cm^{-3} , respectively.

The weak *s* component, which contributes only to the abundances between Fe and Zr, takes place in massive stars ($M > 8M_{\odot}$) during convective core-He burning and subsequently during convective shell-C burning [4, 8]. The more efficient neutron exposure during shell-C burning is characterized by fairly high neutron densities, which start at about $10^{11} - 10^{12} \text{ cm}^{-3}$ and then decrease exponentially.

In contrast to the *s* process, where neutron capture times of the order of days to years confine the reaction path to or close to the valley of β stability, extremely high neutron densities of $> 10^{22} \text{ cm}^{-3}$ are reached in the *r* process, giving rise to capture times of the order of milliseconds, indicating an explosive scenario for the *r* process either related to supernovae or neutron star mergers. In either case, the extreme neutron densities imply that (n, γ) reactions are becoming much more rapid than β decays, thus driving the reaction path close to the neutron drip line at

the limits of stability. After the explosion, the initial, short-lived reaction products decay to the valley of stability.

The reaction paths of the *s* and *r* process are sketched in Fig. 1. The neutron-rich isotopes outside the *s* path can be ascribed to the *r* process. Apart from these *r*-only isotopes the *r* process contributes also to most of the other isotopes, except for those, which are shielded by stable isobars. The corresponding ensemble of *s*-only isotopes is important for the separation of the respective abundance distributions and, more importantly, to set constraints for the overall *s*-process efficiency and for the parameters governing the abundance pattern in *s*-process branchings. The subset of *r*-only nuclei can be used as calibration points for the overall *r*-process distribution. The stable *p* isotopes on the proton-rich side, which cannot be produced by neutron reactions, are also ascribed to supernova explosions [9].

It is to be noted for all *s*-process scenarios that the reaction path is confined to the valley of β stability, because neutron capture times are on average much longer than β half-lives. This feature implies that the evolving *s* abundances are strongly correlated with the respective neutron capture cross sections, which, therefore, constitute the most important nuclear physics input for any *s*-process study.

Neutron reactions and available data

The neutron spectrum typical of the various *s*-process sites is described by a Maxwell-Boltzmann distribution, because neutrons are quickly thermalized in the dense stellar plasma. The effective stellar reaction cross sections are therefore obtained by averaging the experimental data over that spectrum. The resulting Maxwellian averaged cross sections (MACS)

$$\langle \sigma \rangle_{kT} = \frac{2}{\sqrt{\pi}} \frac{\int_0^\infty \sigma(E_n) E_n e^{-E_n/kT} dE_n}{\int_0^\infty E_n e^{-E_n/kT} dE_n}$$

are commonly compared for a thermal energy of $kT = 30$ keV, but for realistic *s*-process scenarios a range of thermal energies has to be considered, from about 8 keV typical of the $^{13}\text{C}(\alpha, n)$ source in low mass AGB stars to about 90 keV during shell-C burning in massive stars. To cover this full range, energy-differential cross sections $\sigma(E_n)$ are needed in the energy region $0.1 \leq E_n \leq 500$ keV.

Although an extensive set of experimental cross sections is meanwhile available, the situation is far from being satisfactory. The data for many of the stable isotopes do not meet the required accuracy of $\leq 5\%$, are discrepant, or cover only part of the energy range of interest. For the set of unstable nuclei, which play an essential role in understanding the *s*-process branchings, reliable data are yet largely missing, because they are very difficult to access experimentally.

The current status has been summarized in the KADoNiS compilation [10, 11], where data sets for 356 isotopes, including 77 radioactive nuclei on or close to the

s-process path are listed. While measurements exist for the (n, γ) cross sections of almost all 277 stable isotopes, experimental data are available for only 14 radioactive nuclei, including the branch points ^{63}Ni , ^{129}I , ^{135}Cs , ^{147}Pm , ^{151}Sm , ^{154}Eu , and ^{163}Ho . However, most measurements on unstable isotopes have been carried out via activation in a quasi-stellar spectrum for $kT = 25$ keV [12], thus requiring extrapolation to lower and higher temperatures by means of theoretical cross sections obtained with the Hauser-Feshbach statistical model that are typically affected by uncertainties of 25 to 30%.

Fig. 2. The *s*-process network in the Sm-Eu-Gd region with branchings of the reaction path at ^{151}Sm , $^{152,154,155}\text{Eu}$, and ^{153}Gd . Due to the competition between neutron capture and β -decay, the abundance patterns of branchings carry important information on neutron flux and temperature at the *s*-process site.

The MACS values of unstable isotopes are crucial for the interpretation of *s*-process branchings as illustrated in Fig. 2. The local abundance pattern produced by the branchings is reflecting the stellar neutron flux that determines the neutron capture flow at the branching point compared to the strength of the β -decay channel. In some cases, however, this mechanism is more complicated, because the effective β -decay rate can be enhanced at stellar temperatures. The branching at ^{151}Sm is of particular interest because the 93 yr half-life of ^{151}Sm decreases by about a factor of 30 in an *s*-process environment [13].

Accordingly, branchings can be considered as diagnostic tools for the stellar interior, provided that temperature and neutron flux can be constrained by reconstructing the abundance pattern on the basis of reliable cross section data for the involved isotopes.

Laboratory work: activation vs. time of flight

Stellar (n, γ) cross sections are commonly measured either by activation in a quasi-stellar neutron field [12] or using the time-of-flight (TOF) technique. The activation method has the advantage of superior sensitivity, thus allowing one to limit the sample material to sub- μg amounts and to avoid excessive backgrounds

from the activity of the sample. For most branch points, however, activation is not applicable, because they are situated between stable isotopes (see ^{151}Sm in Fig. 2).

In view of the relevant keV energies, TOF measurements of stellar cross sections depend on accelerator-based neutron facilities, using either moderated sources produced by high-energy pulsed electron or proton beams or neutrons directly originating from (p, n) reactions with low-energy protons. While moderated sources, e.g. GELINA at Geel/Belgium [14, 15], n_TOF at CERN [16], or LANSCE at Los Alamos [17], cover very wide neutron energy ranges from thermal to nearly the initial energy of the charged particle beam, the neutron spectra produced with low-energy machines can be tailored to the astrophysically relevant keV region, e.g. at the FRANZ facility in Frankfurt/Germany [18, 19].

Fig. 3. Integrated neutron flux in the astrophysically relevant energy interval from 10 to 100 keV at existing TOF facilities and at the future FRANZ accelerator.

Compared to activation measurements the sensitivity of the TOF method suffers from the lower (pulsed) beam currents and from the (much) larger distance between neutron source and sample. These drawbacks must be compensated by increasing the sample mass, by increasing the peak currents of the pulsed neutron sources, or by reducing the flight path. Because the sample mass for unstable isotopes is limited by the specific γ -activity, neutron flux and flight path have to be optimized instead. The key parameter for TOF experiments is the achievable flux at the sample position, which is compared in Fig. 3 for the main existing and future facilities. The expected performance of FRANZ is based on the experience with the Karlsruhe Van de Graaff accelerator, scaled with the beam current on target.

While the 20 m stations at LANSCE and n_TOF represent the best choices among the moderated sources, the shorter flight path at FRANZ of only 1 m will make this design a very attractive option for measurements on unstable samples. Note, however, that additional aspects for such measurements, e.g. the resolution in

neutron energy, the effect of different repetition rates, γ -flash intensities, and facility-specific backgrounds are not considered in this comparison.

Outlook

In summary, intense pulsed sources of keV neutrons are instrumental for a wealth of applications in astrophysics, especially for cross section measurements on unstable isotopes. At present, TOF measurements on a range of *s*-process branch point isotopes appear to be feasible and are partially under investigation at n_TOF/CERN [20]. Apart from these *s*-process aspects, there is also a multitude of requests for MACS data concerning other scenarios with even higher neutron densities. Under certain conditions, stars may experience convective-reactive nucleosynthesis episodes if H-rich material is convectively mixed into a He-burning zone [21, 22], resulting in neutron densities up to about 10^{15} cm^{-3} , a situation intermediate between the *s* and the *r* process. Accordingly, the neutron-capture path is shifted by a few mass units from the valley of stability, as neutron capture times are only minutes or hours. Explosive nucleosynthesis in the *r* and *p* process are another field, where neutron reactions on unstable isotopes are of key importance, essentially during the freeze-out phase when free neutrons are captured by the primary reaction products.

The data requests for these explosive scenarios are yet outside the reach of existing facilities. Once FRANZ starts operation, it might be possible to reduce the flight path to about 10 cm, thus extending the possibilities for measurements on unstable samples significantly. Recently, neutron-induced reactions in inverse kinematics have been proposed [23] by combining a beam of radioactive ions cycling in a storage ring similar to the CRYRING at GSI/FAIR [24] with a reactor core or a cold neutron beam as the neutron target. This method would also provide an option for unstable isotopes with half-lives below about a week, too short for measurements with stationary samples.

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