Nuclear Instruments and Methods in Physics Research A 613 (2010) 343–346

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Radioactive target needs for nuclear reactor physics and nuclear astrophysics

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ARTICLE INFO

Available online 6 October 2009

Keywords:
Radioactive targets
Nuclear data
Element nucleosynthesis
Fission reactors

ABSTRACT

Nuclear reaction cross sections of short-lived nuclei are key inputs for new generation nuclear reactor simulations and for models describing the nucleosynthesis of elements. After discussing various topics of nuclear astrophysics and reactor physics where the demand of nuclear data on unstable nuclei is strong, we describe the general characteristics of the targets needed to measure the requested data. In some cases the half-life of the nucleus of interest is so short that it is not possible to produce a target and perform the measurement. However, some alternative methods have been developed that allow one to obtain neutron-induced cross sections of highly radioactive nuclei. One of these methods is the surrogate reaction technique. We explain the principle of the surrogate method and describe the characteristics of the targets used in surrogate experiments.

1. Introduction

Even though nuclear astrophysics and reactor physics may appear to be rather distant topics at a first view, they have important aspects in common. Both topics are strongly based on computer simulations or models for which the availability of reliable nuclear reaction data is of crucial importance. Reactor physics uses transport codes, e.g. MCNP [1], where the history of the interaction of single neutrons with the different nuclei present in the reactor core is simulated for their complete life cycle ranging from their generation in a nuclear fission process until they diminish due to nuclear reactions. One of the primary goals of nuclear astrophysics is to obtain a detailed understanding of the synthesis of all the elements which make up our world. Theories of nucleosynthesis are tested by calculating isotope abundances with rather complex stellar models and comparing them with observed results. Both topics have also in common that neutron-induced reactions play an essential role. Although in nuclear astrophysics proton-, alpha- and gamma-induced reactions are also important. Typical paths for the formation of nuclei in a reactor or in a star are one or more neutron captures followed by β^- decay. When the nuclei are heavy enough, the interaction with neutrons can lead to fission where the initial nucleus splits into two lighter nuclei. Thus, neutron-induced capture and fission reactions are highly interesting for both topics. Of course, the characteristics (temperature, neutron density, etc.) of a reactor core and those of a star may be very different. Therefore, the neutron-energy domains of interest in astrophysics and in reactor physics can be rather distinct. Finally, another similarity between both topics is the need of cross section measurements of unstable nuclei. Very often the production of a radioactive target as well as the realisation of the measurement in an adapted facility is extremely challenging.

2. Radioactive nuclei of interest in nuclear astrophysics

Elements heavier than ^{56}Fe can be assembled within stars by a slow neutron capture process known as the s-process or in explosive environments, such as supernovae, by the so-called r-process that involves rapid neutron captures and the rp-process, which involves rapid proton captures [2]. In the rp-process consecutive proton captures onto seed nuclei followed by β^+ decay lead to heavier elements. This process occurs in an environment with a high rate of proton captures and high temperatures. The nuclei formed are localised in the neutron-deficient side of the chart of nuclei, see Fig. 1. The s-process occurs at relatively low neutron density and intermediate temperature conditions in stars. As illustrated in Fig. 1, this process produces stable isotopes up to Bi by moving along the valley of beta stability. Heavier nuclei such as thorium or uranium are produced thanks to the r-process. Immediately after a core-collapse supernova, there is an extremely high neutron flux and temperature, such that neutron capture occurs much faster than β^- decays, meaning that the r-process evolves along the neutron drip line, see Fig. 1. Consequently, nucleosynthesis models require proton- and neutron-induced capture and fission cross sections of radioactive nuclei situated in the three regions depicted in Fig. 1.

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doi:10.1016/j.nima.2009.09.071

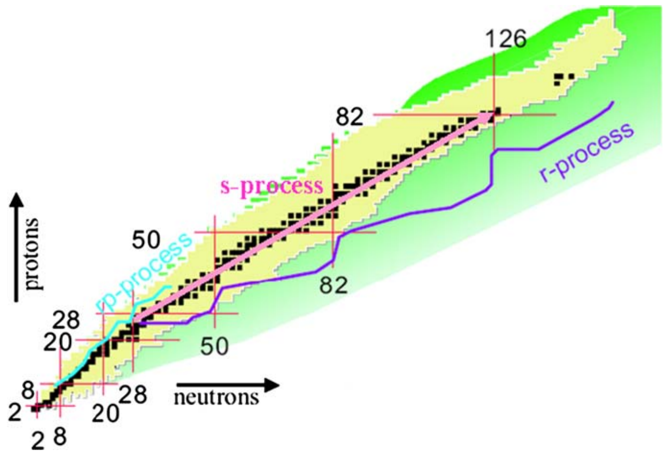


Fig. 1. (Colour online) Chart of nuclei representing the different element nucleosynthesis processes. The nuclei located on the valley of beta stability are represented by the black squares. The rapid proton capture process (rp-process) on the neutron-deficient side of the valley of stability is represented by the light blue line, the slow neutron capture process (s-process) on the stability valley is illustrated by the pink arrow and the rapid neutron capture process (r-process) on the neutron-rich side of the stability valley by the blue line. Figure adapted from Ref. [3]. For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.

Clearly, there is no way to have targets of nuclei directly involved in the r-process and the heavier part of the rp-process. For this reason the experiments aiming at measuring data close to the proton and the neutron drip lines are rather based on the use of radioactive beams in inverse kinematics. On the contrary, for the s-process, there exist very specific demands where the radioactive targets required are still feasible. Indeed, reliable neutron-induced capture cross sections on branching points are crucial for testing and constraining stellar models in terms of the basic physical s-process parameters, i.e. neutron density, temperature, and pressure. Branching points are nuclei with half-lives comparable to the neutron capture time scale, which give rise to branchings in the reaction path. According to the review article by Käppeler and Mengoni [4], the main requests are ^{79}Se, ^{85}Kr, ^{147}Pm, ^{151}Sm, ^{163}Ho, ^{170}Tm, ^{171}Tm, ^{179}Ta, ^{204}Tl and ^{205}Pb. The half-lives of these nuclei vary from 129 days for ^{170}Tm to 1.5×10^7 years for ^{205}Pb. These nuclei are β^+ or β^- emitters, which in some cases emit gamma rays subsequent to β decay. The different possible s-process temperatures require the experimental neutron capture cross sections to cover a wide energy range, from about 300 eV to several hundred keV. Besides the branching points, other neutron capture cross sections of unstable nuclei are also important. The study of ^{60}Fe(n,γ) has been identified to be of great interest for at least two reasons: (i) ^{60}Fe has a half-life of 1.5 million years and was found in deep layers of oceanic crust sediments. It is assumed that a nearby supernova which exploded 2.8 million years ago is responsible for the excess of ^{60}Fe. In order to determine the locus of this event in the solar system it is necessary to investigate the production (via ^{59}Fe(n,γ)^{60}Fe) and destruction mechanism (via ^{60}Fe(n,γ)^{61}Fe) of ^{60}Fe. Furthermore, (ii) ^{60}Fe is one of the few, long-lived γ-ray emitters that could be observed with γ-ray telescopes like INTEGRAL after supernovae explosions. Therefore, the production and destruction of ^{60}Fe is important to compare the observed amount of ^{60}Fe and the calculated values from supernovae models. The preparation of a ^{60}Fe target is treated in Ref. [5].

3. Radioactive nuclei of interest in nuclear reactor physics

In this work we concentrate on two topics that demand strong efforts in terms of nuclear data measurements: minor actinides incineration and the development of the Th–U cycle. Minor actinides (mainly Np, Am and Cm isotopes) are produced by successive neutron captures, beta and alpha decays starting from ^{238}U in the current U–Pu cycle. Because of their very high radiotoxicity, these nuclei represent one of the most harmful types of nuclear waste. At present, two different strategic approaches are proposed for minor actinides waste disposal: direct disposal without any reprocessing and spent fuel reprocessing with the aim to optimise the extraction of minor actinides and then incinerate them. Incineration amounts for the transmutation of minor actinides into less radiotoxic or short-lived species obtained by neutron-induced fission reactions. The reliable design of reactors for incineration requires an accurate knowledge of minor actinides cross sections in a fast neutron flux. However, the available data are rather scarce. The reason for this lack of data is the short half-life of many of these nuclei, which makes it very difficult to produce and to manipulate targets of these isotopes. Reactor simulations require principally neutron-induced fission (n,f), capture (n,γ), elastic (n,n), and inelastic (n,n′) cross sections. However, since the neutron-energy regime of interest extends from few eV to about 20 MeV, also two-neutron emission (n,2n) cross sections are needed. Moreover, other data such as fission fragment isotopic yields, average number of neutrons per fission or number of delayed neutrons are highly interesting because these quantities strongly affect the reactor neutron flux. For most minor actinides the existing fission data are of limited quality. These fission cross sections are often measured with uncertainties greater than 20% and important discrepancies between different sets of data are observed. The situation is even worse for other decay channels such as (n,γ), (n,n′) or (n,2n) where no data at all are available. Some of the minor actinides of interest are ^{237}Np, ^{240,241,242,242m,243}Am and ^{242,243,244,245}Cm. The half-lives of these nuclides vary from 16 h for ^{242}Am to 2.14×10^6 years for ^{237}Np. Among these nuclei we find several β^- emitters but the majority of them are strong alpha emitters and some may even fission spontaneously, which implies the emission of neutrons. Also γ- and X-rays can be emitted. This illustrates how difficult it can be to make and handle targets of these nuclei.

Another issue of interest in reactor physics is the development of the Th–U fuel cycle. The underlying main idea is to replace the ^{238}U of the current fuel by ^{232}Th. Similarly to ^{238}U, ^{232}Th is a fertile nucleus. Indeed, a neutron capture on ^{232}Th followed by two β^- decays leads to the formation of the fissile nucleus ^{233}U. The use of ^{232}Th as a fuel has the advantage that it increases the resources (Th is more abundant than U by a factor 3 or 4). Moreover, the radiotoxicity of the generated nuclear waste is reduced. Since Th is lighter than U one needs more neutron captures to produce minor actinides. The nuclei of interest for the Th–U cycle are ^{232}Th and ^{233}U and several neighbouring isotopes as ^{231}Th, ^{231,233}Pa and ^{232,233,234,235}U. They are mainly β^- and alpha emitters with half-lives varying from 25.52 h for ^{231}Th to 1.4×10^{10} years for ^{232}Th. Whereas for the current U fuel cycle an enormous effort has been undertaken concerning experimental nuclear data, this is not at all the case for the Th–U cycle for which the data are not precise enough or simply missing. Even for the long-lived ^{232}Th the fission cross section needs to be considerably improved between few keV and 1 MeV neutron energy.

4. Target characteristics

The target characteristics depend strongly on the type of measurement, therefore we distinguish between three main types of experiments: neutron-induced capture, neutron-induced fission and indirect measurements with light charged particle beams based on the surrogate reaction method.

4.1. Neutron-induced capture cross sections measurements

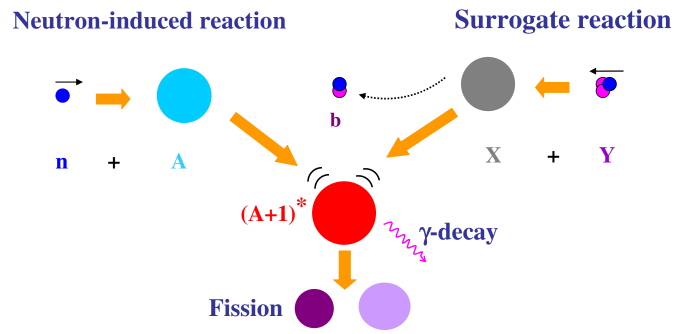
Due to the low neutron fluxes and the reduced gamma detection efficiency, samples of few grams are generally required for this type of experiments. However, the radioactivity of unstable nuclei may limit considerably the sample quantity, which has to be compensated with a particularly efficient γ-detection set-up and an intense neutron source. For example recently, the (n,γ) cross section of ^{241}Am ($T_{1/2}$=432.6y) has been measured at LANSCE in Los Alamos National Laboratory using the DANCE γ-ray array and a sample of only 219 $\pm$ 1 μg [6]. Very often the sample is in form of powder pressed into a container made of Al or Ti [7]. However, in some cases thin targets are used piled-up in a compact manner inside a container [4,8]. A precise knowledge of the isotopical composition of the sample is very important in order to correct for the background generated by possible contaminants.

4.2. Neutron-induced fission cross sections measurements

Fission fragments are heavy nuclei that may lose important amounts of energy when traversing a material. In addition, due to momentum conservation, the two fragments are emitted back-to-back in opposite directions. Therefore, in fission experiments targets should be thin enough to allow at least one of the fission fragments to reach the fission detector. Typical target thicknesses vary between 200 and 500 μg/cm^2 [9]. The sample is evaporated or electrodeposited onto a support. Typical support materials are Al, Ni or Ti. The thickness of the support ranges between few μm to 1 mm. Thin supports of few μm are used if the experiment requires the two fission fragments to be detected. Special care should be made to avoid heavy contaminants that fission, in particular neighbouring isotopes that may have not been completely separated from the raw radioactive material. The size of the targets is very variable with diameters ranging from few mm to various cm [10]. The size depends mainly on the diameter of the neutron beam. For small samples an homogeneity better than 90% is required, for large targets one needs to measure how the matter is distributed all over the target surface. For many actinides this can be done via alpha cartography or radiographic imaging [11].

4.3. Indirect measurements via the surrogate reaction method

In some cases the activity of the targets is such that the fabrication and the manipulation of the sample are impossible due to a lack of availability of the target material or its short half-life. Alternative indirect methods have been developed to overcome these difficulties. One of these methods is the surrogate reaction technique. This method was developed in the 70's by Cramer and Britt [12]. Given a desired neutron-induced reaction n+A that proceeds through a compound nucleus $(A+1)^*$, the surrogate reaction method consists of producing compound nucleus $(A+1)^*$ via an alternative (surrogate) reaction, e.g. a few-nucleon transfer reaction $y+X\rightarrow(A+1)^*+b$ and measuring the decay (e.g. fission or radiative capture) of the compound nucleus $(A+1)^*$ in coincidence or not with the ejectile b. The ratio between the number of decays detected in coincidence with the ejectile (corrected for the decay-detector efficiency) and the total number of ejectiles gives the decay probability of the compound nucleus $(A+1)^*$. The neutron-induced cross section for the corresponding decay channel is then deduced from the product of the decay probability measured in the surrogate reaction and the compound nucleus cross section for the neutron-induced reaction. The latter cross section is obtained from optical model calculations. The principle of the surrogate method is illustrated in Fig. 2. The conditions under which the surrogate method can be applied to infer neutron-induced cross sections have been investigated in Refs. [13,14].



$$\sigma_{n,i}(E_n) = \sigma_{CN}^{cal}(E_n)\cdot P_i^{\exp}(E_n)$$

Fig. 2. Schematic representation of the surrogate method compared with the direct neutron-induced reaction. In this case the surrogate reaction is a transfer reaction. Two possible decay modes "i" (fission and gamma emission) of nucleus $(A+1)^*$ are also represented. The neutron-induced cross-section $\sigma_{n,i}$ for decay channel "i" is obtained by multiplying the calculated cross-section for the formation of nucleus $(A+1)^*$ through neutron absorption σ_{CN}^{cal} with the measured decay probability P_i^{exp}.

The surrogate reaction method presents several advantages:

- The most important: in some cases the target X of the surrogate reaction is less radioactive and easier to produce than the target A required in the neutron-induced measurement. We will see some examples below.
- In charged particle-induced transfer reactions different combinations of nucleons can be exchanged between projectile and target leading to the production of various residues. In contrast to neutron-induced experiments, the simultaneous access to several transfer channels allows one to determine cross sections of various nuclei from just one projectile–target combination.
- Since there are two bodies in the outgoing reaction channel, the excitation energy of the compound nucleus E^* follows a broad probability distribution. The E^* is obtained by measuring the kinetic energy and the angle of the ejectile. The compound nucleus excitation energy can be translated into neutron energy E_n via the relation $E^*=B_n+AE_n/(A+1)$, where B_n is the neutron binding energy of the compound nucleus. Therefore, with fixed beam energy, the surrogate method allows the determination of cross sections over a wide range of neutron energy.

This technique has been applied to the measurement of the neutron-induced fission [15] and capture [16] cross sections of ^{233}Pa (half-life of 27 days) via the transfer reaction ^{232}Th (^{3}He, p)^{234}Pa. The ^{232}Th ($T_{1/2}$=1.4 $\times$ 10^{10} y) target was produced at the SIDONIE separator of the CSNSM. Recently, the surrogate reaction method has been used to determine the neutron-induced fission cross sections of ^{242}Cm($T_{1/2}$=163 d), ^{243}Cm($T_{1/2}$=29.1 y) and ^{241}Am($T_{1/2}$=432.2 y) with a ^{243}Am($T_{1/2}$=7370 y) target and a ^{3}He beam [17]. Two ^{243}Am targets of approximately 106 μg/cm^2 were prepared at the Argonne National Laboratory. Each target was electrodeposited on a 75 μg/cm^2 carbon backing. The surrogate method has been shown to be very successful for extracting (n,f) cross sections of unstable actinides. This technique in principle can be also used to infer neutron capture cross sections. However, this application of the surrogate technique has still to be validated. In Ref. [18] the possibility of using the surrogate method to infer capture cross sections of s-process branching nuclei has been discussed.

The target thickness is limited by the sample activity and by the high elastic scattering cross section of the beam particle on the target nucleus that may cause detector saturation. In fission measurements one may need to detect the two fission fragments in coincidence with the ejectile, this requirement also constrains the target thickness. However, the reduced sample thickness can be largely compensated by the high intensity of the charged particle beam. Typical target thicknesses range between 100 and 250 $\mu g/cm^2$. The sample is evaporated/electrodeposited onto a thin C support (50–100 $\mu g/cm^2$) [19]. The thickness of the support determines the energy straggling of the light ejectiles and thus the energy resolution associated to the surrogate experiment. A thin support is also required to limit the background generated by reactions between the projectile and the nuclei of the backing. The isotopic purity of the sample is extremely important. Neither heavy nor light contaminants should be present since reactions on the contaminants can falsify the number of coincidences decay particle-ejectile and the total number of ejectiles that are used for determining the decay probability. This background is extremely difficult if not impossible to remove.

5. Summary and conclusions

Neutron-induced cross sections of short-lived nuclei are of great importance in reactor physics and nuclear astrophysics. In this work we have outlined some of the regions of the chart of nuclei of interest for both topics. The main target characteristics for neutron-induced capture and fission measurements have been given. The surrogate reaction method, an indirect technique to determine neutron-induced cross sections of unstable nuclei, has been presented together with a description of the required target characteristics. The further development of research domains such as nuclear waste transmutation or the understanding of element nucleosynthesis is strongly linked to the availability of radioactive targets. Therefore, an effort should be made to maintain the current expertise and improve the existing techniques for radioactive target preparation.

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