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Nuclear Instruments and Methods in Physics Research A 556 (2006) 547–555
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
www.elsevier.com/locate/nima

Thermal neutron capture cross-section measurements of ^{243}Am and ^{242}Pu using the new mini-INCA α- and γ-spectroscopy station

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Received 22 September 2005; received in revised form 27 October 2005; accepted 27 October 2005
Available online 29 November 2005

Abstract

In the framework of the Mini-INCA project, dedicated to the study of Minor Actinide transmutation process in high neutron fluxes, an α- and γ-spectroscopy station has been developed and installed at the High Flux Reactor of the Laue-Langevin Institut. This set-up allows short irradiations as well as long irradiations in a high quasi-thermal neutron flux and post-irradiation spectroscopy analysis. It is well suited to measure precisely, in reference to ^{59}Co cross-section, neutron capture cross-sections, for all the actinides, in the thermal energy region. The first measurements using this set-up were done on ^{243}Am and ^{242}Pu isotopes. Cross-section values, at $E_n = 0.025$ eV, were found to be (81.8 ± 3.6) *b* for ^{243}Am and (22.5 ± 1.1) *b* for ^{242}Pu. These values differ from evaluated data libraries by a factor of 9% and 17%, respectively, but are compatible with the most recent measurements, validating by the way the experimental apparatus.

PACS: 25.40.Lw; 28.41.Kw; 29.30.Ep; 29.30.Kv

Keywords: Neutron; Cross-section; ^{243}Am; ^{242}Pu; Spectroscopy; Nuclear waste; Transmutation

1. Introduction

One of the crucial challenges for the nuclear industry in this century is the capability to reduce and to manage the mass inventory and the radio-toxicity of nuclear wastes coming from spent fuel of nuclear power plants. One of the field of research presently under investigation for high-level activities and long-lived nuclear wastes by the scientific community is the transmutation of long-lived isotopes into shorter lived or stable ones. The incineration of nuclear wastes requires not only innovative technological solutions, necessary to built systems providing intense neutron fluxes ($>10^{15}$ n/cm^2/s), but also a reliable set of nuclear data. Accurate fission cross-sections are necessary to estimate the incineration potential of such systems but not only. Accurate neutron capture cross-sections are also needed to follow and to predict precisely the transmutation chain. These data are needed for a wide energy range including the thermal energy region.

Due to their limited role in conventional fuel cycles, minor actinides have not been extensively studied in the past few decades. Their nuclear parameters, tabulated in the three most widely used evaluated nuclear data libraries, JEFF [1], ENDF [2] and JENDL [3], are not always known with the precision required for transmutation dedicated systems [4]. This statement is especially valid in the case of ^{242gs}Am for which a significant discrepancy of a factor of 20 existed between JEF-2.2 (JENDL-3.2) and ENDFB-VI libraries in the capture cross-section value. A recent measurement [5] has confirmed the ENDF-BVI value and that the transmutation of ^{241}Am in an high thermal neutron flux is theoretically feasible.

In this context, and following the measurement of the ^{242gs}Am capture cross-section [5], the mini-INCA (INcineration of Actinides) project has been created [6]. The

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doi:10.1016/j.nima.2005.10.125

objective of the project is to extend the field of investigations to all other minor actinides (1) which play an important role in the nuclear waste inventories for future nuclear energy production scenarios [4], and (2) for which nuclear parameters are determined with significant discrepancies between different libraries [7]. The first two isotopes examined in this project are ^{243}Am and ^{242}Pu as they contribute significantly to the mass inventory of the nuclear spent fuel and as they play a significant role in the ^{241}Am transmutation chain. Indeed, ^{243}Am and ^{242}Pu are connected to form ^{244}Cm isotope, which constitutes one of the major problem when dealing with the handling and the stocking of spent fuel. Moreover, ^{244}Am isotopes have very large fission cross-sections and could then participate to the incineration process of ^{241}Am. But, due to their very short lifetime, fission process is extremely disadvantaged as compared to β-decay process. In consequence, very high neutron fluxes are needed to get a significant contribution of the fission process, and thus, the economy of the incineration system strongly depends on the values of the cross-sections and their accuracy. For ^{243}Am and ^{242}Pu isotopes, discrepancies of about 4% exist on the capture cross-sections between the main data libraries and larger discrepancies for experimental data.

This paper will describe the new experimental installation of the Mini-INCA project at the high flux reactor of ILL, dedicated to off-line α and γ spectroscopies of irradiated samples, and present the results obtained on the capture cross-sections of ^{243}Am and ^{242}Pu. It is divided into two parts: experimental method, including a description of the experimental device, and results.

2. Experimental method

The Mini-INCA set-up is located at the High Flux Reactor (HFR) of the Laue-Langevin Institut (ILL) in Grenoble [10]. It consists of two complementary irradiation facilities: H9 beam channel where activation analysis of short-time irradiated samples are performed, and V4 channel where long-time irradiations are done including on-line fission cross-section measurements. In the present paper, only the H9 experimental set-up is described. It provides, as it will be detailed later, essentially thermal neutrons with an intensity of the order of 6×10^{14} n/cm^2/s. Thanks to such an intense neutron flux, small mass samples ($<100\,\mu$g) can be used with the advantages of reducing local neutron-flux perturbations and α-particles self-absorption in the target. Moreover, the high level of thermalisation available in the H9 channel results in minimizing the contribution of epithermal neutrons in the reaction rate balance for most of the isotopes as shown in Ref. [11].

2.1. Activation method

The activation method is employed to measure capture cross-sections in H9. Samples are irradiated into the H9 beam channel and then automatically transferred into the "Mini-INCA" chamber where α- and γ-spectroscopy analyses are performed. The evolution of a sample in a neutron flux is given by a linear system of differential equations [8]

$$\frac{\mathrm{d}N}{\mathrm{d}t} = -MN \tag{1}$$

where M is a square matrix with diagonal elements representing the total decay rate of the isotope i and the non-diagonal elements are the transition rates from isotope j to isotope i. N is a vector giving the isotope population N_i for a given isotope i. The reaction rates M_{ij} could be written in terms of reaction cross-sections (σ) and decay constant (λ)

$$M_{ij} = \begin{cases} \lambda_i + \sigma_i\phi & \forall j = i \\ -\sigma_j\phi \text{ or } \lambda_j & \forall j \neq i \end{cases} \tag{2}$$

where σ_i is the combustion (capture and fission) cross-section, σ_j is the capture cross-section and ϕ is the integrated flux. In general, these equations are solved numerically but for low neutron fluency they could be solved analytically. In particular, for isotopic samples and short-time irradiations, as in our experiments, where double capture could be neglected, the amount N_{X+1} of isotope $X+1$ produced from the isotope X at the end of irradiation could be expressed as[1]

$$N_{X+1} = \frac{N_X\sigma_c(X)\phi}{\lambda_{X+1} - \sigma_c(X)\phi}[1 - \exp(-(\lambda_{X+1} - \sigma_c(X)\phi)t_{irr})] \tag{3}$$

where N_X is the quantity of initial isotope X measured at the end of irradiation, $\sigma_c(X)$ is the capture cross-section, t_{irr} is the irradiation time. If the condition $\lambda_{X+1} \ll \sigma_c(X)\phi$ is fulfilled, the capture cross-section can be extracted

$$\sigma_c(X) \simeq \frac{1}{\phi t_{irr}} \ln\left(1 + \frac{N_{X+1}}{N_X}\right). \tag{4}$$

Quantities N_{X+1} and N_X are deduced from the measured γ- or α-line intensities I_{X+1} and I_X, respectively, and its writes

$$\begin{cases} N_{X+1} = \dfrac{I_{X+1}}{\lambda_{X+1}\Omega\varepsilon}\exp(\lambda_{X+1}t) \\ N_X = \dfrac{I_X}{\lambda_X\Omega\varepsilon}\exp(\lambda_X t) \end{cases} \tag{5}$$

where t is the time between the end of irradiation and the measurement, and $\Omega\varepsilon$ is the total detection efficiency. The quantity ϕt_{irr} is the so-called neutron fluency seen by the target during the irradiation and is measured directly from the analysis of a neutron flux monitor irradiated together with the sample. In general, the ^{59}Co(n, γ)^{60}Co reaction cross-section is used for the normalisation, so that the measured cross-section (Eq. (4)) could be expressed as a function of the

[1] Assuming that fission cross-section of X and burn-up of $X+1$ could be neglected.