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Thermal neutron cross section and resonance integral measurements for the $^{133}Cs(n,\gamma)^{134m}Cs$ reaction

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ABSTRACT

The thermal neutron cross section (σ_0) and the resonance integral cross section (I_0) for the formation of the isomeric state ^{134m}Cs by the $^{133}Cs(n,\gamma)^{134m}Cs$ reaction were experimentally determined by using an activation method with a single monitor ^{55}Mn. In this study, each of analytical grade MnO_2 and Cs_2CO_3 powders was diluted separately with Al_2O_3 powder to reduce neutron self-shielding effects, and then they were irradiated with and without a cadmium shield case in an isotropic neutron field of the ^{241}Am–Be neutron source. The activities produced in the samples were measured by a high resolution γ-ray spectrometry with a calibrated HPGe detector. The necessary correction factors for γ-ray self-attenuation, true coincidence summing effects, thermal and resonance neutron self-shielding effects, and epithermal spectrum-shape factor were taken into account. The σ_0 and I_0 values were determined to be $\sigma_0 = 2.77 \pm 0.14$ b and $I_0 = 22.8 \pm 1.7$ b for the $^{133}Cs(n,\gamma)^{134m}Cs$ reaction. These results are discussed and compared to the previous measurements and the evaluated data. The present σ_0 value is close to some of the previously reported experimental and evaluated data within the uncertainty limits. However, the present I_0 value was found to be about 28–51% smaller than other experimentally reported ones appeared in the literature. The measured resonance integral cross section value agrees well only with the newest experimental data obtained by Nakamura et al. (1999) for the $^{133}Cs(n,\gamma)^{134m}Cs$ reaction.

1. Introduction

Cesium-134m (^{134m}Cs) is the isomeric state of the ^{134}Cs radioisotope and decays to the ground state of ^{134}Cs completely by isomeric transition (IT) with a half-life of 2.912 h. It is produced both directly as a fission product and via neutron capture by the $^{133}Cs(n,\gamma)^{134m}Cs$ reaction from ^{133}Cs which is the only stable isotope of Cs-element. The thermal neutron cross section and resonance integral for the formation of the isomeric state, ^{134m}Cs via the $^{133}Cs(n,\gamma)^{134m}Cs$ reaction are of great importance for studies concerning interactions of neutrons with matter and other fundamental nuclear research, such as the nuclear data requirements for decay-heat calculations.

In surveying the literature, it appeared that a number of measurements was carried out for the thermal neutron cross section and the resonance integral for the $^{133}Cs(n,\gamma)^{134m}Cs$ reaction. Those values for the thermal neutron cross section are distributed from 2.44 to 2.82 b, except the older value of 0.016 b reported by Seren et al. (1947). The newest experimental value for the thermal neutron cross section is 2.70 b measured by Nakamura et al. (1999). However, there are serious discrepancies amongst the resonance integral cross sections, I_0 ranged from 23.2 to 34.4 b for the $^{133}Cs(n,\gamma)^{134m}Cs$ reaction, which differ from each other up to 48%. Nevertheless, it should be noted that the latest value of $I_0 = 23.2$ b measured by Nakamura et al. (1999) is quite smaller than other experimental values of 30.2 b (Heft, 1978), 29.2 b (Van Der Linden et al., 1974), 30 b (Ryves, 1970; Hayodom et al., 1969), and 34.4 b (Rajput and Mac Mahon, 1995; Baerg and Bartholmew, 1960). It seems that there is not sufficient consistency among σ_0 or I_0 values. Therefore, the aim of the present work is to carry out a new experiment with the state-of-the-art modern neutron metrology in order to clarify the existing differences in the σ_0 and I_0 values for the $^{133}Cs(n,\gamma)^{134m}Cs$ reaction by employing Cd-ratio method with use of the $^{55}Mn(n,\gamma)^{56}Mn$ reaction as a single reference monitor.

For the thermal neutron cross section and the resonance integral measurements of isotopes, the activation method is generally used by comparing the cadmium ratios of isotopes of the elements of interest relative to the cadmium ratio of a reference monitor, for which usually gold is employed. However, ^{55}Mn has been considered to be a convenient alternative monitor with good resonance parameters (Mughabghab et al., 1981; Shibata et al., 2011) for the resonance integral determination in which most of the resonance captures occur at relatively higher neutron energy region at about 337 eV, which is quite far from $1/v$ region. Other advantages of ^{55}Mn as a monitor isotope were explicated

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previously (i.e., Karadag et al., 2003, 2007; Karadag and Yücel, 2004, 2005, 2008; Yücel and Karadag, 2005; Yücel et al., 2007).

2. Experimental

2.1. Samples

The analytical grade Cs_2CO_3 and MnO_2 powders were obtained from Aldrich Inc. In order to minimize the influences due to both thermal and epithermal neutron self-shielding effects, they were mixed independently from each other with sufficient amounts of Al_2O_3 powder. Because ^{27}Al isotope has the lower neutron absorption cross-section. The samples of ($96.3\%Al_2O_3 + 3.7\%Cs_2CO_3$) and ($96.6\%Al_2O_3 + 3.4\%MnO_2$) were prepared by choosing suitable dilution percentages to obtain minimum neutron self-absorptions in the samples. Then, the diluted samples were filled in small polystyrene tubes with the diameter of 6.5 mm, height of 13.1 mm and a wall thickness of 1 mm. Ten (10) diluted Cs-samples having each mass of about 0.8 g were individually prepared. Five (5) of them were used for Cd-covered irradiation and the other five (5) samples were used for obtaining bare irradiation data. The same sample preparation produces were employed for ten (10) Mn- samples.

2.2. Irradiation and activity measurements

The sample irradiations were carried out with and without a cadmium (Cd) box, which is a cylinder box with a 1 mm wall thickness. The irradiator used in this work has an isotropic neutron field provided by three ^{241}Am–Be isotopic neutron sources each of which having 592 GBq activity, immersed in paraffin moderator shielded with lead bricks. Since the irradiation hole has a very large volume compared to the sample volume, the effect of thermal flux depression at the irradiation position could be neglected. The irradiation position was the same for all samples. Thermal and epithermal neutron fluxes were measured to be $(1.5 \pm 0.2) \times 10^4$ and $(1.4 \pm 0.1) \times 10^3$ neutrons/cm^{2}s respectively, and the epithermal neutron spectrum shape factor (α) was experimentally determined as 0.083 ± 0.016 at the sample irradiation position (Yücel and Karadag, 2004). This irradiator was previously described in detail (see Karadag et al., 2003; Yücel and Karadag, 2004; Yücel et al., 2007). For both the $^{133}Cs(n,\gamma)^{134m}Cs$ and $^{55}Mn(n,\gamma)^{56}Mn$ reactions the irradiation times were chosen for a period of four to five half-lives in order to yield enough activity to be measured in the γ-ray counting system. After the irradiations, the suitable waiting times were employed to minimize dead time losses and to eliminate the possible contributions of 843.76 keV γ-ray by minor activity of ^{27}Mg ($T_{1/2}$ = 9.46 min), which may be produced by the $^{27}Al(n,p)^{27}Mg$ reaction.

The samples were measured by a γ-ray counting system equipped with a coaxial n-type high purity Ge detector (Canberra GR2020) with an active volume of about 100 cm^3, which has a measured relative efficiency of 22.6% at 1332.5 keV of ^{60}Co, an energy resolution of 1.80 keV and a peak-to-Compton ratio of 53.5:1 at 1332.5 keV of ^{60}Co. The detector was shielded by a 10 cm thick lead lined with 1 mm copper sheets on all sides. The multi-channel analyzer (MCA) system has a full 8 K channels MCA memory/ADC conversion gain for pulse height analysis, covered up to about 2000 keV in a 4096 MCA channels by setting suitable amplifier gain. The samples were counted on the end-cap of the detector to get more counts in the spectrum due to close counting geometry. For this counting geometry, i.e. a small cylinder placed on the end-cap of the detector, the full-energy detection efficiency as a function of energy for the present HpGe detector was determined by using a multinuclide standard source (EG-ML S/N:1160-39-3) in sand matrix (SiO_2: $\rho = 1.7 \pm 0.1$ g cm^{-3}) spiked with ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{60}Co and ^{88}Y radionuclides, obtained from Eckert&Ziegler/Isotope Products Laboratories Inc., traceable to PTB (Germany). In the efficiency calibration, a small amount of about 0.7 g sand matrix spiked with above mentioned radionuclides was filled in the cylinder by taking an aliquot from the source. This gamma-ray extended source was prepared in the same size cylinder of 6.5 mm diameter × 13.1 mm height which is identical to the sample cylinders. Also, GESPECOR® (ver. 4.2), based on MNCP simulation, was used for obtaining the calculated efficiency values for a small cylinder placed on the end-cap of the detector. The experimental efficiency data agreed very well with the calculated ones.

The counting times predetermined for each measurement varied between 2 and 8 h, depending on mainly the activity level of ^{134m}Cs or ^{56}Mn products in the samples. A typical experimental γ-ray spectrum for the irradiated Cs sample is shown in Fig. 1. The counting periods were high enough to ensure good statistical

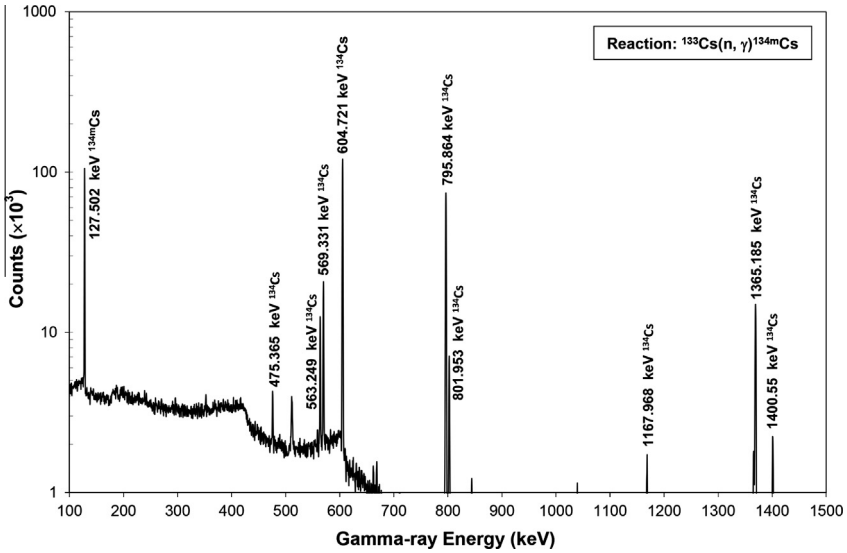

Fig. 1. A typical γ-ray energy spectrum of the irradiated Cs sample.