Annals of Nuclear Energy 69 (2014) 74–89

Contents lists available at ScienceDirect

Annals of Nuclear Energy

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

Europium resonance parameters from neutron capture and transmission measurements in the energy range 0.01–200 eV

G. Leinweber [a,*], D.P. Barry [a], J.A. Burke [a], M.J. Rapp [a], R.C. Block [a], Y. Danon [b], J.A. Geuther [c], F.J. Saglime III [d]

[a] Bechtel Marine Propulsion Corp., Knolls Atomic Power Laboratory P.O. Box 1072, Schenectady, NY 12301-1072, United States
[b] Rensselaer Polytechnic Institute, Gaerttner LINAC Center, 110 8th St., Troy, NY 12180, United States
[c] Kansas State University, 3002 Rathbone Hall, Manhattan, KS 66506, United States
[d] Moog Inc., East Aurora, NY 14052, United States

ARTICLE INFO

Article history:
Received 6 December 2013
Accepted 31 January 2014
Available online 25 February 2014

Keywords:
Europium
Transmission
Capture
Thermal cross section
RPI
Resonance parameters

ABSTRACT

Europium is a good absorber of neutrons suitable for use as a nuclear reactor control material. It is also a fission product in the low-yield tail at the high end of the fission fragment mass distribution. Measurements have been made of the stable isotopes with natural and enriched samples.

The linear electron accelerator center (LINAC) at the Rensselaer Polytechnic Institute (RPI) was used to explore neutron interactions with europium in the energy region from 0.01 to 200 eV. Neutron capture and transmission measurements were performed by the time-of-flight technique. Two transmission measurements were performed at flight paths of 15 and 25 m with ^{6}Li glass scintillation detectors. The neutron capture measurements were performed at a flight path of 25 m with a 16-segment sodium iodide multiplicity detector.

Resonance parameters were extracted from the data using the multilevel R-matrix Bayesian code SAMMY. A table of resonance parameters and their uncertainties is presented.

To prevent air oxidation metal samples were sealed in airtight aluminum cans in an inert environment. Metal samples of natural europium, 47.8 atom% ^{151}Eu, 52.2 atom% ^{153}Eu, as well as metal samples enriched to 98.77 atom% ^{153}Eu were measured.

The measured neutron capture resonance integral for ^{153}Eu is (9.9 ± 0.4)% larger than ENDF/B-VII.1. The capture resonance integral for ^{151}Eu is (7 ± 1)% larger than ENDF/B-VII.1.

Another significant finding from these measurements was a significant increase in thermal total cross section for ^{151}Eu, up (9 ± 3)% from ENDF/B-VII.1. The thermal total cross section for ^{153}Eu is down (8 ± 3)% from ENDF/B-VII.1, but it is larger than that of ENDF/B-VII.0.

The resolved resonance region has been extended from 100 eV to 200 eV for both naturally-occurring isotopes. Uncertainties in resonance parameters have been propagated from a number of experimental quantities using a Bayesian analysis. Uncertainties have also been estimated from fitting each Eu sample measurement individually.

1. Introduction

Europium is important in the design of light-water nuclear reactors for two reasons. First, europium is a fission product in the low-yield tail at the high end of the fission fragment mass distribution. Second, it is a strong neutron absorber that could be employed as a solid oxide control material. Europium is a slow-burning poison due to its 5-member chain of absorbing isotopes, mass numbers 151 through 155. Each of these isotopes has either high fission yield or high thermal neutron cross section.

The purpose of the present work was to determine resonance parameters for europium. The resonance parameters in ENDF/B-VII.1 (Chadwick et al., 2011) were adopted primarily from the measurement of Rahn et al., 1972). The Rahn et al. measurement utilized highly enriched oxide samples, consisted of transmission and self-indication experiments, and employed the synchrocyclotron at Columbia University. The current measurement has better energy resolution and updated analysis methods.

Other prominent experiments include Moxon et al. (1976) who measured capture cross sections averaged over 100 eV-wide

* Corresponding author. Tel.: +1 (518) 276 4006; fax: +1 (518) 2764007.
E-mail address: leinwg@rpi.edu (G. Leinweber).

http://dx.doi.org/10.1016/j.anucene.2014.01.041
0306-4549/

regions in 1976, Widder (1974) performed neutron capture measurements with a Moxon-Rae detector in 1974, Konks et al. (1968) used a lead slowing-down-time spectrometer to determine average capture cross sections in 1968, and Anufrijev et al. (1979) used a reactor in 1979. Recent experiments include Lee et al. (2010) in 2010 using a C_6D_6 capture detector at a 12 m flight path. Their results agreed with the JENDL-4.0 evaluation in the resolved resonance region. They used this agreement as validation of their weighting function. Parker et al. (2007) used the DANCE barium fluoride capture detector at the Los Alamos National Laboratory in 2007.

The release of the ENDF/B-VII.1 library in 2011 included an increase in the thermal total cross section of ^{153}Eu of 14% over the ENDF/B-VII.0 (Chadwick et al., 2006) value. The current measurement supports an increase by a smaller amount as discussed in Section 4.9.

2. Experimental conditions

2.1. Overview

Table 1 gives some details of the experimental conditions including neutron targets, overlap filters, LINAC pulse repetition rates, flight path lengths, and time-of-flight channel widths. The neutron energy for a detected event was determined using the time-of-flight (TOF) technique.

The nominal resolution, pulse width divided by flight path length, was ≈1 ns/m for epithermal transmission and capture measurements.

Thermal and epithermal capture and epithermal transmission were measured at a 25 m flight path. Thermal transmission was measured at 15 m. Thermal and epithermal transmission were measured with ^{6}Li glass detectors (Barry, 2003; Leinweber et al., 2002; Leinweber et al., 2010; Trbovich, 2003). Thermal and epithermal capture were measured with a 16-segment NaI detector (Barry, 2003; Leinweber et al., 2002; Leinweber et al., 2010; Trbovich, 2003), (Block et al., 1988).

The LINAC was used to accelerate electrons into a tantalum target. Bremsstrahlung radiation and photoneutrons were produced. The neutron-producing targets were optimized for each energy range (Danon et al., 1993; Danon et al., 1995; Overberg et al., 1999).

Table 2 gives some sample information including the sample thickness, atom fraction of each isotope, and measurements.

The uncertainties in sample thickness were propagated from multiple measurements of sample weight and diameter. The diameter measurements were the dominant component of the uncertainties. All samples were mounted in aluminum sample cans. The thickness of aluminum on each of the front and rear faces of each sample was 0.38 mm. The influence of these sample cans, as well as all background, was measured by including empty sample cans in all measurements. Background in transmission measurements is discussed in Section 3.2.1.

2.2. Sample Information

There are only two naturally-occurring isotopes of europium (see Table 2). The samples measured were elemental as well as enriched to 98.77 atom% ^{153}Eu. Europium is a highly-reactive metal, and care was taken to prevent oxidation. Metallic samples for both natural and enriched Eu were fabricated, weighed, and encapsulated in an inert atmosphere. X-ray imaging of the encapsulated thin metal disks was performed, and the images were analyzed to identify any non-uniformity of thickness (Geuther et al., 2013).

X-ray images of the samples used in these experiments are shown in Fig. 1. A samarium step-wedge was used to calibrate X-ray image data and quantify the non-uniformity of sample thickness. Samarium was chosen as the step-wedge material because it has nearly the same mass attenuation coefficient as europium. Additionally, samarium is much less reactive in air and could be imaged next to the encapsulated (in 0.38 mm Al) Eu samples. Fig. 1 is reprinted from Ref. (Geuther et al., 2013). The sample thicknesses are given in Table 2. They were determined from measurements of mass and area made at the time of encapsulation, not from the subsequent X-ray images. The X-ray imaging results for average sample thickness provided confirmation of both the thicknesses and the methods. The relative density profiles from the imaging measurements were included in the SAMMY analysis. There were no visible signs of oxidation at the time of encapsulation, which was done in an inert environment. The ^{153}Eu-enriched samples were provided by the Oak Ridge National Laboratory. The natural samples were obtained from the KAMIS Corporation. All samples were certified >99.9 weight percent europium. The results of mass spectrographs performed on the europium samples by their vendors are given in Table 3.

2.3. Capture detector

The capture detector is a gamma detector containing 20 l of NaI(Tl) divided into 16 optically-isolated segments (Block et al., 1988). The scintillation crystals form an annulus around the neutron beam with the sample at its center. The neutron beam was

Table 1
Europium experimental details.

Experiment	Overlap filter	Neutron-producing target	Elec-tron pulse width (ns)	Ave. beam current (μA)	Beam energy (MeV)	Energy region (eV)	Channel width, (μs)	Pulse repetition rate (pulses/s)	Flight path length (m)
Epithermal transmission	Boron carbide	Bare bounce	25	11	58	$E < 15$ $15 < E < 800$ $E > 800$	0.4096 0.1024 0.0256	225	25.590 ± 0.006
Thermal transmission	None	Enhanced thermal target	540	8	55	$E < 0.05$ $0.05 < E < 1.4$ $1.4 < E < 5.6$ $E > 5.6$	26.214 3.2768 0.8192 0.4096	25	14.96 ± 0.02
Epithermal capture	Cadmium	Bare bounce	19	13	56	$E < 15$ $15 < E < 800$ $E > 800$	0.4096 0.1024 0.0256	305	25.564 ± 0.006
Thermal capture	None	Enhanced thermal target	560	8	56	$E < 0.05$ $0.05 < E < 1.4$ $1.4 < E < 5.6$ $E > 5.6$	26.214 3.2768 0.8192 0.4096	25	25.446 ± 0.002