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Rhenium resonance parameters from neutron capture and transmission measurements in the energy range 0.01 eV to 1 keV

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ABSTRACT

Rhenium is a refractory metal with potential uses in nuclear reactor applications, particularly those at very high temperatures. Measurements have been made using natural samples. Natural rhenium consists of two isotopes: ^{185}Re (37.40%) and ^{187}Re (62.60%). The electron linear accelerator (LINAC) at the Rensselaer Polytechnic Institute (RPI) Gaerttner LINAC Center was used to explore neutron interactions with rhenium in the energy region from 0.01 eV to 1 keV. Neutron capture and transmission measurements were performed by the time-of-flight technique. Two transmission measurements were performed at flight paths of 15 m 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. The uncertainties in resonance parameters were propagated from a number of experimental quantities using a Bayesian analysis. Uncertainties were also estimated from fitting each Re sample measurement individually. The measured neutron capture resonance integral for ^{185}Re is (4 ± 1)% larger than ENDF/B-VII.1. The capture resonance integral for ^{187}Re is (3 ± 1)% larger than ENDF/B-VII.1. Other findings from these measurements include: a decrease in the thermal capture cross section for ^{185}Re of (2 ± 2)% from ENDF/B-VII.1; a decrease in the thermal capture cross section for ^{187}Re of (3 ± 4)% from ENDF/B-VII.1; a decrease in the thermal total cross section for ^{185}Re of (2 ± 2)% from ENDF/B-VII.1; and a decrease in the thermal total cross section for ^{187}Re of (6 ± 5)% from ENDF/B-VII.1. Considering the uncertainties, none of the indicated changes in thermal cross sections represents a statistically significant change from ENDF/B-VII.1.

1. Introduction

Rhenium is an extremely rare material that does not occur as a free element in nature. It is extremely dense, has a high melting point, and the highest boiling point of any element (Weast et al., 1987). Natural rhenium consists of one stable isotope, ^{185}Re (37.40% abundance) and one long-lived isotope, ^{187}Re (62.60% abundance; half-life of 4.12 × 10^{10} years)(Baum et al., 2010). As a refractory metal (i.e., extremely resistant to heat and wear), rhenium has potential uses in reactor applications(Friesenhahn et al. 1967). Alloys containing rhenium have potential use in space reactors and fusion reactors (Busby et al., 2007; Craft et al., 2014; Guan et al., 2016).

The purpose of the present work was to determine resonance parameters for rhenium. The resonance parameters in ENDF/B-VII.1 (Chadwick et al., 2011) were adopted from Mughabghab (Mughabghab, 2006). The values published by Mughabghab are based on several historical experiments.

The earliest measurements of rhenium resonance parameters were performed by Melkonian et al. (1953) who performed neutron transmission measurements in natural rhenium samples up to 21.2 eV at Columbia University in 1953. Two years later, Igo (1955)

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conducted neutron transmission measurements between 1 eV and 13 eV with natural rhenium samples using the crystal spectrometer at Brookhaven National Laboratory. In 1965, Vertebnyi et al. (1965) performed neutron transmission measurements up to 110 eV with isotopically enriched samples of ^{185}Re and ^{187}Re along with natural rhenium using the VVR-M reactor with a chopper and published resonance parameters up to 21.46 eV. Friesenhahn et al. (1967) performed neutron transmission, capture yield, and self-indication measurements in 1967 with natural rhenium samples using the Gulf General Atomic linear accelerator for energies up to 30 keV, with resonance parameters evaluated up to 100 eV. Ideno et al. (1972) performed neutron transmission measurements in 1971 with natural rhenium samples using the linear accelerator at the Japan Atomic Energy Research Institute up to 300 eV. In 1976, Namenson et al. (1976) measured neutron resonance parameters up to 2 keV in isotopically enriched rhenium samples using the Oak Ridge National Laboratory linear accelerator (ORELA). More recently (2012), Arboccò et al. (2013) measured the thermal total cross sections of ^{185}Re and ^{187}Re as part of neutron activation analyses performed at the Belgian Reactor 1 at Studiecentrum voor Kernenergie – Centre d'étude de l'énergie nucléaire (BR1, SCK•CEN).

2. Experimental conditions

2.1. Overview

The RPI LINAC was used to accelerate electrons into one of the tantalum neutron-producing targets. 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). 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, 2010; Trbovich, 2003). Thermal and epithermal capture were measured with a 16-segment NaI detector (Barry, 2003; Leinweber et al., 2002, 2010; Trbovich, 2003; Block et al., 1988).

Table 1 gives some details of the experimental conditions including neutron targets, overlap filters (used to prevent overlap of neutron pulses by removing the lowest energy neutrons from each pulse), 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 ≈1.8 ns/m for epithermal transmission and capture measurements.

Table 2 gives some sample information including the sample thicknesses and measurements for which each sample was used. The uncertainties in sample thickness were propagated from multiple measurements of sample mass and diameter. The diameter measurements were the dominant component of the uncertainties in the number densities. All samples were mounted in open aluminum sample cans. 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.

An independent array of neutron detectors was used to monitor for fluctuations in LINAC operation. These detectors, referred to as beam monitors or monitors, were used to perform statistical checks of the measured data, and one monitor was selected as the beam intensity normalization standard for each experiment.

2.2. Sample information

There are only two naturally-occurring isotopes of rhenium, ^{185}Re and ^{187}Re. The samples used in the current measurements were elemental and in metallic form. The samples were obtained from Rhenium Alloys, Inc. and were certified >99.99% pure rhenium. The results of impurity analyses performed on the rhenium samples by the vendor are given in Table 3.

2.3. Capture detector

The capture detector is a gamma ray detector containing 20 cubic decimeters 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 collimated to a diameter of 5.08 cm at the sample position. Neutrons that scatter from the sample are absorbed by a hollow cylindrical liner (0.9 cm thick) fabricated of 98.4 wt% $^{10}B_4C$ ceramic to reduce the number of scattered neutrons reaching the gamma detector. The discriminator on each detector section was set to 100 keV. A total energy deposition of 1 MeV for the epithermal measurement and 2 MeV for the thermal measurement was required to register a capture event. Therefore, the system discriminates against the 478 keV gamma rays from ^{10}B(n;α,γ)

Table 1
Rhenium 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	42 ± 2	13	56	E < 4.0 4.0 < E < 44.4 44.4 < E <258.2 E > 258.2	8.00000 0.50000 0.06250 0.03125	225	25.596 ± 0.0055
Thermal Transmission	None	Enhanced Thermal Target	700 ± 50	7	50	E < 0.36 0.36 < E < 9.00 9.00 < E < 36.81 E > 36.81	16.0000 1.0000 0.2500 0.0625	25	14.973 ± 0.0055
Epithermal Capture	Cadmium	Bare Bounce	47.1 ± 0.3	16	53	E < 4.0 4.0 < E < 44.2 44.2 < E < 255.5 E > 255.5	8.00000 0.50000 0.06250 0.03125	225	25.564 ± 0.0055
Thermal Capture	None	Enhanced Thermal Target	860 ± 130	6.6	48	E < 0.60 0.60 < E < 8.85 8.85 < E < 35.61 E > 35.61	16.0000 1.0000 0.2500 0.1250	25	25.444 ± 0.0055