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Disentangling the ^{16}O cross section using light water and heavy water benchmark assemblies

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HIGHLIGHTS

- Measurement of fast neutron flux penetrating through heavy and light water spheres.
- Effect of different data libraries on calculations.
- Sensitivity analysis of ^{16}O elastic, inelastic and (n, alpha) cross sections.

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ABSTRACT

Fast neutron leakage spectra from the light and heavy water sphere of 30 cm in diameter with neutron source in its centre were measured by a stilbene scintillation detector in the region of 1–10 MeV in the distance of 85 cm from the spheres surface. We use the light and heavy water to eliminate the effect of hydrogen. ^{252}Cf with the approximate emission rate of 5.5E8 n/s was used as a neutron source for all measurements involved and was placed in the centres of the spheres. The measured neutron spectra are compared with MCNP transport code calculations in ENDF/B-VII.0, ENDF/B-VIII.b4 and JENDL-4 nuclear data libraries. Experimental results for both cases follows similar trend. The best agreement is achieved with ENDF/B-VIII.b4 library in both cases. All libraries underestimate experimental measurement in the region of 3–4 MeV. Furthermore, JENDL-4 library overestimates experiment in the region of 4–6.5 MeV. In addition, we performed cross section sensitivity analysis for elastic, inelastic and (n,α) reaction in JENDL-4 and ENDF/B-VIII.b4 libraries since they have almost independent evaluations of ^{16}O.

1. Introduction

Oxygen belongs to the highest priority isotope in terms of nuclear data. Many nuclear data applications require a good knowledge and understanding of cross-sections for oxygen. Since the oxygen is present in light, heavy water and nuclear fuel in the form of oxide, then accurate knowledge of cross-section data and related uncertainties is crucial for the reactor analysis and design and nuclear criticality safety as well as for the processing and disposal of nuclear waste. The elastic cross section for oxygen is important for the fast neutron transport in the water moderating system and the $^{16}O(n,\alpha)^{13}C$ reaction absorbs fast neutrons. Cross-section (XS) for $^{16}O(n,\alpha)^{13}C$ reaction is listed in the High Priority Request List (HPRL, 2017). This reaction affects the reactivity of light water reactors and is responsible for the production of helium in fuel pins and claddings and is therefore important for these parts behavior. Furthermore, the precise calibration of neutron source emission using the manganese sulfate bath technique is also influenced.

It should be noted that, the JENDL-4.0 (Shibata et al., 2011) and ENDF/B nuclear data evaluations for ^{16}O are the only ones which are largely independent. However, the JENDL-4.0 has adopted the ENDF/B-VII.0 (Chadwick et al., 2006) (n,α) cross section below 6.5 MeV (Chadwick et al., 2014). For that reason, we focus on the JENDL-4, ENDF/B-VII.0 and ENDF/B-VIII.0 (Brown, 2017) ^{16}O neutron nuclear data evaluations.

Validation of the energy-dependent differential evaluated data in neutron benchmark fields is an essential step of the evaluation procedure. Such testing may find discrepancies in the evaluated data, especially in energy ranges, where XS is the most sensitive to the results of integral measurements. The neutron benchmark experiments using the ^{252}Cf neutron sources are highly suitable since the ^{252}Cf spontaneous fission spectrum is known with relatively high accuracy and small total uncertainties of integral measurements can be achieved.

In the light of mentioned context we have performed experiments involving measuring fast neutron leakage spectrum in the range of

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1–10 MeV using light and heavy water spherical benchmark assemblies employing ^{252}Cf neutron source in their centres to eliminate the effect of hydrogen. Calculations of neutron leakage spectra with different ^{16}O evaluations are compared with the experimental data. Moreover, the influence of the elastic, inelastic and (n,α) XS of ^{16}O via the sensitivity analysis is assessed since they are the most influential (with the exception of angular distributions which are difficult to separate in the spherical geometry) XS variation.

The emission of the ^{252}Cf neutron source involved in the experiments was approximately 5.5E10^{8} n/s during the experiments. The total emission of the source was measured by manganese sulfate bath method according to the Certificate of Calibration.

2. Description of the experimental and calculation methods

The ^{252}Cf neutron source was placed in the centre of a heavy or light water sphere using a pneumatic flexo-rabbit system. Heavy or light water sphere has diameter d = 29.997 cm and is covered with a steel coating of thickness t = 0.078 cm and in the case of heavy water sphere contains 99.36 wt% of heavy water, rest is a light water (Jánský et al., 1992). Fast neutron leakage spectra were measured 100 cm from the centre of the water sphere using stilbene scintillator detector. The geometry of the experimental arrangement can be found in Fig. 1. The evaluated neutron spectra were determined as an average of the several measurements.

The room effect was estimated by the employing the set of the shielding cones. The resulting neutron spectra arise as a difference between the measurement without and with shielding cones. Shielding cones consists of layers made of borated polyethylene and lead (Jánský et al., 2000).

Leakage neutron spectra were measured by the proton recoil method using a fully digitized two-parameter spectrometric system in the interval from 1.0 to 10.0 MeV in groups with step of 100 keV, for details see Veškrna et al. (2014). The size of the cylindrical stilbene scintillator used for the measurements was 10 × 10 mm. The neutron spectra are evaluated from the measured proton recoil spectra by means of deconvolution using Maximum Likelihood Estimation, see (Cvachovec and Cvachovec, 2008). The 10 × 10 stilbene crystal used in measurements was tested in quasi monoergetic spectra behind Si filter (see Košťál et al., 2017). The significant sources of uncertainty in the measurement were calibration uncertainty 2–5% (it concerns energy calibration), uncertainty in the crystal calibration factor 2.0% (it concerns absolute calibration), and uncertainty in the total emission of the neutron ^{252}Cf source 0.7%. Overall measurement uncertainty, involving also statistical uncertainty and dispersion between measurements, is between approximately 3% and 15% in the measured region for light water sphere and 3–10% for heavy water sphere. Generally, light water sphere measurement has higher uncertainty due to the higher gamma background.

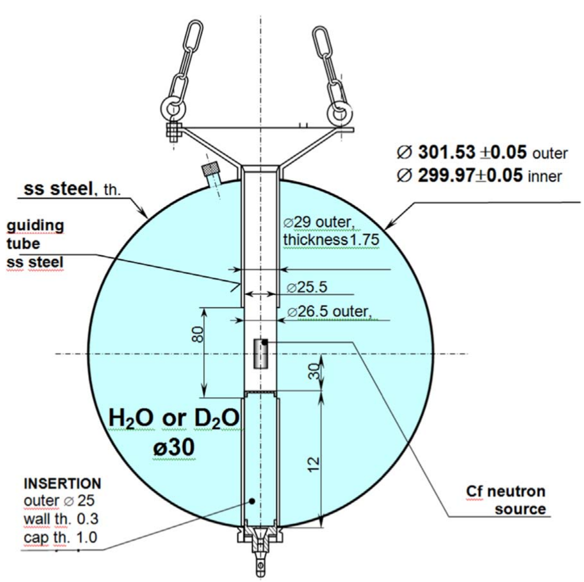

Fig. 1. Light or heavy water sphere design with neutron source. All dimensions shown are in mm. The spheres were hanged up to a distance of 2 m from the floor to minimize the room effect.

All calculations were performed with the MCNP6 transport code (Goorley et al., 2012) using ENDF/B-VII.0 (Chadwick, 2006) and ENDF/B-VIII.b4 (Brown, 2017) nuclear data transport libraries in the ACE format. JENDL-4 files were processed by the NJOY-99.049_296 code (MacFarlane and Kahler, 2010). All calculations were performed at the room temperature. The input ^{252}Cf spontaneous neutron fission spectrum was taken from the IRDFF webpage (IRDFF, 2016). The influence of the nuclear data libraries onto nuclear transport problem was investigated by changing of only ^{16}O XS ACE file. The other ACE files employed were taken from ENDF/B-VII.0 library. The XS sensitivity analysis was performed using the PERT card. Elastic, inelastic and (n,α) XS of ^{16}O were varied. Again, only ^{16}O XS was varied in the library under study, the rest ACE files were taken from ENDF/B-VII.0 library. It should be noted that the results depend on the choice of the other ACE files as well.

Overall MCNP tally statistical calculations uncertainties were always under 0.5%. The other sources of uncertainties were uncertainty of the detector position 0.5%, uncertainty in the sphere dimensions 0.2%, uncertainty in the position of the neutron ^{252}Cf source 1.0% and uncertainty in the total emission of neutron ^{252}Cf source 1.3% giving total calculation uncertainty of 1.9%. All other sources of uncertainty were found negligible.

3. Results

Fig. 2 compares neutron flux density for the light and heavy water sphere in 100 cm from the centres of the spheres. It can be seen that light water neutron leakage spectrum is harder than in the case of heavy water.

Fig. 3 shows the C/E ratios for the light water sphere. It compares various libraries calculations, where Gaussian energy broadening function was implemented. Good agreement is achieved with all libraries under study in the region of 1–3 MeV. However, all libraries underestimate experiment up to 15% in the region of 3–4 MeV. For higher energies, ENDF/B libraries achieve good agreement in the region of 4–6.5 MeV unlike JENDL-4 library which overestimates experimental results. Good agreement is achieved for energies higher than 6.5 MeV with all libraries. Fig. 4 shows the C/E ratios for the heavy water sphere. Again, Gaussian energy broadening function was implemented for all calculated spectra. The trend for all libraries is very similar as in the light water case. Overall, the agreement is best for the ENDF/B-VIII.b4 library. Again, all libraries underestimate experiment up to 14% in the region of 3–4 MeV. JENDL-4 library significantly overpredicts measured data in the region of 4–7 MeV.

The elastic angular distribution for emitted neutrons determines the energy lost during collisions and thus affects the neutrons slowing down and their removal to below the given detection energy limit of 1 MeV. The observed neutron fluence difference between JENDL-4 and ENDF/B-VIII.b4 libraries can be caused by this. The angular distributions differ significantly as it is shown in Fig. 5.

Fig. 6 explores the possible correlations between the light and heavy water calculations by means of the difference between C/E ratios for heavy and light water. Very interesting is the fact that JENDL-4 and ENDF/B-VIII.b4 libraries follow the same trend and the difference is almost independent on the used library.

Next, we performed sensitivity analysis to 2% rise of elastic or inelastic or (n,α) XS (varied) and compared it with the unperturbed calculation (standard) by means of C/E ratios for JENDL-4 and ENDF/B-