Measurement and simulation of the leakage neutron spectra from Fe spheres bombarded with 14 MeV neutrons

Authors: Zhao, Qi, Nie, Yangbo, Ding, Yanyan, Zhang, Shiyu, Ren, Jie, Xu, Kuozhi, Pan, Xinyi, Ruan, Xichao, Nie, Yangbo

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Abstract

Iron is commonly used as a structural and shielding material in nuclear devices. The accuracy of its nuclear data is critical for the design of nuclear devices. The evaluation data of 56Fe isotopes in the latest version of the CENDL-3.2 library from China was significantly updated. This new data must be tested before it can be used. To test the reliability of this data and assess the shielding effect, a shielding benchmark experiment was conducted with natural Fe spherical samples using a pulsed deuterium- tritium neutron source at the China Institute of Atomic Energy (CIAE). The leakage neutron spectra from the natural spherical iron samples with different thicknesses (4.5, 7.5, and 12 cm) were measured between 0.8-16 MeV after interacting with 14 MeV neutrons using the time-of-flight method. The simulation results were obtained by Monte Carlo simulations by employing the Fe data from the CENDL-3.2, ENDF/B-VIII.0, and JEDNL-5.0 libraries. The measured and simulated leakage neutron spectra and penetration rates were compared, demonstrating that the CENDL-3.2 library performs sufficiently overall. The simulation results of the other two libraries were underestimated for scattering at the continuum energy level.

Full Text

Preamble

Measurement and simulation of the leakage neutron spectra from Fe spheres bombarded with 14 MeV neutrons

Qi Zhao[1], Yangbo Nie*[1],[2], Yanyan Ding[1], Shiyu Zhang[2], Jie Ren[1], Kuozhi Xu[1], Xinyi Pan[1], Xichao Ruan[1],[2]

[1]Key Laboratory of Nuclear Data, China Institute of Atomic Energy, Beijing 102413, China
[2]School of Nuclear Science and Technology, Lanzhou University, Lanzhou 730000, China

*Corresponding author. E-mail address: nieyb@163.com

Abstract

Iron is commonly used as a structural and shielding material in nuclear devices, and the accuracy of its nuclear data is critical for reactor design. The evaluation data for ^{56}Fe isotopes in the latest CENDL-3.2 library from China were significantly updated and must be tested before application. To assess the reliability of this data and evaluate shielding effectiveness, a shielding benchmark experiment was conducted using natural Fe spherical samples with a pulsed deuterium-tritium neutron source at the China Institute of Atomic Energy (CIAE). Leakage neutron spectra from natural iron spheres of different thicknesses (4.5, 7.5, and 12 cm) were measured between 0.8–16 MeV after interaction with 14 MeV neutrons using the time-of-flight method. Monte Carlo simulations were performed using Fe data from the CENDL-3.2, ENDF/B-VIII.0, and JENDL-5.0 libraries. Comparison of measured and simulated leakage neutron spectra and penetration rates demonstrated that the CENDL-3.2 library performs sufficiently well overall, while the other two libraries underestimated scattering at the continuum energy level.

Keywords: Iron sphere, CENDL-3.2, shielding benchmark experiment, pulsed 14 MeV neutron source, time-of-flight method

1. Introduction

A complete set of nuclear data is frequently required as input for simulation calculations in reactor design, nuclear fuel cycles, nuclear medicine, and other nuclear technology applications [1-2]. Most countries with nuclear power programs have created evaluated libraries to meet application requirements, including the ENDF/B (United States), JEFF (Western Europe), BROND (Soviet Union), JENDL (Japan), FENDL (IAEA), and CENDL (China) libraries, which have been developed since the 1960s. The latest CENDL-3.2 library was released in 2020 [3], while JENDL-5.0 was released in 2021 [4].

Nuclear evaluation data represent the results from assessments of nuclear processes and can be used to evaluate the safety and efficacy of nuclear technology. These data include measurements of radioactive materials, radiation levels, and other parameters related to nuclear events, and are widely used for microscopic cross-section measurements [5-7] as well as accelerator and reactor designs [8]. Such data are crucial for nuclear safety assessment, particularly for developing and operating nuclear power plants and other facilities handling radioactive materials, as these technologies require careful monitoring to minimize safety risks and ensure effective, responsible use [9]. Benchmark experiments are critical for determining data accuracy and represent an important step in analyzing nuclear data before distribution to users [10]. Macroscopic sample measurements are an essential prerequisite for benchmark experiments, and for certain critical evaluation data, a series of such measurements must be performed to verify reliability. These experiments are vital for validating nuclear data essential for numerous nuclear applications.

A benchmark experiment involves comparing predictions made using nuclear data with experimental measurements under controlled conditions. By comparing calculation results with experimental data, scientists can evaluate the accuracy of nuclear data in predicting nuclear behavior under various conditions. This process is critical for ensuring the safety and reliability of nuclear applications, as inaccurate data can lead to errors in reactor design and operation and pose potential safety risks [11]. Therefore, benchmark experiments are essential for improving understanding of nuclear processes and developing more accurate predictive models.

Iron plays a critical role in the nuclear industry due to its unique properties [12], including its ability to absorb and decelerate neutrons. Iron is extensively used in nuclear reactor construction, particularly as steel, an iron-based alloy. In nuclear reactors, iron-based materials manufacture various components such as reactor pressure vessels, steam generators, and fuel cladding, which are crucial for safe operation and controlling nuclear reactions. Iron is also used in nuclear fuel production as a matrix material to encapsulate fuel pellets, helping contain radioactive material and prevent environmental leakage. Overall, iron provides a reliable and durable material for nuclear facility construction and component manufacturing. Improving the accuracy of Fe isotope evaluation data, particularly for ^{56}Fe, is highly significant for reducing shielding design redundancy and improving device miniaturization. The penetration problem—particle transport through matter-filled geometries—is a common challenge in radiation shielding calculations, requiring high-quality ^{56}Fe evaluation data to improve calculation accuracy for deeply penetrating neutrons.

Several Fe shielding benchmark experiments have been conducted to test evaluation data, including measurements of Fe sphere leakage neutron spectra using D-T neutron sources at Lawrence Livermore National Laboratory (LLNL) [13], angular leakage neutron spectra from slab samples at Japan Atomic Energy Research Institute (JAERI) [14], and leakage neutron and γ spectra from slab samples at Technical University of Dresden (TUD) [15]. Absolute reaction rates in spherical Fe systems were measured at the Institute of Nuclear Physics and Chemistry of CAEP [16-17], and leakage neutron spectra from slab samples were measured at CIAE [18]. These results provide important experimental data for analyzing and improving Fe evaluation data in various libraries [19].

To assess the quality of ^{56}Fe evaluation data in CENDL-3.2 and provide high-quality data for deep penetration shielding calculations, this study conducted a shielding benchmark experiment using Fe spheres with a fusion D-T neutron source. Using the time-of-flight method, leakage neutron spectra were measured for D-T neutrons passing through iron spheres of 4.5, 7.5, and 12 cm thickness, with measured energies ranging from 0.8–16 MeV. MCNP-4C simulations were performed using Fe evaluation data from CENDL-3.2, ENDF/B-VIII.0, and JENDL-5.0 libraries. Comparison and analysis of simulated and experimental results demonstrated that CENDL-3.2 simulations agree sufficiently with experimental results, and the evaluated Fe data from CENDL-3.2 can be used as optimal data for nuclear device shielding calculations.

2. Experiment Setup

The China Institute of Atomic Energy established an integral experimental system for verifying nuclear evaluation data [20-24]. Since neutron-hydrogen elastic scattering has only one reaction channel and its cross-section serves as an international standard, leakage neutron spectra from hydrogen-containing materials (standard samples) provide the primary means for checking experimental system integration. The CIAE system was tested by measuring leakage neutron spectra from polyethylene and water samples [25]. The experimental schematic for measuring Fe sphere leakage neutron spectra is shown in Figure 1 [Figure 1: see original paper], including a neutron source, sample, collimated shield, detectors, and electronic acquisition system. Main experimental parameters are listed in Table 1 .

2.1 Neutron source

Pulsed 300 keV D ions were generated using a Cockcroft-Walton accelerator at the Key Laboratory of Nuclear Data in CIAE, bombarding a tritium-loaded titanium (T-Ti) target to produce approximately 14 MeV pulsed neutrons through the D(T, n)^{4}He reaction [26]. The T-Ti target had a diameter of 2.2 cm, an active zone diameter of 1.6 cm, and thickness of approximately 2.6 m. D-T neutron source parameters included beam intensity of ~10 A, pulse width of 2.5 ns, frequency of 1.5 MHz, and neutron yield of 2×10^8/s. Neutron yields were acquired using the associated particle method, measuring ^{4}He particles with an Au-Si surface barrier detector positioned 135° from the beamline [27-28].

2.2 Samples

Polyethylene and natural iron samples were selected, with polyethylene used to test measurement system accuracy. The spherical Fe shell model is shown in Figure 2 [Figure 2: see original paper], consisting of two joined hemispherical shells with two holes to accommodate the neutron source target tube, which holds associated ^{4}He particles to enable the T-Ti target to penetrate the shell center. Dimensions are listed in Table 1, and elemental mass ratios of the iron sphere sample are listed in Table 2 . A new spherical shell (Fe sphere No. 3) was composed of Fe spheres Nos. 1 and 2. To minimize the influence of the hemisphere fitting plane on the leakage neutron spectrum, the fitting plane was rotated approximately 15° when placing the sample, ensuring the detector views a complete iron sphere at a stereo angle.

2.3 Collimator

During experiments, neutrons interact not only with the sample but also with other laboratory equipment, and these scattered neutrons may enter the detector and create background counts. To reduce background influence, a combined collimating and shielding system was used, consisting of a pre-collimator and an additional collimator embedded in the wall [29]. By placing the detector outside the wall collimator, the measured background spectrum was significantly lower than other spectra, as shown in Figure 3 [Figure 3: see original paper].

2.4 Detector systems and data processing

Time-of-flight spectra of leakage neutrons passing through the spherical Fe sample were measured using an EJ-301 liquid scintillator detector (5.08 cm diameter, 2.54 cm thick) positioned 769$±$1 cm from the neutron source at 90° to the beam. The electronic circuit is shown in Figure 4 [Figure 4: see original paper]. The CAMAC bus-based acquisition system unified EJ-301 detector and beam pick-up signals into the KMAX system to obtain charge integration (QDC), pulse shape discrimination (PSD), and time-of-flight (TOF) spectra of leakage particles.

Signal QDC was used as particle energy (E), with energy scale calibrated using ^{137}Cs and ^{22}Na γ radioactive sources. For PSD, particle discrimination was performed based on different zero-crossing times of pulses generated by different particles, accomplished using a 2160A module from CANBERRA. TOF used the detector anode signal as the start time signal. A copper ring in the accelerator tube generated an induced charge when D-ion pulse beam clusters passed through, forming a pick-up signal used as the stop signal after appropriate delay. To determine actual neutron flight time, one cycle time was deducted from the observed time.

Neutrons and γ rays were screened through offline analysis using Kmax software to obtain neutron and γ time-of-flight spectra, where γ time-of-flight spectra provided the time distribution of pulsed neutrons. Final time-of-flight spectra were obtained using:

$$TOF_{real} = \frac{TOF_{meas} \times K}{\varepsilon_d \times N_\alpha \times S \times N_s}$$

where TOF_{real} is the actual neutron spectrum entering the detector, ε_d is detector efficiency, K is the coefficient from associated particle method neutron yield measurement, N_α is the number of measured alpha particles, S is detector area, TOF_{meas} is neutron events detected, and N_s is neutron source count. The final time-of-flight spectrum was obtained as neutron flux per unit neutron per unit area (cm^{-2}).

3. Simulation

Experimental simulations are an important component of shielding benchmark research. Simulations were implemented using the MCNP code to obtain neutron time-of-flight spectra using evaluation data from specific libraries, enabling direct comparison with experimental results. The neutron source, sample, and detector are the most important experimental parameters, and accurate description of the neutron source (energy spectrum distribution, angular flux distribution, and pulsed time distribution) and detector (neutron detection efficiency curve) is critical for ensuring simulation accuracy [30].

3.1 Energy spectra and angular distribution of the neutron source

MCNP simulations require neutron source input information, and description precision directly affects output accuracy. While energy spectrum and angular distribution can be obtained through both experimental measurement and simulation, accurate experimental measurements are difficult in an accelerator experimental hall. Therefore, the TARGET program was used to calculate the neutron source energy spectrum [31]. Developed by PTB in Germany, TARGET calculates energy spectrum and angular distribution of D-T neutron sources based on incident D-particle and target parameters (energy, pulse width, and tritium target thickness). Results are shown in Figure 5 [Figure 5: see original paper], where neutron energy decreases as exit angle increases. The neutron source spectrum and angular distribution calculated by TARGET can be directly used as MCNP neutron source parameters.

3.2 Time distribution of the pulsed neutron

Simulation of time-of-flight spectra requires precise description of neutron source pulsed time distribution. Pulse-time distributions for different samples were obtained by extracting leakage γ-particle time-of-flight spectra from measurements. In offline data analysis, γ-particles were selected to extract time-of-flight spectra, which after appropriate processing served as pulsed neutron time distributions, as shown in Figure 6 [Figure 6: see original paper].

3.3 Detector efficiency

A liquid scintillator detector measured fast neutron energy spectra, and the detector efficiency curve is an important parameter for neutron spectrum measurement. The efficiency curve was calculated using the NEFF program developed by PTB (Germany) [32], obtained after describing detector geometry, light response curve, and energy threshold. The threshold energy for simulating the detector efficiency curve was chosen as 0.3 times the Compton edge energy of ^{137}Cs, corresponding to 0.8 MeV neutron energy. Simulation results are shown in Figure 7 [Figure 7: see original paper] and stored as detector efficiency in the MCNP input card.

3.4 The time-of-flight spectrum

The energy spectra, angular flux distribution, pulse-time distribution of source neutrons, and liquid scintillator detector efficiency curve were described in the MCNP-4C code input card [33]. The experimental hall environment was complex and difficult to model accurately, so only neutrons within the detector solid angle to the sample were considered. Based on background measurements shown in Figure 3 [Figure 3: see original paper], neutrons scattering into the detector from walls, ground, and other facilities could be ignored. Structures significantly influencing incoming detector neutrons included the target, sample, and collimators. Therefore, the entire experimental hall was simplified as an air-filled cylinder, with target, collimators, and modeled according to actual structures and dimensions. Polyethylene and Fe data were adopted from CENDL-3.2, ENDF/B-VIII.0 [34], and JENDL-5.0 libraries, while other structural material nuclei information was obtained from CENDL-3.2. MCNP directly provided simulated time-of-flight spectra. A ring detector was used in simulation, and the obtained time-of-flight spectrum indicates neutron flux per unit area (cm^{-2}), enabling direct comparison with experimental measurements. The calculation model is shown in Figure 8 [Figure 8: see original paper].

4. Uncertainties Analysis

The final experimental time-of-flight spectrum is given by Equation 1, where the coefficient k of the associated particle method is corrected by comparing neutron source measurements and simulations. Thus, the value of K is:

$$K = \frac{N_{n,meas}}{N_{\alpha,meas} \times \varepsilon_d \times N_{n,sim} \times S}$$

where $N_{n,meas}$ is the neutron count from measuring the neutron source time-of-flight spectrum, $N_{\alpha,meas}$ is the corresponding associated α count, ε_d is detector efficiency from simulation, $N_{n,sim}$ is flux of the simulated neutron source time-of-flight spectrum, and S is detector area. According to Equations 1 and 2, TOF_{real} is:

$$TOF_{real} = \frac{TOF_{meas} \times N_{n,meas} \times N_{\alpha,sim}}{N_{\alpha,meas} \times N_{n,sim} \times N_s}$$

Since experimental data are finally normalized using relative coefficients, and the ratio of experimental neutron source measurements to simulation results serves as relative coefficient K, most systematic errors are reduced or eliminated, including angular errors from associated α particle and time-of-flight spectrum measurements. Remaining uncertainties include: statistical error from neutron source simulation ($ 0.5 0.5 0.2 $3%), and statistical uncertainties in neutron counts per channel [35].

5. Results and Discussion

5.1 Neutron source measurement results

The D-T neutron source TOF spectrum was measured directly, with results shown in Figure 9 Figure 9: see original paper. The leakage neutron spectrum comprises two components: direct neutron emission contribution and scattered neutron contribution. Direct contribution from the neutron source occurred primarily between 120–206 ns, while scattered neutrons appeared mainly between 206–620 ns, with target tube-scattered, air-scattered, and collimator-scattered neutrons constituting the majority.

The model in Figure 8 [Figure 8: see original paper] simulated the neutron source time-of-flight spectrum. Table 3 lists integral values for experimental and simulated results in both intervals and the C/E ratio (simulated/experimental). Experimental and simulated results for the direct contribution interval showed excellent agreement, confirming measurement system reliability. Experimental results exceeded simulations for the scattered neutron interval, with Figure 9(a) showing a peak near 350 ns (~2.5 MeV). This primarily results from unreacted deuterium (D) ions continuously depositing in the target during the experiment, subsequently reacting with incident D ions behind via the D(d, n)^{3}He reaction to produce ~2.5 MeV neutrons. This reaction probability increases with target usage time. Additionally, the experimental hall situation was more complex than the simulation model, making it impossible to account for all scattered neutrons, leading to lower simulation results.

5.2 Measurement results of the polyethylene sample

To further ensure measurement system reliability, leakage neutron spectrum measurements and simulations were performed for a standard polyethylene sample. CENDL-3.2, ENDF/B-VIII.0, and JENDL-5.0 library data were used for polyethylene material in simulations, with results shown in Figure 9 Figure 9: see original paper.

By calculating the energy distribution of emitted neutrons after interaction with C and H nuclides, the leaking neutron spectrum from 14 MeV neutrons interacting with polyethylene spheres can be approximately divided into three intervals: (1) The elastic scattering interval (120–200 ns, depending on source neutron pulse width) primarily includes C and H elastic scattering, with minimum neutron energy from C elastic scattering approximately 10.5 MeV (180°). (2) The H elastic scattering interval (200–250 ns) is dominated by H elastic scattering. (3) The low-energy neutron interval includes C inelastic scattering, the C(n, n' 3α) reaction, H elastic scattering, and multiple scattering.

Table 4 lists experimental result integral values for each interval, simulated integral values for different libraries, and C/E values. Using hydrogen elastic scattering cross-section as the standard, the comparison shows CENDL-3.2 and JENDL-5.0 C/E deviations in the H elastic scattering interval are within 3%, while ENDF/B-VIII.0 simulation deviation is less than 1%, confirming measurement system reliability. Notably, ENDF/B-VIII.0 simulation results in the low-energy neutron interval are significantly underestimated by approximately 9% compared to experimental results, primarily because the C(n, n' 3α) reaction neutron energy spectrum in ENDF/B-VIII.0 is significantly lower than in other libraries.

5.3 Measurement results of the Fe sample

Time-of-flight spectra of leakage neutrons from deuterium-tritium pulsed neutrons interacting with spherical Fe samples of three thicknesses (4.5, 7.5, and 12 cm) were measured using the time-of-flight method. MCNP simulations calculated leakage neutron spectra for spherical Fe samples using CENDL-3.2, ENDF/B-VIII, and JENDL-5.0 libraries. Figure 10 [Figure 10: see original paper] compares experimental and simulated results.

14 MeV neutrons interact with natural iron through four main reaction channels: elastic scattering (n, el), discrete energy level inelastic scattering (n, inl)D, continuous energy level inelastic scattering (n, inl)C, and (n, 2n) reactions. NDPlot software can plot the secondary neutron energy spectrum for natural iron samples incident with 14 MeV neutrons, as shown in Figure 11 [Figure 11: see original paper]. Combining experimentally obtained pulse time widths, the time-of-flight spectrum was divided into four intervals based on different reaction channel energy ranges, as listed in Table 5 .

According to Figure 11, the entire experimental energy spectrum is divided into four intervals: 12.5-16 MeV for elastic scattering (n,el), 8.5-12.5 MeV for discrete energy level inelastic scattering (n,inl)D, 2.4-8.5 MeV for continuous energy level inelastic scattering (n,inl)C, and 0.8-2.4 MeV for the (n,2n) reaction.

Experimental and simulated spectra were integrated according to these energy intervals, yielding experimental integrated values, simulated integrated values, C/E values, and neutron penetration rates for different thicknesses, as listed in Table 6 . Table 7 shows C/E values for different thicknesses.

Based on Tables 6 and 7 results, the following conclusions can be drawn: (1) The neutron penetration rate in the 0.8-16 MeV interval slightly exceeds 1 at 4.5 cm thickness because the relatively small thickness prevents neutrons from decelerating below 0.8 MeV, and neutron multiplication occurs via the (n,2n) reaction. As sample thickness increases, neutrons continuously decelerate, neutrons below 0.8 MeV become more abundant, and penetration in the 0.8-16 MeV interval decreases. (2) The neutron scattering interval between 4-16 MeV includes elastic, discrete energy-level inelastic, and continuous energy-level inelastic scattering. Since leakage neutrons in this interval are primarily affected by scattering neutron cross-sections, transmittance exhibits exponential decay. The penetration rate follows the exponential decay law $y = e^{-Ax}$, where y is penetration rate and x is Fe sample thickness. Fitting results using experimental data from Table 6 are shown in Figure 12 [Figure 12: see original paper], with fitting parameter $A = 0.15495$ for the elastic scattering region and 0.08423 for the 2.4–16 MeV region. (3) Figure 13 [Figure 13: see original paper] shows C/E value variation for 0.8–16 MeV neutrons with different spherical iron thicknesses using various libraries. CENDL-3.2 provides a slightly larger total cross-section, ENDF/B-VIII.0 provides a smaller one, and this deviation increases with thickness. JENDL-5.0 simulation results agree sufficiently with experimental results, with overall deviation controlled within 3%. (4) Figure 3 and Table 6 demonstrate that as thickness increases from 4.5 cm to 12.5 cm, leakage neutrons in the elastic scattering interval gradually decrease. When thickness increases from 4.5 cm to 7.5 cm, leakage neutron count in the inelastic scattering interval significantly reduces, but does not change significantly from 7.5 cm to 12 cm. This occurs because leakage neutrons in the inelastic scattering interval increase as high-energy neutrons decelerate but decrease as thickness increases; the net change results from both effects. These competing effects also apply to the (n,2n) interval counting law. Moreover, since only neutrons above 0.8 MeV were measured, many neutrons decelerated below this threshold and were not counted, causing the overall (n,2n) interval leakage neutron count trend to decrease then increase with thickness. (5) C/E values from all three libraries gradually decreased with increasing thickness for continuous energy-level inelastic scattering intervals, while C/E values for discrete energy-level inelastic scattering intervals gradually increased, suggesting that inelastic scattering cross-sections in all three libraries may be slightly large while elastic scattering cross-sections are slightly small. (6) Simulated values for the continuum energy level inelastic scattering interval from both JENDL-5.0 and ENDF/B-VIII.0 libraries were approximately 10% lower than experimental values for all three thicknesses, indicating that the continuum energy level inelastic scattering cross-sections for 14.5 MeV incident neutrons on natural iron are too small in these libraries. As shown in Figure 14 [Figure 14: see original paper], the secondary neutron energy spectrum from ENDF/B-VIII.0 is larger between 0–3 MeV but smaller between 3–8 MeV, which may cause underestimation of simulation results in this interval. CENDL-3.2 simulation results showed varying biases in the (n,2n) interval.

6. Summary

This study conducted an integral sphere Fe shielding experiment using a D-T pulsed neutron source at the China Institute of Atomic Energy. Leakage neutron spectra from Fe spheres with thicknesses of 4.5, 7.5, and 12 cm were measured using the time-of-flight method. To test iron data reliability in CENDL-3.2, ENDF/B-VIII.0, and JENDL-5.0 libraries, time-of-flight spectra were simulated using MCNP-4C code. In addition to the physical model, the simulation fully considered detector efficiency, neutron source energy and angular distributions, and pulsed neutron time distributions, producing time-of-flight spectra with improved fine structure that facilitates comparison for short time intervals. The entire time-of-flight spectrum was divided into four intervals: elastic scattering (n, el), discrete energy level inelastic scattering (n, inl)D, continuous energy level inelastic scattering (n, inl)C, and (n, 2n) reaction intervals. Simulation and experimental results in different intervals were compared to verify data for various reaction channels.

Comparison results indicate: (1) CENDL-3.2 simulation results show the best agreement with experimental results. (2) JENDL-5.0 simulations were approximately 8% lower in the continuum energy level inelastic scattering interval, while ENDF/B-VIII.0 simulations were approximately 10% lower, likely because both libraries underestimate the secondary neutron energy spectrum in the high-energy portion of continuum energy level inelastic scattering.

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