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2026-01-06

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Abstract

The cross sections of the ^{113,115}In(n,γ) reactions play a critical role in s-process nucleosynthesis in nuclear astrophysics and in the performance of Ag-In-Cd control rods in commercial nuclear reactors. Neutron capture cross sections of indium from 1 eV to 1 MeV were measured at the Back-n facility of the China Spallation Neutron Source (CSNS) using the time-of-flight (TOF) technique. In the resolved resonance region, the resonance parameters of ^{113}In and ^{115}In between 1 eV and 2000 eV were extracted with the R-matrix code SAMMY, and discrepancies between the present results, previous experimental data, and evaluated nuclear data libraries were analyzed. In the unresolved resonance region, astrophysical Maxwellian-averaged cross sections (MACSs) of the ^{113,115}In(n,γ) reactions were derived over the temperature range relevant for s-process nucleosynthesis, based on the newly determined averaged cross sections. At a typical s-process thermal energy of $kT = 30$ keV, the MACS of ^{115}In(n,γ) is found to be about 20% higher than the value recommended in the Karlsruhe Astrophysical Database of Nucleosynthesis in Stars (KADoNiS).

Full Text

Preamble

Neutron Capture of Indium at s-process Temperatures at the CSNS Back-n

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The cross sections of the ^{113,115}In(n, γ) reaction play a critical role in s-process nucleosynthesis in nuclear astrophysics, Neutron Resonance Capture Analysis for determining elemental and isotopic composition of archaeological and cultural heritage objects, and Ag-In-Cd control rods in commercial nuclear reactors. We measured the neutron capture cross sections of indium from 1 eV to 1 MeV at the Back-n facility of the Chinese Spallation Neutron Source (CSNS) using the time-of-flight (TOF) method. In the resolved resonance region, resonance parameters of ^{113,115}In were extracted with the R-matrix code SAMMY from 1 eV to 2000 eV, and inconsistencies between available experimental data and evaluated libraries were discussed. In the unresolved resonance region, astrophysical Maxwellian-averaged cross sections (MACSs) of the ^{113,115}In(n, γ) reaction were calculated within the temperature range of s-process nucleosynthesis models based on the new averaged cross sections. At a typical s-process temperature of kT = 30 keV, the MACS of ^{115}In(n, γ) is found to be about 20% higher than the recommended value in the Karlsruhe Astrophysical Database of Nucleosynthesis in Stars.

Keywords: Neutron capture cross section, China Spallation Neutron Source (CSNS), Pulse Height Weighting Technique (PHWT), C6D6 detector, Maxwellian Averaged Cross Section (MACS)

Introduction

Stellar nucleosynthesis theory generally holds that heavy elements in the Universe are produced mainly through neutron capture processes in stars, including the slow neutron capture process (s-process) and the rapid neutron capture process (r-process). Indium, as a heavy element, is predominantly synthesized through the s-process occurring over thousands of years in stars with masses between 0.6 and 10 solar masses.

Natural indium has two stable isotopes: ^{113}In (4.29%) and ^{115}In (95.71%). The s-process path near indium isotopes is shown in [Figure 1: see original paper]. The origin of ^{113}In, classified as a p-nucleus, remains partially unresolved—while it can be produced directly in both s- and r-processes as well as through the extremely slow β-decay of ^{113}Cd (with an extraordinary half-life of approximately 8 quadrillion years), these known nucleosynthetic mechanisms appear insufficient to fully explain its observed cosmic abundance. ^{113}In serves as one of the sensitive indicators for overproduction by models of the p- and s-processes.

Through successive neutron captures, ^{112}Cd transforms into ^{115}Cd that then decays into the radioactive isotope ^{115}In, accounting for its relatively high cosmic abundance compared to stable ^{113}In. A key point is that ^{115}In cannot be produced via the ^{114}In(n, γ)^{115}In reaction due to the short half-life of ^{114}In

($t_{1/2}$ = 71.9 s). Its rapid β^- decay to ^{114}Sn effectively bypasses this direct path. Upon capturing a neutron, ^{115}In forms ^{116}In. Precise measurements of the ^{115}In(n, γ)^{116}In neutron capture cross section are crucial for understanding the nucleosynthesis of ^{115,116}Sn in the Universe.

The existence of resonance structures in neutron-induced reaction cross sections constitutes the fundamental principle underlying Neutron Resonance Transmission Analysis (NRTA) and Neutron Resonance Capture Analysis (NRCA). As robust non-destructive analytical techniques, NRTA and NRCA enable precise determination of both elemental and isotopic compositions of diverse materials and specimens. Ancient bronze artifacts frequently contain substantial quantities of impurities, including arsenic, antimony, indium, cobalt, and nickel, with certain elements present at notable concentrations. These bronze relics thus represent ideal subjects for investigation via NRCA. In specific experimental contexts, NRCA is further integrated with Time-of-Flight Neutron Diffraction (TOF-ND) measurements, and the resonance structures of the ^{113,115}In(n, γ) cross sections form the basis of NRCA for determining elemental and isotopic composition of archaeological and cultural heritage objects.

Indium serves as an important material in nuclear technology due to its significant thermal neutron capture cross section. Primarily utilized as a neutron-absorbing material in components such as reactor control rods, it is also employed for neutron flux monitoring in reactor dosimetry. The initial core mass composition of Ag–In–Cd control rods is typically specified as 80% silver (Ag), 15% indium (In), and 5% cadmium (Cd). Consequently, obtaining accurate neutron capture cross-section data for indium is essential for optimizing the design and performance of both neutron spectrometers and nuclear reactor control systems.

However, previous neutron capture experiments on indium and evaluated data in major libraries are not determined with high precision. In the resolved resonance region, data from J. Yoon et al., obtained via time-of-flight using a BGO detector, cover energies from 3.09 meV to 23.7 keV, but do not provide clear resonance structures above 30 eV. E. Haddad et al. employed a liquid scintillation counter to obtain data in the energy range from 1.05 eV to 21.3 keV, which conformed well to evaluated data between 1 eV and 100 eV in terms of peak size and position. The data from Yu. P. Popov et al. cover the widest energy region of 1 to 21 keV, but their energy resolution is relatively limited. In the unresolved resonance region above 2 keV, numerous previous measurements exist, with little difference between data from different evaluation libraries. The data from V. N. Kononov et al. and D. Kompe are in good agreement with the evaluation libraries.

In this work, the neutron capture cross sections of indium in the 1 eV–1 MeV energy region were measured employing the CSNS Back-n white neutron source with improved accuracy and wider energy coverage. The experimental details are described in Sec. II, data preprocessing methods in Sec. III, results and discussion in Sec. IV, and conclusions in Sec. V.

II. Measurements

CSNS is China's first high-power proton accelerator-based neutron source, which generates high-intensity white neutrons through spallation using a 1.6 GeV proton beam incident on a tungsten target. It aims to provide a powerful platform for both fundamental scientific research and high-tech development across many application fields.

The Back-n white neutron source at CSNS is specifically designed for nuclear data measurements through time-of-flight techniques and multidisciplinary neutron-based applications. Back-n provides the world' s highest flux white neutron beam (10^7 n cm^{-2} s^{-1} at 55 m from experimental station ES#1) with a continuous energy spectrum from 0.5 eV to 200 MeV.

Neutron capture cross-section measurements were conducted at experimental station ES#2 of Back-n, situated approximately 76 meters from the spallation target, as shown in [Figure 2: see original paper]. The double-bunch operation mode of CSNS was applied in this work to generate higher neutron flux and save experimental time for material science, life science, resource environment, new energy, and other applications. Under this mode, the accelerator produces proton beams with two bunches separated by 410 ns at a pulse frequency of 25 Hz. The resulting temporal overlap of neutrons from the double bunches was resolved to the single-bunch spectrum using a Bayesian-based double-bunch unfolding method.

To shape the neutron beam into a circular Gaussian profile, a neutron shutter is placed at 20 m from the spallation target and two neutron collimators are positioned in front of each experimental station. A cadmium (Cd) foil in the neutron shutter absorbs neutrons with energies below 0.3 eV to avoid overlap between neutron pulses.

Four samples were used in this study: natural indium (natIn), natural carbon (natC), natural lead (natPb), and an empty target. natC and natPb are used to determine the background from scattered neutrons and in-beam γ-rays. The ^{197}Au sample serves as the standard to calculate the relative cross section of natIn(n,γ) in the energy range from 1 eV to 1 MeV. The sample parameters are shown in . Additionally, a Ta-Ag-Co filter with thickness of 1.0 mm + 0.4 mm + 1.4 mm was used to reveal the in-beam γ-ray level in the beam using the black resonance method at experimental station ES#1.

The time-of-flight (TOF) method is essential for determining neutron energy in a white neutron source, as shown in Eq. (1):

$$E_n = m_n c^2 \left(\frac{1}{\sqrt{1-(L/T_n)^2/c^2}} - 1 \right)$$

where L is the effective neutron flight length and T_n is the neutron flight time. The γ-rays emitted by decay of the compound nucleus are detected by the C6D6 system, placed at experimental station ES#2. The detector system consists of four C6D6 detectors with aluminum detector brackets and an aluminum sample holder. The C6D6 detectors are liquid scintillation detectors that use deuterated benzene as the scintillating material, featuring low neutron sensitivity and fast time response.

III. Data Preprocessing

Data preprocessing includes detection efficiency correction using the pulse height weighting technique (PHWT), background subtraction to remove unwanted counts, and correction for multiple scattering and self-shielding effects.

The pulse height weighting technique is essential for (n,γ) cross-section measurements using the C6D6 detection system. The weighting function WF, approximated by a fifth-order polynomial, makes the detection efficiency $\epsilon_{\gamma i}$ proportional to the incident γ-ray energy $E_{\gamma i}$, as:

$$\sum_i \epsilon_{\gamma i}(E_d, E_{\gamma i}) WF(E_d) = \kappa E_{\gamma i}$$

$$E_{\gamma i} = S_n + E_n$$

where E_d is the deposited energy in the C6D6 detectors, $\epsilon_{\gamma i}(E_d, E_{\gamma i})$ is the detector response function to the ith γ with energy $E_{\gamma i}$ determined by GEANT4 Monte Carlo simulations, and κ is a proportionality constant. S_n is the neutron binding energy and E_n is the kinetic energy of the incident neutron in the center-of-mass system.

The original spectra for different targets are shown in [Figure 3: see original paper]. To obtain the true counts of the (n,γ) reaction from these data, background subtraction is essential. The background can be divided into two categories: target-independent background $N_{\text{empty}}(t_n)$, which can be eliminated by measuring the spectrum of an empty target, and target-dependent background $N_{\text{sample}}(t_n)$, which varies between nuclides and comprises activation background N_{ac}, scattered neutron background $N_{\text{sn}}(t_n)$, and in-beam γ background $N_{\text{s}\gamma}(t_n)$. Thus, the total background composition is:

$$B_{\text{total}}(t_n) = N_{\text{ac}} + N_{\text{empty}}(t_n) + N_{\text{sample}}(t_n)$$

$$N_{\text{sample}}(t_n) = N_{\text{sn}}(t_n) + N_{\text{s}\gamma}(t_n)$$

The neutron activation background, arising from decay of isotopes produced by neutron capture, is determined by fitting the plateau observed at flight times $<$

10.7 ms (< 0.3 eV). In this region, beam γ-rays are negligible and a cadmium filter removes neutrons, eliminating scattering and capture effects.

The scattered neutron background is caused by neutrons scattered from the sample into the environment. The neutron capture cross section of carbon is relatively small compared to its neutron scattering cross section, and its in-beam γ scattering is also weak, making it useful for subtracting the neutron scattering background. Taking the indium target as an example, its neutron scattering background can be expressed as:

$$N_{\text{sn,In}} = \frac{Y_{\text{el,In}}}{Y_{\text{el,C}}}(N_C - N_{\text{empty}} - N_{\text{ac,C}})$$

The in-beam γ-rays are generated by neutron capture reactions in the water moderator around the spallation target. Each sample has a different in-beam γ-ray background, which can be normalized by a factor K. Lead has a large γ scattering cross section and a small neutron capture cross section. The black resonance filtering method is used to obtain K by measuring spectra of the Pb target and In target with an Ag-Co-Ta-Co filter. The filter creates absorption valleys at 4.28 eV, 124.787 eV, and 5.016 keV, where counts are contributed mainly by activation and in-beam γ-rays:

$$N_{\text{s}\gamma,\text{In}} = K(N_{\text{Pb}} - N_{\text{empty}} - N_{\text{sn,Pb}} - N_{\text{ac,Pb}})$$

where $Y_{\text{el,C}}$ and $Y_{\text{el,In}}$ are the neutron scattering yields of the carbon and indium targets, respectively, N_C is the spectrum of the carbon target, and $N_{\text{ac,C}}$ is the activation background. N_{Pb} is the spectrum of the lead target, and $N_{\text{sn,Pb}}$ is the neutron scattering background from the lead target. The effect of this processing procedure is shown in [Figure 3: see original paper]. Finally, the true counts of the (n,γ) reaction for the target can be expressed as:

$$N_{\text{In,net}} = N_{\text{In}} - B_{\text{total}}$$

After processing with PHWT, the yield Y_w of the studied target can be expressed as:

$$Y_w = \frac{N_{\text{In,net}}}{\epsilon_\gamma I F}$$

where ϵ_γ denotes the probability of detecting neutron capture events from Eq. (2), I denotes the neutron flux, and F is the normalization factor. Finally, the neutron capture cross section σ_γ can be obtained by:

$$\sigma_\gamma = \frac{Y_w}{1 - e^{-N_s \sigma_{\text{tot}} f_c}}$$

where σ_{tot} is the total neutron cross section from evaluated data ENDF/B-VIII.1, N_s is the atomic surface density of the sample, and f_c is the correction factor for multiple scattering and self-shielding effects.

In the resonance region, neutron multiple scattering increases the effective target thickness, while self-shielding causes the neutron flux I to vary with target thickness. In the resolved resonance region (RRR), these effects are considered in the corrections of the SAMMY code. In the unresolved resonance region (URR), the corrections can be treated as an energy-dependent factor f_c, determined by GEANT4 simulations.

The main uncertainty sources are shown in , including experimental conditions uncertainty, data analysis method uncertainty, and experimental statistical uncertainty. The neutron energy spectrum uncertainty was determined by Y. Chen et al. based on analysis of the Back-n neutron beam spectrum using calibrated Li-Si detectors and a fission chamber. Before the experiment, uncertainties in the sample target' s geometric parameters were determined, and target material purity was considered, controlling the total sample target uncertainty within 0.5%. During the experiment, the proton beam current uncertainty was recorded at about 1.5%. The PHWT and double-bunch unfolding method uncertainties are estimated to be less than 1.1% and 2.0%, respectively. The normalization uncertainty remains less than 1.5%. The statistical uncertainty stays below 3.47%. Therefore, the experimental results demonstrate relatively high precision, with errors controlled within approximately 10% across the entire 1 eV to 1 MeV energy range.

IV. Results and Discussions

A. Resolved Resonance Region

This study used the R-matrix code SAMMY to analyze neutron capture yields and extract resonance parameters from 1 eV to 2 keV. The capture yields of natural indium were fitted within the multilevel R-matrix framework using the Reich-Moore approximation, with systematic corrections applied for resolution broadening, multiple scattering, self-shielding, and moderation length. During resonance parameter extraction, parameters from evaluated nuclear data libraries such as ENDF/B-VIII.1 and JENDL-5 were used as initial priors, ultimately yielding experimental resonance parameters including resonance energy E_R, resonance spin J, radiative width Γ_γ, neutron width Γ_n, and orbital angular momentum L. All resonance parameters have been deposited in the Science Data Bank. Due to coupling between Γ_γ and Γ_n, the directly measurable quantities are often limited to the resonance energy E_R and the total capture kernel k, which is proportional to the area under an isolated resonance:

$$k = g\frac{\Gamma_\gamma \Gamma_n}{\Gamma_\gamma + \Gamma_n}$$

where g is a statistical factor defined as:

$$g = \frac{2J+1}{(2s+1)(2I+1)}$$

with $J = |L+S|$, $S = s+I$, I representing the spin of the target nucleus in its ground state (determined based on specific sample nucleus characteristics), and s denoting the intrinsic spin of the neutron (1/2).

In the SAMMY code, reaction cross sections for different channels can be precisely calculated by incorporating resonance parameters with the R-matrix formalism, along with transformations of intermediate parameters and matrices. The SAMMY fit results for capture yields are shown in [Figure 4: see original paper]. The fitted neutron capture yields Y_{SAMMY} were converted to neutron capture cross sections of natIn(n,γ) in the RRR, as shown in [Figure 5: see original paper]. These results provide insight into points of contention in previous evaluation libraries, with discrepancies mainly concentrated below 500 eV.

Our experimental data are in good agreement with prior measurements by J. Yoon et al. and Yu. P. Popov et al. across the 0–30 eV energy range. However, beyond 30 eV, their datasets contain too few points to show clear resonance peaks, whereas our experiment provides well-resolved resonance structures above 30 eV. E. Haddad et al. show very good agreement with this work between 1 eV and 100 eV in terms of peak size and position.

The resonance structure identified at 26.78 eV in our measurements and in other evaluated nuclear data libraries is not reported in CENDL-3.2. The present results show better agreement with JEFF-3.3 and JENDL-5 than with ENDF/B-VIII.1 in terms of peak height at 73.04 eV and 86.32 eV. The resonance peaks observed in this experiment at 224.12 eV and 227.61 eV align more closely with peak positions in ENDF/B-VIII.1 and TENDL-2023, and are located to the right of those in JENDL-5. At 228.10 eV and 250.45 eV, the peak positions and resonance heights in this work show closest agreement with TENDL-2023. At 289.19 eV and 294.10 eV, the measured resonance positions match those in ENDF/B-VIII.1 and TENDL-2023. At 267.18 eV, the peak positions in the present results show closest agreement with TENDL-2023. The measured resonances at 354.29 eV, 362.71 eV, 371.54 eV, 383.29 eV, and 384.23 eV also align more closely with ENDF/B-VIII.1 and TENDL-2023 than with JENDL-5. The peak heights of this result at 366.67 eV and 379.38 eV show good agreement with ENDF/B-VIII.1 and TENDL-2023. The same trend is confirmed for resonances at 449.11 eV, 454.27 eV, and 457.14 eV, whose positions show better agreement with ENDF/B-VIII.1 and TENDL-2023, providing additional evidence for systematic deviation from JENDL-5. At 422.75 eV, the peak position in the present result is intermediate between those in the four evaluation libraries. The peak height of this result at 472.84 eV shows closest agreement with JEFF-3.3 and JENDL-5.

In summary, our experimental data show systematic agreement with ENDF/B-VIII.1 and TENDL-2023 across the measured energy range, while exhibiting discernible peak position offsets relative to JENDL-5. Due to peak broadening caused by limited energy resolution in certain regions, some resonance structures, particularly those with cross sections below 1 barn, cannot be reliably identified. Hence, facilities with higher energy resolution and further experimental investigation are strongly recommended using isotopically enriched samples.

B. Unresolved Resonance Region

The averaged experimental neutron capture cross sections from this work are compared with evaluation libraries and previous experiments in [Figure 6: see original paper]. The experimental data in the unresolved resonance region (URR) from 2 keV to 1 MeV are available at https://doi.org/10.57760/sciencedb.33726.

Since 1960, more than ten experiments have been conducted for natural indium according to the EXFOR database. Notably, the energy region of this result is wider than all previous measurements, spanning three orders of magnitude. The natIn(n,γ) result agrees basically with four evaluation libraries below 200 keV within uncertainty limits. Good agreement is observed between our result and values measured by V. N. Kononov et al. and D. Kompe, while some discrepancies exist with other measurements. Data from H. Xu et al. are lower than this work, whereas results reported by L. M. Spitz et al. and J. Hellström are slightly higher.

The TALYS code was employed to compute average cross sections of natIn(n,γ) from 2 keV to 1 MeV. Given the absence of experimental resonance parameters for individual isotopes ^{113,115}In, the average radiation width $\langle\Gamma_\gamma\rangle$ and s-wave average resonance level spacing D_0 from ENDF/B-VIII.1 and JENDL-5 were adopted as key constraints in TALYS calculations. The theoretical cross section for natIn(n,γ) was subsequently derived from the abundance-weighted average of the two isotopic cross sections. Results indicate good agreement between TALYS-calculated cross sections for ^{113,115}In(n,γ) and both experimental measurements and evaluated data in the unresolved resonance region. Furthermore, experimental data from this work are consistent with TALYS-based theoretical predictions within a 10% margin of error.

The resulting theoretical average neutron capture cross sections for the ^{113,115}In(n,γ) reaction are presented in [Figure 6: see original paper] (b) and (c). For ^{113}In, our results lie between JEFF-3.3 and JENDL-5 evaluated libraries. Within the 400–1000 keV range, the cross sections of ^{113}In(n,γ) are in good agreement with data from H. A. Grench and H. O. Menlove. For ^{115}In, our results show best agreement with JEFF-3.3 and JENDL-5 evaluated libraries. The ^{115}In(n,γ) cross section at 24 keV has been measured by W. W. Campbell and N. Yamamuro et al. based on the Sb-Be photoneutron source, and our results are consistent with Campbell' s experiment at this energy point.

In the 4-400 keV energy region, our ^{115}In(n,γ) results are consistent with V. N. Kononov et al. and J. S. Brzosko.

C. Maxwellian-Averaged Cross Section

To investigate the s-process involving ^{113,115}In within stellar environments, it is essential to integrate experimentally determined (n,γ) cross sections with the kinetic energy distribution of neutrons (E_n) present in stellar plasma. This integration is performed using Eq. (13) to derive MACS:

$$\mathrm{MACS} = \frac{\int_0^\infty \sigma(E_n) E_n e^{-E_n/kT} dE_n}{\int_0^\infty E_n e^{-E_n/kT} dE_n}$$

where $\sigma(E_n)$ is the averaged experimental capture cross section calculated by TALYS code in this work. The MACS from kT = 5-100 keV computed in this work are shown in [Figure 7: see original paper]. Data from the Karlsruhe Astrophysics Database of Nucleosynthesis in Stars (KADoNiS), previous experimental values, and evaluated libraries are also shown for reference. MACS values from this work and KADoNiS v0.3 are listed in for comparison.

Differences exist between this work and evaluated values. For ^{113}In, the present results show good agreement with ENDF/B-VIII.1 and KADoNiS v0.3. For ^{115}In, the results are slightly higher than values in KADoNiS v0.3 and all four evaluation libraries.

At the thermal energy of kT = 30 keV relevant to nuclear astrophysics, the MACS values for ^{113,115}In(n,γ) are 818.1 ± 81.8 mb and 841.2 ± 84.1 mb, respectively. Comparisons between this work and evaluated values, theoretical values, and experimental values including KADoNiS v0.3 are illustrated in [Figure 7: see original paper]. For ^{113}In, this work aligns with the majority of evaluated values and the KADoNiS v0.3 recommended value, except for values provided by B. J. Allen et al. The theoretical values of J. A. Holmes et al., S. Goriely (2002), and S. Goriely (2005) deviate evidently from this work' s experimental results. For ^{115}In, the present results are consistent with most evaluated values and several experimental values. The MACS values from this work are found to be about 20% higher than the KADoNiS v0.3 recommended value.

V. Conclusions

This study presents new high-precision measurements of neutron capture cross sections of natural indium in the energy range of 1 eV to 1 MeV, conducted at the Back-n white neutron beamline of the China Spallation Neutron Source. The experiment employed C6D6 liquid scintillation detectors combined with the time-of-flight technique to achieve high-resolution data. We provide a brief description of the experimental setup, detector system, and sample characteristics, and discuss the data preprocessing procedure and analysis methods, including the pulse height weighting technique, double-bunch unfolding method, and background subtraction.

In the RRR, the R-matrix code SAMMY was employed to derive accurate resonance parameters in the energy range of 1 eV–2 keV, providing new insights that help resolve discrepancies among existing evaluation data libraries. In the URR, averaged neutron capture cross sections were obtained from experimental measurements. Finally, MACSs within the thermal temperature kT range of s-process nucleosynthesis models were calculated through numerical integration. The present results for the ^{115}In(n,γ) reaction are found to be about 20% higher than the recommended value used in nucleosynthesis calculations in KADoNiS v0.3. This is a significant result for the role of the s-process in nuclear astrophysics and should motivate additional experiments to verify this result and nucleosynthesis calculations to study its impact on the s-process.

These results provide critical benchmarks for s-process nucleosynthesis of ^{113,115}In in nuclear astrophysics, neutron resonance capture analysis for determining bulk composition of materials, research and development of neutron-absorbing materials in commercial nuclear reactors, and nuclear data evaluations.

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