Neutron Resonance Transmission Analysis for Isotope Content Quantification at CSNS Back-n

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Abstract

This study uses experimental validation of Neutron Resonance Transmission Analysis (NRTA) utilizing the Back-n beamline at the China Spallation Neutron Source. Natural silver is used as a surrogate material for 99 Tc, with 1 mm and 2 mm thick samples configured to simulate 50% variations in isotopic content. The EJ-270 plastic scintillator detector, combined with the time-of-flight method, is utilized to acquire neutron energy spectra. During data processing, black resonance techniques are applied for background subtraction, while Doppler broadening and energy resolution function for correction to cross-section are implemented to mitigate thermal motion effects of nuclei and beamline structural impacts. A gradient descent algorithm is employed for least-squares fitting of experimental data to invert isotopic areal density and theoretical transmission. Results confirming NRTA' s efficacy in isotopic content quantification and achieving effective identification of 50% isotope content changes. This investigation validates NRTA' s potential for spent nuclear fuel transmutation monitoring in an Accelerator Driven Sub-critical System (ADS), and the research results indicate that NRTA is sensitive to the resonance of neutron beamline materials themselves and the quality of statistical data.

Full Text

Preamble

Neutron Resonance Transmission

Analysis

Isotope Content Quantification Back-n Yaoxuan Xueying -Zhang Yonghao-Chen Liang -Chen Yongqin Zhichao Yanbin -Zhang Hongling Zhendong-An Wenli Zhenhua-Wu Hanyu-Deng

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Abstract

study experimental validation Neutron Resonance Transmission

Analysis

(NRTA) utilizing Back-n beamline China Spallation Neutron Source.

Natural silver surrogate material thick samples configured simulate variations isotopic content.

EJ-270 plastic scintillator detector, combined time-of-flight method, utilized acquire neutron energy spectra.

During processing, black resonance techniques applied

background

subtraction, while oppler broadening energy resolution function correction cross section implemented mitigate thermal motion effects nuclei beamline structural impacts. gradient descent algorithm employed least-squares fitting experimental invert isotopic

areal density theoretical transmission.

Results

onfirming NRTA' s efficacy isotopic content quantification achieving effective identification isotope content changes. investigation validates NRTA' s potential spent nuclear transmutation monitoring Accelerator Driven Sub-critical System (ADS), research

results

indicate sensitive resonance neutron beamline materials themselves quality statistical

Keywords

NRTA, CSNS, Isotope identification, Nondestructive

Introduction

global low-carbon energy transition aimed addressing climate crisis, nuclear energy owing energy density carbon emissions plays pivotal carbon neutrality strategies According International Atomic Energy Agency, total installed capacity operational nuclear power reactors worldwide reached China installed nuclear capacity surpassed reflecting steady year-on-year growth 1.86%. annual nuclear electricity generation risen accounting 4.86% national power avoiding approximately million tonnes emissions compared coal-fired generation Despite these significant contributions carbon reduction, sustainable development nuclear energy faces major challenges: tight supply nuclear long-term risks associated geological disposal high-level radioactive waste.

These issues remain critical technical barriers expansion nuclear power Accelerator Driven Sub-critical System (ADS) employs broad-spectrum spallation neutron external neutron source sustain transmutation spent within subcritical reactor. system enables generation energy output recovers nuclear materials while converting long-lived fission products (LLFPs) minor actinides (MAs) medium-short-lived, low-radioactive isotopes through spallation neutron bombardment.

Therefore, effectively minimizes waste valuable resources fissile materials other isotopes, significantly reduces production high-level radioactive waste, which important direction future development reactors

During operation essential implement offloading procedures transmutation-qualified assemblies prevent neutron resource wastage, while continuing transmutation non-transmutation-qualified assemblies. quantitative criteria transmutation qualification characterized specific nuclide concentrations within fuel, including LLFPs

237 Np

assembly deemed transmutation-qualified concentrations these characteristic nuclides decrease predetermined threshold levels.

Given inherent radioactivity

background

assemblies requirement secondary transmutation non-transmutation-qualified assemblies, imperative develop non-destructive detection techniques capable accurately quantifying characteristic nuclide concentrations without destroying geometric integrity assembly, order dynamic management assembly.

Neutrons serve non-destructive probes analyzing material internal characteristics owing their exceptional transmission capabilities.

Priesmeyer first proposed application Neutron Resonance Transmission

Analysis

(NRTA) non-destructive

method

determining isotopic abundances nuclear rods[11]. fundamental principle involves utilizing time-of-flight (TOF) techniques measure energy spectrum white neutron transmitted through sample determine content material elements observing position intensity transmission dips. differences neutron interaction cross-sections among different nuclides, unique fingerprint nuclides provides physical basis technology identify nuclide types contents conducted non-destructive

analysis

copper samples complex geometries using technology

results

showed technology combined derived analytical expressions achieve non-destructive isotope

analysis

complex geometric samples, fitted areal density agreement reference value Harufumi Tsuchiya colleagues developed integrated active neutron non-destructive

analysis

system 2023, which conducted experiments metal plate samples.

results

confirmed system could accurately determine nuclear material areal densities, validating feasibility compact systems quantitative

analysis

nuclear constituents 2024, conducted feasibility study based Back-n beamline China Spallation Neutron Source (CSNS), demonstrating application potential field nondestructive isotopic detection 2026, further completed measurement study neutron cross-sections natural silver Back-n beamline, further validating capability applications NRTA, non-destructive

method

nuclide identification quantification, enables absolute

analysis

without requiring calibration. characteristic makes particularly suitable monitoring nuclide concentrations assemblies. validate experimental measurement

method

post-processing technology NRTA, assess measurement accuracy sensitivity concentration variations, research conducted using natural silver samples varying thicknesses Back-n facility.

Experimental setup

experiment

carried using Institute Energy Physics, Chinese Academy Sciences. proton energy pulse repetition frequency bombard tungsten targets, generating high-flux neutrons through spallation reactions.

Back-n beamline ability provide neutron beams broad energy ranging mainly serving precise measurement nuclear advanced nuclear technology application research Figure illustrates schematic spatial layout Back-n beamline, experimental flight length approximately meters.

Figure.1 Back-n beamline

experiment

adopts non-radioactive nuclide substitution scheme solve problems limited access radioactive materials radiation safety hazards.

shown Figure typical exhibits neutron resonance cross-sections within energy range similar natural silver.

Natural silver non-radioactive consists stable isotopes, forming natural isotope composite system. allows verify technology capability multi-isotope quantification.

Based these characteristics, natural silver selected substitute study. experimental sample consisted natural silver sheets, thick sample groups thicknesses obtained stacking, equivalent change isotope content.

Three measurement samples experiment: sample, single-layer sample thick), double-layer sample thick). samples placed Station Figure.2 Cross section natural silver EJ-270 plastic scintillator detector selected

experiment

neutron detection capabilities ability effectively discriminate between neutrons gammas pulse shape discrimination detector operated working voltage during experiment. avoid interference in-beam neutrons gamma photomultiplier detector, detector arranged perpendicular line. detector positioned Station during measurement.

experiment

utilized DT5751 Acquisition System (DAQ) developed signal acquisition, stored format local storage media.

Under neutron conditions, system generates substantial streams; insufficient write speeds buffer saturation trigger protective pause mechanisms, resulting valid Concurrently, neutron

induces signal pile-up effects, significantly complicating subsequent processing.

experiment

smallest collimator aperture combination Back-n beamline minimize neutron solve above problem beamline accompanies intense gamma-ray, combined EJ-270 detector’ s gamma sensitivity, exacerbated stream volume signal pile-up, impairing neutron signal acquisition.

Therefore, shield installed neutron window attenuate gamma-ray.

Additionally, avoid coincidence thermal neutrons previous cycle neutrons cycle, cadmium plate installed window filter thermal neutrons.

Experimental Principles Processing Neutron flight energy During process proton bombardment tungsten target, gamma flash narrow width intensity induced. event manifests multi-peak superposition feature waveform acquired DT5751. gamma flash signal effectively identified processing developed based Python.

Therefore, processing, neutron start obtained calculating gamma flash signal recorded EJ-270 detector, other signals acquired detector taken time.

Then, calculation formula neutron time-of-flight (TOF) expressed where which other signals arrive detector, time; gamma flash arrives detector; flight distance target station detector; speed light.

Using calculate start neutron. Since neutron energy range analyzed exhibited significant relativistic effects, neutron energy calculation performed using Equation

$= 1\ 2$

where neutron mass. Experimental transmission experimental transmission obtained ratio sample energy spectrum no-sample energy spectrum

background

contributions deducted:

푄푃 $=$

background

determined using black resonance technique. technology inserts samples resonance resonance filter) beam. filter, resonance observed spectrum.

Under resonance conditions, theoretical transmission position approaches near-zero values (theoretical transmission sufficient) However, practical measurements reveal non-zero counts position, which attributed

background

contributions. Therefore, considered count non-zero

background

count, fitting curve

background

obtained fitting positions spectrum through least squares method. variation

background

fitted analytical formula consisting constant exponential terms:

= 0 + 1 퍐 − 1 + 2 퍐 − 2 # 4

Parameter represents contribution time-independent term. first exponential originates gamma generated capture neutrons hydrogen elements, while second exponential primarily arises neutrons scattered internally within station where detector located. sample utilized

experiment

maximum thickness Silver exhibits resonance peaks 5.19, 16.30, 30.60, 51.56, 71.00, theoretical transmission within these resonance range getting maximum value satisfies black resonance technique

requirement transmission below enabling sample black resonance filter. shown Figure neutron spectrum fitted

background

spectrum presented. Notably, several non-silver resonance overlap fitted

background

spectrum spectrum. phenomenon presence spallation target CSNS, these isotopes exhibit resonance peaks introduce extraneous resonance features.

Figure.3 Transmission neutron spectrum fitted

background

spectrum

Doppler broadening

thermal motion atomic nuclei induces relative kinetic energy shifts. thermal motion forming statistical process energy distribution, resonance peaks undergo broadening. phenomenon termed Doppler broadening. cross section Doppler broadening assumes adherence Maxwell-Boltzmann distribution gases, which Doppler broadening equation derived shown Eq(4)

’ = 1

Where represents effective cross section temperature, incident particle velocity. corresponds relative cross section velocity between incident particle target nucleus.

Doppler broadening

required cross sections performed using processing, section broadening.

Figure.4 Doppler broadening non-broadening total cross section resonance Energy resolution function nominal neutron flight distance

experiment

approximately meters. shown Figure neutrons generated different spatial positions within spallation target, majority requiring complex scattering processes within target before exiting evacuated tube. event detector, neutrons interact scintillator varying regions, generating signals.

Consequently, actual flight distance neutrons generally exceeds nominal value exhibits distinct spatial distribution. addition, proton non-zero width, resulting neutrons being generated simultaneously, rather being continuously distributed certain period time.

These factors introduce systematic deviations between neutron energy spectrum measured

method

energy spectrum. difficult invert neutron energy spectrum energy spectrum obtained through measurement processing.

Figure.5 influence neutron flight process practical applications, energy resolution function characterize factors flight distance distribution distribution.

Subsequently, convolutional broadening treatment applied theoretical cross section, shown Equation thereby incorporating these factors analysis.

Where energy resolution function, cross section after Doppler broadening, cross section after

energy resolution broadening.

Different experimental devices different energy resolution functions structural differences. instance, n_{TOF} GELINA neutron sources utilize function characterize energy resolution, ORELA facility adopts function. function developed Jiang Back-n beamline study energy resolution broadening processing cross section performed through Python code.

Least squares fitting gradient descent

method

Least Squares

Method

(LSM) numerical fitting approach obtains optimal parameters iteratively updating parameters theoretical models minimize discrepancies between experimental theoretical values. fundamental widely applied techniques parameter estimation, extensively nonlinear fitting.

method

employs experimentally measured (e.g., neutron transmission spectra NRTA) theoretical model unknown parameters (e.g., theoretical formula transmission calculation). continuously iterating theoretical parameters, squared residuals between theoretical experimental values minimized. parameters minimized squared residuals regarded optimal fitting parameters.

NRTA, theoretical transmission uniform sample expressed Where, neutron energy, areal density nuclide fitted, where atomic volume number density nuclide, sample thickness. neutron cross section nuclide includes effects Doppler broadening

energy resolution broadening.

squared residuals between experimental transmission theoretical transmission given

퓺 2 =

reduce influence experimental values relatively large errors, weighted least squares adopted, which weights assigned residuals different points weights generally taken reciprocal measurement errors,

퓺 2 =

where measurement error calculated Equation

퓺 = 1

where denotes effective neutron count. Owing existence types count spectra, namely transmitted neutron spectrum sample neutron spectrum without sample, error experimental transmission propagated Equation after experimental transmission obtained using Equation (10).

퓺퓐 = 퓺

where represent points errors transmitted neutron spectrum sample neutron spectrum without sample, respectively. fitting process nonlinear fitting, Figure presents program chart calculation nonlinear least squares fitting.

Since nonlinear least squares fitting requires iterative computation numerical methods, iterative algorithms include Gauss-Newton method, Levenberg-Marquardt method, Gradient Descent method, among others.

Among these,

method

features simple principle, implementation, requirement matrix inversion, making suitable datasets large number points.

Figure.6 program chart Essentially,

method

adjusts parameters fitted along "descending direction" (also referred downhill direction negative gradient direction) squared residuals function" , ultimately reaching "lowest point" (minimum value) function.

Therefore, implementation principle Gradient Descent

method

expressed

1 +1 = 1 − 휄

2 +1 = 2 − 휄

+1 =

where denotes areal density particles nuclide, initial value assigned initiate iterative calculation. subscripts refer iteration iteration, respectively. represents update rate, which controls magnitude parameter adjustment iteration code: excessively large value prevent convergence optimal value, while excessively small value convergence speed. function, which function characterize magnitude discrepancies between theoretical experimental values, multiple optional forms.

Equation adopted function study iterative calculation process essentially consistent program chart shown Figure difference being process "adjusting parameters" therein implemented using method.

Equation indicates calculated iteration; then, under control gradient reassigned along direction update parameter

+1 , until convergence to the minimum

decreasing function become squared residuals achieved. criterion convergence change nuclide content threshold Based Python programming language, procedural implementation gradient descent iterative algorithm completed, applied numerical solution process nonlinear least squares fitting.

Discussion

exhibits double-bunch distribution terms time. unfolding based Bayes theorem developed invert measured energy spectrum under double-bunch conditions equivalent

result

single-bunch However, processing already taken account influence double-bunch energy resolution function double-bunch relatively small impact energy range involved study.

Therefore, there perform double-bunch unfolding operation energy spectrum here.

Figure shows count spectra different samples, resonance induced natural silver observed.

Figure.7 Count spectra different samples After performing

background

subtraction, cross section broadening, least-squares fitting during processing, Table presents fitting

results

obtained using different neutron libraries,

results

represented particle number areal density n/barn. values parentheses table denote fitting

results

ratio true. closer ratio unity, better fitting performance. value derived sample thickness, density, combined isotopic abundance.

Table.1 Fitting

results

different library

Thickness ENDF/B-VIII.1 JEFF-4 TENDL-2023 11.02% 10.81% 10.82%

107 Ag

(1.10) (1.25) (1.24) 7.93% 7.82% 7.80%

109 Ag

(1.09) (1.01) (1.05) 10.36% 10.29% 10.33%

107 Ag

(1.12) (1.17) (1.16) 7.69% 7.77%

109 Ag

(1.09) (1.01) (1.05) According Table content obtained fitting JEFF-4 library close value, whereas content derived fitting ENDF/B-VIII.1 library consistent value.

Figure presents comparison between fitted transmission distribution (plotted using cross-section JEFF-4 library cross-section ENDF/B-VIII.1 library) experimental data, transmission curve areal density included. shown Figure fitted transmission distribution extremely close transmission distribution, which indicates algorithm effectively achieves inversion experimental results.

Figure.8 Fitting

results

different samples

background

subtraction relatively level, measurement duration single sample approximately hours during experiment.

results

several hundred effective neutron counts experimental points, leading relatively large statistical uncertainty. experimental featuring uncertainty,

results

fitted algorithm study still consistency values, which fully demonstrates effectiveness algorithm employed research inverting isotopic contents. effective

neutron count further improved through experimental optimization sample measurement duration appropriately extended, statistical characteristics experimental enhanced, accuracy fitting

results

expected further improved. Table shows that, regardless nuclear database used, relative uncertainty fitted

results

significantly higher phenomenon mainly caused interference tungsten components spallation target CSNS.

Figure.9 Comparison transmission distribution between natural Tungsten Silver silver sample, theoretical transmission curve natural tungsten thickness

results

indicate multiple resonance exhibit significant overlap resonance tungsten.

Given actual thickness tungsten spallation target considerably greater black resonance absorption region formed tungsten almost completely cover resonance Consequently, effective neutron count resonance positions decreases significantly during measurements, ultimately leading larger uncertainty fitted

results

study, total thickness silver sample reduced adjusting stacking amount silver foils, which equivalently achieved reduction content silver isotopes.

Table lists relative changes fitted contents where relative change defined 퓲퓟
퓞 퓳퓞퓟표 퓲퓟 퓞

fitting

results

obtained using ENDF/B-VIII.1 library, while those derived JEFF-4 library.

Table.1 Relative changes isotopic content Relative changes -50.73% -50.04%

results

demonstrate technique exhibits recognition sensitivity relative change isotopic contents fitted values agreement actual results, whereas those slightly poorer correspondence, which attributed relatively measurement accuracy isotopic content.

result

confirms, certain extent, technique applied nondestructive quantitative detection relative changes isotopic contents during transmutation spent fuel, thereby providing indirect monitoring

method

transmutation process.

Conclusion

Transmission experiments based natural silver samples Back-n beamline achieve nondestructive quantitative detection isotopic content.

Experimental

results

indicate exhibits detection capability relative variations isotopic content, measurement precision meeting application requirements. efficacy gradient descent algorithms fitting experimental validated, confirming their capacity effectively isotopic content information.

results

reveal absolute non-destructive quantitative technique requires prior calibration provide effective non-destructive testing solution monitoring spent transmutation.

Experimental

results

indicate resonance characteristics materials constituting neutron source target station exert influence measurement accuracy technique. dedicated detection apparatus, selection optimization target station structure constituent materials should fully taken account early design stage, minimize interference impact resonance absorption characteristics target station materials final measurement results, thereby enhancing measurement accuracy reliability.

addition, shows sensitivity statistical uncertainties experimental data, necessitating advancements experimental methodologies mitigate challenge.

Proposed optimizations include: employing low-gamma sensitivity detectors implementing shielding structure optimizations. reduce detector, thereby increasing effective neutron count achieving higher precision isotope content measurement. future, further research conducted samples cylindrical complex isotopic compositions.

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Note: Figure translations are in progress. See original paper for figures.