



Neutron absorption spectroscopy for identification of light elements in actinides

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Abstract

We are developing cryogenic high energy resolution fast-neutron spectrometers using superconducting transition-edge sensors (TES) for nuclear science and non-proliferation applications. Fast neutrons are absorbed in 94% enriched ${}^6\text{LiF}$ single crystals with volumes of $\sim 1\text{ cm}^3$ in an exothermic ${}^6\text{Li}(n,\alpha){}^3\text{H}$ capture reaction. The neutron energy is measured from the subsequent temperature rise with a Mo/Cu multilayer TES. Fast-neutron spectra from a ${}^{252}\text{Cf}$ source show an energy resolution of 55 keV. Here we discuss the instrument performance, with emphasis on the identification of light elements in actinide matrices. © 2005 Elsevier Science. All rights reserved

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1. Introduction

Calorimetric fast-neutron spectroscopy based on exothermic (n,α) reactions is an alternative method to conventional approaches like time-of-flight, ${}^3\text{He}$ ionization chambers or proton-recoil proportional counters [1 - 4]. Fast neutrons with kinetic energy E_n deposit a total energy $E_{\text{total}} = E_n + Q_{\text{reaction}}$ in an absorber crystal. If E_{total} is distributed among the reaction products with stopping ranges of a few microns, E_{total} can be measured from the rise in

absorber temperature with a superconducting TES. This method offers high energy resolution, a simple response function and easy gamma-ray discrimination. While neutron-emitting actinides can be identified even with low energy resolution spectrometers, the presence of light-element inclusions like C, F or O in the actinide matrix can only be detected from the unique elemental signatures resulting from neutron scattering and absorption resonances in the MeV energy range [5, 6]. These measurements require high energy resolution to detect the narrow resonances, which can be achieved at detector operating temperatures of $\sim 0.1\text{ K}$ [7, 8].

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2. Monte Carlo simulations

As an example, consider the presence of oxygen in plutonium. The neutrons emitted by spontaneous or induced fission of Pu interact with Pu and O nuclei. The neutron transport for a simple unshielded spherical PuO₂ sample has been simulated with Monte Carlo N-Particle code (MCNP) [9] (Fig. 1). A spectrometer with an energy resolution of $\Delta E_{FWHM} = 10$ keV can easily visualize spectral features due to the ¹⁶O elastic scattering resonances that are not detectable with $\Delta E_{FWHM} = 100$ keV.

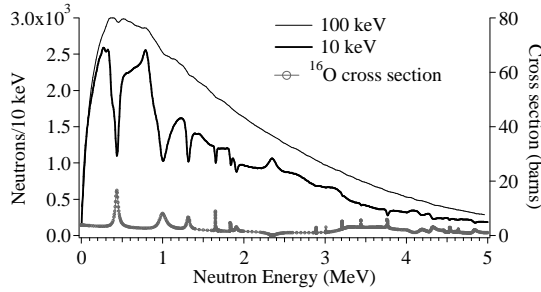


Fig 1. MCNP simulation of the neutron spectra of PuO₂ for different instrumental energy resolution. Resonances in the neutron scattering cross section of ¹⁶O (bottom trace) cause the dips in the neutron spectra at the corresponding energies [10].

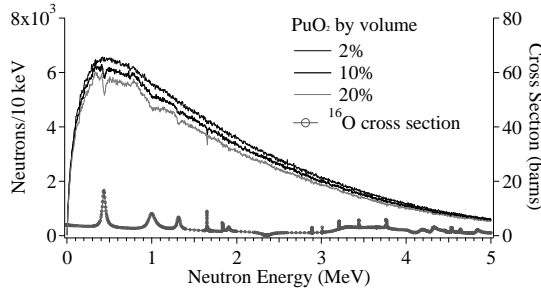


Fig 2. MCNP simulation of a metallic Pu sample with different PuO₂ content at $\Delta E_{FWHM} = 10$ keV.

A minimum of $\sim 10\%$ PuO₂ by volume distributed as a thin layer around a metallic Pu sample can be identified with a resolution on the order of 10 keV (Fig. 2). In addition, the effects of shielding can be quantified for high-Z materials like Pb and Fe, while low-Z thermalizing materials like polyethylene smooth out spectral features (Fig. 3). A statistical noise below 20% per 10 keV FWHM energy bin requires a minimum of 5×10^5 total counts to achieve the resolution in Fig. 1. With a target count rate of about 100 Hz for our spectrometer, such a spectrum can be acquired in about 2 hours.

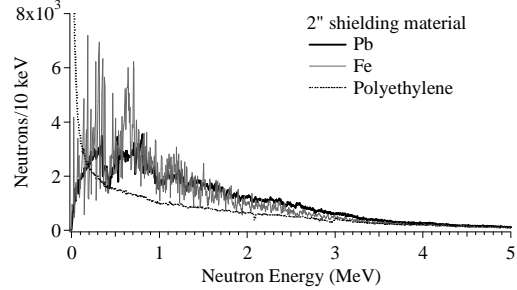


Fig 3. Effect of shielding on the PuO₂ neutron spectrum seen with $\Delta E_{FWHM} = 10$ keV. Densely packed, narrow resonances in Fe produce large fluctuations of the neutron flux.

3. Experiment

In an earlier demonstration experiment, a prototype neutron detector based on the ¹⁰B(n, α)⁷Li reaction in a 1 mm³ metallic TiB₂ absorber has produced $\Delta E_{FWHM} = 5.5$ keV with thermalized neutrons from a ²⁵²Cf neutron source [11]. A similar energy resolution with a factor of 1000 increase in detection efficiency is possible by using dielectric LiF crystals with similar heat capacity of a few nJ/K at 0.1 K and with volumes on the order of cm³.

In this experiment, we use the ⁶Li(n, α)³H reaction ($Q_{\text{reaction}} = 4.782$ MeV) in a 94% ⁶Li-enriched 0.7 \times 0.7 \times 0.4 cm³ ⁶LiF crystal. The crystal was grown by the vertical Bridgman technique, cut and polished to optical quality [12]. The sensor is a 200 nm Mo/Cu multilayer TES deposited on a 2 \times 2 \times 0.5 mm³ SiN-coated Si substrate glued with GE-7031 varnish onto the ⁶LiF crystal. An epoxy-coated Al screw in contact with the Si chip is used to control the thermal conductivity G_{TES} to the cold bath and thus the signal decay time by applying variable mechanical tension. The crystal is installed on 1/32" sapphire balls at the end of a cold finger inside the refrigerator [13]. The TES is voltage-biased, and signals are read-out with a commercial JeSEF SQUID preamplifier and optimally filtered off-line.

Fast-neutron spectra from a 2 μ Ci ²⁵²Cf source shielded with 1 cm of Pb are obtained for two different values of G_{TES} by adjusting the mechanical tension on the Al screw. Shorter pulses have been obtained for bigger tension, corresponding to a larger value of G_{TES} . The spectra have the same $\Delta E_{FWHM} = 55$ keV obtained from the thermal neutron peak at $Q_{\text{reaction}} = 4.782$ MeV, with an electronic noise of 6.5 keV (Fig. 4).

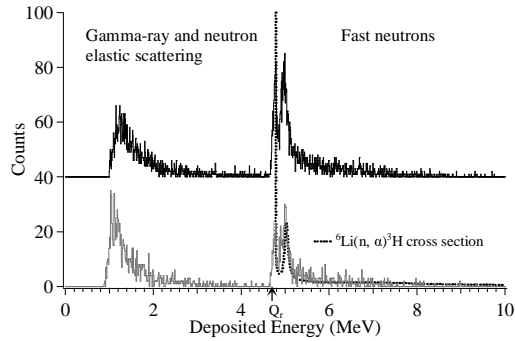


Fig. 4. Detector response to a ^{252}Cf fast-neutron source for detectors with different decay times τ_d . The spectra reflect the energy dependence of the $^6\text{Li}(n,\alpha)^3\text{H}$ reaction cross section (dotted line). The short pulse device (top) has $\tau_d = 7.6$ ms and $\tau_r = 0.17$ ms, the long pulse device (bottom) has $\tau_d = 18.6$ ms and $\tau_r = 1.12$ ms. The top spectrum is shifted by 40 counts for clarity.

The $1/e$ rise time τ_r is set by phonon propagation inside the absorber with width W and phonon transport across the absorber-Si interface, $\tau_r \approx W/v_{\text{ph}} + (C_{\text{Si+TES}} + C_{\text{LiF}})/G_{\text{abs}}$. For our high quality ^6LiF crystals, the phonon propagation is ballistic, and a phonon velocity of 4.9×10^3 m/s causes the energy to spread throughout the crystal within a few μs [14]. The thermal coupling $1/G_{\text{abs}} = 1/G_K + 1/G_v$ between the absorber and the Si chip is due to the Kapitza thermal contact resistance $G_K = 1.6 \times 10^{-3} \text{T}^3/\text{A}$ assuming a contact area $A = 4 \text{ mm}^2$, plus the thermal conductivity of the varnish $G_v = 105 \mu\text{W/K}$ [15]. The heat capacities $C_{\text{LiF}} = 0.19 \text{ nJ/K}$ and $C_{\text{Si+TES}} = 11 \text{ pJ/K}$ at $T_c = 155 \text{ mK}$ are calculated from the Debye law [16]. The theoretical rise time is $\tau_r = 28 \mu\text{s}$, a factor of 5 less than the smallest experimental value in Fig. 4. This may be due to reduced phonon transport across the varnish, as suggested by the reduction of τ_r when tightening the Al screw and by the shorter τ_r for events absorbed in the Si chip.

The decay time is $\tau_d = (C_{\text{Si+TES}} + C_{\text{LiF}})/G_{\text{TES}}$, and G_{TES} can be determined by irradiating the detector with gamma-rays only, which will interact with higher probability in the Si chip and then decay to the cold bath with a decay time $\tau_{d,\text{gamma}} \approx C_{\text{Si+TES}}/G_{\text{TES}}$. For $\tau_{d,\text{gamma}} \approx 1$ ms we obtain $G_{\text{TES}} \approx 12.5 \text{ nW/K}$ and thus $\tau_d \approx 11$ ms, in agreement with the observations.

In summary, we have built a cryogenic fast-neutron spectrometer with a 94% ^6Li -enriched ^6LiF

absorber crystal with an energy resolution of 55 keV FWHM. Its pulse decay time can be reduced without loss in energy resolution by adjusting the thermal conductivity to the TES chip with an Al screw. This allows the detection of light elements in actinide matrices through their nuclear scattering resonances within a few hours.

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