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TITLE: LEAD 207, 208 (N, XN GAMMA) REACTIONS FOR NEUTRON ENERGIES UP TO 200 MEV

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SUBMITTED TO: ORNL, International Conference on Nuclear Data for Science and Technology
Gatlinburg, TN USA
May 9-13, 1994

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$^{207,208}\text{Pb}(n,xn\gamma)$ REACTIONS FOR NEUTRON ENERGIES UP TO 200 MeV

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ABSTRACT

High-resolution γ -ray spectra from the interaction of neutrons in the energy range from 3 to 200 MeV with $^{207,208}\text{Pb}$ were measured with the white neutron source at the WNR facility at Los Alamos National Laboratory. From these data, excitation functions for prominent γ transitions in $^{200,202,204,206,207,208}\text{Pb}$ were derived from threshold to 200 MeV incident neutron energy. These γ -production cross sections represent formation cross sections for excited states of the residual nuclei. The results are compared with the predictions of nuclear reaction calculations based on the exciton model for precompound emission, the Hauser-Feshbach theory for compound nuclear decay, and coupled channels calculations to account for direct excitation of collective levels. Good agreement was obtained over the entire energy range covered in the experiment with reasonable model parameters. The results demonstrate that multiple preequilibrium emission has to be taken into account above about 40 MeV, and that the level density model of Ignatyuk should be used instead of the Gilbert-Cameron and back-shifted Fermi-gas models if excitation energies exceed about 30 MeV.

I. INTRODUCTION

Until now, almost no data existed for cross sections of neutron-induced reactions for the formation of specific residual nuclei for neutron energies above 30 MeV. This is mostly due to the fact that the activation method requires monoenergetic neutron sources which do not exist in this energy range. One way to study specific reaction channels at higher energies is to use a pulsed white neutron beam and to measure the prompt γ -radiation emitted in neutron-induced reactions using high resolution Ge-detectors. The neutron energy is determined by time-of-flight (TOF) and the residual nuclei can be identified by the characteristic γ energies for transitions between

low-lying levels. In general, neutron-induced reactions leave the residual nucleus in a highly excited state which decays via a γ cascade of typically 3 to 4 steps. For even-even nuclei direct transitions to the 0^+ ground states are strongly inhibited and often more than 90% of the γ cascades proceed to the ground state (gs) via the first 2^+ levels. For these nuclei we get approximately the total production cross section by measuring the first $2^+ \rightarrow \text{gs}$ γ -ray transition. If the residual nucleus is not even-even, partial production cross sections are obtained from the production cross sections for a given γ -ray transition, which are also of interest, because they indicate the population of an individual state in the same sense as the production of isomeric states. Here, however, the states have short half-lives, whereas the less numerous isomeric states have long half-lives.

$^{207,208}\text{Pb}$ samples were selected for a comparison between theory and experiment because a large amount of data already exists to restrict the parameter space for the nuclear model calculations. In the ^{208}Pb region there are also very large shell effects in the nuclear level density, and it is thus possible to study the dependence of these shell effects on excitation energy with high sensitivity. Finally, there exists very good spectroscopic information on the discrete levels and γ -branching ratios for all lead isotopes reached by (n,xn) reactions up to $x = 9$ (^{200}Pb).

II. EXPERIMENT AND DATA REDUCTION

The experiments were performed on the 30° left flight path of the WNR facility.¹ The general features of the 30° left Ge spectrometer setup and of the beam pulse structure are described in Ref. 2. A schematic diagram of the flight path collimation and shielding is shown in Fig. 1. The isotopically enriched Pb samples (92.78% ^{207}Pb and 99.56% ^{208}Pb , respectively) were mounted at a distance of 41.48 m from the neutron production target on a thin

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plastic frame. Two high-purity coaxial Ge detectors with active volumes of approximately 70 cm³ and 140 cm³ were used at γ -emission angles of 90° and 125°. The collimators were steel tubes filled with tungsten powder, because the usual lead shielding emits the same γ rays as the isotopic lead targets when excited by scattered neutrons.

Fig. 1. Experimental setup.

In this paper we report data from the 125° detector only. This position was chosen because the value of the P_2 Legendre polynomial function is zero at that angle. The angle integrated cross section can then be approximated as 4π times the measured cross section at $\theta=125^\circ$, provided the coefficients of the higher-order polynomials are small.

The neutron energy range from 3 to 200 MeV was divided into 53 groups with increasing widths according to the energy resolution of the experiment. The neutron flux was measured with a fission chamber positioned at a distance of 37.30 m from the production target. It contained both ²³⁵U and ²³⁸U fission foils. The neutron fluence for each energy group was determined from the two-dimensional (neutron TOF versus fission pulse height) fission chamber spectra. The ²³⁵U(n,f) and ²³⁸U(n,f) cross sections given by Lisowski et al.³ were used.

Two-dimensional spectra, neutron TOF versus γ pulse-height, were recorded for the Ge detector. The time resolution, determined from the γ -ray flash from the neutron-production target, varied from 10 ns FWHM for $E_\gamma = 200$ keV to 5 ns FWHM for $E_\gamma = 3$ MeV. The γ -ray energy resolution obtained during the experiments was 2.8 keV FWHM at a γ -ray energy of 803 keV. For each neutron energy group a one-dimensional γ pulse-height spectrum was derived from the two-dimensional spectrum and analyzed. Relative excitation functions were derived for each γ transition studied. The differential cross sections at $\theta=125^\circ$ were converted to total γ -production

cross sections by multiplying them by 4π . Corrections were applied for the attenuation of the γ rays within the samples and for contributions of secondary neutrons to the γ production cross sections.

Because of uncertainty in our knowledge of the Ge detector dead time and the absolute flux intercepted by the irregularly shaped samples, the relative cross sections were normalized to data obtained in a separate 14-MeV measurement,⁴ performed at the Institute of Physics of the Slovak Academy of Sciences. Normalization factors were derived from the cross section of prominent γ -transitions in (n,n' γ) and (n,2n γ) reactions.

The γ -ray transitions analyzed in this work are listed in Table I. The 569.7-keV and 897.8-keV transitions from the ²⁰⁷Pb(n,n' γ) reaction (and the not resolved 899.1-keV transition from the ²⁰⁷Pb(n,4n γ) reaction) were not analyzed because low energy neutrons from the preceding micropulse (frame overlap) gave significant contributions to the measured γ production cross sections. Due to the high excitation energy (2614.6 keV) of the first excited level of ²⁰⁸Pb, the frame-overlap problem did not occur in studying the ²⁰⁸Pb(n,n' γ) reaction.

In the residual nuclei ^{200,202,204,206,207}Pb long-lived isomeric states ($t_{1/2} > 2$ ns) are involved in the cascade. When isomers are present in the cascade, the γ radiation from the first excited level $\rightarrow$ gs transition is not emitted promptly. Such delayed transitions cannot be properly correlated with the neutron energy because the measured TOF includes the decay delay. Therefore correction procedures had to be applied to ensure that the derived cross sections relate only to the prompt emission of γ rays. The cross sections associated with the prompt part of the γ cascade are the production cross sections for the relevant transitions minus the contributions from the long-lived isomers. According to the level schemes and γ -ray intensities of ^{200,202,204,206,207}Pb these contributions equal γ -production cross sections related to the decay of the lowest energy isomers. Therefore prompt γ -production cross sections for the first excited level $\rightarrow$ gs transitions can be expressed as differences of production cross sections for individual transitions as listed in the last column of Table I. (The influence of some weaker transitions in the decay of the isomers of ²⁰³Pb and ²⁰⁴Pb was neglected because their effect is small compared to the uncertainties of this experiment.) The delayed contributions to the measured first excited level $\rightarrow$ gs transition were determined from the time distributions of the decay of the isomers by analyzing the corresponding γ rays.⁵ In ²⁰⁴Pb the $2_1^+ \rightarrow 0_1^+$ transition ($E_\gamma = 899.1$ keV) was not resolved from the $E_\gamma = 897.8$ -keV $\rightarrow$ gs transition in ²⁰⁷Pb. Therefore we report the sum of these two cross

sections minus the contribution from the 1273.9-keV 4^+ isomer of ^{204}Pb .

III. CROSS SECTION CALCULATIONS

Extensive calculations of the γ -ray production cross sections studied experimentally were performed by means of the computer code GNASH,⁶ which describes the precompound mechanism of nuclear reactions by the exciton model, and compound nucleus decay by the Hauser-Feshbach model, and allows the addition of contributions from direct reactions. The aim of the cross section calculations was not to reproduce the experimental results merely by varying a large number of parameters but to test the ability of the simple exciton precompound model⁷ and the Hauser-Feshbach model of compound nuclear reactions to describe cross sections for specific reaction channels at higher energies than have been considered in the past. Calculations of $(n, x\gamma)$ cross sections for particular γ -ray transitions provide exacting tests of the nuclear models because such transitions select only a part of the angular momentum distribution involved in the overall reaction process.

Most model parameters were not varied but were kept at values typical of a large number of analyses of a variety of data at lower energies. For example, the matrix element normalization constant that describes the competition between precompound particle emission and internal transitions to higher exciton states in preequilibrium emission was fixed at a typical value of 135 MeV³. Direct collective excitation also was considered for a number of levels in ^{208}Pb , and was accounted for by the coupled-channels calculations performed with the code ECIS.⁸

For the nuclear level densities, the Ignatyuk model⁹ was chosen instead of the simpler Gilbert-Cameron¹⁰ and other Fermi-gas models that incorporate an energy-independent level density parameter. By the use of this energy-dependent level density parameter, the Ignatyuk model includes the theoretically expected disappearance of shell effects in the nuclear level densities at higher excitation energies. Within this model the nuclear moment of inertia was given the value of the full rigid body moment of inertia. More details about the various parameters used can be found in Ref. 5.

Three extensions in the modeling of preequilibrium reactions were installed in GNASH to improve the physics for calculations at higher energies. Until now it was assumed that after the first preequilibrium particle is emitted, the remaining particle-hole states proceed to equilibrium via a series of nucleon-nucleon collisions before decaying. We have modified this assumption to allow the

particle-hole states left after primary preequilibrium emission to decay by "multiple preequilibrium" emission. The second modeling improvement was to calculate spin distributions for the residual states formed in preequilibrium reactions using angular momentum distributions based on the exciton model.¹¹ And finally, we have incorporated the excitation-energy dependence of the Ignatyuk level-density formula into the particle-hole state densities used in the exciton model calculations. Cross-section calculations were performed with this *a priori* "best choice" parameter set and also with some modified parameter sets in order to illustrate the sensitivity of calculated cross sections to various model parameters.

IV. RESULTS AND DISCUSSION

Reasonable agreement is found between our measurements and the GNASH calculations performed with the *a priori* standard parameter set for all reactions. There are, however, some discrepancies which are discussed in Ref. 5 in detail.

As examples our results for the $^{208}\text{Pb}(n, 2\gamma)^{207}\text{Pb}$ and the $^{208}\text{Pb}(n, 7\gamma)^{202}\text{Pb}$ reactions are given in Fig. 2. The figure illustrates clearly the influence of multiple preequilibrium (PE) emission. Above neutron energies of about 40 MeV multiple precompound emission dramatically increases the $(n, 2n)$ and $(n, 3n)$ cross sections. As more high-energy neutrons are emitted, a larger fraction of the available incident energy is in two or three neutron emission steps, and correspondingly the cross sections for multi-particle emission decrease. The measured data also clearly support the use of the Ignatyuk level density model for energies above 30 MeV. As seen from Fig. 2b the use of the Gilbert-Cameron level density formula results in an effective reaction threshold much higher than observed in the experiment for $(n, 7\gamma)$ reactions as in the Gilbert-Cameron model considerably more energy is needed to produce a (n, xn) reaction with a specific x than in the Ignatyuk model (see Ref. 5). Calculations were also performed with a nuclear moment of inertia reduced to 60% of the rigid body value. The standard calculation gives a somewhat better overall agreement with the experimental data, but we cannot exclude the possibility of a reduced moment of inertia.

In summary, we have demonstrated the effectiveness of white source $(n, x\gamma)$ measurements, in which prompt γ rays are detected, for supplying data that provides useful tests of nuclear models over a wide energy range. Our calculations demonstrate the effectiveness of preequilibrium models at higher energies, and the importance of multiple preequilibrium particle emission and of the correct choice of the level density formula to accurately describe the data.

Fig. 2. Prompt γ production cross sections:
 (a) $^{208}\text{Pb}(n,2n\gamma)^{207}\text{Pb } 5/2^- \rightarrow \text{gs}$ transition ($E_\gamma = 569.7$ keV)
 (b) $^{208}\text{Pb}(n,7n\gamma)^{202}\text{Pb } 2^+ \rightarrow \text{gs}$ transition ($E_\gamma = 960.7$ keV)

Table I: Nuclear reactions and gamma transitions investigated

Reaction	γ transition	γ energy [keV]	Measured γ -production cross section
$^{207}\text{Pb}(n,2n\gamma)^{206}\text{Pb}$	$2^- \rightarrow \text{gs}$	803.1	$\sigma(803.1) - \sigma(516.2)$
$^{207}\text{Pb}(n,6n\gamma)^{202}\text{Pb}$	$2^+ \rightarrow \text{gs}$	960.7	$\sigma(960.7) - \sigma(657.5) - \sigma(787.0)$
$^{207}\text{Pb}(n,8n\gamma)^{200}\text{Pb}$	$2^- \rightarrow \text{gs}$	1026.5	$\sigma(1026.5) - \sigma(245.1)$
$^{208}\text{Pb}(n,n\gamma)^{208}\text{Pb}$	$3^- \rightarrow \text{gs}$	2614.6	$\sigma(2614.6)$
$^{208}\text{Pb}(n,2n\gamma)^{207}\text{Pb}$	$5/2^- \rightarrow \text{gs}$	569.7	$\sigma(569.7) - \sigma(1063.7)$
$^{208}\text{Pb}(n,3n\gamma)^{206}\text{Pb}$	$2^+ \rightarrow \text{gs}$	803.1	$\sigma(803.1) - \sigma(516.2)$
$^{208}\text{Pb}(n,5n\gamma)^{204}\text{Pb}$	$2^+ \rightarrow \text{gs}$	899.1 ^a	$\sigma(899.1) + \sigma(897.8) - \sigma(374.4)^a$
$^{208}\text{Pb}(n,7n\gamma)^{202}\text{Pb}$	$2^+ \rightarrow \text{gs}$	960.7	$\sigma(960.7) - \sigma(657.5) - \sigma(787.0)$
$^{208}\text{Pb}(n,9n\gamma)^{200}\text{Pb}$	$2^+ \rightarrow \text{gs}$	1026.5	$\sigma(1026.5) - \sigma(245.1)$

^a The 899.1-keV (^{204}Pb) and 897.8-keV ($3/2^- \rightarrow 1/2^-$ in ^{207}Pb) γ lines were not resolved.

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