TITLE: LEAD 208 (n,pxn gamma) REACTIONS FOR NEUTRON ENERGIES UP TO 200 MEV

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SUBMITTED TO: NEANC Specialist's Meeting on Measurement, Calculation and Evaluation of Photon Production Data
Bologna, Italy
November 14-17, 1994

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208Pb(n,pxrγ) Reactions for Neutron Energies up to 200 MeV

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ABSTRACT

The prompt gamma-radiation from the interaction of fast neutrons with enriched samples of 208Pb was measured using the white neutron beam of the WNR facility at Los Alamos National Laboratory. The samples were positioned at about 40 m distance from the neutron production target. The spectra of the emitted gamma-rays were measured with a high-resolution HPGe detector. The incident neutron energy was determined by the time-of-flight method and the neutron fluence was measured with a 238U fission chamber. In addition to the primary purpose of this experiment, the study of (n,xny) reactions leading to various lead isotopes, gamma transitions in the residual nuclei 207,205,203,201Tl were analyzed. From these data gamma-production cross sections in the neutron energy range from the effective thresholds to 200 MeV were derived. The lines for the analysis had to be chosen carefully as the (n,pnxy) cross sections are rather small and the interference with unresolved lead lines (even weak ones) would cause significant errors. The effect due to isomers with half-lives exceeding a few nanoseconds was taken into account and corrected for, if necessary. The measured cross sections were compared with the results of nuclear model calculations based on the exciton model for preequilibrium particle emission and the Hauser-Feshbach theory for compound nucleus decay. Unlike in the case of (n,xny) reactions the calculated results in general did not give a good description of the measured cross sections.

1. Introduction

One method to measure photon-production cross sections in neutron induced reactions is the use of a "white" neutron spallation source and high-resolution gamma-ray spectroscopy. The incident neutron energy is determined by the time-of-flight method and gamma-ray production cross sections can be measured simultaneously for a wide neutron energy range. Recently such measurements have been performed at the Weapons Neutron Research facility (WNR) at the Los Alamos National Laboratory in an energy range covering several hundreds of MeV. One of the recent experiments was a study of (n,xny) reactions on 207,208Pb in the neutron energy range from 3 to 200 MeV. The (n,xny) reactions were analyzed first, as neutron emission is the dominant reaction channel. Good agreement was found between the experimental results and model calculations performed with the code GNASH. These calculations are based on the Hauser-Feshbach formalism for compound nucleus decay and the exciton model for preequilibrium particle emission. Multiple preequilibrium particle

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emission was taken into account and the level density formula given by Ignatyuk\textsuperscript{5} was used. More details on these calculations can be found in Ref. 2.

The objective of this work was to identify gamma-rays related to less intense reaction channels in the already measured spectra. We analyzed \(^{208}\text{Pb}(n,p\gamma\gamma)\) reactions leading to various isotopes of Tl. As our nuclear model calculations can reproduce \((n,x\gamma\gamma)\) cross sections very satisfactorily it was the goal of this work to check these calculations for a minor channel.

2. Experiment

The experiment was performed on the 30° left flight path of the WNR facility and is described in detail in Ref. 2. A schematic diagram of the flight path collimation and shielding is shown in Fig. 1. The isotopically enriched Pb sample (99.56% \(^{208}\text{Pb}\)) was mounted at a distance of 41.48 m from the neutron production target on a thin plastic frame. Two high-purity coaxial Ge detectors with active volumes of approximately 70 cm\(^3\) and 140 cm\(^3\) were used at \(\gamma\)-ray emission angles of 90° and 125°. The collimators were steel tubes filled with tungsten powder, because the usual lead shielding emits the same gamma rays as the isotopic lead targets when excited by scattered neutrons. The detector position of 125° 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 4\(\pi\) times the measured cross section at \(\theta = 125^\circ\), provided the coefficients of the higher-order polynomials are small. Gamma rays from Tl isotopes were analyzed in the spectra measured with the 125° detector only.

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 containing a \(^{238}\text{U}\) fission foil centered on the beam at a distance of 37.30 m from the production target. The neutron fluence for each energy group was determined from the two-dimensional (neutron TOF versus fission pulse height) fission chamber spectra using \(^{238}\text{U}(n,f)\) cross sections given by Lisowski et al.\textsuperscript{6}

![Experimental setup](image-url)
Two-dimensional spectra, neutron TOF versus gamma 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.

3. Data Reduction

Guided by the results of model calculations performed with the code GNASH, such transitions were considered for analysis where rather large cross sections (more than about 10 mb in the maximum) were expected. As even weak gamma-lines from \(^{208}\text{Pb}(n,x\gamma)\) reactions reach such cross section values it was essential to select only such Tl-lines which could clearly be separated from the Pb lines in the gamma-ray spectra. Finally there must also be no interfering background lines, especially from the tungsten collimator.

A major problem in such white source experiments is the presence of isomers with half-lives exceeding a few nanoseconds in the residual nuclei. When isomers are present in the cascade preceding the gamma-transitions investigated, the measured gamma-radiation is not emitted promptly. Such delayed transitions may be detected in this type of experiment but they cannot be properly correlated with the neutron energy because the measured TOF includes the decay delay. Therefore correction procedures have to be applied to ensure that the derived cross sections relates only to the prompt emission of gamma rays. Due to the pulse structure of the WNR-facility the actual correction procedure depends on the half-life of the isomer. If the life-time of the isomer is large compared to the duration of a macropulse (typically 800 µs), most of the delayed γ-rays (about 98%) would be emitted between the macropulses when the detector electronics is gated off, and thus will not be counted. If the half-life of an isomer in the cascade is comparable to the duration of a macropulse, the delayed γ-rays are observed in the measured spectrum with a uniform distribution in time and can be subtracted as a constant background. The background rate then can be determined from the measured intensities below the effective reaction threshold. For isomers with shorter half-lives the delayed contribution to the measured transition can be determined from the time distributions of the decay of the corresponding isomers. (See Ref. 2 for details). The applicability of this last method depends on the decay scheme, γ-ray intensities and energies of transitions related to the decay of the isomers. It was not possible to use this method in any of the Tl-nuclei studied in this work. If the life-time of the isomer is smaller than the time resolution of the experiment the delay can be neglected.

According to the decay schemes\(^7\) and the γ-ray intensities, the transitions listed in Table I were chosen for analysis. Table I also shows the isomers which might cause delayed emission of the γ-rays.

As seen from Table I long-lived isomers have to be considered for the γ-transitions in \(^{205}\text{Tl}\) and \(^{201}\text{Tl}\). The isotope \(^{203}\text{Tl}\) does not have any known isomers, from \(^{207}\text{Tl}\) a prompt transition was chosen for analysis. No correction was necessary for the 203.8-keV isomer in \(^{205}\text{Tl}\) with a half-live of 1.46 ns. The average life-time of this isomer is well below the time resolution of our experiment and can therefore be neglected. To get a coarse estimate of the effect of the 1484.0-keV isomer (\( t_{1/2} = 4.5 \) ns) in \(^{205}\text{Tl}\) the data analysis was redone under the assumption the 203.7-keV and 720.1-keV γ-rays were partially delayed by a constant delay time equal to the mean life-time of this isomer. The fraction of the γ-ray intensity delayed was estimated using the results of model calculations. This procedure resulted in somewhat
different cross sections. On the average the differences were about 4%, much smaller than the statistical uncertainties of about 15% to 30%. Because correction factors could be estimated with large uncertainties only (due to the poor statistics), we neglected also this correction for the delay caused by this 4.5-ns isomer.

If significant intensity of the 203.7-keV and 720.1-keV lines is delayed by the 3290.6-keV level \((t_{1/2} = 2.6 \, \mu s)\) in \(^{205}\text{Tl}\), this would result in significant intensity below the effective thresholds of the relevant excitation functions. As this intensity is not observed, the measured cross sections for the 203.7-keV and 720.1-keV transitions can be considered a good approximation to the total \(\gamma\)-ray production cross sections for these lines.

### Table I: Nuclear reactions and gamma transitions investigated.

<table>
<thead>
<tr>
<th>Reaction investigated</th>
<th>(\gamma) Transition (Level energies in keV)</th>
<th>(\gamma) Energy</th>
<th>Isomers in (\gamma) cascade</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{208}\text{Pb}(n,p\gamma)^{207}\text{Tl})</td>
<td>1682.7 (\rightarrow) 351.0</td>
<td>1331.7</td>
<td>none</td>
</tr>
<tr>
<td>(^{208}\text{Pb}(n,p3\gamma)^{205}\text{Tl})</td>
<td>203.7 (\rightarrow) gs</td>
<td>203.7</td>
<td>(3/2^+) 203.7 1.46 ns</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(11/2^-) 1484.0 4.5 ns</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(25/2^+) 3290.6 2.6 (\mu s)</td>
</tr>
<tr>
<td>(^{208}\text{Pb}(n,p3\gamma)^{205}\text{Tl})</td>
<td>923.8 (\rightarrow) 203.7</td>
<td>720.1</td>
<td>(11/2^-) 1484.0 4.5 ns</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(25/2^+) 3290.6 2.6 (\mu s)</td>
</tr>
<tr>
<td>(^{208}\text{Pb}(n,p5\gamma)^{203}\text{Tl})</td>
<td>680.5 (\rightarrow) 279.2</td>
<td>401.3</td>
<td>none</td>
</tr>
<tr>
<td>(^{208}\text{Pb}(n,p7\gamma)^{201}\text{Tl})</td>
<td>331.2 (\rightarrow) gs</td>
<td>331.2(^a) (9/2(^-)) 919.5 2.035 ms</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(13/2(^-)) 2015.0 2.9 ns</td>
</tr>
<tr>
<td>(^{208}\text{Pb}(n,p7\gamma)^{201}\text{Tl})</td>
<td>1571.7 (\rightarrow) 1238.8</td>
<td>332.9(^a) (13/2(^-)) 2015.0 2.9 ns</td>
<td></td>
</tr>
</tbody>
</table>

\(^a\) The 331.2-keV and 332.9-keV \(\gamma\) lines in \(^{201}\text{Tl}\) were not resolved.

In the residual nucleus \(^{201}\text{Tl}\) there are two \(\gamma\)-ray transitions with 331.2 and 332.9 keV \(\gamma\)-ray energy. These two lines cannot be resolved by our experiment. The delay from the 2015.0-keV isomer can be neglected due to the short half-life of 2.9 ns. The half-life of the 919.5-keV level (2.035 ms) is long enough that more than 98% of the delayed intensity is emitted outside the time range (corresponding to the neutron energy range between reaction threshold and 200 MeV) which was actually analyzed. There is in addition also no significant intensity (within the uncertainty limits) observed in the neutron energy range below the threshold. Therefore corrections need not be performed and the measured cross section for the residual nucleus \(^{201}\text{Tl}\) is the sum of the \(\gamma\)-ray production cross sections for the 332.9-keV transition and the prompt part of the 331.2-keV transition.

To derive \(\gamma\)-ray production cross sections, a one-dimensional \(\gamma\) pulse-height spectrum was derived from the two-dimensional spectrum, neutron TOF versus \(\gamma\) pulse-height, for each neutron energy group. Fig. 2 shows an example of a spectrum for the neutron energy range 90 to 100 MeV. Four of the lines actually analyzed are marked in the figure. The number of counts in the \(\gamma\)-peak areas were obtained by adding the channel contents within the peak and subtracting a smooth (linear) background. As the choice of the peak limits and the
background region is somewhat subjective, an additional uncertainty component was added quadratically to the statistical uncertainties. An estimate of this uncertainty was obtained by comparing the peak areas determined by different summing limits and background regions. Corrections were applied for the attenuation of the $\gamma$ rays within the samples.

![Gamma-ray spectrum](image)

**Figure 2:** Part of the gamma-ray spectrum for the neutron energy group 90 - 100 MeV. The marked $\gamma$-lines were analyzed.

From the peak areas, the neutron fluence, and the $\gamma$ detector efficiency, relative excitation functions were derived for each $\gamma$ transition analyzed. The differential cross sections at $\theta = 125^\circ$ were converted to total $\gamma$-production cross sections by multiplying them by $4\pi$.

Because of uncertainty in our knowledge of the Ge detector dead time and the absolute flux intercepted by the irregular shaped samples, the results were normalized to data obtained in a separate 14-MeV experiment performed at the Institute of Physics of the Slovak Academy of Sciences. Normalization factors were derived from the cross sections of prominent transitions in $^{208}\text{Pb}(n,n'\gamma)$ and $^{208}\text{Pb}(n,2\gamma\gamma)$ reactions.

The total uncertainties were obtained by adding statistical and estimated systematic uncertainties in quadrature. Uncertainties in the range from 35% to 50% were estimated for $\gamma$-ray production cross sections of the 1331.7-keV and 401.3-keV transitions in $^{207}\text{Tl}$ and $^{203}\text{Tl}$, respectively. For the measured $\gamma$-ray production cross sections in the residual nuclei $^{205}\text{Tl}$ and $^{201}\text{Tl}$ the total uncertainties were in the range from 15% to 35%.
4. Results and Discussion

Extensive calculations of the γ-ray production cross sections in $^{208}\text{Pb}(n,x\gamma)$ reactions were performed by means of the code GNASH$^4$ in the course of the investigation of $^{208}\text{Pb}(n,x\gamma)$ reactions (see Ref. 2). 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. This assumption was modified 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. And finally, we have incorporated the excitation-energy dependence of the Ignatyuk level-density formula$^5$ 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 (see Ref. 2 for details). For the nuclear level densities, the Ignatyuk model was chosen instead of the simpler Gilbert-Cameron$^9$ and other Fermi-gas models. The energy-dependent level density parameter of the Ignatyuk model accounts for 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.

The same "best choice" parameter set, which gave good agreement with the experimental cross sections in $^{208}\text{Pb}(n,x\gamma)$ reactions leading to various lead nuclei was used in the calculations of γ-ray production cross sections in Tl nuclei from $^{208}\text{Pb}(n,p\gamma)$ reactions. The results for transitions in $^{207,203,201}\text{Tl}$ are given in Figs. 3 to 5. In Fig. 5, from the sum of the calculated γ-ray production cross sections for the 331.3-keV and 332.9-keV transitions, the production cross section for the 919.5-keV level ($t_{1/2} = 2.035$ ms) was subtracted to correct for the fact that only the promptly emitted 331.2-keV γ-rays are observed in the experiment.

There are rather large uncertainties, and also some approximations made regarding the effect of long-lived isomers on our experimental results. Because model calculations without special parameter adjustments can predict experimental results in general not better than within about 20% to 30% the experimental results are suitable quantities for comparison with calculations.

Other than in the case of reactions with neutron emission only (see Refs. 2 and 3), there is no agreement between experimental results and model calculations for the majority of the transitions analyzed. For the analyzed transitions in $^{207}\text{Tl}$ (Fig. 3) and $^{205}\text{Tl}$ (Figs. 4 and 5) the model calculation overestimates the measured cross section for neutron energies above about 70 MeV. The calculations result in cross sections about a factor of 3 to 4 higher than the experimental ones. There is rather good agreement for the 401.3-keV transition in $^{203}\text{Tl}$ and finally the calculated cross sections are smaller than the experimental results for γ-ray transitions in $^{201}\text{Tl}$ for high neutron energies. It seems that our calculation gives a too hard spectrum of the emitted protons. Proton emission is important for the preequilibrium stage of the reaction only, as proton emission from compound nucleus decay is strongly suppressed by the Coulomb barrier. An average energy of the emitted protons from the preequilibrium stage that is too high results in higher cross sections for reactions with the subsequent emission of only a few neutrons, and in lower cross sections for reactions with multiparticle emission as less energy is available after emission of the proton. As seen from Figs. 3 to 7 this effect is observed in the present study. It should be mentioned that the experimental information on
discrete levels and γ-ray branching in Tl nuclei is not as good as for Pb. As the information on discrete levels and γ-ray branching is essential for the calculation of γ-ray production cross sections for individual transitions, incomplete level scheme information might also contribute to some of the observed discrepancies.

Since neutron emission is such a dominant reaction channel the results of the previous investigations of $^{208}$Pb(n,xrry) reactions are virtually independent of the description of protons in the exit channel. The present study of $^{208}$Pb(n,pnγ) reactions gives us experimental information, which may help to improve the modeling of (n,pxn) reactions for lead.

Acknowledgments

This work was supported by the Fonds zur Förderung der wissenschaftlichen Forschung in Österreich (Project P 7908-TEC), and the U.S. Department of Energy under contracts W-7405-ENG-36 and W-7405-ENG-48.

Figure 3: $^{208}$Pb(n,pnγ)$^{207}$Tl cross section for the $5/2^+ (1682.7 \text{ keV}) \rightarrow 3/2^+ (351.0 \text{ keV})$ transition ($E_\gamma = 1331.7 \text{ keV}$) in $^{207}$Tl. Symbols: Present experiment. Solid line: GNASH calculation.
Figure 4: $^{208}$Pb($n$,p$3\gamma$)$^{205}$Tl cross section for the $3/2^+ (203.7 \text{ keV}) \rightarrow 0^+$ transition ($E_\gamma = 203.7 \text{ keV}$) in $^{203}$Tl. Symbols: Present experiment. Solid line: GNASH calculation.

Figure 5: $^{208}$Pb($n$,p$3\gamma$)$^{205}$Tl cross section for the $7/2^+ (923.8 \text{ keV}) \rightarrow 3/2^+ (203.7 \text{ keV})$ transition ($E_\gamma = 720.1 \text{ keV}$) in $^{203}$Tl. Symbols: Present experiment. Solid line: GNASH calculation.
Figure 6: $^{208}\text{Pb}(n,p5\gamma)^{203}\text{Tl}$ cross section for the $5/2^+ (680.5 \text{ keV}) \rightarrow 3/2^+ (279.2 \text{ keV})$ transition ($E_\gamma = 401.3 \text{ keV}$) in $^{203}\text{Tl}$. Symbols: Present experiment. Solid line: GNASH calculation.

Figure 7: $^{208}\text{Pb}(n,p7\gamma)^{201}\text{Tl}$ cross section. Sum of the cross sections for the $13/2^- (1571.7 \text{ keV}) \rightarrow 3/2^+ (1238.8 \text{ keV}) 3/2^+$ transition ($E_\gamma = 332.9 \text{ keV}$) and for the prompt part of the (331.2 keV) $\rightarrow$ gs transition ($E_\gamma = 331.2 \text{ keV}$) in $^{201}\text{Tl}$. Symbols: Present experiment. Solid line: GNASH calculation.
References


