Photonuclear reactions for detection of nuclear materials and nonproliferation

Mark B. Chadwick [a,1] and Marie-Laure Giacri [b]

[a] *T-16, Nuclear Theory Group, Los Alamos National Laboratory, Los Alamos, NM 87545, USA*

[b] *SPhN, CEA Saclay, F-91191 Gif Yvette, France*

Abstract

New techniques are being developed to detect the presence of fissile materials using active interrogation, with photons made from electrons on a bremsstrahlung conversion target. We describe work at Los Alamos to develop a radiation transport simulation code to model such processes, to help guide the technology development and to help interpret measurements. Nuclear physics plays a significant role in this project, since nuclear cross sections and decay properties are needed by the simulation code. We describe nuclear theory and models to help us predict the photonuclear and photofission cross sections, including the emission spectra of the neutrons following photoabsorption. Delayed neutrons from the beta-decay of the fission products are evaluated based on models of the fission yields, the half lives, and the delayed-neutron fractions.

1 Introduction

It is well known that certain special nuclear materials (SNM) such as ^{235}U are difficult to detect because of the small quantities of radiation they emit (neutron and photon leakage). This presents a serious problem in nonproliferation efforts, for preventing the spread of nuclear materials, and has led to research programs at many laboratories to develop "active interrogation" methods, to detect SNM using technologies that can be deployed at customs and border points. These technologies involve an external probe of radiation that impinges on a container, and subsequent emitted radiations are detected

[1] E-mail: mbchadwick@lanl.gov

Preprint submitted to Elsevier Science *24 June 2003*

and used to characterize the material present. Both incident neutron and incident gamma-ray probes are being studied; this paper focuses on gamma-ray probes from electron bremsstrahlung.

At Los Alamos we are developing a capability to detect SNM using bremsstrahlung gamma-rays as the active interrogation probe. A compact betatron produces 8 MeV electrons, which impinge on a bremsstrahlung conversion target to produce photons with energies up to 8 MeV. A fraction of these photons have energies above the photofission threshold for actinides (typically about 5 MeV). When photofission occurs, both prompt fission neutrons and delayed fission neutrons are produced, and are multiplied by the fissile materials. The key signature being investigated is the emission of delayed neutrons following the beta-decay of the fission products, since this is a unique signal indicating the presence of SNM.

New simulation capabilities are needed to help develop this technology. This is important to help experimentalists design and optimize the accelerator, conversion target, and neutron detectors. Simulations also provide valuable information on the photon transport through various container configurations, and the spatial distribution of fission events that produce delayed neutrons. Simulations are also important for interpreting measurements. In order for the Los Alamos MCNP transport code to be used for such simulations, it has had to be extended in certain ways [1]; furthermore, nuclear physics research to produce cross section databases of photonuclear reactions is needed. These physics developments are described below.

2 Photonuclear reactions

Photonuclear cross sections for absorption, neutron emission, and fission, have been measured in the Giant Resonance regime at many laboratories. Work at Livermore, Illinois, Saclay, [2] and Russia, has been particularly important [3]. For these data to be useful in simulation codes, nuclear data evaluation work and nuclear modeling is required. Such work helps resolve inconsistencies in measured cross sections from different labs. Very importantly, nuclear modeling is required for providing predictions on the emitted neutron energy and angular spectra, which is needed for transport simulations using complete sets of evaluated cross sections. Such spectra have rarely been measured for monoenergetic photon sources. Additionally, the importance of delayed neutrons in this application has stimulated calculations of the delayed neutron production, time-dependence, and energy spectra, using our CINDER burn-up code together with information on the fission product yield distributions, Pn values and beta-decay half lives.

An International Atomic Energy Agency collaboration project (Coordinated Research Program) focused on developing a set of evaluated ENDF photonuclear cross sections, for use in transport simulations [4]. In this project we evaluated and modeled data for over 160 isotopes, and this has been an important resource for the present project. Los Alamos' contribution to this work was the evaluation of some high priority materials in accelerator applications and in dosimetry: isotopes of C, O, Al, Ca, Fe, Cu, Ta, W, Pb [5]. Also, the Los Alamos GNASH code was the basis of the South Korean evaluations, which provided the major contribution to this library. The actinides were evaluated by the Obninsk group, though they did not include delayed-neutron emission information.

This paper focuses on recent work to evaluate and model nuclear data on important actinides: ^{235,238}U, ^{239}Pu, ^{237}Np. Whilst the photoneutron and photofission cross sections in the Obninsk evaluations appear to be of good quality, we are re-evaluating these data so as to take advantage of our ability to predict accurate prompt neutron spectra that are consistently determined with the cross sections. This includes both prompt neutron spectra from $(\gamma, 1n)$ and $(\gamma, 2n)$ processes, as well as prompt fission neutron spectra calculated with the Madland-Nix model. We are also evaluating delayed neutron data for these nuclides using two approaches: (1) A longer-term project is to evaluate delayed neutron data, with the help of the CINDER code, using "microscopic" summation calculations using photonuclear fission yield, Pn, and half-life data; (2) A short-term, but approximate, approach is to adapt photon projectile information from existing equivalent neutron information as a surrogate, i.e. use neutron data on the A-1 target at an incident energy shifted by the neutron separation energy. This is approximate because even though the surrogate neutron reaction forms the same compound nucleus at the same energy, the spin-distribution is different, and this may lead to different yield distributions, which will impact the delayed neutron production.

3 GNASH nuclear reaction modeling

A model of photonuclear reactions must account for a number of different nuclear reaction mechanisms involved in the initial photonuclear excitation process, and the subsequent decay of the excited nucleus by particle emission. At low energies, below about 30 MeV, the Giant-Dipole Resonance (GDR) is the dominant excitation mechanism, where a collective bulk oscillation of the neutrons against the protons occurs. At higher energies, where the wavelength of the photon decreases, photoabsorption on a neutron-proton (quasi-deuteron (QD)) becomes important. In these processes, the initial nuclear excitation can be described by particle-hole excitations ($1p1h$ for the GDR; $2p2h$ or $2p1h$, as we discuss later, for QD processes) and thus it is natural to use a preequilibrium theory to describe the processes of preequilibrium emission, and damping to equilibrium, during the evolution of the reaction. The present work can be used to calculate photonuclear reactions for incident photons with energies below 140 MeV, which is the threshold for pion production.

3.1 The Photoabsorption Model

If experimental data exists for the total photoabsorption cross section, it can be used in GNASH calculations via an input file. The most useful type of experimental data here are from photon absorption experiments which really measure the total photoabsorption cross section. For heavy nuclei, compilations of photoneutron total cross section such as that of Dietrich and Berman [2] can be used to approximate the photoabsorption cross sections, since contributions from photoproton reactions (and other reactions producing complex charged particles) are suppressed by the Coulomb barrier. However, in light nuclei this approach cannot be used since the photoproton cross section is no longer small, and in some cases exceeds the photoneutron cross section. In such cases one should rely only on absorption-type data.

3.2 Preequilibrium and compound nucleus emission

Particle and hole excitations are produced in the nucleus following photoabsorption. Particle emission can occur from such a state yielding the typically high-energy preequilibrium emission; or alternatively, a nucleon-nucleon interaction may occur producing more particle-hole excitations. The composite nuclear system passes through such stages of increasing complexity towards equilibrium, and the total preequilibrium emission is the sum of contributions from all the preequilibrium stages.

Wu and Change [6] and Blann [7] have successfully applied preequilibrium physics modeling to describe the emission of high-energy nucleons following photoabsorption. Wu and Chang used an initial $2p2h$ state in the preequilibrium cascade, while Blann argued that the two holes are correlated through the QD mechanism, and therefore can be approximately treated as one degree of freedom, i.e. a $2p1h$ initial state. We follow Blann's prescription, and use the Kalbach exciton model in GNASH [8] to calculate the preequilibrium emission of fast nucleons. Once the incident photon energy exceeds about 50 MeV, multiple preequilibrium emission (MPE) becomes important, where more than one fast particle is emitted. For this we use the generalized MPE model of Ref. [9]. For both primary and multiple preequilibrium emission, we modified the neutron-proton distinguishability factors to reflect the initial particle-hole type appropriate for the QD mechanism (i.e. of the two particles, one is a neutron and the other a proton).

After the preequilibrium phase of the reaction, the excited residual nucleus is allowed to decay by (sequential) Hauser-Feshbach compound nucleus emission.

3.3 Photofission

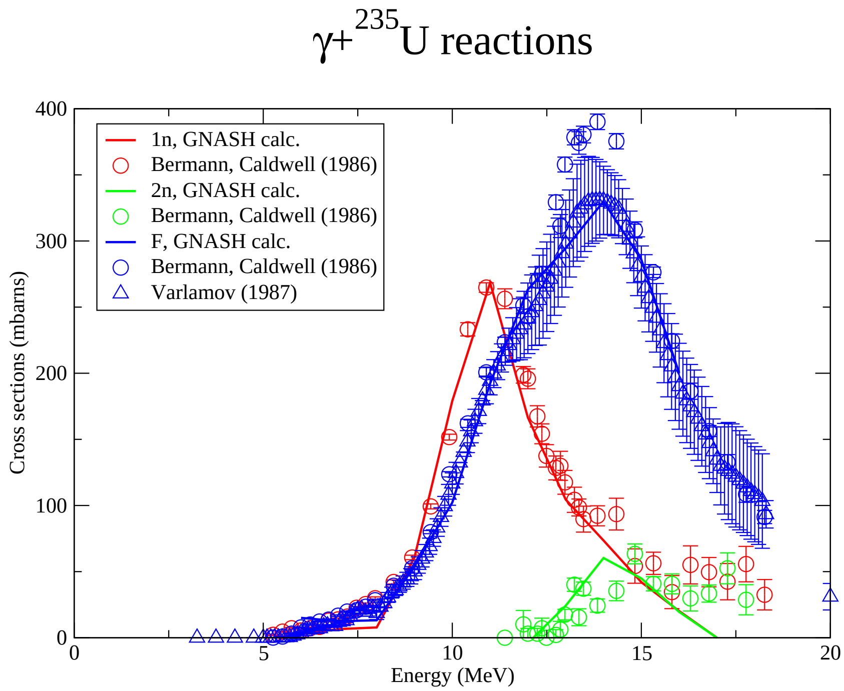

Fig. 1. Calculated and measured photonuclear and photofission cross sections for ^{235}U. In the case of photofission, our GNASH calculations agree reasonably well with Valamov's evaluated data.

We have recently adapted our GNASH nuclear reaction code to model photofission reactions. The extensions required were minimal: we had already developed a (non-actinide) photonuclear modeling capability (see previous subsection), and we have extensive experience in using GNASH to model *neutron*-induced fission reactions [10]. Our earlier neutron-induced analyses for isotopes of uranium, plutonium, and neptunium, had led to a set of input parameters describing fission barriers within a double-humped parabolic model, fission transition states and level densities, transmission coefficients, etc., which could be directly used in the present photo-fission studies. Using these parameters led to accurate predictions on the measured $(\gamma, 1n)$, $(\gamma, 2n)$, and (γ, F) data. In some cases we made small adjustments to the fission barrier parameters to further optimize agreement between calculation and experiment, but the adjustments needed were modest. This builds confidence in the overall nuclear reaction mechanism modeling we apply.

4 Results and Discussion

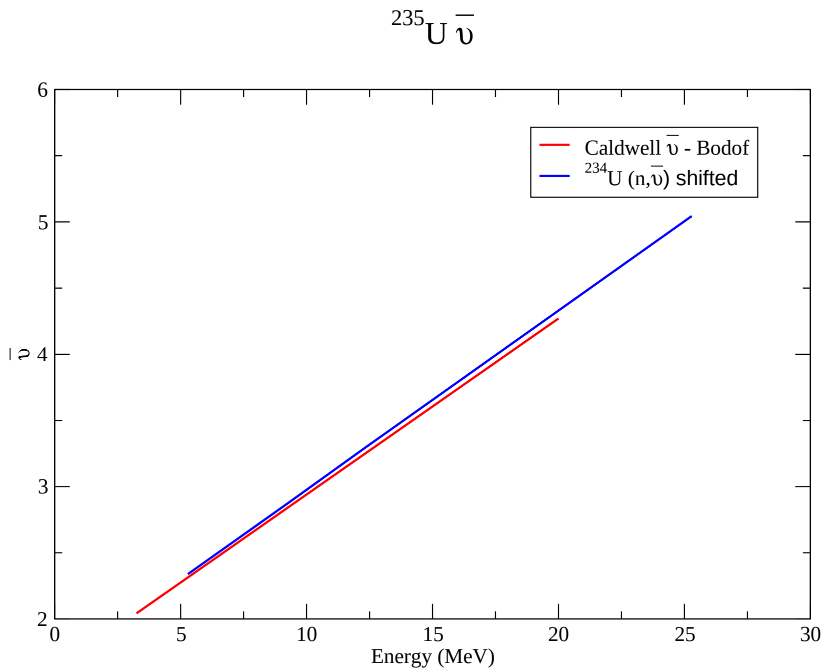

Fig. 2. Measured prompt fission neutron multiplicity for photons on ^{235}U, compared with equivalent data from the neutron reaction on ^{234}U.

Figure 1. shows our results for photonuclear and photofission cross sections for ^{235}U. The measured data for the fission cross section are somewhat discrepant; our calculation is in good agreement with the evaluated data from Varlamov (which aimed to reconcile many sets of measured data). The good agreement seen between theory and experiment provides some validation for the nuclear reaction mechanisms implemented in GNASH, i.e. the competition between preequilibrium, equilibrium, and fission decay processes. Furthermore, our ability to model these cross sections accurately provides some indirect validation of the emission spectra predicted by the code, which are needed for the transport applications.

An important ingredient in an evaluated nuclear cross section database is the evaluation of nu-bar, the average number of prompt neutrons emitted in fission. Measurements of nu-bar for ^{235}U have been made at Livermore by Caldwell, and are shown in Fig. 2. These data are compared with equivalent (surrogate) neutron data for ^{234}U, shifted by the neutron separation energy.

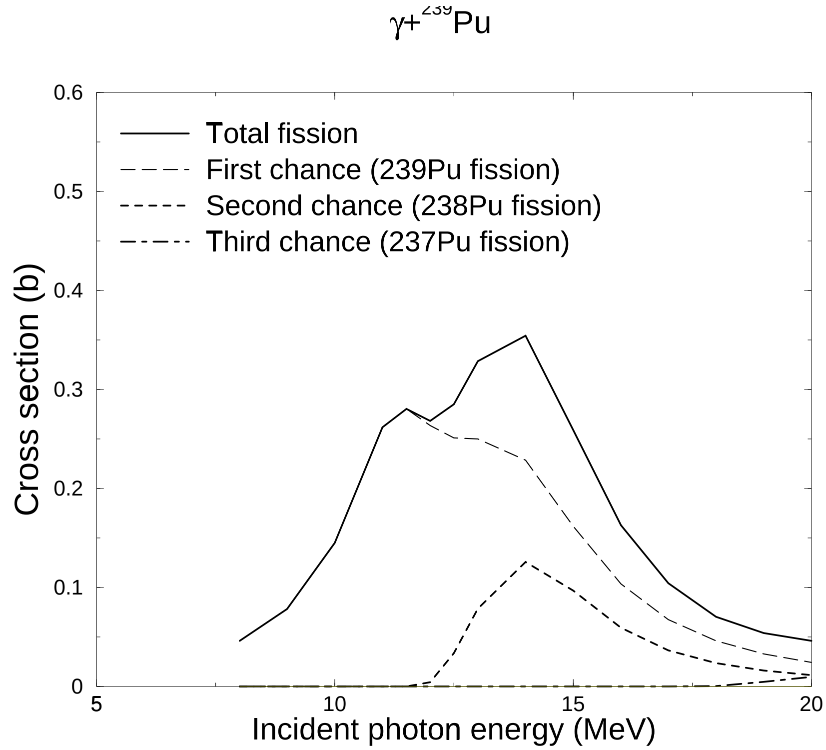

Fig. 3. Contributions of first, second, and third-chance fission to the total fission cross section for photons on ^{239}Pu, as calculated by GNASH.

The close agreement between the directly measured data, and from the surrogate shifted ^{234}U neutron data, is to be expected. We see such agreement in this case, but here are other cases we are studying that show discrepancies, and we are trying to understand this.

As the nuclear reaction proceeds, the compound nucleus can fission immediately (first-chance), or after the emission of a neutron (second-chance), or after the emission of two neutrons (third chance), *etc.* The contributions, as predicted by our GNASH calculation, for plutonium are illustrated in Figure 3. Thus, in order to model accurately the $(\gamma, 1n)$, $(\gamma, 2n)$ cross section accurately, one needs to accurately model the (multi) chance contributions to the total fission cross section as well as the total fission cross section data. We use fission barriers from numerous sources of information, including neutron fission measurements and direct reaction fission probability measurements [11].

5 Conclusion

We have developed our nuclear reaction modeling capabilities to include photofission reactions on actinides. This allows us to build cross section ENDF databases for the cross sections, spectra, as well for prompt and delayed fission neutron emission. Together with advances in our MCNP and CINDER simulation codes, this work provides the necessary tools to be able to simulate photofission detection of special nuclear materials.

We gratefully acknowledge useful discussions with William Wilson, Robert MacFarlane, Peter Moller, Pavel Oblozinsky, Robert Little, Morgan White, Marshall Blann, and Holly Trellue. We thank Bill Johnson for useful discussions and for support of this work.

References

[1] M. C. White *et al.*, Nucl. Sci. Eng. **144**, 174 (2003).

[2] S. S. Dietrich and B. L. Berman, Atomic Data and Nuclear Data Tables **38**, 199 (1988).

[3] A. V. Varlamov, V. V. Varlamov, D. S. Rudenko, and M. E. Stepanov, Technical Report No. INDC (NDS)-394 (1999), International Atomic Energy Agency, Vienna, Austria.

[4] IAEA, Technical Report No. IAEA-TECDOC-1178, International Atomic Energy Agency, http://iaeand.iaea.or.at/photonuclear/, Vienna, Austria.

[5] M. B. Chadwick, P. G. Young, R. E. MacFarlane, M. C. White, and R. C. Little, Nucl. Sci. Eng. **144**, 157 (2003).

[6] J. R. Wu and C. C. Chang, Phys. Rev. C **16**, 1812 (1977).

[7] M. Blann, B. L. Berman, and T. T. Komoto, Phys. Rev. C **28**, 2286 (1983).

[8] P. G. Young, E. D. Arthur, and M. B. Chadwick, in *Proc. of the IAEA Workshop on Nuclear Reaction Data and Nuclear Reactors - Physics, Design, and Safety*, Trieste, Italy, April 15 - May 17, 1996, edited by A. Gandini and G. Reffo (World Scientific Publishing, Ltd., Singapore, 1998), pp. 227–404.

[9] M. B. Chadwick, P. G. Young, D. C. George, and Y. Watanabe, Phys. Rev. C **50**, 996 (1994).

[10] P. G. Young and E. D. Arthur, in *Proceedings Nuclear Data for Science and Technology*, Jülich, Germany, May 13-17, 1991, edited by S. Qaim (Springer-Verlag, New York, NY, 1992), p. ???

[11] A. Gavron, H. C. Britt, E. Konecny, J. Weber, and J. B. Wilhelmy, Phys. Rev. C **13**, 2374 (1976).