Studying photonuclear reactions using the activation technique

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1. Introduction

Nuclear reactions triggered by photons, or photonuclear reactions, are an important subject of studies in nuclear physics. Experimental studies of photonuclear reactions have started in 1950s but some major questions still remain open:

- Although numerous systematic measurements have already been made, the existing datasets are still incomplete. In the June 2013 version of the EXFOR\textsuperscript{[1]} database there are as many as 1350 reaction entries for the $(\gamma,n)$ reaction, 313 entries for the $(\gamma,2n)$ reactions, 64 for $(\gamma,3n)$, and only 14 for $(\gamma,4n)$. There are 789 entries for the $(\gamma,p)$ reaction and just 7 for the $(\gamma,2p)$ reaction. Another experimental database, the IAEA Photonuclear Data Library\textsuperscript{[2]} contains 146 evaluated total photoproduction cross-sections, 21 entries for $(\gamma,2n)$ reactions, 12 entries for $(\gamma,p)$ reactions, and 8 for $(\gamma,2p)$ reactions (the IAEA library omits duplicated and redundant datasets, hence the large difference in numbers). A number of practical applications of photonuclear interactions depend on availability of as full datasets as possible, yet properties of the reactions are measured only for a limited range of nuclei. It can be seen that very little data is available on reactions with high multiplicities.
- A widely recognized problem of existing data is the problem of data validation that needs to be performed for at least some results of past large-scale programs to measure photonuclear cross-sections. There are known systematic discrepancies in existing measurements and independent experiments are needed to resolve the discrepancies and to elaborate reliable data evaluations\textsuperscript{[3–5]}. The purely electromagnetic character of the interaction between photons and nuclei also means that accurate experimental data are required for refinement and further development of theoretical models and computation packages\textsuperscript{[6,7]}. Photodisintegrations play a fundamental role in actively studied phenomena like the astrophysical p-process. There are 32 so-called p-nuclei which depend on photonuclear reactions during stellar nucleosynthesis. Current theoretical calculations cannot accurately predict their abundances\textsuperscript{[8]}. Experimental laboratory studies of photonuclear reactions are required to refine the calculations.
- Finally, applications, like transport security scanners\textsuperscript{[9]}, photon activation analysis (refer to bibliography in Ref.\textsuperscript{[10]}), production of radioisotopes, and so on require high quality experimental and evaluated data for specific photonuclear reactions that they depend on.

There is a strong need for systematic measurement of photonuclear reactions on different nuclei. To perform these measurements and to obtain accurate and reliable data a specialized system specifically tailored for this kind of systematic measurements is needed. This paper describes the setup built for this purpose at the Skobeltsyn Institute of Nuclear Physics of the Moscow State University (SINP MSU).
2. Experimental technique

There are established techniques of experimental measurements of photonuclear reaction properties which rely on different types of sources of incident photons and different ways of detection of occurring reactions. The measurements described in this work are based on the activation technique, that is, the experiments are split into two stages: the irradiation stage and the residual activity measurement stage.

Since the cross-sections of photonuclear reactions are typically of the order of 0.1 b or smaller, a high intensity photon source is required to collect reasonable statistics. We use bremsstrahlung radiation as the source of photons. The general view of the bremsstrahlung production setup is shown in Fig. 1. Depending on the required energy either the 67.7 MeV racetrack microtron [11] or the 55 MeV racetrack microtron [12] is used, both of which are modern compact electron accelerators developed and operated by a collaboration at the SINP MSU. The main parameters of the accelerators are listed in Table 1.

The accelerators are installed in a radiation-shielded room within the experimental hall. A modular digital control system is used to operate the accelerators. Variations of the beam current are measured with a calibrated ionization chamber in the beamline and a Faraday cup and are recorded in a web-accessible database for use during analysis via an analog-to-digital converter card and a LabView program. An example of recorded beam current is shown in Fig. 2.

To produce gamma radiation a radiator target made of tungsten or lead is used. Tungsten is a common convertor material and using lead as a bremsstrahlung production material has the advantage that the target thickness can be easily adjusted. We use bremsstrahlung radiation to maximize the number of photons in the energy range of the giant dipole resonance which dominates the photonuclear cross-section from resonance to 20–30 MeV. An optional aluminum absorber can be placed behind the radiator to remove the remaining electrons from the bremsstrahlung beam. To simulate the angular and energy distribution of the bremsstrahlung radiation produced in a thick radiator, an absorber, and the studied target itself a GEANT4 [13] model of the irradiation geometry from Fig. 1 is used later during analysis of results. Electric charge collected on the target is also digitized and used to measure the beam current in addition to the ionization chamber and the Faraday cup. No specialized cooling systems are installed as even long (several hours) irradiations do not result in significant overheating of radiators or targets.

The generated bremsstrahlung irradiates targets made of the isotopes to be measured. The radiator, the ionization chamber, the electron absorber and the target are mounted in the beamline in a special rigid frame in order to ensure reproducible geometry in all experiments. A web camera is used to monitor position of the beam with the help of a fluorescent screen and to correct placement of targets.

The duration of irradiation is typically 0.5–10 h and depends on the cross-section of the reaction to be studied and on the properties of the reaction products. We use the so-called activation technique to determine the rate of photonuclear reactions in the target. This technique relies on the presence of relatively long-lived unstable nuclei with identifiable gamma lines in their decay spectra among the products of the reaction. As soon as the irradiation ends and radiation levels in the experimental hall become safe the target is transported to a high-purity germanium spectrometer (HPGe) which measures photon spectra of residual activity. We use the Canberra GC 3019 HPGe detector with 30% relative efficiency and an energy resolution of 0.9 keV at 122 keV and 1.8 keV at 1.332 MeV. The detector enclosed in a radiation shielding chamber is located in a dedicated room, which allows us to decrease the natural background by ≈3 orders of magnitude. A procedure of routine energy and efficiency calibrations for the detector is established. To obtain the detector efficiency in different geometries and to calculate correction coefficients for coincidence summing of photons a GEANT4 model of the detector cryostat and the measurement chamber is used. The Livermore electromagnetic package option is used in the model as it leads to the most precise reproduction of the detector response function. The model is matched against the real detector and its environment using the standard sources $^{137}$Cs, $^{60}$Co, $^{152}$Eu, $^{133}$Ba, $^{241}$Am.

![Fig. 1. Schematic view of the setup that is used to irradiate targets with bremsstrahlung radiation.](image)

![Fig. 2. Time dependence of the beam current during a 30 min irradiation measured using the ionization chamber.](image)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>67.7 MeV microtron</th>
<th>55 MeV microtron</th>
</tr>
</thead>
<tbody>
<tr>
<td>Output energy</td>
<td>14.8–67.7 MeV</td>
<td>55.5 MeV</td>
</tr>
<tr>
<td>Beam energy spread</td>
<td>0.1 MeV at 67.7 MeV (from beam dynamics simulation)</td>
<td>1.1 MeV at 10 MeV (measured at 2nd orbit)</td>
</tr>
<tr>
<td>Output current at max. energy</td>
<td>5 mA</td>
<td>10 mA</td>
</tr>
<tr>
<td>Pulse duration</td>
<td>5–40 μs</td>
<td>5 μs</td>
</tr>
<tr>
<td>Orbits</td>
<td>14</td>
<td>11</td>
</tr>
<tr>
<td>Energy gain</td>
<td>4.8 MeV/orbit</td>
<td>5 MeV/orbit</td>
</tr>
<tr>
<td>Max duty factor</td>
<td>0.4%</td>
<td>0.025%</td>
</tr>
<tr>
<td>RTM dimensions</td>
<td>2.2 × 1.8 × 0.9 m</td>
<td>2.7 × 1.7 × 0.8 m</td>
</tr>
</tbody>
</table>
calculate true coincidence summing corrections we use the TrueCoinc software [14] with total and partial absorption efficiencies calculated using the model.

Clocks on the data acquisition and analysis PCs are synchronized via the NTP protocol with a locally run NTP network time server up to several ms accuracy. As an example Fig. 3 shows two spectra of residual activity measured after an irradiation of a palladium target.

The specialized computer system which is used to automatically acquire, store and analyze the induced activity spectra is described in the next section.

3. Automatic data acquisition and analysis system

The activation technique relies on detection of residual radioactivity of unstable nuclei produced in the reactions during the irradiation phase. By measuring the residual activity with a high resolution spectrometer one obtains characteristic gamma ray spectra of decaying unstable reaction products. Specific reactions can be identified by energies and relative intensities of spectral lines of their products and the reaction rates can be calculated from the observed intensities of lines. Although it is possible to use this technique by manually performing a small number of spectrum measurements, a more reliable and comprehensive analysis is available if a large series of spectra is measured automatically. Not only is the analysis more reliable in this case but also reaction rates can be calculated even when the products form complex decay chains. From the statistical point of view it is desirable to measure as many spectra in a series as possible to get the best estimates of reaction yields.

We use a special computer system which allows us to measure several thousands of gamma ray spectra after each irradiation, process the measured data, and automatically generate decay curves of produced nuclei.

The system is described in detail in Ref. [15]. It is based on a networked PostgreSQL database, which stores the acquired spectra and various meta-information: acquisition time and duration of measurement, detector dead time, properties of the sample, detector calibration coefficients, irradiation conditions, and so on. A custom-developed driver is used to read spectra from the Canberra InSpector 1250 multi-channel analyzer over RS232 link every 3 s, whereas the vendor-supplied tools only allow an interval of 7 s or more. Thus a nearly continuous detection of gamma radiation is achieved. Typical duration of measurement of the induced activity is from several days to several weeks, which results in tens of thousands of spectra after each irradiation. The measured spectra are also stored in the database in a binary format for performance reasons.

To process a large number of spectra measured after each irradiation and to simplify the processing workflow the system features a web-based analysis interface. The user can browse available measurements, request series of spectra measured during a certain period of time, perform summation of 3-second spectra over a longer period, view the requested series of spectra or sums of spectra as a series of graphics or in a text form. The generated series of spectra or sums of spectra can then be simultaneously analyzed with a peak fitting program also within the web interface. The program is based on the Minuit2 [16] minimization package and provides both automatic and user-controlled modes of operation. The automatic peak search is performed in three stages. At the first stage the spectrum is separated into two parts, corresponding to the continuum regions and peaks. At the second stage positions of the peaks are refined using information from other spectra in the current analysis and a pattern matching algorithm to split multiple overlapping peaks. At the third stage the peaks and background are fitted with a flexible fitting function, for which a user can select options of different peak shapes and background approximations. Analysis of a 10-spectrum dataset takes several seconds and the parameters can be adjusted afterwards to re-run the program if needed. The quality of automatic peak search depends strongly on the measurement conditions, since one has to deal with count rates from 0.1 to 100,000 s−1, but on the average the percentage of good peak search in automatic mode is about 90–95%. In the generated report page the program displays each spectrum with fitted peaks, a table of peak parameters (energy, area, FWHM, background, fit quality, errors, and so on) for each spectrum, and a summary table containing energy and area of each peak in all processed spectra versus measurement time. Each entry of the summary table can be displayed in a graphical form as a decay curve (normalized by the measurement time). Also at this stage automatic fit of the decay curve with exponential function is performed, and estimates of activity and half-life are shown to the user.

4. Analysis of decay chains

At energies higher than thresholds of the most simple reactions with one outgoing nucleon an increasingly large number of other reaction channels open. Products of the reactions are frequently connected with each other through complex sequences of mutual decays. The number of different nuclei in these decay chains is especially large if the studied target consists of multiple isotopes. Due to the decay chains decay curves may become more complex than a simple exponential function, and a more thorough analysis is needed to restore the reaction yields.

The analysis system of the SINP MSU includes a program for generating decay chains based on the ENSDF database [17] and a special method is used to obtain statistically reliable reaction yields from analysis of complex decay curves.

The decay chain generation program also runs within a web interface. The user enters the desired isotopic composition of the target and the electron beam energy and the program builds a list of nuclei that can be produced by photonuclear reactions. Decay
data from the spectroscopy database ENSDF is used then to connect the produced nuclei with decay relationships. Branching ratios of decays are also calculated at this point and excited isomeric states are taken into account for both decaying and produced nuclei. The decay chains can be shown either graphically or in a form of text table containing the corresponding branching ratios or probabilities of particular decays in the decay chain. In addition there is a simulation mode where photonuclear cross-sections are calculated using the [6] model and folded with the GEANT4 bremsstrahlung spectrum yielding theoretical reaction yields. Then a Java applet calculates accumulation and decay of each element of a corresponding decay chain and the result is shown in a graphical form.

Branching ratios calculated by the program are used as an input for the automatic analysis procedure described in Ref. [18]. The system of ordinary differential equations that describes time evolution of the numbers of different nuclei in the target

$$\frac{dn_i}{dt} = y_i I_\text{t}(t) - \lambda_i n_i + \sum_j k_{ij} n_j$$  \hspace{1cm} (1)

(where $n_i$ is the number of nuclei of type $i$ in the target, $y_i$ are relative reaction yields, $I_\text{t}(t)$ is the recorded accelerator current ($I_\text{t}(t) = 0$ when $t > T_{\text{rad}}$, where $T_{\text{rad}}$ is the irradiation time), $\lambda_i$ are decay constants, and $k_{ij}$ is the branching ratio of the $j \to i$ decay) is constructed and solved, yielding a linear statistical model

$$Ay = x + \epsilon$$

for every vector $x$ composed of measured areas of a certain peak at different moments of time (that is, decay curve) normalized by its intensity and detector efficiency, where the unknown parameter vector $y$ represents reaction yields, $\epsilon$ is the random error vector, and the design matrix $A$ is calculated numerically from the ODE system according to the relationship

$$A_{cd} = \int_{t_{d1}}^{t_{d2}} \lambda(t) \, dt, \quad c = 1, \ldots, M, \quad d = 1, \ldots, N, \quad y_k = \begin{cases} 0, & k \neq M \\ 1, & k = M \end{cases}$$  \hspace{1cm} (2)

where $N$ is the number of measurements in $x$, and $t_{d1}$ and $t_{d2}$ are the times when the $d$th measurement started and stopped, and $M$ is the number of decay chain elements. The linear system is solved using a standard generalized least squares method and reaction yields are obtained:

$$y^o = (A^T \Omega^{-1} A)^{-1} A^T \Omega^{-1} x$$  \hspace{1cm} (3)

where the specific form of the covariance matrix $\Omega$ depends on the method of solution: either feasible least squares or iteratively reweighted least squares method is used.

It should be emphasized that at no stage of the analysis procedure one has to write down explicitly rather complex differential equations and their solutions and also that usage of potentially unstable iterative nonlinear curve fitting algorithms is avoided.

5. Examples of measurements performed using the described system

5.1. Photonuclear reactions on $^{89}$Y

Fig. 4 shows the decay chain formed by $^{87}$Y and $^{87}$Sr and their corresponding isomer states produced in a $^{89}$Y target by $(\gamma,2n)$ and $(\gamma,np)$ reactions. Half-lives and branching ratios extracted from ENSDF are also shown. It is clear from the scheme that the activity of the $^{87}$Sr isomeric transition decay depends on decays of $^{87}g.s.-^{126}$Y. Fig. 5 shows the time dependence of the count rate of the 388 keV peak from $^{87m}$Sr decay measured after 3 h of activation of yttrium target, which differs significantly from the usual exponential shape (bremsstrahlung end-point energy was 55 MeV, produced in a 2.3-mm thick tungsten convertor). The decay chain analysis program automatically builds and solves the corresponding system of ODEs producing the linear statistical model matrix, and the latter is used to obtain estimations of relative yields: $y^{89Y}(\gamma,2n)^{87m}$Sr] = 0.3657(5), $y^{89Y}(\gamma,np)^{87m}$Sr] = 0.6343(5), $y^{89Y}(\gamma,2n)^{87m}$Sr] = 0.07498 (7).

5.2. Photoneutron reactions on $^{209}$Bi

Simultaneous measurement of different reaction channels allows us to increase accuracy and to obtain yields differing up to 5–6 orders of magnitude. As an example, Table 2 shows relative yields of photoneutron reactions on $^{209}$Bi induced by bremsstrahlung photons with end-point energy 67.7 MeV [19]. Reactions with up to 7 outgoing neutrons were observed and their yields were obtained simultaneously.

6. Conclusions

The system for experimental studies of photonuclear reactions that has been used to measure yields and cross-sections of photonuclear reactions on a number of nuclei, including isotopes of hafnium [19], tin [20], palladium [21], uranium [22], and others is described. The usage of modern electron accelerators, HPGe detector with low-background shielding, and web-based automatic analysis software make it possible to use non-monoisotopic
targets and to reconstruct independent yields of multiple reactions, whose products form complex decay chains. Extremely rare reactions up to $^\gamma\text{Bi}(\gamma,7n)$ and $^\gamma\text{Bi}(\gamma,3\alpha)$ and production of nuclei far from the beta stability region can be studied in a routine fashion. The activation technique is free from multiplicity sorting problems of conventional neutron detectors and, therefore, yields of several multiparticle reactions can be measured simultaneously and normalized to a common level.

### References


Table 2

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Rel. yield</th>
<th>$T_{1/2}$ of product</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{209}\text{Bi}(\gamma,2n)^{207}\text{Bi}$</td>
<td>1.00(5)</td>
<td>31.55 years</td>
</tr>
<tr>
<td>$^{209}\text{Bi}(\gamma,3n)^{206}\text{Bi}$</td>
<td>0.15(3)</td>
<td>6.243 days</td>
</tr>
<tr>
<td>$^{209}\text{Bi}(\gamma,4n)^{205}\text{Bi}$</td>
<td>0.09(2)</td>
<td>15.31 days</td>
</tr>
<tr>
<td>$^{209}\text{Bi}(\gamma,5n)^{204}\text{Bi}$</td>
<td>0.017(3)</td>
<td>11.22 h</td>
</tr>
<tr>
<td>$^{209}\text{Bi}(\gamma,6n)^{203}\text{Bi}$</td>
<td>0.007(2)</td>
<td>11.76 h</td>
</tr>
<tr>
<td>$^{209}\text{Bi}(\gamma,7n)^{202}\text{Bi}$</td>
<td>0.00012(6)</td>
<td>1.71 h</td>
</tr>
</tbody>
</table>