EPJ Web of Conferences **69**, 00005 (2014)
DOI: 10.1051/epjconf/20146900005

Development of an experimental set-up for the measurement of neutron-induced fission and capture cross sections of highly radioactive fissile nuclei

Iulia Companis[1], Ludovic Mathieu[1a], Mourad Aïche[1], Peter Schillebeeckx[2], Jan Heyse[2], Gérard Barreau[1], Serge Czajkowski[1], Quentin Ducasse[1], Frank Gunsing[3], Beatriz Jurado[1], Gregoire Kessedjian[4], Julie Matarranz[1] and Igor Tsekhanovich[1]

[1]Centre d'Etudes Nucléaires Bordeaux Gradignan, CNRS/IN2P3, Univ. Bordeaux 1, Chemin du Solarium, F-33175 GRADIGNAN, France
[2]EC-JRC, Institute for Reference Materials and Measurements (IRMM), Retieseweg 111, B-2440 Geel, Belgium
[3]CEA/Saclay - DSM/DAPNIA/SPhN, F-91191 Gif-sur-Yvette, France
[4]Laboratoire de Physique Subatomique et de Cosmologie, CNRS/IN2P3, Univ. Joseph Fourier, INPG, 53 avenue des Martyrs, F - 38026 Grenoble Cedex, France

Abstract. The measurement of neutron-capture cross sections of many actinides is complicated by the difficulty in separating capture γ-rays from the large fission-fragment prompt γ-ray background. For example, current estimates of the capture cross section of ^{233}U show large discrepancies, with differences of more than 20%. To improve the accuracy of data, a new experimental set-up for the simultaneous measurement of the neutron-induced capture and fission cross sections was designed, assembled and optimized. The measurements will be performed at the GEel LINear Accelerator (GELINA) neutron time-of-flight facility in Belgium, where neutron cross sections can be measured over a wide energy range with high energy resolution. The fission detector consists of a dedicated multi-plate high-efficiency ionization chamber (IC). The γ-rays produced in capture reaction are detected by an array of C_6D_6 scintillators. Fission γ−rays are distinguished from capture γ−rays by the anticoincidence signals from the IC and the C_6D_6 detectors. For the undetected fission events a correction has to be applied based on the efficiency of the IC that should be high and known with a high accuracy. The performance of the IC during dedicated test experiments is presented, focusing on the determination of the detection efficiency.

a Corresponding author: mathieu@cenbg.in2p3.fr

1 Introduction

Nuclear energy could be part of the solution to the world energy demand and global warming crises. Nevertheless some issues of nuclear energy have to be solved first, among which is the generation of very radioactive and long-lived waste. The next generation of nuclear reactors will allow the incineration of the most radiotoxic ones: the minor actinides produced by radiative capture on the fuel elements. They will also be able to breed more fuel than they consume, through neutron capture on a fertile element, namely ^{232}Th or ^{238}U. The development of these reactors requires a precise knowledge of the cross sections involved in the process, especially the fission and capture cross sections of actinide isotopes [1,2]. Nowadays, ^{238}U/^{239}Pu is partially used in nuclear reactors and is rather well known. On the contrary ^{232}Th/^{233}U has been much less studied, with only specific tests in few reactors (MSRE [3], Shipping port [4,5]).

The ACEN (Aval du Cycle et Energie Nucléaire) group of the CENBG (Centre d'Etudes Nucléaires de Bordeaux Gradignan) has been involved for several years in the measurement of the capture and fission cross sections of ^{233}U, in close collaboration with IRMM in Geel (Belgium), since the final experiment will be carried out at the GELINA (GEel LINear Accelerator) neutron Time-of-Flight (TOF) facility [6].

In the present contribution, the context and principle of the experiment will be summarized, as well as the functioning of the fission detection. Different methods to measure the Ionization-Chamber (IC) efficiency will be presented. We will focus on a method based on the detection of the prompt fission neutrons. This method will be discussed and the first results will be presented.

2 Experimental method and set-up

2.1 ^{233}U capture cross section

The breeding capabilities of a nuclear reactor are directly linked to the average capture and fission cross sections of the fissile isotope, which is ^{233}U for the thorium cycle. They characterize the proportion of neutrons lost in the sterile ^{233}U radiative capture process. Thus a well known capture cross section is required for accurate nuclear plant simulations, especially in the resonance region. There are only two high-quality experimental data sets for ^{233}U in this energy region, those by Weston [7,8] and Berthoumieux [9]. Figure 1 shows these capture cross sections as well as three evaluations (ENDF/B-VI, ENDF/B-VII and JEFF-3.1). Some clear discrepancies can be seen on these plots. In particular, there are shifts in the energy and differences in amplitude between the capture resonances of the two data sets. The evaluations directly reflect these discrepancies, with significant differences between them.

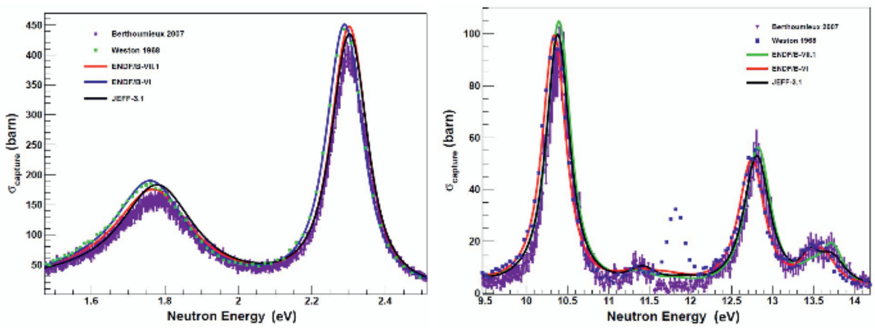

Figure 1. Capture cross sections from different experimental data and evaluated databases in the epithermal region around 2 eV (left) and 12 eV (right). A capture resonance was seen by Weston et al. at 11.8 eV but not by Berthoumieux et al. It was probably due to a contaminant and was not taken into account in the databases.

2.2 Response of the Ionization Chamber

Measuring the capture cross section of a fissile nucleus is a difficult task because the emitted γ-rays are hidden in a very large background of prompt fission γ-rays. A standard method to solve this issue is to detect the fission events and to use them as a veto on the γ-ray detection (VETO method). Nevertheless, no fission detector has 100% efficiency and some fission events remain undetected. The most efficient type of fission detectors are Ionization Chambers (IC) which detect the Fission Fragments (FF) escaping from the target and depositing their energy in an ionizing gas.

The IC developed for the ^{233}U capture cross-section measurement performed by our group is quite simple. Ten thin targets of ^{233}U (250 μg/cm²) will be used in order to maximize the exit probability of the FF without lowering the total amount of material. To keep a quite compact geometry, the chambers are made of two parallel plates without Frisch-grid placed at a distance of 5 mm, and filled with a standard 90% Ar – 10% CH_4 gas mixture. The target sample is deposited on the cathode. The cathode-anode distance does not allow a good energy resolution, but a good timing resolution was obtained: the bias voltage (100V) was chosen to ensure the maximum drift velocity of the electrons in the gas, leading to a faster response of the detector (full details of the experimental setup can be found in [10]). Figure 2 shows a typical spectrum obtained with our detector with a ^{252}Cf source. The high peak at low energy is due to alpha radioactivity, while the large peak is due to FF. In between these two peaks lies a deep valley, that we will call the α-FF valley in the following.

The heavy/light peaks of the usual FF distribution can only be barely seen because of the poor energy resolution: the spectrum shape is characteristic of the incomplete FF energy deposition, since the range is much larger than the cathode-anode distance (few centimeters versus 5 mm). The deposited energy therefore depends on the FF emission angle:
- The steep increase at low energy (around channel 300) is due to FF emitted perpendicularly to the target, which have the shortest path in the IC. Their energy deposition is therefore quite small.
- As the emission angle increases, the path and the energy deposition increase. Maximum energy depositions of the heavy and light fragments are barely seen at channel 750 and 1000, respectively.
- Higher deposited energies can be obtained when, due to straggling effects on the FF emitted in the backward direction, both FF are detected at the same time.
- However, at grazing angles, the FF loses a significant part of its energy in the target itself and/or in the backing irregularities. Depending of the target thickness, the backing surface state and the angular straggling, the energy deposited in the gas greatly varies. Thus, the high-energy tail and the bottom of the α-FF valley are both due to FF emitted at grazing angles.

The α pile-up is moderate in this case since the ^{252}Cf activity is moderate (370 kBq), and the detector and electronics used are quite fast (rise time of about 50 ns, shaping time of 0.25 μs). The α pile-up populates the high-energy tail of the α peak. With a slower detection system or higher activity, this tail would grow and would dramatically reduce the width and the depth of the α-FF valley.

2.3 IC efficiency

To identify fission events, one has to select FF without including α particles. This is done by setting a threshold in the α-FF valley. This threshold must be set carefully as there is an overlap between α and FF:
- α particles with amplitudes above the threshold will be interpreted as fission events. This will induce a small systematic error on the cross section measurement.
- FF with amplitudes below the threshold will be lost, but these "undetected fission" events can be corrected. Their proportion represents $1-\varepsilon_{IC}$, where ε_{IC} is the fission efficiency of the IC. By knowing ε_{IC}, one can subtract the "undetected fission" contribution from the capture γ-ray spectrum obtained after the VETO operation.

A deep and wide α-FF valley is required to achieve a clear discrimination, and greatly reduce the sensitivity of ε_{IC} to a possible electronic drift of the IC amplitude.

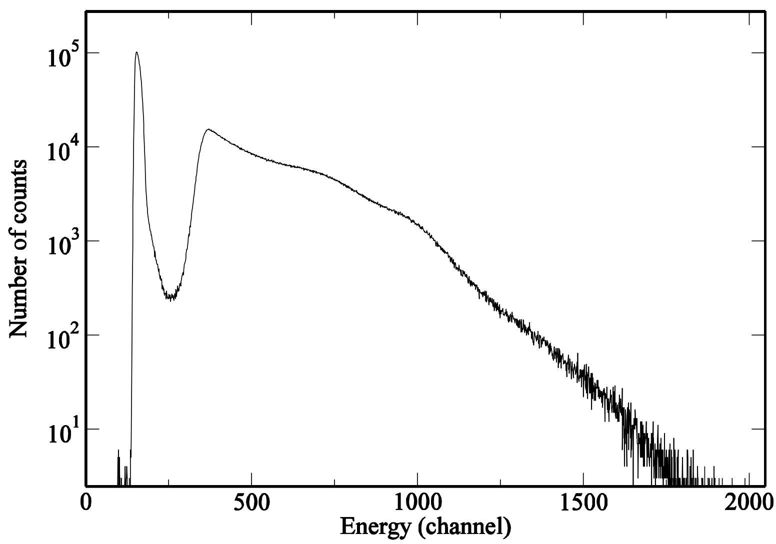

Figure 2. Pulse height spectrum of our ionization chamber obtained with a ^{252}Cf source. The peak on the left is due to the alpha radioactivity, partly eliminated with an electronic threshold. The very broad peak is due to incomplete fission fragment energy deposition.

The fission efficiency ε_{IC} must be known with an excellent precision and accuracy. Indeed, for ^{233}U, capture γ-rays may represent only 3% of the total γ-rays emitted by the nucleus. This proportion depends on the cross sections and is obviously very low for pure fission resonances. As some capture cross-section measurements are based on such pure fission resonances (e.g. the Total Absorption Calorimeter method [7]) these energy regions are of particular interest. In fact, an important systematic error arises if a small capture resonance is hidden. An uncertainty of 1% on ε_{IC} leads to an uncertainty in the determination of the undetected fission events and thus on the capture cross section, that varies from 15% up to 30% in a pure fission resonance. Therefore, the IC efficiency has to be determined with a tremendous accuracy for the capture cross-section to be measured with a good accuracy even for pure fission resonances.

3 Methods to determine the IC efficiency

3.1 Commonly used methods

Depending on the sample nucleus (half-life and spontaneous-fission half-life), knowledge of its fission cross section, and the IC features (presence of the Frisch grid), several methods can be used to measure the fission efficiency. The simplest case occurs when the nucleus of interest fissions spontaneously, like ^{252}Cf, and the fission rate depends only on the amount of material which is easily obtained via alpha spectrometry. The uncertainty sources of this method are the solid angle of the alpha counting set-up and the dead-time correction of the fission measurement. For certain nuclei, the spontaneous fission yield y_{SF} may also be a significant source of uncertainty (for instance the y_{SF} of ^{240}Pu has an uncertainty of 3.5% [11]). For ^{252}Cf, an uncertainty of 1% for the fission efficiency can be achieved.

If the nucleus does not fission spontaneously and the fission-fragment angular distribution is isotropic, the best solution is to use a Frisch-gridded IC. This device allows one to estimate the emission angle of the FF and the missing FF at grazing angles can be inferred through a simple constant fit to the measured FF angular distribution [12]. The uncertainty of this method can be as low as 1%.

In the cases where no Frisch-grid is used (as is our case due to compactness constraints) or when the angular distribution is not isotropic, the fission rate is much more complex to measure. This quantity depends on the neutron flux and has to be normalized to the cross section. The determination of the flux is usually a strong source of uncertainty, since it relies on the detection of a reference reaction ($^{235}U(n,f)$, $^{10}B(n,\alpha)$, $^{6}Li(n,t)$…). This reference itself is function of the amount of material, the detection efficiency and the reaction cross-section. In the end, the accuracy of the IC efficiency is not better than few percent with this method.

3.2 Method based on prompt fission neutrons

A much simpler and more accurate method relies on using the prompt neutrons emitted by the fission fragments just after the fission process. These fast neutrons can be detected by scintillators via neutron scattering and recoil-nucleus energy deposition. Due to differences in the energy-deposition mechanisms, the signals from γ-rays and neutrons can be discriminated via an analysis of the Pulse Shape (PSD). The detected neutrons can be detected in coincidence with a FF, and the IC efficiency is then given by the simple equation:

$$\varepsilon_{IC} = N_{n\_coinc\_FF} / N_{n\_tot} \tag{1}$$

Where:
$N_{n\_coinc\_FF}$ is the number of neutrons detected in coincidence with a FF, and
N_{n_tot} is the total number of detected neutrons

This method is quite robust since in a first approximation, it does not depend on other factors. There is no dead-time correction needed because these quantities are measured in the same experiment. Moreover, there is no need to know the exact position of the scintillators (distance and angle). Nevertheless, one has to carefully check that the selected neutrons are free from any contribution (neutrons from other sources, wrongly discriminated γ-rays…), otherwise the N_{n_tot} term would be overestimated, leading to an underestimation of the efficiency. The accuracy that can be obtained with this method is very good, with an uncertainty for the efficiency below 1%.

In principle, prompt fission γ-rays may be used in exactly the same way. However they suffer from contribution from other strong sources that may reduce the accuracy (contamination from background γ-rays) or even induce systematic errors (contribution from other physical processes such as radiative capture).

4. Experiments and Results

Experiments have been carried out to measure the efficiency of our IC with a ^{252}Cf source. Six C_6D_6 liquid scintillators were placed around the chamber to detect prompt neutrons. In the standard configuration, all the scintillators are located at 90° with respect to the target normal. To test other configurations, C_6D_6 #4 and #6 were placed at 0° and 45°, respectively, at different distances from the target, while the other scintillators remained at 90°. Figure 3 compiles the IC efficiencies measured for each scintillator. Black dots are obtained from C_6D_6 located at 90°, while red and blue dots are obtained from scintillators at 0° and 45° respectively. The error bars are only statistical and are particularly small due to a strong correlation between the two terms of Eq. (1).

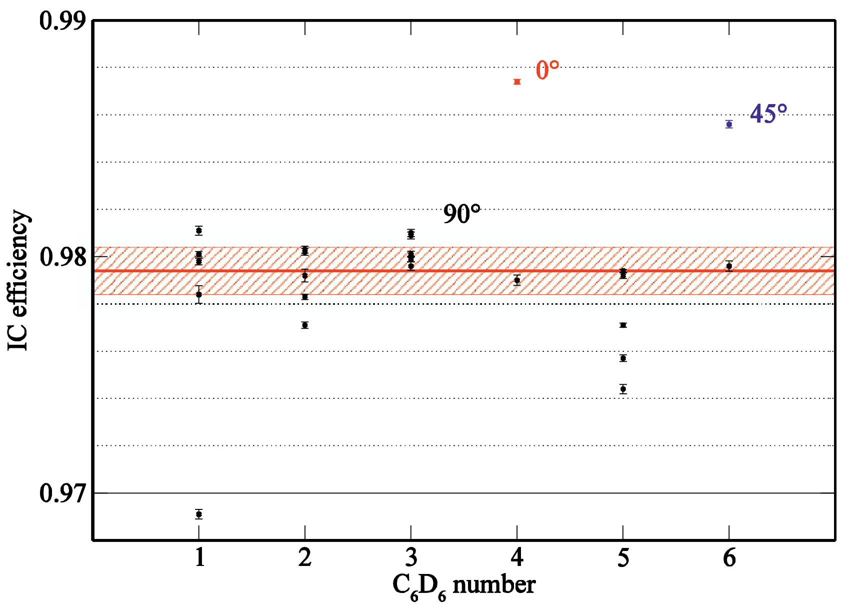

Figure 3. Ionization Chamber efficiencies obtained from neutrons detected in the different C_6D_6 scintillators in different configurations, with their statistical error bars. Black points are obtained in the same experimental conditions (C_6D_6 at 90° with respect to the target normal). The two points indicated as "0°" and "45°" were obtained with C_6D_6 #4 and #6 respectively placed at these angles. The average value is indicated with its standard deviation for measurements at 90°, excluding C_6D_6 #5 and one point of C_6D_6 #1.

Figure 3 shows that C_6D_6 #1 and #5 give values for the efficiency that are significantly below the majority of the other values. One value of the efficiency of C_6D_6 #1 is 10 times lower than the standard deviation of the other results and the discrepancy of the C_6D_6 #5 result is twice the discrepancy of the others. This implies that the efficiency measurement requires the use of several detectors and that care must be taken to ensure their stability. Except these discrepant points, there is a very good reproducibility of the results, with an average value for the efficiency of 97.98% ± 0.10%.

Scintillators at 0° and 45° show a clear discrepancy, with much higher efficiencies. This increase is due to a kinematic effect: fission neutrons are slightly focused in the direction of the FF. Therefore, they have a higher probability of being detected in the C_6D_6 located around the FF emission angle. The IC pulse height spectra in coincidence with neutron detection in a C_6D_6 depend on the scintillator position as can be seen on the left side of Figure 4. The C_6D_6 located at 0° will favor coincidences with FF emitted at 0°, and the C_6D_6 located at 90° favors FF emitted at 90°. As there are less FF below the threshold in the "coinc with 0°" spectrum, the measured efficiency is higher. A simulation code has also been developed, taking into account the kinematic effect of the neutron emission. Results presented in the right part of Figure 4 show three IC spectra in coincidence with neutron detectors at

0°, 45° and 90°. Very strong similarities in spectra shapes can be seen between simulated and experimental data.

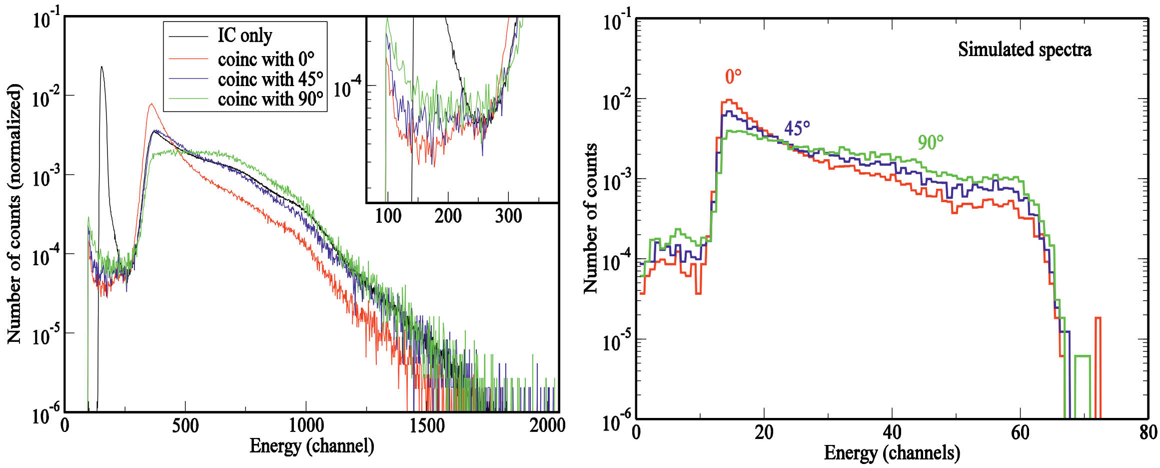

Figure 4. (Left) Spectra of the IC, in single event or in coincidence with different C_6D_6 scintillators. For comparison, all spectra are normalized to 1 FF. Details at low energy are emphasized in the inset. (Right) Simulated spectra of the IC, in coincidence with a neutron in detectors at 0°, 45° and 90°, normalized to 1 FF.

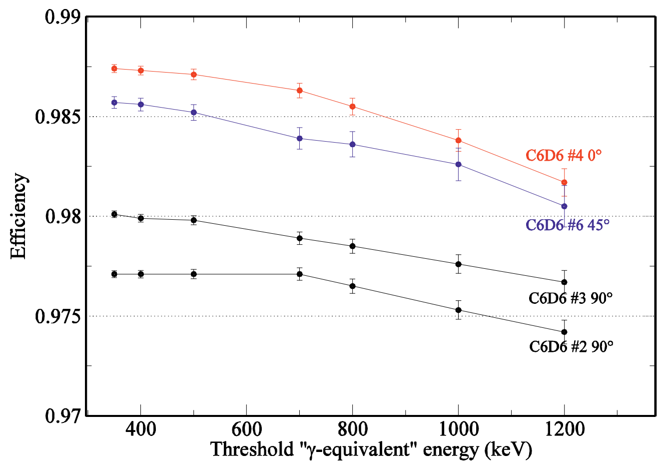

Figure 5. Efficiency of the IC from different C_6D_6 scintillators, as a function of the neutron threshold γ-equivalent energy.

To obtain a better estimation of the efficiency, one has to take into account this angular dependence, as if the neutrons were detected in a 4π detector. Assuming the isotropic emission of prompt neutrons in the laboratory system (which is a valid assumption for ^{252}Cf spontaneous fission), the averaged value over the cos(θ) distribution gives an efficiency of 98.4% ± 0.2%. There are still other factors that have a small influence on the efficiency, such as the neutron energy threshold (see on Figure 5) or the C_6D_6 distance to the target. As can be seen, the efficiency measured decreases with the threshold independently of the C_6D_6 angle. This is probably due to a cosmic-neutron background. This background, negligible compared to fission neutrons at low energy, is of growing importance as the neutron energy increases. More investigations are needed in order to correct this effect and to determine the best estimate of the real efficiency.

5. Conclusion

The fission detection efficiency ε_{IC} is a key parameter for the measurement of the capture cross section of a fissile nucleus with the VETO method. Due to the large number of fission γ-rays compared to capture γ-rays, ε_{IC} has to be high and known with an excellent accuracy. This allows one to obtain a clean fission subtraction and a low uncertainty on the capture cross section. The efficiency measurement via the prompt neutrons emitted by the fission fragments and detected in scintillators is very suited to obtain a very accurate value. Several corrections of few tenths of percent have to be taken into account, which include the dependence on the scintillator angular position. Further investigations are in progress, with the aim of reaching an uncertainty below 0.5%.

References

1. L. C. Leal et al., ORNL/TM-2000/372 (2001)
2. A. Bidaud, O. Meplan, A. Nuttin, S. David, J.N. Wilson, F. Michel-Sendis, P. Guillemin, N. Capellan, Proc. of Nucl. Data for Sci. and Tech. 2007, Nice, France, p. 919 (2007)
3. P. Haubenreich, J.R. Engel, Nucl. Appli. and Tech. **8**, p. 118 (1970)
4. W.J. Babyak, L.B. Freeman, H.F. Raab, Nucl. News, p. 114 (1988)
5. J.C. Clayton, *Westinghouse Bettis Atomic Power Laboratory* WAPD-T-3007 (1993)
6. W. Mondelaers and P. Schillebeeckx, *Notiziaro Neutroni e Luce di Sincrotone* **11**, p. 19 (2006)
7. L.W. Weston et al., Nucl. Sci. Eng. **34**, p.1 (1968)
8. L.W. Weston et al., Nucl. Sci. Eng. **42**, p.143 (1970)
9. E. Berthoumieux et al., Proc. of Nucl. Data for Sci. and Tech **1**, p. 571, Nice, France (2007)
10. I. Companis, M. Aïche, L. Mathieu, G. Kessedjian, P. Schillebeeckx, G. Barreau, G. Boutoux, S. Czajkowski, B. Haas, B. Jurado, A.J.M. Plompen, V. Simutkin, I. Tsekhanovich, Proc. of the Third Int. Workshop on Compound-Nuclear Reactions and Related Topics CNR11, Prague, Czech Republic (2011)
11. J.K. Tuli, BNL-NCS-51655-01/02-Rev **2** (2001)
12. C. Budtz-Jorgensen, H.-H. Knitter and G. Bortels, Nucl. Instr. and Meth. A **236**, p. 630 (1985)