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# Extending the panel of inelastic scattering reactions availaible for dosimetry: Definition of the experimental campaign at the JSI TRIGA reactor

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**Abstract.** The epithermal to fast neutron domain lacks reactions to enhance the knowledge of spectrum in this energy range (1 keV to 1 MeV). Previous studies on this topic focused on $^{94}Zr(n,\gamma)$ capture and $^{117}Sn(n,n')$ inelastic reactions. In the framework of a research collaboration between the French Atomic and Alternative Energies Commission (CEA) and the Jožef Stefan Institute (JSI), a new project consists in finding new inelastic reactions suitable for dosimetry. This paper describes the selection process of new reactions to be tested in an experimental campaign at JSI.

## 1 Introduction

Improving the experimental characterization of the neutron spectrum in an irradiation channel is a key point for reactor dosimetry. The epithermal to fast neutron domain lacks reactions to enhance the knowledge of spectrum in this energy range (1 keV to 1 MeV). Previous studies on this topic at *Dosimetry, Sensors, Instrumentation Laboratory (LDCI)* focused on $^{94}Zr(n,\gamma)$ [1] capture and $^{117}Sn(n,n')$ [2] inelastic reaction. In the framework of a research collaboration between the French Atomic and Alternative Energies Commission (CEA) and the Jožef Stefan Institute (JSI), the teams have launched a research project to extend the panel of inelastic scattering reactions available to dosimetry community, with an extended range of applications (surveillance capsule, Material Testing Reactors (MTR), Generation-IV reactors and fusion facilities).

## 2 State of art and search for new isomers

### 2.1 State of art: Inelastic reactions available to the community

The inelastic reactions available today for dosimetry are the few listed in Table 1. The natural element contains generally only one isotope (Rh, Nb). For indium case, the $^{115}In$ is the major

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isotope without stable isotopes on each side (114,116), so it can be nearly considered as a unique isotope. The mercury reaction is not used any more (short half-life, toxicity, fast neutron facilities availability). A reaction is considered to be at high level of confidence if the cross section is available in IRDFF[3].

**Table 1.** Inelastic reactions $X(n,n')X^m$ available for dosimetry.

| Isotope | Abundance | Isomer level (keV) | Half-life | X or $\gamma$ rays (keV) | Use |
|---|---|---|---|---|---|
| $^{93}Nb$ | 100% | 30.8 | 16.12 yr | 16.5 ($X_{K\alpha}$), 18.6 ($X_{K\beta}$) | MTR, Surveillance |
| $^{103}Rh$ | 100% | 39.8 | 56.1 min | 20 ($X_{K\alpha}$), 22 ($X_{K\beta}$) | ZPR |
| $^{115}In$ | 95.7% | 336.2 | 4.486 h | 336.2 | ZPR, MTR at low power |
| $^{113}In$ | 4.3% | 391.7 | 99.1 min | 391.7 | new in IRDFF |
| $^{199}Hg$ | 16.87% | 632.5 | 42.67 min | 158.3, 374.1 | Past use in YAYOI, JMTR |
| $^{117}Sn$ | 93% (enr.) | 314.6 | 14.0 d | 156.02, 158.6 | Medical, ZPR , MTR Not yet in IRDFF |

The measured isomers mainly decay by internal transition (IT). Table 1 shows also that for isomers with a low energy level, the gamma line is strongly converted to X-ray ($X_{K\alpha}$, $X_{K\beta}$). Isomer measurement relies on good knowledge of atomic and nuclear data: fluorescence yield ($\omega_K$) and internal conversion coefficient (ICC) [4]. The X-measurement has been improved for years by the laboratory [5]. Consequently, searching isomers with a low energy level enhance the difficulty of measurement as it requires a dedicated low energy detector and the development of associated methods.

In a first attempt of finding new reactions, several experiments have been carried on the $^{117}Sn(n,n')^{117m}Sn$ reaction (last line of Table 1) [2]. The main conclusions of the studies on this reaction were:

- The use of enriched isotope is needed to avoid interferences.
- $^{117m}Sn$ half-life value must be updated in some evaluations.
- The $^{117}Sn(n,n')^{117m}Sn$ cross section needs a global re-evaluation to be included in IRDFF.

### 2.2 Search for reactions and associated isomers

The first step is to identify isomers of natural isotopes with a sufficient half-life to be counted on a $\gamma$ and/or X-ray detector after the irradiation (off-line measurements). A global search using NUDAT database[6] gives the followings results (Table 2). Isomer level is more reliable data for the first step of analysis than a pseudo-threshold computed on cross section librairies with all potential flaws in ENDF format especially the LFS flag[7].

The first criterion is to remove the gaseous elements (Kr, Xe) because the measurement facilities available at Cadarache and JSI are dedicated to solid samples measurement. Osmium is also removed because of its high toxicity. Cadmium is also removed as it is a well known material with very high capture cross section.

Table 2 shows a great variety of natural abundance for the isotope of interest. Some elements have also a great number of natural isotopes (10 for tin, 8 for Te, 7 for Ba). Interferences may arise and can prevent the use of the inelastic reaction.

**Table 2.** First list of isotopes and isomer main characteristics (level / half-life).

| Isotope | Abundance [8] | Isomer level (keV) | Half-life | | Main rays (intensity) (keV) (%) |
|---|---|---|---|---|---|
| $^{83}Kr$ | 11.5% | 41.55 | 1.83 | h | 12 (25.25%, $X_{K\alpha}$) |
| $^{87}Sr$ | 7% | 388.53 | 2.815 | h | 388.53 (82.2%) |
| $^{111}Cd$ | 12.80% | 396.21 | 48.5 | min | 245.4 (94%),150.8 (29.1%) |
| $^{113}Cd$ | 12.22% | 263.73 | 14.1 | yr | 263.7 (0.03%) |
| $^{117}Sn$ | 7.68% | 314.6 | 14.0 | d | 158.6 (86.4%), 156.02 (2.1%) |
| $^{119}Sn$ | 8.59% | 89.53 | 293.1 | d | 23.8 (16.5%), 25 (22.8%, $X_{K\alpha}$) |
| $^{123}Te$ | 0.89% | 247.5 | 119.2 | d | 159 (84.3%), 27 (41.4%, $X_{K\alpha}$) |
| $^{125}Te$ | 7.07% | 144.78 | 57.4 | d | 35.5 (7.3%), 27 (97.6%, $X_{K\alpha}$) |
| $^{129}Xe$ | 26.40% | 236.14 | 8.88 | d | 196.5 (4.6%), 39.6 (7.5%) |
| $^{131}Xe$ | 21.23% | 163.93 | 11.84 | d | 163.9 (2%), 29 (44.3, $X_{K\alpha}$) |
| $^{135}Ba$ | 6.59% | 268.22 | 28.7 | h | 268.22 (16%) |
| $^{189}Os$ | 16.15% | 30.82 | 5.81 | h | 8.91 (21.8% $X_L$) |
| $^{193}Ir$ | 62.7% | 80.24 | 10.53 | d | 9.18 (21.9% $X_L$), 64 (0.4%, $X_{K\alpha}$) |
| $^{195}Pt$ | 33.78% | 259.08 | 4.01 | d | 98.9 (11.7%), 65 (59.1%, $X_{K\alpha}$) |

## 3 Interferences and activation computations

For multi-isotopes elements, we have to estimate the competing reactions that produce the isomer. We consider the following main inelastic reaction of interest (1).

$$ {}^{A}_{Z}X + n \rightarrow {}^{A}_{Z}X^{m} + n' \tag{1} $$

The non-threshold capture reaction (2) produce the same isomer. If ${}^{A-1}_{Z}X$ isotope abundance has the same order of magnitude than ${}^{A-1}_{Z}X$ abundance, the thermal capture may result in predominant activity at the end of irradiation.

$$ {}^{A-1}_{Z}X + n \rightarrow {}^{A}_{Z}X^{m} + \gamma \tag{2} $$

The ${}^{A-1}_{Z}X(n,2n){}^{A}_{Z}X^{m}$ reaction should also be considered, but due to its higher threshold, cross sections are lower than inelastic ones.

The feedback from known dosimeters (Rh, Nb and In) shows that other reactions may interfere in the measurement of the isomer:

- The ${}^{A}_{Z}X(n,\gamma)$ produce a strong gamma emitter with a short half-life hiding the isomer ($^{104}Rh$, $^{116m}In$). Measurements have to be delayed waiting for the decay of this emitter.
- The ${}^{A}_{Z}X(n,\gamma)$ produce a gamma emitter that induces fluorescence in the material ($^{94}Nb$, $^{95}Nb$). Computations are required to estimate the contributions when cooling down not is not possible ($^{94}Nb$).

As a general method and in order to evaluate the contributions of interfering isotopes or competing reactions, activation computations have been performed on the selected elements with the CEA code DARWIN[9] using the previously established neutron spectrum of the pneumatic irradiation channel of the JSI TRIGA MARK-II reactor. Table 3 summarize the activation parameters of the study.

Enriched compositions are computed just as for $^{117}Sn$ studies. An enriched material may lower interferences as required and enhance the reaction of interest. These computations are done only if the enriched material is available from suppliers.

The neutron filtered/bare activity ratio computation has a double objective:

**Table 3.** Activation computation parameters.

| Parameter | Values | Comment |
|---|---|---|
| Isotopic compositions | IUPAC[8]<br>enriched in isotope | natural element<br>if available commercially |
| Outputs of the code | activities, gamma spectrum | check all gamma emitters |
| Neutron filter | bare and boron nitride (BN) | impact of capture reactions |
| Cooling times | From min. to days. | optimize measurements |

**Table 4.** Activity ratio (BN Filtered/Bare) for a selection of computations.

| Computation | Enrichment | Isomer | Ratio |
|---|---|---|---|
| Sr | natural | $^{87m}Sr$ | 29% |
| Sr87* | 92.6% | $^{87m}Sr$ | 85% |
| Sn | natural | $^{117m}Sn$ | 86% |
| Sn | natural | $^{119m}Sn$ | 92% |
| Sn117* | 92.8% | $^{117m}Sn$ | 99.6% |
| Sn119* | 96.3% | $^{119m}Sn$ | 99.8% |
| Te | natural | $^{123m}Te$ | 49% |
| Te | natural | $^{125m}Te$ | 93% |
| Te125* | 99.89% | $^{125m}Te$ | 99.8% |
| Ba | natural | $^{135m}Ba$ | 81% |
| Ba135* | 94.9% | $^{135m}Ba$ | 99.6% |
| Ir | natural | $^{193m}Ir$ | 99% |
| Pt | natural | $^{195m}Pt$ | 76% |

- When using natural element, the computation gives the yield of fast reactions.
- When using enriched element, the computation indicates if a neutron filter is still necessary. The previous experimental campaign with enriched $^{117}$Sn showed experimentally that the $^{117}$Sn foil can be irradiated bare [10].

Table 4 shows the activity ratio for a selected set of computations. The computations are experimental design computations so no uncertainties are given at this stage. The computations marked with stars indicate the presence of enriched compounds. The second column indicates the enrichment of the isotope of interest.

For some reactions, the ratio is close to 100% even with natural element. The production of $^{193m}Ir$ is entirely from inelastic reaction because the only stable isotopes for iridium are $^{191}Ir$ and $^{193}Ir$. But the iridium computation gives a strong $^{192}Ir$ activity produced by $^{191}Ir(n,\gamma)$. This activity cannot be lowered enough using enriched compound and neutron filter. This ratio is one but not the only indicator for the selection of isotopes.

The activities at different cooling times are computed to determine the optimal measurement time window just as cooling times are required for Rh and In foils.

## 4 Material considerations and gamma detector modelling

The common shape for dosimeters are foils or wire. When using foils, the thickness is one of the most important parameter to compute the self-absorption of the $\gamma$ or X rays in the dosimeter[5]. So, when measuring new isomers, we have to point out the following items:

- As for Nb and Rh, nuclear and atomic data have to be checked with attention for computing corrections on raw measurements. Some values, not recently evaluated, may introduce a large uncertainty.
- If the material is not available in thin foils, like common dosimeters, the shape of the new dosimeter may also create uncertainty. The previous experiments with tin[2] have shown that a ductile metal like tin gives simple ways of manufacturing a dosimeter. Just as indium, tin has a low melting point and is not suitable for some experiments.
- For some elements (Te, Sr, Ba), enriched isotope is not available as foil but as powder. It has to be encapsulated in a suitable way to avoid contamination in the facilities. Furthermore, encapsulation has to be similar as a foil (disk) to lower the uncertainties of the computed corrections. Thickness and apparent density are here key parameters to obtain a measured activity without a too large uncertainty. Measurement may also include a destructive method such as $^{93m}Nb$ activity measurement [11].

To estimate corrections, a Monte Carlo modelling of the dosimeter and the detector with GEANT4[12] can estimate the requested parameters. GEANT4 includes a radioactive decay module that can describe a source as a mixture of isotopes using the results from DARWIN activation computations.

The simulations can estimate:

- The $\gamma$ spectrum emitted to the detector,
- Peak summing corrections, especially (X,$\gamma$) which may occur in close geometry with low energy detectors (Figure 1 shows the ($\gamma$,$\gamma$) sum peak at 314 keV and the (X,$\gamma$)) sum peaks above the main $\gamma$ peak at 158 keV),
- Fluorescence corrections,
- Self absorption corrections.

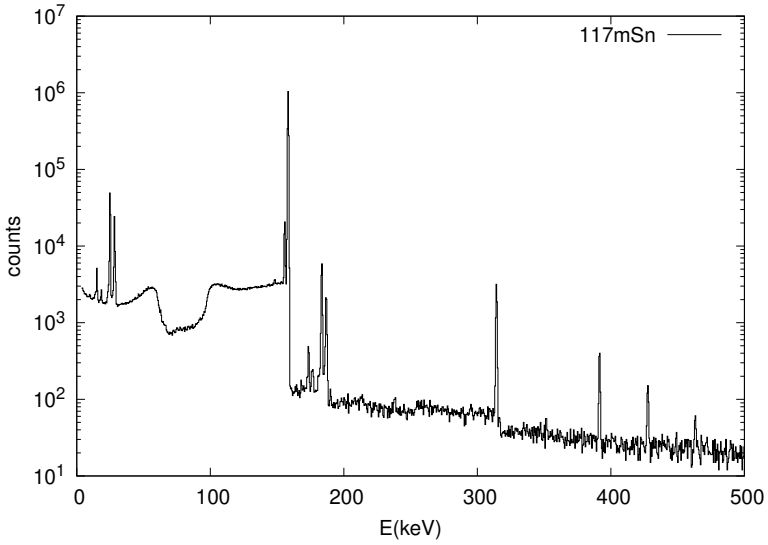


**Figure 1.** $^{117m}Sn$ measured gamma spectrum with ($\gamma$,$\gamma$) and ($\gamma$,$X$) sum peaks.

The gamma spectrum, even computed with a non optimized model of dosimeter/detector, gives a complementary information to activation computations to indicate if the peaks of the isomer are detectable with other activations products in the mixture.

## 5 Final selection of dosimeters

The final selection of foils have been made considering all the items described in the previous sections. As already mentioned gaseous elements were removed (Kr,Xe). The cadmium and iridium were also removed because their isomers were not detectable due to other activations reactions and their low emitting isomers. Osmium toxicity is not compatible with facilities safety rules.

The selected dosimeters are shown in Table 5 with respectives advantages and drawbacks. They are planned to be irradiated in the pneumatic tube channel of the TRIGA MARK II at JSI along with reference monitors (Al-Co, Al-Au, Ni) as references. Standard niobium and rhodium foils will also be added to the panel.

**Table 5.** Final selection of dosimeters.

| Material | Isomer | Advantages | Drawbacks |
|---|---|---|---|
| $^{87}Sr*$ | $^{87m}Sr$ | strong gamma emitter<br>enriched material | short half-life<br>$SrCO_3$ powder<br>neutron filter |
| $^{117}Sn*$ | $^{117m}Sn$ | half-life, gamma emitter,<br>ductile metal | (X,$\gamma$) summing peaks |
| $^{119}Sn*$ | $^{119m}Sn$ | ductile metal, half-life | X and very low energy $\gamma$ emitter |
| Te | $^{123m}Te$,<br>$^{125m}Te$ | long half-life<br>long half-life | (X,$\gamma$) summing peaks<br>X and low intensity $\gamma$ emitter<br>have to wait $^{131}I$ decay |
| $^{125}Te*$ | $^{125m}Te$ | long half-life<br>enriched material | X and low intensity $\gamma$ emitter<br>Te powder |
| $^{135}Ba*$ | $^{135m}Ba$ | strong gamma emitter<br>enriched material | short half-life<br>$BaCO_3$ powder |
| Pt | $^{195m}Pt$ | relatively short half-life<br>foils available | low energy $\gamma$ emitter<br>no enriched material |
| Hg | $^{199m}Hg$ | gamma emitter | short half-life<br>$HgO_2$ powder, toxicity<br>neutron filter |

## 6 Conclusion and future work

The planned experimental campaign has two major objectives. The first objective is to demonstrate the measurement feasibility of isomers produced by inelastic reactions. A comprehensive analysis of all data needed to provide a reliable activity value is necessary to establish the uncertainty budget of the measurements. This analysis, with Monte-Carlo code, will point out the key values that have to be reevaluated (half-life, emission probabilities, atomic data,...). The associated recommendations will focus on the improvement of these values as already done with niobium and rhodium. The uncertainties induced by dosimeter

shapes (thickness, density, foil or not) can define a future work on new manufacturing processes of the selected materials. The low energy emitters may benefit from recent progress on nano-materials technologies (low thickness deposits of materials methods for example).

The second objective is the feedback on cross sections. After obtaining consolidated activity values, a joint analysis will be carried out, comparing the experimental $(n, n')$ reaction rates to the calculated results with the irradiation conditions in the channel[13]. The comparison between the measured reaction rates and the calculations and different nuclear data libraries (ENDF/B-VIII.0[14], JEFF 3.3[15] and TENDL[16] libraries) may give a rough indication of the quality of the (n,n') reaction cross section data. In case there is good agreement between the measurements and the calculations for some candidate reactions, proposals will be made to include these reactions into IRDFF. For the reactions where there are discrepancies between the measurements and final calculations, the measurement data will provide assistance to the nuclear data evaluation community in pinpointing the problematic energy regions in the evaluations.

Feedback on measurements and cross sections are necessary to go towards a routine use of the new dosimeters in the experimental facilities.

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