Dosimetry Reactions which need updating or were recommended for evaluation and inclusion in IRDFF at RCM-1 and RCM-2.

Following the recommendations adopted by RCM-1 (July 2013, <u>INDC(NDS)-0639</u>, p. 19-20) and RCM-2 (March 2015, <u>INDC(NDS)-0682</u>), advices from the Fusion community, and an assessment of possibility to implement new (re)evaluations made by K. Zolotarev and NDS/IAEA staff we come to following summary.

Summary

(i) we decided to do for the next version of IRDFF:

- re-evaluate ${}^{23}Na(n,\gamma)$ and ${}^{23}Na(n,2n){}^{22}Na$ (the closest alternatives/perspectives are ${}^{37}Cl(n,\gamma){}^{38}Cl$ or ${}^{127}I(n,\gamma){}^{128}I$);
- newly evaluate ${}^{27}\text{Al}(n,2n){}^{26}\text{Al}$.
- (ii) the high energy applications evaluate and eventually include in IRDFF like the high threshold reactions:

(n,xn) on ¹⁹⁷Au(n,3-5n), ¹⁶⁹Tm(n,2-3n), ²⁰⁹Bi(n,3-10n) (see <u>Evaluation</u> made by V. Pronayev), ⁵⁹Co(n,3-5n), ⁸⁹Y(n,2-4n), ⁹³Nb(n,2-4n), ¹⁰³Rh(n,4-8n), ¹³⁹La(n,4-10n), ¹⁷⁵Lu(n,2-4n), ⁶³Cu(n,?);

composite reactions resulting to the production of the same radioactive residual: ${}^{27}Al(n,x){}^{24}Na, {}^{nat}Fe(n,x){}^{54}Mn, {}^{nat}Fe(n,x){}^{56}Mn, {}^{nat}Fe(n,x){}^{51}Cr, {}^{nat}Ti(n,x){}^{47}Sc, {}^{nat}Ti(n,x){}^{48}Sc, {}^{nat}Ni(n,x){}^{57}Co, {}^{nat}Ni(n,x){}^{60}Co, {}^{nat}Ni(n,x){}^{57}Ni, natCu(n,x){}^{64}Cu, as well as {}^{10}B(n,X){}^{4}He, {}^{11}B(n,X){}^{4}He, {}^{6}Li(n,X){}^{4}He, and {}^{7}Li(n,X){}^{4}He.$

(iii) include the ⁵⁸Co/^{58m}Co energy dependant branching ratio in IRDFF (ratio data are available in V. Semkova et al. Nucl Phys A730(2004)255)

Noticed deficiencies in the IRDFF evaluations.



1. ¹⁸¹Ta(n,γ) - thousands times underestimation at 14 MeV was "discovered" in Nov 2015:

Existing Experimental total 181 Ta (n,γ) 182 Ta cross sections at 14.5 MeV

 $= 1.0 \pm 0.2$ mb (C. Necheva, I. Phys. G: Nucl. Pan. Phys. 20(1994)L33)

 $= 1.13 \pm 0.17$ mb (M. Budnar, INDC(YUG)-6,1979)

Current source of the ¹⁸¹Ta(n,γ) evaluation in IRDFF is IRDF-2002, which in turn has adopted the JENDL/D-99 evaluation!

Detailed assessment of the dosimetry reactions status and possibility of further updating or new evaluations.

I. IRDFF reactions which need further update.

- 23 Na(n, γ) 24 Na, 23 Na(n,2n) 22 Na, 181 Ta(n, γ) 182 Ta and 63 Cu(n, γ) 64 Cu reactions excitation functions and related uncertainties **need re-evaluation**.

The 23 Na(n, γ) 24 Na reaction excitation function in the current IRDFF version was taken from the ENDF/B-VII.1 library. The cross section in ENDF/B-VII.1 was calculated in the energy range 1.000E-05 eV - 600 eV using a Breit-Wigner shape and parameters for the first resonance: $E_r =$ 2.81 keV, $\Gamma_n = 376$ eV, and $\Gamma_v = 0.353$ eV. In the energy range 600 eV - 500 keV the capture cross section is described by single level Breit-Wigner resonance parameters. From 500 keV to 20 MeV the capture cross section is the same as in ENDF/B-IV file.

As it evident the evaluation for the ${}^{23}Na(n,\gamma){}^{24}Na$ reaction included in the IRDFF v-1.03 library does not take into account the results of new measurements of Firestone et al. at 0.0253 eV [1] and experimental information obtained after 1975 years.

The 23 Na(n,2n) 22 Na reaction excitation function was adopted from the JENDL/D-99 library.

JENDL/D-99 evaluation was carried out in August 1996 and does not take into account the later measurements [2-4].

The 181 Ta (n,γ) 182 Ta reaction data for IRDFF v-1.03 were taken from JENDL-3.2. The 181 Ta(n, γ) 182 Ta reaction excitation function presented in JENDL-3.2 library was evaluated by N. Yamamuro in March 1987. In the RR region 1.00E-05 eV – 1 keV the 181 Ta(n, γ) cross-section was described by MLBW resonance parameters. In the energy interval 1 - 100 keV cross-section are is represented by unresolved parameters.

The evaluation carried out for ${}^{181}Ta(n,\gamma){}^{182}Ta$ reaction in JENDL-3.2 library does not take into account the measurements [5-13], which provide additional experimental information in the energy interval 1.97 keV – 3 MeV and at 14 MeV. Uncertainty in the JENDL-3.2 capture cross section for ¹⁸¹Ta(n,y) in the energies range 1.000E-05 eV – 20 MeV are given by simple LB = 1 matrix. The ¹⁸¹ $Ta(n, \gamma)^{182}Ta$ reaction excitation function presented in JENDL-3.2 library significantly underestimates the cross sections in the energy range above 7 MeV. At 14.5 MeV point the evaluated capture cross section is 6003 times lower than equivalent experimental data of *C. Necheva et al.* [10].

II. New dosimetry reactions for evaluation and inclusion in IRDFF

II.1. Fission Reactors

The participants of the 1-st RCM on "Testing and Improving the International Reactor Dosimetry and Fusion File (IRDFF)" elaborated a list of the new perspective reactions which are interesting for the dosimetry applications:

- 27 Al(n, γ)²⁸Al often present in facilities, short-lived (2.24 min);
- 94 Zr(n, γ) 95 Zr and 96 Zr(n, γ) 97 Zr first resonance at high/low (2.3/0.3 keV) energies; _
- 70 Zn(n, γ)⁷¹Zn first resonance at energy above 10 keV but low abundance (0.62%);
- ¹¹⁷Sn(n,n')^{117m}Sn low threshold (0.3 MeV), convenient $T_{1/2} = 14$ days and γ -energy 314 keV; ⁹³Nb(n, γ)^{94g+m}Nb and ⁹⁴Nb(n, γ)^{94m}Nb for burn up calculations;
- 93m Nb(n, γ)⁹⁴Nb due to 300 keV threshold is interesting for the fast flux measurements

- 113 In(n, γ) 114m In – perspective reaction for reactor dosimetry due to the appropriate half-life 49.51 ± 0.01 days and lonely decay line 190 keV with intensity 15.56 ± 0.15%. Its rate, measured using the nat In activation detector, will give additional information for 115 In(n, γ) 116m In.

After RCM-1 the excitation function of the 113 In $(n,\gamma)^{113m}$ In reaction was re-evaluated in the energy range 1.000E-05 eV – 20 MeV [18] and included in the IRDFF-1.05.

The results of new evaluation of the 94 Nb(n, γ) 94m Nb reaction excitation function in the energy range 1.000E-05 eV – 20 MeV were reported at the 1-st RCM. Evaluated data file has been included in IRDFF-1.03.

Comments on the ¹¹⁷Sn(n,n')^{117m}Sn reaction. It is attractive for dosimetry application due to low threshold 317 keV and convenient decay parameters: $T_{1/2} = (14.00 \pm 0.05)$ days and two gammalines 156.02 keV (I $\gamma = 2.113\%$) and 158.56-keV (I $\gamma = 86.4\%$), which can be used for the reaction rate measurement. Usage of the activation detectors made of natural tin does not allow to measure correctly the ¹¹⁷Sn(n,n')^{117m}Sn reaction rate due to processes ¹¹⁸Sn(n,2n)^{117m}Sn, ¹¹⁸Sn(γ ,n)^{117m}Sn, ¹¹⁹Sn(n,3n)^{117m}Sn leading to production of the same isomer ^{117m}Sn. The abundance of ¹¹⁷Sn, ¹¹⁷Sn, ¹¹⁷Sn in natural tin is equal (7.68 ± 0.07) At.%, (24.22 ± 0.09) At.%, and (8.59 ± 0.047) At.%, respectively. This negative impact can be bypassed by using the high enriched ¹¹⁷Sn samples. The experimental information about the ¹¹⁷Sn(n,n')^{117m}Sn reaction cross section is scarce and covered neutron energies range 0.524 – 15.9 MeV. Regrettable the integral experimental data, which can be used for testing of the ¹¹⁷Sn(n,n')^{117m}Sn evaluated reaction excitation function are absent today.

All this currently does not allows an evaluation of the ${}^{117}Sn(n,n'){}^{117m}Sn$ reaction cross section for dosimetry application.

Request from RCM-2 (March 2015):

There is an interest for the ¹¹⁷Sn(n,n')^{117m}Sn reaction because of its unique characteristics combination: $E_{Threshold} = 0.314$ MeV, $T_{1/2} = 14$ d, $E_{g1} = 158$ keV ($I_{g1} = 86\%$). Experimental irradiations of this dosimeter have been done with enriched Tin (93% at. ¹¹⁷Sn) in different spectra. However, nuclear data need additional measurements (see *IRDFF HPRL*) and upgrades (lack of uncertainties, discrepancies between library evaluations) to allow this reaction to be used.

- ${}^{37}Cl(n,\gamma){}^{38}Cl$ and ${}^{127}I(n,\gamma){}^{128}I$ reactions are useful for the reactor dosimetry application and can be **added** to IRDFF.

The usage of the NaCl sample makes possible the measurement of two reactions simultaneously: ${}^{23}\text{Na}(n,\gamma){}^{24}\text{Na}$ and ${}^{37}\text{Cl}(n,\gamma){}^{38}\text{Cl}$. On other hand, the NaI sample gives an option to measure simultaneously the ${}^{23}\text{Na}(n,\gamma){}^{24}\text{Na}$ and ${}^{127}\text{I}(n,\gamma){}^{128}\text{I}$ reaction rates. It is also worthwhile to note that in several neutron radiative capture cross-sections experiments, the ${}^{127}\text{I}(n,\gamma){}^{128}\text{I}$ reaction was used as standard in energy interval 10 keV – 1 MeV.

The isotopic abundance of ³⁷Cl in natural chlorine is (24.22 ± 0.0010) atom percent [14]. ³⁸Cl produced in the (n,γ) reaction undergoes 100% β^{-} decay mode with a half-life of (37.230 ± 0.014) minutes. The ³⁷Cl $(n,\gamma)^{38}$ Cl reaction rate can be measured by detecting 1642.43 keV ($I_{\gamma} = 0.333 \pm 0.007$) and 2167.54 keV ($I_{\gamma} = 0.444 \pm 0.000$) gamma radiations. The ³⁷Cl $(n,\gamma)^{38}$ Cl reaction rate can be measured also by detecting β^{-} particles with end-point energies 1106.3 keV ($I_{\beta} = 0.331 \pm 0.008$),

2748.9 keV ($I_{\beta} = 0.113 \pm 0.005$) and 4916.5-keV ($I_{\beta} = 0.556 \pm 0.009$). Recommended decay data for ³⁸Cl (half-life, gamma and β ⁻ emission probabilities per decay of ³⁸Cl) were taken from Ref. [15].

The isotopic abundance of ¹²⁷I in natural iodine is 100 atom percent [14]. ¹²⁸I produced in the (n,γ) reaction undergoes two decay modes with a half-life of (24.99 ± 0.02) minutes. The $(93.1 \pm 0.8)\%$ of disintegrations occurs by β^{-} decay mode and $(6.9 \pm 0.8)\%$ of disintegrations via the ϵ -capture mode. The ¹²⁷I $(n,\gamma)^{128}$ I reaction rate can be measured by detecting 442.90 keV ($I_{\gamma} = 0.169 \pm 0.017$) and 526.56 keV ($I_{\gamma} = 0.0159 \pm 0.0017$) gamma radiations. Recommended decay data for ¹²⁸I (half-life and gamma emission probability per decay of ¹²⁸I) were taken from Ref. [16].

II.2. Fusion Reactors

- ²⁸Si(n,p)²⁸Al – an appropriate activation reaction for the 14 MeV neutron flux monitoring during planned fusion experiments at JET and ITER

Evaluation of this and contributing ${}^{29}Si(n,x){}^{28}Al$ reactions was done (see K.I Zolotarev INDC(NDS)-0668) and included in IRDFF-1.05.

- ${}^{64}Zn(n,2n){}^{63}Zn$, ${}^{90}Zr(n,p){}^{90}Y$, ${}^{186}W(n,2n){}^{185}W$, ${}^{197}Au(n,p){}^{197}Pt$ – are sensitive to 14 MeV neutrons and look suitable for the fusion applications.

Reaction ⁶⁴Zn(n,2n)⁶³Zn can be used as an activation detector. The ⁶³Zn produced in the (n,2n) reaction undergoes 100% ε -capture mode with a half-life of (38.47 ± 0.05) minutes. The ⁶⁴Zn(n,2n)⁶³Zn reaction rate can be measured by detecting 511 keV (I_{γ} = 1.855 ± 0.017), 669.62 keV (I_{γ} = 0.082 ± 0.000), and 962.06 keV (I_{γ} = 0.065 ± 0.004) gamma radiations. Decay data for ⁶³Zn (half-life and gamma emission probability per decay of ⁶³Zn) were taken from Ref. [19].

Regarding amount of experimental data available in EXFOR the ${}^{64}Zn(n,2n){}^{63}Zn$ reaction excitation function can be evaluated for dosimetry application in the energy range from threshold to 40 MeV.

Reactions 90 **Zr**(**n**,**p**) 90m **Y** and 90 **Zr**(**n**,**p**) ${}^{90m+g}$ **Y** can also be used for dosimetry application. The isomeric state of 90m **Y** undergoes 100% IT mode with a half-life of (3.19 ± 0.06) hours [20]. Isomeric transition is accompanying by emission of 202.53-keV and 497.51-keV gamma-rays with intensities I_{γ1} = (97.3 ± 0.4)% and I_{γ2} = (90.74 ± 0.05)% [20]. The 90g **Y** undergoes 100% β⁻ decay mode with a half-life of (64.053 ± 0.020) hours [20]. The 90 Zr(n,p) ${}^{90m+g}$ **Y** reaction rate can be measured only by detecting β⁻ particles with end-point energy 2280.1-keV (I_β = 99.9885 ± 0.0014)% [20].

On the basis of experimental data given in EXFOR the ${}^{90}Zr(n,p){}^{90m}Y$ and ${}^{90}Zr(n,p){}^{90m+g}Y$ reaction excitation functions can be evaluated for dosimetry application in the energy range from threshold to 20-21 MeV.

Reaction ¹⁸⁶W(n,2n)^{185m+g}W can be used as an activation detector. The isomeric state of ^{185m}W decays via 100% IT transition with a half-life of (1.63 ± 0.03) min [20]. The ^{185g}W undergoes 100% β^{-} decay with a half-life of (75.1 ± 0.3) days [21]. The ¹⁸⁶W(n,2n)^{185m+g}W reaction rate can be measured only by detecting β^{-} particles with end-point energy 432.5 keV (I_β = 99.928 ± 0.008)% [21].

Experimental data for the ${}^{186}W(n,2n){}^{185m+g}W$ reaction cross section cover the energies range only from 7.41 to 14.8 MeV, that make difficult its evaluation.

²⁷Al(n,2n)²⁶Al. is perspective reaction for dosimetry in fusion applications as a long term fluence monitor.
Moreover it proved to be measured by the AMS technique [23] (other recommended reactions are ⁵⁶Fe(n,2n)⁵⁵Fe, ²³⁸U(n,3n)²³⁶U) and thus can serve for validation of the ²⁵²Cf(s.f.) and ²³⁵U(n,f) prompt fission spectra above reaction threshold 13.55 MeV (see also: http://www-nds.iaea.org/IRDFFtest/Cf252U235_HighThreshold.pdf).

The residual nucleus ²⁶Al undergoes 100% decay via the ε -capture mode with a half-life of 717000 years [17]. The ²⁷Al(n,2n)²⁶Al reaction rate can be measured by detecting 511 keV (I_{γ} = 163.5 ± 0.4)% and 1808.65 keV (I_{γ} = 99.76 ± 0.04)% gamma radiations. The ²⁷Al(n,2n)²⁶Al reaction rate can be measured also by detecting β ⁻ particles with end-point energy 1173.42 keV (I_{β} = 81.73 ± 0.21)%.

¹⁹⁷Au(n,p)^{197m}Pt and ¹⁹⁷Au(n,p)^{197m+g}Pt reactions are suitable for activation measurements in the Fusion facilities.

The isomeric state of ^{197m}Pt decays with a half-life of (95.41 ± 0.18) minutes by two modes [22]. The (95.41 ± 0.18)% of disintegrations occurs by the IT transition. The ¹⁹⁷Au(n,p)^{197m}Pt reaction rate can be measured by detecting of 346.5 keV gamma radiation ($I_{\gamma} = 11.1 \pm 0.3$)% [22] and X-rays. The ground state of ^{197g}Pt undergoes 100% β⁻ decay mode with a half-life of (19.8915 ± 0.0019) hours [21]. The ¹⁹⁷Au(n,p)^{197m+g}Pt reaction rate can be measured by detecting 77.35 keV ($I_{\gamma} = 17.0 \pm 2.3\%$) and 191.437 keV ($I_{\gamma} = 37\%$) gamma radiations. Uncertainty of intensity for the second gamma-line is not determined.

The fusion community have also interest to the following reactions planned to be used as detectors in TBMs of ITER:

¹⁴⁰Ce(n,2n)^{139m}Ce; ¹⁴⁰Ce(n, α)^{137m}Ba, ²⁷Al(n, γ)²⁸Al, ²⁷Al(n,p)²⁷Mg, ⁵²Cr(n,p)⁵²V, ⁵³Cr(n,p)⁵³V, ⁵⁴Cr(n,p)⁵⁴V, ⁵⁴Cr(n, α)⁵¹Ti, ⁹³Nb(n,g)^{94m}Nb, ⁹³Nb(n, α)^{90m}Y, ⁹³Nb(n, $n\alpha$)^{89m}Y and ⁹³Nb(n,2n)^{92m}Nb. Evaluated data for the ²⁷Al(n,p)²⁷Mg, ⁹³Nb(n, γ)^{94m}Nb and ⁹³Nb(n,2n)^{92m}Nb reactions are already available in the IRDFF v-1.03 library.

The ${}^{93}Nb(n,\alpha){}^{90m}Y$ reaction can be also appropriate for the neutron dosimetry in fission reactors.

The ²⁷Al(n,γ)²⁸Al, ⁵²Cr(n,p)⁵²V, ⁵³Cr(n,p)⁵³V, ⁵⁴Cr(n,p)⁵⁴V and ⁵⁴Cr(n,α)⁵¹Ti reactions, which leads to production short-lived residual nucleus, could be re-evaluated for ITER application. Regrettably the integral experimental data which could be used for the testing of the evaluated ¹⁴⁰Ce(n,α)^{137m}Ba reaction excitation functions are not available nowadays.

The lack of such experimental information currently not allows an evaluation of the ${}^{140}Ce(n,2n)^{139m}Ce$ and ${}^{140}Ce(n,\alpha)^{137m}Ba$ reaction cross sections for ITER dosimetry application.

The same can be said about ${}^{93}Nb(n,n\alpha)^{89m}Y$ reaction.

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