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Irradiation damages in tantalum spallation target

The irradiation by a high intensity proton beam of the spallation target in Accelerator Driven Systems (ADS) gives rise to structural damages that must be considered in the design and handling of such materials. In this work, some of these damages are evaluated in the case of the irradiation of a tantalum target. Tantalum was recently proposed as solid target in the case of low power systems. Unfortunately, not enough experimental data of spallation reactions in this material are available. Therefore, our calculations were made using MCNPX and the stand-alone version of the spallation codes INCL4 + ABLA. Results of this work are the concentrations of spallation residues produced in the target after one year of irradiation and calculations of the activity derived. Also displacements per atom in the target material have been evaluated. Different components (elastic scattering and non-elastic reactions) of the displacement cross-sections were calculated with two different methods.

Strahlenschäden in einem Spallationstarget aus Tantal. *Die Bestrahlung eines Spallationstargets in einem Beschleunigergetriebenen System (ADS) mit einem Protonenstrahl hoher Intensität führt zu Strukturschäden welche beim Entwurf und bei der Handhabe berücksichtigt werden müssen. In der vorliegende Arbeit werden einige dieser Schäden untersucht für den Fall einer Bestrahlung eines Targets aus Tantal. Tantal war vor einigen Jahren vorgeschlagen worden als Feststoff Target in ADS mit niedriger Leistung. Leider sind für dieses Material nur ungenügende experimentelle Daten verfügbar. Deshalb wurden Simulationsrechnungen durchgeführt mit dem Monte Carlo Code MCNPX und mit der Stand-alone Version des Spallations-Codes INCL4 + ABLA. Die Ergebnisse dieser Untersuchungen sind die Konzentrationen der Spallationsprodukte nach einem Jahr der Bestrahlung und Berechnung der resultierenden zeitabhängigen Radioaktivität. Weiter wurden „Displacements per Atom" (DPA) im Targetmaterial ermittelt. Dazu wurden verschiedene Komponenten (elastische Streuung und nicht-elastische Reaktionen) des DPA Querschnitts mit zwei verschiedenen Methoden bestimmt.*

1 Introduction

Tantalum has recently been proposed as target material for the TRADE project. This project was a common international initiative aiming to validate the coupling of an accelerator and spallation target to the existing 1 MW TRIGA reactor of the ENEA-Casaccia Centre [1, 2].

Due to the low power of the reactor, a solid target cooled by water was chosen. The choice of the material of the spallation target is based on different criteria as the neutron yield, the final activation, the thermal conductivity, mechanical properties, behavior under strong temperature constraints, corrosion etc.

In the case of tantalum chosen as target material, not many experimental data are available for predicting some of the consequences due to the proton beam irradiation. In this work, we present calculations concerning some of these problems: the concentration of spallation residues produced in the target after one year of irradiation that can give rise to corrosion or embrittlement problems, the activation of the target during and after irradiation and the displacements per atom (dpa) created by the spallation residues.

2 The spallation residues in the target

We considered in this work the spallation target isolated (no sub-critical reactor core is present) and being irradiated by a proton beam of 0.14 mA, at three different energies: 140, 300 and 1 000 MeV. The final concentration of a given residue in the target is the result of the creation rate by spallation reactions modified by the radioactive decay during the irradiation time and the activation created by the spallation neutrons. These factors have been calculated by combining different codes.

2.1 Residues created by proton irradiation

In order to calculate the spallation rate, version 2.5 of MCNPX has been used. MCNPX allows evaluating the interaction probability between the proton and the tantalum target taking into account the elastic scattering and the shape and thickness of the irradiated target (for details see [15]).

For these calculations we have chosen realistic irradiation conditions proposed in the TRADE experiment. The geometry of the target used for the MCNPX calculation is represented in Fig. 1 and it corresponds to measures given in different solutions for the TRADE target [3]. The beam tube has an outer diameter of 37.3 mm and an inner diameter of 28.22 mm. The tantalum target has a conical shape with an active part of 370 mm. This sharp cone angle (4.36°) is intended to maximize the axial distribution of neutron production and minimize the global and local power densities of the target.

The proton beam has a Gaussian shape with $\sigma_x = \sigma_y =$ 5.5 mm and an intensity of 0.14 mA. The energy considered in the TRADE experiment was, in a first time, 140 MeV and then, in further considerations 300 MeV. As mentioned, we will present here results at both energies and the same intensity of the proton beam, in order to compare damages created by the two different beams. Also the spallation residues for 1 GeV proton have been evaluated as this is the reference energy in most of the ADS project.

In MCNPX, the incident proton and all the particles produced in the reactions are transported through the target. When spallation reaction between one of the particles and a

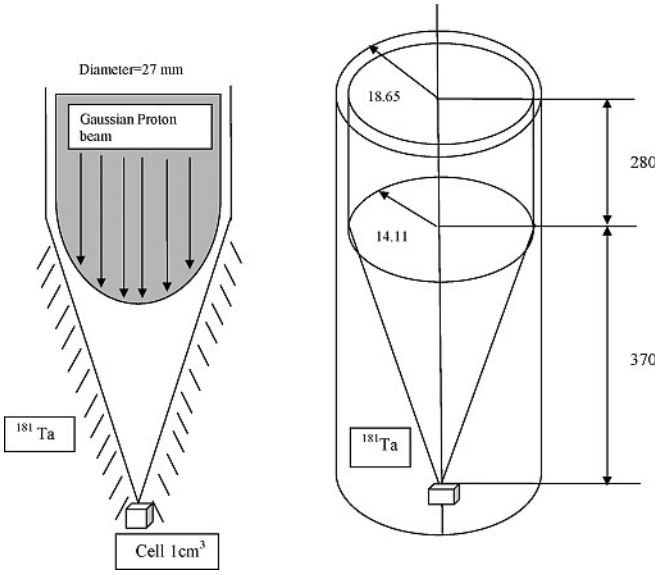

Fig. 1. Schematic design of the conical tantalum target used in the MCNPX input file. Measures are taken from the TRADE project and are given in mm. The proton beam has a Gaussian shape with $\sigma_x = \sigma_y = 5.5$ *mm and 0.14 mA of intensity*

target atom can take place, it is simulated by one of the different codes available in MCNPX for the description of the spallation reaction. Those spallation reactions are described by two different phases, each of them modeled by different codes that are used coupled: the intranuclear cascade and the de-excitation of the pre-fragment. In this work two intranuclear cascade codes were used: the Bertini [4] code and the new version of the Liege Intra Nuclear Cascade Code INCL4 [5] coupled with two different de-excitation models: Dresner code [6] and ABLA code [7] respectively. Both intranuclear cascade and de-excitation codes are available in the new version of MCNPX 2.5.e (INCL4/ABLA has been recently introduced).

The Bertini intranuclear cascade code coupled with the Dresner evaporation code is one of the most ancient Monte Carlo codes for the description of the spallation reactions and although it has been shown important deficiencies in the spallation products predictions it is still one of the most used in the application area. INCL4 + ABLA, on the other hand, are new codes still under development being compared to new experimental data. They have been proved to reproduce quite well recent data concerning residual nuclei of spallation reactions on Pb and Au [5].

Unfortunately, not many experimental data concerning the spallation residues production cross-sections on tantalum are available. Therefore we cannot present here a comparison of the prediction of these two codes with experimental values of spallation products in ^{181}Ta.

2.2 *Evolution of the concentrations during the irradiation time*

As it was mentioned above, during the irradiation time the concentration of the different residues created by spallation reactions in the target changes due to their radioactive decay.

The evolution of the concentration of a given element in the target during the irradiation time can be written as:

$$\frac{dNi}{dt} = -\lambda_i N_i + \sum_{j\neq i} a_{ji}\,\lambda_{j\to i}\,N_j + F_i - \langle\sigma_{i\to j}\rangle\,\phi_{tot}\,N_i + \sum_{j\neq i}\beta_{ji}\langle\sigma_{j\to i}\rangle\phi_{tot}\,N_j \quad (1)$$

where the first term corresponds to the radioactive decay of the element N_i with decay constant λ_i, the second term increases the concentration with all other residues which decay into N_i, the third term represents the feed of a given element by the spallation reaction on tantalum (we use here data obtained with MCNPX for protons and neutrons with energy above 20 MeV), fourth term describes the decrease of the N_i element due to the activation by neutrons with energy below 20 MeV and fifth term the contribution of other elements transformed into N_i by the same procedure.

The most important contribution to the time evolution of the concentration of a given residue in the tantalum target is given by the first three terms in Eq. (1). These contributions have been calculated for three different cases: a proton beam of 140, 300 and 1 000 MeV with two different programs ORIHET3 (that can only calculate the evolution due to these three terms) and KAPROS (that can also take into account the activation due to the spallation neutrons).

ORIHET3 is an adaptation of the Oak Ridge Isotope Generation and depletion code ORIGEN designed to study the buildup and decay of activity in any system for which the nuclide production rates are known.

In ORIHET3 two decay data libraries are provided with the code, the original ORIHET library and NUBASEX [8] derived from the NUBASE [9] evaluated compilation. For our calculations, the NUBASEX library was used. This library has data for 3 738 nuclides and uses more than 40 decay modes including decay to isomeric states of the daughter. Major details about the differences between NUBASEX and NUBASE libraries are given in Ref. [9].

The KAPROS code (Karlsruhe program system) [10] is a modular system composed by a "kernel" and different modules that can be used in free order to perform nuclear calculations as:

- Determination of atomic number densities for material compositions,
- Calculation of multigroup cross-sections,
- Neutron Diffusion calculations,
- Neutron Transport calculations,
- Reactor kinetics parameters, based on perturbation theory,

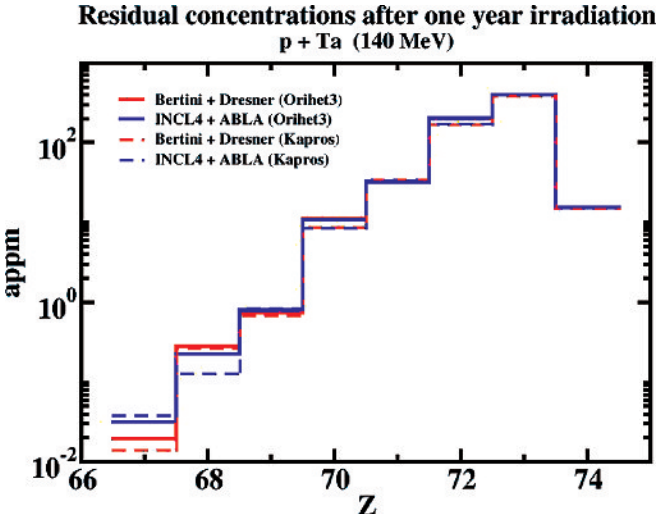

Fig. 2. Concentration in appm (atoms of impurity per million atoms in the target) of chemical impurities in the ^{181}*Ta target after one year of irradiation by a 140 MeV proton beam of 0.14 mA of current intensity. Results are calculated using ORIHET3 depletion code (solid line) and KAPROS code (doted line)*

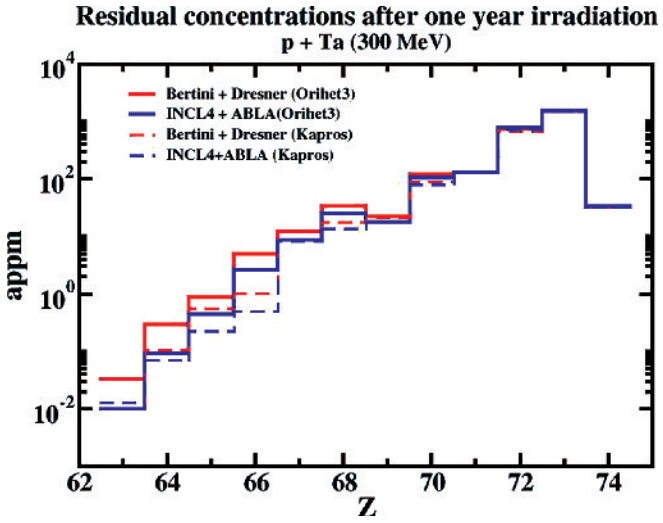

Fig. 3. Concentration in appm (atoms of impurity per million atoms in the target) of chemical impurities in the ^{181}Ta target after one year of irradiation by a 300 MeV proton beam of 0.14 mA of current intensity Results are calculated using ORIHET3 depletion code (solid line) and KAPROS code (doted line)

– Burn up and depletion calculations,
– Etc.

In this case, the KAPROS code was used for the calculation of the evolution of the different residual nuclei concentrations in order to compare results obtained with ORIHET3. For this purpose, three different libraries were used containing decay data for light elements, heavy isotopes and fission products. Existing files for fission reactor burn-up are based on the libraries used by the stand-alone code KORIGEN [17]. For spallation target applications a special library was created based on the NUBASE [9] file to replace the original file for light elements. For each element the information concerning their half life time and their different radioactive decay modes are recorded.

The evolution of the concentrations has been calculated dividing the total irradiation time (365 days in this case) in 100 irradiation time steps (each of 3.65 days) and "feeding" (F_i) the concentrations with their spallation production (calculated with MCNPX) each irradiation step.

A comparison of results obtained with ORIHET3 (solid lines) and KAPROS (dotted lines) for the three energies of the proton beam are given in Figs. 2, 3 and 4 (140, 300 and 1 000 MeV respectively). These figures show the concentration of a given chemical element after one year of irradiation. Values of the concentrations of the residual nuclei in the target at the three energies of the proton beam are shown in Tables 1, 2 and 3.

This comparison between the two evolution codes shows excellent agreement for the residues close to tantalum, which are the most important produced elements. The agreement is generally very good (less than 30 % difference) for all the elements except for some particular cases (Z = 68, 66). In these cases the difference can arise from the fact that the most important contributors to these concentrations are isotopes generated by a radioactive decay chain involving very short lived isotopes (short compared to the total irradiation time, 1 year). Indeed, the time steps defined in the KAPROS calculation for the evolution of concentrations must be adapted to the time of life of the residue we are interested on calculating its concentration. Therefore, it is not possible to obtain good results for the final concentration of isotopes with very long and very short life time in the same calculation with the same time step definition.

For each of the two evolution codes ORIHET3 and KAPROS, the calculations have been made with the two spallation codes mentioned above: INCL4 + ABLA (blue lines in figures) and Bertini + Dresner (red lines in figures). Residues produced at 1 GeV of the proton beam show differences between the two spallation codes that are on the order of 30 % in the case of the fission products and that, for evaporation residues with charge number $50 < Z < 60$ can achieve a factor of 40. For evaporation products with a charge close to the charge of tantalum (which are the only elements produced for 140 and 300 MeV), the difference between the two spallation physical models is less than a factor of 2. In all the cases, the differences between the two spallation codes are more important than the differences between the two different evolution codes ORIHET3 and KAPROS.

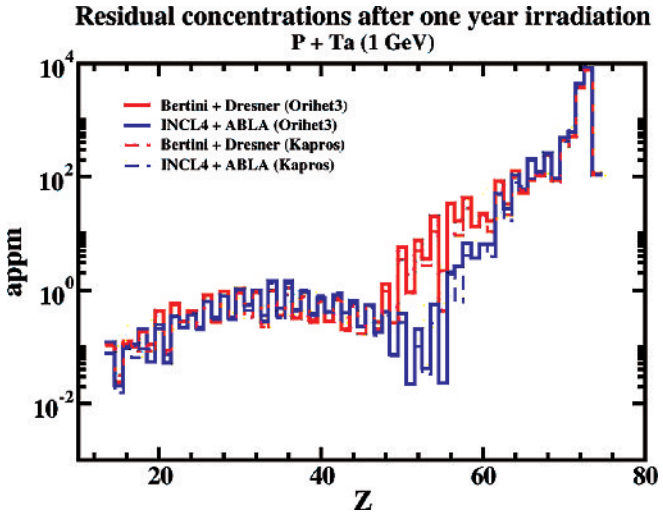

Fig. 4. Concentration in appm (atoms of impurity per million atoms in the target) of chemical impurities in the ^{181}Ta target after one year of irradiation by a 1 GeV proton beam of 0.14 mA of current intensity results are calculated using ORIHET3 depletion code (solid line) and KAPROS code (doted line)

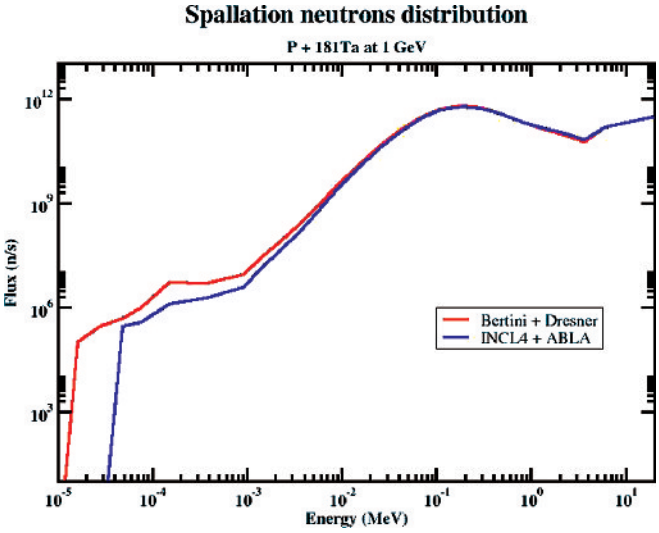

Fig. 5. Mean neutron flux with E < 20 MeV in the tantalum target when bombarding by a 1 GeV proton beam of 0.14 mA of intensity. The flux has been divided in 69 energy groups (0–20 MeV)

Table 1. Concentrations of residual nuclei in the target at 140 MeV [appm]

Element	ORIHET3 (Bertini + Dresner)	ORIHET3 (INCL4 + ABLA)	KAPROS (Bertini + Dresner)	KAPROS (INCL4 + ABLA)
Ho Z = 67	1.98E-2	3.17E-2	1.435E-2	3.83E-2
Er Z = 68	0.28	0.23	0.27	0.13
Tm Z = 69	0.74	0.81	0.68	0.84
Yb Z = 70	11.24	10.90	8.83	8.48
Lu Z = 71	33.02	31.95	34.4	33.42
Hf Z = 72	200.81	205.26	168.94	174.39
Ta Z = 73	399.94	400.87	393.38	396.65
W Z = 74	15.55	15.47	14.85	15.24

Table 2. Concentrations of residual nuclei in the target at 300 MeV [appm]

Element	ORIHET3 (Bertini + Dresner)	ORIHET3 (INCL4 + ABLA)	KAPROS (Bertini + Dresner)	KAPROS (INCL4 + ABLA)
Eu Z = 63	3.28E-2	1.00E-2	1.28E-2	–
Gd Z = 64	0.30	9.14E-2	0.11	7.09E-2
Tb Z = 65	0.89	0.44	0.55	0.22
Dy Z = 66	5.00	2.64	1.01	0.51
Ho Z = 67	12.29	8.65	12.06	8.41
Er Z = 68	34.40	24.92	17.83	13.83
Tm Z = 69	22.05	18.00	21.38	17.57
Yb Z = 70	120.73	105.65	88.93	79.37
Lu Z = 71	128.75	130.77	131.92	132.74
Hf Z = 72	744.08	807.65	686.99	751.72
Ta Z = 73	1 518.26	1 545.43	1 498.21	1 521.58
W Z = 74	32.84	33.72	32.53	32.94

One of the main advantages of the KAPROS procedure is that it allows taking into account the activation due to neutrons in the target (last terms in Eq. (1). In MCNPX calculations for thick targets, residues are created by incident protons and also by all the protons and neutrons with energy above 20 MeV, resulting from spallation reactions. The mean flux of neutrons with energy below 20 MeV created by the proton irradiation in the target has been computed and divided in 69 energy groups to be treated by the program KAPROS in order to calculate their contribution to the total concentration of a given residue. The value of the mean flux in these 69 energy groups is shown in Fig. 5 in the case of an incident proton beam of 1 GeV and 0.14 mA of intensity (for the others two energies, the values of the flux is less important). The calculations are performed with the two codes: INCL4 + ABLA and Bertini + Dresner.

In the particular case treated in this work, the tantalum target is only being irradiated by the proton beam and it is not placed within a sub-critical core, therefore, the spectrum of neutrons presented in Fig. 5 represents an spallation neutron spectrum with an important component of fast neutrons. The total flux of spallation neutrons is $6 \cdot 10^{12}$ n/s and its influence in the different residual concentrations is always less than 1 %, so, negligible in our case. As already mentioned, neutron contribution is negligible in this case but it is not at all in the case of a spallation target within a subcritical core with flux around $10^{14} – 10^{15}$ n/s.

3 The activation of the target

Some of the residues created by spallation reactions are radioactive products that, during and after the irradiation, generate an activity of the target material.

We have calculated this activation (using ORIHET3) during one year of irradiation and the cooling after this irradiation: 1 min, 1 hour, 1 day, 1 month, 1 year, 10 years, 10^2 years, 10^3 years, 10^4 years and 10^5 years. Calculations corresponding to the three energies of the proton beam (140, 300 and 1 000 MeV) are shown in Fig. 6.

The left graphics on this figure show the results of the activity in the spallation target after one year of irradiation calculated with the two different codes (Bertini + Dresner and INCL4 + ABLA). In the right part of the figure, the cooling is presented. ^{178}Ta is the most important radioactive isotope responsible of the activation of the target during the irradiation time at all energies. For the cooling time, different isotopes are responsible of the activation of the target at each time step. All these important radioactive isotopes are detailed in Table 4, where also their concentration after the irradiation is calculated.

As it can be seen from this figure, the difference between predictions of the two codes are negligible in the evaluation of the activation of the target and only little discrepancies are seen after very long time (10 years) of the cooling. This difference is also attenuated with the increase of the proton beam energy.

4 Displacements per atoms

The irradiation of the tantalum target by the proton beam gives rise to collision cascades between particles (incident protons or light particles created in the reactions) and the target atoms or even between residual nuclei created by spallation reactions and the tantalum atoms in the target. These collision cascades displace the tantalum target atoms of its original place in the crystal lattice being at the origin of fragilization and embrittlements problems in the irradiated material.

An evaluation of this irradiation problem can be made using the concept of displacement per atom (dpa). The number of dpa in the material is calculated from the total displacement cross-section σ_d:

$$\sigma_d = \sum_i \int_{E_d}^{T\max} \frac{d\sigma(Z_i, A_i, Z_t, A_t)}{dT_i} \cdot n_d(T_i, Z_i, A_i, Z_t, A_t)\, dT_i \qquad (2)$$

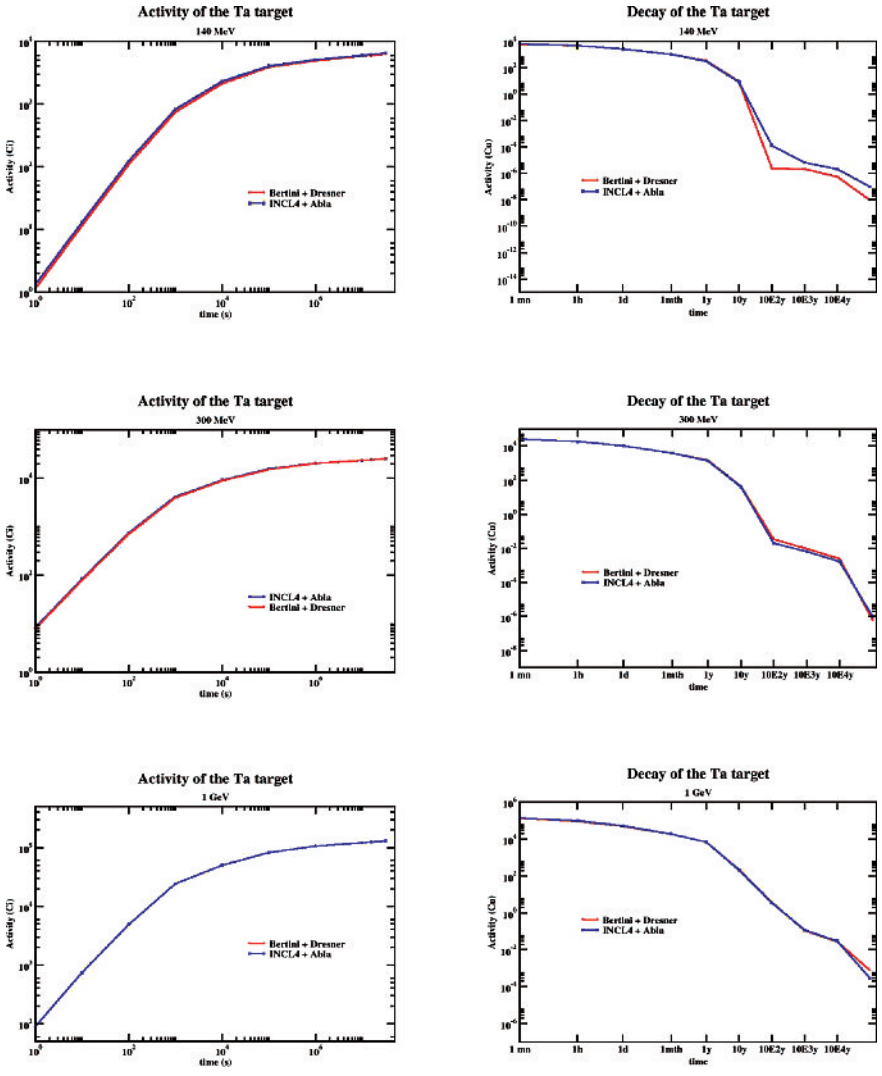

Fig. 6. Activation of the ^{181}Ta target irradiated by a Gaussian proton beam of 0.14 mA and 140, 300 and 1000 MeV, respectively. In left figures activation (in Ci) during one year of irradiation is shown using two different spallation codes for the evaluation of the residues: Bertini + Dresner and INCL4 + ABLA. The main isotope responsible of the activation of the target is ^{178}Ta. Right figures show the decay of the activation of the target as a function of the cooling timealso calculated with both codes. The most important contributors to the activation of the target at each time step of the cooling are detailed in Table 4

Where $ds_i(Z_i, A_i, Z_t, A_t)/dT_i$ is the energy spectrum of the i-th recoil atom (Z_i,A_i) produced in the interaction of the incident particle with the target atom (Z_t, A_t), and $n_d(T_i, Z_i, A_i, Z_t, A_t)$, the number of Frenkel pairs produced by this primary knock-on atom, (PKA) with energy T_i. The energy recoil spectrum extends between E_d which is the effective threshold displacement energy and $T_{\max}$, the maximal energy of the PKA spectrum. In the case of the tantalum target, E_d is taken equal to 90 eV.

Therefore, the calculation of the displacement cross-section is related to the calculation of the recoil spectra for the PKA production and the evaluation of the number of displacements that it produces. The nature of the PKA atom can be the same as that of the atoms composing the target if the reaction between the incident particle and the target is elastic or a different atom if the first reaction has given rise to a spallation residue.

The PKA dissipates its initial kinetic energy in two ways: some energy is lost by exciting the electrons of the medium and the other part can be used to make elastic collisions with atoms of the target. An atom with which the PKA collides sometimes receive enough kinetic energy to displace it from its lattice site and make similar collisions of its own giving rise to the cascade collisions.

A way to evaluate the number of defects (Frenkel pairs vacancy-interstitial) produced in an irradiated material was proposed in 1975 by Norgett, Robinson and Torrens [11–13]. This model allows evaluating the part of the kinetic energy of the PKA that is going to be used to create the cascade defaults T_{dam} if one takes into account looses due to the electronic stopping power.

Table 3. Concentrations of residual nuclei in the target at 1 GeV [appm]

Element	ORIHET3 (Bertini + Dresner)	ORIHET3 (INCL4 + ABLA)	KAPROS (Bertini + Dresner)	KAPROS (INCL4 + ABLA)
Si Z = 14	0.12	7.75E-2	0.11	0.12
P Z = 15	2.34E-2	2.07E-2	3.18E-2	1.55E-2
S Z = 16	0.11	0.10	0.13	9.42E-2
Cl Z = 17	0.10	0.11	8.59E-2	6.59E-2
Ar Z = 18	0.20	0.20	0.19	9.69E-2
K Z = 19	0.11	5.34E-2	8.61E-2	9.09E-2
Ca Z = 20	0.43	0.21	0.45	0.26
Sc Z = 21	7.53E-2	5.20E-2	8.99E-2	7.23E-2
Ti Z = 22	0.58	0.35	0.56	0.34
V Z = 23	0.28	0.22	0.28	0.22
Cr Z = 24	0.43	0.37	0.39	0.38
Mn Z = 25	0.22	0.22	0.21	0.20
Fe Z = 26	0.81	0.63	0.76	0.62
Co Z = 27	0.29	0.33	0.27	0.30
Ni Z = 28	0.95	0.78	0.97	0.84
Cu Z = 29	0.33	0.32	0.33	0.30
Zn Z = 30	1.05	1.04	0.91	1.01
Ga Z = 31	0.56	0.53	0.34	0.44
Ge Z = 32	0.93	1.00	0.98	0.96
As Z = 33	0.27	0.36	0.23	0.30
Se Z = 34	0.99	1.44	0.98	1.32
Br Z = 35	0.36	0.48	0.35	0.32
Kr Z = 36	1.10	1.41	0.85	1.32
Rb Z = 37	0.32	0.43	0.31	0.47
Sr Z = 38	0.90	0.95	0.78	1.04
Y Z = 39	0.30	0.38	0.27	0.35
Zr Z = 40	0.68	0.74	0.63	0.81
Nb Z = 41	0.28	0.41	0.29	0.40
Mo Z = 42	0.69	0.83	0.66	0.90
Tc Z = 43	0.20	0.40	0.20	0.29
Ru Z = 44	0.59	0.66	0.51	0.63
Rh Z = 45	0.23	0.23	0.18	0.22
Pd Z = 46	0.54	0.54	0.49	0.50
Ag Z = 47	0.27	0.18	0.21	0.16
Cd Z = 48	1.30	0.42	0.97	0.36
In Z = 49	0.26	7.42E-2	0.27	6.97E-2
Sn Z = 50	5.80	0.39	3.57	0.27

Table 3. (continued)

Element	ORIHET3 (Bertini + Dresner)	ORIHET3 (INCL4 + ABLA)	KAPROS (Bertini + Dresner)	KAPROS (INCL4 + ABLA)
Sb Z = 51	0.90	2.19E-2	1.96	9.97E-2
Te Z = 52	7.44	0.20	4.90	0.10
I Z = 53	3.48	4.15E-2	2.73	3.38E-2
Xe Z = 54	20.08	0.55	10.98	0.27
Cs Z = 55	0.43	2.31E-2	2.23	0.14
Ba Z = 56	33.34	2.11	28.26	1.94
La Z = 57	16.40	2.61	9.33	0.62
Ce Z = 58	41.69	6.63	28.02	4.05
Pr Z = 59	13.39	3.64	13.13	3.65
Nd Z = 60	22.06	6.41	21.68	6.50
Pm Z = 61	16.87	6.46	10.87	3.93
Sm Z = 62	84.84	48.52	60.82	31.83
Eu Z = 63	32.39	26.51	21.27	17.09
Gd Z = 64	126.03	105.52	98.87	80.11
Tb Z = 65	52.08	59.26	59.73	69.67
Dy Z = 66	197.57	207.73	87.75	95.19
Ho Z = 67	105.95	119.89	104.02	128.34
Er Z = 68	236.12	257.83	130.78	159.19
Tm Z = 69	85.78	92.70	82.87	97.07
Yb Z = 70	443.32	487.88	349.00	427.54
Lu Z = 71	509.83	611.53	513.04	665.43
Hf Z = 72	3 801.81	4 399.36	3 676.78	4 585.26
Ta Z = 73	7 813.56	8 360.159	7 696.99	8 857.31
W Z = 74	111.10	108.16	108.81	113.99

The damage energy, giving rise to displacements, can be calculated from the kinetic energy of the PKA as:

$$T_{dam} = T_i \cdot \xi \tag{3}$$

being ξ the damage efficacy that can be written as:

$$\xi = \frac{1}{1 + k_L \cdot (\varepsilon + 0.40244\,\varepsilon^{3/4} + 3.4008\,\varepsilon^{1/6})} \text{ with } \varepsilon = T_i/E_L \tag{4}$$

Where k_L and E_L are parameters introduced in the Lindhard's theory about the atomic collisions in solids [12]. Once the damage energy is calculated, the number of Frenkel pairs in the NRT model is evaluated with the following expression:

$$N_{NRT}(T) = \frac{0.8}{2E_d} T_{dam}(T) \tag{5}$$

In principle, the displacements in the target material can be created by the elastic reactions of protons and neutrons in the target and by the residues created in the spallation reactions. In the next section we have restricted our calculations to the dpa due to spallation residues created directly by the incident protons in the target using two different calculation methods: results given by MCNPX with different spallation codes and integrating the results evaluated with the INCL4 + ABLA stand alone version. An evaluation of the elastic reaction displacement cross-section created by protons and neutrons at energies from 10^{-5} eV up to 1 GeV can be found in references [14–15]. These calculations show that, above 50 MeV, the contribution to the total displacement cross-section (elastic + inelastic reactions) does not exceed 10–13 %.

4.1 Calculation of the inelastic displacement cross-section with MCNPX2.5.e

The MCNPX code allows the calculation of both the production cross-section of the spallation residues and their damage energy spectra needed to the evaluation of the damage cross-sections. These two data can be calculated using different in-

Table 4. Most important elements contributing to the activation of the ^{181}Ta target during the cooling time after one year of irradiation by a Gaussian proton beam of 140, 300 and 1 000 MeV respectively calculated with Bertini + Dresner (B + D) and INCL4 + ABLA (I + A) models. In columns 4–9, the concentrations of the elements at the end of the irradiation are given

Element	Half life	Decay	[appm] 140 MeV (B + D)	[appm] 140 MeV (I + A)	[appm] 300 MeV (B + D)	[appm] 300 MeV (I + A)	[appm] 1 GeV (B + D)	[appm] 1 GeV (I + A)
^{179}Ta	1.82 y	EC	44.7	42.0	170.14	159.10	1 019.0	948.4
^{178}Ta	9.31 mn	EC	$1.16 \cdot 10^{-3}$	$1.3 \cdot 10^{-3}$	$3.8 \cdot 10^{-3}$	$4.7 \cdot 10^{-3}$	0.02	0.03
^{177}Ta	56.56 h	EC	0.35	0.37	1.09	1.24	5.30	6.71
^{175}Hf	70 d	EC	7.47	7.42	23.90	25.34	—	–
^{163}Ho	$4.5 \cdot 10^{3}$ y	EC	$9.7 \cdot 10^{-4}$	$2.6 \cdot 10^{-3}$	4.25	2.95	37.83	54.84
^{157}Tb	71 y	EC	–	–	0.39	0.18	31.67	35.84
^{154}Dy	$3 \cdot 10^{6}$ y	α	–	–	0.08	–	–	–
^{137}La	$6 \cdot 10^{4}$ y	EC	–	–	–	–	9.36	1.47
^{99}Tc	$2.1 \cdot 10^{5}$ y	β-	$2.4 \cdot 10^{-4}$	–	–	–	–	–
^{94}Nb	$2.0 \cdot 10^{4}$ y	β-	–	$1.4 \cdot 10^{-3}$	–	–	–	–
^{90}Sr	28.78 y	β-	–	$1.7 \cdot 10^{-3}$	–	–	–	–
^{81}Kr	$2.3 \cdot 10^{5}$ y	EC	–	–	–	$5.8 \cdot 10^{-3}$	–	–

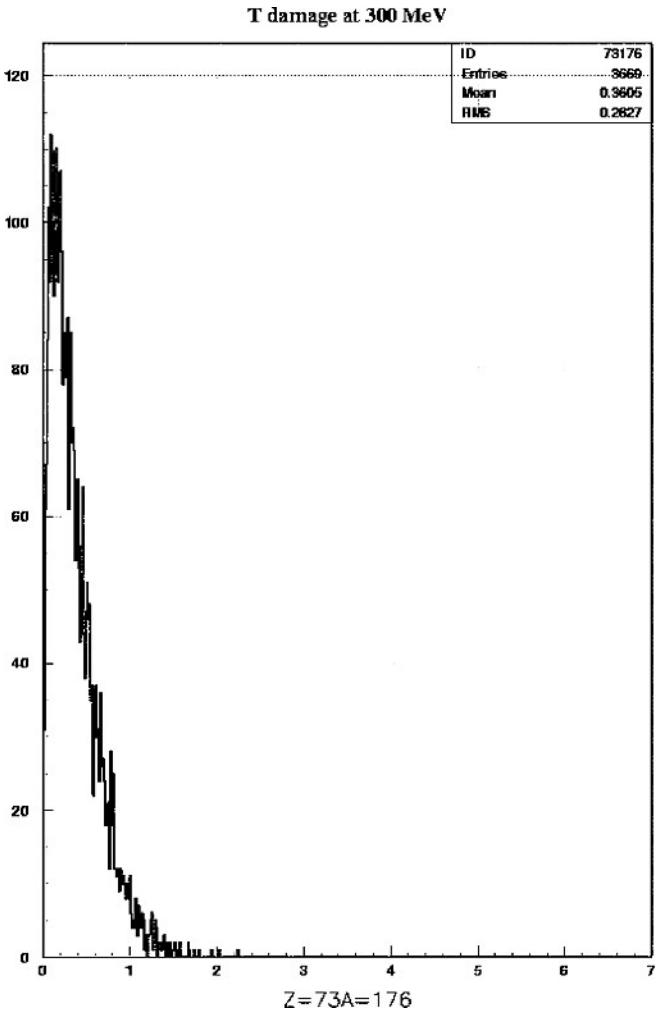

Fig. 7. Number of events creating a ^{176}Ta spallation residue (from 300 000 incident protons of 300 MeV kinetic energy) versus damage energy of this isotope (in MeV)

tranuclear and de-excitation codes. In this work we have chosen the intranuclear cascade of Bertini + Dresner evaporation and INCL4 + ABLA implemented in MCNPX2.5.e.

In the first step, the production cross-sections of the residues are calculated with MCNPX in a thin target. Then, using the history file created the damage energy spectra of the residues are evaluated using the NRT model but with little changes in the equations presented above:

$$k_L = \frac{0.133745\, Z_i^{2/3} Z_t^{1/2}}{A_i^{3/2}\, A_t} \left(\frac{A_i + A_t}{2}\right)^2 \left(\frac{2}{Z_i^{2/3} + Z_t^{2/3}}\right)^{3/4} \tag{6}$$

$$\varepsilon = \frac{0.03252\, A_t\, T_i}{(A_i + A_t)\, Z_i\, Z_t (Z_i^{2/3} + Z_t^{2/3})^{1/2}} \tag{7}$$

The total damage energy of these spallation residues is thus directly obtained and the number of Frenkel pairs can be calculated from Eq. (5). In order to obtain the nonelastic displacement cross-section one should multiply this result by the reaction cross-sections that has been obtained.

Results obtained for the two different spallation codes in MCNPX2.5.e are shown in Table 5 for three different energies of the incident proton beam. The difference between values obtained with the two spallation codes is of 4 % at maximum which shows an excellent agreement.

4.2 Calculation of the inelastic displacement cross-section with the INCL4 + ABLA stand alone version

As described above, MCNPX allows the calculation of the displacement cross-section in a direct way. We have chosen to compare these results to the calculations done with the INCL4 + ABLA stand alone version that gives just the information needed to the calculation of the displacement cross-section.

In the INCL4 + ABLA stand alone version a matrix of results is created in an ntuple format that can be read by the PAW [16] analysis system.

Table 5. Inelastic displacement cross-section in a ^{181}Ta target for different energies of the incident proton. These values were calculated with MCNPX2.5.e using two different spallation models INCL4 + ABLA and Bertini + Dresner

Energy	INCL4 + ABLA	Bertini + Dresner
140 MeV	2 322 b	2 345 b
300 MeV	3 556 b	3 543 b
500 MeV	4 943 b	4 978 b
1 GeV	7 247 b	7 553 b

Table 6. Inelastic displacement cross-sections calculated from the information registered in the n-tuple of the INCL4 + ABLA stand alone version for different energies of the incident proton. The difference with the displacement cross-sections calculated with MCNPX2.5.e using INCL4 + ABLA spallation model is also presented

Energy	Inelastic displacement cross-section	Difference MCNPX (%)
140 MeV	1 840 b	20
300 MeV	3 043 b	14
500 MeV	4 441 b	10
1 GeV	6 492 b	10

This matrix contains the following information for each event:

1) Mass of the target nuclei
2) Charge of the target nuclei
3) Incident energy of the proton
4) Multiplicity of the cascade
5) Multiplicity of the evaporation
6) Total multiplicity
7) Impact parameter
8) Angular momentum of the pre-fragment (after the cascade phase)
9) Charge, mass and energy of the fission fragment
10) Charge, mass and energy of the evaporation residue
11) Momentum in the laboratory frame of the spallation residue
12) θ and φ angles in the laboratory frame of the spallation residue
13) As it can be seen from Eq. (2), the information needed for the calculation of the inelastic displacement cross-section, the energy spectra of the spallation residue and its production cross-section, is given in this matrix.

For the evaluation of the displacement cross-section from data cumulated in the ntuple, we have applied Eq. (3 and 4) to the recoil energy of the residues created, in order to calculate the damage energy for each of them. The result for a given residue ($Z = 73$, $A = 176$) is shown in Fig. 7 for an incident proton of 300 MeV. This figure shows the number of events creating a ^{176}Ta spallation residue versus the damage energy of this isotope.

The displacement cross-section for this residue is calculated by integrating, channel per channel in the histogram, the number of events (related to the production cross-section) multiplied by the value of the damage energy (multiplied by $0.8/2 \cdot 90$ (eV) in order to obtain the number of Frenkel pairs), that means calculating exactly Eq. (2).

Table 7. Number of DPA created by the irradiation of an incident Gaussian proton beam of different energies and 0.14 mA in 1 cm^3 cell of ^{181}Ta, after one year of irradiation

Energy	DPA created by protons per year	DPA created by neutrons per year
140 MeV	16.14	2.00
300 MeV	25.09	2.78
500 MeV	34.78	3.77
1 GeV	52.15	6.12

Then, an addition is made over all the residues produced by the incident proton in order to obtain the total nonelastic displacement cross-section.

The results obtained using this method, are shown in Table 6 for different energies of the incident proton beam. In this table we also present the difference with the value obtained using MCNPX2.5.e with INCL4 + ABLA spallation code.

The difference between the two values can arise from the method to calculate the damage energy from the recoil energy of the residue, as it has been seen MCNPX2.5.e does not use exactly the same formulation used here. In any case, this difference is rather low if comparing with other approaches to calculate the displacement cross-section as the Molecular Dynamics method (MD) where a factor of 3–4 can be obtained between the MD method and the NRT method used here. Using this method for the calculation of the displacement cross-sections allows a real control of the different parameters taking part in this evaluation.

4.3 DPA created in the tantalum target

The displacement cross-sections are used in order to calculate the number of DPA that are going to be produced in an irradiated material. Many processes can give rise to these displacements in the crystal lattice of the material: when an incident proton impinges the target nuclei two different reactions can be produced, an elastic scattering of the proton or an inelastic reaction or spallation reaction.

Elastic scattering gives little recoil energy to the target nuclei and produces short displacement cascades. In general, as mentioned before, it has been proved [14–15] that their contribution to the total displacements created during the irradiation does not exceed 10–13 %.

The inelastic reactions (spallation reactions) in the target produce residual nuclei with high recoil energy that start the most important displacement cascades in the material. The number of DPA can be calculated by multiplying the inelastic displacement cross-section by the flux of incident protons.

Nevertheless, one must take into account the fact that when an incident proton of a given energy impinges the nuclei target a certain number of neutrons and protons with different energies are produced in the cascade and the evaporation phases. These nucleons can, themselves, produce new displacements with other target nuclei both by elastic and inelastic interactions.

In this work we have calculated the total number of displacements created in a cell of 1 cm^3 situated just in the center of the incident beam (see Fig. 1). This is the cell that is going to suffer the most important irradiation problems as it is going to receive a flux of $5\,577 \cdot 10^{14}$ p/s · cm^2 in the case treated here (Gaussian beam of 0.14 mA). We have taken into account not

only the incident protons but also all other nucleons (neutrons and protons) created in spallation reactions with the target.

In order to achieve this goal, we used MCNPX2.5.e for the evaluation of the mean flux of protons and neutrons created in the cell by each incident proton composing the Gaussian beam. The flux of protons and neutrons were divided in different groups each of them of a given energy in order to use the damage reaction cross-sections calculated in [14–15]:

- 50 energy groups were needed for proton fluxes created from an incident beam of 140 MeV and 24 groups for neutrons.
- 54 energy groups were needed for proton fluxes created from an incident beam of 300 MeV and 34 groups for neutrons.
- 58 energy groups were needed for proton fluxes created from an incident beam of 500 MeV and 38 groups for neutrons.
- 68 energy groups were needed for proton fluxes created from an incident beam of 1 GeV and 48 groups for neutrons.

Then, it is necessary to multiply the mean flux calculated by MCNPX2.5.e by the incident proton flux (the result is Φ_i) and the total displacement cross-section at the energy of the given group σ_{di} calculated in [14–15]. We thus obtain the number of displacements created in the cell for each energy group (i) and per second. An addition of these values gives us the total number of displacements created in the cell per second.

$$\sum_i \sigma_{di} \cdot \Phi_i \tag{8}$$

In Table 7, the result of theses calculations are presented for the four different energies of the incident proton beam after one year of irradiation of the 1 cm^3 cell.

As we have already shown in the precedent section, the different methods used to calculate the displacement cross-sections using the NRT approximation, can be evaluated to 10–20 % depending on the energy of the proton beam. These values give us an idea of the error that can be considered for the results presented in Table 7.

One should remark at this point that the evaluation of the number of DPA has been made taking into account the incident beam as the only origin of irradiation problems in the target. In a real ADS target, the material is also surrounded by an important neutron flux produced in the reactor. The characteristics of this neutron flux depend on the reactor chosen for the transmutation (thermal or fast spectrum). Therefore, this contribution should also be taken into account in the same manner for the calculation of the total DPA produced in the target material.

5 Conclusion

Some of the irradiation problems that a tantalum spallation target can suffer have been presented in this work. The shape of the tantalum target chosen for the calculation is based on precise descriptions of the TRADE target. Also, the Gaussian shape of the beam and its energy foreseen for this experiment (140 or 300 MeV). Calculations have been extended to 1 GeV of the proton beam which is the most adapted energy to run a high power ADS.

Spallation reactions in the target create residual nuclei that, during the irradiation time, increase the concentration of impurities in the material. These concentrations can give rise to corrosion and embrittlement problems in the material that must be taken into account when running the ADS. In this work we have presented an evaluation of these residual nuclei concentrations calculated after one year of irradiation by different energies proton beams. The calculations of the residual production cross-sections were made using MCNPX2.5.e with two different spallation models: the well-known Bertini intranuclear cascade + Dresner evaporation code and the new implemented intranuclear cascade INCL4 followed by the ABLA de-excitation code. The radioactive decay of different products during the irradiation time was performed with ORIHET 3 and data of the NUBASEX library and was compared to results given by the KAPROS code. In the case studied in this work, were the activation created by the spallation neutron flux is negligible, both codes give good agreement of the final residual concentrations, Nevertheless, for the case of a spallation target within a sub-critical core, this contribution is not longer negligible and the KAPROS code can take it into account in the evaluations.

The result of the final residual concentrations shows that, for lower energies of the proton beam (140 and 300 MeV), the most important productions correspond to residues with a charge near the charge of the tantalum target. These concentrations can be of the order of 10^3 appm (atoms of impurities per million atoms in the target) and the difference between the two used codes is rather small. Nevertheless, for residual nuclei with a charge between 50 and 65 the two codes can be different from a factor of 40 at 1 GeV. Even if the production concentration of these residues is small (at maximum some ten appm), this large difference can be important for some residues and shows important discrepancies in the physical description of the evaporation model. On the other hand, fission products are predicted with good agreement between the two codes.

The knowledge of the residual nuclei production in the target allows the calculation of the activation of the tantalum spallation target during and after the irradiation time for all energies of the incident proton beam. This is a useful determination in order to predict the security measures necessary to handle the target after the irradiation. We show in this case that there are not important differences between results obtained with the two models, as the isotopes contributing the most to the activation of the target are residues with a charge close to the one of the target material.

Damage production rate (dpa) in the target material for all energies considered in this work has also been determined. The most important contribution to the dpa is the displacements produced by the residual nuclei formed in the spallation reactions. Two methods have been shown for the evaluation of the inelastic displacement cross-section: using MCNPX or the stand alone version of the INCL4 + ABLA code and integrating directly the data contained in the resulting matrix. Differences between the two methods go from 10 to 20 % in the final result.

Finally, the total displacement cross-sections (elastic + non-elastic) calculated with MCNPX2.5.e have been used for the calculation of the total number of dpa created in a cell of 1 cm^3 irradiated directly by the Gaussian beam at its central position and at different energies. In this calculation we have considered dpa produced by the incident protons in the cell added to the dpa due to the protons and neutrons created in these reactions at all possible energies. In this case, the tantalum target was considered to be isolated (not inside a reactor core) and no other neutron flux coming from the reactor was added to the one created by the proton irradiation. This total number of DPA can be now evaluated in order to determine the real consequences that it could have in the life-time of the target material.

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