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# Analysis of nuclear reactions to determine the radionuclides generated and its activity in various devices

José Ródenas, Gumersindo Verdú

Departamento de Ingeniería Química y Nuclear, Universitat Politècnica de València, Spain

#### 1. INTRODUCTION

In a nuclear reaction X (a, b) Y an incident particle a impinges a target nucleus X, normally at rest, resulting another particle b and a new nuclide Y. If Y is radioactive, the reaction generates radionuclides and it is called an activation reaction. This type of reactions are important because it produces radioactive contamination, appearing radioactive substances where there were not. Therefore, it causes a problem of irradiation or contamination and the environment should be protected. On the other hand, the production of radionuclides can be of interest to be used in some industrial or medical applications. In any case, it is necessary to know the activity produced in the reaction.

The generated activity depends on the reaction cross section, incident particle flux and spectrum as well as particle flux distribution, concentration of the target nuclide  $\mathbf{X}$  and reaction time. When the reaction is finished, the activity decreases with disintegration constant and cooling time.

Most of the activation reactions are induced by neutrons since the addition of one neutron produces normally an instability in the nucleus, but other particles like protons can also be used to produce radionuclides.

Nuclear reactors and accelerators are some of the most used devices to host nuclear reactions to produce radionuclides for various applications. A nuclear reactor is an important source of neutrons to produce radionuclides. Accelerators generate particles that can produce different reactions. An important type of accelerator is a cyclotron that can be used to produce positron emitter radionuclides, necessary in a Positron Emission Tomography (PET) device for applications of Nuclear Medicine.

A summary of the simulation of nuclear reactions using the Monte Carlo (MC) method is presented in this work.

# 2. CALCULATION OF THE GENERATED ACTIVITY

To estimate the generated activity, the reaction rate Q should be calculated according to eq. (1). It depends on the incident particle flux  $\Phi(E)$  and the macroscopic cross section  $\Sigma(E)$  of the reaction. The Monte Carlo method can be used to estimate  $\Phi(E)$  while cross sections are in the program library provided the reaction is identified.

Different MC codes can be used for this proposal: MCNP5 (X-5 Monte Carlo Team, 2005), MCNP6 (Pelowitz, Fallgren, McMath (Eds.), 2014), GEANT (Giani, S. et al., 1998), or FLUKA (Ferrari, Fassò, Ranft, Sala, 2005) among others.

$$Q = \int \Phi(E) \Sigma(E) dE$$
 (°1)

where

Q is the reaction rate per volume unity (reactions/cm<sup>3</sup>-s)

 $\Phi$  (E) is the particle flux (particles/cm<sup>2</sup>-s); and

 $\Sigma$  (E) is the macroscopic cross section (cm<sup>-1</sup>) of the reaction X (a, b) Y.

The macroscopic cross section can be expressed by equation (2).

$$\Sigma (E) = N \sigma(E) \tag{2}$$

being

N the atomic density of the target X (at/cm<sup>3</sup>); and

 $\sigma$  (E) the microscopic cross section of the reaction (barn), with 1 barn =  $10^{-24}$  cm<sup>2</sup>.

Thus, equation (1) can be rewritten as follows:

$$Q = C \int \Phi(E) \, \sigma(E) \, dE \tag{3}$$

being C a normalization factor depending on the target concentration. C is equivalent to the atomic density N of the target in atoms/barn-cm. If the nucleus X corresponds to the isotope j of an element n contained in the sample

(for instance, an alloy like stainless steel), C can be determined from density, composition and isotopic abundances, according to eq. (4).

$$C = \left(\frac{\rho}{M}\right) \left(\frac{X_n}{100}\right) \left(\frac{X_j}{100}\right) 10^{-24} N_A$$
 (4)

being

 $\rho$  the density of the target sample (g/cm<sup>3</sup>)

M the atomic or molecular weight (g/mol)

X<sub>n</sub> the percentage of element n in the sample

 $X_i$  the percentage of isotope j in element n

10<sup>-24</sup> the equivalence for cross section units (cm<sup>2</sup>/barn)

N<sub>A</sub> the Avogadro number (atoms/mol)

For each radionuclide Y generated in the reaction, a matter balance can be done, each reaction producing one nucleus:

$$\frac{\mathrm{dY}}{\mathrm{dt}} = \mathbf{Q} - \lambda_{\mathbf{Y}} \mathbf{Y} \tag{5}$$

being

Y the concentration of Y (nuclei/cm<sup>3</sup>)

Q the reaction rate per volume unity (reactions/cm<sup>3</sup>-s equal to nuclei/cm<sup>3</sup>-s)

 $\lambda_{\rm Y}$  the disintegration constant of the radionuclide Y (s<sup>-1</sup>)

Integrating equation (5), the concentration of Y is obtained, being t<sub>i</sub> the irradiation time:

$$Y(t) = \left(\frac{Q}{\lambda_{Y}}\right) \left(1 - e^{-\lambda_{Y} t_{i}}\right)$$
 (6)

For a cooling time  $t_c$  the concentration Y(t) becomes:

$$Y(t) = \left(\frac{Q}{\lambda_Y}\right) \left(1 - e^{-\lambda_Y t_i}\right) e^{-\lambda_Y t_c}$$
(7)

Multiplying now by  $\lambda_Y$ , the volumetric activity (Bq/cm<sup>3</sup>) is obtained:

$$A_{Y}(t) = Q \left( 1 - e^{-\lambda_{Y} t_{i}} \right) e^{-\lambda_{Y} t_{c}}$$
(8)

To obtain the total activity  $A_{Ytotal}$  (t) it is necessary to multiply eq. (8) by the sample volume V. The maximum activity will be the asymptotic value, Q (multiplied by the target volume), considering an irradiation time very long ( $\sim \infty$ ) and neglecting the cooling time.

# 3. APPLICATION OF THE MONTE CARLO METHOD

Neutrons induce most of the activation reactions. So in the next analysis, neutrons are considered as the incident particles used to produce activation reactions.

A Monte Carlo code such as MCNP can calculate the neutron flux  $\Phi$  (E) from the neutron source and its characteristics: geometry, dimensions, materials and so on. Values of  $\sigma$  (E) for materials involved in the considered reactions are extracted from the code libraries. If values of C are provided, reaction rates Q are calculated according to equation (3) for each case considered. MCNP uses a tally F4 for this type of problem with a multiplier card FM4 including values of C, material X identifier and reaction identifier.

All tallies obtained with MCNP are normalized to be per starting particle. Therefore, the activity is calculated per emitted particle, neutrons in this case. As eq. (5) needs Q in nuclei/cm<sup>3</sup>-s, eq. (3) should calculate Q per volume unity (reactions /cm<sup>3</sup>-s) using the flux  $\Phi$  (neutron/cm<sup>2</sup>-s). However, the tally F4 gives fluence (neutron/cm<sup>2</sup>), so it is necessary to consider a particle rate, that is, the instantaneous neutron population  $\dot{N}$  (neutron/s) that can be calculated by eq. (9).

$$\dot{\mathbf{N}} = \overline{\mathbf{P}} \, \mathbf{c} \, \mathbf{v} \tag{9}$$

where

P is the mean power (W)

c is equal to 3.12 E+10 fissions/W-s; and

v is the mean number of neutrons emitted per fission, equal to 2.47 for U-235.

Therefore, the final activity (in Bq) will be the product of equations (8) and (9) by the sample volume V. That is:

$$A_{\text{Ytotal}}(t) = A_{\text{Y}}(t) \dot{N} V \tag{10}$$

A similar analysis can be done for other type of particles causing a nuclear reaction.

## 4. SOME SPECIFIC CASES OF APPLICATION OF THIS METHODOLOGY

In previous papers MCNP5 (X-5 Monte Carlo Team, 2005) was applied to estimate the activity generated in control rods of a BWR (Ródenas et al., 2010a, 2010b) as well as the dose produced around the irradiated fuel pool where control rods are stored after being withdrawn from the reactor (Ródenas et al., 2010c, 2010d). It is very important to calculate these doses because they can affect to plant workers in the area.

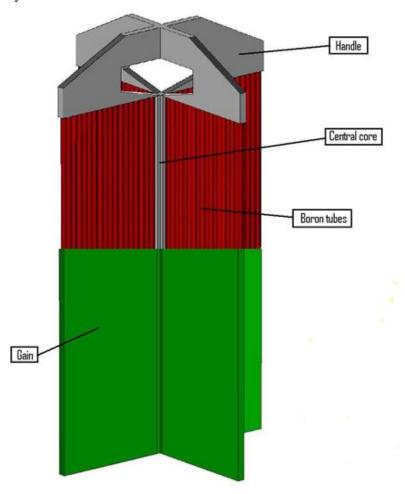
Later, a similar analysis was applied to the irradiation of a sample of stainless steel (Ródenas, Gallardo, Weirich, Hansen, 2014) as the activation of control rods is mainly produced in the stainless steel tubes containing the absorber. More exactly in the components of this alloy. The comparison of simulation results with experimental measurements allows the validation of developed models.

Another application is to simulate the neutron activation to adjust some parameters (reactor power, irradiation time, cooling time and sample volume) to obtain exempt activities (Ródenas, Aledó, 2018). When radionuclides are used like tracers, it is convenient that its activity be below exempt values so that the marked product can be considered as a conventional waste. As well, the dose potentially received by workers in the process should be below limits, since they will not work in a controlled area.

Finally, in a recent work an analysis of reactions with protons in a cyclotron to produce positron emitters for a PET has been performed (Jabaloyes, 2018).

# 4.1. Activity generated in control rods of a BWR

Control rods are activated by neutron reactions into the reactor. The dose produced by this activity is not important inside the reactor, but it has to be taken into account when the rod is withdrawn from the reactor. Activation reactions produced have been modelled with the MCNP5 code obtaining the number of reactions, which can be converted into activity.



#### Fig. 1. Model of the control rod.

Major activation reactions will be produced in stainless steel components. A model of the control rod with its different parts, handle, tubes, gain, and central core, can be seen in figure 1. The reactions taken into account and generated isotopes are listed in Table 1. Nevertheless, only those isotopes emitting gamma rays have interest from the point of view of dose calculation. Consulting disintegration schemes at the JANIS database (Janis, 2005), the list is simplified. They remain those listed in Table 2 where calculated activities are shown. The activity generated by neutron irradiation of control rods in a BWR is important for some components of stainless steel, in particular those producing Co-60 and Mn-54.

Table 1. Activation reactions in stainless steel.

N14 (n, p) C14	Mn55 (n, 2n) Mn54
Fe54 (n, p) Mn54	Fe54 (n, γ) Fe55
Co59 (n, γ) Co60	Ni58 (n, γ) Ni59
Ni58 $(n, \alpha)$ Fe55	Ni60 (n, p) Co60
Ni62 (n, γ) Ni63	Cu63 (n, α) Co60
Nb93 (n, γ) Nb94	Mo92 (n, γ) Mo93
Ag107 (n, γ) Ag108m	Ag109 (n, γ) Ag110m
Eu151 (n, γ) Eu152	Eu153 (n, γ) Eu154
Hf177 (n, γ) Hf178	Cl35 (n, p) Cl36
Cl37 (n, 2n) Cl36	Ti46 (n, p) Sc46
Al27 (n, γ) Al28	$Zn64 (n, \gamma) Zn65$

Table 2. Activation results (Bq) for the model of control rod.

	gains	central core	tubes	handle
Mn54	2.59E+13	4.45E+13	1.78E+14	2.60E+14
Co60	1.48E+14	1.14E+14	7.15E+14	1.97E+15
Nb94	1.70E+08	2.35E+08	1.04E+09	1.88E+09
Ag108m	3.70E+09	4.48E+09	1.91E+10	4.61E+10
Ag110m	6.62E+11	7.65E+11	4.74E+12	8.89E+12
Eu152	2.50E+10	1.13E+10	9.12E+10	3.74E+11
Eu154	1.66E+09	1.49E+09	8.72E+09	1.98E+10
Hf178	1.44E+11	1.47E+11	8.79E+11	1.71E+12
Sc46	1.84E+08	3.29E+08	1.28E+09	1.88E+09
<b>Zn65</b>	1.18E+12	1.30E+12	5.86E+12	1.48E+13

The activity generated in control rods of BWR was not measured because of its high value. Instead the dose produced by this activity was measured into the pool where rods are stored. Results of simulation of activation were used for another MC model to calculate doses produced at different distances and depths into the pool. Agreement was satisfactory (6% deviation) for the uppermost point of the control rod, that is, the handle, where the highest dose is produced. Therefore, an indirect validation was done (6).

The handle of the control rod is the piece more activated because it is exposed to neutron flux most of the time while the rest of the rod can be only partially introduced into the core. Therefore, an inversion of the position of control rods stored into the irradiated fuel pool will produce an important reduction of the dose around the pool. Results show an important reduction of dose rate, by a factor of 2000 approximately, in the zones where operators are usually working. Therefore, it is very advisable to introduce the appropriate changes in the hanger devices in order to achieve this important dose reduction (Ródenas et al., 2010d).

# 4.2. Activity generated in a sample of stainless steel

As stated above, most of the activation is produced in stainless steel components of the control rod. Therefore, the Monte Carlo model can be applied to the activation produced in a piece of stainless steel exposed to some neutron flux in a reactor. So, a stainless steel sample was irradiated in the training reactor AKR-2 of the Technical University Dresden (Hansen and Wolf, 2009). It is a zero power, thermal reactor moderated by solid polyethylene. The fuel elements consist of a homogeneous mixture of moderator and uranium oxide fuel enriched 19.8%. It has

a maximum power of 2 W and the maximum neutron flux in the central experimental channel is  $\Phi_{max} = 5$  E+07 n/cm<sup>2</sup> s. For the experiment, the reactor is driven at a power level of P = 0.59 W. This corresponds to a measured neutron flux of 2.5 E+07 n/cm<sup>2</sup> s in the central experimental channel.

A stainless steel sample type X8CrNiTi18.10 is irradiated for 10 hours in the central experimental channel of the reactor. The sample has cylindrical shape with a radius of 1 cm and a length of 7 cm. It has a volume of  $21.99 \text{ cm}^3$  and a density  $\rho$ =  $7.9 \text{ g/cm}^3$ . Thus, it has a mass of 173.73 g. The composition of the stainless steel sample is listed in Table 3 (Thyssen Krupp Materials International, 2007) and the reactions produced in the sample can be read in Table 4.

Table 3. Composition of the sample.

Element	weight fraction %	Molar mass g/mol
Cr	19.000	51.9961
Ni	12.000	58.6934
C	0.100	12.0107
Si	1.000	28.0855
Mn	2.000	54.938
P	0.045	30.973
S	0.015	32.065
Ti	0.400	47.867
Fe	65.440	55.845

Table 4. Reactions produced in the sample.

$P^{31}(n, \gamma) P^{32}$	Fe <sup>58</sup> (n, γ) Fe <sup>59</sup>
$Cr^{50}(n, \gamma) Cr^{51}$	$\mathrm{Ni}^{58}\left( \mathrm{n},\gamma\right) \mathrm{Ni}^{59}$
$Cr^{54}$ $(n, \gamma)$ $Cr^{55}$	$\mathrm{Ni}^{58}\left(\mathrm{n},\alpha\right)\mathrm{Fe}^{55}$
Mn <sup>55</sup> (n, 2n) Mn <sup>54</sup>	Ni <sup>60</sup> (n, p) Co <sup>60</sup>
$\mathrm{Mn^{55}}\left(\mathrm{n},\gamma\right)\mathrm{Mn^{56}}$	$\mathrm{Ni}^{62}\left(\mathrm{n},\gamma\right)\mathrm{Ni}^{63}$
Fe <sup>54</sup> (n,p) Mn <sup>54</sup>	$\mathrm{Ni}^{64}\left( \mathrm{n},\gamma\right) \mathrm{Ni}^{65}$
$Fe^{54}$ (n, $\gamma$ ) $Fe^{55}$	${ m Mo^{92}}({ m n},\gamma){ m Mo^{93}}$

Results from MCNP5 simulation are listed in Table 5 for a cooling time of 10 min. The most important activity is obtained for Mn-56. It is due to the high value of the cross section for the reaction  $(n, \gamma)$  in manganese and to the short half-life of Mn-56 (2.582 h) that permits this radionuclide to reach equilibrium during an irradiation time of 10 hours. The longer half-lives of some radionuclides, like Ni-59 (80 a), Ni-63 (100 a) and Mo-93 (3 a) causes that a low activity is generated because equilibrium is far to be reached.

The MC model to simulate the irradiation of a stainless steel sample in the training reactor AKR-2 was also validated by means of another MC model calculating the dose produced around the irradiated sample. Results of the second simulation were compared with dose measurements and the maximal deviation was 8%. (9)

Irradiation and cooling times have importance depending on the half life of the nuclides produced. Radionuclides with short half life arrive quickly to asymptotic equilibrium, but its activity decreases also quickly with the cooling time. It is the opposite for radionuclides with higher values of half life.

Irradiation of reactor components in power reactors is very long (up to 15 years in average) compared to the time of irradiation of the sample in a training reactor like AKR2. Thus, also radionuclides with high half-lives build up during the irradiation in a power reactor, although they were hardly present in the current problem. This is however not downgrading the model. As it works for short lived nuclides, it will also be applicable for long lived nuclides.

Table 5. Activities calculated for tc=10 min

			A (Bq)	
nuclide	Tally F4	A (Bq/cm <sup>3</sup> )	per neutron	A total (Bq)
$\mathbf{P}^{32}$	7.777E-09	1.556E-10	3.422E-09	1.56E+02

$Cr^{51}$	5.793E-06	6.004E-08	1.320E-06	6.00E+04
Cr <sup>55</sup>	6.088E-08	8.691E-09	1.911E-07	8.69E+03
$\mathrm{Mn}^{54}$	2.226E-08	2.056E-11	4.522E-10	2.06E+01
$\mathbf{Mn}^{56}$	1.310E-05	1.167E-05	2.566E-04	1.17E+07
Fe <sup>55</sup>	4.274E-06	1.252E-09	2.754E-08	1.25E+03
Fe <sup>59</sup>	1.061E-07	6.846E-10	1.505E-08	6.84E+02
$\mathrm{Co}^{60}$	3.902E-10	5.855E-14	1.288E-12	5.85E-02
Ni <sup>59</sup>	1.594E-05	1.577E-13	3.467E-12	1.58E-01
$Ni^{63}$	2.733E-06	2.162E-11	4.754E-10	2.16E+01
Ni <sup>65</sup>	6.985E-08	6.246E-08	1.373E-06	6.24E+04
$\mathrm{Mo}^{93}$	2.648E-06	6.983E-13	1.536E-11	6.98E-01

## 4.3. Analysis of some parameters to obtain exempt activities

Another simulation of the neutron activation was performed to adjust some parameters (reactor power, irradiation time, cooling time and sample volume) to obtain exempt activities, so the radionuclides generated can be used as tracers in a non-controlled area.

The study is based on some data obtained from measurements performed many years ago during a project developed at IRISIB, Brussels (Belgium) for a Diesel motor (Manderlier, Tondeur, Guillaume, Gerardy, 2003). In that work, they tried to control particles escaping from the filter by measuring the activity of the marked compound. Some additives marked with radionuclides were added to the motor oil. Three additives were chosen containing respectively Na-24, Mo-99 and Sb-124, which were obtained by neutron activation. Therefore, three elements, Na, Mo and Sb, were used for the experience at IRISIB. Their significant characteristics are listed in table 6.

Na Mo Sb Density (g/cm<sup>3</sup>) 0,968 10,28 6,697 Natural isotopic abundance Na-23 100% Mo-98 24,39% Sb-123 42,79% Na-23  $(n,\gamma)$  Na-24 Mo-98  $(n,\gamma)$  Mo-99 Activation reaction Sb-123 (n, $\gamma$ ) Sb-124 Half-life 14,997 h Na-24 65,976 h Mo-99 60,2 d Sb-124 **Exempt activity** 0,1 MBq Na-24 1 MBq Mo-99 1 MBq Sb-124

Table 6. Characteristics of the elements chosen for the experience

The generated activity is directly proportional to the reactor power as it is clear from equations (9) and (10). Therefore, the value of the power, neutron population indeed, is crucial to obtain a certain activity. According to Eq. (10) the variation of the activity with the sample volume is also linear. Consequently, a lower power and small sample volumes will be considered.

A longer irradiation time will permit to reach the activation equilibrium, but it depends on the radionuclide half-life. As for the cooling time, it should be very short for Na-24 due to its short half-life, while it is less important for Mo-99 and Sb-124.

The volume of the additive containing Na-24 was 330 cm<sup>3</sup>. It is quite a large volume, but the short half-life of Na-24 implies that an exempt activity can be obtained with not too long irradiation times and short cooling times. The variation of the Na-24 activity with the irradiation time is presented in figure 2 for three different cooling times: 0, 1 and 3 hours. From this figure, it can be deduced that an exempt activity can be produced with irradiation times less than 100 hours, and a cooling time of 1 or 3 hours.

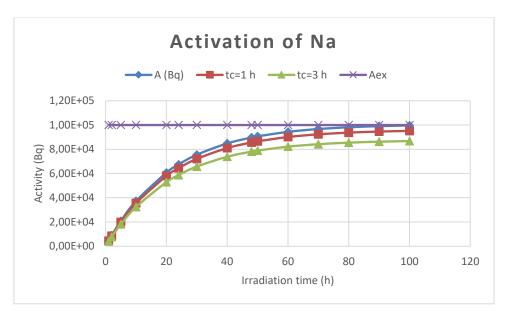


Fig. 2. Variation of the Na-24 activity with the irradiation time.

To obtain Mo-99 a volume of Mo equal to 0.85 cm<sup>3</sup> has been considered for three different values of the reactor power: 2, 50 and 1000 W. The cooling time is not important due to the half-life of Mo-99. For the higher values of the power, 50 W and 1 kW, it is not possible to get exempt activities, even with some cooling time. For the lowest power considered, 2 W, an exempt activity can be obtained with irradiation times lower than 8 hours. Therefore, if the power is higher it will be necessary a smaller volume. Results can be seen in Table 7.

Table 7. Activities (Bq) of Mo-99 for different powers varying the irradiation time.

t <sub>i</sub> (hr)	$P_1 = 2 W$	$P_2 = 50 \text{ W}$	$P_3 = 1 \text{ kW}$	Aex
1	1.30E+05	3.24E+06	6.48E+07	1.00E+06
2	2.58E+05	6.45E+06	1.29E+08	1.00E+06
3	3.85E+05	9.63E+06	1.93E+08	1.00E+06
4	5.11E+05	1.28E+07	2.55E+08	1.00E+06
5	6.35E+05	1.59E+07	3.18E+08	1.00E+06
6	7.58E+05	1.90E+07	3.79E+08	1.00E+06
7	8.80E+05	2.20E+07	4.40E+08	1.00E+06
8	1.00E+06	2.50E+07	5.00E+08	1.00E+06
9	1.12E+06	2.80E+07	5.60E+08	1.00E+06
10	1.24E+06	3.09E+07	6.19E+08	1.00E+06

For obtaining Sb-124, the volume has been set at  $0.75 \text{ cm}^3$ , and no cooling time is included as the half-life is very long. Again three different values of the reactor power: 2, 50 and 1000 W, have been considered. Simulation gives values of activity for a variation of the irradiation time up to 100 hours. Results are shown in figure 3 for P = 2 W and in figure 4 for higher powers. It can be seen that exempt activities cannot be got with higher values of the power, while it is possible for 2 W. For a higher power, the volume should be reduced.

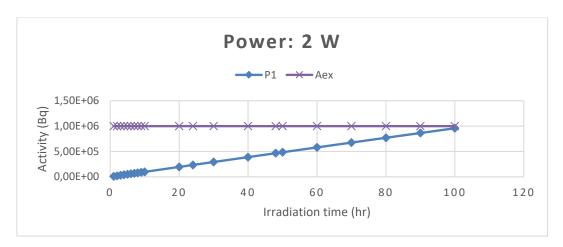


Fig. 3. Variation of the Sb-124 activity with the irradiation time for low power.

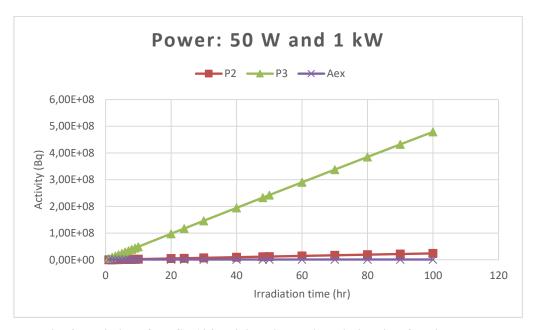


Fig. 4. Variation of the Sb-124 activity with the irradiation time for high power.

# **4.4. Reactions with protons**

The last case presented considers an analysis of reactions with protons in a cyclotron to produce positron emitters for a PET. The analysis is centered on the production of Ga-68 and Cu-64. Ga-68 has a half-life of 68 m and emits positrons with 1.9 MeV. Half-life of Cu-64 is 12.7 hr and emits positrons with 0.653 MeV. They can be produced in a cyclotron by the reactions:

It is important to underline that these reactions are produced in liquid targets, usually a nitric dissolution. This can improve results, as many times it is difficult to extract the radionuclide from a solid target where reactions are produced.

To simulate reactions with protons it is necessary to use MCNP6 (Pelowitz, Fallgren, McMath (Eds.), 2014) and the appropriate cross section libraries. Results of the simulation have been compared with experimental measurements performed at the University of Coimbra, Portugal (Alves, 2017). Discrepancies between calculated and measured activities are lower than 10 %.

# 5. CONCLUSIONS

An analysis of nuclear reactions to determine the activity of radionuclides generated has been presented. Neutrons have been the particles considered to induce reactions, and mainly nuclear reactors as the devices where reactions are performed, but also protons and a cyclotron have been considered.

The Monte Carlo model developed is suitable to determine radionuclides produced and its activity.

Results have been compared with experimental values and models have been validated.

Further developments are foreseen to consider other reactions, varying the device where they are produced, as well as the incident particle and the target.

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