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Experimental assessment and analysis of calculations accuracy for the neutron-induced radio-isotope creation in copper parts of radiotherapy accelerators

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ABSTRACT:

Induced radioactivity in particle accelerators is currently not sufficiently addressed, particularly for radiotherapy electron LINACs when the activation occurs via secondary neutrons created in photo-nuclear reactions.

Methodologies based on Monte-Carlo simulations coupled with activation calculation codes are often used to address this issue. Their accuracy, which is potentially affected by the limited capabilities of the calculation tools to simulate nuclear reactions at the origin of the materials activation, can be deeply analyzed only through comparisons with experimental values but the latter are not widely available.

New macroscopic data for the activity induced by neutrons in the materials commonly present in medical electron LINACs are presented. Neutron-induced activation has been measured by sample irradiation at SPIRAL2/NFS facility in accurately characterized configurations. Experimental results were compared to the simulations predictions allowing to assess the precision that can be achieved by a fully simulation-based methodology, mainly related to the energy-dependent reaction cross-sections.

KEYWORDS: electron LINAC, neutron-induced reactions, activation, Monte-Carlo simulations, γ spectrometry

1. INTRODUCTION

Due to induced radioactivity in facilities using particle accelerators, their dismantling generates radioactive waste that needs appropriate management in terms of precise characterization and identification of storage and recycling paths. The components of these accelerators and the biological shielding potentially become radioactive following activation directly by the beam itself and by secondary particles created through nuclear reactions. IAEA Specific Safety Guide SSG-49 requires that “In the planning stage for decommissioning, the degree and extent of contamination in a facility should be clearly determined, characterized, evaluated and classified”. Furthermore, IAEA Specific Safety Guide SSG-46 requires the following: “When designing accelerators producing high energy X-ray beams (>10 MV), manufacturers should minimize potential hazards from neutron activation of patients and materials in the treatment room (induced radioactivity secondary to radiation therapy)”. Council directive 2013/59/EURATOM states that

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the undertaking has appropriate arrangements for radioactive waste management. Moreover, as the clearance and exemption levels are nuclide-specific, measurements based on γ radiation dose rate are not sufficient for decision-making.

Radiotherapy represents the main medical application using particle accelerators with about 4.000 facilities in use in European countries (more than 500 facilities are presently in use in France, where the number increases at a rate of 14% over the last five years) and about 13.000 facilities over the world. Most of these facilities use linear electron accelerators, whilst cyclotrons are used by only a few facilities, dedicated to hadron-therapy and to the production of radio-isotopes for nuclear medicine.

There is presently an increasing demand from facilities operators for appropriate management of the radioactive waste generated by the dismantling of medical electron accelerators. Considering the large number of medical facilities that are due to arrive at the end of their life cycle, the regulatory authorities and waste management operators started to look closely at the aspect of accurate characterization of the waste generated, in terms of level of activity and radio-isotopes created, in order to identify the correct waste management options. Particle accelerators used for research are generally high-scale and are producing beams with high energy and intensity. Considering the large amount of waste generated at the accelerator facilities, the optimization of its management is crucial to reduce costs, especially by accurately identifying the waste with an activation level low enough to be considered as conventional, which could then be recycled. In some EU countries, the national legislation recently requires for this process to be fulfilled even before the commissioning, which in the absence of an appropriate methodology could be an important impediment in further developments.

These aspects cannot be presently addressed with the required accuracy since the only method available consists of the control of parts during dismantling by simple detectors available on-site. This method gives only an indication of the activation level and dose rate associated to γ emitters, while β emitting radio-isotopes are not taken into account. Therefore, if the pure β emitters happen to be the main contributors to the total radiological inventory of a component, this method is completely inappropriate. Therefore, no well-consolidated decision can be taken based on this simple approach for the best strategies to be adopted in waste management. Moreover, the dose rate measurements do not always give information about the activity concentration in a macroscopic object, especially if mainly low-energy γ radiation is emitted from a large component or if the component is not homogeneously activated.

The French metrology laboratory for ionizing radiation (LNHB) recently started to address this issue by developing a method to calculate the complete radioactive inventory created by activation of the accelerator's components and infrastructure, consisting of accurate modeling of the geometry and of particle interactions with matter [1]. Considering the complexity of the geometries and the diversity of the materials, the method based on computational tools is the only viable option able to provide a complete answer in assessing the induced radioactivity and it is nowadays extensively used by the community [2,3]. In order to ensure the accuracy of the results obtained by such fully calculation-based methodologies and consequently the appropriate actions to be undertaken when comes to the radiological aspects related to the waste management and to the exposure to residual doses, calculations results must be validated against experimental data. The strongest validations can be achieved by activating specific material samples in neutron beams that are accurately characterized in terms of flux and energy spectrum. When comparing the obtained data with the results from the calculation, one can validate the accuracy of the computational tools and identify possible needs for improvements in the models or evaluated libraries used by the codes. This work presents new data obtained by measurements of neutron-induced activation in copper, paving the way for a broader approach including other materials of interest for radiotherapy accelerators and subsequently allowing improved predictive calculation tools. This could support practice for future clearance and decommissioning of radiotherapy electron accelerators.

2. INDUCED RADIOACTIVITY IN MEDICAL ELECTRON ACCELERATORS

Most radiotherapy treatments are based on accelerators producing electron beams, that can be used directly in some particular cases but more often (around 90% of the treatments) they are converted, via bremsstrahlung reaction with a high Z target, in photon beams which are further used to irradiate the cancerous tumors.

The activation of materials in this case is related to one specific process, namely the nuclear reactions induced by photons (or photo-nuclear reactions) following the interaction of photons with the materials composing the accelerator. The photo-nuclear (γ, Xn) reactions producing neutrons are responsible for the direct creation of radioactive isotopes from stable nuclei after losing one neutron. On the other hand, the neutrons produced can subsequently induce nuclear reactions by interacting with the materials and this is estimated to be the major contribution to the induced activation in a medical accelerator's component, especially those not being directly exposed to the photon beam it produces.

Considering this specific mechanism responsible for the activation of components and the materials used, the case of a medical accelerator is rather particular. On one hand, the level of induced activation is expected to be relatively low, mainly because of the energy threshold of photo-nuclear reactions (the lowest threshold is 1.67 MeV for the ${}^9\text{Be}(\gamma, Xn)$ reaction) and the variation of their cross-section with the photon energy. Figure 1 below shows two examples obtained using different data sets from the reference [4], for a light and for a heavy nucleus. It is important also to mention that in the operating regimes of the conventional radiotherapy accelerators the electron energies don't exceed 25 MeV.

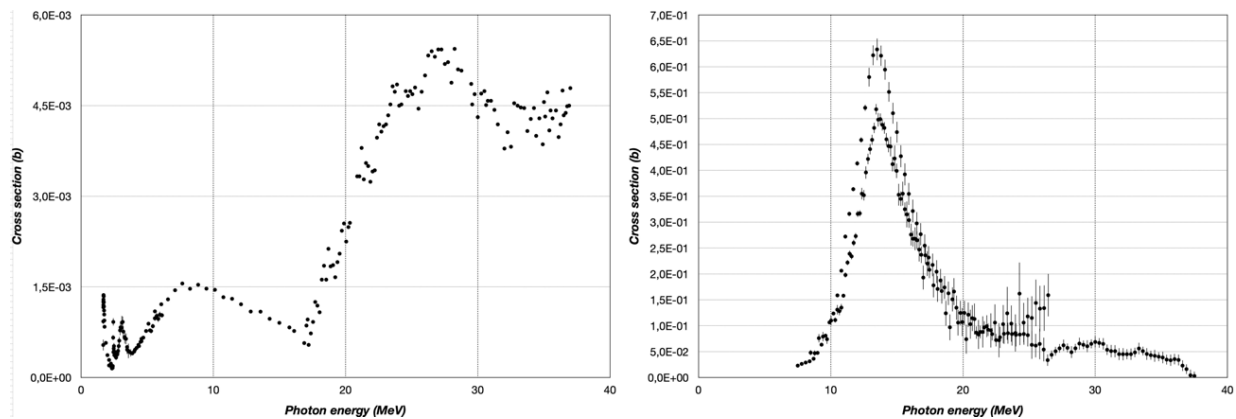


Figure 1: Experimental cross-sections from EXFOR data library for neutron production in photo-nuclear (γ, Xn) reactions on ${}^9\text{Be}$ (left) and ${}^{208}\text{Pb}$ (right).

On the other hand, the low-level waste generated, although not highly radioactive, still has to be managed and in high quantities. As an example, the total weight represented by the main components of a radiotherapy electron accelerator is typically above 1 Ton and when considering all 4000 facilities currently operating in Europe the amount of material needing appropriate waste management after dismantling is highly significant. The aspect of recycling becomes more important in this context since only the waste coming from some components of the accelerator will have to be considered as radioactive and appropriately stored. However, the authorizations for clearance from the safety authorities need an accurate assessment of the radioactive inventory. This can only be achieved by a consolidated methodology based on validated computational tools.

The method proposed by LNHB to simulate the particle interaction with matter and to calculate the production of photons and neutrons by radiotherapy accelerators is based on the state-of-the-art Monte-Carlo tools including MCNP [5], FLUKA [6], and PHITS [7]. In Figure 2 below are presented typical calculated spectra at the patient location of total photon and neutron radiation produced by a VARIAN

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TrueBeam radiotherapy accelerator operating at 20 MV. The neutron energies are distributed over a wide range, from thermal up to a maximum of around 10 MeV.

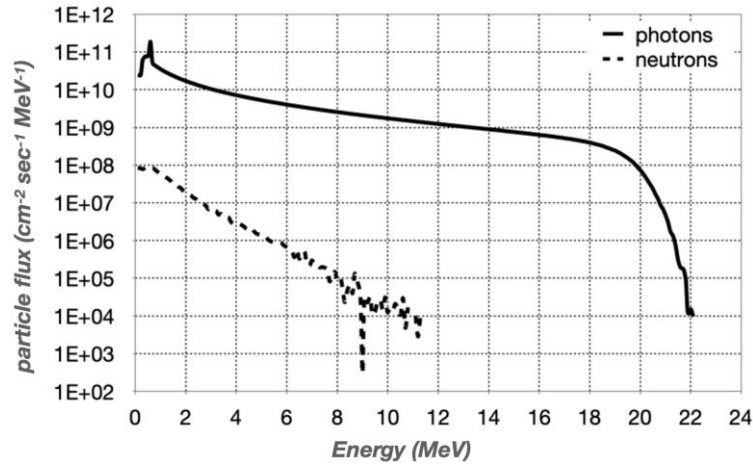


Figure 2: Spectra of photons and neutrons produced by a radiotherapy accelerator with an electron beam of 20 MeV as calculated by the MCNP6.2 Monte-Carlo code.

The activity induced in the materials by neutron-nucleus reactions can be determined using dedicated activation calculation tools, among which one of the most important is FISPACT-II [8]. This code is continuously maintained and regularly updated with the most recent libraries, including several options for cross-section values for neutron-induced reactions from TENDL, ENDF/B.VIII.0, JEFF-3.3, JENDL-4.0, and CENDL-3.1 international libraries, which are complemented with the latest decay data to calculate the activity related to the radio-isotopes created. While any libraries could be used, the development of FISPACT-II over the last few years has run in parallel with the development of the evaluated cross-section library TENDL based on the nuclear reactions code TALYS [9], which is recommended by the authors as the source of evaluated cross-sections to be used in the calculations.

As powerful and complete as they are intended to be, the activation calculation tools don't always provide good accuracy of the results for a specific application. The evaluated cross-section libraries they are using to obtain the probability of a given neutron-induced reaction leading to a radioactive nucleus and the decay data to calculate the related activity are sometimes incomplete or inaccurate, depending on the neutron energies and the composition of the materials. The complexity related to neutron-nucleus reactions is illustrated in Figure 3 which shows an example, obtained using TENDL evaluated cross-section library, of the variation of the reaction probability as a function of neutron energy. It is important to underline the existing difference between capture (n,) and elastic reactions and reactions at the origin of the emission of charged particles. The first two reactions are considered without threshold being therefore dominant at low neutron energies and characterized by a region of resonances while the others occur for neutron energies beyond a certain threshold and whose probability increases with the neutron energy.

Considering their wide energy range as shown in Figure 2, neutrons being produced by a radiotherapy accelerator can induce a large variety of nuclear reactions at the origin of radioactive isotopes creation. The complexity of all these phenomena at play implies that the precise determination, by a methodology based on calculation, of the activity induced by neutrons in the components of radiotherapy electron accelerators, requires a good knowledge of the probabilities of the different nuclear reactions at its origin and their appropriate integration into the calculation codes used. The lack of data however can strongly limit the accuracy of the calculation-based methods as the models they are using are not necessarily fulfilling this condition as it will be discussed in detail in the subsection 4. 2. This constitutes the main motivation of the present work, as new data must be obtained in order to validate the predictive power of the models used, or if important disagreement is found, to provide the community with new information helping to improve the

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use of calculation tools for the assessment of induced activity in the components of radiotherapy electron accelerators.

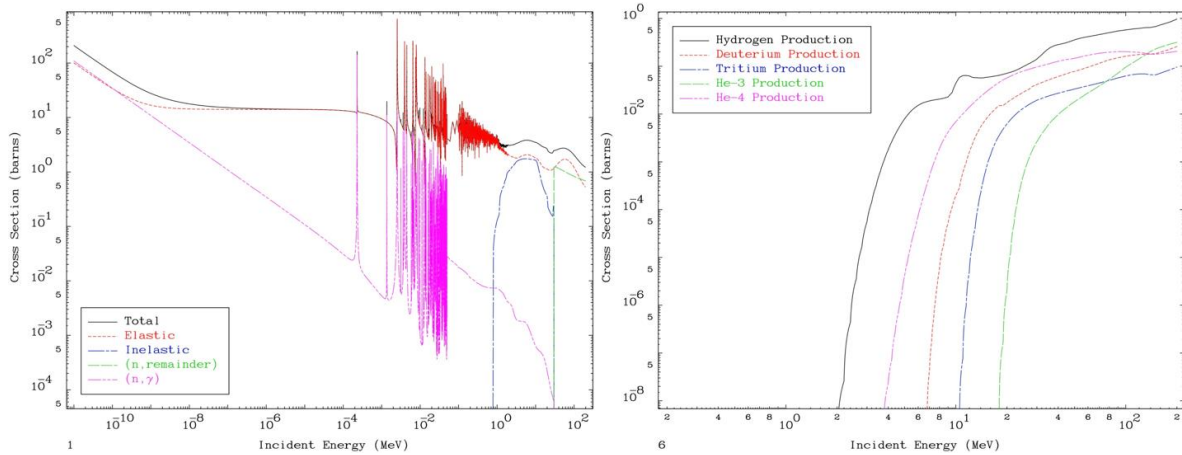


Figure 3: Cross-sections of main neutron-induced reactions on ^{65}Cu .

3. EXPERIMENTAL PROCEDURE

Experiments were performed at the Neutrons For Science (SPIRAL2/NFS) facility of GANIL (France) described in detail in the reference [10]. Samples of six materials in their natural isotopic compositions, (carbon, aluminum, steel, copper, tungsten and lead), selected for being abundantly present in the components of medical electron accelerators, were irradiated under the neutron flux produced at SPIRAL2/NFS in a particular configuration in order to match as close as possible to the expected neutron distribution at a medical electron accelerator which is rather continuous and Maxwellian-shaped up to a maximum energy of about 10 MeV as shown in Figure 2.

One of the methods used to produce neutrons at SPIRAL2/NFS is based on the $^9\text{Be}(d,Xn)$ reaction and is supposed to fulfill this criteria. Indeed, the measured double differential distributions from of the neutrons produced by these reactions in a thick target show a rather continuous behavior as presented in Figure 4. The flux values presented are obtained from the experimental neutron yields measured in the reference [11] and refer to the neutron production from the target irradiation meaning the values are corrected for the background contributions namely from neutron scattering in the surrounding environment.

However, the emission of neutrons at forward angles with respect to the incident deuterons is dominated by the direct break-up reaction giving a pronounced increase of the production probability around 20 MeV in the neutron spectrum. Direct reactions are however strongly related to the direction of the incident particle and therefore the break-up contribution is almost inexistent at angles above 90 degrees. The neutron distribution in the backward direction is therefore dominated by the evaporation process with a typical Maxwellian shape which is more in agreement with the irradiation conditions required for the present study. Figure 4 also shows the distributions of neutrons emitted at 0 and 90 degrees simulated in this work with the MCNP6.2 Monte-Carlo code which are in rather satisfactory agreement with the measurements from the reference [11]. This comparison allows to conclude that MCNP6.2 is relatively accurate in simulating the $^9\text{Be}(d,Xn)$ reactions being therefore suitable for the subsequent simulations for the needs of the present study as it will be discussed in the next section.

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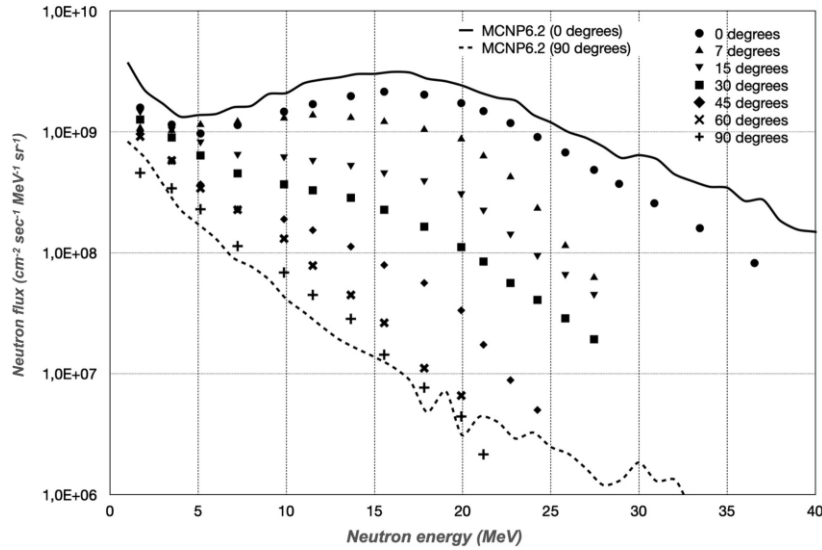


Figure 4: Double differential distributions of neutrons produced by the interaction of 40 MeV deuterons with a thick beryllium target. Data from the reference [11] (symbols) are compared to the MCNP6.2 simulations (curves).

Finally, MCNP simulated neutron energy distributions presented in Figure 5 illustrate the above-mentioned considerations on the similarities achieved, by the adequate selection of the position at 90 degrees with respect to the incident deuterons, between the neutron spectrum obtained at SPIRAL2/NFS and the one predicted in a copper part (conversion target support) of a VARIAN TrueBeam medical electron accelerator's head. The angle of 90 degrees has been selected as the best compromise allowing an enough decrease of the contribution from fast neutrons from the deuteron break-up while preserving a reasonably high neutron flux which would have been much lower at higher backward angles. Despite neutrons produced at SPIRAL2/NFS still being more energetic considering the nuclear reaction used for their production, one can consider the shapes of the two distributions close enough to simulate the neutron-induced mechanisms leading to the activation of materials during the operation of a medical electron LINAC by the activation of samples in the neutron flux produced at SPIRAL2/NFS.

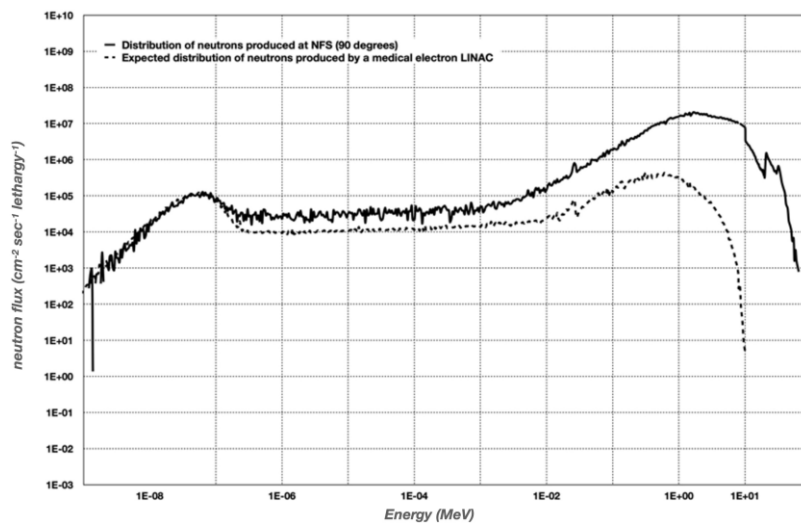
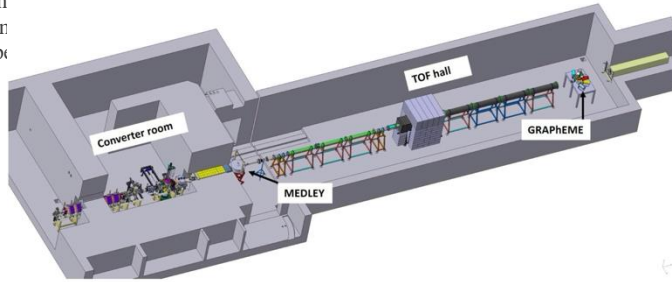


Figure 5: Simulated energy distribution of neutrons available for sample irradiation at SPIRAL2/NFS, compared to the estimated distribution of neutrons produced by a radiotherapy electron accelerator.

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A general layout of the SPIRAL2/NFS facility [10] is given in Figure 6. It is

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composed of two main areas,



Figure 6: Schematic view of the SPIRAL2/NFS facility and a picture of the inside of the converter cave.

After being exposed for a duration of 15 minutes in the converter room to neutrons with energy distribution as discussed above and at an average flux of the order 10^9 neutrons/cm²/sec (estimated from the deuteron beam current of 20 μ A monitored during the irradiation and the measured neutron yields from the reference [11]), the samples were quickly transferred through a pneumatic system [12] to the TOF area in front of a

Canberra coaxial n-type liquid nitrogen cooled high-purity germanium (HPGe) detector for the measurements of the γ spectra. Samples consisted in disks of 4 mm thick and 15 mm in diameter and traveled in small 3D-printed plastic containers fitted to the dimensions of the pneumatic pipes. Spectra were acquired during 15 minutes and began approximately 15 seconds after the irradiation ended, allowing the measurement of contributions from short-lived radio-isotopes. This is illustrated by the identification in the γ spectra discussed in detail in the next section of the radio-isotope ^{16}N (half-life of 7.13 seconds), produced by the $^{16}\text{O}(n,p)^{16}\text{N}$ reaction in the plastic material of the sample containers and the air trapped inside.

4. RESULTS AND DISCUSSION

4.1 Simulation of the experimental conditions

The experimental set-up has been simulated using the MCNP6.2 code with a detailed geometry model taking into account the target system including the beryllium conversion target and its copper cooler as well as all the elements present in the converter room strongly influencing the flux and the energy distribution of neutrons: the concrete shielding with walls, roof and ceiling, the magnets of the deuteron beam line, the neutron collimator separating the converter cave from the TOF hall. The simulations result in terms of 2D neutron flux map are presented in Figure 7. They allow the neutron flux and its energy distribution at the exact location where the samples were irradiated to be determined. As expected, based on the agreement with experimental data found in Figure 4, the value of the simulated neutron flux at the sample location agrees with the one estimated during the irradiation as discussed above of the order of 10^9 neutrons/cm²/sec. The neutron energy distribution has been calculated with MCNP6.2 using the specific CCFE scheme with 709 energy groups, allowing further activation calculations to be performed with the FISPACT-II code using the option of neutron-induced reactions cross-sections from the TENDL library.

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FISPACT-II allows us to accurately reproduce the irradiation and decay sequences as they were recorded during the experiments. Based on the neutron flux value and the energy distribution previously calculated by the simulations with the MCNP6.2 code at the sample location and considering the known irradiation duration, it uses the energy-dependent cross-section values from the TENDL library to calculate the yields of radio-isotopes produced in each sample by the neutron-induced nuclear reactions on the stable nuclei present in its initial composition. Decay data libraries are further used to calculate the weight of these radio-isotopes at the time after the sample irradiation corresponding to the γ spectrometry measurements. The calculations predictions, thus simulating as accurately as possible the real experimental conditions, can then be directly compared to the data obtained by the experiments performed in this work.

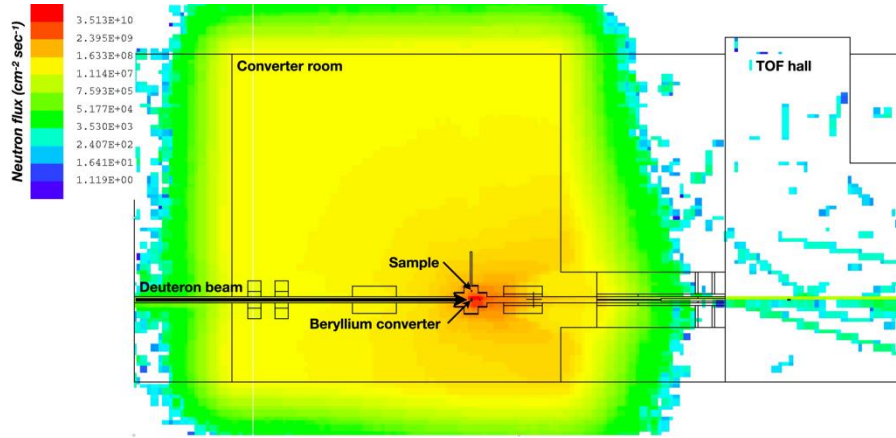


Figure 7: Spatial distribution of neutrons produced at SPIRAL2/NFS by the interaction of 40 MeV deuterons with the beryllium converter.

4.2 Analysis of the results

Gamma spectrometry measurements of the irradiated samples allowed to obtain the results presented in Figure 8. The energy calibration curve of the HPGe detector obtained using an europium (Eu) source is given in the inset of the figure. The resolution of the detector has been also determined from the measurements with the same source and ranges from 0.3% at the high energies to 3% at the low energies of the Eu spectrum.

For the carbon sample, no data above the background has been recorded suggesting that no radioactive isotope is created in significant amount in this material when exposed to a neutron energy distribution typical for a medical electron LINAC. The identification of the radio-isotopes from the energies of the peaks in the spectra for the activated samples has been performed based on state-of-the-art nuclear data library developed in the frame of the DDEP project (Decay Data Evaluation Project) and maintained and continuously updated by LNHB [13].

The analysis has been done for each sample individually. For the sake of clarity and to stay in line with the main objective of this work, being to explore the possibility to deeply studying the neutron activation mechanisms at electron medical accelerators by reproducing the conditions for irradiation of materials for this specific application using the neutron source produced at SPIRAL2/NFS, only one case is discussed in detail in this paper. The example we selected concerns the case of copper, as it is a material which can generally be always found in particle accelerators.

Several radio-isotopes were identified in the γ spectrum acquired after neutron irradiation of the copper sample (Figure 8), created from both ^{63}Cu and ^{65}Cu natural isotopes through various neutron-nucleus reaction channels as discussed below.

Other radio-isotopes were also present due to activated background. For instance, ^{28}Al , ^{27}Mg , and ^{24}Na were identified as created by (n,γ) , (n,p) , and (n,α) reactions respectively on ^{27}Al , as is the material constituting

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the walls of the tube in which neutrons are travelling in the TOF area. The origin of the three gamma peaks attributed at this stage to ^{116}In need further investigation, while the remaining peaks were associated to naturally occurring radio-isotopes such as ^{40}K and ^{208}Tl .

Activation calculation performed predicts the creation in the copper sample as irradiated at SPIRAL2/NFS of the ten radio-isotopes given in Table 1 for which the weight in the total activity is the highest. Highlighted radio-isotopes in the first column are the ones which were also identified by the analysis of the corresponding γ spectrum from Figure 8. Six radio-isotopes out of ten were found both experimentally and by calculations. $^{60\text{m}}\text{Co}$, ^{61}Co , ^{61}Cu and ^{63}Co produced respectively by the reactions $^{63}\text{Cu}(n,\alpha)^{60\text{m}}\text{Co}$, $^{65}\text{Cu}(n,n\alpha)^{61}\text{Co}$, $^{63}\text{Cu}(n,3n)^{61}\text{Cu}$ and $^{65}\text{Cu}(n,^3\text{He})^{63}\text{Co}$ were only predicted by FISPACT-II and not identified in the measured γ spectrum, three of them having however a negligible contribution to the calculated total activity of the sample.

The possible radio-isotope production paths from the main neutron interactions with the two natural copper isotopes are schematically shown in Figure 9. The two natural copper isotopes ^{63}Cu and ^{65}Cu are at the origin of creation of respectively 5 and 4 radio-isotopes through various neutron-induced reaction channels indicated in the figure and those probabilities are discussed below individually. It is worth mentioning the case of ^{60}Co and ^{63}Ni which can be produced in theory respectively by the reactions $^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$ and $^{63}\text{Cu}(n,p)^{63}\text{Ni}$ but were neither identified by γ spectrometry nor calculated by FISPACT-II. This is explained by the irradiation times used in our case which at 15 minutes were too short to produce a significant amount of these radio-isotopes given their long half-life of 5.27 years and 101.2 years respectively.

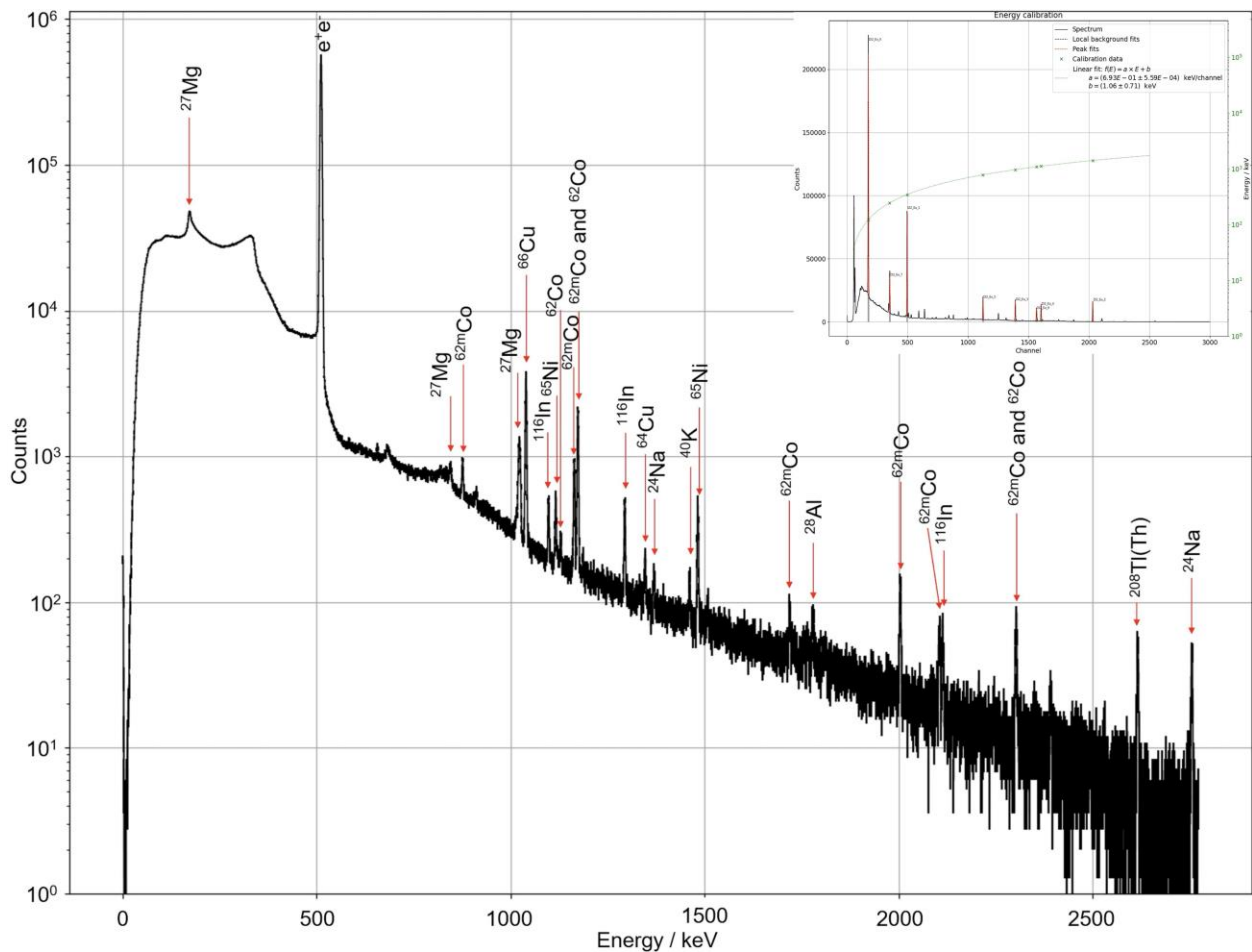


Figure 8: Measured γ spectrum for the neutron irradiated copper sample after background subtraction.

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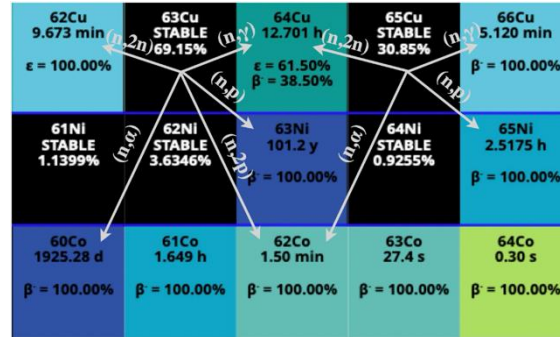
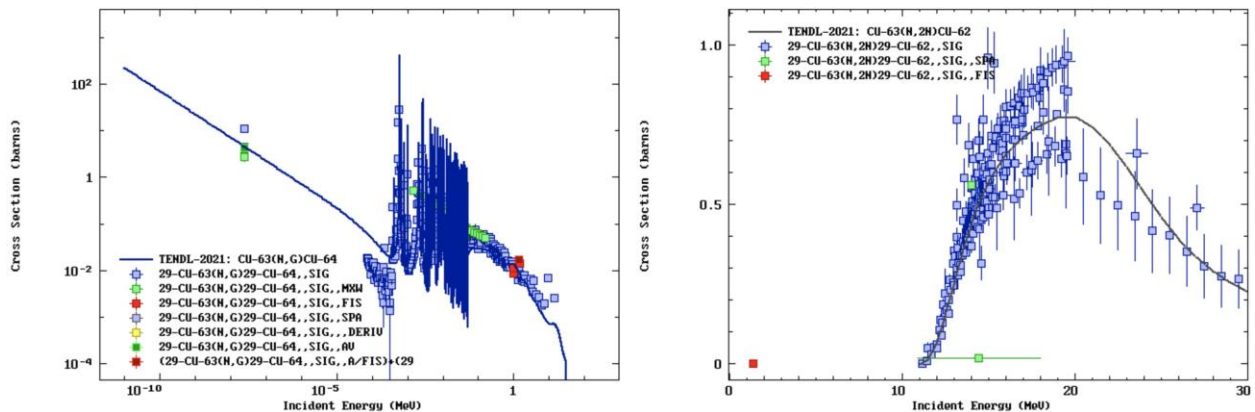


Figure 9: Main neutron-induced reactions leading to the production of radio-isotopes in copper.

In order to test the accuracy of the calculation by the code FISPACT-II of the radio-isotopes production probability, we further analyzed the corresponding neutron-induced reactions cross-sections behavior and compared the values from the TENDL library used by FISPACT-II with the data available in the literature. As shown in Figure 10, among the three neutron-induced reactions on the ^{63}Cu natural isotope producing measurable radio-isotopes given our irradiation times, the available experimental cross-sections found in EXFOR database for the reactions $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$ and $^{63}\text{Cu}(n,2n)^{62}\text{Cu}$ are relatively abundant and the evaluated cross-sections from TENDL library show globally a good agreement despite discrepancies being identified in some neutron energy domains. In the case of the other possible reaction $^{63}\text{Cu}(n,2p)^{62}\text{Co}$ no data was found in the literature which doesn't allow to test the accuracy of the cross-section values from TENDL library in this particular case.

Concerning the reactions on ^{65}Cu (Figure 11), experimental cross-sections are available for all of them and the evaluated values from the TENDL library are in some cases in significant disagreement, namely for the reaction $^{65}\text{Cu}(n,\alpha)^{62}\text{Co}$, for the reaction $^{65}\text{Cu}(n,p)^{65}\text{Ni}$ at neutron energies below 10 MeV and for the reaction $^{65}\text{Cu}(n,\gamma)^{66}\text{Cu}$ in the thermal neutrons region. A very good agreement was found on the other hand for the reaction $^{65}\text{Cu}(n,2n)^{64}\text{Cu}$.

The contribution of each reaction to the total radio-isotope yield when several reactions are at the origin of its production are also given in the figures as the discrepancies found between the evaluated cross-sections used to calculate it and the experimental values may have a significant impact on the accuracy of the calculated radio-isotope inventory. A relevant example on this matter concerns the radio-isotope ^{62}Co which is produced mainly (at 70%) by the reaction $^{65}\text{Cu}(n,\alpha)^{62}\text{Co}$ for which evaluated cross-sections are in significant disagreement with the experimental values.



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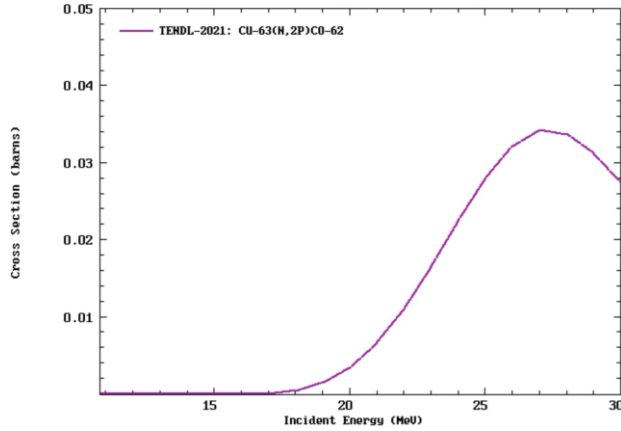


Figure 10: Cross-sections of the neutron-induced reactions on ^{63}Cu from the TENDL library used by the code FISPACT-II (continuous lines) compared with the experimental values available in the literature (symbols).

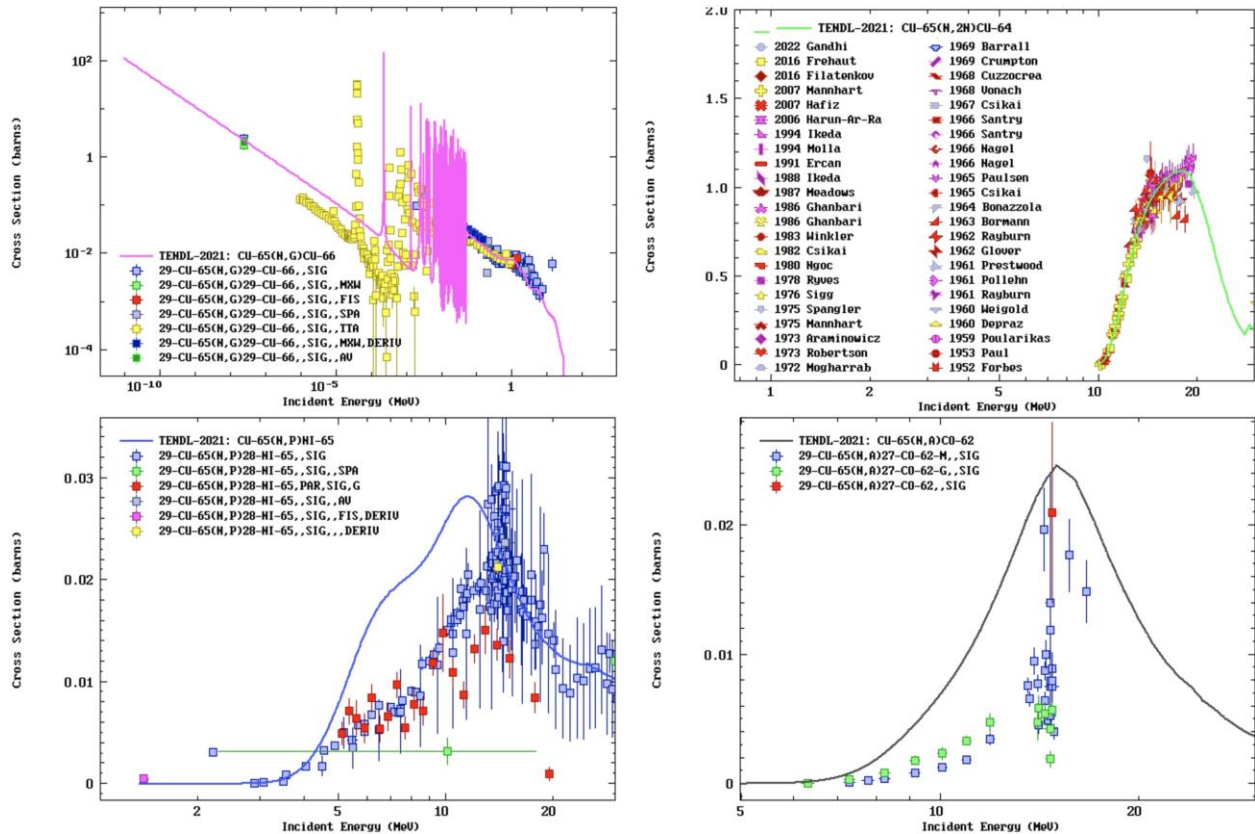


Figure 11: Cross-sections of the neutron-induced reactions on ^{65}Cu from the TENDL library used by the code FISPACT-II (continuous lines) compared with the experimental values available in the literature (symbols).

4.3 Assessment of activation calculations accuracy

Provided the sufficient knowledge of the experimental neutron flux and its energy distribution as well as the sample composition and the irradiation-decay sequences when calculating the radioactivity induced by neutrons in the samples with the FISPACT-II code, the main source of uncertainty affecting the calculations results is related to the reaction cross-sections used to determine the probability of production of each radio-

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isotope. The experimental conditions in which the samples were irradiated at SPIRAL2/NFS and subsequently analyzed by gamma spectrometry are well-known and faithfully reproduced in the simulations performed in this work as discussed in the subsection 4.1. The next step was the estimation of the calculation accuracy based on the comparisons concerning the neutron-induced reactions cross-sections, between the values from the TENDL library used by FISPACT-II and the experimental data available in the literature (Figures 10 and 11).

The method proposed in this work consists of the analysis by a specially developed algorithm for the experimental and evaluated cross-section values represented in Figures 10 and 11. The first step of the method consisted in calculating the existing gaps in terms of cross-section value for each of the neutron-induced reactions presented in the plots, by simply comparing the evaluated cross-sections from the TENDL library to the measured ones. Since not all the neutrons used for sample irradiation at SPIRAL2/NFS are contributing equally to the global spectrum (Figure 5), the previously deduced cross-section gaps were subsequently weighted for each neutron energy bin of the spectrum to account for its actual contribution to the given reaction. The so obtained weighted gaps are therefore reflecting, for each neutron energy bin, the cross-section related inaccuracy of the calculated production probabilities of the radio-isotopes by the code FISPACT-II. Then for a given radio-isotope, the resulting global uncertainty affecting its total production is obtained by integration over the energy range of these energy dependent uncertainties.

The values are given in the last column of Table 1 and turned out to be significant as they range from 7% to around 28%. The highest uncertainty concerns the extreme case of ^{62}Co production by the reaction $^{63}\text{Cu}(n,2p)^{62}\text{Co}$ characterized by a complete lack of experimental cross-section values and for which the uncertainty has been consequently maximized.

Table 1: Radio-isotopes created by neutron-induced reactions in the copper sample.

	Weight (%)	Calculation uncertainty
^{62}Cu	0,59	7,65 %
^{66}Cu	0,28	7,50 %
$^{60\text{m}}\text{Co}$	0,07	17,20 %
^{64}Cu	0,03	4,83 %
^{62}Co	0,01	27,80 %
$^{62\text{m}}\text{Co}$	0,006	17,32 %
^{65}Ni	0,003	23,93 %
^{61}Co	0,002	17,40 %
^{61}Cu	0,0014	9,13 %
^{63}Co	0,0002	16,27 %

5. CONCLUSIONS AND PERSPECTIVES

The need for a well-consolidated method to accurately determine the radioactive inventory created by neutron-induced activation in accelerators components is increasingly growing. For radiotherapy accelerators this is motivated by their specificity in terms of materials used and operating regimes but also from the point of view of their use as medical devices. The technology related to radiotherapy is continuously improving in order to increase the quality of treatments and so the lifetime of a medical accelerator is relatively short. This leads to relatively low activity levels in their components and makes them good candidates for waste recycling but this can only be done on the basis of an accurate knowledge of the radio-isotopes created.

Experimental assessment and analysis of calculations accuracy for the neutron-induced radio-isotope creation in copper parts of radiotherapy accelerators

The first measurements performed using neutrons produced at the SPIRAL2/NFS facility presented in this work are related to this specific application, but they must be seen as the first stage of a broader scientific program to be continued in the future.

New macroscopic neutron activation data of copper have been measured and compared to the predictions of the activation calculations code FISPACT-II in terms of radioactive species being produced. Although the main contributing radio-isotopes to the total activation predicted by the code were also identified from the gamma spectrometry of the irradiated sample, we showed that their actual calculated yields can be significantly inaccurate when the probabilities of the responsible neutron-induced reactions are in strong disagreement with the experimental values or when the later are not at all available in the existing literature. This finding underlines the need for new data to be measured allowing to increase the predictive power of the calculation codes and consequently the accuracy that can be reached by the simulation-based methodologies used to assess the radioactive inventory of electron accelerators in particular.

The measurements performed in the present work constitutes a first step in this direction. Despite the unavailability for this first experiment of accurate data namely on the efficiency of the HPGe detector used for γ spectrometry, mandatory for a proper determination of radio-isotopes yields from the measured γ spectrum, a relative approach for the radio-isotope production was still possible. Moreover, the strong focus on the identification of their creation paths allowed a detailed analysis of the associated probabilities-related accuracy that can be presently achieved by the activation calculations. These results are therefore above all useful in pointing out particular reactions that need further improvement in terms of cross-section data and by doing so, additional measurement and evaluation needs to be further addressed in order to produce new data for the integration in the computational tools. The final purpose is to improve the accuracy of the codes used in the methodologies based fully on simulations to assess the activation of accelerator components, as it is presently the only approach available and therefore extensively used by the community allowing a complete characterization of facilities in terms of their residual radiological impact.

Finally, this work provided evidence that the neutron beams available at SPIRAL2/NFS offer enough flexibility in terms of energy distribution, considering the kinematics of the reactions used for their production, to cover a wide range of application related needs in terms of neutron-induced radioactivity. Other specific needs can thus be considered, related to the deep study of neutron-induced activation in other materials of interest such as silicon, iron, cobalt, nickel, or niobium. Other extensively used accelerator-based applications characterized by different energy spectra of neutrons produced can also be addressed, such as cyclotrons for the production of isotopes for nuclear medicine. Large-scale research facilities using particle accelerators such as synchrotrons and spallation sources also have specific needs in terms of neutron-induced activation, especially when considering the large quantities of potentially activated materials.

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