# DIN - DEPARTMENT OF INDUSTRIAL ENGINEERING

## PhD Course in Mechanics and Advanced Engineering Sciences (DIMSAI)

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# *ACTIVATION AND DECOMMISSIONING OF BIOMEDICAL CYCLOTRONS*

## Introduction

The applications of particle accelerators in biomedical field has grown significantly in the last two decades and is still growing, especially for cancer diagnosis and treatment. During the operational lifetime accelerators and their surrounding structures are activated by primary and secondary particles, in a longer term this represent a decommissioning issue. Only in recent years the attention on the generation of radioactive waste and on radiological hazards associated with decommissioning started to be relevant. Regulations today require that decommissioning must be considered as part of the design and planning phase of an accelerator facility. Nevertheless there are no specific international standards or guideline documents and cases of accelerator decommissioning have been sporadically described in the technical literature.

This work is focused on PET cyclotrons facility activation assessment. In Italy there are 36 PET cyclotrons (Figure 1), most of these have been in use for 10-15 years, it is therefore expected that in the coming years some events of decommissioning or partial decommissioning for the replacement of some components, will take place. When considering the dismantling of a PET cyclotron facility considerable amount of low level solid radioactive waste has to be characterized and disposed of. Secondary neutrons, generated during the routinely production of $^{18}F$ by the $^{18}O(p,n)^{18}F$ reaction, are mainly responsible for activation. The level of activity produced varies considerably, depending on the type of accelerator, on its use and on the specific structure of the bunker, for this reason, each facility needs a specific decommissioning strategy. This work aimed at developing a Monte Carlo approach to preliminary assessing activation in order to define an "ad hoc" decommissioning strategy.

Figure 1 – PET cyclotron facilities in Italy

## Materials & Methods

In this work two main case study were analysed (Table 1): The GE PETtrace facility of Sant'Orsola-Malpighi Hospital (Bologna) and the IBA CYCLONE 18/9 facility of Inselspital (Bern). The Monte Carlo code FLUKA was used to accurately modelled the two facilities for activation assessment. Activity was scored at different positions, depth and for different life expectancy of cyclotrons.

Different campaigns of experimental measurements of neutron fluence, neutron spectrometry and materials activation were performed in order to compare and to prove the consistency between simulated and experimental results.

Table 1 – Main features of the two case study analysed

| | Energy | Acceleration plan | Fluorine targets | N° of irradiations per day | Typical irradiations conditions | Workload in a year | Installation year |
|---|---|---|---|---|---|---|---|
| PETtrace | 16.5 MeV | Vertical | 1 | 1 | 60 µA for 100 min | ≈ 25000 µAh | 2002 |
| CYCLONE 18/9 | 18 MeV | Horizontal | 4 | 3 | 70 µA for 90 min | ≈ 59000 µAh | 2012 |

## Monte Carlo Models

### GE PETtrace facility of Sant'Orsola-Malpighi Hospital (Bologna)

### IBA CYCLONE 18/9 facility of Inselspital (Bern)

**Geometry**

Detailed MC models of Cyclotrons and bunkers were realized in order to allow for an accurate transport of neutrons. The models includes all the major components of the cyclotrons that are expected to interact with the generated particles.

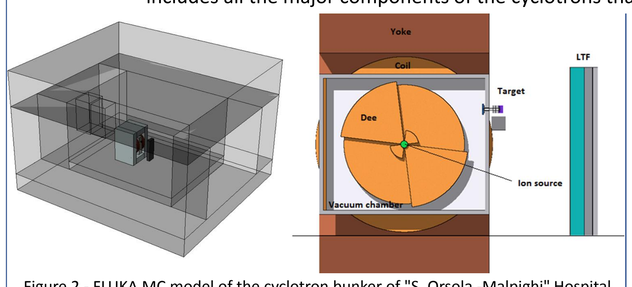

Figure 2 - FLUKA MC model of the cyclotron bunker of "S. Orsola -Malpighi" Hospital

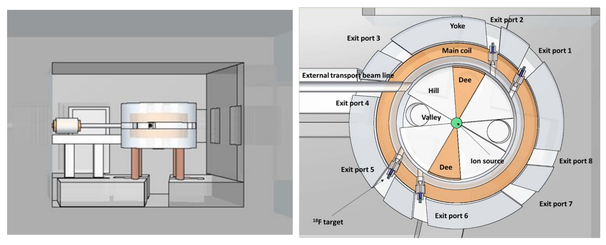

Figure 3 - FLUKA MC model of the cyclotron bunker of Inselspital

**Source**

Because of the rather low neutron production rate an efficient approach was developed: 1) secondary neutrons released by the target in proton irradiation were studied in a separate and simplified simulation; 2) the neutron spectrum obtained was then used directly as the source term using an external subroutine

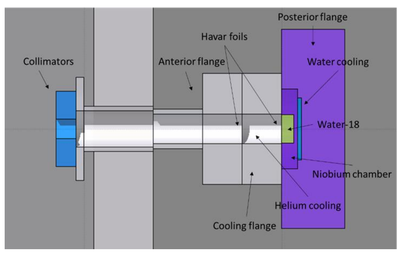

Figure 4 - MC model of the GE niobium target assembly

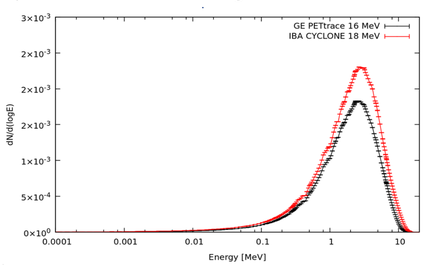

Figure 5 - Comparison between neutron energy spectra generated by the $^{18}O(p,n)^{18}F$ reaction in PETtrace target (in black) and in CYCLONE target (in red)

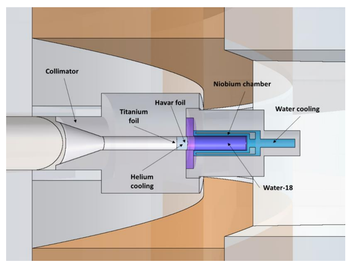

Figure 6 - MC model of IBA NirtaR Liquid Target large volume

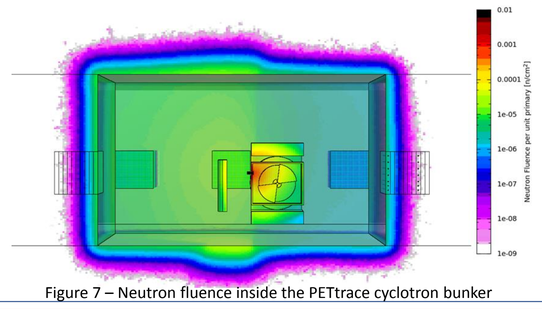

Figure 7 – Neutron fluence inside the PETtrace cyclotron bunker

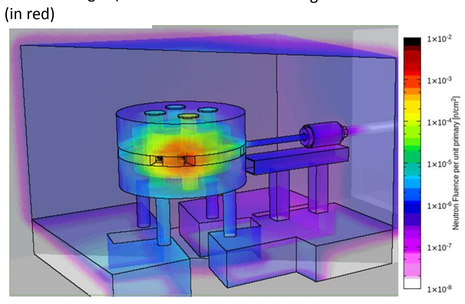

Figure 8 – Neutron fluence inside the CYCLONE cyclotron bunker

## Experimental Measurements

### GE PETtrace facility of Sant'Orsola-Malpighi Hospital (Bologna)

**Neutron dosimetry**

Measurement of the neutron ambient dose equivalent H*(10) in 12 points (Figure 9) were performed using:

- a neutron rem-counter FHT-757 (Thermo Scientific) provided with a $BF_3$ proportional-counter and a PE-moderator
- A set of 12 TLD dosimeters, type CR39

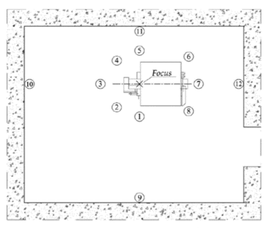

Figure 9 – Experimental setup

**Non-destructive activation assessment using a portable CZT detector**

As long as an accelerator is operational, experimental measurements for material activation assessment are problematic due to the impossibility, in most cases, to perform core drilling. For this reason we define a non-destructive in-situ measurement methodology using a very compact, USB powered, CdZnTe (CZT) detector: Kromek GR1 Gamma Ray Spectrometer™ (by Kromek™ , Sedgefield, UK). The efficiency calibration of the detector in non-standard geometries, which represent a critical aspect for this kind of measurements, was performed via Monte Carlo simulations.

Figure 10 – Kromek GR1

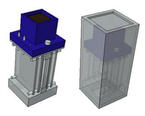

Figure 11 – MC model

Figure 12 – Experimental setup

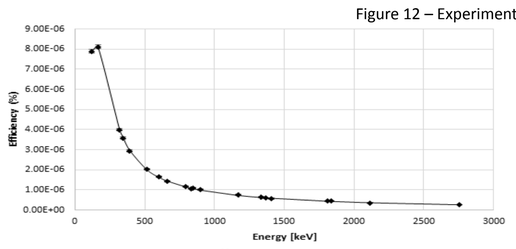

Figure 13 – Efficiency calibration curve

### IBA CYCLONE 18/9 facility of Inselspital (Bern)

**Neutron spectrometry**

Neutron spectrometry was performed using a set of six Bubble Detector Spectrometer (BDS, manufactured by BTI Bubble Technology Industries) with six different energy threshold (10, 100, 600, 1000, 2500, 10000 keV). Four different measurements were performed irradiating the four different targets and placing the detectors in front of the irradiated target. An iterative unfolding procedure was developed to obtain an estimation of neutron spectral distribution.

Figure 14 – Bubble detector

Figure 15 – Experimental setup

**Bunker sample activation assessment using an HPGe detector**

During the maintenance of CYCLONE 18/9 it was possible to perform three core drilling on the bunker walls. Those samples were then measured in gamma spectroscopy with an HPGe detector for activation assessment. Dedicated analysis sequences and nuclide Libraries were created using the spectroscopy software Genie™ 2000 for spectra analysis. The calibration of the detector was performed using the well known and validated LabSOCS code.

Figure 16 – Core drilling

Figure 17 – Concrete sample

Figure 18 – HPGe detector

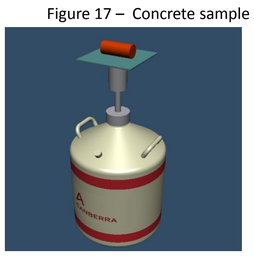

Figure 19 – LabSOCS geometry composer for efficiency calibration

## Results

In the following some of the main results are reported. Experimental and simulated results prove to be consistent, discrepancies in activation assessment are mainly due to the fact that concrete activation is strongly dependent on trace element concentration that is heterogeneous and usually unknown. The literature values used in simulations could be quite different respect to real trace element concentration. Nevertheless aim of this work was to define a methodology to preventively have an idea of the order of magnitude of the activation level depending on materials, positions, bunker design and type, workload, life expectancy of the cyclotron.

### GE PETtrace facility of Sant'Orsola-Malpighi Hospital (Bologna)

Table 2 – Comparison of neutron ambient dose equivalent obtained with simulation and experimental measurements

| Position | Dose ± Uncertainty [mSv/µAh] | | |
|---|---|---|---|
| | FHT-752 | CR39 | FLUKA |
| 1 | 330 ± 130 | 380±200 | 308.8±0.5 |
| 2 | 430±170 | 300±160 | 464.4±0.6 |
| 3 | 90±40 | 60±30 | 97.82±0.26 |
| 4 | 420±70 | 340±180 | 480.0±0.6 |
| 5 | 330±130 | 340±180 | 330.9±0.5 |
| 6 | 27±11 | 6±5 | 20.87±0.11 |
| 7 | 32±13 | 19±14 | 75.69±0.24 |
| 8 | 23±9 | 6±5 | 18.87±0.10 |
| 9 | 55±22 | 31±19 | 52.93±0.19 |
| 10 | 45±18 | 38±22 | 46.61±0.17 |
| 11 | 190±80 | 130±70 | 169.6±0.3 |
| 12 | 22±9 | 50±30 | 38.00±0.17 |

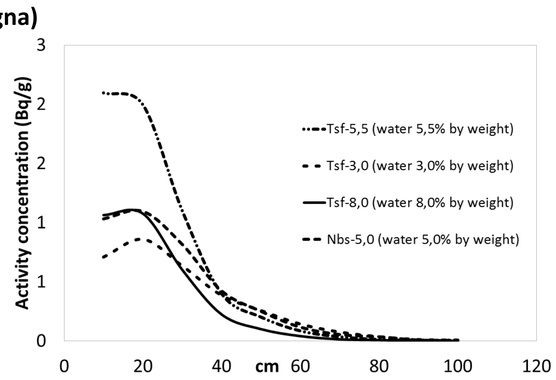

Figure 20 – Comparison of $^{152}Eu$ in-depth activation profiles obtained varying concrete composition. Compositions chosen for this evaluation were taken from the reference neutron shielding materials of Report NCRP 51.

Table 3 – Comparison of activity concentrations obtained with simulation and experimental non-destructive measurements using Kromek GR1

| Nuclide | Activity concentration ± Uncertainty [Bq/g] | | | | | |
|---|---|---|---|---|---|---|
| | Wall 1 | | Wall 2 | | Wall 3 | |
| | Kromek | FLUKA | Kromek | FLUKA | Kromek | FLUKA |
| $^{152}Eu$ | 0.12 ± 0.13 | 0.75 ± 0.04 | 0.36 ± 0.11 | 1.71 ± 0.07 | < 0.04 | 0.80 ± 0.04 |
| $^{60}Co$ | 0.26 ± 0.03 | 0.21 ± 0.01 | 0.18 ± 0.02 | 0.48 ± 0.01 | 0.04 ± 0.01 | 0.13 ± 0.02 |
| $^{54}Mn$ | 0.25 ± 0.03 | 0.06 ± 0.01 | 0.08 ± 0.01 | 0.20 ± 0.01 | < 0.01 | 0.002 ± 0.001 |
| $^{134}Cs$ | 0.06 ± 0.01 | 0.01 ± 0.01 | 0.07 ± 0.01 | 0.03 ± 0.01 | < 0.01 | 0.01 ± 0.01 |

### IBA CYCLONE 18/9 facility of Inselspital (Bern)

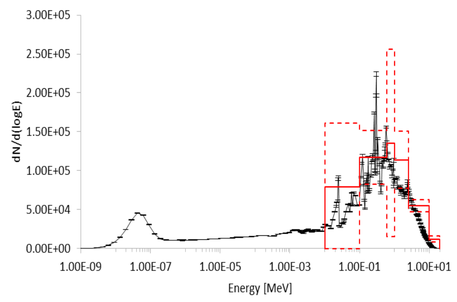

Figure 20 – Comparison of neutron spectra obtained with Fluka (in black) and bubble detectors (in red) with related uncertainties

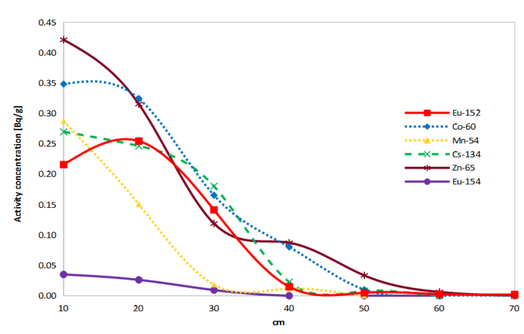

Figure 21 – Example of in-depth activation profiles in concrete of the main radionuclides

Table 3 – Comparison of activity concentrations obtained with simulation and HPGe measurements

| Nuclide | Activity concentration ± Uncertainty [Bq/g] | | | | | |
|---|---|---|---|---|---|---|
| | Sample 1 | | Sample 2 | | Sample 3 | |
| | HPGe | FLUKA | HPGe | FLUKA | HPGe | FLUKA |
| $^{152}Eu$ | 0.103 ± 0.002 | 0.101 ± 0.013 | 0.612 ± 0.005 | 0.217 ± 0.010 | 0.406 ± 0.008 | 0.207 ± 0.017 |
| $^{60}Co$ | 0.171 ± 0.005 | 0.150 ± 0.010 | 0.248 ± 0.008 | 0.348 ± 0.031 | 0.214 ± 0.006 | 0.360 ± 0.011 |
| $^{54}Mn$ | 0.008 ± 0.001 | 0.008 ± 0.052 | 0.145 ± 0.006 | 0.208 ± 0.026 | 0.083 ± 0.003 | 0.366 ± 0.010 |
| $^{134}Cs$ | 0.032 ± 0.001 | 0.033 ± 0.023 | 0.218 ± 0.004 | 0.218 ± 0.053 | 0.209 ± 0.004 | 0.228 ± 0.013 |
| $^{65}Zn$ | 0.017 ± 0.001 | 0.104 ± 0.013 | 0.114 ± 0.004 | 0.367 ± 0.013 | 0.056 ± 0.005 | 0.393 ± 0.015 |
| $^{154}Eu$ | 0.013 ± 0.001 | 0.001 ± 0.071 | 0.072 ± 0.002 | 0.013 ± 0.001 | 0.053 ± 0.002 | 0.018 ± 0.001 |

## Conclusions

The level of induced radioactivity of a cyclotron and of its bunker varies considerably from case to case and depends strongly on the type of accelerator, on its use and on the specific structure of the bunker. For this reason, the definition of a methodology for preliminary activation assessment is fundamental for the identification of an ad hoc decommissioning strategy. In comparison to outdated, troublesome analytic methods, MC simulations allow to accurately reproduce the real geometry of an accelerator inside its bunker. This is of the uttermost relevance both in the design phase, to achieve an optimal site planning, and at the time of decommissioning, to allow proper definition of an "ad hoc" strategy.

In conclusion, provided a detailed model is developed, Monte Carlo approach to the study of bunker activation level allows to characterize with satisfactory accuracy the radioactive waste from the decommissioning of a specific cyclotron facility and to identify any critical issue or possible countermeasures to be taken in order to decrease future dismantling costs. Nevertheless Monte Carlo assessment must always be validated by experimental measurements to prove the reliability of results.



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IL SEGUENTE MATERIALE È RISERVATO AL PERSONALE DELL'UNIVERSITÀ DI BOLOGNA E NON PUÒ ESSERE UTILIZZATO AI TERMINI DI LEGGE DA ALTRE PERSONE O PER FINI NON ISTITUZIONALI