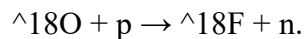


Viability of Medical Cyclotron as Gamma Radiation Source for Research on Nuclear Materials

Lola O'Regan

Radiation-induced changes in the corrosion behaviour of nuclear materials is an important consideration in the development of materials used in applications like the storage of nuclear waste in a deep geologic repository. Exposure to ionizing radiation can alter the chemical environment surrounding a material and influence corrosion through processes like radiolysis. Unfortunately, conventional radiation sources like research reactors and dedicated cobalt-60 GammaCells can be expensive to construct and maintain. Medical cyclotrons, however, are widely available because of their use in medical isotope production. Approximately 228 research reactors, 500 ^{60}Co GammaCells, and 1500 cyclotron facilities are estimated to exist worldwide, highlighting the potential accessibility of cyclotrons as alternative radiation sources for nuclear materials research.¹ In this project, the viability of a medical cyclotron as a radiation source for nuclear materials research is explored. In particular, the project focused on characterizing the radiation environment surrounding the GE PETtrace 880 cyclotron at the St. Joseph's Hospital Cyclotron Facility, quantifying absorbed radiation dose using Fricke dosimetry, and determining whether the absorbed dose could be correlated with the activity of the cyclotron target. A further objective was to establish experimental methods for investigating radiation-induced corrosion of carbon steel and SS316L stainless steel and to compare cyclotron irradiation with irradiation from a conventional ^{60}Co GammaCell.

Medical cyclotrons are primarily used to produce short-lived radionuclides for medical imaging such as fluorine-18, widely used in positron emission tomography (PET).² It is produced by bombarding an oxygen-18 target with protons, releasing fast neutrons according to the equation



In addition to the primary nuclear reaction, secondary reactions occurring in and around the target produce additional radiation, including gamma radiation. The resulting environment is therefore not equivalent to irradiation by a pure gamma source. Previous work by Jeffries *et al.* demonstrated that medical cyclotrons used for ^{18}F production can generate substantial fast-neutron fields, which

were characterized during ^{18}F production using activation foils and Monte Carlo simulations.³ However, limited work has focused specifically on absorbed gamma radiation dose within the cyclotron and its potential relationship to the corrosion of nuclear materials via radiolysis.

Fricke dosimetry was used to quantify the absorbed radiation dose within the cyclotron environment. A Fricke dosimeter is a chemical dosimeter based on the radiation-induced oxidation of ferrous ions, Fe^{2+} , to ferric ions, Fe^{3+} . The concentration of Fe^{3+} generated by irradiation can be determined spectrophotometrically from its absorbance at 304 nm. This provides a method of relating a measurable chemical change to the energy deposited by ionizing radiation.

The absorbed dose was calculated according to

$$D = A / (\epsilon x p G(\text{Fe}^{3+}))$$

where D is the absorbed dose in gray (J kg^{-1}), A is the absorbance measured at 304 nm, ϵ is the molar extinction coefficient, x is the optical path length, p is the solution density, and $G(\text{Fe}^{3+})$ is the radiation chemical yield for ferric-ion production.

Fricke dosimetry was performed in 3-day intervals over the course of 1 month, giving 10 measurements. The measured absorbed doses varied considerably depending on the irradiation targets used during the production of ^{18}F by the facility. The values ranged from 20 to 436 Gy, with a total absorbed dose of 1855 Gy over 1 month (Figure 1). Blank samples of the unirradiated Fricke solutions were also measured as a control.

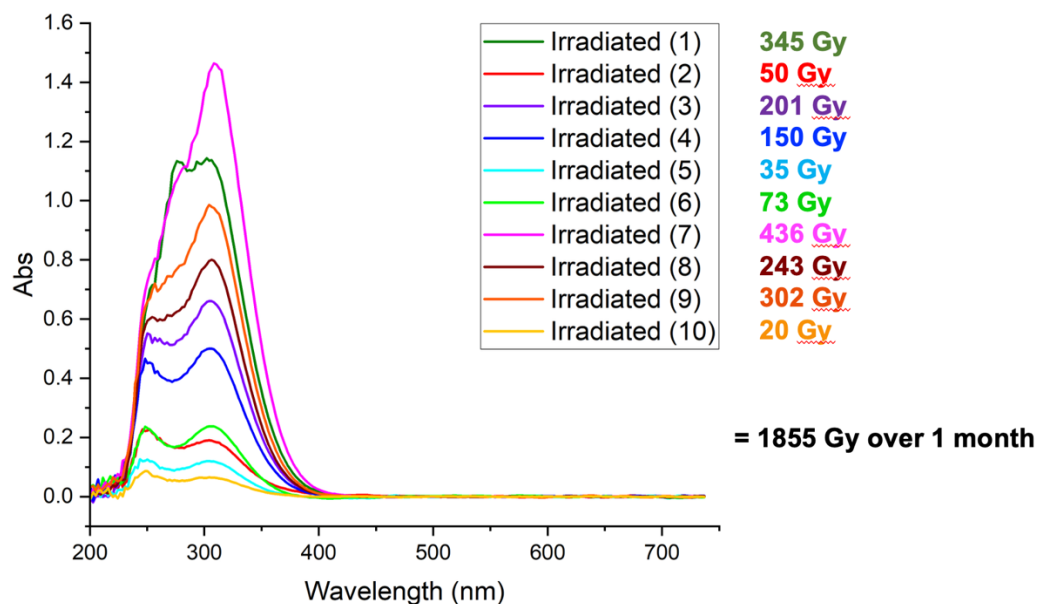


Figure 1. Fricke dosimetry data of samples exposed to 3 days of radiation within the cyclotron.

After establishing the absorbed dose using Fricke dosimetry, the next objective was to determine whether the dose could be related to the activity of the cyclotron targets. Target activity values recorded in the cyclotron log obtained from St. Joseph's cyclotron facility were compared with the corresponding Fricke measurements. The primary factor that seemed to be contributing to the large variation in absorbed doses was the usage of either target 1 or target 4 during ^{18}F production. Due to the geometry of the targets in relation to the sample (Image 1), target 1 could deliver a much higher dose to the sample. When just considering the activity of target 1 compared to the absorbed dose, the data produced a linear relationship described by $y = 0.0114x + 35.18$ with an R^2 value of 0.8314 (Figure 2). However, the relationship should be interpreted as an empirical correlation for the conditions investigated rather than as a universally applicable dose-prediction model. The geometry of the target and sample location is of critical importance in this case. The activity-dose relationship is useful because target activity is routinely recorded during cyclotron operation. If the relationship is validated through additional measurements, it could allow future experiments to estimate absorbed dose from cyclotron operating data without requiring Fricke dosimetry for every exposure. This would simplify the planning of prolonged irradiation experiments and allow samples to be exposed according to a predetermined target dose.

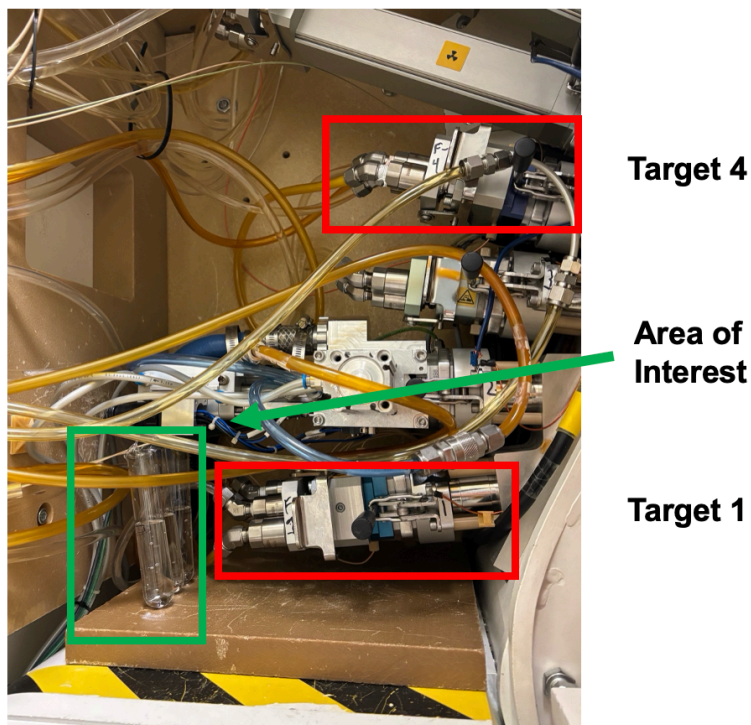


Image 1. Depiction of cyclotron targets and sample placement.

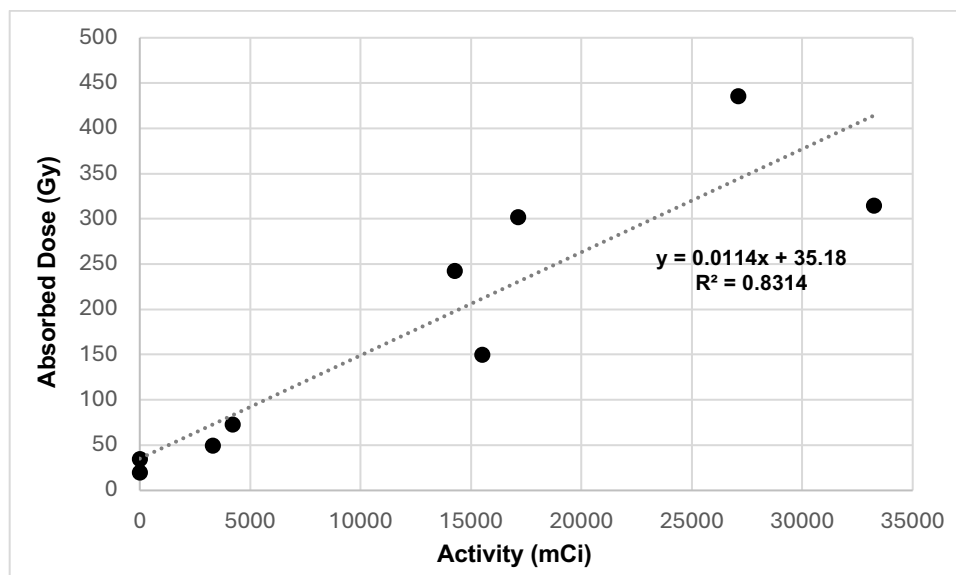


Figure 2. Absorbed dose vs. calculated activity of target 1.

An important question was whether the dose achievable in the cyclotron was sufficient to produce measurable changes in corrosion behaviour. Other work in the literature demonstrates that radiation-induced corrosion effects can occur at substantially higher absorbed doses than the approximately 1855 Gy accumulated during the initial cyclotron experiment. Aljohani *et al.*

reported that absorbed doses of 75 kGy decreased the corrosion resistance of austenitic stainless steel, while doses of 150 and 250 kGy increased corrosion resistance under their experimental conditions.⁴ Smart *et al.* reported that an absorbed dose of 44 kGy increased corrosion of carbon steel, with the observed response depending on dose rate. Winsley *et al.* reported no difference in corrosion of steels irradiated to 138–150 kGy at a lower dose rate of 25 Gy/h.⁵ The approximately 1855 Gy accumulated during the initial cyclotron experiment is below this range. The primary purpose of the initial exposure was therefore to establish the radiation environment and experimental methodology rather than to conclusively demonstrate radiation-induced corrosion.

A ⁶⁰Co GammaCell was used to provide a controlled gamma-radiation exposure for comparison with the cyclotron. The GammaCell operated at a dose rate of 533.8 Gy/h, and an irradiation time of 3.5 h produced a total absorbed dose of approximately 1868 Gy. This dose was selected because it was comparable to the approximately 1855 Gy accumulated in the cyclotron over approximately one month.

All standardized corrosion samples were prepared from carbon steel and SS316L stainless steel, selected as representative materials for investigating the effects of radiation on corrosion behaviour. Samples were cut to 0.5 × 0.5 × 1 cm and were manually polished on all sides to 1 μm before being imaged and weighed. Each sample was then placed in a 50 mL test tube containing 25 mL of 0.1 M NaCl solution and an approximately equal volume of headspace and then sealed by fusing the upper glass lip into an ampule.

Following GammaCell irradiation, corrosion was assessed using mass loss and ICP-OES analysis. Mass loss measurements were taken after removing the sample from solution and cleaning excess corrosion products from the surface (Figure 3). ICP – OES was performed on both the decanted solution from the ampule and the cleaning solution. Solid corrosion products were digested with acid (Figure 4).

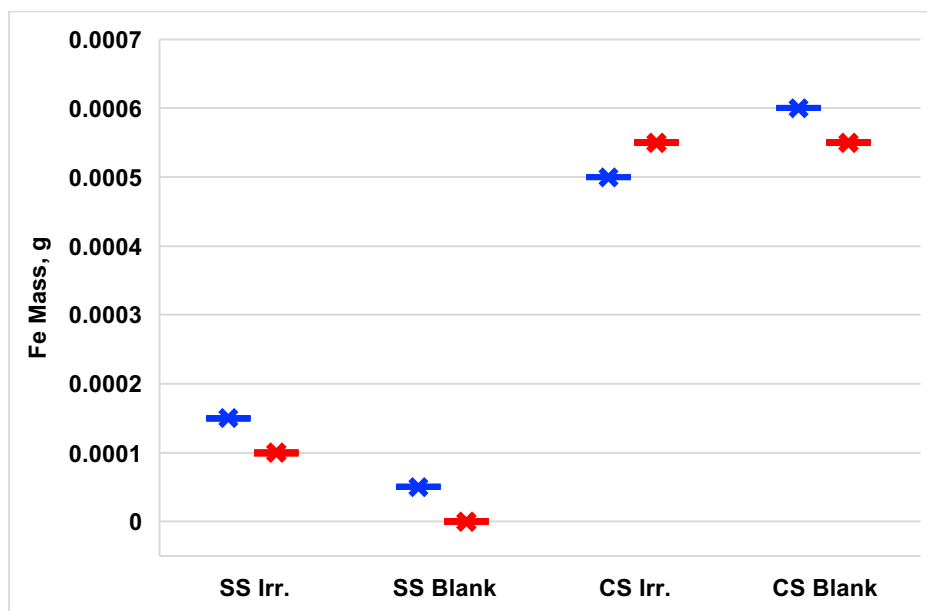


Figure 3. Mass loss measurements after GammaCell irradiation

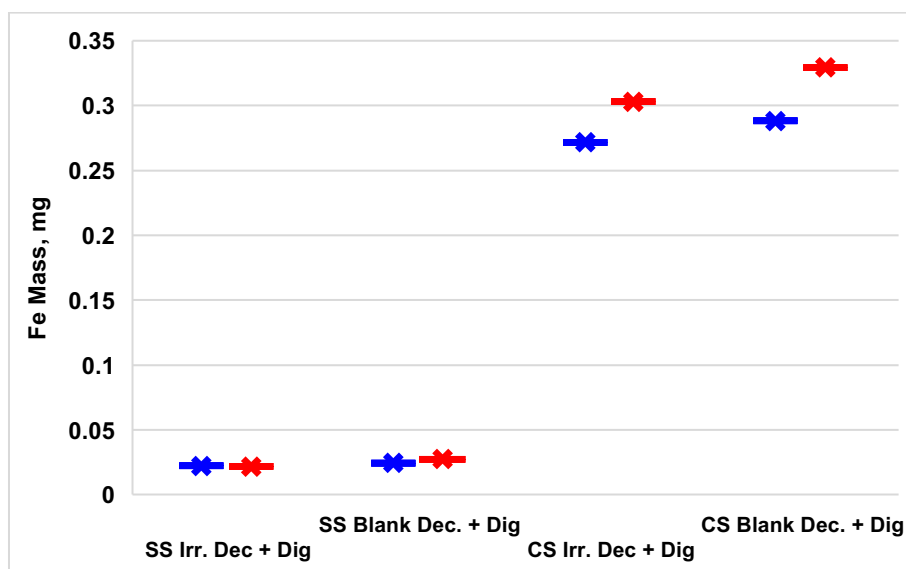


Figure 4. ICP OES results after GammaCell irradiation

These preliminary results cannot be interpreted as definitive evidence of radiation-induced corrosion at such a low dose rate. The differences between irradiated and blank samples are not statistically significant, and the experiment represents an early stage of method development. The results instead demonstrate that mass-loss measurements and ICP-OES can be incorporated into the experimental framework for future, longer-duration exposures within the cyclotron.

In conclusion, this project established a quantitative foundation for investigating the use of a medical cyclotron as a radiation source for nuclear materials research. Fricke dosimetry measured varying absorbed doses over 3-day intervals, with an accumulated absorbed dose of approximately 1855 Gy over one month. A relationship between target activity and absorbed dose was established for Target 1, suggesting that target activity may be useful for estimating future radiation exposures under comparable operating conditions. This work therefore establishes the dosimetry, sample preparation, irradiation, and analytical framework required to evaluate the medical cyclotron for nuclear materials research. Prolonged exposures, equivalent GammaCell comparisons, and characterization of the fast and thermal neutron components of the cyclotron radiation field will be necessary to determine how the mixed cyclotron radiation environment affects nuclear materials and whether existing medical cyclotron infrastructure can be effectively used for radiation-induced corrosion research. Future work will involve completing corrosion measurements after prolonged exposure in the cyclotron and comparing the results with equivalent GammaCell irradiations. Additional characterization of the cyclotron radiation field is also required, particularly for fast and thermal neutrons. Separating the contributions of gamma radiation and neutron irradiation will be important for understanding the mechanisms responsible for any observed changes in material behaviour.

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