CHARACTERIZATION AND UTILIZATION OF NEUTRON RADIATION FROM A PETRACE CYCLOTRON

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ABSTRACT

For the past 10 years, the University of Missouri Research Reactor Center has hosted a cyclotron facility which conducts daily irradiations of O-18 targets for the production of F-18 fludeoxyglucose. The General Electric PETtrace Cyclotron accelerates protons to 16 MeV with an available current of 100 μA. The cyclotron is situated in a vault which permits extracted proton and deuteron beams, as well as access to the neutrons resulting from the daily proton irradiation of O-18 targets. This fast neutron flux can be used for materials and electronics testing, as well as for the production of other isotopes. An MCNPX model of the cyclotron vault has been made and irradiation activation foils have been used to unfold the fast neutron flux spectrum. Calculated isotope production will be given for radioisotopes of interest in Nuclear Medicine which are difficult to produce by other means. We will present motivation for and production rates of isotopes including Ra-223, Ac-227, and Th-227.

KEYWORDS
Cyclotron, neutron flux, radioisotope production

1 INTRODUCTION

In this work we investigate an approach to produce radioisotopes for use in radiopharmaceuticals and various other applications. This technique would take advantage of the neutron flux generated as an inevitable byproduct of an accelerator that is itself used for the production of a different radioisotope.

A secondary target placed near the cyclotron undergoes transmutation or modification through the interaction of the neutron field. For example, a target can absorb a neutron and undergo subsequent decay, or interact with a high energy neutron and lose one or more neutrons, again with a possible subsequent decay.

The major benefits of this proposed technique are that isotopes can be passively irradiated inside of a target utilizing pre-existing accelerator systems commonly associated with medical facilities, taking advantage of the neutrons produced as a byproduct of conventional radiopharmaceutical production methods.

Though reactors can irradiate such targets, the use of the inevitable neutron field from an accelerator offers a number of advantages. If a target is itself radioactive, failure or leakage of the target deep within a reactor is a possible regulatory issue for the reactor facility. Such a target in an accelerator facility would have the benefit of immediate accelerator shut down and technical staff access. Accelerators are often sited in hospital settings, so the production of short lived isotopes, favored for medical procedures, can be conducted near patients, removing the need to transport isotopes from reactor sites to hospitals. Since the cost of accelerator operations are already being supported by the existing isotope production, use of this neutron field is “free”, reducing overall radiopharmaceutical cost. This technique could be extended to the production of any number of radio-isotopes from various target isotopes.

In this work we investigate a specific application involving the production of Radium-223. Short-lived radiopharmaceuticals that mimic calcium in the human body, like radium, are prime candidates for non-invasive palliative treatment of various types of bone cancer [1]. This proposed technique would take advantage of the neutron flux generated as an unintended byproduct of a cyclotron that is itself used for the

production of Flourine-18, a radioisotope used in various PET imaging agents, particularly in fludeoxyglucose (FDG).

The neutron flux would be allowed to interact with a target composed of a radium compound sealed inside of a container. Radium-226 can capture a neutron producing the unstable daughter product Radium-227. Radium-227 beta decays to Actinium-227 and Actinium-227 decays primarily by beta emission to Thorium-227. Thorium-227 alpha decays to the desired isotope Radium-223.

With an 11.4 day half-life, Radium-223 decays with three alpha-particles in rapid succession. Alpha-particles are desirable for internal treatment of cancers because though energetic and destructive of nearby cancer cells, they penetrate relatively short distances, which limits the potential for collateral damage to healthy cells in the vicinity. Short-lived isotopes are preferable because deleterious effects associated with radioactive compounds in the body are mitigated by their short half-lives. Thus, materials and methods for producing short-lived radioisotopes whose decay routes lead to a multiplicity of alpha particles are of considerable medical advantage.

2 METHOD

The University of Missouri Research Reactor Center hosts a cyclotron facility which conducts daily irradiations of Oxygen-18 targets for the production of Flourine-18 FDG. The General Electric PETtrace Cyclotron accelerates protons to over 16 MeV with an available current of up to 100 μA. The cyclotron is situated in a vault which permits extracted beams, as well as access to the neutrons resulting from the daily proton irradiation of Bruce Technologies O-18 targets.

An MCNPX Version 2.7 model has been made describing the neutron environment within the vault while the cyclotron is driving the Oxygen-18 targets, the code is described elsewhere [2]. Figure 1 shows the accelerator, the Bruce Technologies target, and a scaffold to hold targets to be irradiated from neutrons produced in the Bruce Technologies target [3].

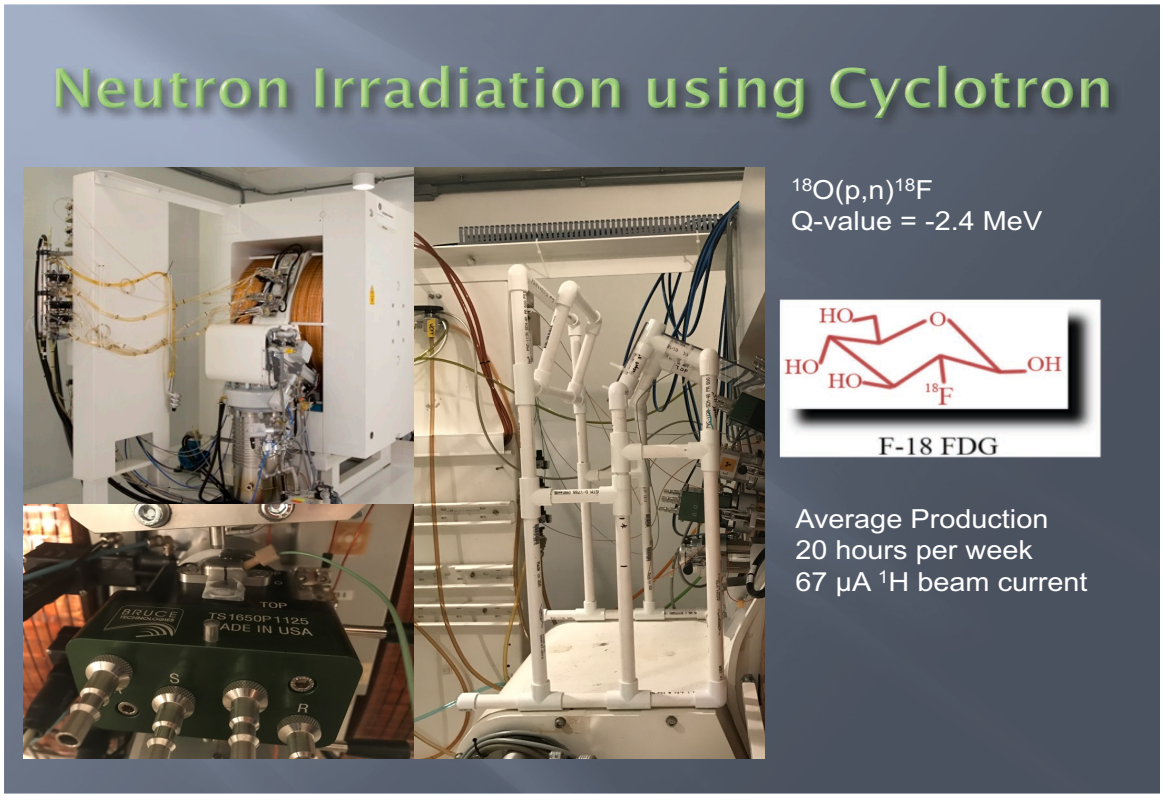

Figure 1. Cyclotron, Bruce Technologies target, neutron irradiation scaffold, F-18 FDG production.

The production pathway to Radium-223 from Radium-226 described in the introduction has a possible complication at the intermediate isotope Actinium-227. Actinium-227, which has approximately a 22-year half-life, has a substantial neutron capture cross section. Neutron capture would interrupt the production pathway to Radium-223 and should be avoided. Fig. 2 shows the neutron capture cross section for both Radium-226 and Actinium-227. Others have suggested that to maximize Radium-223 production, Radium-226 targets should be irradiated with an epithermal spectrum of neutrons and thermal neutrons should be shielded [1]. The production calculations assume that Actinium was chemically separated from Radium following neutron irradiation. The production calculations used ENDF VIII cross sections with the exception of TENDL 2015 cross sections for oxygen-18 and Actinium-227. The cross sections used in this work for Actinium-227 activation and Radium-226 activation are shown in Figure 2. MCNPX was used to calculate the cross section for Actinium-227 activation to Actinium-228. The production of Radium-227 was calculated using MCNPX. Subsequent decay and activation rates were calculated using the Bateman equations [4,5,6].

Figure 3 depicts the set up for the MCNPX simulation and the activity results of the calculation. The blue block in the simulation is the Bruce Technologies target with the proton beam entering the target from the right and striking the H_2O-18 reservoir as indicated in the figure. The red blocks are polyethylene moderators used to slow the fast neutron spectrum from the target. Two simulations were run, one with boron doped polyethylene to substantially reduce thermal neutrons, and one with undoped polyethylene.

The one gram mass $^{226}RaCO_3$ targets were exposed in the simulation to neutron flux originating from the interaction of the proton beam and the H_2O-18 reservoir. Target 1 was approximately 225mm from the reservoir, and target 2 was approximately 100mm from the reservoir. Target 2 was as close as practically possible to the reservoir, essentially on top of the Bruce Technologies target with a 25 mm layer of polyethylene moderator in between. The code ran simulating 1000 hours of proton beam irradiation at 67 μA, approximately a year's operation in normal production of clinical F-18 FDG.

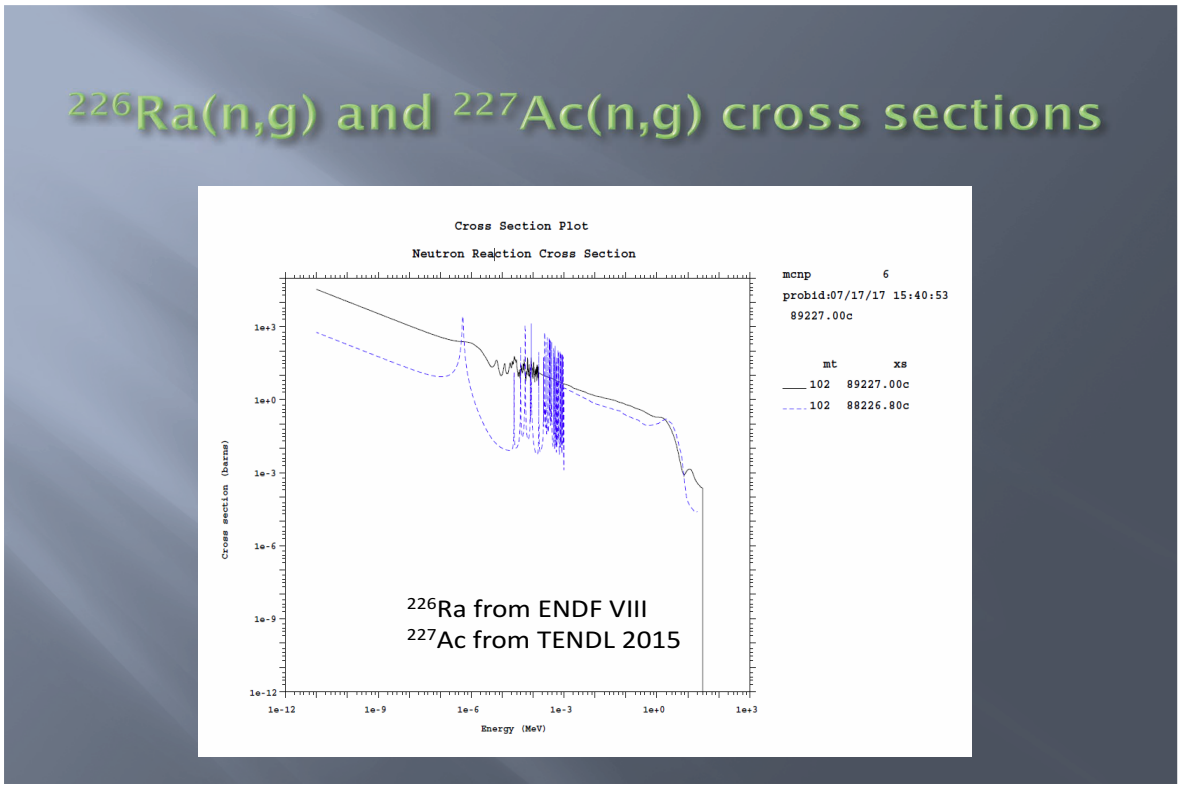

Figure 2. Radium-226 and Actinium-227 neutron capture cross sections.

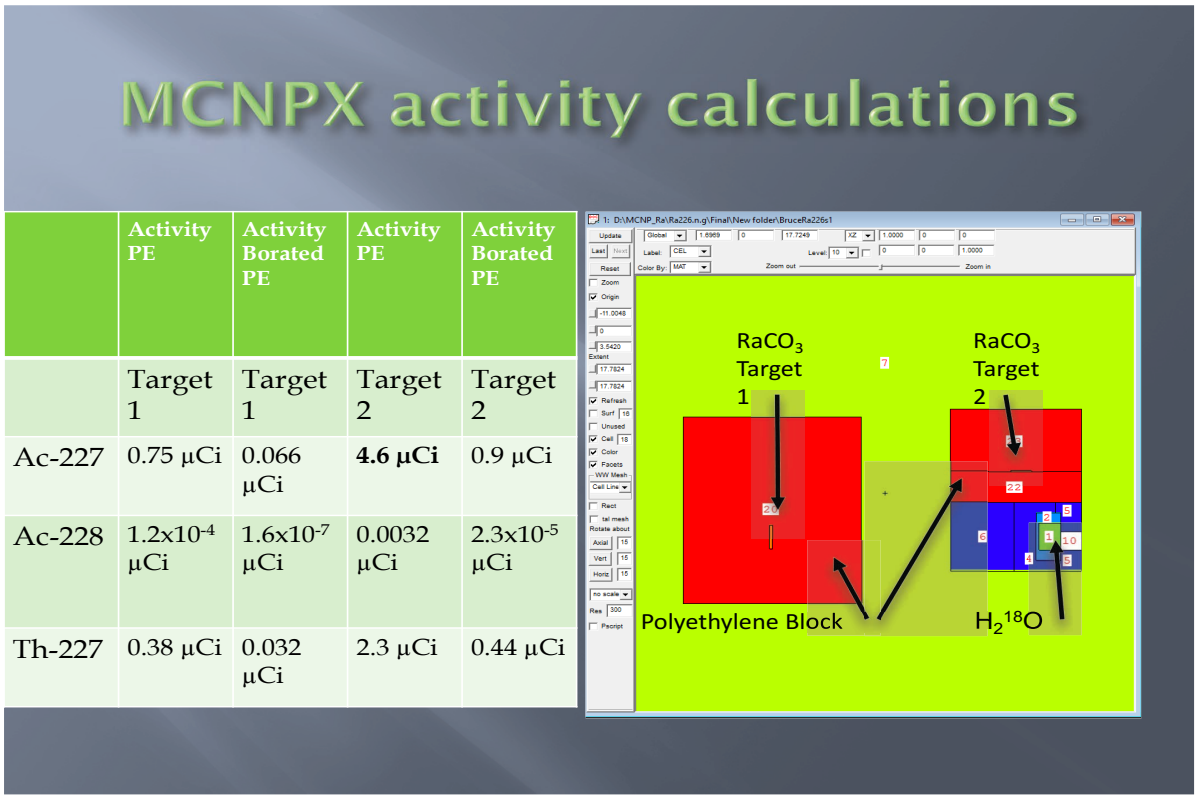

Figure 3. MCNPX simulation and the activity results calculated using the Bateman equations.

3 RESULTS

Unsurprisingly, Figure 3 indicates that Ac-227 production is greatest in the $^{226}RaCO_3$ target (target 2) nearest the neutron source. Boronated polyethylene reduces the production of unwanted ^{228}Ac but also reduces significantly the production of Ac-227. Production of Ac-228 in any case is extremely low, consequently there seems to be no need to shield thermal neutrons from the $^{226}RaCO_3$ target.

Figure 4 shows Ra-223 production from a $^{226}RaCO_3$ target irradiated for 1000 hours. Again, Ra-223 results from the decay of Ac-227 which has a half-life of approximately 22 years. The decay of the Ac-227 would result in the production of approximately 3 µCi of Ra-223 every 50 days.

4 CONCLUSIONS

Obviously, locating the $^{226}RaCO_3$ target closer to the source of neutrons could significantly increase the production of Ac-227 and subsequently the production of Ra-223. This would require a relatively modest modification of existing commercial FDG targets. Simply increasing the mass of the target, or enlisting the use of multiple cyclotrons would also increase Ra-223 production. This would raise the question of the availability of Ra-226

In any case, this simple simulation and calculation has shown that there is real potential to use the neutron field from a cyclotron, the inevitable byproduct of PET isotope production with a proton beam, to produce yet another isotope.

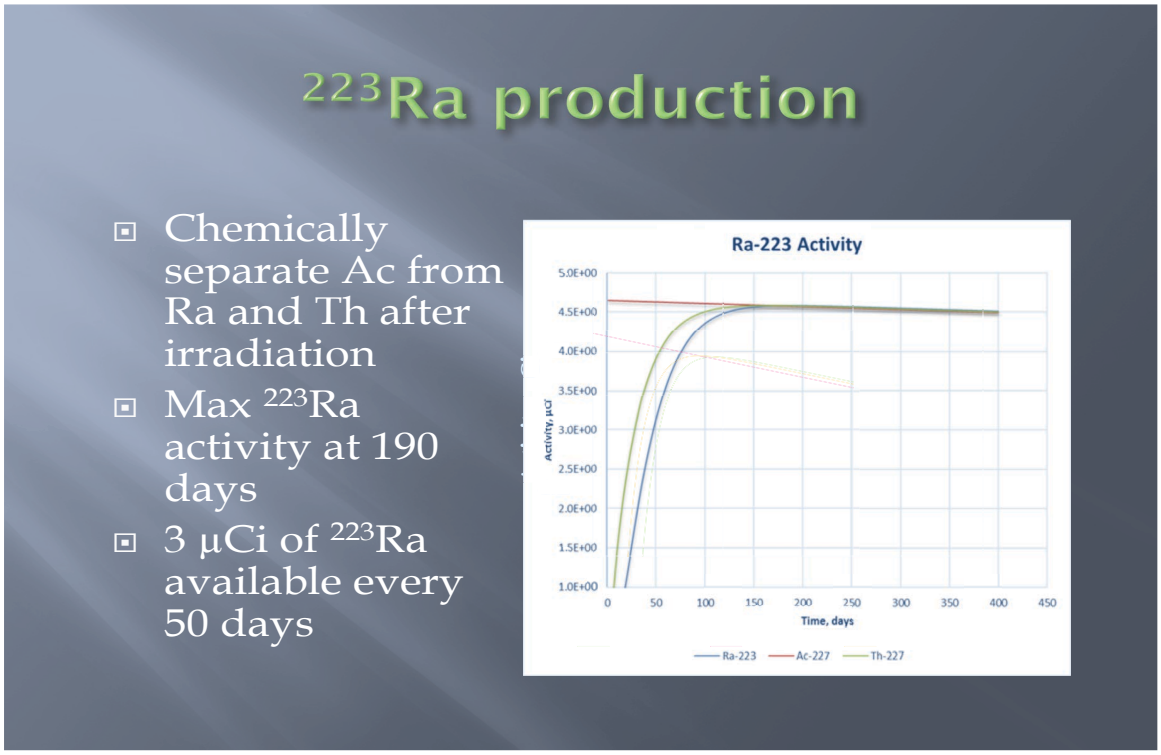

Figure 4. Ra-223 production

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