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ABSTRACT

This paper describes a proof of principle experiment to produce ${}^{13}N$ using an inertial electrostatic confinement (IEC) fusion device. This radioisotope is often used in positron emission tomography scans to image the heart. The 10-minute half-life of ${}^{13}N$ limits its use to those areas and clinics that possess an accelerator. A portable IEC device could be brought to remote locations, however, and produce short-lived PET isotopes on-site. Using the 14.7 MeV protons produced from the D-³He fuel cycle, the University of Wisconsin IEC device was used to produce approximately 4 - 8 Bq of ${}^{13}N$ during two separate experiments.

I. INTRODUCTION

Radioisotopes have been used in various medical imaging procedures for several decades with increasing regularity. In particular, positron emission tomography (PET) scans have recently experienced significant growth, and indications are that the number of these procedures administered each year will continue to grow¹. As this procedure expands, so to will the demand for the radioisotopes used. Short-lived isotopes (half-life < 60 minutes) are becoming popular to reduce the radiation dose to the patient. Moreover, many of the short-lived radioisotopes have attractive chemical properties that enable their incorporation into a variety of compounds. Because of the numerous commercial vendors of radioisotopes, many nuclear medicine clinics do not create their own radioisotopes. These commercial manufacturers use large, immobile accelerators and cyclotrons for isotope production². The time required to travel from the production site of the PET isotopes to the patient, however, limits their ability to deliver short-lived isotopes to less than about 100 miles. In contrast, an inertial electrostatic confinement (IEC) fusion device can be portably configured, and could be used to produce radioisotopes on-site at even the most remote locations.

The University of Wisconsin IEC device uses a second-generation fuel cycle to produce high-energy protons³⁻⁵:

$$D + {}^{3}He \rightarrow p(14.7 \text{ MeV}) + {}^{4}He(3.7 \text{ MeV}).$$

The 14.7 MeV protons can be used to produce a variety of radioisotopes^{1,6,7}. Specifically, ¹³N is produced via the following reaction by irradiating a water target:

$${}^{16}O(p,\alpha){}^{13}N$$

The objective of this paper is to discuss how a proof of principle experiment to produce ¹³N was done using fusion energy. The ¹³N isotope was selected for several reasons. First, there is currently no significant commercial production of the isotope due to its short half-life. Second, the demand for ¹³N PET scans should substantially increase with the anticipated Medicare/Medicaid insurance coverage of this procedure. Third, the ¹⁶O(p, α)¹³N reaction cross sections⁸ match the D-³He proton energies as shown in Figure 1.



Figure 1. Proton cross section for ${}^{16}O(8)$.

II. METHODS

The ¹³N production method focuses on the use of thin-walled containers through which the water is circulated. This container is placed inside the IEC chamber, a schematic diagram of which is shown in Figure 2. The container is then irradiated with 14.7 MeV protons from D-³He reactions. To date, three different water containment apparatus have been developed. All three are based on the same design and use numerous 0.64 cm diameter thin-walled tubes, connected to manifolds at either end, as the



Figure 2. University of Wisconsin IEC Chamber.

containment method (see Figure 3). Two versions, models Al-M1 and Al-M2, use aluminum tubing with a wall thickness of 380 microns; the third model, SS-M1, uses 304 stainless steel tubing with a wall thickness of 125 microns. All three models are approximately 60 cm by 60 cm.

The radioisotopes are produced in a batch process, i.e. the same volume of water is circulated through the containment system rather than using a continuous flow of fresh water. Additionally, the water contains 10 milli-molar of ethyl alcohol in order to promote the formation of ¹³NH₃⁺ ammonia ions once the ¹³N has been created. This is necessary for two reasons. First, it facilitates the extraction of the ¹³N from the water; second, ¹³N is administered as an ammonia solution during PET scans.

Figure 4 is a schematic diagram of the containment system. Water is pumped through the system at approximately 1 liter per minute. The water enters one manifold, is distributed among the numerous tubes, and passes through to the next manifold. From there, the water exits the manifold, flows out the chamber and passes through a heat exchanger.

The heat exchanger is required because the operating conditions necessary for radioisotope production introduce four kilowatts or more of power into the IEC chamber. Some of this heat is absorbed by the water and must be removed to prevent steam production. This is accomplished by passing the water through a length of copper tubing wound into coils and submerged in a chilled water bath. The water inside the containment system exits the heat exchanger, passes through a pressure relief system that also accommodates the volumetric expansion of the water, and flows back into the pump. The water then reenters the apparatus in the chamber to again be irradiated by the high-energy protons. Figure 5 shows the model SS-M1 apparatus mounted in the University of Wisconsin's IEC chamber.

After the water mixture has been irradiated for the necessary length of time, the containment system is pressurized with helium. A series of valves is opened or closed as required to direct the water containing the NH_3^+ ions into a column of ion exchange resin. Ion exchangers are polymer compounds possessing numerous ionic sites that can reversibly exchange either cations or anions with similarly charged ions in the surrounding solution without altering the ion's chemical properties⁹.



Figure 3. Model SS-M1 water containment apparatus.

For this experiment, approximately 25 grams of DOWEX 50WX8 (100-200) cation exchange resin is The DOWEX resin employs SO₃⁻ ions used. (covalently bound to the resin polymer) as ionic sites, and H^+ ions (ionically bound to the SO₃⁻) as the vehicles for ion exchange. Before use, this resin is rinsed with approximately 100 mL of 0.1 M NaOH in order to replace the H⁺ ions with Na⁺ ions at each SO₃⁻ site. This is done to facilitate the capture of NH_3^+ ammonia ions since the NH_3^+ ions have a higher bonding affinity with the SO_3^- than the Na^+ ions. When the water solution is passed through the column of ion exchange resin, the SO_3^- sites will give up their Na^+ ions and bond with NH_3^+ ions, removing all ¹³N-labeled ammonia from the water.

Because the resin will collect a variety of cations and because the number of ions the resin will capture is finite, the water solution should be as free of contaminant ions as possible. This experiment uses deionized 18 M Ω -cm water, although the water is certainly not this pure once inside the apparatus as a



Figure 4. Schematic drawing of isotope production system.



Figure 5. Model SS-M1 apparatus mounted in the University of Wisconsin IEC chamber.

result of exposure to carbon dioxide in the atmosphere during transfer. Moreover, metal ions from the system itself undoubtedly enter the water. Circulating several batches of 18 M Ω -cm water through the system prior to the experiment minimizes the metal ions' influence. Once all of the water solution is passed through the ion exchange column, the column activity is measured.

A 3 x 3 NaI detector and multichannel analyzer are used to count the 511 keV gamma rays created by the annihilation of the ¹³N positrons. Each positron annihilation results in the emission of two 511 keV gamma rays in opposite directions. During counting, the resin column is placed on the face of the 3 x 3 NaI detector, which is located within a lead vault to shield background radiation. Given the modest fusion reaction rates achieved in the proof of principle experiment, the activity of ¹³N produced was expected to be less than 20 Bq. To achieve reasonable certainty in the count data from this low activity, the resin column is counted in 180-second intervals. Using a calibrated ²²Na source, a region of interest (ROI) is established with the MCA counting software from 470 keV to 570 keV. The 511 keV photopeak falls within these energies, and the counting software sums the total counts within this ROI to determine the gross activity. The measured background activity is subtracted from this to determine the net activity of the resin column. Finally, the net activity is divided by the detector efficiency, measured to be 17%, to yield the true activity of ¹³N. These results are used to create a decay curve. This curve is used to calculate the decay constant of the radionuclide and to determine the quantity produced at the end of irradiation¹⁰.

III. RESULTS

Two separate experiments were conducted to produce ¹³N using a proton flux created from D-³He fusion. During both experiments, the D-³He fusion reaction rate varied considerably over the experiment operation time. This was due to a number of factors, including short operational shutdowns and a significant increase in the outgassing of chamber components as they were heated. The water target was irradiated for approximately 22 minutes in the first experiment, and for approximately 10 minutes in the second experiment. When the proton irradiation was terminated, the water target was passed through the ion exchange resin column and then transferred to the 3 x 3 NaI detector. The resin column was counted for five 180second intervals following each experiment. These data points are show below in Figures 6 and 7, respectively. Each figure also includes an exponential fit of the activity data (heavy line), as well as a dashed line representing the true decay path of ^{13}N .

The fit of the first experiment's data yields a decay constant λ of 9.141 x 10⁻⁴ s⁻¹, which differs from the true λ for ¹³N by 21%. The activity of ¹³N created at the end of irradiation is calculated to be

approximately 7.5 Bq. The dashed curve passes through all the error bars of the count data in Figure 6, lending confidence to the belief that the measured activity in the resin column is ¹³N. An exponential fit of data from the second experiment yields a decay constant of 1.144 x 10⁻³ s⁻¹, which differs from the true λ by only 1.3%. The activity of ¹³N at the end of irradiation was calculated to be approximately 4.4 Bq. Although small, these quantities satisfy the goals of this proof of principle experiment.



Figure 6. Measured ¹³N activity in the ion exchange resin from the first isotope production experiment.



Figure 7. Measured ¹³N activity from the second isotope production experiment.

IV. SUMMARY

This proof of principle experiment demonstrates that fusion power has the potential to create ¹³N for PET scans. Moreover, the portability of IEC devices makes them uniquely suited to the production of short-lived radioisotopes for medical use at remote clinical locations. The reaction rate in current IEC chambers must increase by many orders of magnitude, though, to produce radioisotopes in clinical quantities. The anticipated increase in demand for ¹³N PET scans may help stimulate the improvements needed for IEC devices to become commercially viable isotope production machines.

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