Nuclear Instruments and Methods in Physics Research B 410 (2017) 230–239

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Measurement of the ^{64}Zn,^{47}Ti(n,p) cross sections using a DD neutron generator for medical isotope studies

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ARTICLE INFO

Article history:
Received 26 April 2017
Received in revised form 7 August 2017
Accepted 17 August 2017

Keywords:
DD neutron generator
Medical isotope production
Scandium (Sc) and copper (Cu) radioisotopes
Indium
Ratio activation
Theranostics

ABSTRACT

Cross sections for the ^{47}Ti(n,p)^{47}Sc and ^{64}Zn(n,p)^{64}Cu reactions have been measured for quasi-monoenergetic DD neutrons produced by the UC Berkeley High Flux Neutron Generator (HFNG). The HFNG is a compact neutron generator designed as a "flux-trap" that maximizes the probability that a neutron will interact with a sample loaded into a specific, central location. The study was motivated by interest in the production of ^{47}Sc and ^{64}Cu as emerging medical isotopes. The cross sections were measured in ratio to the ^{113}In(n,n′)^{113m}In and ^{115}In(n,n′)^{115m}In inelastic scattering reactions on co-irradiated indium samples. Post-irradiation counting using an HPGe and LEPS detectors allowed for cross section determination to within 5% uncertainty. The ^{64}Zn(n,p)^{64}Cu cross section for $2.76^{+0.01}_{-0.02}$ MeV neutrons is reported as 49.3 ± 2.6 mb (relative to ^{113}In) or 46.4 ± 1.7 mb (relative to ^{115}In), and the ^{47}Ti(n,p)^{47}Sc cross section is reported as 26.26 ± 0.82 mb. The measured cross sections are found to be in good agreement with existing measured values but with lower uncertainty (<5%), and also in agreement with theoretical values. This work highlights the utility of compact, flux-trap DD-based neutron sources for nuclear data measurements and potentially the production of radionuclides for medical applications.

1. Introduction

There has been significant interest in the past several years in exploring the use of neutron-induced reactions to create radionuclides for a wide range of applications. This interest is due to the volumetric absorption of neutrons as compared to charged particle beams (ranges of g/cm^2 as compared to 10's of mg/cm^2), together with the fact that isotope production facilities often produce large secondary neutron fields. Particular interest has been paid to (n,p) and (n,α) charge-exchange reactions since these reactions produce high-specific activity radionuclide samples without the use of chemical carriers in the separation process.

Two other potential neutron sources for (n,x) reactions exist in addition to the secondary neutron fields generated at existing isotope production facilities: reactors and neutron generators that utilize the D(T,n)α ("DT") and D(D,n)^{3}He ("DD") reactions. While reactors produce copious quantities of neutrons, their energy spectra are often not well-suited to the preparation of high-purity samples due to the co-production of unwanted activities via neutron capture, in addition to the significant start-up costs and proliferation concerns involved in their commissioning [1]. Similarly, while the higher energy 14–15 MeV neutrons produced at DT generators are capable of initiating (n,p) and (n,α) reactions, their higher energy opens the possibility of creating unwanted activities via (n,pxn) and (n,αxn) reactions that cannot easily be separated from the desired radionuclides. DT generators may also often be limited by the restricted use of tritium at many institutions.

In contrast, the neutron spectrum from a DD reaction, which ranges from approximately 2–3 MeV, is ideally suited to (n,p) radionuclide production. However, the lower achievable flux from these generators limits their production capabilities. An additional complication is the relative paucity of high-quality, consistent cross section data for neutrons in the 2–3 MeV DD energy range.

The purpose of the present work is to explore the potential to use high-flux neutron generators to produce high-specific activity samples of radionuclides at the mCi level for local use in the application community. The research group at UC Berkeley has

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http://dx.doi.org/10.1016/j.nimb.2017.08.021
0168-583X/

developed a High Flux Neutron Generator (HFNG) that features an internal target where samples can be placed just several millimeters from the neutron producing surface in order to maximize the utilization of the neutron yield for the production of a desired radionuclide [2–4]. The HFNG uses the D(D,n)^{3}He reaction to produce neutrons with energies near 2.45 MeV together with a self-loading target design to maintain continuous operation without target replacement. In addition to the generator itself, efforts are underway to design neutron reflection capabilities to allow scattered neutrons multiple opportunities to interact with an internally mounted target. While these design efforts are underway, the HFNG can be used to better characterize production cross sections at the appropriate neutron energy.

The present work features a pair of cross section measurements for the production of two emerging non-standard medical radionuclides: the positron emitter ^{64}Zn(n,p)^{64}Cu and the single-photon emission computed tomography (SPECT) tracer ^{47}Ti(n,p)^{47}Sc. ^{64}Cu ($t_{1/2}$ = 12.7 h) undergoes β^+ decay (61.5% branching ratio) to ^{64}Ni or β^- decay (38.5% branching ratio) to ^{64}Zn [5]. The emitted short-range 190-keV β^- particle makes this an attractive therapeutic radionuclide, which also has the possibility for simultaneous positron emission tomography (PET) imaging for real-time dose monitoring and verification. This makes ^{64}Cu particularly desirable for emerging radiation therapy protocols [6–9]. In addition, copper radiochemistry is well developed, and many existing ligands and carriers may be used for selective delivery of the radionuclide to different sites in patients. The second radionuclide studied, ^{47}Sc ($t_{1/2}$ = 3.35 d), undergoes β^- decay to ^{47}Ti, emitting a high-intensity (63.8%) 159-keV gamma ray in the process [10]. This radionuclide is attractive as an emerging diagnostic isotope, due to the similarity of the emitted gamma ray to that of the well-established ^{99m}Tc [11–14]. Due to the short half-life ($t_{1/2}$ = 6.0 h) of and dwindling supplies of ^{99m}Tc, ^{47}Sc stands poised as a potential solution to this shortage, due to its longer half-life and multiple production pathways without the need for highly enriched uranium [15]. In addition, when paired with ^{44}Sc, ^{47}Sc forms a promising "theranostic" pair for use in simultaneous therapeutic and diagnostic applications [16,17].

Current methodology in radiochemistry has shown recovery of upwards of 95% of produced ^{64}Cu [18,19] and ^{47}Sc [20–22] from solid target designs, without the need for additional carrier. By expanding the base of efficient reaction pathways, great advances are possible in making production of medical radionuclides more efficient and affordable for those in need. It is this desire to improve the options available for modern medical imaging and cancer therapy which has motivated the campaign of nuclear data measurements for isotope production at the UC Berkeley HFNG.

2. Experiment

2.1. Neutron source

Neutron activation was carried out via irradiation in the High-Flux Neutron Generator (HFNG), a DD neutron generator at the University of California, Berkeley. This generator extracts deuterium ions from an RF-heated deuterium plasma (using ion sources similar to designs from the Lawrence Berkeley National Laboratory [4]) through a nozzle, whose shape was designed to form a flat-profile beam, 5 mm in diameter. This deuterium beam is incident upon a water-cooled, self-loading titanium-coated copper target [2,3], where the titanium layer acts as a reaction surface for DD fusion, producing neutrons with a well-known energy distribution as a function of emission angle [23]. While the machine's design features two deuterium ion sources impinging from both sides of the target, only a single source was used in the present work. Irradiation targets are inserted in the center of the titanium layer deuteron target, approximately 8 mm from the DD reaction surface, prior to startup. Fig. 1 displays a cut-away schematic of the HFNG. A 100 keV deuterium beam was extracted at 1.3 mA, creating a flux of approximately $1.3 \cdot 10^7$ neutrons/cm^{2}s on the target.

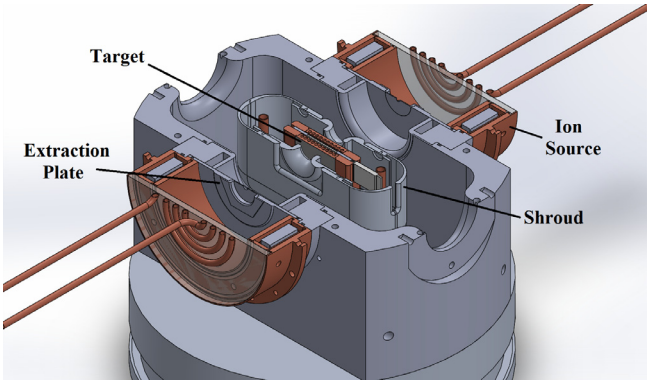

Fig. 1. Cut-away schematic of the HFNG. The ion source is approximately 20 cm in diameter.

2.2. Cross section determination by relative activation

The approach used in both measurements was to irradiate foils of zinc or titanium, which were co-loaded with indium foils in order to determine their (n,p) cross sections relative to the well-established ^{113}In(n,n′)^{113m}In and ^{115}In(n,n′)^{115m}In neutron dosimetry standards [24,25]. Table 1 lists physical characteristics of each foil for the various irradiations. In each experiment, the co-loaded foils were irradiated for 3 h at nominal operating conditions of 1.3 mA and 100 kV. After irradiation, the foils were removed and placed in front of an appropriate High-Purity Germanium (HPGe) gamma-ray detector and time-dependent decay gamma-ray spectra were collected.

One cm diameter, 1-mm thick natural abundance zinc and titanium targets were employed for the measurement. Each of these was co-loaded with a natural abundance Indium foil of 1 cm diameter and 0.5 mm thickness in a recess cut into a 2-mm thick polyethylene holder, as seen in Fig. 2, which was mounted in the HFNG target center. Prior to loading, each foil was washed with isopropanol and dried, to remove any trace oils or residue that could become activated during irradiation.

2.3. Determination of effective neutron energy

The D(D,n)^{3}He reaction at 100 keV lab energy produces neutrons with energies ranging from 2.18 to 2.78 MeV, over an angular range of 0–180° in the lab frame-of-reference with respect to the incident deuteron beam. This distribution has been well documented [23] and is shown in Fig. 3 for 100 keV incident deuteron energy.

Since the samples are separated by only 8 mm from the DD reaction surface they subtend a fairly significant (~17°) angular range in a region of high (approximately $1.3 \cdot 10^7$ neutrons/cm^{2}s) neutron flux. This stands in contrast to other measurements which feature collimated beams and significantly lower total neutron flux.

The Monte Carlo N-Particle transport code MCNP6 [26] was used to model the neutron energy spectrum incident upon target foils co-loaded into the HFNG (see Fig. 4). The neutron spectral distribution is also broadened by the temperature of the target. This