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Excitation functions of ^{nat}Zn(p,x) nuclear reactions with proton beam energy below 18 MeV

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HIGHLIGHTS

- Measurement of excitation functions of ^{61}Cu, ^{66}Ga, ^{67}Ga and ^{65}Zn of ^{nat}Zn(p,x) reactions.
- Comparison of these measurements with the published literature, where possible.
- Thick target yield of each radionuclide was calculated from measured data.

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ABSTRACT

We measured the excitation functions of ^{nat}Zn (p,x) reactions up to 17.6 MeV, using the stacked-foils activation technique. High-purity natural zinc (and copper) foils were irradiated with proton beams generated by an 18 MeV isochronous cyclotron. Activated foils were measured using high-purity Ge gamma spectroscopy to quantify the radionuclides ^{61}Cu, ^{66}Ga, ^{67}Ga, and ^{65}Zn produced from the reactions. Thick-target integral yields were also deduced from the measured excitation functions of the produced radioisotopes. These results were compared with the published literature and were found to be in good agreement with most reports, particularly those most recently compiled.

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1. Introduction

The majority of medium-energy (11–18 MeV) proton cyclotrons operating in a biomedical setting are used for producing short-lived radioisotopes (i.e., ^{18}F, ^{11}C, ^{13}N and ^{15}O) for routine diagnostic applications of positron emission tomography (PET). They might also be used to produce less conventional radioisotopes for clinical and preclinical research; for example radiometal and radiohalide PET isotopes. One class of interest represents those radioisotopes produced by proton-induced reactions in natural zinc (^{nat}Zn), principally ^{61}Cu, ^{66}Ga, ^{67}Ga, ^{68}Ga and ^{65}Zn. These radioisotopes are used in PET or single-photon emission computed tomography (SPECT) diagnostic or preclinical studies, depending on their radiation characteristics.

For example, it is useful to investigate a reliable and cost effective method for the production of ^{61}Cu via the reaction ^{nat}Zn(p,α)^{61}Cu below 18 MeV, even though the radioactive by-products co-produced in the same target must be separated out in the purification process. The relatively short half-life of ^{61}Cu (3.33 h) makes it suitable for the radiolabelling of small molecules such as ATSM and PTSM for hypoxia and blood flow measurements, respectively (Jalilian et al., 2008, 2009). Gallium-67 ($t_{1/2}$=3.26 d) is the longest half-life radionuclide produced by proton irradiation of ^{nat}Zn, arising from ^{67}Zn (natural occurrence

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4.1%) with a relatively poor yield plus other radioisotopic products. It can also be produced from ^{68}Zn (18.8%; [p,2n] reaction) above 10.3 MeV (Szelecsényi et al., 2012) (Table 1), and is widely used in SPECT imaging (Ćwikła et al., 1999). Although it is not widely employed, the isotope ^{66}Ga ($t_{1/2}$=9.49 h) is suitable for labelling of monoclonal antibodies (Goethals et al., 1990) or blood cells (Jalilian et al., 2006). Gallium-68 ($t_{1/2}$=67.6 min) can be produced directly by a cyclotron (e.g.; Szelecsényi et al., 1998) but is usually sourced for practical purposes from a ^{68}Ge/^{68}Ga generator (lifetime of several months), so that PET imaging studies can be performed without the need for a co-located cyclotron (Shetty et al., 2010).

Of relevance to this study there have been several reports of proton-induced excitation functions for reactions in a ^{nat}Zn target over energies that include the range 0–18 MeV (Barrandon et al., 1975; Little and Lagunas-Solar, 1983; Nortier and Mills, 1991; Kopecky, 1990; Hermanne, 1997; Szelecsényi et al., 2005; Al-Saleh et al., 2007; Uddin et al., 2007). For example, Al-Saleh et al. (2007) reported cross section data for ^{66}Ga, ^{67}Ga, ^{68}Ga, ^{62}Zn and ^{65}Zn by proton-induced reactions in ^{nat}Zn up to 27.5 MeV. Uddin et al. (2007) reported the ^{nat}Zn(p,x) reactions for protons up to 40 MeV for the formation of ^{61}Cu, ^{66}Ga, ^{67}Ga, ^{68}Ga, ^{62}Zn and ^{65}Zn. Szelecsényi et al. (2005) published the excitation functions of proton-induced reactions in ^{nat}Zn up to 100 MeV. The nuclear and decay properties of isotopes produced from ^{nat}Zn(p,x) reactions below 18 MeV proton energy are shown in Table 1.

In this study we measured ^{nat}Zn(p,x) excitation functions of ^{61}Cu, ^{66}Ga, ^{67}Ga and ^{65}Zn for energies ranging from the effective threshold of the particular reaction up to 17.6 MeV, using the stacked-foils activation technique. Reliable cross section measurements were acquired from combining independent experiments in triplicate.

2. Experimental methods

Excitation functions for the ^{nat}Zn(p,x) reactions were measured by the stacked-foils activation technique (Qaim et al., 1977). High-purity natural copper monitor foils of 25 mm diameter were employed to measure beam flux and energy in the stack using the characteristics of the well documented ^{63}Cu(p,n)^{63}Zn and ^{65}Cu(p,n)^{65}Zn reactions (IAEA, 2001). These foils had a thickness of 0.025 ± 0.004 (SD) mm (99.99+% elemental purity; Goodfellow Metals Ltd, UK). The ^{nat}Zn target foils were of 25 mm diameter (99.99% elemental purity; Goodfellow) and a thickness of 0.025+0.003 mm, with an isotopic composition of ^{64}Zn (48.6%), ^{66}Zn (27.9%), ^{67}Zn (4.1%), ^{68}Zn (18.8%) and ^{70}Zn (0.6%). The foil stacks consisted of combinations of the Cu and Zn foils, with one monitor Cu foil at the target front face. A typical stack of consisted of four units of four Zn foils sandwiched between single Cu monitor foils (Cu–4*x*Zn–Cu), with the last (fifth) stack consisting of (Cu–8*x*Zn–Cu) so that the exit energy of the beam from the final stack was well below the threshold energy of the studied reaction. For each experiment, a stack was inserted into an external solid targetry beam line, where the target was cooled in front by helium gas flow and from behind by a cooled water jet (Scharli et al., 2012). The proton source employed was an IBA 18/18 MeV isochronous cyclotron (IBA, Louvain le Neuve, Belgium).

Foil stacks were irradiated with a primary beam of energy 17.6 MeV, accounting for beam degradation by an obligatory 0.025 ± 0.0005 (SD) mm-thick Havar® foil vacuum window. Irradiation was for 3 min using a target beam current of 5 μA. Activities of the irradiated foils were measured at 3 h following end of bombardment (EOB) by cryo HPGe gamma spectroscopy (Canberra, Meriden, USA; S5000 MCA & GC1018 Ge detector), using an instrument calibrated with standard sources of ^{60}Co, ^{57}Co, ^{137}Cs, ^{65}Zn, ^{109}Cd and ^{133}Ba, with an uncertainty in the intrinsic efficiency curve of 5%. The energy degradation within each foil including the Havar® foil was calculated by SRIM2008 software (Ziegler et al., 2011). The distance between the specimen and the end cup of the HPGe detector was set at 50 cm, in order to reduce the dead time of each measurement to less than 5%.

The uncertainty in the derived reaction cross section data is a function of the independent uncertainties encountered in the act of measurement; beam current (10–12%), detector efficiency (5%), counting statistics (4–8%), foil thickness (4–7%) and time of irradiation (1–2%). Thus, the total uncertainty in these data ranged from 13 to 16%. The uncertainty in the proton energy at a point along the beam path was derived from analysis of ^{63}Zn and ^{65}Zn product reactions in the monitor Cu foils. Since three replicate independent experiments were performed for each cross section data point the uncertainty in the energy can be estimated from the spread of these calculated energy values. This uncertainty ranged from ± 0.25 MeV (SD) at 17.6 MeV to ± 0.85 MeV at around 3.2 MeV.

3. Results and discussion

The cross sections for those product isotopes accessible to this study from ^{nat}Zn(p,x) reactions are shown in Table 2. Comparisons

Table 1
Nuclear and decay properties of isotopes produced from ^{nat}Zn (p,x) reactions below 17.6 MeV.

Radionuclide	Half-life[a]	Decay mode[a]	$E\gamma$ (keV)[a]	$I\gamma$ (%)[a]	Reactions[b]	E_{th} (MeV)[b]
^{61}Cu	3.33 h	EC (38.5%); β^+ (61.4%)	282.956	12.20	^{64}Zn(p,α)	6
			656.008	10.77		
^{64}Cu[c]	12.70 h	EC (43.1%); β^+ (17.4%)	1345.77	0.776	^{68}Zn(p,αn)	8.5
					^{67}Zn(p,α)	9.5
^{67}Cu[c]	61.83 h	β^- (99.0%)	91.26	7.0	^{70}Zn(p,α)	7.7
			93.31	16.1		
			184.57	48.7		
^{65}Zn	244.26 d	EC (98.6%); β^+ (1.4%)	1115.5	50.60	^{66}Zn(p,d)	8.9
						11.2
					^{66}Zn(p,n+p)	9.7
					^{67}Zn(p,t)	
^{66}Ga	9.49 h	EC (45.3%); β^+ (56%)	833.537	5.88	^{66}Zn(p,n)	6
			1039.231	36.90	^{67}Zn(p,2n)	13.6
^{67}Ga	3.26 d	EC (99.8%)	184.6	21.2	^{67}Zn(p,n)	1.8
			300.2	16.8	^{68}Zn(p,2n)	10.3
^{68}Ga[c]	67.63 min	EC (38%); β^+ (62%)	1077	3	^{68}Zn(p,n)	3.75

$E\gamma$ energies of γ-ray emissions. $I\gamma$, probability of γ-ray emission per nuclear disintegration. E_{th}, threshold energy of reaction.

[a] Data from table of nuclides; http://atom.kaeri.re.kr/.

[b] Data from Experimental Nuclear Reaction Data (EXFOR).

[c] Not reported in this study (see text).

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