Accelerator production of ^{71}As from metal germanium targets and cross section measurement of ^{70}Ge(d,n)^{71}As reaction

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Introduction

The radioisotopes ^{72}As ($t_{1/2}$ = 26.0 h, 88% β+) and ^{71}As ($t_{1/2}$ = 65.3 h, 28% β+), shown in Figure 1, have applications in positron emission tomography (PET) imaging when bound to nanoparticle- [1–3], peptide- [4], and antibody-based [3,5] radiopharmaceuticals. These diagnostic radionuclides have theranostic potential when paired with β⁻-emitting therapeutic ^{77}As or, due to the homologous relationship between arsenic and antimony, the Meitner-Auger-electron- (MAe–) emitter ^{119}Sb. Our previous work reported new production and isolation techniques for the positron-emitter ^{71}As. However, our measured yield of ^{71}As does not agree well with the theoretical physical yield computed from ^{70}Ge(*d,n*)^{71}As cross section measurement performed by K.Otozai et al. [6]. Therefore, we propose a new measurement of the ^{70}Ge(d,n)^{71}As cross section at the Notre Dame Nuclear Science Laboratory.

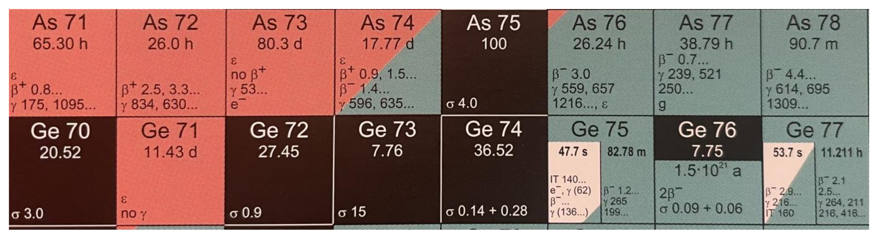

Figure 1: Section of the chart of the nuclides showing the isotopes of germanium and arsenic. Nuclides in black are stable, red decay by β+/ec, and blue decay by β–.

Methods

- Thick ^{70}Ge coin targets (10 mm diameter, 150-300 μm thick and ~70 mg) were **irradiated** with 35 μA of 8 MeV deuterons (GE PETtrace cyclotron) and ^{71}As **quantified** by high purity germanium (HPGe) gamma spectroscopy.
  - Use IAEA - Nuclear Data Sheet to monitor the ^{70}Ge(*d,n*)^{71}As cross section for **theoretical physical yield calculation**.
- ^{70}Ge(*d,n*)^{71}As excitation function will be measured via stacked-foil activation method
  - Stacks containing two target foils (0.41±0.02 μm ^{nat}Ge on 25.4 μm Kapton, Astral Technology Unlimited, Inc), a beam monitor foil (15 μm ^{nat}Ni, Goodfellow Cambridge Ltd), and an energy degrader foil (50 μm ^{nat}Al, Goodfellow Cambridge Ltd) will be assembled in custom aluminum frames (Figure 2).
  - Use ^{nat}Ni (d,x) ^{61}Cu or ^{56}Co to **monitor** beam intensity [7] (Figure 3.).
  - Identical foil stacks, shown in Figure 2c, are planned for irradiation at incident deuteron energies of 4, 7, 8 and 10 MeV with a HV FN type Pelletron tandem accelerator in August 2023.
  - **The energy of deuterons** in each foil were estimated based on SRIM ion stopping range calculations [8].
  - ^{71}As, ^{61}Cu, and ^{56}Co will be **characterized** by HPGe gamma spectroscopy measurements.
  - The cross-section will be calculated by **the activation equation** (**eq 1,** where I is the number of incident particles per unit time, n is the target nuclei per unit volume, x is the target thickness and σ is the cross section).

$$A(t) = R\left(1 - e^{-\lambda t}\right) \quad and\ R = Inx\sigma$$

Eq 1: The activation equation

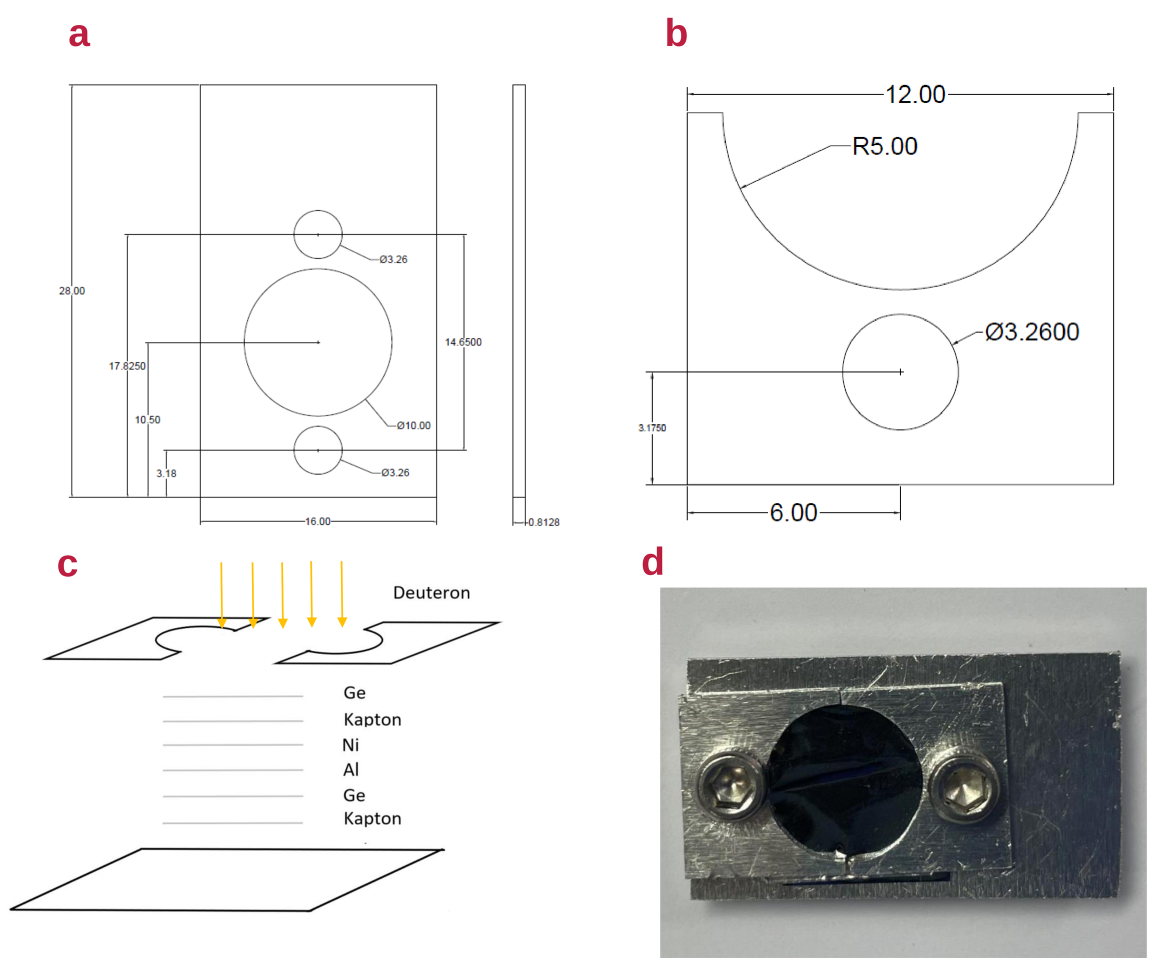

Figure 2: (a) The dimension of custom foil stack holding frame (b) The vertical view of the foil stack show the stacking order of each foil and the arrows indicate incident direction of deuteron (c) An assembled foil stack frame.

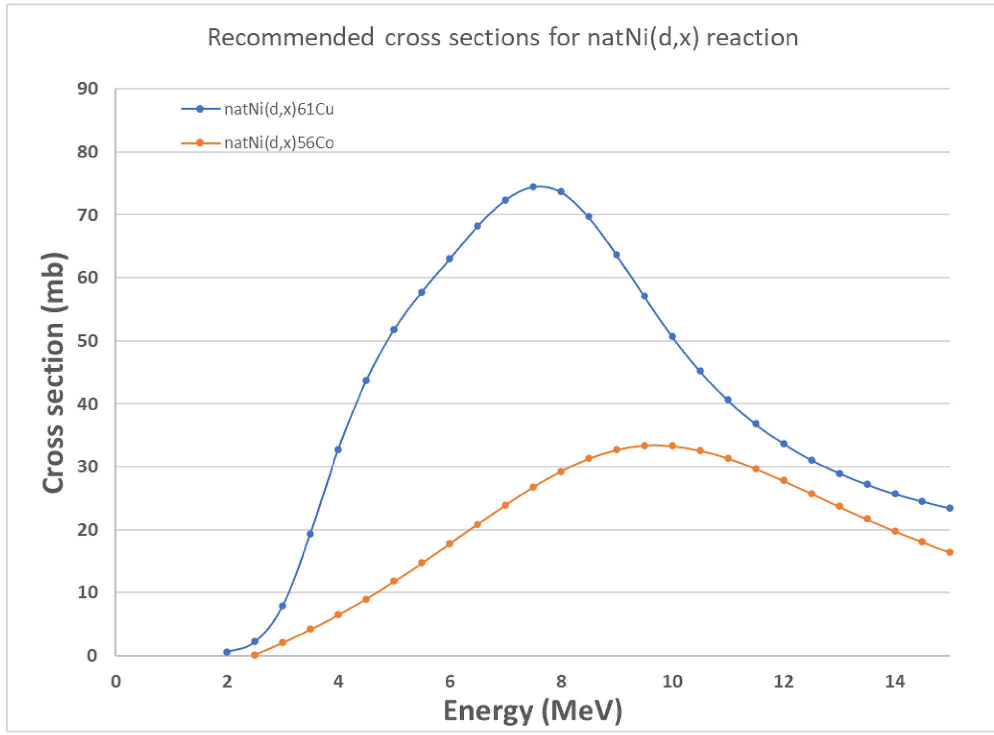

Figure 3: The ^{nat}Ni (d,x)^{61}Cu cross section as function of deuteron energy (Provided by IAEA - Nuclear Data Sheet)

Conclusions

The ^{70}Ge(d,n)^{71}As physical production yield showed inconsistency with theoretical yield, which motivates measurement of the cross section. The cross section measurement at Notre Dame will be performed in August 2023.

Acknowledgments

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Results

Thick target irradiation

- **The thick Ge$_{(m)}$ targets** (fully and not fully covered) are shown in ***Figure 4***.
- After **deuteron irradiation**, Ge$_{(m)}$ coin-type targets survived well (***Figure 3c***) with ^{71}As radioarsenic purity >99% .
- The **physical yield** is shown in ***Table 1*** with the maximum compared with the theoretical physical yield based on calculations using K.Otozai group data [11].
- **The experimental physical yield (Table 1)** from thick ^{70}Ge$_{(m)}$ coin targets showed inconsistency with the predicted physical yield (6.3 MBq·μA^{-1}·h^{-1}).

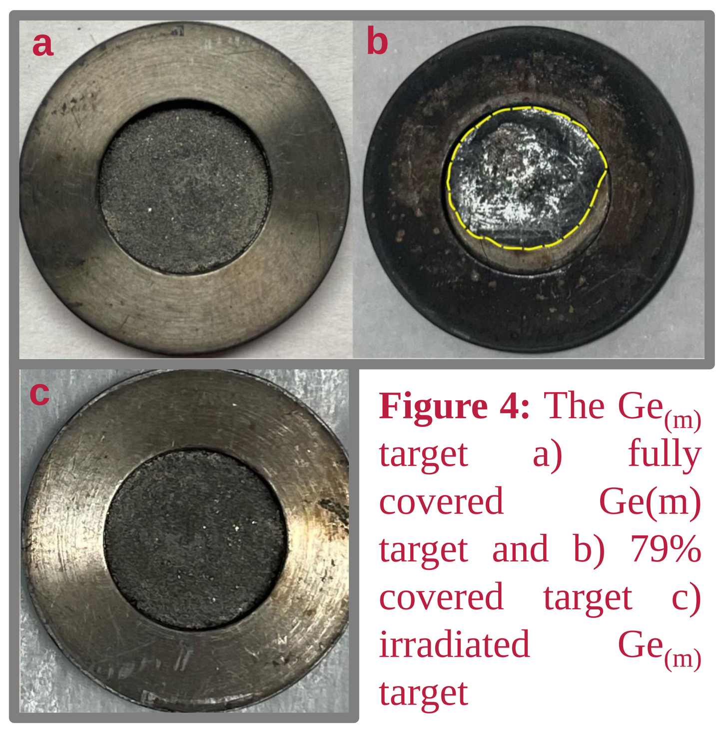

Figure 4: The Ge$_{(m)}$ target a) fully covered Ge(m) target and b) 79% covered target c) irradiated Ge$_{(m)}$ target

Estimated coverage (%)	Ge mass (mg)	Ge thickness (μm)	Experimental ^{71}As physical (MBq·μA-1·h-1)	Theoretical ^{71}As physical yield (MBq·μA-1·h-1)	n=
100	100.1 ± 0.1	285 ± 22.5	6.1 ± 0.1	6.29	2
100	68.8 ± 0.2	152 ± 6	5.91 ± 0.04		2
78 ± 7	87.4 ± 5.2	150 ± 30	5 ± 1		4
79 ± 8	69.1 ± 0.5	280 ± 50	4.1 ± 0.5		4

Table 1: Cyclotron yields at 8 MeV from coin-type ^{70}Ge targets

Thin foil cross section measurement

Material	thickness (μm)	Energy (MeV)	Activity (kBq)
			^{71}As
Ge	0.4	10.0	2.10
		9.5	2.23
		8.2	2822.50
		8.0	2.49
		7.6	2.44
		6.8	2.25
		5.8	1.64
		5.5	1.41
		4.2	0.45
		2.2	0.01

Table 2: The energy of deuteron for ^{nat}Ge foil and the calculated activity for ^{nat}Ge(d,x)^{71}As.

Material	thickness (μm)	Energy (MeV)	Activity (kBq)	
			^{61}Cu	^{56}Co
Ni	15	9.4	2822.50	4.25
		8.9	3144.22	4.13
		7.3	3545.18	3.25
		6.0	3028.43	2.26
		4.5	2125.21	1.17

Table 3: The energy of deuteron for ^{nat}Ni foil and the calculated activity for ^{nat}Ni(d,x)^{61}Cu and ^{nat}Ni(d,x)^{56}Co

- **The average deuteron energy, and expected ^{71}As, ^{61}Cu, and ^{56}Co activities** [6,7] in each target foil (**Table 2**) and beam monitor foil (**Table 3**) after a 4 h, 0.1 μA irradiation.
- The ^{70}Ge(d,n)^{71}As activation function and planned incident deuteron energies are shown in **Figure 5**.

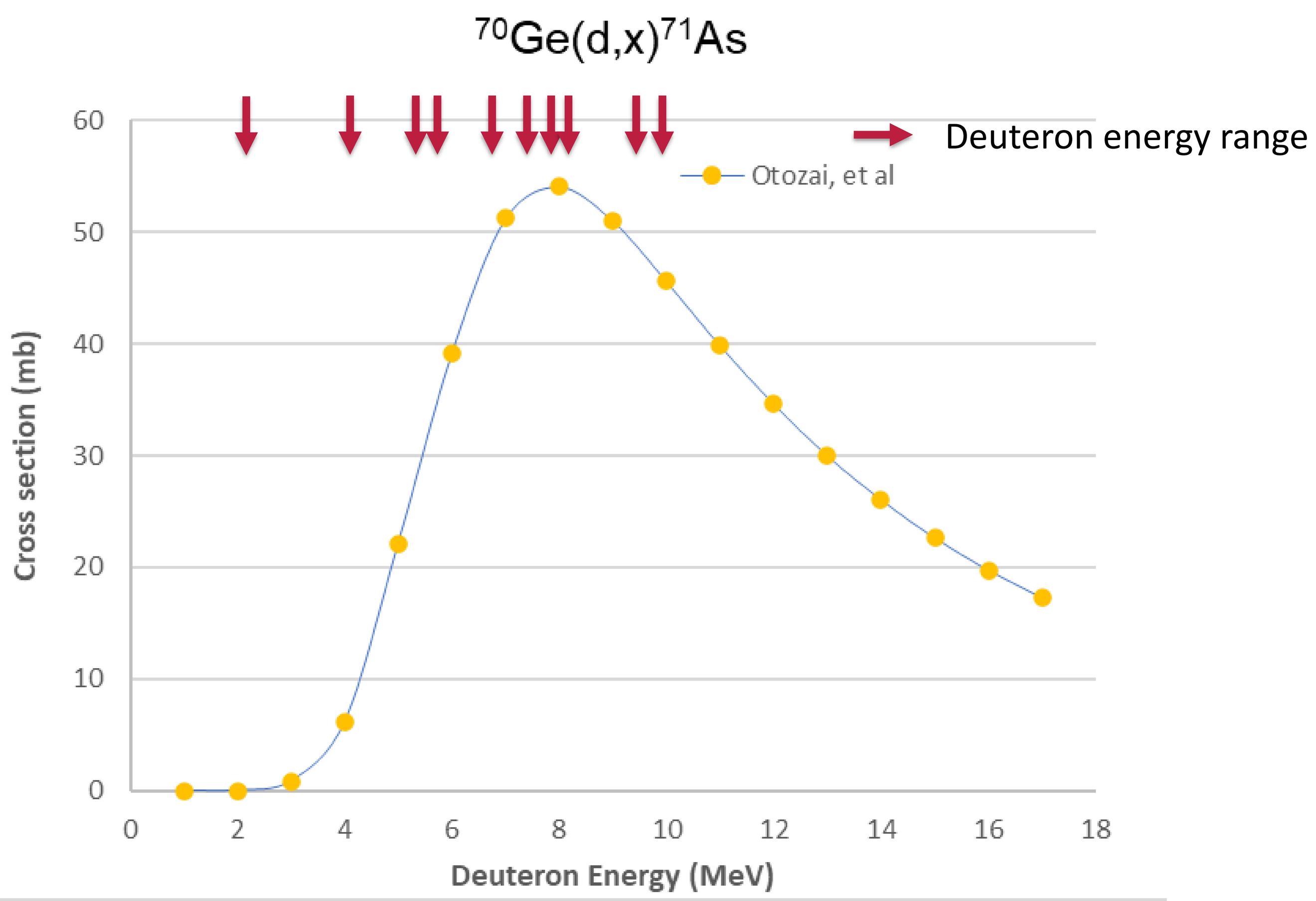

Figure 5: The ^{70}Ge(d,x)^{71}As activation cross section as function of incident deuteron energy

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