Ga-Co Compounds for Production of Germanium Radioisotopes

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Introduction:

Production of the theranostic isotope pair ^{69}Ge ($T_{1/2}$ = 39 hr, 21% β^+, E_{max} = 1205 keV) and ^{71}Ge ($T_{1/2}$ = 11 d, 100% EC) from elemental gallium targets is challenging but offers the prospect of a novel positron-emitting radionuclide for diagnostic experiments [1]. In its elemental form, gallium melts at 30 °C and the liquid reacts strongly with most metals, compromising many standard accelerator target systems. To avoid elemental Ga targets and improve thermal tolerance and survivability in the beam of the targets, Ga-Ni compounds have been synthesized previously using different techniques [1,2]. However incorporation of the nickel precursor, because of the (p,n) nuclear reactions, results in radiogallium overwhelmed by the presence of ^{60}Cu and as well as other activation products. Therefore, we have attempted to make Ga-Co intermetallic compounds to reduce undesirable, coproduced radionuclidic activities while increasing the gallium atom fraction in the target and the target's melting point. We report our preliminary investigation of the production of ^{69}Ge from these compounds.

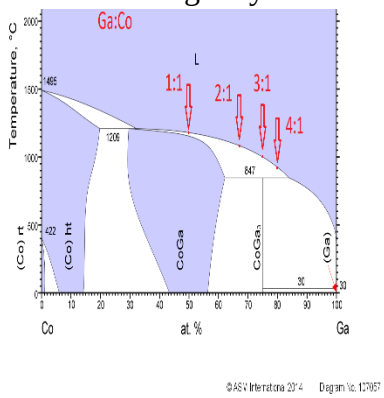


Figure 1: Phase Diagram for the Ga-Co binary system, with arrows at each of the proposed atom ratios highlighting the intersection of the liquidus line [3]

Methods:

Cobalt powder, gallium pellets, niobium rod stock and concentrated nitric acid (c. HNO_3) were obtained in high purity for this investigation. Samples of cobalt and gallium with atom ratios of Ga:Co varying from 2:1 to 4:1 (n=3 each) were placed in quartz tubes, flushed with argon gas, evacuated, and sealed into ampules. The quartz glass ampules were then placed in a muffle furnace at 1100 °C for approximately 2 hours, then rapidly quenched in a water bath. The resulting intermetallic pellets were hot-pressed into in-house machined niobium crucibles using a boron nitride tamper under argon gas flow at 1100 °C and quenched in a water bath. Targets were covered with 200 µm molybdenum degrader foils and irradiated on a GE PETrace at 5 µA for 1 hour. Post irradiation high purity germanium (HPGe) gamma spectrometry of the targets analyzed radionuclidic purity and activation yields. Following irradiation with ~13 MeV protons, radiochemical isolation of the germanium isotope was carried out by single column extraction chromatography using an extraction resin functionalized with N,N,N',N'-tetrakis-2-ethylhexyldiglycolamide (branched DGA, Eichrom) [4]. Irradiated targets were dissolved in 1-2 hours with 5 mL of c. HNO_3. Aliquots of the dissolved targets were diluted with c. HNO_3 and loaded onto approximately 200 mg of resin. The column was subsequently rinsed with c. HNO_3 and eluted with approximately 1.5 mL 0.1 M HNO_3. Contents of the load, wash, and elution fractions were quantified by HPGe to track the retention of the $^{69}Ge^{4+}$ ions on the resin columns and in the different eluates.

Results:

The masses of cobalt powder and chunks of gallium sealed in the ampules were observed to have formed orange-hot, glowing pellets when removed from the furnace immediately prior to quenching. The average change in mass during the pellet forging in the ampules observed initially was less than 5%. The average change in mass during the pellet pressing observed initially was less than 1%. None of the targets showed any appreciable melting or mass loss during the irradiation with up to 5 μA of ~13 MeV protons; evaluation of target beam current tolerance is ongoing. For ^{69}Ge, the first 3:1 atom ratio target irradiated showed an experimental yield of 0.36 mCi/(μA h). Evaluation of the activation yields for all the atom ratios is ongoing. Targets dissolve in c. HNO_3 and produce a dark reddish-brown solution. Initial separation results show over 60% retention of the ^{69}Ge on the branched DGA resin; further evaluation of the radiochemical isolation of ^{69}Ge from irradiated Ga-Co via single column extraction chromatography is ongoing.

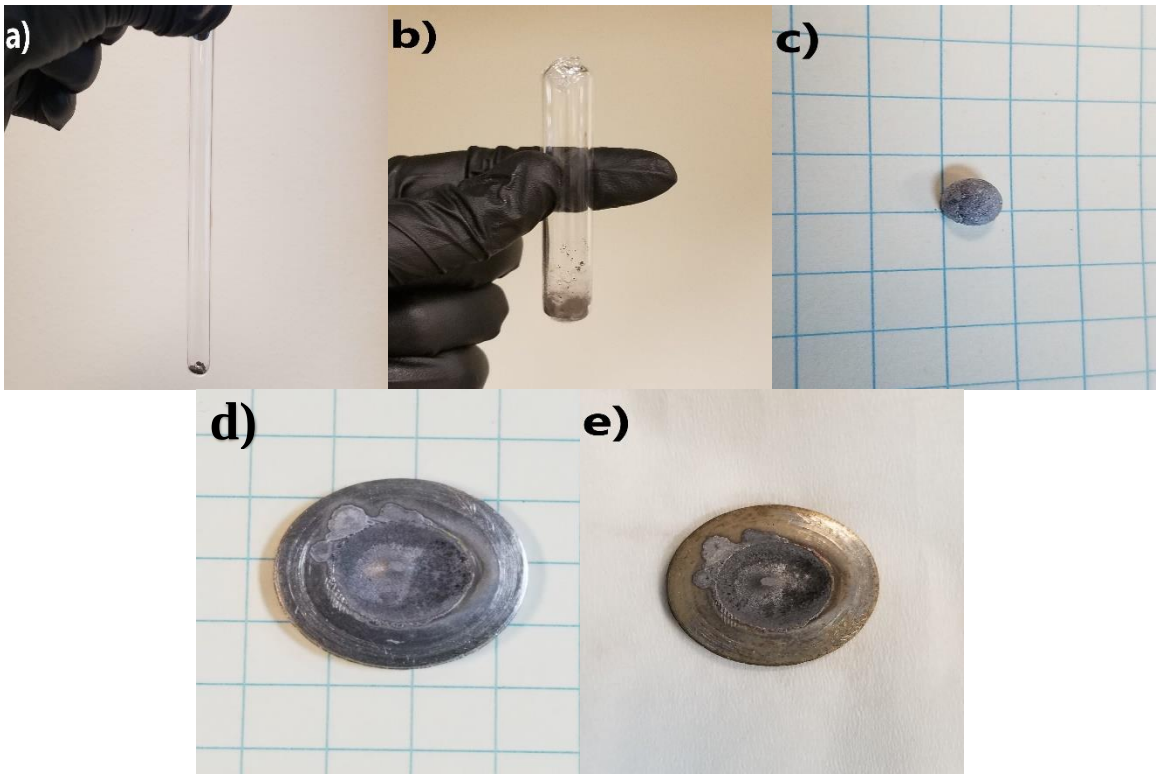
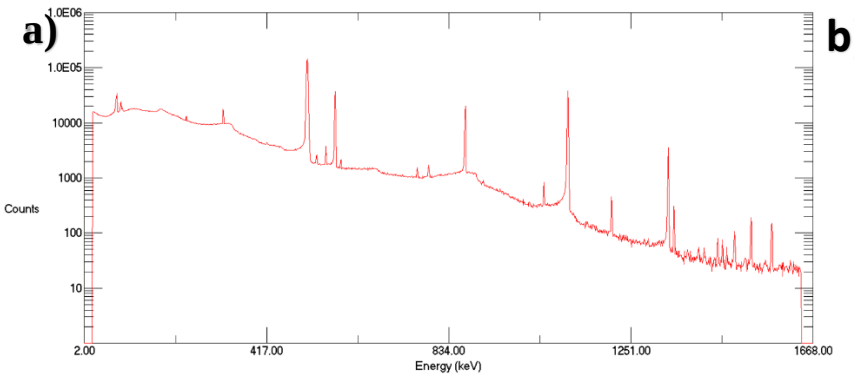


Figure 2: a) Cobalt and germanium loaded in quartz tube, b) Sealed ampoule, c) Pellet resulting from furnace forging, d) Ga-Co target formed from hot-pressing, e) Ga-Co target post-irradiation

a)



b)

Ge-69 Gamma Emission List			
E_gamma (keV)	I_gamma (%)	E_gamma (keV)	I_gamma (%)
234.79	0.37	912.7	0.061
318.63	1.55	1052.02	0.43
419.07	0.072	1106.77	36
511	47	1207.21	0.39
532.66	0.27	1336.6	4.5
553.35	0.69	1349.89	0.32
574.11	13.3	1404.7	0.018
587.4	0.32	1449.54	0.047
762.49	0.23	1487.96	0.097
788.14	0.34	1525.84	0.27
871.98	11.9	1572.85	0.23

Figure 3: a) Gamma spectrum obtained from irradiated Ga-Co target, and b) Table of prominent ^{69}Ge gamma peaks

Conclusions:

The Ga-Co compounds made here are promising accelerator targets. Improving the target's homogeneity and optimization of the Ga-Co atom ratio is expected to increase ^{69}Ge production yields, and optimization of the dissolution and chromatographic separation will allow for the preparation of no-carrier-added, high radiochemical purity ^{69}Ge for use in preclinical diagnostic imaging applications.

References:

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2. H.F. Valdovinos, S. A. Graves, T.E. Barnhart et al. Journal of Nuclear Medicine **55**, abstract 216 (2014)
3. A. Chari, A. Garay, R. Arróyave. Thermodynamic remodeling of the Co-Ga system **34(2)**,189-195 (2010)
4. H.F. Valdovinos. Production, labeling and in vivo studies with the theranostic positron-emitting radiometals ^{44}Sc, $^{55/58m/58g}$Co, $^{61/64}$Cu, ^{86}Y and ^{69}Ge. PhD Dissertation, University of Wisconsin-Madison, Madison, WI, (2017)