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PRODUCTION OF COSMOGENIC NUCLIDES IN THICK TARGETS BY ALPHA BOMBARDMENT I - SHORT-LIVED RADIOISOTOPES

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Production of short-lived cosmogenic nuclides in planetary surfaces and remote spacecraft detectors was simulated by bombarding "thick" C, Mg, Al, Si, SiO_2, Fe, Ni, and Ge targets with 60, 90 and 120 MeV alpha particles. Gold foils were used to monitor alpha particle fluence; product nuclides were measured by gamma ray spectroscopy. The results were used to calculate production yields and average cross sections.

Introduction

Cosmic ray particles interact with terrestrial and extraterrestrial matter to produce a large number of stable and radioactive nuclides, the so-called cosmogenic nuclides. Investigations of cosmogenic nuclide abundances in extraterrestrial matter (meteorites, cosmic dust, and planetary surfaces) provide a means of studying the history of both the irradiated matter and the cosmic radiation itself. Studies of cosmogenic nuclides in terrestrial matter have a wide variety of applications in the

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earth sciences. Unfortunately interpretation of cosmogenic nuclide data for both terrestrial and extraterrestrial samples is often hampered by a lack of knowledge of the production rates of these nuclides from the target materials present.

Another important application for cosmogenic nuclide studies lies in the area of gamma-ray astronomy and planetary gamma-ray spectroscopy [1]. When gamma-ray detectors are launched into space, build up of cosmogenic nuclides occurs due to cosmic ray bombardment. Corrections for this radiation contamination may be possible only after production rates of radionuclides from detector crystal materials have been studied in detail.

Two distinct classifications of cosmic radiation exist: galactic cosmic ray (GCR) particles, which originate outside the solar system, and solar cosmic ray (SCR) particles , which are emitted by the sun during solar flares. Both SCR and GCR particles are predominantly protons, with about 10% alpha particles and 1% heavier nuclides. The energies of most GCR particles range from 100 to 3000 MeV, while most SCR particles have energies below 100 MeV. The mean flux of SCR particles at 1 AU greatly exceeds that of GCR particles [2].

Production of cosmogenic nuclides in meteorites and planetary surfaces has been simulated by bombarding suitable target materials (e.g. Fe, Al, Si, Ni) with particle beams spanning the energy ranges of SCR and GCR particles [3-8]. Similar experiments may be performed to study radionuclide production from bombardment of radiation detector crystals (e.g. Si, Ge). Thus far, however, the majority of these experiments have been carried out using proton beams. Experimental data for production of cosmogenic nuclides in targets by alpha particle induced reactions are sparse [3], and data for alpha particle induced reactions on Ge containing targets are extremely rare [9]. Since alpha particles constitute a significant fraction of the total cosmic ray flux, their role in cosmogenic nuclide production must be studied in detail. In this investigation we simulated production of cosmogenic nuclides by SCR alpha particles by irradiating suitable target materials in alpha particle beams with energies $\leq$ 120 MeV.

Experimental

In order to study the production rates of cosmogenic nuclides from SCR alpha particles on planetary surfaces and in remote spacecraft detectors, we prepared thick targets for irradiation from the following materials: C, Mg, Al, Si, SiO_2, Fe, and Ni (all highly abundant in planetary surfaces), as well as Ge (commonly used in radiation detectors). All thick targets were cylindrical, each target being of sufficient thickness to act as a beam stop. We also prepared approximately 40 thin Au foils (0.001 mm thickness) to be used as flux monitors. All targets were prepared from ultrapure materials.

Irradiations of all targets were performed with the 88 Inch Cyclotron at Lawrence Berkeley Laboratory. Each thick target was irradiated for 10-45 minutes with 60, 90 or 120 MeV alpha particles using beam currents of 1.0, 0.4-0.6, and 0.5-0.6 μA respectively. Targets were mounted in a Faraday cup, the total charge accumulating in the cup during each irradiation being used to monitor the beam fluence. Gold foils were also used to monitor beam fluence. For some irradiations two gold foils were used - one mounted in front of the target (monitor) and a second foil mounted upstream (cover) to make up for recoil losses in the Au monitor foil. In other irradiations the cover foil was omitted.

After a suitable cooling time, the targets were removed to a well-shielded area and counted by gamma-ray spectroscopy. Gamma-ray spectra for selected targets are shown in Figure 1. Each target was counted 3 or 4 times in order to establish decay curves for the product nuclides. Measurements were taken using four Ge detectors in conjunction with a PC based multichannel acquisition system. Detector efficiency was determined using standard calibrated sources of ^{133}Ba, ^{152}Eu, ^{56}Co, ^{60}Co, and ^{88}Y; efficiencies were fitted to an exponential curve. Fits were better than ± 15% for all measured efficiencies. Peak fitting and integration for all gamma-ray spectra were carried out using the GELIFT program.

Alpha particle fluence (total incident alpha particles) for each irradiation was calculated using the total electric charge accumulated in the Faraday cup during irradiation; the results are given in Table 1. The accuracy of the current integration measurements was checked by calculating beam fluences using ^{196}Au activities measured in the Au foils and previously determined values for the $^{197}Au(\alpha,\alpha n)^{196}Au$ cross sections for 60, 90 and 120 MeV alpha particles [8]. While particle fluences calculated from Au cover foil data appear to be consistently low, as would be expected due to recoil losses, fluences calculated using Au monitor data are generally in good agreement (± 15%) with current integration values. Current integration measurements were also checked against the product of beam current with irradiation time; agreement between these values was better than ± 10% for each irradiation.

Production yields for radioactive nuclides produced in the thick targets were then calculated as number of nuclei produced/incident alpha particles. Average cross sections, $\sigma(E)$, for the production of these nuclides over the energy ranges 120-90 MeV and 90-60 MeV were then calculated using the equation

$$\sigma(E) = \frac{\text{Yield(E2)} - \text{Yield(E1)}}{E} * \frac{dE}{dX}$$

where Yield(E2) and Yield(E1) are the production yields for the nuclide at the upper and lower energies of the range, E is the energy range in MeV, and dE/dX is the average total stopping power of the alpha particles in MeV*cm^2/atom [10].

Results and Discussion

Table 2 gives halflifes, as well as energies and intensities of major gamma rays, for selected nuclides produced in the targets. Calculated production yields and average cross sections are given in Table 3.

Uncertainties in the calculated production yields were estimated from counting statistics, efficiency curve fits and uncertainties in calculated beam fluences. Uncertainties in the production yields for most nuclides are estimated at $\pm$ 15%.

A major problem with the germanium targets was their fragility. The Ge target irradiated at 90 MeV was broken prior to counting. Hence because of larger errors in counting efficiency (due to a change in counting geometry) and the possibility of target material loss prior to counting, uncertainties in production yields for nuclides in this target may be higher than reported.

Prior data for cross sections of ^{4}He-induced reactions in these target materials are sparse; those literature values which do exist are given in Table 3. Our cross sections for the production of ^{7}Be from C are in fair agreement with cross sections given by Michel [3]. While cross sections obtained by Michel for the production of ^{7}Be from Si using 90-60 MeV alpha particles (~4 mb) are also in good agreement with our value (4.0 mb), Michel's cross sections for production of this nuclide using 120-90 MeV alphas (6-7 mb) are more than 50% higher than our value (4.0 mb).

Conclusions and Work in Progress

Production yields and average cross sections for many short-lived radioisotopes produced by 60, 90 and 120 MeV alpha particle irradiations of C, Mg, Al, Si, SiO_2, Fe, Ni and Ge targets have been reported for the first time, providing basic data for cosmogenic nuclide production calculations and for Ge- detector radionuclide buildup in space. Long-lived cosmogenic nuclides produced in these targets (^{10}Be, ^{26}Al) have been isolated using chemical techniques described by Theis and Englert [11], and measured by accelerator mass spectrometry. Production yields for these nuclides will be reported in a subsequent paper.

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Table 1 - Calculated Beam Fluences for Thick Target Irradiations.

Target	Alpha Energy (MeV)	Beam Current (μA)	Irradiation Time (s)	Total Charge (μC)	Beam Fluence (Total α)
C	60	1.0	2040	1876	5.86×10^{15}
	90	0.6	1500	876	2.73×10^{15}
	120	0.6	1260	676	2.11×10^{15}
Mg	60	1.0	2100	1985	6.20×10^{15}
	90	0.6	1020	599	1.87×10^{15}
	120	0.6	780	400	1.25×10^{15}
Al	60	0.35	2700	981	3.06×10^{15}
	90	0.4	2160	804	2.51×10^{15}
	120	0.6	900	554	1.73×10^{15}
Si	60	1.0	1920	1721	5.37×10^{15}
	90	0.6	1560	800	2.50×10^{15}
	120	0.5	1080	549	1.71×10^{15}
SiO_2	60	1.0	1980	1810	5.65×10^{15}
	90	0.6	1080	602	1.88×10^{15}
	120	0.5	780	400	1.25×10^{15}
Fe	60	1.0	2100	2131	6.65×10^{15}
	90	0.6	1440	810	2.53×10^{15}
	120	0.5	1380	668	2.08×10^{15}
Ni	60	1.0	1800	1894	5.91×10^{15}
	90	0.6	780	500	1.56×10^{15}
	120	0.5	840	402	1.25×10^{15}
Ge	60	1.0	1920	1805	5.63×10^{15}
	90	0.6	1440	833	2.60×10^{15}
	120	0.5	1080	549	1.71×10^{15}

Table 2 - Radionuclides Observed in the Irradiated Thick Targets.

Product Nuclide	Halflife	Gamma Energy (keV)	Gamma Intensity (%)	Target Element(s)
^{7}Be	53.3 d	477	10.4	C,O,Mg,Al,Si
^{22}Na	2.62 y	1274	99.9	Mg,Al,Si
^{28}Mg	21.1 h	401	35.9	Mg,Al,Si
^{48}V	16 d	984	100	Fe,Ni
^{51}Cr	27.7 d	320	9.80	Fe,Ni
^{52}Mn	5.59 d	1434	100	Fe,Ni
^{54}Mn	312 d	835	100	Fe,Ni
^{56}Mn	2.58 h	1811	30.0	Fe,Ni
^{52}Fe	8.2 h	169	100	Fe,Ni
^{55}Co	17.5 h	1408	16.9	Fe,Ni
^{56}Co	78.8 d	846	100	Fe,Ni
^{57}Co	271 d	122	85.6	Fe,Ni
^{58}Co	70.8 d	810	99.4	Fe,Ni
^{60}Co	5.3 y	1333	100	Ni
^{56}Ni	6.1 d	158	99.0	Fe,Ni
^{57}Ni	36 h	1377	77.9	Fe,Ni
^{61}Cu	3.41 h	283	13.1	Ni
^{62}Zn	9.2 h	548	15.3	Ni,Ge
^{65}Zn	244 d	1115	50.7	Ni,Ge
^{72}Ga	14.1 h	834	95.6	Ge
^{68}Ge	288 d	1077	3.30	Ge
^{69}Ge	39 h	574	11.0	Ge
^{71}As	61 h	175	83.6	Ge
^{72}As	26 h	834	80.1	Ge
^{73}As	80.3 d	53	10.5	Ge
^{74}As	17.8 d	596	60.3	Ge
^{76}As	26.3 h	559	44.6	Ge
^{72}Se	8.4 d	834	92.0	Ge
^{73}Se	7.1 h	361	96.7	Ge
^{75}Se	119 d	265	58.2	Ge

Table 3 - Production Yields and Cross Sections for Selected Radionuclides in the Thick Targets.

Product Nuclide	Target Material	Production Yields Nuclei/incident particles			Average Production Cross Section (mb)†			
		60MeV	90MeV	120MeV	120-90MeV		90-60MeV	
^{7}Be	C	1.47×10^{-4}	6.37×10^{-4}	1.17×10^{-3}	26	*30-35*	32	*35*
^{28}Mg	Mg	1.54×10^{-6}	3.76×10^{-6}	5.05×10^{-6}	0.11		0.26	
^{24}Na	Mg	9.60×10^{-5}	3.15×10^{-4}	5.33×10^{-4}	19		25	
^{22}Na	Mg	4.78×10^{-4}	1.32×10^{-3}	2.07×10^{-3}	66		98	
^{7}Be	Mg	5.06×10^{-6}	4.69×10^{-5}	1.19×10^{-4}	6.4		4.9	
^{28}Mg	Al	1.28×10^{-6}	3.68×10^{-6}	5.45×10^{-6}	0.17		0.30	
^{24}Na	Al	3.39×10^{-5}	3.39×10^{-4}	6.82×10^{-4}	32		38	
^{22}Na	Al	1.60×10^{-4}	3.50×10^{-4}	7.79×10^{-4}	41		23	
^{7}Be	Al	2.33×10^{-6}	3.05×10^{-5}	6.97×10^{-5}	3.7		3.5	
^{28}Mg	Si	8.38×10^{-8}	1.04×10^{-6}	2.27×10^{-6}	0.13		0.13	
^{24}Na	Si	3.14×10^{-6}	6.06×10^{-5}	1.94×10^{-4}	14		7.7	
^{22}Na	Si	1.00×10^{-5}	3.33×10^{-4}	5.79×10^{-4}	25		43	
^{7}Be	Si	4.41×10^{-6}	3.39×10^{-5}	7.29×10^{-5}	4.0	*6-7*	4.0	*4*
^{28}Mg	SiO_2	3.94×10^{-8}	5.06×10^{-7}	1.07×10^{-6}	0.13		0.14	
^{24}Na	SiO_2	6.64×10^{-7}	2.61×10^{-5}	9.06×10^{-5}	15		7.7	
^{22}Na	SiO_2	1.79×10^{-6}	1.25×10^{-4}	2.70×10^{-4}	33		37	
^{7}Be	SiO_2	1.68×10^{-5}	1.09×10^{-4}	2.68×10^{-4}	37		28	
^{57}Ni	Fe	3.65×10^{-5}	4.96×10^{-5}	5.60×10^{-5}	1.1		2.9	
^{56}Ni	Fe	1.37×10^{-6}	3.68×10^{-6}	8.23×10^{-6}	0.77		0.51	
^{58}Co	Fe	1.08×10^{-3}	1.32×10^{-3}	1.39×10^{-3}	12		53	
^{57}Co	Fe	1.15×10^{-3}	1.45×10^{-3}	1.89×10^{-3}	75		67	
^{56}Co	Fe	2.00×10^{-4}	7.15×10^{-4}	1.12×10^{-3}	69		115	
^{55}Co	Fe	1.31×10^{-5}	6.71×10^{-5}	1.33×10^{-4}	11		12	
^{52}Fe	Fe	5.20×10^{-7}	2.61×10^{-6}	7.98×10^{-6}	0.91		0.47	
^{56}Mn	Fe	5.04×10^{-6}	3.33×10^{-5}	6.51×10^{-5}	5.4		6.3	
^{54}Mn	Fe	3.47×10^{-4}	8.00×10^{-4}	1.55×10^{-3}	127		101	
^{52}Mn	Fe	2.42×10^{-5}	3.14×10^{-4}	6.96×10^{-4}	65		64	
^{51}Cr	Fe	5.76×10^{-5}	2.62×10^{-4}	1.08×10^{-3}	139		45	
^{48}V	Fe		1.85×10^{-4}	1.57×10^{-4}	24			
^{62}Zn	Ni	4.35×10^{-5}	5.02×10^{-5}	5.65×10^{-5}	1.1		1.6	
^{61}Cu	Ni	4.13×10^{-4}	5.48×10^{-4}	6.44×10^{-4}	17		32	
^{57}Ni	Ni	6.30×10^{-5}	1.85×10^{-4}	3.69×10^{-4}	33		29	
^{56}Ni	Ni	2.99×10^{-6}	1.11×10^{-5}	2.68×10^{-5}	2.8		1.9	
^{58}Co	Ni	2.94×10^{-4}	9.13×10^{-4}	1.62×10^{-3}	127		145	
^{57}Co	Ni	3.73×10^{-4}	1.01×10^{-3}	2.08×10^{-3}	193		149	
^{56}Co	Ni	2.51×10^{-4}	5.76×10^{-4}	1.15×10^{-3}	103		76	
^{55}Co	Ni	1.39×10^{-5}	1.05×10^{-4}	2.17×10^{-4}	20		21	
^{52}Fe	Ni		1.45×10^{-6}	5.69×10^{-6}	0.76			
^{56}Mn	Ni		2.17×10^{-6}	1.13×10^{-5}	1.6			
^{54}Mn	Ni		1.65×10^{-4}	4.92×10^{-4}	59			

Product Nuclide	Target Material	Production Yields Nuclei/incident particles			Average Production Cross Section (mb)†	
		60MeV	90MeV	120MeV	120-90MeV	90-60MeV
^{52}Mn	Ni	$3.94x10^{-6}$	$1.02x10^{-4}$	$2.88x10^{-4}$	34	23
^{51}Cr	Ni		$7.58x10^{-6}$	$3.28x10^{-4}$	4.5	
^{48}V	Ni		$7.27x10^{-6}$	$3.84x10^{-5}$	5.6	
^{76}As	Ge	$6.75x10^{-5}$	$1.35x10^{-4}$	$1.65x10^{-4}$	6.0	18
^{74}As	Ge	$2.59x10^{-4}$	$6.86x10^{-4}$	$9.61x10^{-4}$	65	111
^{71}As	Ge	$2.14x10^{-4}$	$6.25x10^{-4}$	$1.10x10^{-3}$	95	107
^{69}Ge	Ge	$9.06x10^{-5}$	$5.31x10^{-4}$	$1.08x10^{-3}$	109	114
^{68}Ge	Ge	$4.23x10^{-5}$	$1.75x10^{-4}$	$4.50x10^{-4}$	49	35
^{65}Zn	Ge	$7.73x10^{-6}$	$7.17x10^{-5}$	$3.11x10^{-4}$	48	17

†Literature values are in italics.

Figure Captions

Figure 1. Gamma-ray spectra for targets irradiated with 60 MeV alpha particles:

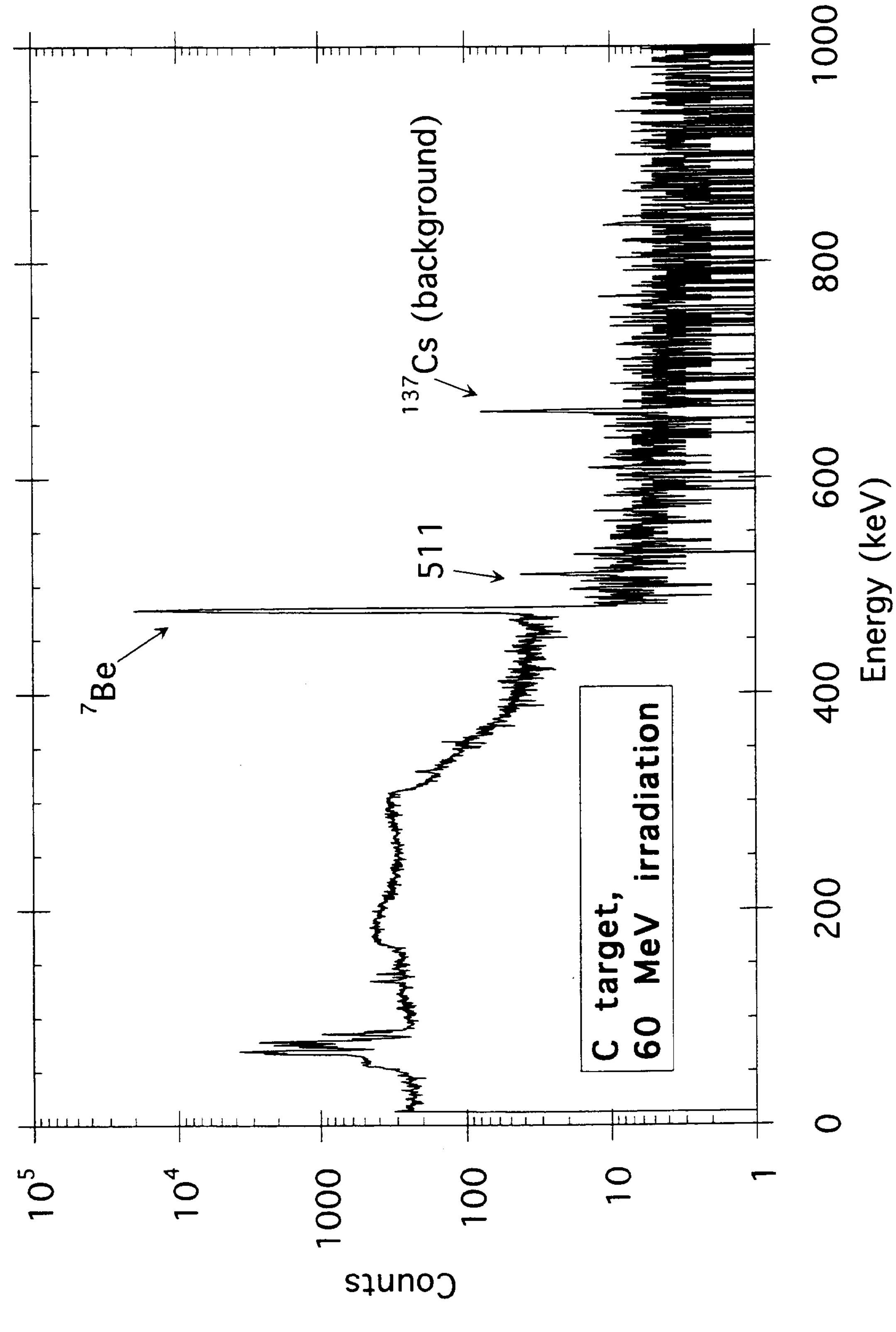
a) carbon

b) magnesium

c) silicon

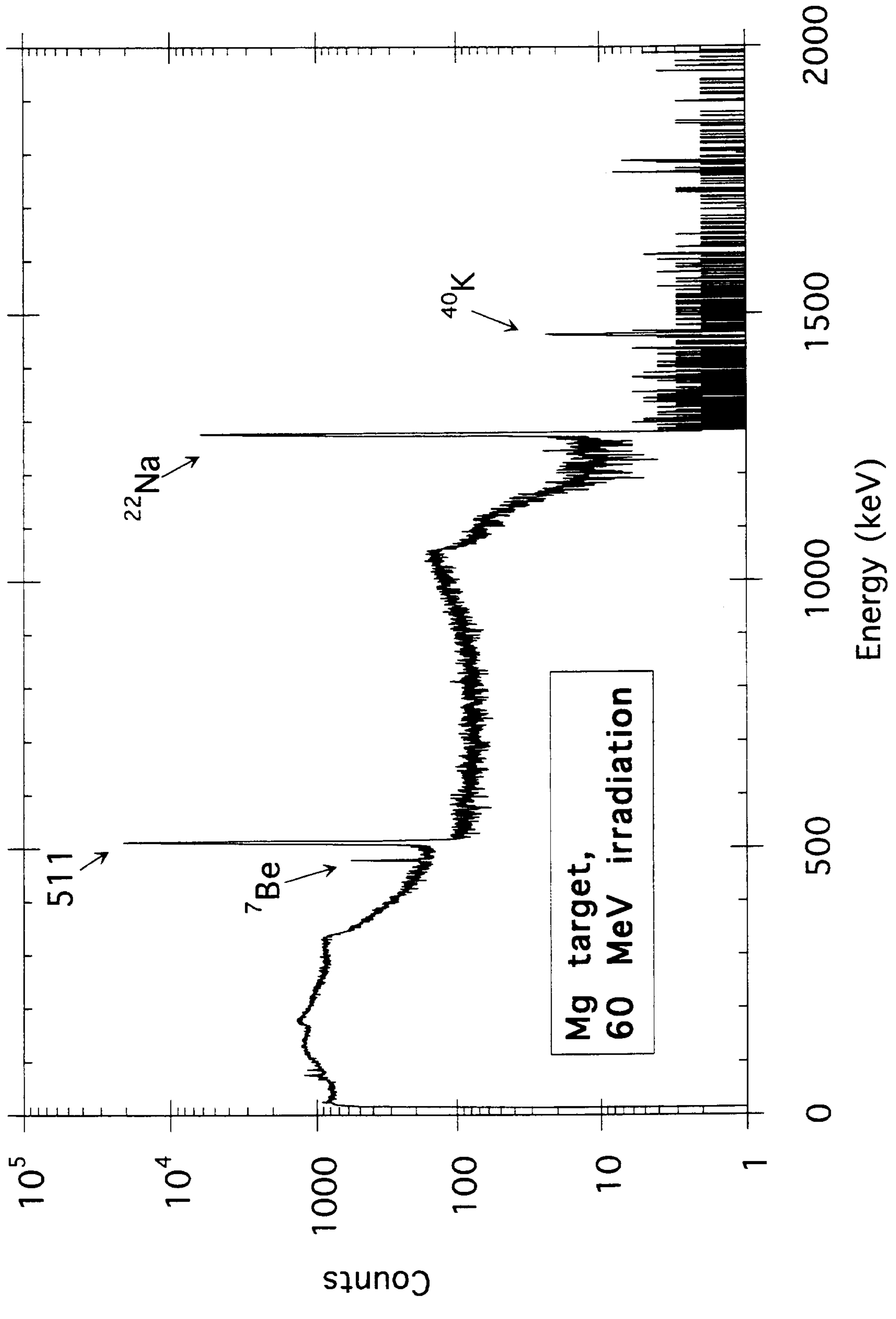
d) iron

e) germanium.



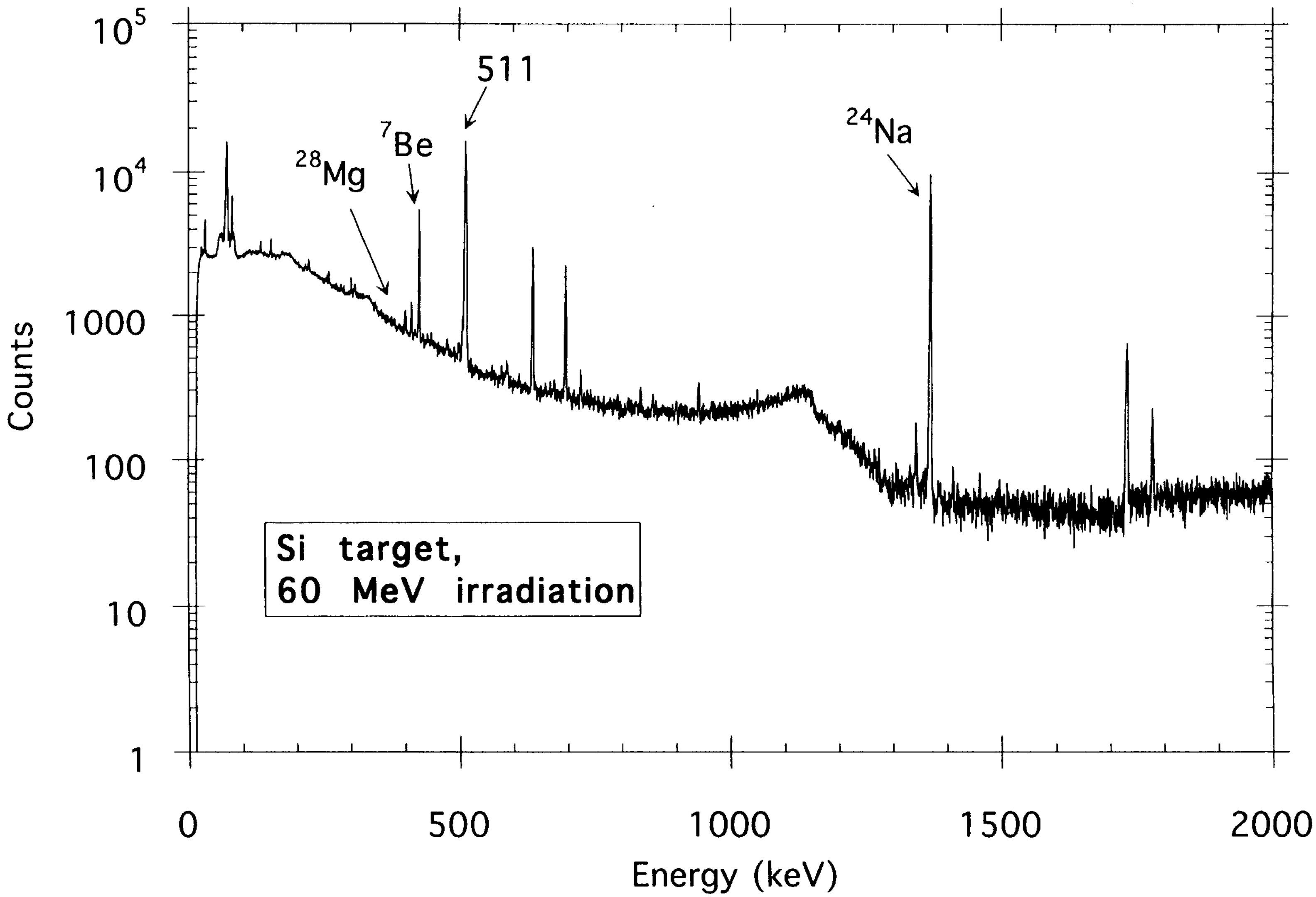
C target,
60 MeV irradiation
7Be
511
137Cs (background)
Energy (keV)
Counts
0
200
400
600
800
1000
1
10
100
1000
10^4
10^5

Figure 1a



Mg target,
60 MeV irradiation
511
7Be
22Na
40K
Counts
Energy (keV)
10^5
10^4
1000
100
10
1
0
500
1000
1500
2000

Figure 1b.



511
^{7}Be
^{28}Mg
^{24}Na
Si target,
60 MeV irradiation
Counts
10^5
10^4
1000
100
10
1
0
500
1000
1500
2000
Energy (keV)

Figure 1c

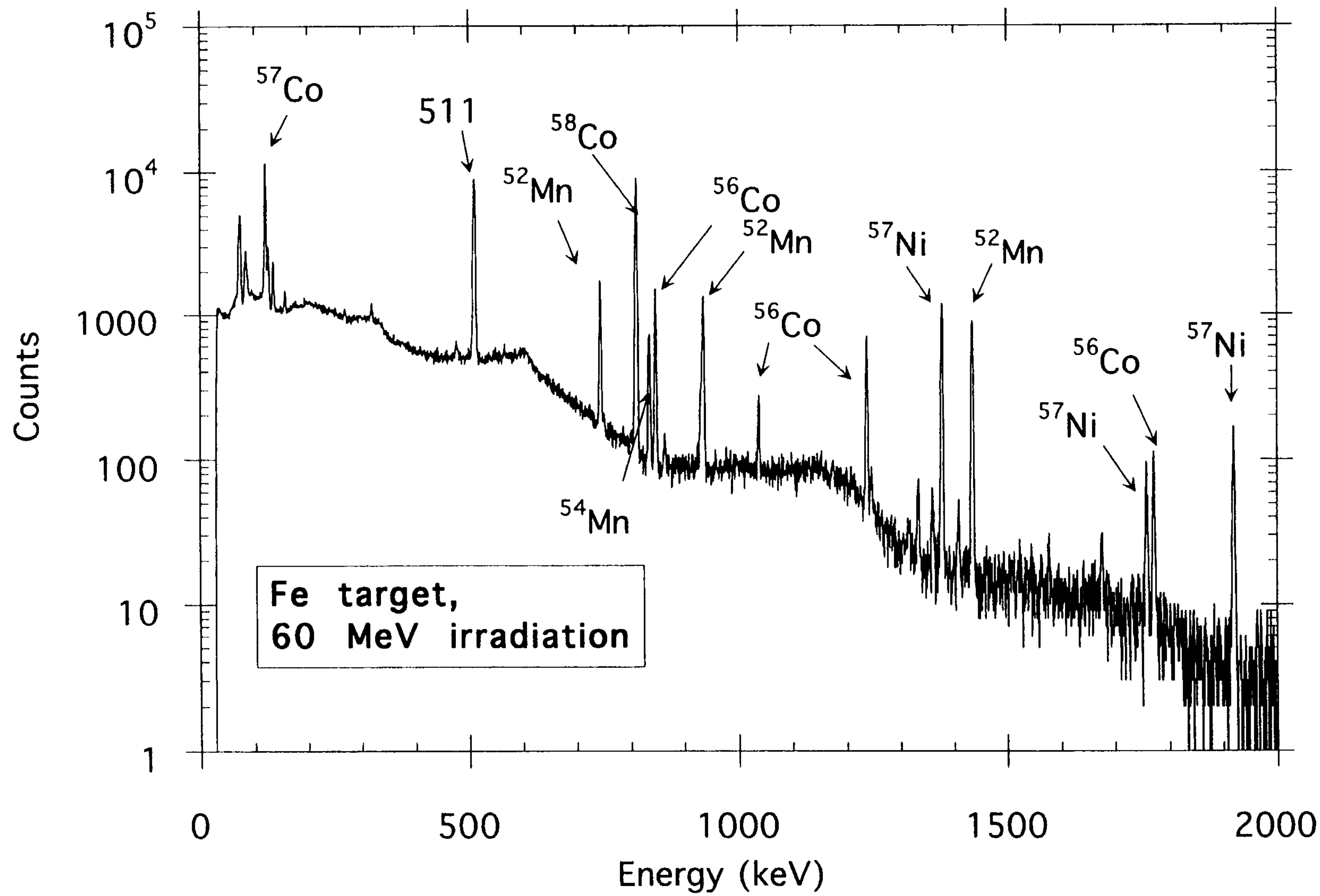

57Co
511
58Co
52Mn
56Co
52Mn
57Ni
52Mn
56Co
56Co
57Ni
57Ni
54Mn
Fe target,
60 MeV irradiation
Counts
Energy (keV)

Figure 1d

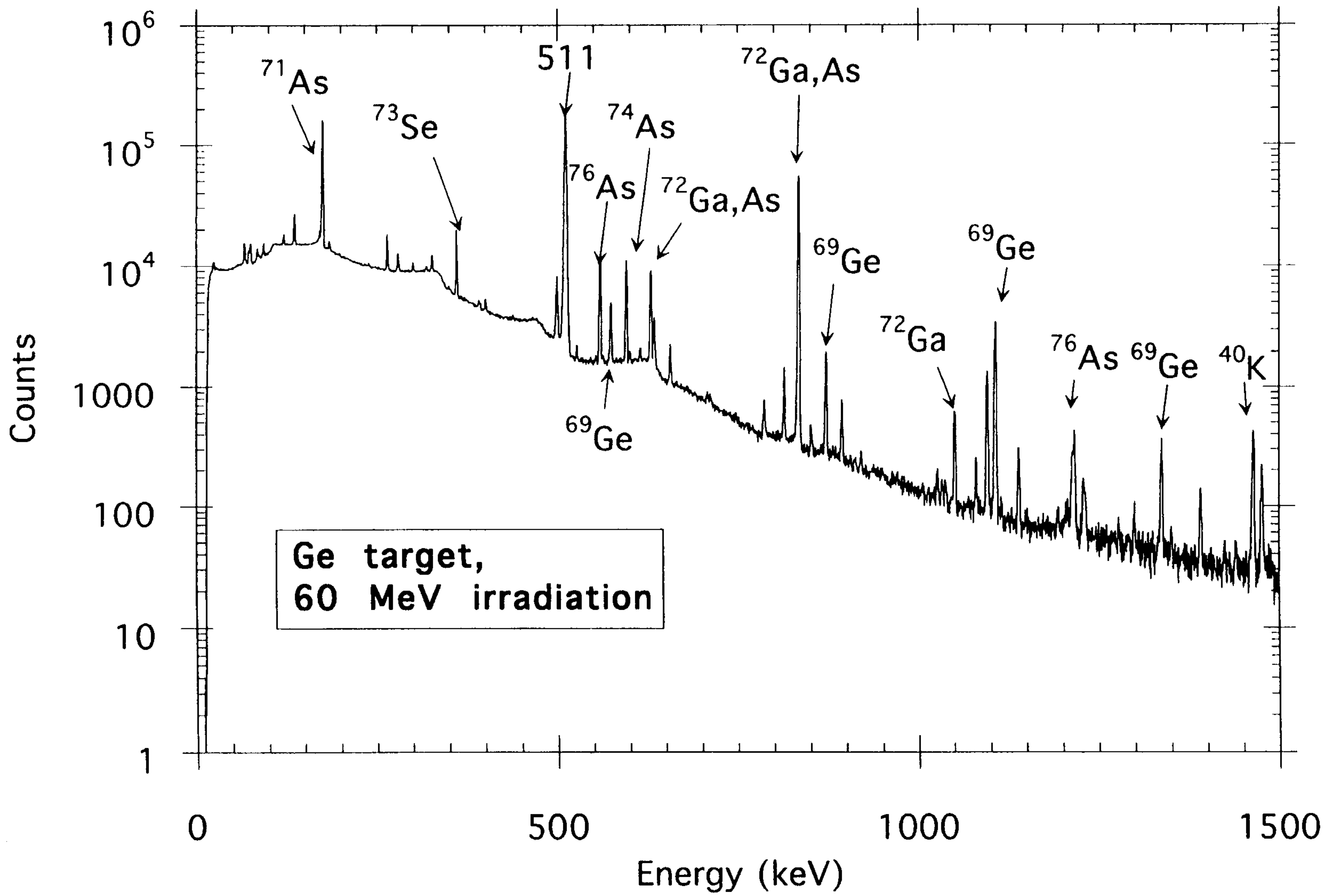

Counts
Energy (keV)
10^6
10^5
10^4
1000
100
10
1
0
500
1000
1500
^{71}As
^{73}Se
511
^{76}As
^{74}As
^{72}Ga,As
^{69}Ge
^{72}Ga,As
^{69}Ge
^{72}Ga
^{69}Ge
^{76}As
^{69}Ge
^{40}K
Ge target,
60 MeV irradiation

Figure 1e