Michigan State University

National Superconducting Cyclotron Laboratory

DETERMINATION OF RELATIVE RADIOACTIVE ABUNDANCES IN NEUTRON ACTIVATED FACILITY PARTS AT THE NSCL

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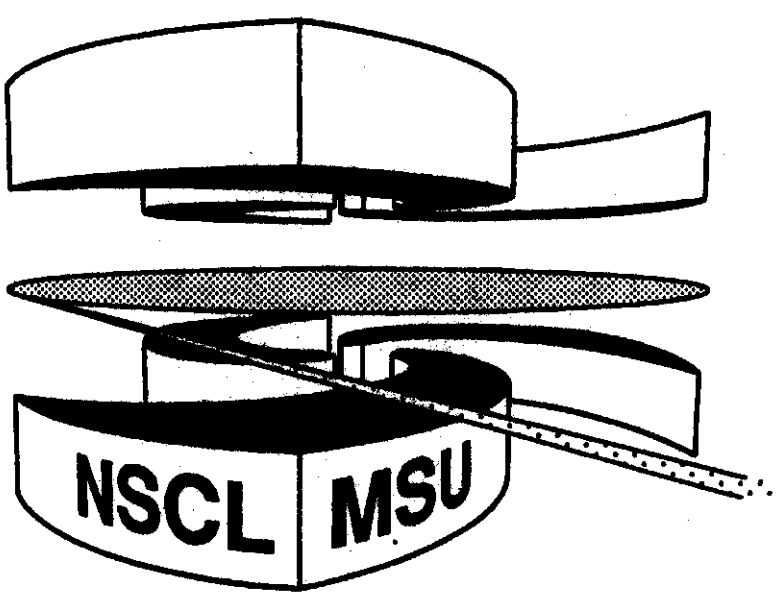

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Determination of Relative Radionuclide Abundances in Neutron Activated Facility Parts at the NSCL

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Abstract

Facility parts from the NSCL were analyzed by gamma-ray spectroscopy to determine relative radionuclide abundances produced by neutron activation during normal accelerator operations. A high-purity germanium detector calibrated for energy and efficiency was used to obtain spectra from brass, copper, stainless steel, aluminum and a group of miscellany from an electrical junction box. The isotopes ^{57}Co, ^{58}Co, ^{54}Mn, ^{56}Co, ^{65}Zn and ^{60}Co were identified in the activated copper; activation of brass produced ^{65}Zn and ^{60}Co; activation of stainless steel produced ^{57}Co, ^{51}Cr, ^{58}Co, ^{54}Mn, ^{65}Zn, ^{60}Co and ^{56}Co. The isotopes ^{58}Co, ^{54}Mn, ^{65}Zn and ^{60}Co were identified in the activated electrical junction box.

I. Introduction

The disposal of radioactive waste materials presents many problems for the research community. These include expense, storage space, environmental considerations, public health issues and security. Most often the volume of radioactive waste is reduced by segregating the waste into specific radionuclides and concentrating the waste material. However, engineering control of experimental conditions may be also used to limit the production of radioactive waste.

The production of waste materials by accelerator laboratories is unique in that most waste is in the form of activated facility (accelerator, beam-line and experimental apparatus) parts. The purpose of this study was to identify the radionuclides produced in facility parts at the NSCL and to determine their relative abundance. The identification of radionuclides within the activated parts is necessary to determine decay times for storage purposes. But, studies such as this can be used for engineering control over what materials are used for facility parts in high neutron flux environments, so that the amount of waste produced can be minimized.

II. Experimental

A. Detector information and calibration

High resolution gamma ray spectroscopy was used to identify radionuclides in some representative materials used at the NSCL. All gamma ray spectra were obtained using an Ortec high-purity germanium detector, model GEM 90220-P, at 2500 V positive bias, supplied from an Ortec 459 bias supply. For signal conversion, a Canberra Model 2020 amplifier, a Canberra Model 8075 ADC and a PC-based Canberra System 100 multichannel analyzer [1] were used.

The detector was calibrated for energy with a known nuclide source, (NIST standard SRM 4275C-69, September 1, 1988) using the following photopeaks: ^{155}Eu, 86.6keV; ^{154}Eu, 123.1keV; ^{125}Sb, 176.4keV; ^{154}Eu, 248.0keV; ^{125}Sb, 427.4keV; ^{125}Sb, 463.4keV; ^{154}Eu, 1274.4keV; and ^{154}Eu, 1596.4keV. This yielded Energy (keV) = (0.6984) Ch + 6.2556 as the energy calibration equation.

The photopeak efficiency calibration was performed using the same spectral peaks with their known intensities and uncertainties. The efficiency equation for the energy range 80 keV to 2000 keV was determined to be

$$\ln(Eff) = -2.26 - 0.258(\ln E) - 0.0262(\ln E)^2$$

for a source-to-detector distance of 3.5 inches. A plot of the efficiency equation and the individual calibration points is presented in Figure 1. The uncertainty in the efficiency calibration was taken to be an average of the peak uncertainties from the standard source calculated by the Spectran [1] calibration software algorithm.

B. Experimental Set-up

The detector was placed inside a shield constructed of 2 inch-thick lead bricks, with both ends open. Two layers of sheet metal were fitted around the detector sides. The outermost layer was copper, 0.013 inch thick, to shield from lead x-radiation. The innermost layer was cadmium, 0.023 inch thick, for additional shielding from radiation induced within the copper metal. The end plate of the detector was left exposed to the source. Calibrations using the gamma-ray standard, and all data collection, were performed with the sources 2 inches from the lead shield, centered to the detector face, giving an overall source-to-detector distance of 3.5 inches.

C. Sample Description

Five different materials were analyzed by gamma-ray spectroscopy. These were brass, stainless steel, copper, aluminum and a collection of miscellany from and including an electrical junction box. The parts were from the NSCL's A1200 radioactive ion beam facility and were neutron-activated during normal accelerator operations. It was assumed that they decayed without further activation from February 7, 1994, when operations ceased to allow an upgrade of the A1200 facility.

The brass samples were two groups of brass fixtures. The first group, labelled B1, consisted of two brass quick-disconnect air-line fittings, each with an overall length of 4.25 inches, and a straight brass tube with fittings, having an overall length of 8.5 inches. All three pieces had an average outside diameter of 0.75 inch. The total mass for the brass content of B1 was 298.75 grams. The second group of brass, labelled B2, consisted of two brass fittings, each with an overall length of 2.25 inches, and an outside diameter of 0.75 inch, and a threaded brass tube, having a length of 0.88 inch, and an outside diameter 0.5 inch. The total mass of brass in the sample B2 was 753.68 grams.

The stainless steel sample was a t-shaped tube having a length of 5.75 inches, a height of 3.25 inches, an inside diameter of 1.38 inches, and outside diameter of 1.44 inches. The total mass of stainless steel was 1407.4 grams.

The copper sample was a group of conflat vacuum seal rings, composed of soft-annealed, oxygen-free high conductivity copper, all having a thickness of 0.06 inch; one small ring had an inside diameter of 1.44 inches and outside diameter of 1.88 inches, 6 medium sized rings had inside diameters of 4 inches and outside diameters of 4.75 inches, 3 large rings had inside diameters of 6 inches and outside diameters of 6.75 inches. The total mass of copper was 617.15 grams.

The aluminum sample was a beam-line vacuum pipe that was 20 inches long, having an inside diameter of 3.5 inches and an outside diameter of 3.75 inches. The mass of aluminum was 1589.0 grams. A "hot spot" on the large aluminum tube was determined with a NaI detector. This region was centered toward the detector face. This activity was possibly from direct beam irradiation, rather than from neutron activation.

A background spectrum was collected for 7200 live seconds. In each case sample spectra were also collected for 7200 live seconds. All sample spectra were analyzed after background subtraction.

Elemental analysis was performed by comparison of the measured gamma ray energies with those in the database, Evaluated Nuclear Structure Data File (ENSDF), found within the GDISP [2] software, and by cross-referencing to the Table of Radioactive Isotopes [3].

III. Results

The resulting stripped spectra are presented in figures 2 through 7. Photopeaks from decays of the most abundant radionuclides are labeled with their emission energies. The identified radionuclides from the spectra obtained are presented in Table 1.

For each of the spectra obtained, a calculation of relative abundance at the date of the last activation was performed. First the activity was measured by determining the total counts per second in the photopeak, then dividing this by the detector efficiency and by the decay probability. The rate of disintegration is then divided by the decay constant λ, where $\lambda = \frac{\ln 2}{t_{1/2}}$. Here, $t_{1/2}$ is the half-life of a given radionuclide. This calculation yields a representative number of atoms present at the time of spectral analysis. The actual number of atoms present in a given sample was not calculated due to the geometry of these extended sources. To determine a representative number of atoms of each radionuclide present at the time activation ceased, the equation $N = N_0 e^{-\lambda t_d}$ was used, where t_d represents the time of decay. Corrections for peak summing were also performed. The abundances calculated are presented in Table 2.

Consistent with the findings of Thomas [4], activated copper contained ^{57}Co, ^{58}Co, ^{54}Mn, ^{56}Co, ^{65}Zn and ^{60}Co; the activated brass contained ^{65}Zn and ^{60}Co; the activated stainless steel contained ^{57}Co, ^{51}Cr, ^{58}Co, ^{54}Mn, ^{65}Zn, ^{60}Co and ^{56}Co. The parts from the electrical junction box contained ^{58}Co, ^{54}Mn, ^{65}Zn and ^{60}Co.

The relative abundance was determined for each radionuclide identified in a given sample. This was calculated by totalling the representative number of atoms of all radionuclides within a sample, dividing the representative number for each radionuclide by the total and multiplying by 100 to obtain a percentage. Results of relative abundance calculations are presented in Table 3 and graphically represented in Appendix B, pages 1 to 5.

IV. Conclusions

Neutron activated facility parts in the beam-line present a unique problem in determining their absolute radionuclide abundance, since they are of odd shapes and sizes, and compensations for self-absorption must be made. However, relative abundances can be determined. All materials studied here, with the exception of aluminum, contained ^{60}Co as the longest-lived radionuclide ($t_{1/2}$=5.3 years). The presence of radionuclides with such long half-lives determines storage time and disposal procedures.

Future studies on this project will be concerned with developing a method for the determination of absolute radionuclide abundance based upon the geometry of the activated part, the attenuation of the material and build-up effects.

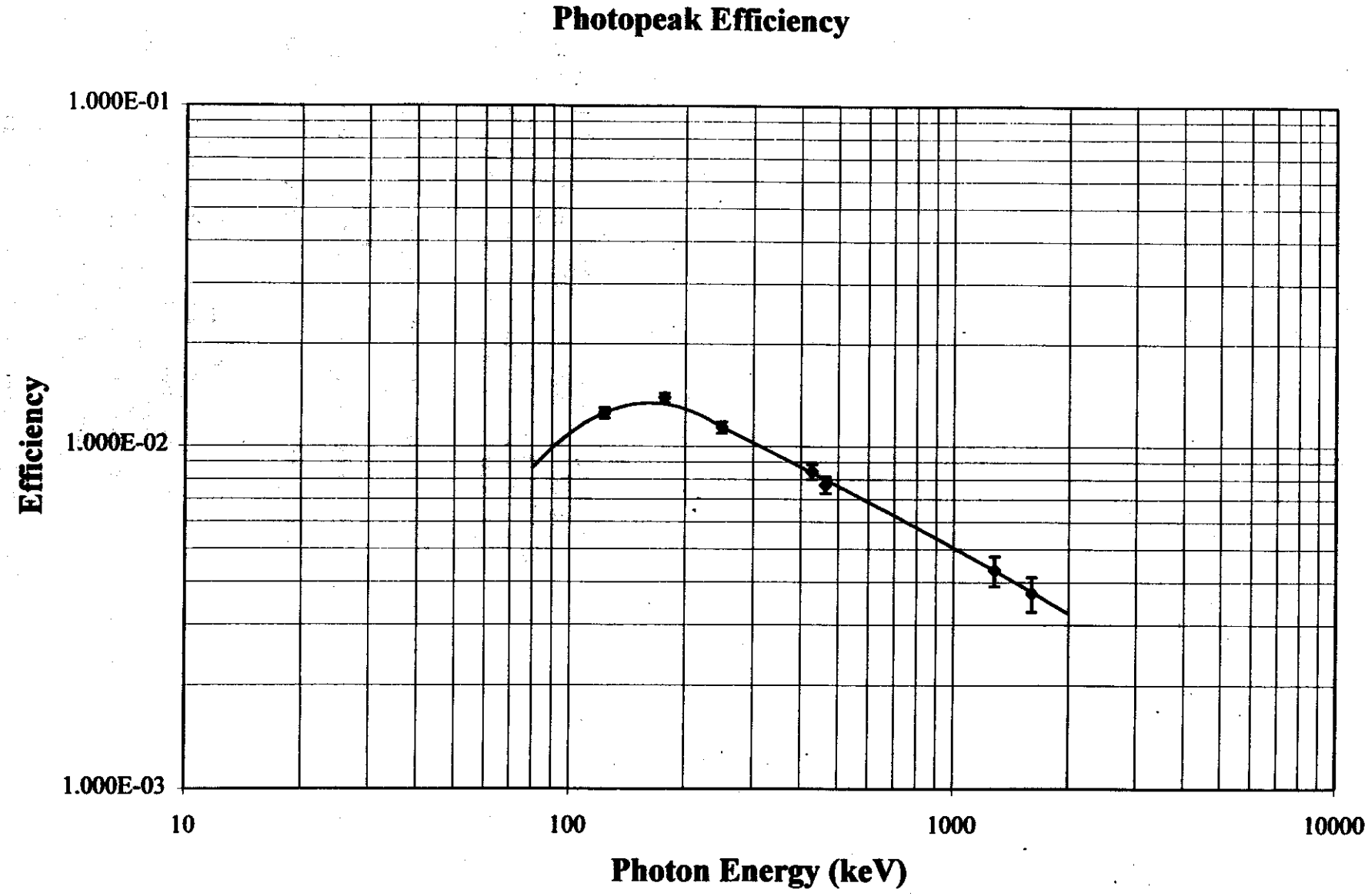
References

[1] Canberra System 100 Software, Canberra Industries, Inc., Meriden, CT.

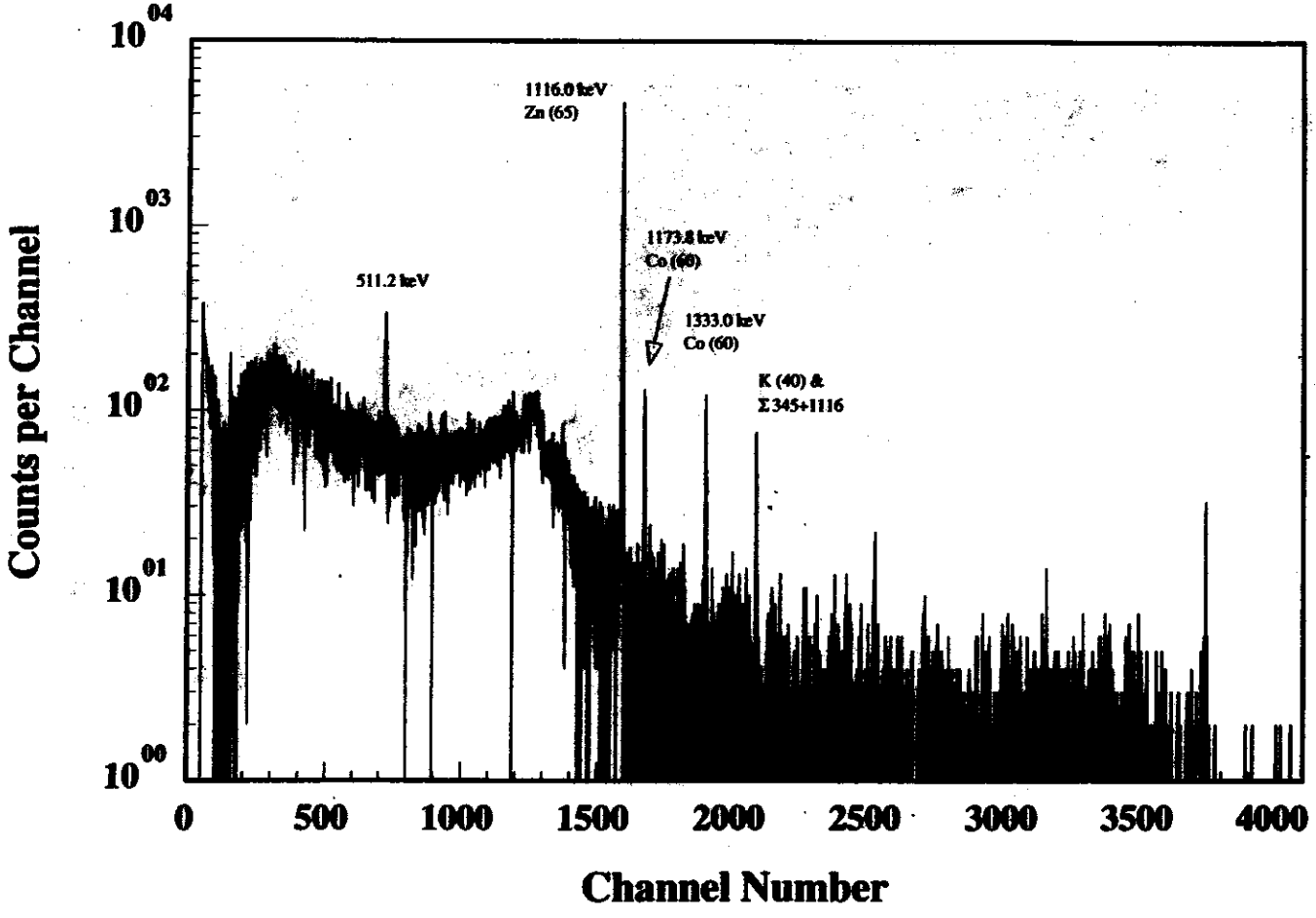
[2] Spanier, L. & Ekstrom, P., Computer Code GDISP, vers 1.2, July 1990. Department of Nuclear Physics, Solvegatan 14, S-223 62 Lund, Sweden. Data are taken from ENSDF (the Evaluated Nuclear Structure Data File) from 27 February 1990. ENSDF is edited and maintained by the National Nuclear Data Center, Brookhaven National Laboratory, on behalf of the International Network for Nuclear Structure Data Evaluation.

[3] Browne, E. & Firestone, R., Table of Radioactive Isotopes @1986. J.Wiley & Sons, Inc., New York.

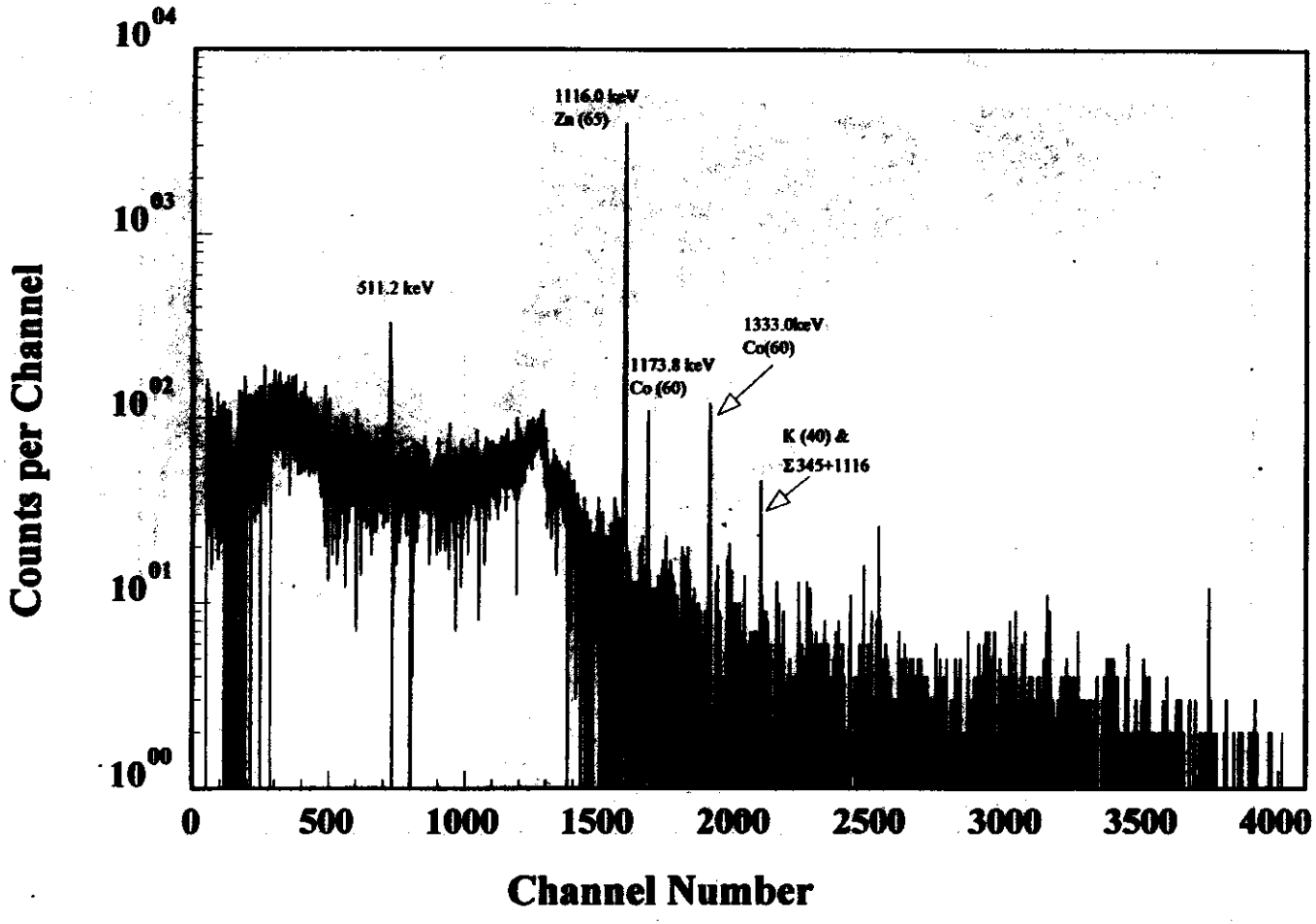
[4] Thomas, R.H., Health Physics Practices at Research Accelerators, Lawrence Berkeley Laboratory Report LBL-4655, Feb. 1976.


Photopeak Efficiency
1.000E-01
1.000E-02
1.000E-03
Efficiency
10
100
1000
10000
Photon Energy (keV)

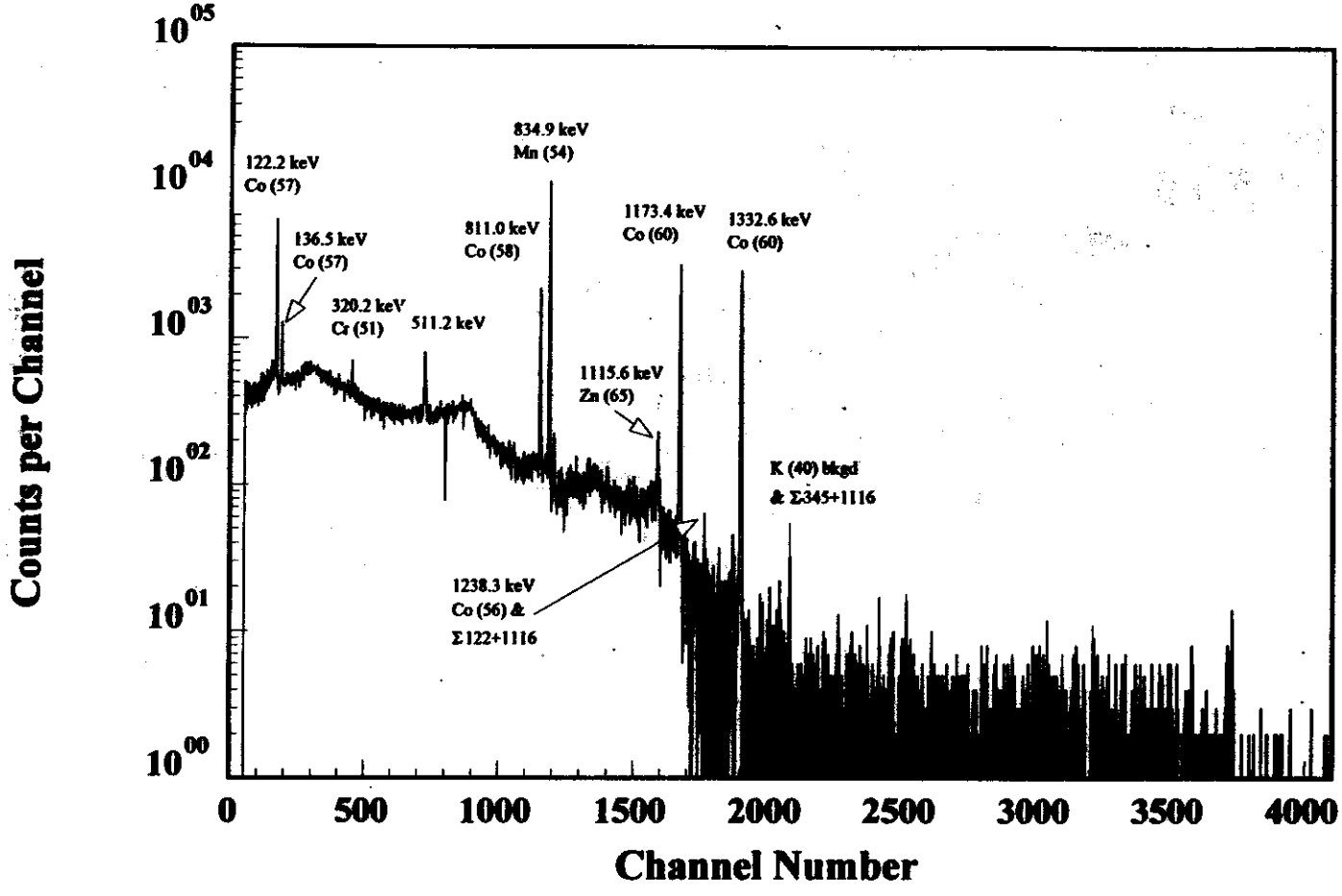
Brass B1


Counts per Channel
Channel Number
10^04
10^03
10^02
10^01
10^00
0
500
1000
1500
2000
2500
3000
3500
4000
1116.0 keV
Zn (65)
1173.8 keV
Co (60)
1333.0 keV
Co (60)
511.2 keV
K (40) &
Σ345+1116

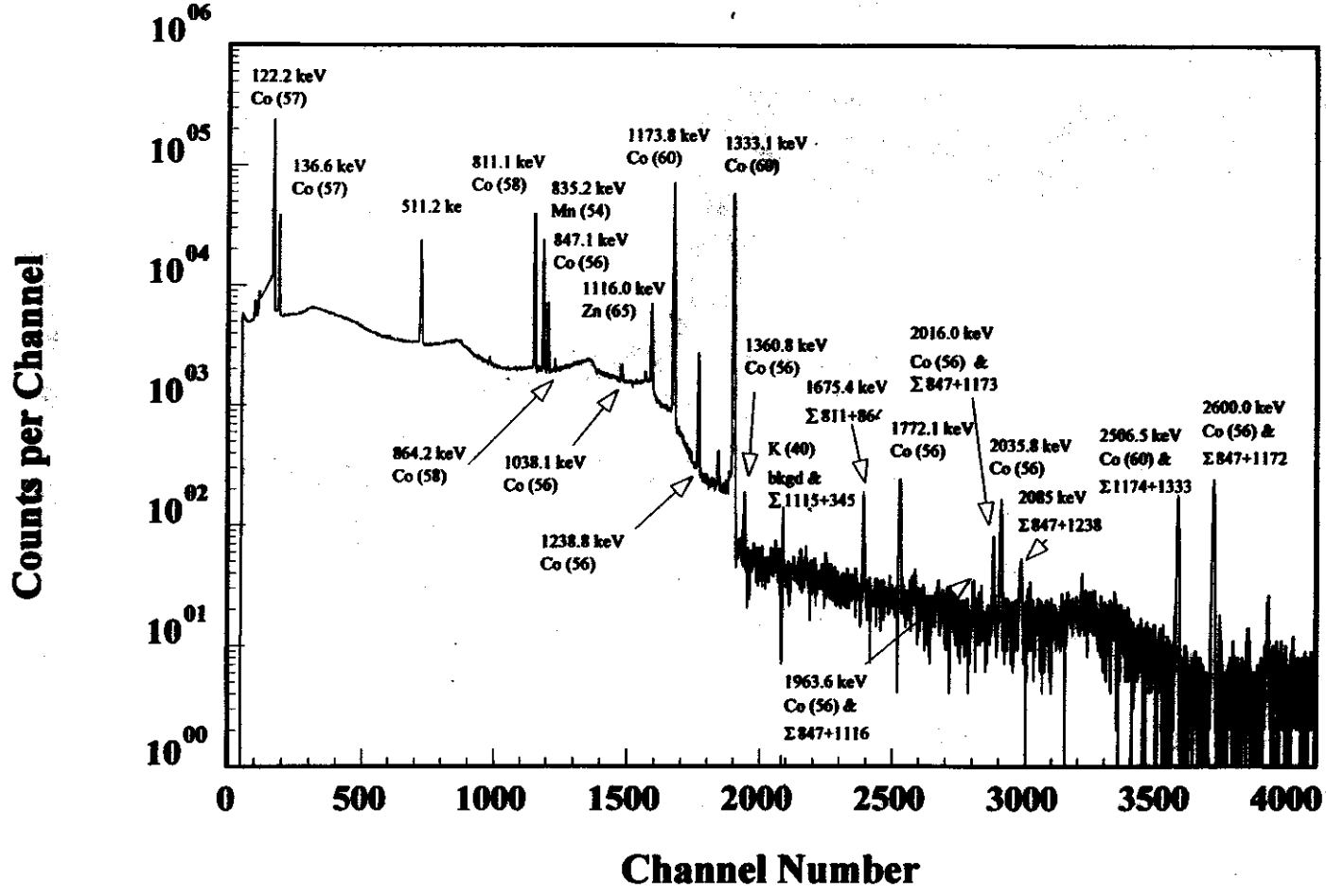
Brass B2


Counts per Channel
Channel Number
10^04
10^03
10^02
10^01
10^00
0
500
1000
1500
2000
2500
3000
3500
4000
511.2 keV
1116.0 keV
Zn (65)
1173.8 keV
Co (60)
1333.0keV
Co(60)
K (40) &
Σ345+1116

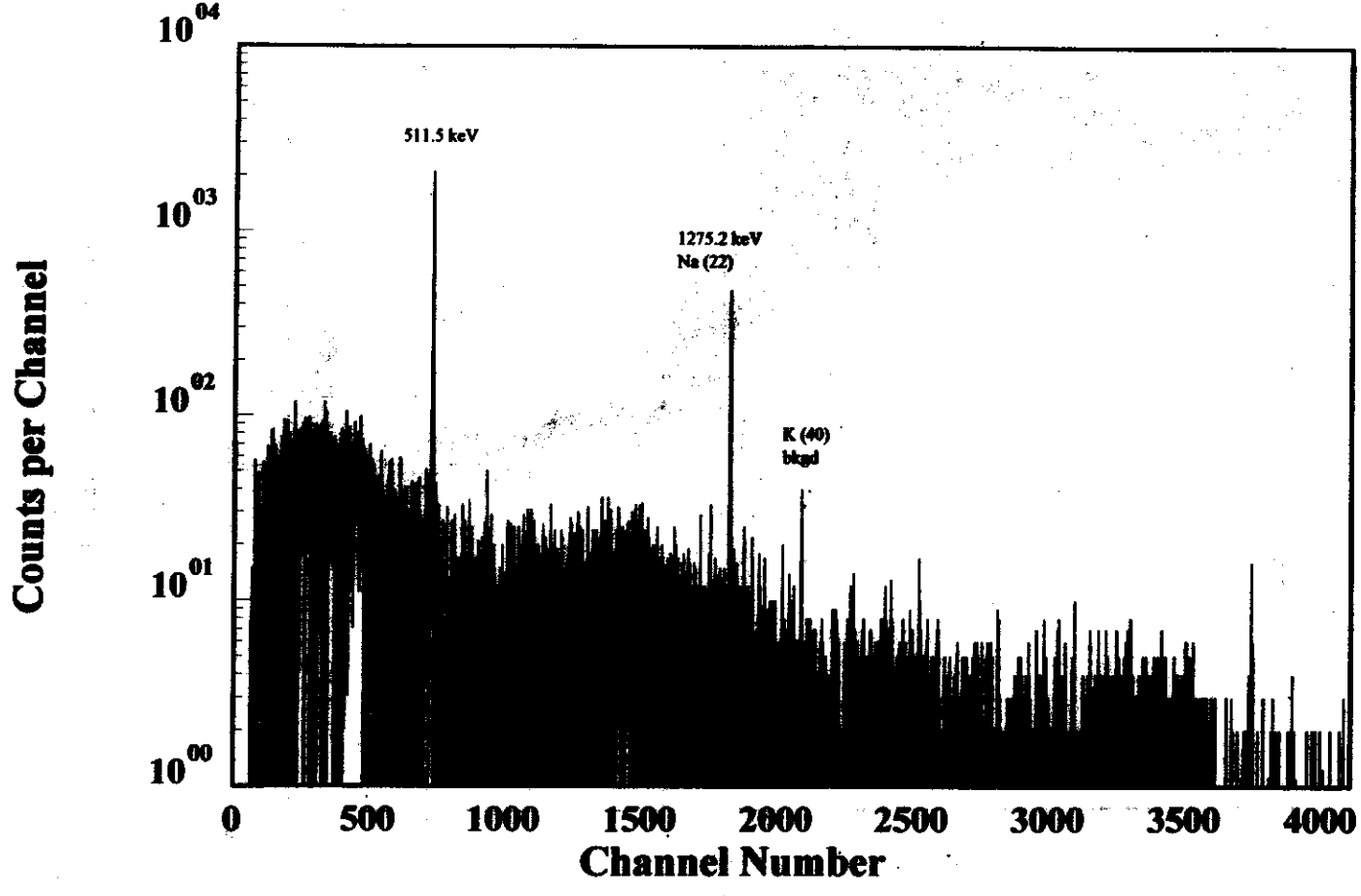
Stainless S2



Copper C2


Counts per Channel
Channel Number
122.2 keV
Co (57)
136.6 keV
Co (57)
511.2 ke
811.1 keV
Co (58)
835.2 keV
Mn (54)
847.1 keV
Co (56)
1116.0 keV
Zn (65)
1173.8 keV
Co (60)
1333.1 keV
Co (60)
1360.8 keV
Co (56)
1675.4 keV
Σ811+86
K (40)
bkgd &
Σ1115+345
1772.1 keV
Co (56)
2016.0 keV
Co (56) &
Σ847+1173
2035.8 keV
Co (56)
2085 keV
Σ847+1238
2506.5 keV
Co (60) &
Σ1174+1333
2600.0 keV
Co (56) &
Σ847+1172
864.2 keV
Co (58)
1038.1 keV
Co (56)
1238.8 keV
Co (56)
1963.6 keV
Co (56) &
Σ847+1116

Aluminum A3


Counts per Channel
10^{04}
10^{03}
10^{02}
10^{01}
10^{00}
511.5 keV
1275.2 keV
Na (22)
K (40)
bkgd
0
500
1000
1500
2000
2500
3000
3500
4000
Channel Number

Junction Box

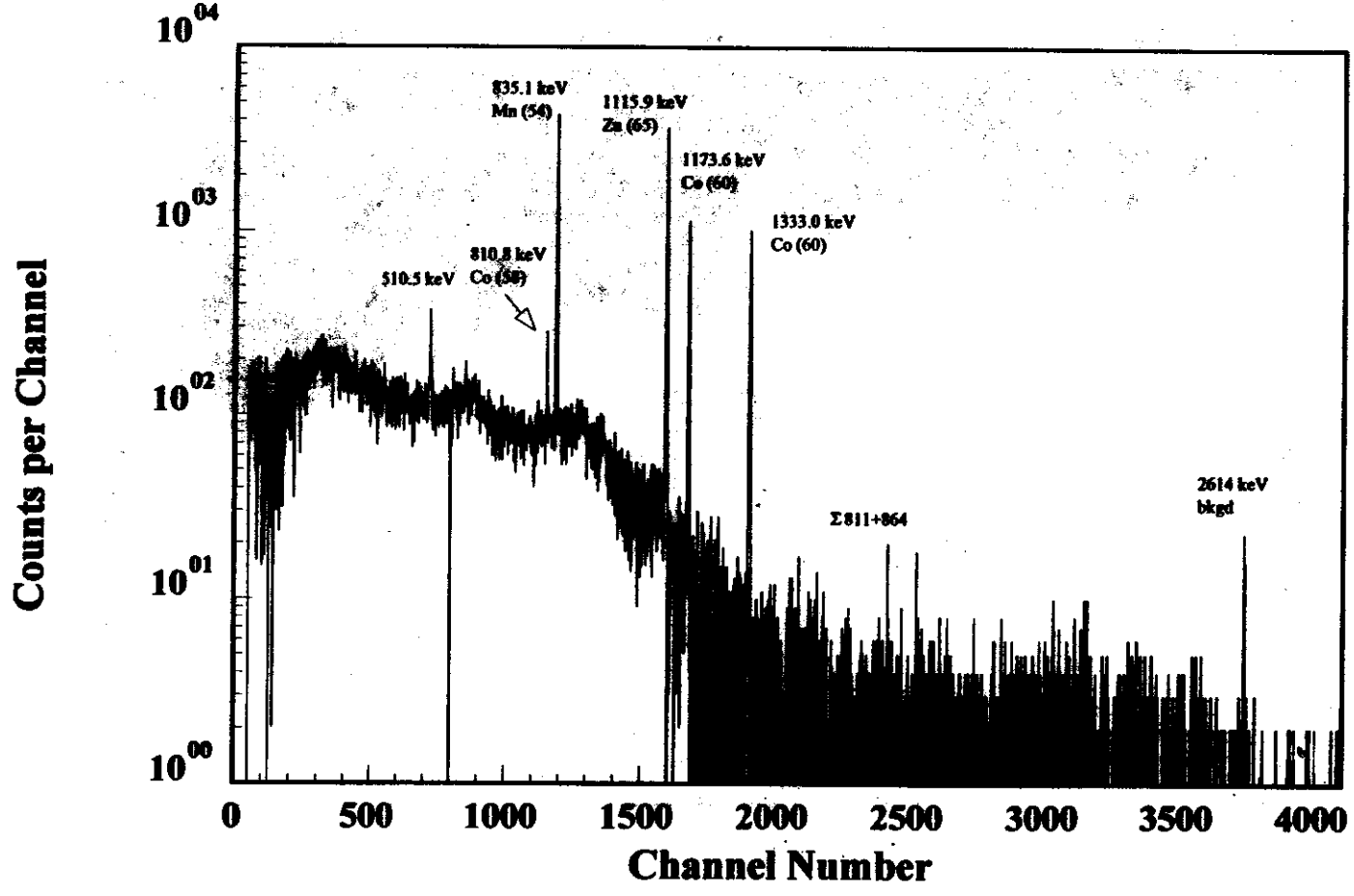


Table 1

Radionuclides identified in neutron activated materials at the NSCL.

Material	*Nuclide*	*Gamma Energy (keV)*	*T1/2[ref. 3]*
Brass	^{65}Zn	1116.0	244 days
	^{60}Co	1173.7	5.27 years
		1333.0	5.27 years
Stainless Steel	^{57}Co	122.1	272 days
		136.5	
	^{51}Cr	320.2	27.7 days
	^{58}Co	811.0	70.9 days
	^{54}Mn	834.9	312 days
	^{65}Zn	1115.6	244 days
	^{60}Co	1173.4	5.27 years
		1332.6	
	^{56}Co	1238.3	77.8 days
Copper	^{57}Co	122.2	272 days
		136.6	
	^{58}Co	811.1	70.9 days
	^{54}Mn	835.2	312 days
	^{56}Co	847.1	77.8 days
		1038.1	
		1238.8	
		1360.8	
		1772.1	
		1963.6	
		2016.0	
		2035.8	
		2600.0	
	^{65}Zn	1116.0	244 days
	^{60}Co	1173.8	5.27 years
		1333.1	5.27 years
Junction Box	^{58}Co	810.8	70.9 days
	^{54}Mn	835.1	312 days
	^{65}Zn	1115.9	244 days
	^{60}Co	1173.6	5.27 years
		1333.0	5.27 years

Table 2

Results of Calculations

Brass B2

Nuclide	*Energy (keV)*	*Representative Number Initial (2/7/94)*
^{65}Zn	1116	5.62(23)E+10
^{60}Co	1173	3.39(22)E+09
^{60}Co	1333	4.69(29)E+09

Brass B1

Nuclide	*Energy (keV)*	*Representative Number Initial (2/7/94)*
^{65}Zn	1116	6.57(26)E+10
^{60}Co	1173	3.94(24)E+09
^{60}Co	1333	4.42(28)E+09

Stainless S2

Nuclide	*Energy (keV)*	*Representative Number Initial (2/7/94)*
^{57}Co	122	7.36(10)E+09
^{57}Co	136	9.15(23)E+09
^{51}Cr	320	3.27(14)E+10
^{58}Co	811	1.06(04)E+10
^{54}Mn	834	7.06(24)E+10
^{65}Zn	1115	1.80(11)E+09
^{60}Co	1173	1.25(05)E+11
^{56}Co	1238	5.52(42)E+08
^{60}Co	1332	1.30(06)E+11

Junction Box

Nuclide	*Energy (keV)*	*Representative Number Initial (2/7/94)*
^{58}Co	811	2.38(19)E+08
^{54}Mn	835	2.42(08)E+10
^{65}Zn	1116	1.01(04)E+11
^{60}Co	1174	4.40(19)E+10
^{60}Co	1333	4.55(21)E+10

Table 2

Results of Calculations

Copper C2

Nuclide	*Energy (keV)*	*Representative Number Initial (2/7/94)*
^{57}Co	122	3.12(04)E+11
^{57}Co	137	3.93(05)E+11
^{58}Co	811	3.15(10)E+11
^{54}Mn	835	1.92(07)E+11
^{56}Co	847	2.47(09)E+10
^{56}Co	1038	2.65(11)E+10
^{65}Zn	1116	7.70(30)E+10
^{60}Co	1174	2.78(11)E+12
^{56}Co	1239	2.64(12)E+10
^{60}Co	1333	2.83(13)E+12
^{56}Co	1361	2.78(16)E+10
^{56}Co	1772	2.86(16)E+10
^{56}Co	1964	4.35(47)E+10
^{56}Co	2016	2.74(21)E+10
^{56}Co	2036	2.72(18)E+10
^{56}Co	2600	2.83(20)E+10

Aluminum A3

Nuclide	*Energy (keV)*	*Representative Number*
^{22}Na	1275	1.10E+10

Table 3

Relative Abundances

Brass B2

Nuclide	*Relative Abundance (%)*
^{60}Co	6.7
^{65}Zn	93.3

Brass B1

Nuclide	*Relative Abundance (%)*
^{60}Co	6.0
^{65}Zn	94.0

Stainless S2

Nuclide	*Relative Abundance (%)*
^{51}Cr	13.0
^{54}Mn	28.0
^{56}Co	0.2
^{57}Co	3.3
^{58}Co	4.2
^{60}Co	50.7
^{65}Zn	0.7

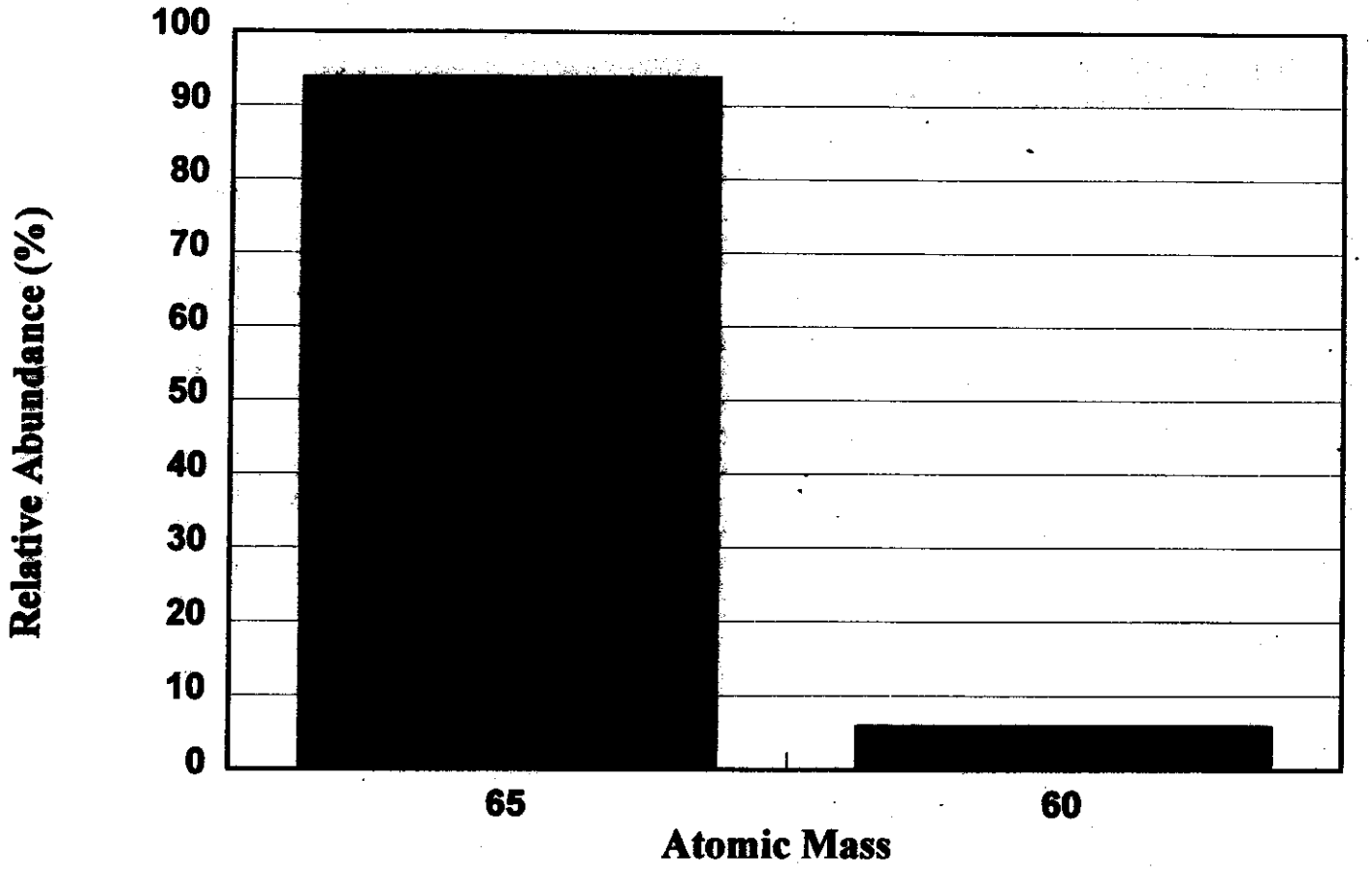
Junction Box

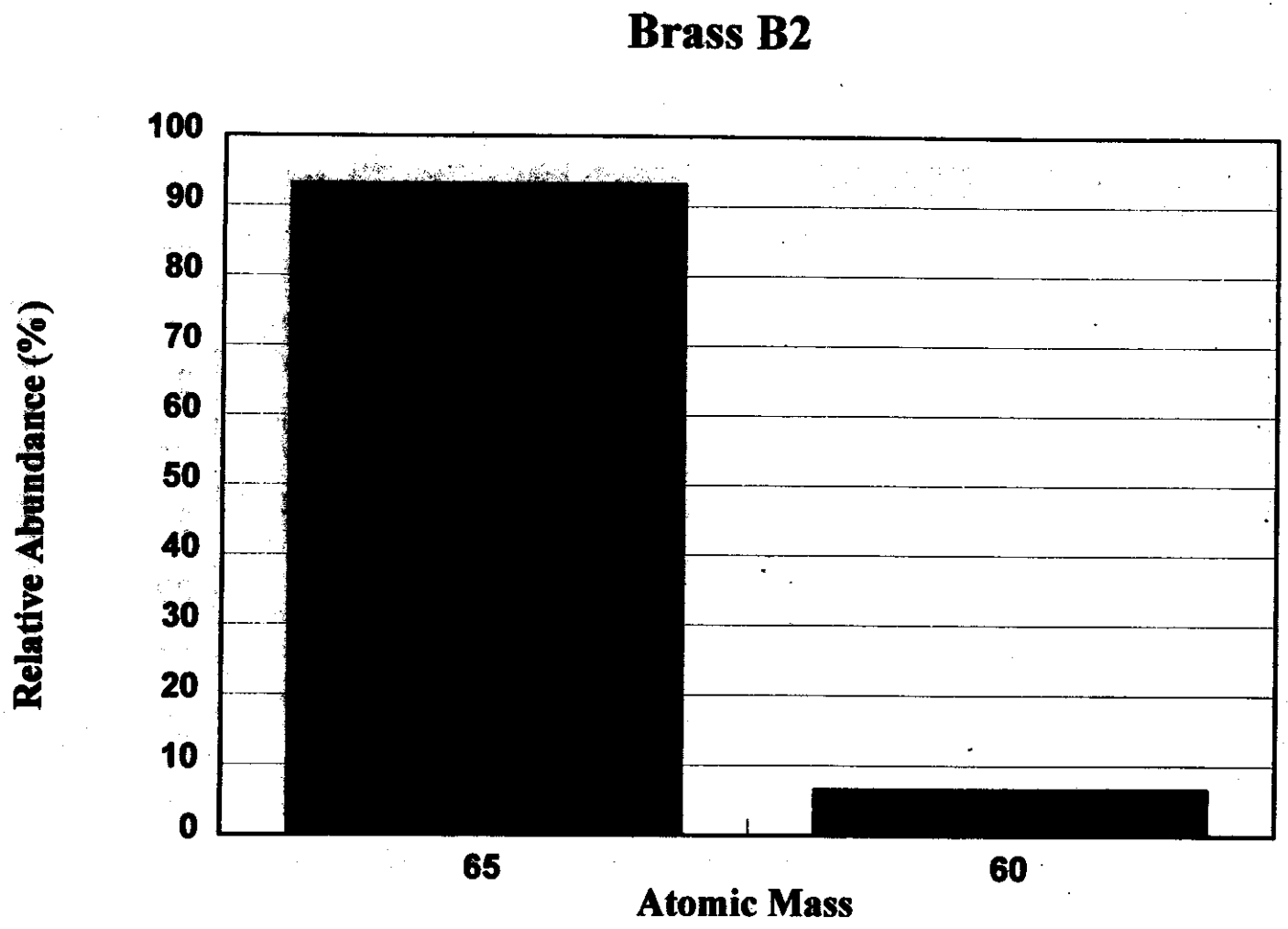
Nuclide	*Relative Abundance (%)*
^{54}Mn	14.2
^{58}Co	0.1
^{60}Co	26.3
^{65}Zn	59.3

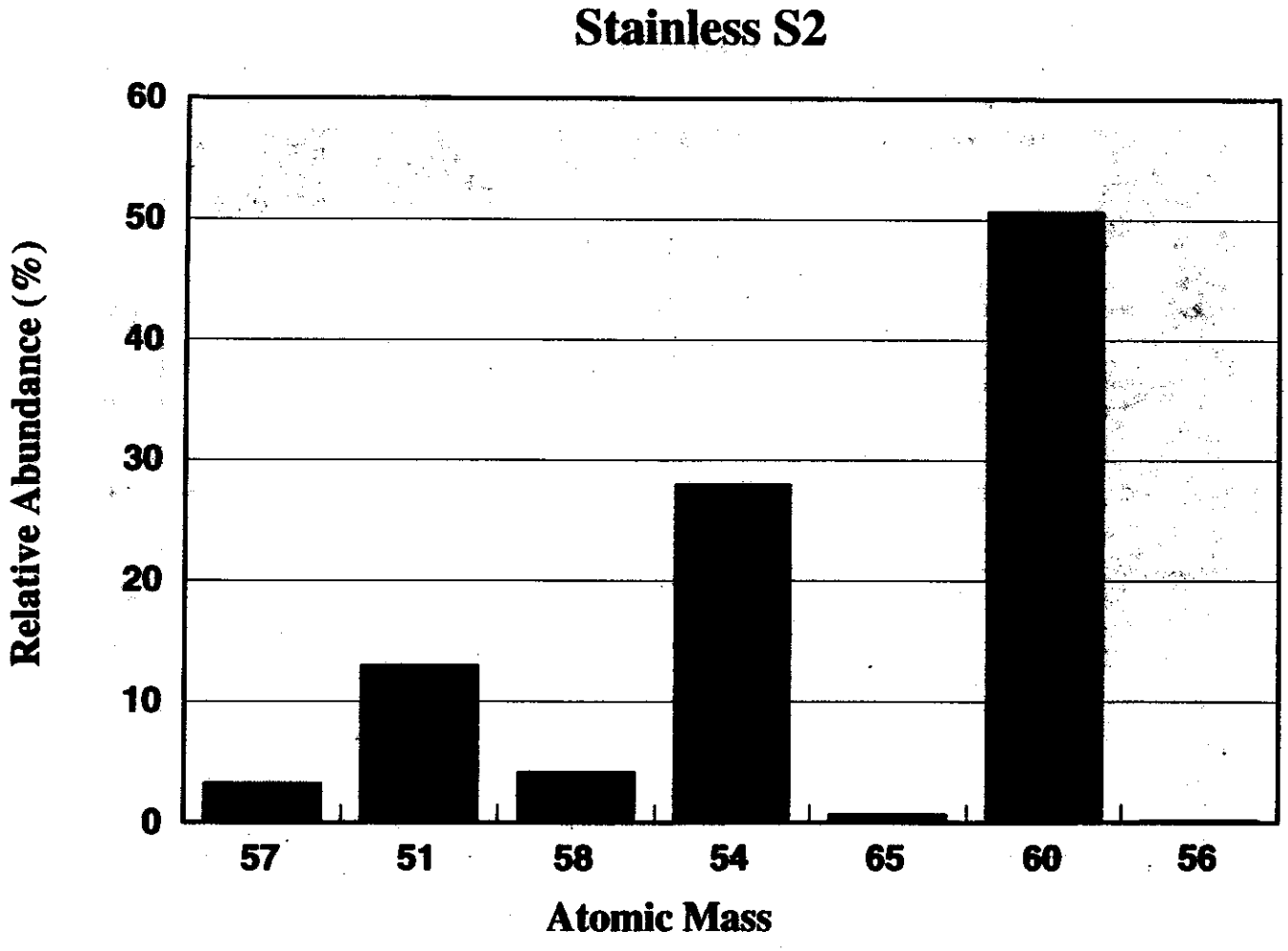
Copper

Nuclide	*Relative Abundance (%)*
^{54}Mn	5.1
^{56}Co	0.7
^{57}Co	9.4
^{58}Co	8.4
^{60}Co	74.5
^{65}Zn	2.1

Brass B1


Relative Abundance (%)
100
90
80
70
60
50
40
30
20
10
0
65
60
Atomic Mass


Brass B2
Relative Abundance (%)
100
90
80
70
60
50
40
30
20
10
0
65
60
Atomic Mass


Stainless S2
Relative Abundance (%)
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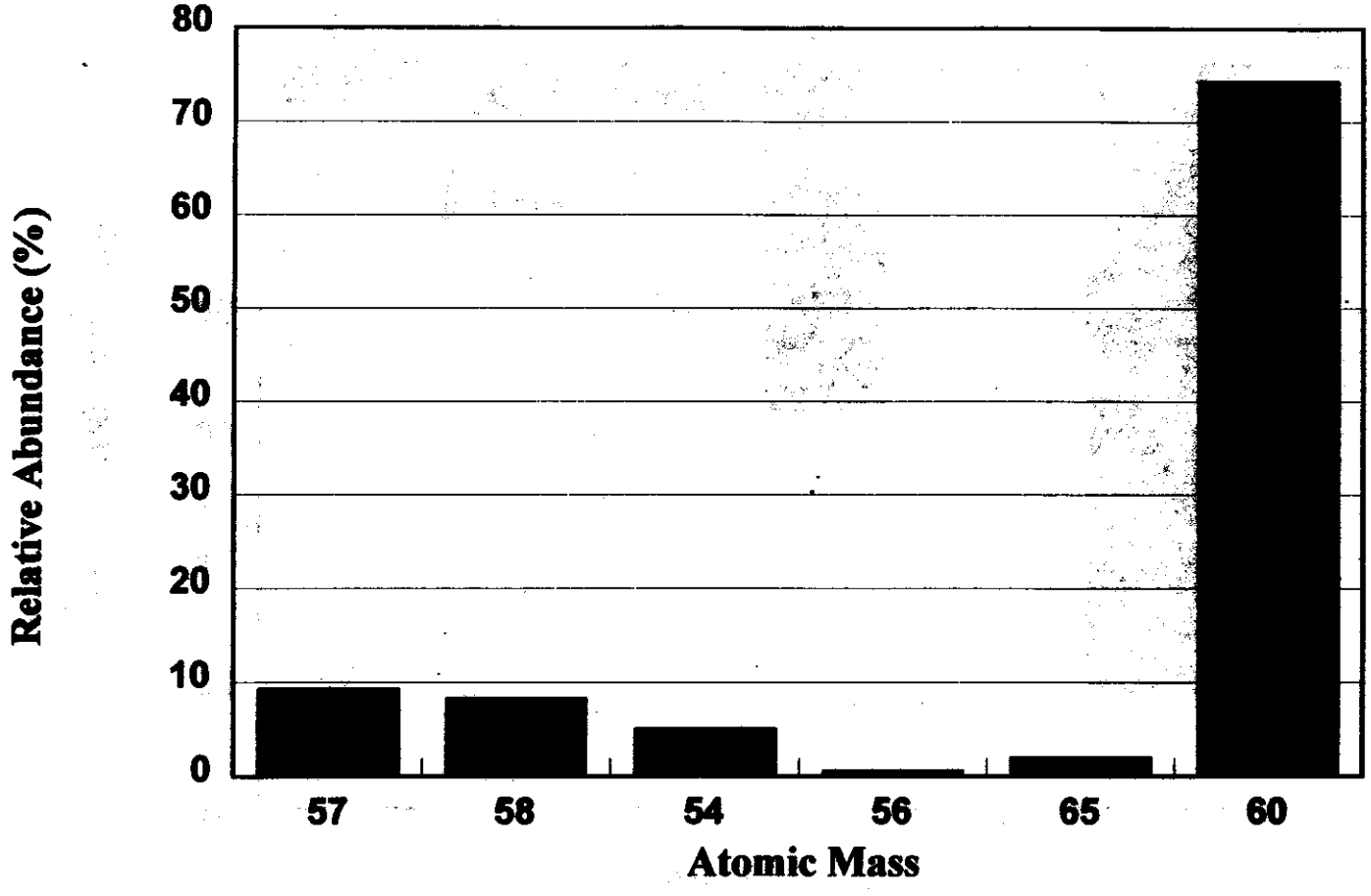
50
40
30
20
10
0
57
51
58
54
65
60
56
Atomic Mass

Copper C2


Relative Abundance (%)
80
70
60
50
40
30
20
10
0
57
58
54
56
65
60
Atomic Mass

Junction Box