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Study of Radionuclides Created by ^{nat}Ni(γ,x*n* y*p*) Reactions for Bremsstrahlung Photons Produced by 140-MeV Electrons

J. K. Dickens

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Engineering Physics and Mathematics Division

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STUDY OF RADIONUCLIDES CREATED BY $^{nat}Ni(\gamma,xn\ yp)$ REACTIONS FOR BREMSSTRAHLUNG PHOTONS PRODUCED BY 140-MeV ELECTRONS

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ABSTRACT

Twelve radionuclides from ^{48}V to ^{57}Ni produced by photon interactions with a sample of elemental Ni have been studied to obtain relative yields of these reaction products. Precision half lives were determined for ^{56}Ni and ^{57}Ni. For ^{56}Ni, $T_{1/2} = 6.08 \pm 0.03$ days, in agreement with previous measurements. For ^{57}Ni, $T_{1/2} = 35.54 \pm 0.05$ hr, in agreement with the 1982 measurement of Grutter but ~0.5 hr shorter than earlier measurements. Relative yields of the 127-keV, 1758-keV, and 1919-keV gamma rays following decay of ^{57}Ni were determined with respect to the 1338-keV gamma ray of that decay and the results agree better with measurements of Grutter and of Gatrousis et al. than they do with the Lederer et al. evaluation.

INTRODUCTION

Although the Oak Ridge Electron Linear Accelerator (ORELA) has been used almost exclusively as a neutron source, there have been several recent scoping experiments designed to utilize the ORELA in other modes. These include (direct) electron and photon interactions with materials wherein the photons are bremsstrahlung created in a Ta target by the 140-MeV electron beam of the ORELA. Since several radionuclides created by Ni + γ reactions have conveniently long half lives, elemental Ni is a useful material to be used to define the photon "beam" which emanates from the small ORELA Ta target. Indeed, Ni may become a useful photon "beam" monitor should such photon-reaction experiments be further pursued at the ORELA. As a first step toward such a possible program, the gamma-ray-assay study described in this report was done on a piece of Ni foil irradiated in a preliminary experiment by ORELA 140-MeV electron-produced bremsstrahlung.

EXPERIMENTAL DETAILS

The 140-MeV electron beam from the ORELA struck a small Ta target of about 5 mm thickness, and the resulting bremsstrahlung struck a Ni foil of ~0.1 mm thickness. The irradiation at ~30 kW of power lasted for about 3 hours. After a period of cooling, the sample was removed from the target room. A small piece of area ~4 cm^2 was cut from the center of the foil and mounted on a piece of fibreboard about 0.4 mm thick.

The sample thus mounted could be accurately positioned (to ± 1 mm) in a horizontal plane vertically above a well-calibrated Ge(Li) detector mounted on a vertical dipstick. This detector is maintained in a relatively low-background environment. Its specifications are (a) 15% efficiency and (b) 2.1-keV resolution for $E_\gamma = 1.33$ MeV.

Gamma-ray spectral measurements commenced about 24 hours following the irradiation when the Ni sample source strength was approximately 4×10^5 Bq. The last and sixty-first measurement was initiated 65 days following the irradiation, at which time the overall source strength was approximately 4×10^3 Bq. Included in these measurements were a series at the same sample-to-detector distance to determine accurate values for the half lives of ^{56}Ni and ^{57}Ni, since for the latter there were experimental disagreements in the literature.[1-3] In particular, the most recent measurements of Grütter[3] differ from earlier measurements by Ebrey and Gray[1] and by Rothman et al.[2]

A comparatively long measurement was taken for the purpose of obtaining a spectrum having "good statistics," so as to clearly define the dominant peaks and also to search for weakly excited transitions. The raw spectral data obtained from this measurement are presented in Figs. 1 to 4. This measurement commenced 4.1 days following the end of the irradiation, and it required 60000 sec of accumulation time. The sample-to-detector distance was 10 cm, and the dispersion was ~0.82 keV per channel. A perusal of the peak in Fig. 2 corresponding to detection of $E_\gamma = 1377.6$ keV indicates a full-width-at-half-maximum resolution (FWHM) of about 2.5 keV for the "working" resolution for long counting periods. Nearly all of the observed peaks in this spectrum were identified as being due to detection of gamma radiation emanating from radioisotopes which could have been produced by Ni + γ reactions and which have half lives long enough for observation after four days of cooling.

The spectral data were first reduced using an up-to-date version of the documented[4] code TPASS. This analysis was followed by manual data reduction for certain of the data, especially those used to determine half lives plus small peaks which were likely to correspond to interesting transitions but which the computer program did not locate.

Following peak identifications and then efficiency-corrected yield determinations, the TPASS program utilized a table look-up procedure to assign gamma rays to appropriate decay transitions. Finally, the yields of the transitions which were observed were then combined to deduce the overall yield of each responsible radioisotope. Corrections were made for dead times in the analyzer and for random summing effects which are count rate dependent. Real summing effects were also observed and were a part of the data reduction. For example, the small peak labelled "(1504.7)" in Fig. 2 is one such peak, being the sum peak corresponding to the 127-keV transition in coincidence with the 1378-keV transition. For this particular spectrum, the ratio of the yield of the (127+1378)-keV sum peak to the yield of the 1378-keV peak is 0.00087, which, in turn, requires a fractional increase of ~0.002 to the 1378-keV peak. Overall yields for twelve isotopes are collected in Table 1.

For the determinations of half lives for ^{56}Ni and ^{57}Ni, spectra were obtained starting 5 days following the end of the irradiation and continuing for 60 days, or about ten times the ^{56}Ni half life. The counting times were increased with increasing cooling time to provide adequate peak yields for analysis. Using documented[5] mathematical analysis methods, half-life determinations were done individually for gamma rays representing the dominant transitions following decay of ^{56}Ni and ^{57}Ni. For ^{56}Ni, study of the 158-keV peak yielded a half life of 6.08 ± 0.03 d which may be compared with 6.10 ± 0.02 d in the evaluation of Lederer et al.[6] For ^{57}Ni, the data and analysis of the 1378-keV peak are shown in Fig. 5 and lead to a half life of 35.54 ± 0.05 hr. A similar analysis was performed for the 1919-keV data of ^{57}Ni, resulting in a half life of 35.55 ± 0.10 hr. Comparison of the present measurement with earlier measurements[1-3] is given in Table 2. The Lederer et al.[6] evaluation recommends a value of 36.1 hr.

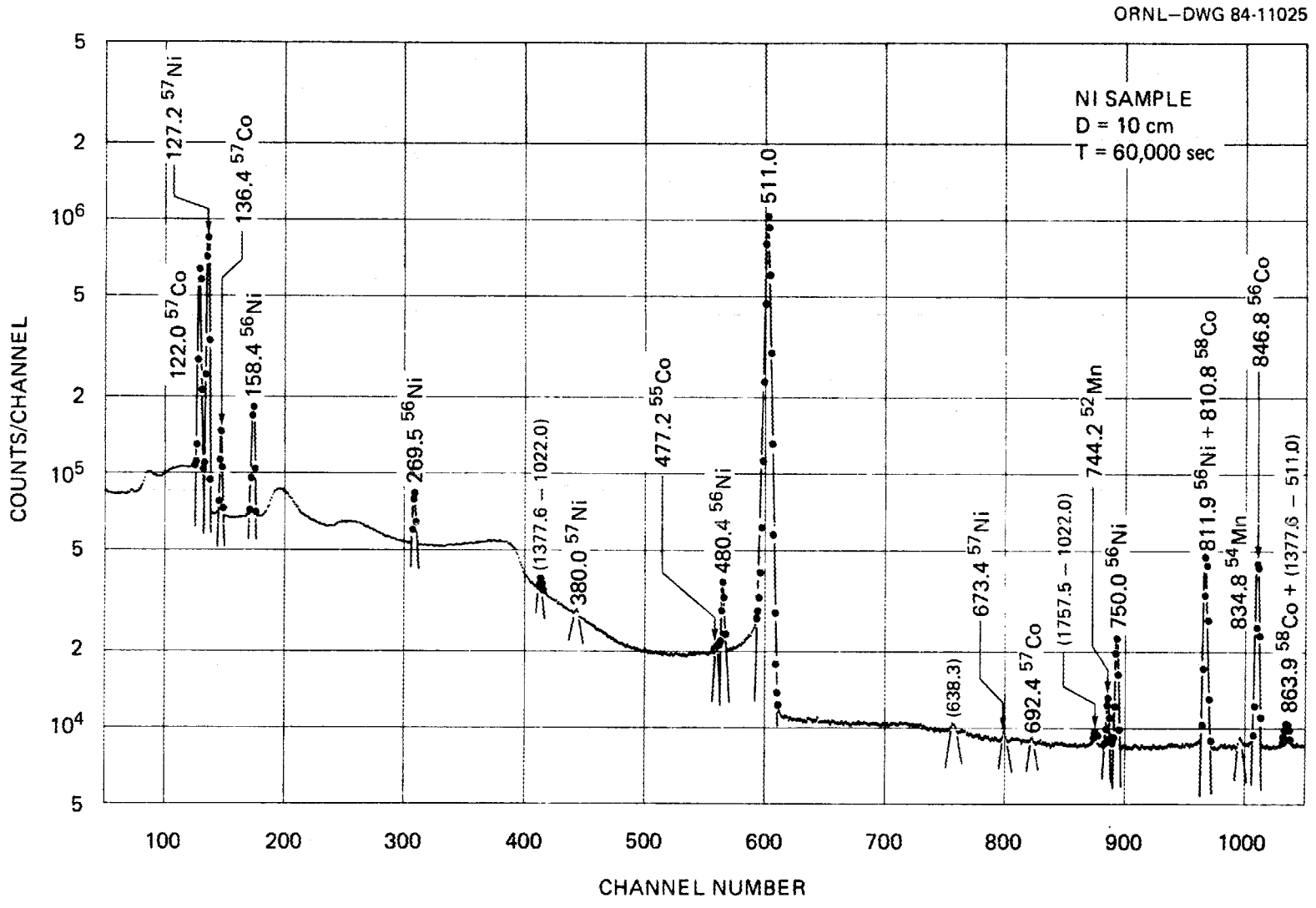

Fig. 1. Low-energy segment of the spectrum obtained commencing about four days after the end of a 3-hr irradiation of an elemental Ni foil by bremsstrahlung from the ORELA. Peaks are labelled with detected gamma-ray energy, in keV, and the assigned radioisotope. (Some of the individual points have been enhanced so that they can be observed.) The weak peak at $E_\gamma = 638$ keV is likely the sum peak for ^{57}Ni decay of $E_\gamma = 127$ keV combined with $E_\gamma = 511$ keV.

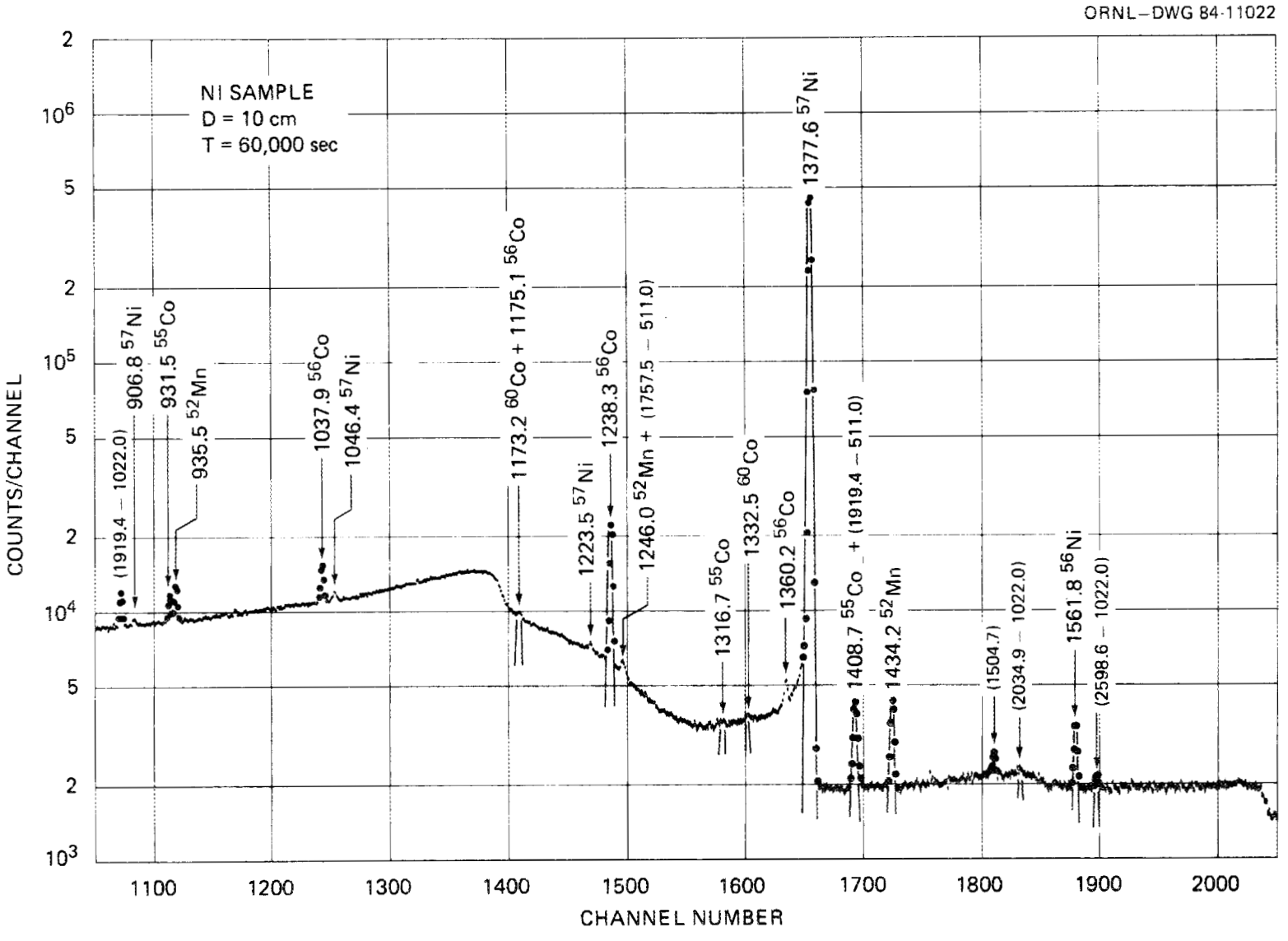

Fig. 2. Next higher energy segment of the spectrum. Escape peaks are labelled with the responsible gamma-ray energy, in keV, minus 511 keV for the single-escape mode and minus 1022 keV for the double-escape mode. The weak peak at $E_\gamma = 1505$ keV is likely the sum peak for ^{57}Ni decay of $E_\gamma = 127$ keV combined with $E_\gamma = 1378$ keV.

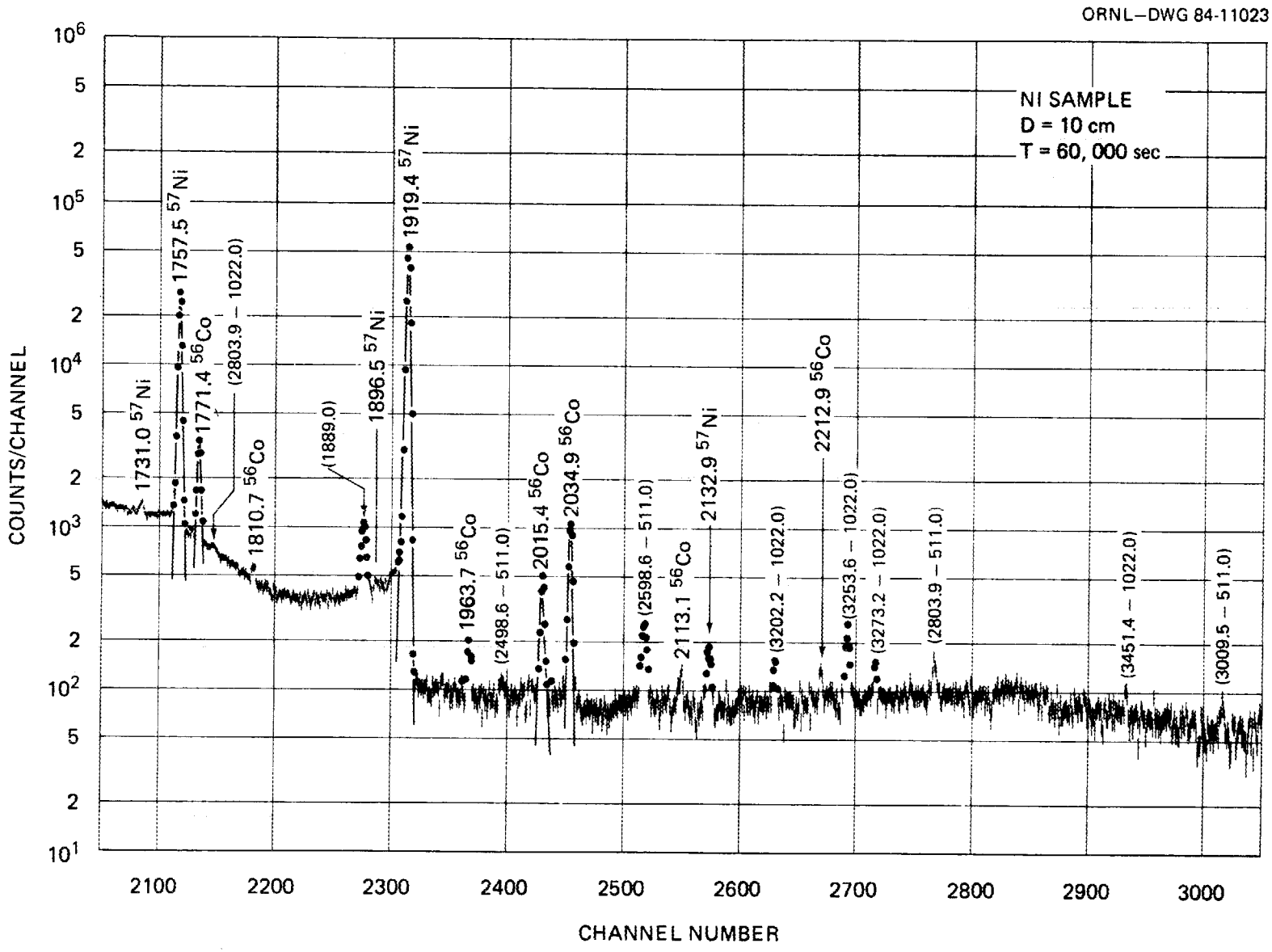

Fig. 3. Next larger-energy segment of the spectrum. The weak peak at $E_\gamma = 1889$ keV is likely the sum peak for ^{57}Ni decay of $E_\gamma = 511$ keV combined with $E_\gamma = 1378$ keV.

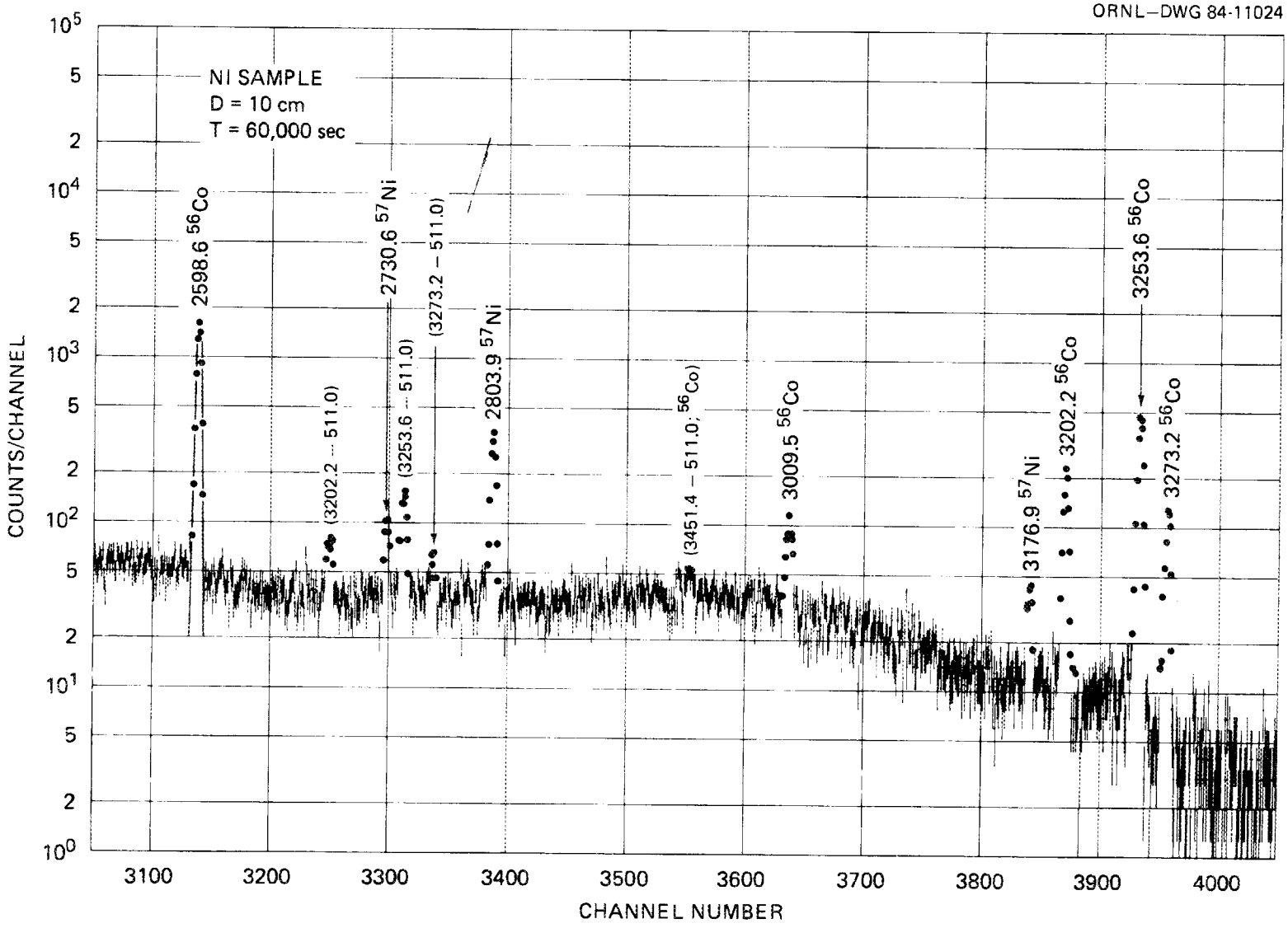

Fig. 4. Highest-energy portion of the spectrum.

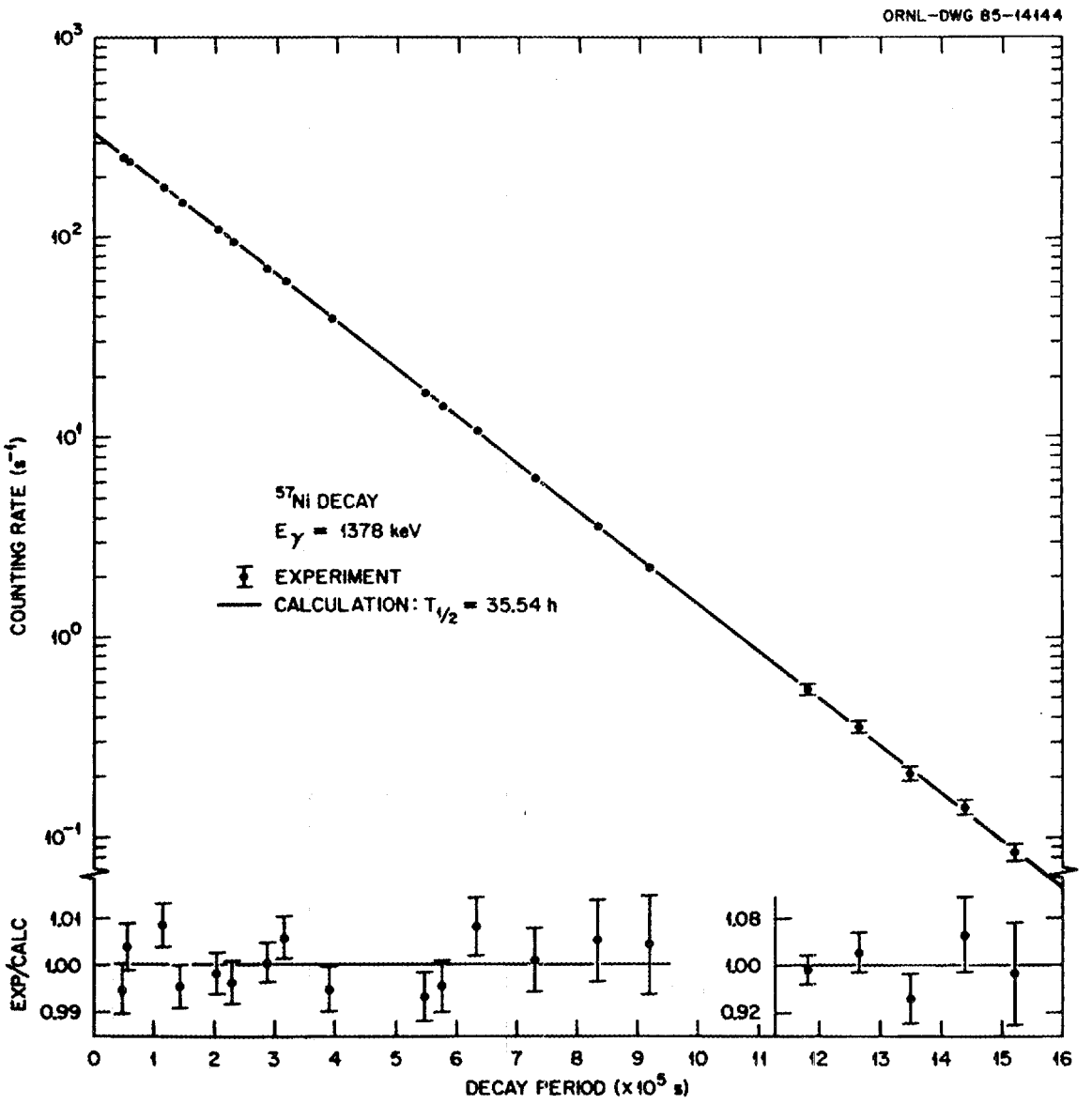

Fig. 5. Analysis of the decay of ^{57}Ni. The upper portion of the figure exhibits the experimental counting rate data for the 1378-keV gamma ray following the decay of ^{57}Ni as a function of decay time. The computed decay rate for a 35.54-hr half life is shown as the solid line, and the experimental data are shown as solid points and uncertainties. The lower portion of the figure exhibits the ratio of the experimental data (and uncertainties) to the calculated data using an expanded scale.

Table 1. Yields of isotopes created by Ni + γ reactions. The yields are in number of atoms of the isotope plus isotonic precursors as of the end of the irradiation.

Isotope	Yield
^{48}V	$(1.5 \pm 0.3) \times 10^{7}$
^{51}Cr	$(3.4 \pm 0.7) \times 10^{8}$
^{52}Mn	$(2.05 \pm 0.10) \times 10^{8}$
^{54}Mn	$(9.85 \pm 0.30) \times 10^{8}$
^{59}Fe	$(1.27 \pm 0.32) \times 10^{7}$
^{55}Co	$(9.0 \pm 0.3) \times 10^{8}$
^{56}Co	$(1.57 \pm 0.05) \times 10^{10}$
^{57}Co	$(1.90 \pm 0.06) \times 10^{11}$
^{58}Co	$(8.5 \pm 0.3) \times 10^{9}$
^{60}Co	$(1.25 \pm 0.13) \times 10^{9}$
^{56}Ni	$(1.24 \pm 0.05) \times 10^{9}$
^{57}Ni	$(6.02 \pm 0.15) \times 10^{10}$

Table 2. $T_{1/2}$ (hr) for ^{57}Ni from several experiments.

Year	$T_{1/2}$ (hr)	Reference
1965	35.99 ± 0.12	Ebrey and Gray (ref. 1)
1974	36.16 ± 0.11	Rothman et al. (ref. 2)
1982	35.65 ± 0.05	Grutter (ref. 3)
1985	35.54 ± 0.05	Present results

The present data were also studied for gamma-ray branching ratio information for the decay of ^{57}Ni. Precision results were obtained for the four largest yields. The present results are compared with prior measurements[3,7-8] in Table 3; the Lederer et al.[6] evaluation recommends the Heath[7] branching-ratio measurements.

Table 3. Relative branching ratios (%) for decay of ^{57}Ni

E_γ (keV) =	127	1338	1758	1919	Reference
	16.6 ± 1.0	100	9.1 ± 0.8	18.9 ± 1.2	Heath (ref. 7)
	20.0 ± 0.6	100	7.70 ± 0.24	17.0 ± 0.5	Gatrousis et al. (ref. 8)
	20.3 ± 0.5	100	7.63 ± 0.20	17.0 ± 0.45	Grutter (ref. 3)
	20.2 ± 0.5	100	7.87 ± 0.16	17.0 ± 0.4	Present results

DISCUSSION OF RESULTS

As shown in Tables 2 and 3, the results for ^{57}Ni from the present study are in good agreement with the measurements of Grutter[3] and of Gatrousis et al.,[8] but not in good agreement with the evaluation of Lederer et al.,[6] which was based on earlier data.[1,2,7]

The results given in Table 1 are rather more interesting, albeit less definitive. A portion of the periodic table is shown in Fig. 6, and the relative positions of several measured isotopes may be observed in this figure. In particular, if the reactions are considered as ^{A}Ni(γ,xn yp) reactions (where A = 58, 60, 61, 62, or 64), one obtains some estimations of yields as functions of x and y. From the present results, the most probable reaction has $x = 0$ and $y = 1$ (i.e., ^{58}Ni(γ,p)^{57}Co), with the next most probable having $x = 1$ and $y = 0$. Two-nucleon reactions are the next most probable. However, barring a contaminant in the Ni sample (which was not tested for impurities), the experiment does yield information on at least one ten-nucleon reaction. Of course, some care must be observed, since, for example, ^{57}Co could also be produced by the ^{60}Ni(γ,$2np$) reaction. The cross section for that reaction may well be larger than the ^{58}Ni(γ,$2np$)^{55}Co reaction because of the two extra neutrons in ^{60}Ni. However, the yield of ^{55}Co is a factor of ~70 smaller than the yield of ^{57}Co, and one would not *a priori* expect more than perhaps a factor of 5 for the ratio of the ^{60}Ni(γ,$2np$) cross section to the ^{58}Ni(γ,$2np$) cross section for the photon energies of this experiment.

The overall results, therefore, suggest a decreasing yield as a function of increasing $x + y$. Such a conclusion is not really surprising for this particular experiment. The incident photon spectrum is characterized by a continuously decreasing yield as a function of increasing photon energy. Hence, in the total incident photon fluence, there will be substantially more incident photons having energies sufficient for only *one* photo-nucleon reaction than incident photons having energies sufficient for two photo-nucleon reactions, and similarly for incident photons having energies sufficient for two photo-nucleon reactions but not three photo-nucleon reactions, etc.

CONCLUSIONS

The present study of radionuclides created by photon interactions with ^{nat}Ni was a scoping experiment having the first goal of observing what was there to be observed. The data yielded definitive information concerning the decay of ^{57}Ni which, although not "new" information, does support more recently published measurements which were in disagreement with earlier measurements.

PARTIAL CHART OF ISOTOPES

Ni 53	Ni 54	Ni 55 <5s	Ni 56 6.1d E_γ=158	Ni 57 35.6h E_γ=1378	Ni 58 68.3	Ni 59 8×10^4y no γ	Ni 60 26.1	Ni 61 1.1	Ni 62 3.6	Ni 63 100y no γ	Ni 64 0.9
Co 52	Co 53	Co 54 <2m	Co 55 17.5h E_γ=931	Co 56 78.5d E_γ=847	Co 57 271d E_γ=122	Co 58 70.8d E_γ=811	Co 59 100	Co 60 5.27y E_γ=1173	Co 61 1.65h E_γ=67.4	Co 62 <15m	Co 63 <1m
Fe 51	Fe 52 8.3h E_γ=168.7	Fe 53 8.5m	Fe 54 5.8	Fe 55 2.7y no γ	Fe 56 91.8	Fe 57 2.1	Fe 58 0.3	Fe 59 44.6d E_γ=1099	Fe 60 $\sim10^5$y E_γ=58.6	Fe 61 6.0m	
Mn 50 <2m	Mn 51 46m	Mn 52 5.6d E_γ=1434	Mn 53 4×10^6y no γ	Mn 54 312d E_γ=834	Mn 55 100	Mn 56 2.58h E_γ=847	Mn 57 1.6m	Mn 58 <2m			
Cr 49 42m	Cr 50 4.35	Cr 51 27.7d E_γ=320	Cr 52 83.79	Cr 53 9.50	Cr 54 2.36	Cr 55 3.6m	Cr 56 6m				
V 48 16d E_γ=984	V 49 331d no γ	V 50 0.25	V 51 99.75	V 52 3.8m							

Fig. 6. Portion of the periodic table including radioisotopes observed in this experiment. In each box the chemical symbol and isotopic mass are shown at the top. If the isotope is stable, the second line gives the percentage of the natural element for that isotope. If the isotope is radioactive, the half life is shown on the third line, and the energy (in keV) of the principal decay gamma ray is given on the fourth line.

Data were observed for every radioisotope[6] exhibited in Fig. 6 having (1) a half life long enough to be observed some 24 hours after the end of the irradiation, (2) a half life short enough to provide measurable activity, (3) a gamma-ray decay transition having energy $E_\gamma > 100$ keV, and (4) a plausible production reaction for photon interaction with at least one of the five stable isotopes of Ni. This lack of selectivity, while not unexpected, does indicate that the bremsstrahlung beam will be a difficult beam to monitor. However, the lack of selectivity does suggest that the bremsstrahlung beam could be used to produce radioisotopes several nucleons from the line of stability for basic or applied studies. Indeed, a number of follow-on experiments seem to be natural continuations of the present study. Several will be given consideration for future work.

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