Neutron Capture γ-Ray Spectra from the Elements $Z=17$–30 and $Z=45$–57

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A NaI(Tl) scintillation counter and a 5819 photomultiplier have been used in conjunction with a very accurate and flexible twenty-channel pulse-height discriminator to study thermal neutron capture gamma-ray spectra. The energy range 20 kev–10 Mev was studied for the elements of atomic numbers 17–30 and 45–57 excluding the noble gases. In the case of chlorine, four lines are found below 2.1 Mev. These probably correspond to the first four excited states of Cl^{36}. A neutron binding energy of 8.57 ± 0.03 Mev is found for Cl^{36} by combining these lines with pair spectrometer data on the high energy lines. The method yields information on only the most prominent transitions in the capture gamma-decay schemes. No systematic distribution of these prominent lines is evident from the limited number of lines that can be resolved by this technique.

I. INTRODUCTION

RECENTLY a small measure of progress has been made in the study of the instantaneous gamma-rays that are emitted following thermal neutron capture. One new method that has been used is the nuclear emulsion technique.[1] The emulsions are loaded with deuterium in some form and capture gamma-rays are detected by observing photoproton tracks in the emulsion. This method is useful only above 3 Mev and has very poor energy resolution.

A much more fruitful technique has been the use of a pair spectrometer[2] by Kinsey and his colleagues at Chalk River. Although the method has a low energy cutoff between 2.5 and 3.5 Mev, it has about a 1.5–2 percent energy resolution in the 6–10 Mev range. Many interesting and surprising results (Pb^{208}) have been found by this technique.

The development of scintillation counters with high gamma-ray detection efficiencies has made it possible to study capture gamma-ray spectra[3,4] below the low energy cutoff of the methods mentioned above. In fact, we have now made measurements down to 20 kev by this technique.

We have begun a systematic study of thermal neutron capture gamma-ray spectra by using a scintillation counter gamma-ray spectrometer. The program is intended to study groups of elements in the low, intermediate and high atomic number regions of the periodic table. Whenever possible, a series of elements of adjacent atomic numbers is studied. It is hoped that in this way some systematic features of the spectra will be made evident. The present paper deals with the two groups of elements $Z=17$–30 and $Z=45$–57 excluding the noble gases, argon and xenon. The results[3] on chlorine, manganese, and iron which were previously reported have been extended to lower energies with higher resolution and greater accuracy.

II. APPARATUS

The detecting apparatus consists of a probe made of a NaI(Tl) crystal, a 5819 photomultiplier, and a 2-stage preamplifier. The crystal is $\frac{3}{8}$-inch high and $1\frac{1}{2}$ inches in diameter. It is packaged according to the method of Swank and Moenich.[5] This packaging technique is very successful and the crystals seem to last indefinitely. A $\frac{1}{2}$-inch Lucite light pipe is used between the crystal package and the photomultiplier.

The pulses from the probe are fed into a window amplifier and then in turn into a twenty-channel pulse-height discriminator and a twenty-channel recording console. Each recording channel has a decade scaler which can be cascaded into the adjacent channel. Channel No. 20 acts as an integrating channel and records the integrated counting rate of all gamma-rays above the region of the spectrum that is being searched.

The window amplifier chassis contains an accurate auxiliary pulse generator whose output may be taken in one or two volt steps for setting the twenty channels to be as nearly equal to each other as possible. The window amplifier has a gain of 5 when set for 1-volt intervals and a gain of 2.5 when set for 2-volt intervals; i.e., its output pulses are always 5-volt pulses.

The window amplier contains appropriate attenuators so that any energy range may be studied. For example, there are five gain settings, A–E. The attenuation of pulse height that is controlled by each gain setting allows the use of the following convenient scheme.

By choosing the correct photomultiplier voltage, the gain settings A–E can be made to correspond to 5, 10, 12.5, 25 and 50-kev/volt pulse height. By raising the voltage properly, gain A can be used to give 2.5 kev/volt and by lowering the voltage, gain E can be set to give 100 kev/volt. On all gain settings, a voltage range of 0–100-volts pulse height is available and since the 20 channels can be made to correspond to 1 or 2 volt intervals, a 20 or 40-volt interval can be studied. The

[1] B. Hamermesh, Phys. Rev. **76**, 182 (1949); **80**, 415 (1950); **81**, 487 (1951).
[2] Kinsey, Bartholomew, and Walker, Phys. Rev. **77**, 723 (1950); **78**, 77 (1950); **79**, 218 (1950); **81**, 150 (1951); **82**, 380 (1951); **83**, 519 (1951).
[3] B. Hamermesh and V. Hummel, Phys. Rev. **83**, 663 (1951).
[4] Thornton, der Mateosian, Motz, and Goldhaber, Phys. Rev. **86**, 604 (1952).
[5] R. Swank and J. Moenich, Rev. Sci. Instr. **23**, 502 (1952).

20 or 40-volt interval may be located anywhere along the 0–100-volt interval. In this manner a very flexible searching scheme is available.

The spectrometer is calibrated by use of gamma-ray sources having simple decay schemes. The instrument is usually calibrated with gamma-rays from Au^{198} (0.411 Mev), Cs^{137} (0.669 Mev) and Na^{24} (1.38 and 2.76 Mev). The first two sources provide sharp peaks due to the photoelectric effect in NaI. The Na^{24} provides a total of five peaks, three of which are sharp. These three are the photoelectric peaks from each of the two gamma-rays and the pair production peak (with escape of both annihilation quanta) from the 2.76-Mev line. The Na^{24} gives three good calibration values in a single counting sequence.

Various other sources have been used for calibration and resolution determinations. It has been found that the resolution[6] is given by the relation

$$\Delta E/E=(a/\sqrt{E})+b,$$

where ΔE is the full width of a line at half-maximum, E is the energy of the line, and a and b are constants. For the work described here, with E in kev, a was 1.64 and 2.05 and b was 0.05 and 0.036, respectively, for the two probes used.

III. METHOD

Figure 1 shows the experimental arrangement used in these experiments. A narrow beam of neutrons from the Argonne heavy water moderated pile is allowed to impinge on the capturing material. The capture gamma-rays generated are emitted in all directions and a narrow beam passes through the slit onto the NaI crystal.

The crystal and photomultiplier are inside of a heavy bismuth shield. There are 8 inches of bismuth on the side of the shield facing the pile and 4 inches on all other sides. The block of bismuth containing the $1\frac{1}{2}$-inch circular aperture is 2 inches thick. The sides of the aperture are lined with $\frac{1}{4}$ inch of steel. This is needed to attenuate the annihilation quanta that arise from capture gamma-rays and scattered pile gamma-rays that strike the bismuth sides of the aperture. The whole shield is wrapped in 30-mil cadmium. The aperture is free of cadmium but $\frac{1}{4}$ inch of LiF inside a thin cardboard container is placed over the aperture to absorb scattered thermal neutrons from the capturing samples. The crystal is in line with the capturing samples and the aperture. Back of the crystal (in a downward direction) is a gap of two feet to the floor of the pile room.

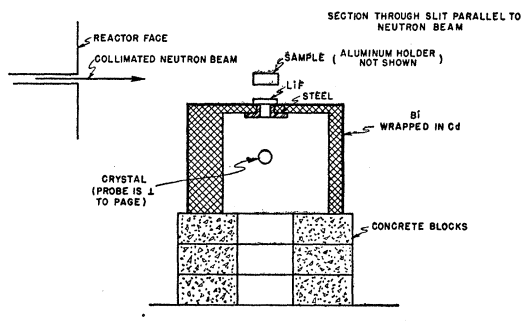

FIG. 1. Schematic arrangement of apparatus.

[6] V. Hummel and B. Hamermesh, A.E.C.U. 2020.

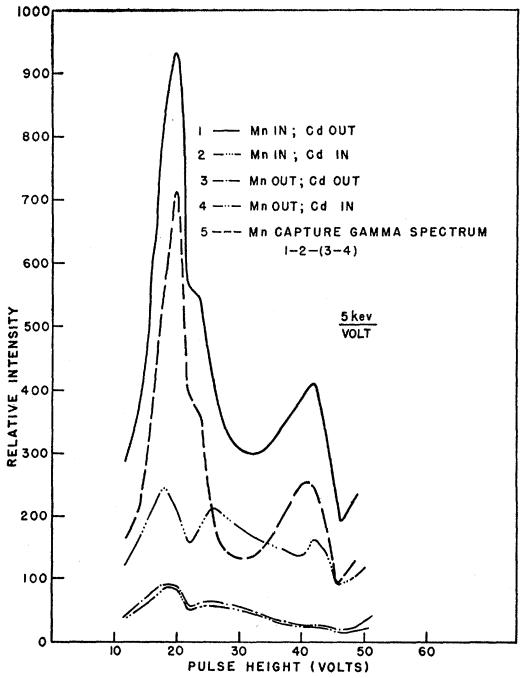

FIG. 2. Pulse-height distribution from low energy capture gamma-rays from manganese.

The samples used were in the shape of disks $\frac{3}{4}$ inch in diameter where the solid element was available. If powders were used, they were placed inside of $\frac{3}{4}$-inch diameter lusteroid test tubes. The thickness of a sample (in the direction of the neutron beam) was made as large as possible, consistent with conservative counting rates (except for Sc where only 5 grams of the oxide was available). The criteria used were not to allow any mechanical recorder to count faster than 250 clicks per minute and, in addition, not to allow the last channel (the integrating channel), to count more than 25,000 counts per minute. These limits were found to be very safe.

The neutron flux on the target was monitored with a BF_3 pulse chamber placed in a hole in the reactor near the beam hole used in these experiments. It was found that equal monitor intervals usually occurred in equal time intervals.

IV. RESULTS

In order to make a measurement at a given region of a spectrum, a cycle of four measurements was required. These measurements are:

1. Source in—no cadmium in the beam.
2. Source in—cadmium in the beam.
3. Source out—no cadmium in the beam.
4. Source out—cadmium in the beam.

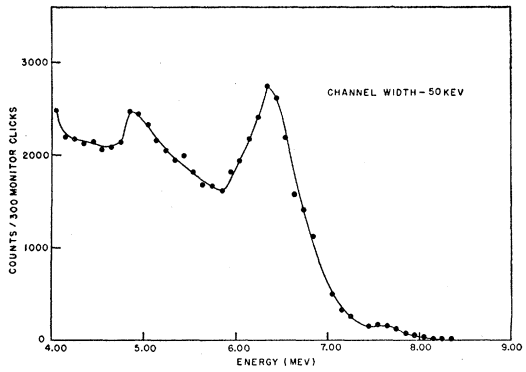

FIG. 3. High energy pulse distribution from neutron capture gamma-rays of iron.

The cadmium was placed in the beam near the pile, i.e., far from the photomultiplier shield, so as to prevent cadmium capture gamma-rays from seeing the aperture in the shield. The effect of the capture gamma-rays from a given sample is related to the four measurements given above by the relation 1–2–(3–4). Figure 2 shows a curve for manganese in the low energy region on gain A (5 kev/volt) set for 2-volt intervals. The curves 3 and 4 are so nearly the same (as they should be if the beam is well collimated) that steps 3 and 4 may be omitted, and the effect can be found by taking data with and without cadmium in the beam with the capturing sample in place. The data of Fig. 2 show two very intense lines, one at 90 kev and the other at 190 kev,

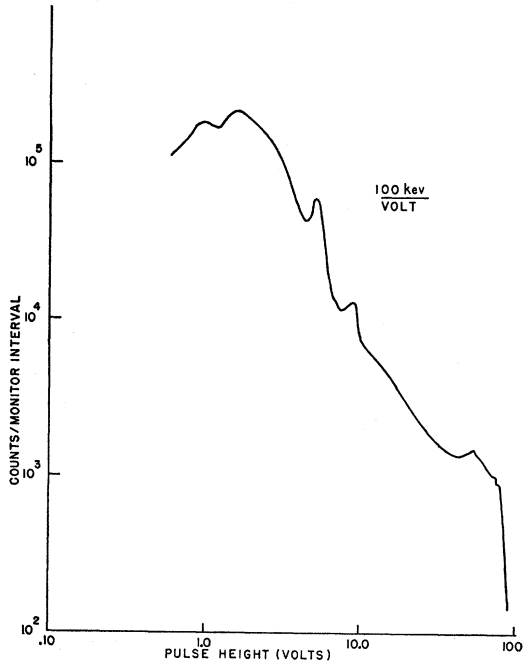

FIG. 4. Pulse-height distribution from capture gamma-rays from chromium.

emitted by the rapidly decaying[7] Mn^{56} compound nucleus formed on neutron capture.

Figure 3 shows a typical high energy portion of a spectrum from an iron sample. There are three peaks present. They arise from pair production in NaI caused by gamma-rays at 6.0, 7.4, and 8.5 Mev. The 6.0- and 7.4-Mev lines are most likely from Fe^{57}, whereas the 8.5-Mev line, which is much weaker, is probably from a ground-state transition in Fe^{55}. The very poor high energy resolution (5–6 percent) makes it difficult to observe very much of the structure of the high energy parts of the spectra such as is seen by a pair spectrometer.

In the lower energy portions of the spectra a very serious difficulty arises due to Compton scattering of the high energy lines in the NaI. Consider a low energy

TABLE I. Prominent thermal neutron capture gamma-rays from the elements $Z=17-30$.

Element	Sample	Gamma-ray energy (Mev)	Remarks
Cl	C_2Cl_6	0.784±0.010 1.15 ±0.010 1.59 ±0.013 2.00 ±0.025 6.2±0.2, 7.7±0.2	
K	Fluoride	6.0, 8.2	
Ca	Metal	6.8, 8.2	
Sc	Oxide	0.152, 0.220, 7–9	0.152 line is from the 20-sec isomer of Sc^{46}. This has been measured as 0.142 ±0.005 in a separate experiment. No crossover of 0.152 and 0.220 lines is observed. Not enough material was available to study high energy region. Curves show possible lines in 7–9-Mev region.
Ti	Oxide	1.0, 1.38, 5.0, 6.5–7.0	The 1.0 line is probably a Compton peak from the 1.38-Mev line. There are two lines between 6.5 and 7.0 Mev.
V	Metal	5.3, 5.7, 6.8, 7.4	A line at 1.4 Mev from V^{52} beta-decay is observed.
Cr	Metal	0.880, 5–6, 8.0–8.5, 8.5–9	Spectrum is very complex at high energies.
Mn	Metal	0.090, 0.190, 5.0, 7.2	No crossover of the 90- and 190-kev lines is observed. A 900-kev line from the 2.6-hour Mn^{56} activity is observed.
Fe	Metal	0.425, 6.0, 7.4, 8.5	No line at 1.4 Mev is observed. (See reference 3.)
Co	Oxide	0.220, 1.1, 1.5, 5.8, 7.0	
Ni	Metal	6.5–7.5, 8.5–9	Very complex spectrum.
Cu	Metal	0.150, 6.5–7, 7–8	
Zn	Metal	1.0, 7.5	

[7] The nucleus emitting the capture spectrum will be the one usually listed unless otherwise stated.

TABLE II. Prominent thermal neutron capture gamma-rays from the elements $Z=45-57$.

Element	Sample	Gamma-ray energy (Mev)	Remarks
Rh	Metal	0.080, 0.160	Gamma-rays probably from Rh^{104} 4.34-min and Rh^{104} 44-sec activities.
Pd	Metal	Unresolved	
Ag	Metal	0.187	
Cd	Metal	0.558, 8.5	See reference 4.
In	Metal	0.160, 0.256	No crossover observed.
Sn	Metal	7.5–8.0	
Sb	Metal	Unresolved	
Te	Metal	0.609	
I	CHI_3	Unresolved	
Cs	Nitrate	Unresolved	
Ba	Metal	Unresolved	
La	Metal	4.5	

portion of a capture spectrum. Then all of the higher energy lines feed their Compton electrons (formed in NaI) into lower energy parts of the spectrum. Therefore, the lower energy lines sit on top of a rapidly rising background. Figure 4 shows the complete normalized counting rate *vs* energy curve for the capture gamma-rays from chromium. The rapid rise toward low energies means that only the most prominent lines of the capture spectra can be observed.

Tables I and II contain those lines that stand out above the rapidly rising background that is present with all elements as one goes toward low energies. Gamma-rays following beta-decay are also observed. In all cases listed the spectra were explored from the region of 20 kev up to the point where no counts were detectable above background. All lines below 700 kev were measured to ± 10 kev. In the case of chlorine, the lines were measured with the accuracy indicated in Table I.

The results given in the tables show no systematic structure of the spectra. In the case of chlorine, however, interesting conclusions can be drawn since the spectrum has recently been studied with the pair spectrometer by Kinsey, Bartholemew, and Walker.[8] The decay scheme of Cl^{36} that is presented in Fig. 10 of their paper proposes that the first four excited states have the energies 0.79 ± 0.04, 1.14 ± 0.04, 1.58 ± 0.04, and 1.94 ± 0.07 Mev, respectively. These energies are obtained by subtracting from the neutron binding energy of Cl^{36} the energies of the lines which Kinsey *et al.*, label *B–E*. The neutron binding energy has been reported as 8.54 Mev by Shrader and Pollard[9] and 8.49 Mev by Ennis.[10] Kinsey *et al.*,[8] find that the most energetic gamma-ray from Cl^{36} has an energy of 8.56 ± 0.03 Mev. This line probably arises from a ground-state transition from the initial state of Cl^{36} that is formed upon neutron capture.

The lines that we find at 0.784, 1.15, 1.59, and 2.00 Mev correspond to the transitions to the ground state from the levels reached by Kinsey's line *B–E* in Cl^{36}. The line at 6.2 Mev is probably Kinsey's line *F* and the line at 7.7 Mev is probably his line *B*. The 7.7-Mev line and the 0.784-Mev line are in cascade. The error in our measurement of the 7.7-Mev line does not permit us to make a good binding energy determination from our data alone.

[8] Kinsey, Bartholomew, and Walker, Phys. Rev. **85**, 1012 (1952).
[9] E. F. Shrader and E. Pollard, Phys. Rev. **59**, 277 (1941).
[10] W. W. Ennis, Phys. Rev. **82**, 304 (1951).

TABLE III. Neutron binding energy of Cl^{36} from combined scintillation spectrometer and pair spectrometer data.

Line	Scintillation spectrometer	Pair spectrometer	Binding energy of Cl^{36}
A	···	8.56 ± 0.03 Mev	8.56 ± 0.03 Mev
B	0.784 ± 0.010 Mev	7.77 ± 0.03 Mev	8.55 ± 0.03 Mev
C	1.15 ± 0.010 Mev	7.42 ± 0.03 Mev	8.57 ± 0.03 Mev
D	1.59 ± 0.013 Mev	6.98 ± 0.03 Mev	8.57 ± 0.03 Mev
E	2.00 ± 0.025 Mev	6.62 ± 0.06 Mev	8.62 ± 0.06 Mev
		Weighted average	8.57 ± 0.03 Mev

If we label our four lowest lines *B–E* and combine each of them with Kinsey's line of the corresponding letter, then we find the values for the neutron binding energy of Cl^{36} shown in Table III.

The results in Table III indicate that these four branches of the capture gamma-decay scheme of Cl^{36} involve only two steps. Of course, the order of emission of the two lines in a branch is not definitely known. However, on the basis of our present knowledge (dependence of intensity on energy difference), it seems more likely that the more energetic transition in a given branch will occur first. This would mean that the four lowest lines that we observe arise from the first four excited states of Cl^{36}.

V. CONCLUSIONS

The use of a NaI gamma-ray spectrometer for the study of the capture gamma-ray spectra of the elements $Z=17-30$ and $Z=45-57$ yields data on only the most prominent lines in these spectra. The poor high energy resolution plus the rapidly rising background at lower energies makes it difficult to observe more than the most prominent lines. The low energy lines that are observed probably arise from states very near the ground state. In the case of chlorine, it is very likely that the line at 784 kev is emitted from the first excited state of Cl^{36} and that the lines at 1.15, 1.59, and 2.00 Mev arise from the next three excited states. No apparent systematics of the spectra are evident.

It is with great pleasure that we extend our thanks to our colleagues from various divisions at the Argonne National Laboratory who have helped make this work possible. We are especially grateful to Robert Swank of our Instrument Research Division and Robert Schumann of our Electronics Section. Mr. Swank provided the excellently packaged, high resolution crystals and the selected photomultipliers and a very stable single-channel analyzer which was used in the very early stages of our investigation. Mr. Schumann designed and built the 20-channel analyzer.