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Gamma-Ray Attenuation-Coefficient Measurements*

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Total γ-ray attenuation coefficients have been measured at nine energies in the range of 88 keV to 2.75 MeV for the following elements: Be, C, Mg, Al, S, Ti, Fe, Ni, Cu, Zn, Zr, Nb, Mo, Ag, Sn, La, Gd, Hf, W, Au, Pb, Th, U, and Pu. Radioactive isotopes were used as sources of monoenergetic γ radiation in a narrow-beam-collimated geometry. Experimental errors are less than 1%.

INTRODUCTION

A significant number of photon attenuation-coefficient measurements, calculations, and compilations has been published recently.[1-18] Appreciable discrepancies between some of the experimental and theoretical values indicate the need for additional accurate and consistent measurements of total attenuation coefficients. The initial results of comprehensive measurements in the energy range of 25 to 130 keV were reported by McCrary *et al.*[19] The present paper presents additional results extending the range of attenuation-coefficient measurements to 2.75 MeV.

The narrow-beam total attenuation coefficient μ, with units of cm^2/g, is defined by the relation

$$I = I_0 e^{-\mu x}, \tag{1}$$

where I_0 is the incident beam intensity, I is the transmitted intensity, and x is the sample thickness in g/cm^2. In the present work, μ was measured at nine energies in the range of 88 keV to 2.75 MeV for 24 elements with atomic numbers ranging from 4 to 94. Radioactive isotopes were used as sources of monoenergetic γ radiation. Both the incident and transmitted beams were collimated.

APPARATUS

Figure 1 is a schematic representation of the experimental apparatus. The γ source is located in a collimated steel source holder having a slot which provides for about 2 in. of incident beam collimation. The sample is placed between the source collimator and the main collimator, which is a 15-in.-long steel slab having a 0.1-in.^{2} collimating slot. For a typical sample, this collimation system reduces the maximum solid angle of scattering from sample to detector to less than 0.5×10^{-4} sr. The source, sample, collimators, and detector are shielded by approximately 8 in. of lead. The detector is a $1\frac{1}{2}$-in.-diam by $\frac{1}{2}$-in.-long NaI(Tl) crystal with a 0.005-in. Be window mounted on a RCA 6342A photomultiplier tube. Signals from the photomultiplier are amplified and fed through a single-channel analyzer to a preset time scaler.

SOURCES

The nine radioactive isotopes used as γ-ray sources are listed in Table I. The γ-ray energy, half-life, and method of production of each source are indicated. The γ-ray energies used are considered to be the most accurate values from the

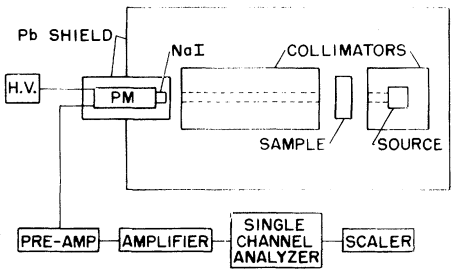

FIG. 1. Schematic diagram of experimental apparatus.

current literature and are referenced individually. Five of the sources, ^{177}Lu, ^{198}Au, ^{65}Zn, ^{140}La, and ^{24}Na are multiple γ emitters. The highest-energy γ ray was used with ^{177}Lu, ^{65}Zn, and ^{24}Na. The ^{198}Au source emits two γ rays of higher energy than the 411.80-keV γ ray used in the measurements. These γ rays are emitted at 676 keV (1%) and 1.088 MeV (0.2%). ^{140}La emits one higher-energy γ ray at 2.53 MeV (3%). Thus, for the ^{198}Au and ^{140}La sources, it is possible that an electron resulting from the Compton scatter (in the NaI crystal) of a higher-energy γ ray may be detected at the photopeak energy of the principal γ ray. Corrections for this effect were found to be negligible for ^{198}Au, while only a small correction was necessary for ^{140}La, as discussed in the data-analysis section.

SAMPLES

Most of the samples were machined as right circular cylinders with diameters 0.5–0.75 in. and thicknesses approximately 0.2–2.0 in. The Ag and Au samples were cut in rectangular shapes from rolled plates of 0.08–0.25 in. and 0.005–0.10 in. thicknesses, respectively. The *S* samples were hot pressed into cylindrical shapes. The Pu samples were machined from a Pu rod to thicknesses of 0.006–0.25 in., and were clad with 0.0004-in. Ni for health protection. The area and weight of each sample were measured and the average thickness in g/cm^2 was calculated.

A specimen from the material used for preparing the samples of each element was subjected to spark-source mass spectrometric analysis and/or chemical analysis to determine the quantities of all impurities in the samples. In general, the samples were very pure, but for some elements small impurity corrections to the measured attenuation coefficients were necessary.

EXPERIMENTAL PROCEDURE

In addition to the sample thickness, three quantities are measured to calculate μ, namely, the incident beam intensity, the attenuated beam intensity, and the background. Incident beam intensities varied from 500 counts/min with the ^{109}Cd source to 50 000 counts/min with the ^{198}Au source. Attenuated beam intensities were measured with the sample in a movable holder which positioned the sample normal to the incident beam. The sample could be moved with respect to the beam so that any part of the sample surface could be exposed to the γ rays. Errors introduced by deviations in the average thickness of the sample were then minimized by measuring the attenuated beam intensity at a large number of positions over the sample area. The background count was taken

TABLE I. Gamma sources.

Source	Energy (keV)	Half-life	Method production[a]
^{109}Cd	88.09[b]	453*d*	*C*
^{141}Ce	145.41[c]	32.5*d*	*C*
^{177}Lu	208.36[d]	6.7*d*	*N*
^{203}Hg	279.12[d]	46.9*d*	*N*
^{198}Au	411.80[d]	2.7*d*	*N*
^{137}Cs	661.6[d]	30*y*	*C*
^{65}Zn	1115.5[e]	245*d*	*C*
^{140}La	1598.0[d]	40.2*h*	*N*
^{24}Na	2753.9[d]	15*h*	*N*

[a] *C* is commercially produced source; *N* is produced by thermal neutron capture from irradiation in LASL reactor.

[b] C. Foin, A. Gizon, and J. Oms, Nucl. Phys. A113, 241 (1968).

[c] D. H. White and D. J. Groves, Nucl. Phys. A91, 453 (1967).

[d] C. M. Lederer, J. M. Hollander, and I. Perlman, Table of Isotopes (John Wiley & Sons, Inc., New York, 1968), 6th ed.

[e] W. W. Black and R. L. Heath, Nucl. Phys. A90, 650 (1967).

by placing a 4.0-in.-long depleted uranium rod in the sample position.

Counting times from 1 to 5 min were used, depending on the source strength. For each sample, counts were taken in the following sequence: background, no sample, sample, sample, sample, no sample, background, no sample, sample, and so on. The sample was moved between "sample" counts. The counting sequence was continued in most cases until counting statistics contributed less than 0.6% error to the measured value of μ. In obtaining this statistical level, it was necessary to make from 9 to 135 sample counts for a single element and energy. Most efficient use of counting time was made by selecting a sample thickness which gave a value of I/I_0 in the range of $0.1 < I/I_0 < 0.4$. For the radioactive samples Th, U, and Pu, counts were also taken with the sample in place and the source removed, in order to correct for any γ contribution by the sample to the sample counts. In taking the Pu data, the "no sample" counts were made with two layers of 0.0004-in. Ni in the beam to correct for attenuation caused by the Ni cladding on the Pu samples.

DATA ANALYSIS

From a sequence of counts taken for a given γ-ray energy, a series of transmission ratios $R = I/I_0$ was calculated. The procedure was as follows: Let the sequence of counts be denoted by B_1, N_1, S_1, S_2, S_3, N_2, B_2, N_3, S_4, S_5, and so on, where B is the background count, N is the no-sample count, and S is the sample count. For the short half-life sources the data-recording cycle was adjusted so that a linear decay correction was sufficiently accurate over the period of a single cycle. All counts for a given sample are for the same counting time. Then we have

$$
\begin{aligned}
R_1 &= (S_1 - B_1)/(N_1 - B_1)\,, \\
R_2 &= [S_2 - \tfrac{1}{2}(B_1 + B_2)]/[\tfrac{1}{2}(N_1 + N_2) - \tfrac{1}{2}(B_1 + B_2)]\,, \\
R_3 &= (S_3 - B_2)/(N_2 - B_2)\,, \\
R_4 &= (S_4 - B_2)/(N_3 - B_2)\,, \\
R_5 &= [S_5 - \tfrac{1}{2}(B_2 + B_3)]/[\tfrac{1}{2}(N_3 + N_4) - \tfrac{1}{2}(B_2 + B_3)]\,. \\
&\vdots
\end{aligned}
\tag{2}
$$

In this manner, a set $\{R_i\}$ was calculated for each sample at each energy. From the mean transmission $\overline{R}$ of the set, a value of the attenuation coefficient μ was calculated:

$$\mu = -x^{-1} \ln \overline{R}\,, \tag{3}$$

where x = average sample thickness in g/cm^2. From the standard deviation δ_R calculated from the set $\{R_i\}$, a standard deviation of μ, δ_μ, was calculated:

$$\delta_\mu = \left| \frac{\partial \mu}{\partial R} \right| \delta_R\,. \tag{4}$$

Where applicable for the radioactive samples Th, U, and Pu, a sample correction C_s was subtracted from the numerator of R_i, where

$$C_s = (\text{sample background-room background}).$$

These counts were taken, with the source removed, for a long period of time and corrected for the appropriate counting time. The sample correction for Pu at the 88-keV energy of the ^{109}Cd source resulted in an increase in μ of about 2%. Sample corrections for Pu at the other energies and for Th and U at all energies were negligible.

The measured values of μ at the ^{140}La source energy of 1.60 MeV were corrected for Compton electron counts resulting from the 2.53-MeV γ rays which were scattered within the NaI detector. The correction was made by subtracting, from the numerator and denominator of each R_i, the fraction of the counts due to Compton electrons which are detected in the analyzer window width of about 1.60 MeV. For the sample count due to both the 1.60- and 2.53-MeV γ rays, we have

$$
\begin{aligned}
S \propto\, & f(1.6)\, p_t(1.6)\, p_d(1.6) \\
& + f(2.53)\, p_t(2.53)\, p_d(2.53)\,,
\end{aligned}
$$

where $f(1.6)$ is the fraction of source γ rays with 1.6-MeV energy, $p_t(1.6)$ is the probability of transmission through sample for 1.6-MeV γ ray, and $p_d(1.6)$ is the probability of detection in NaI for 1.6-MeV γ ray. These quantities are defined similarly for the 2.53-MeV γ ray. For the "no-sample" count due to both γ rays,

$$N \propto f(1.6)\, p_d(1.6) + f(2.53)\, p_d(2.53)\,.$$

The source fractions are $f(1.6) = 0.96$ and $f(2.53) = 0.03$. The transmission probabilities are

$$p_t(1.6) = e^{-\mu(1.6)x} \quad \text{and} \quad p_t(2.53) = e^{-\mu(2.53)x}\,,$$

where μ in cm^2/g and x in g/cm^2 are the attenuation coefficient at the appropriate energy and the sample thickness, respectively. The detection probabilities are

$$p_d(1.6) = \mu_\gamma^{\,\mathrm{NaI}}(1.6),$$

and $p_d(2.53) = \mu_e^{\mathrm{NaI}}(1-\epsilon_{0.93}^{\mathrm{NaI}})$,

where $\mu_\gamma^{\mathrm{NaI}}(1.6)$ is the photopeak cross section in the NaI detector for a 1.60-MeV γ ray, and μ_e^{NaI} is the cross section for production of a 1.60-MeV electron resulting from the Compton scatter of a 2.53-MeV γ ray. $\epsilon_{0.93}^{\mathrm{NaI}}$ is the NaI detector efficiency[20] for the 0.093-MeV scattered γ ray ($h\nu' = h\nu_0 - T_e$) so that $(1-\epsilon_{0.93}^{\mathrm{NaI}})$ is the probability that the scattered γ ray escapes the detector, resulting in the detection of only the 1.60-MeV electron. This correction was applied to all samples at the ^{140}La source energy and resulted in an average increase in μ of 0.5%. The correction was applied for the two higher-energy γ rays from the ^{198}Au source and was found to be negligible for all samples at the ^{198}Au source energy, which is in agreement with the experimental and calculated results of Colgate.[9]

For certain combinations of energy, sample material, and scattering angle, a γ ray may be coherently scattered through a small angle and be degraded so slightly in energy that the scattered γ ray reaches the detector and is incorrectly counted as part of the unattenuated "sample" count. While this Rayleigh scattering is generally not considered in broad beam conditions, it must be accounted for in the interpretation of experimental attenuation coefficient data.[18] Using the calculated differential scattering cross-section data of Brown[21] to interpolate at small angles and the data of Veigele[22] for extrapolation to energies above 1 MeV, the Rayleigh scattering was found to contribute a maximum of less than 0.05% to the measured attenuation coefficients, so that no coherent scattering corrections were applied to the data.

Using the Klein-Nishina formula[23] for Compton scattering, it was found that incoherent scattering in the sample made an insignificant contribution to the "sample" count.

From the quantitative analysis of each sample, corrections in μ because of impurities were applied as follows:

$$\mu_{\mathrm{element}} = (\mu_{\mathrm{meas}} - \textstyle\sum_i \mu_i f_i)/(1 - \sum_i f_i) \quad , \quad (5)$$

where μ_i is the attenuation coefficient for the ith impurity and f_i is the weight fraction of the ith impurity element in the sample. Since the values of f_i were small, the required accuracy of values of μ_i was not as great as the desired accuracy in μ_{element}. This was demonstrated by applying second-order corrections in (5), i.e., impurity corrected values of μ_i were used to recalculate μ_{element}. The second-order corrections made no changes in any of the values of μ_{element}. All impurities as large as 10 ppm were included in (5).

RESULTS

The results of the present series of measurements are listed in Table II. The errors are 1 standard deviation in the corrected value of μ, where the corrections are those discussed in the data-analysis section.

It should be borne in mind that absorption coefficients in cm^2/g are unsuitable for Z interpolation and should be converted to units such as b/atom before use in this regard.

The attenuation coefficients of five representative elements (Be, Al, Fe, Gd, and Pu) are plotted in Fig. 2 along with the results of McCrary *et al.*[19] to illustrate consistency between the x-ray and the γ-ray measurements in the energy region where the two sets of measurements overlap.

The present measurements are in general agreement with the results of Wyard[8] in the energy range of 279 keV to 1.51 MeV, while agreement with the results of Davisson and Evans[10] and Colgate[9] is seen with the exception of some measurements at 2.62 and 2.75 MeV, respectively.

ERRORS

Sources of random errors include counting statistics, uncertainties in sample thickness, and errors in applying sample impurity corrections. Counting statistics for most of the measurements

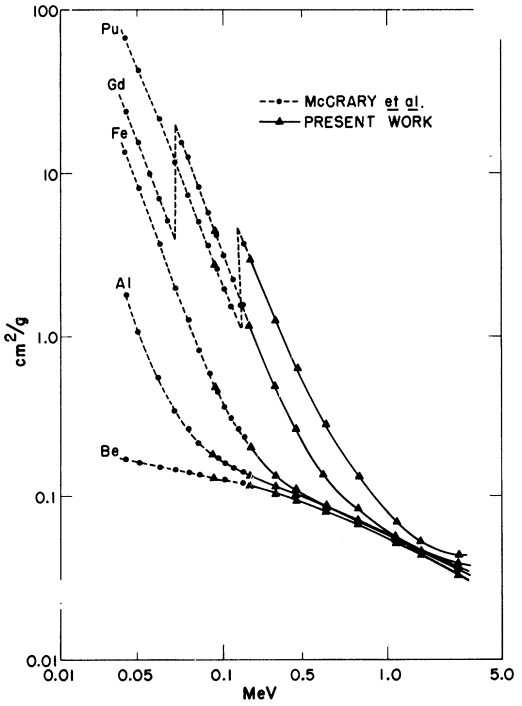

FIG. 2. Attenuation coefficients (cm^2/g) versus photon energy (keV) for Be, Al, Fe, Gd, and Pu.

TABLE II. Experimentally determined values of γ-ray attenuation coefficients in cm^2/g.

Element	88.09 keV	145.41 keV	208.36 keV	279.12 keV	411.80 keV
Beryllium	0.1360 ± 0.0008	0.1193 ± 0.0003	0.1070 ± 0.0005	0.09686 ± 0.00031	0.08295 ± 0.00035
Carbon	0.1560 ± 0.0007	0.1357 ± 0.0008	0.1209 ± 0.0002	0.1087 ± 0.0002	0.09401 ± 0.00014
Magnesium	0.1817 ± 0.0007	0.1403 ± 0.0007	0.1205 ± 0.0004	0.1090 ± 0.0005	0.09320 ± 0.00019
Aluminum	0.1827 ± 0.0008	0.1382 ± 0.0008	0.1198 ± 0.0008	0.1064 ± 0.0005	0.09058 ± 0.00043
Sulfur	0.2274 ± 0.0012	0.1527 ± 0.0008	0.1286 ± 0.0005	0.1115 ± 0.0003	0.09513 ± 0.00031
Titanium	0.3384 ± 0.0035	0.1711 ± 0.0009	0.1267 ± 0.0004	0.1072 ± 0.0005	0.08851 ± 0.00037
Iron	0.4841 ± 0.0026	0.2036 ± 0.0006	0.1392 ± 0.0003	0.1127 ± 0.0003	0.09064 ± 0.00021
Nickel	0.5754 ± 0.0036	0.2225 ± 0.0021	0.1499 ± 0.0013	0.1187 ± 0.0003	0.09442 ± 0.00035
Copper	0.6005 ± 0.0038	0.2297 ± 0.0007	0.1473 ± 0.0011	0.1165 ± 0.0006	0.09115 ± 0.00038
Zinc	0.6546 ± 0.0045	0.2420 ± 0.0016	0.1538 ± 0.0007	0.1185 ± 0.0005	0.09281 ± 0.00022
Zirconium	1.333 ± 0.007	0.3994 ± 0.0015	0.2062 ± 0.0007	0.1392 ± 0.0009	0.09755 ± 0.00037
Niobium	1.468 ± 0.007	0.4254 ± 0.0015	0.2151 ± 0.0006	0.1456 ± 0.0008	0.09976 ± 0.00038
Molybdenum	1.518 ± 0.007	0.4533 ± 0.0018	0.2227 ± 0.0009	0.1485 ± 0.0005	0.1008 ± 0.0004
Silver	2.031 ± 0.013	0.5771 ± 0.0024	0.2707 ± 0.0010	0.1697 ± 0.0005	0.1075 ± 0.0004
Tin	2.384 ± 0.017	0.6458 ± 0.0040	0.2928 ± 0.0011	0.1772 ± 0.0005	0.1092 ± 0.0005
Lanthanum	3.276 ± 0.014	0.8879 ± 0.0023	0.3829 ± 0.0023	0.2165 ± 0.0008	0.1227 ± 0.0004
Gadolinium	4.376 ± 0.026	1.184 ± 0.004	0.4946 ± 0.0039	0.2691 ± 0.0009	0.1405 ± 0.0005
Hafnium	5.696 ± 0.042	1.567 ± 0.011	0.6457 ± 0.0042	0.3413 ± 0.0019	0.1684 ± 0.0007
Tungsten	6.148 ± 0.030	1.687 ± 0.005	0.7053 ± 0.0040	0.3689 ± 0.0007	0.1775 ± 0.0005
Gold	7.006 ± 0.039	1.982 ± 0.012	0.8236 ± 0.0023	0.4248 ± 0.0021	0.1998 ± 0.0012
Lead	6.576 ± 0.027	2.146 ± 0.009	0.9017 ± 0.0040	0.4621 ± 0.0024	0.2130 ± 0.0013
Thorium	2.426 ± 0.013	2.622 ± 0.012	1.093 ± 0.006	0.5575 ± 0.0030	0.2548 ± 0.0009
Uranium	2.573 ± 0.011	2.756 ± 0.009	1.156 ± 0.004	0.5876 ± 0.0023	0.2671 ± 0.0013
Plutonium	2.766 ± 0.019	2.929 ± 0.012	1.213 ± 0.006	0.6290 ± 0.0011	0.2844 ± 0.0012

Element	661.6 keV	1115.5 keV	1598.0 keV	2753.9 keV
Beryllium	0.06836 ± 0.00030	0.05296 ± 0.00021	0.04420 ± 0.00023	0.03288 ± 0.00014
Carbon	0.07713 ± 0.00036	0.06043 ± 0.00027	0.04953 ± 0.00019	0.03729 ± 0.00014
Magnesium	0.07653 ± 0.00044	0.06023 ± 0.00023	0.04890 ± 0.00018	0.03760 ± 0.00019
Aluminum	0.07436 ± 0.00025	0.05807 ± 0.00045	0.04808 ± 0.00016	0.03682 ± 0.00018
Sulfur	0.07822 ± 0.00043	0.06079 ± 0.00020	0.05100 ± 0.00019	0.03877 ± 0.00013
Titanium	0.07130 ± 0.00023	0.05590 ± 0.00029	0.04632 ± 0.00019	0.03613 ± 0.00019
Iron	0.07258 ± 0.00022	0.05610 ± 0.00018	0.04637 ± 0.00022	0.03751 ± 0.00011
Nickel	0.07497 ± 0.00028	0.05786 ± 0.00021	0.04857 ± 0.00022	0.03779 ± 0.00013
Copper	0.07285 ± 0.00038	0.05521 ± 0.00027	0.04630 ± 0.00017	0.03713 ± 0.00014
Zinc	0.07296 ± 0.00030	0.05606 ± 0.00021	0.04638 ± 0.00022	0.03720 ± 0.00017

TABLE II. (*continued*)

Element	661.6 keV	1115.5 keV	1598.0 keV	2753.9 keV
Zirconium	0.07160 ± 0.00028	0.05393 ± 0.00021	0.04520 ± 0.00014	0.03715 ± 0.00019
Niobium	0.07290 ± 0.00021	0.05517 ± 0.00030	0.04561 ± 0.00021	0.03767 ± 0.00010
Molybdenum	0.07304 ± 0.00024	0.05467 ± 0.00017	0.04502 ± 0.00019	0.03747 ± 0.00018
Silver	0.07585 ± 0.00030	0.05547 ± 0.00022	0.04567 ± 0.00021	0.03796 ± 0.00018
Tin	0.07441 ± 0.00035	0.05348 ± 0.00022	0.04445 ± 0.00022	0.03703 ± 0.00016
Lanthanum	0.07796 ± 0.00022	0.05398 ± 0.00026	0.04480 ± 0.00016	0.03735 ± 0.00018
Gadolinium	0.08317 ± 0.00033	0.05619 ± 0.00025	0.04532 ± 0.00021	0.03862 ± 0.00016
Hafnium	0.09202 ± 0.00042	0.05823 ± 0.00021	0.04693 ± 0.00026	0.03978 ± 0.00019
Tungsten	0.09600 ± 0.00047	0.05964 ± 0.00028	0.04753 ± 0.00023	0.04070 ± 0.00018
Gold	0.1031 ± 0.0004	0.06205 ± 0.00032	0.04893 ± 0.00020	0.04202 ± 0.00017
Lead	0.1074 ± 0.0005	0.06324 ± 0.00032	0.04940 ± 0.00023	0.04151 ± 0.00020
Thorium	0.1202 ± 0.0004	0.06721 ± 0.00033	0.05164 ± 0.00019	0.04356 ± 0.00020
Uranium	0.1244 ± 0.0005	0.06876 ± 0.00040	0.05246 ± 0.00026	0.04365 ± 0.00016
Plutonium	0.1323 ± 0.0007	0.07176 ± 0.00031	0.05404 ± 0.00017	0.04549 ± 0.00022

were less than 0.6%. Errors due to variations in sample thickness were kept to a minimum by measuring the transmission at a large number of positions over the sample area. Since the sample impurity corrections were small, errors in applying this correction were correspondingly small. For all samples except Ti, the average impurity correction to μ was 0.06%. The correction for Ti at 88 keV was 8%, while corrections for Ti at all other energies averaged 0.4%. The impurity corrections for Ti were due to the presence of 0.44 weight % W in the Ti samples. There are no known sources of systematic errors in the present work.

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