Natural Alpha Radioactivity in Medium-Heavy Elements*†

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A large cylindrical ionization counter accommodating samples up to 1200 cm² in area has been used for measurements of natural alpha radioactivity in medium-heavy elements. Low-background techniques and multichannel pulse analysis are employed. The method has greater energy resolution and yields better counting statistics than the nuclear emulsion technique, but does not have as great a sensitivity.

The results obtained from measurements on natural elements and isotopically enriched samples are:

Nuclide	Alpha-particle energy (Mev)	Specific activity of natural element (dis sec^{-1} g^{-1})	Half-life or limit for nuclide (year)
Ce^{142}	···	<0.0002	$>5\times10^{16}$
Nd^{144}	1.83±0.03	0.0093±0.0011	$(2.4\pm0.3)\times10^{15}$
Sm^{146}	···	<0.013	$<3\times10^{8}$
Sm^{147}	2.23±0.02	116±5	$(1.15\pm0.05)\times10^{11}$
Sm^{148}	···	<0.04	$>2\times10^{14}$
Sm^{149}	···	<0.01	$>1\times10^{15}$
Gd^{152}	2.14±0.03	0.00156±0.00010	$(1.08\pm0.08)\times10^{14}$
Hf^{174}	2.50±0.03	0.000065±0.000013	$(2.0\pm0.4)\times10^{15}$
W^{180}	···	<0.00013	$>9\times10^{14}$
Pt^{190}	3.11±0.03	0.012±0.001	$(6.9\pm0.5)\times10^{11}$
Hg^{196}	···	<0.0007	$>1\times10^{14}$

I. INTRODUCTION

ALPHA disintegration is a prominent mode of radioactivity among the heavy elements, where the energy available is large, half-lives being observed over a large range from 10^{-7} second to 10^{10} years. For the elements immediately below bismuth the energy available is considerably smaller, and this mode of disintegration is a comparatively rare phenomenon. The elements lighter than cerium are energetically stable against alpha emission.

Hoffmann,[1] in 1921, was the first to detect alpha radioactivity belonging to an element with atomic number less than 83, when he observed what he believed to be natural alpha radioactivity in platinum. Eleven years later Hevesy and Pahl[2] detected the prominent alpha activity in natural samarium.

In 1949, Thompson, Ghiorso, Rasmussen, and Seaborg[3] produced artifically short-lived alpha emitters of gold, mercury, gadolinium, and dysprosium. Since then, extensive study by Rasmussen and co-workers[4–6] has resulted in characterization of a number of synthetic alpha emitters in the rare earth region.

With the development of the modern nuclear emulsions a very sensitive means of detecting weak natural alpha emitters was made available. By this method natural alpha emitters of bismuth,[7–9] tungsten,[10] neodymium,[11,12] platinum,[12] cerium,[13] lead,[14] gadolinium,[15] and hafnium[15] have been reported.

The first objective of the present work was to develop a method for studying weak alpha activities which would give more precise alpha-particle energies and specific activities than the nuclear emulsion technique. It was felt that a gas ionization counter would accomplish these objectives. At the same time an attempt was made to approach the extremely high sensitivity of the nuclear emulsion technique by the use of large samples and low-background techniques.

The next objective was to confirm the existence of the weak natural alpha emitters which had been reported by the nuclear emulsion technique, and to obtain more precise measurements of their alpha-particle energies and specific activities. A reliable value of the energy of the Sm^{147} alpha particle, often used as a calibration standard, was particularly to be desired.

Another general objective was to attempt to detect

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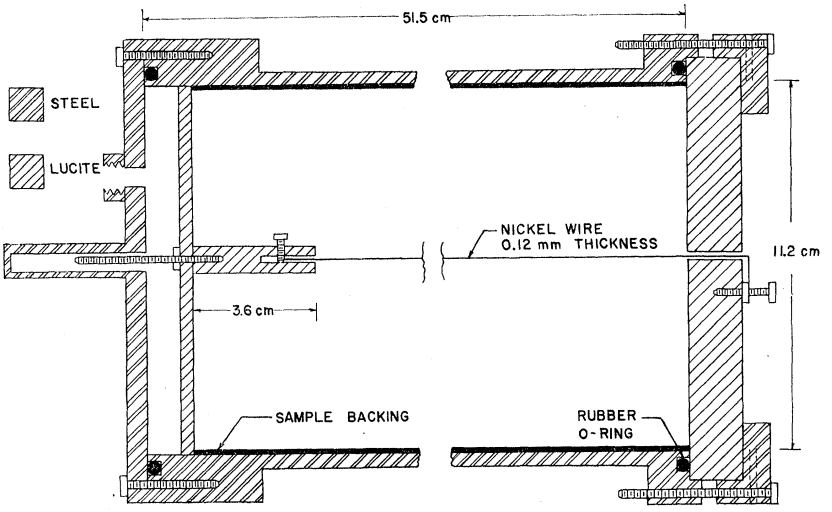

FIG. 1. Diagram of ionization chamber.

natural alpha activity in certain other nuclides in which it was expected as a possibility on the basis of semitheoretical considerations[16] and mass spectrographic data.[17]

The question of the natural alpha activity of tungsten has been a puzzle since it was first reported by Porschen and Riezler.[10] They originally determined the alpha-particle energy to be ~3.2 Mev, corresponding to a theoretical half-life of $\sim 10^9$ years, which, if correct, rules out any of the known natural isotopes of tungsten as being responsible for the activity. They postulated that it might be due to a rare neutron-deficient isotope of tungsten, for example, W^{178}. Kohman and Saito[18] pointed out, however, that the uncertainty in the value of the alpha-particle energy did not rule out the possibility that W^{180}, the lightest known naturally occurring isotope, was the active nuclide. In this study a sample of tungsten enriched in W^{180} was measured to determine whether that isotope is perceptibly alpha active.

Finally, a search was made in natural samarium for alpha activity of Sm^{146}, whose experimentally determined half-life of $\sim 5\times 10^7$ years[19] is sufficiently uncertain as not to exclude the possibility of its existence as a primary natural radionuclide.

II. EXPERIMENTAL ARRANGEMENT

The ion chamber, Fig. 1, was a cylindrical counter 51.5 cm long with an inside diameter of 11.2 cm. The anode was a 0.12-mm nickel wire strung the full length of the counter axis. Samples were deposited uniformly over the surface of a copper or stainless steel sheet which fitted concentrically into the counter against the wall. The total active surface area was 1200 cm^2.

The counting gas consisted of 94% argon, 5% ethylene, and 1% nitrogen. The organic component increases the pulse size in comparison to pure argon[20–22] and stabilizes against upward drift due to easily ionizable impurities. The nitrogen reduces the sensitivity to oxygen impurity[23,24] and stabilizes against downward drift due to electron-capturing impurities. The effect of each additive was found to hold in the presence of the other, and both were necessary to give good drift stability, which was maintained for periods as long as 72 hours.

Optimum conditions for resolution of the alpha-particle spectra were found to be a pressure of 160 cm(Hg) and an anode potential of +1400 volts. The counter was then operating in the proportional region with a very low gas gain (~1.5×).

The output of the chamber was fed into a Los Alamos Model 100-type preamplifier, then to a Higinbotham-type nonoverloading amplifier. From the latter the pulse-size spectrum was recorded with a 24-channel pulse-height analyzer with anticoincidence control.

The background of the chamber in the region above 1 Mev was reduced to a low level by employing massive shielding and anticoincidence guarding. The background from 1 to 3 Mev averaged around 17 counts per hour without massive shielding or anticoincidence, 15 counts per hour with massive shielding only, 12 counts per hour with anticoincidence only, and 9 counts per hour with massive shielding and anticoincidence.

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TABLE I. Isotopic composition of isotopically enriched samples.

Element	Mass number and atomic percent (Asterisk indicates isotope of major enrichment)						
Cerium	136	138	140	142*			
	<0.01	<0.01	9.92±0.06	90.08±0.06			
Neodymium	142	143	144*	145	146	148	150
	1.06±0.057	1.18±0.078	62.62±0.299	2.80±0.068	31.51±0.252	0.66±0.107	0.17±0.042
Samarium	144	147	148*	149	150	152	154
	0.1±0.02	6.4±0.2	83.1±0.3	7.3±0.1	1.2±0.1	1.3±0.1	0.6±0.06
Samarium	144	147	148	149*	150	152	154
	···	1.0±0.05	4.0±0.1	88.8±0.1	3.5±0.1	1.9±0.05	0.9±0.05
Gadolinium	152*	154	155	156	157	158	160
	14.96±0.28	9.75±0.13	27.26±0.27	19.32±0.19	10.08±0.18	11.67±0.21	6.97±0.24
Hafnium	174*	176	177	178	179	180	
	10.14±0.10	19.28±0.13	28.87±0.17	21.72±0.06	7.14±0.06	12.86±0.08	
Tungsten	180*	182	183	184	186		
	6.95±0.02	42.16±0.03	14.15±0.04	22.22±0.06	14.52±0.03		
Platinum	190*	192	194	195	196	198	
	0.76±0.02	5.80±0.03	45.32±0.19	29.64±0.14	15.63±0.11	2.85±0.03	
Mercury	196*	198	199	200	201	202	204
	1.46±0.01	7.48±0.06	9.03±0.04	13.15±0.07	7.87±0.04	25.24±0.09	35.78±0.09

Details of the construction and operation of the apparatus are given elsewhere.[25]

III. SAMPLES AND PREPARATION

It was necessary to prepare a thin uniform film of each sample over a large area (~1200 cm^2). The electrolytic plating bath of Pfanhauser[26] was used in preparing platinum films on copper-coated type 304 stainless-steel sheet. Rare earth oxide and hafnium oxide films were prepared by depositing alcoholic solutions of the rare earth or hafnium nitrate on the inside of a stainless steel sheet rolled into a cylinder, evaporating to dryness under heat lamps while rotating the cylinder, and subsequently heating the sheet strongly with a Meeker burner to decompose the nitrate. Samples of WO_3 and HgS were prepared by slurrying the powder in alcohol and spreading over the stainless steel sheet in the same manner.

In all of this work, samples which were "thin" in relation to the ranges of the alpha particles were used, so that the alpha groups would produce reasonably sharp peaks. "Thick" samples would give larger counting rates, but would form continuous rather than peaked spectra, and in the case of separated isotopes could not be afforded anyway. Film thicknesses mentioned in the following are simply the sample weights divided by the supporting areas, and were usually between 10 and 100 μg cm^{-2}. The actual effective thicknesses of the compounds were probably greater, but the spectra obtained indicate that clumping could not have been serious.

All the enriched isotope samples were obtained from the Isotopes Division of the Oak Ridge National Laboratory, Union Carbide Nuclear Company, Oak Ridge, Tennessee. Listed in Table I are their isotopic analyses.

IV. ALPHA-PARTICLE ENERGY DETERMINATION

An energy versus pulse-height calibration curve, Fig. 2, was obtained by placing various alpha emitters in the chamber and determining the pulse height corresponding to the various alpha energies. Table II gives the known energies.[27–32] Additional points were obtained below 3 Mev by measuring the alpha and triton peaks from the $Li^6(n,\alpha)H^3$ and $B^{10}(n,\alpha)Li^7$ reactions. For the latter measurements the counter was submerged in water, and a Po–Be neutron source of ~20 millicuries Po^{210} activity was placed alongside the counter. All of the points fell close to a straight line which intersected the energy axis at +158 kev, substantiating the findings Cranshaw and Harvey[33] and Hanna[34] that the ionization is linearly dependent on but not proportional to the energy. A least squares analysis of the data gave the following equation for the curve:

$$E=[(0.0830\ \text{volt}^{-1})\times V+0.158]\ \text{Mev}, \qquad (1)$$

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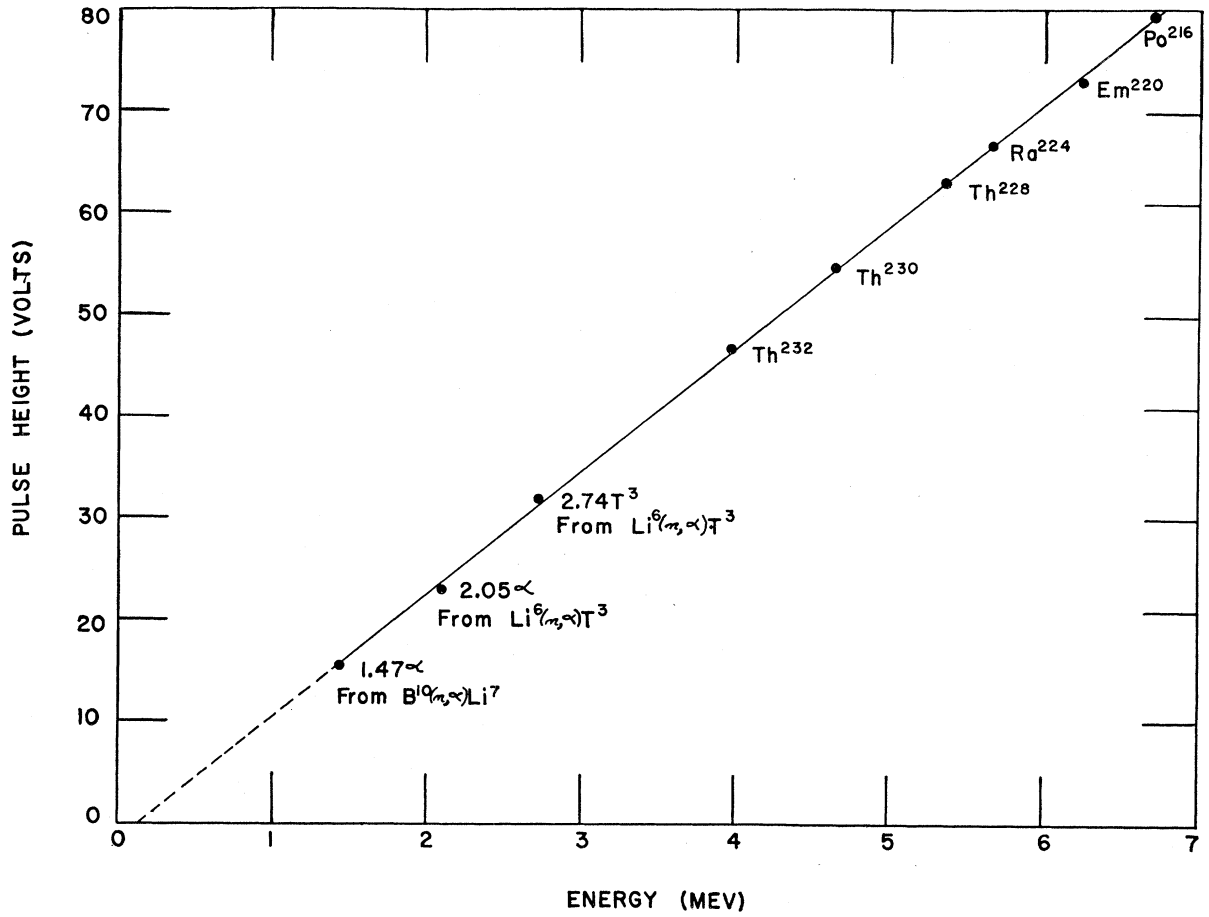

FIG. 2. Energy vs pulse-height calibration curve.

where E is the alpha-particle energy and V the pulse height. The energy intercept should be a constant depending only on the gas used. The slope of the curve, however, is dependent on the degree of pulse amplification. For each run, the slope of the line was determined by obtaining simultaneously the energy spectrum of an alpha emitter of known energy as well as the unknown. From Eq. (1) can be derived the relation:

$$E=E^*+(E_0-E^*)\times V/V_0, \qquad (2)$$

where E_0=alpha-particle energy of standard, V_0=pulse height of standard, E=alpha-particle energy of unknown, V=pulse height of unknown, and E^*=energy intercept=158 kev.

V. DETERMINATION OF SPECIFIC ACTIVITY

The gross alpha spectrum which was obtained from a sample had to be resolved into its components in order to determine the net counting rate due to the particular nuclide of interest. Contaminating activities such as Sm^{147} were resolved from the spectrum using as a guide the shape of the Sm^{147} alpha spectrum from a sample of comparable thickness. The alpha spectrum of the nuclide of interest was resolved from the gross spectrum by assuming it had the same shape. For the rare-earth alpha emitters, whose energies lie close to that of Sm^{147}, the effective counting yield was obtained by determining the counting rate of a known amount of Sm^{147} under the same counting conditions with the

TABLE II. Particle energies used in energy calibration.

Emitter	Particle energy (Mev)	Branching fraction	Weighted average energy for peak (Mev)	Reference
Po^{216}	6.77	1.00	6.77	27
Em^{220}	6.28	1.00	6.28	27, 28
Ra^{224}	5.68	0.95	5.68	27, 28, 29
Th^{228}	5.42 5.34	0.72 0.28	5.40	27, 29
Th^{230}	4.68 4.61	0.76 0.24	4.67	30
Th^{232}	4.01 3.95	0.76 0.24	4.00	31
$Li^6(n,\alpha)H^3$:α	2.05	1.00	2.05	32
:H^3	2.74	1.00	2.74	
$B^{10}(n,\alpha)Li^7$:α	1.47	0.96	1.47	

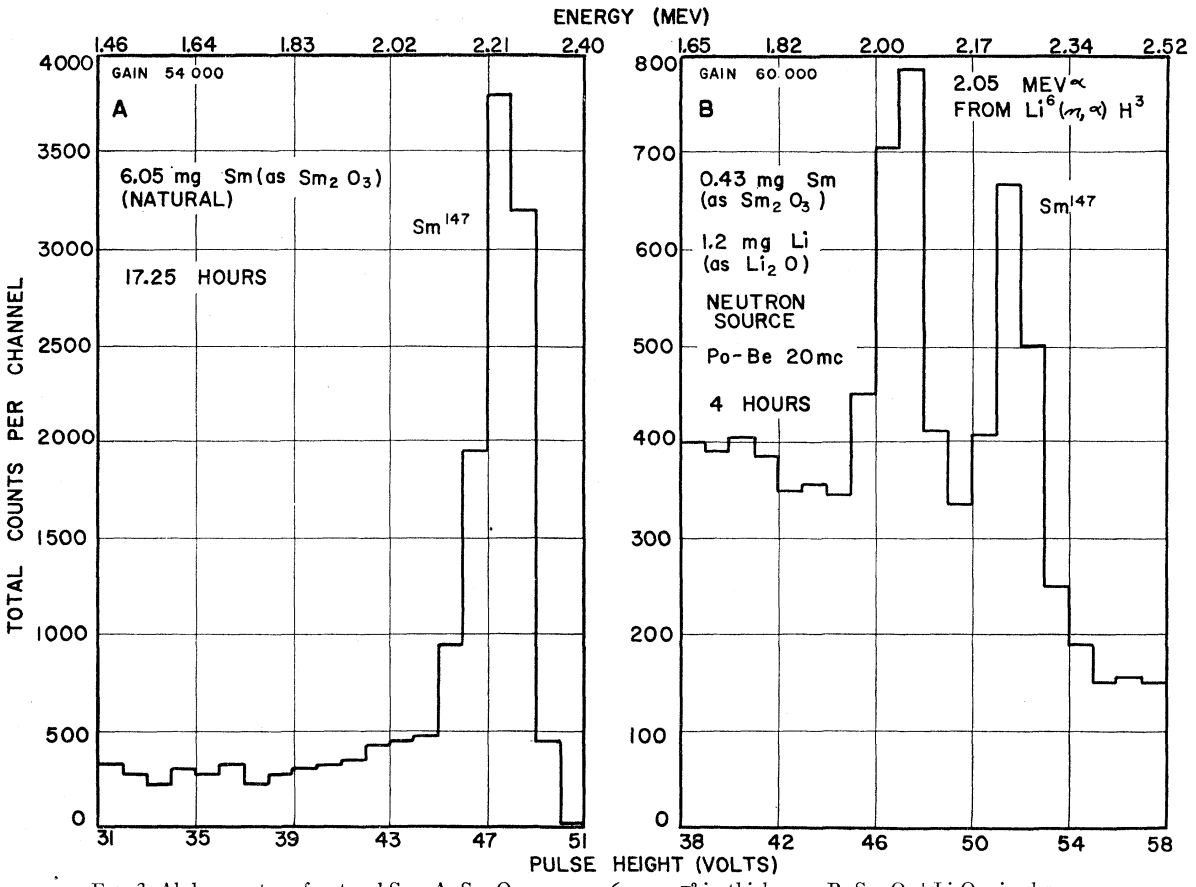

FIG. 3. Alpha spectra of natural Sm: A, Sm_2O_3 source $\sim 6\ \mu g\ cm^{-2}$ in thickness; B, $Sm_2O_3+Li_2O$ mixed source $<2\ \mu g\ cm^{-2}$ in thickness with neutron source.

same low-energy cutoff as the spectrum of the unknown sample.

For Sm^{147} and Pt^{190}, the counting rates were high enough so that an extrapolation of the energy spectrum to zero energy was possible. The counting yields of these activities were obtained by assuming 2π geometry and correcting for self-absorption, back scattering, and chemical yield. The data of Gobeli[35] were used to obtain the range-energy relationship for low-energy alpha particles.

In cases where no activity was found, upper limits for specific activity were obtained by estimating the level of activity which would have given a significant peak of the expected shape in the spectrum, usually several times the standard deviation of the average counting rate per channel.

TABLE III. Summary of results.

Nuclide	Alpha-particle energy (Mev)	Specific activity of natural element (dis sec^{-1} g^{-1})	Half-life or limit for nuclide (year)
Ce^{142}	···	<0.0002	$>5\times10^{16}$
Nd^{144}	1.83±0.03	0.0093±0.0011	$(2.4\pm0.3)\times10^{15}$
Sm^{146}	···	<0.013	$<3\times10^{8}$
Sm^{147}	2.23±0.02	116±5	$(1.15\pm0.05)\times10^{11}$
Sm^{148}	···	<0.04	$>2\times10^{14}$
Sm^{149}	···	<0.01	$>1\times10^{15}$
Gd^{152}	2.14±0.03	0.00156±0.00010	$(1.08\pm0.08)\times10^{14}$
Hf^{174}	2.50±0.03	0.000065±0.000013	$(2.0\pm0.4)\times10^{15}$
W^{180}	···	<0.00013	$>9\times10^{14}$
Pt^{190}	3.11±0.03	0.012±0.001	$(6.9\pm0.5)\times10^{11}$
Hg^{196}	···	<0.0007	$>1\times10^{14}$

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VI. RESULTS

The alpha energies, specific activities, and half-lives of the various nuclides derived in this work, together with lower limits for the half-lives of those nuclides for which no activity was observed, are summarized in Table III.

Sm^{147}

Figure 3(A) represents the alpha spectrum of a 6-mg sample of natural samarium (as Sm_2O_3), used for calculation of the specific activity and half-life of Sm^{147}. Independent measurements of the specific activity and half-life were also made from the samarium samples enriched in Sm^{148} and Sm^{149}. The tabulated values are weighted means of the three measurements.

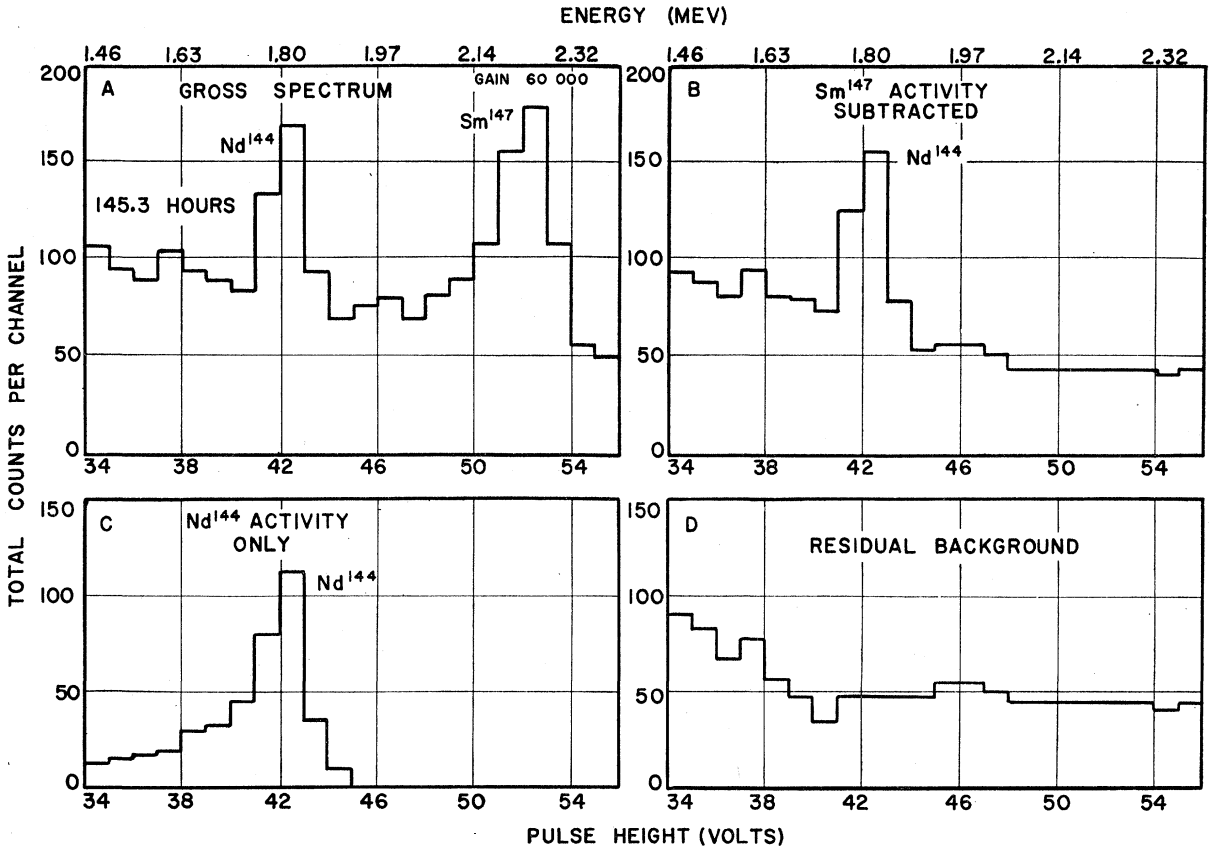

FIG. 4. Alpha spectrum of enriched Nd^{144} sample (A) and its resolution into components (B,C,D).

The alpha-particle energy of Sm^{147} was measured by comparison with the 2.05-Mev[32] alpha arising from the $Li^6(n,\alpha)H^3$ reaction. Figure 3(B) is the spectrum of the Sm^{147} alpha and the comparison alpha from a very thin mixed Sm-Li source in the presence of a thermal neutron flux.

Nd^{144}

Figure 4 is the alpha spectrum which was obtained from 82.8 mg of Nd_2O_3 enriched in Nd^{144}. The sample thickness was 0.069 mg cm^{-2}. The small samarium impurity present served as a standard for the Nd^{144} alpha energy determination.

Gd^{152}

Figure 5(A) is the alpha spectrum obtained from Gd_2O_3 enriched in Gd^{152}. An alpha peak was observed at 2.14 Mev. In order to determine whether this was due to Gd^{152} or Sm^{147} impurity, a sample of Sm_2O_3 was put in the counter with the gadolinium and a second spectrum recorded. Auxiliary experiments showed that the response of the counter was the same for the large Gd_2O_3 and the small Sm_2O_3 sample positions. Two peaks were observed, Fig. 5(B), indicating that at least the major part of the activity observed in Fig. 5(A) was not due to Sm^{147}. The specific activity and half-life were determined from Fig. 5(A) and the alpha-particle energy from Fig. 5(B) using Sm^{147} as a standard.

Hf^{174}

Spectra obtained from HfO_2 enriched in Hf^{174} are shown in Fig. 6. In the first experiment the thickness of the deposit was 0.10 mg cm^{-2} and a weak Po^{210} standard was added. A small peak was observed at 2.50 Mev. In a second experiment, some of the same HfO_2 sample was mixed with a small amount of Sm_2O_3. This run yielded the alpha-particle energy more precisely, using Sm^{147} as the standard.

Pt^{190}

In the spectrum of natural Pt [Fig. 7(A), 1.60 mg cm^{-2}] a distinct peak was observed at 3.1 Mev and another at 5.3 Mev, the latter presumably due to Po^{210} contamination. In a spectrum [Fig. 7(B)] obtained with enriched Pt^{190} (0.045 mg cm^{-2}), the specific activity and half-life calculated for Pt^{190} were in good agreement with the values calculated from the natural Pt, proving that the observed activity was due to Pt^{190}. Because of the better counting statistics obtained with the enriched

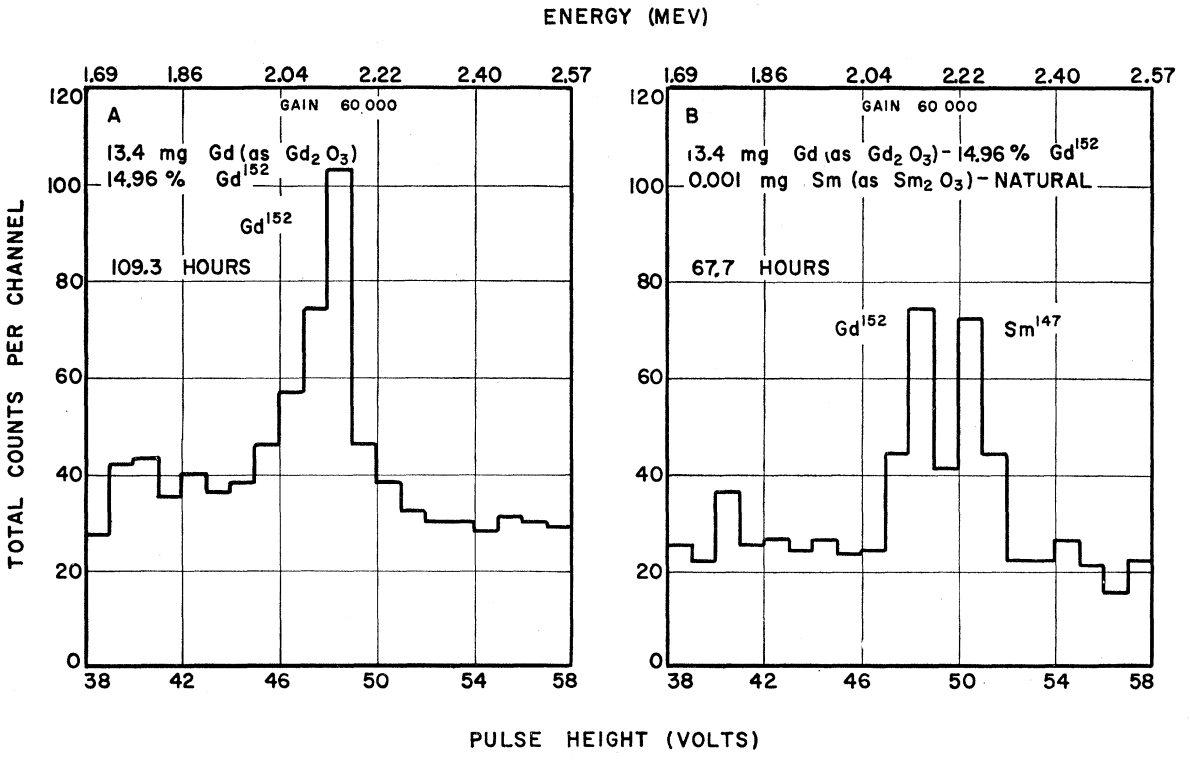

FIG. 5. Alpha spectra of enriched Gd^{152} sample: A, as received from supplier; B, with added Sm_2O_3 source.

Pt^{190} sample, the results obtained were weighted a factor of 3 relative to the natural Pt^{190} results in obtaining an average value of the specific activity and half-life.

The alpha-particle energy of Pt^{190} was determined by comparison with the alpha of Sm^{147} [Fig. 7(C) and Th^{232} [Fig. 7(D)].

Ce^{142}

Figure 8 shows the spectrum from CeO_2 enriched in Ce^{142}. The sample thickness was 0.048 mg cm^{-2}. A small amount of samarium was mixed with the sample to provide an energy calibration. No peak was found that could be attributed to a Ce^{142} activity. The level of activity expected based on the claim of Riezler and Kauw[13] is indicated in the figure.

Sm^{146}

A sample of natural Sm_2O_3 was counted for seven days for the purpose of looking for alpha particles which might arise from Sm^{146}. Figure 9 is a spectrum of the pulses obtained. No group was observed which could be attributed to Sm^{146} alpha activity, the energy of which is given as 2.55 ± 0.03 Mev from artificial production.[19,36] The upper half-life limit in Table III was calculated assuming that nucleosynthesis occurred continuously from $\sim12\times10^9$ yr ago to $\sim4.7\times10^9$ yr ago.[37,38]

Sm^{148}

A 19.6-mg sample of Sm_2O_3 enriched with Sm^{148} to 83.1% was counted for 44.3 hours. The sample also contained 6.4% Sm^{147}. No peak was observed in the sample which could be attributed to Sm^{148} alpha activity, which should have a somewhat lower energy than Sm^{147}.

Sm^{149}

A 30.8-mg sample of Sm_2O_3 enriched in Sm^{149} to 88.8% was counted for 78 hours. The isotopic fraction of Sm^{147} in the sample was 1.0%. No peak was observed in this sample which could be attributed to Sm^{149} alpha activity, which should also have a lower energy than Sm^{147}.

W^{180}

In the measurement of enriched W^{180} (sample thickness 0.083 mg cm^{-2}) a weak source of Po^{210} was also present. In the spectrum (Fig. 10) no peak was observed which could be assigned to tungsten alpha activity. The expected level based on the Porschen and Riezler[10] value of the specific activity and the assumption that W^{180} is the active isotope is also indicated in the figure.

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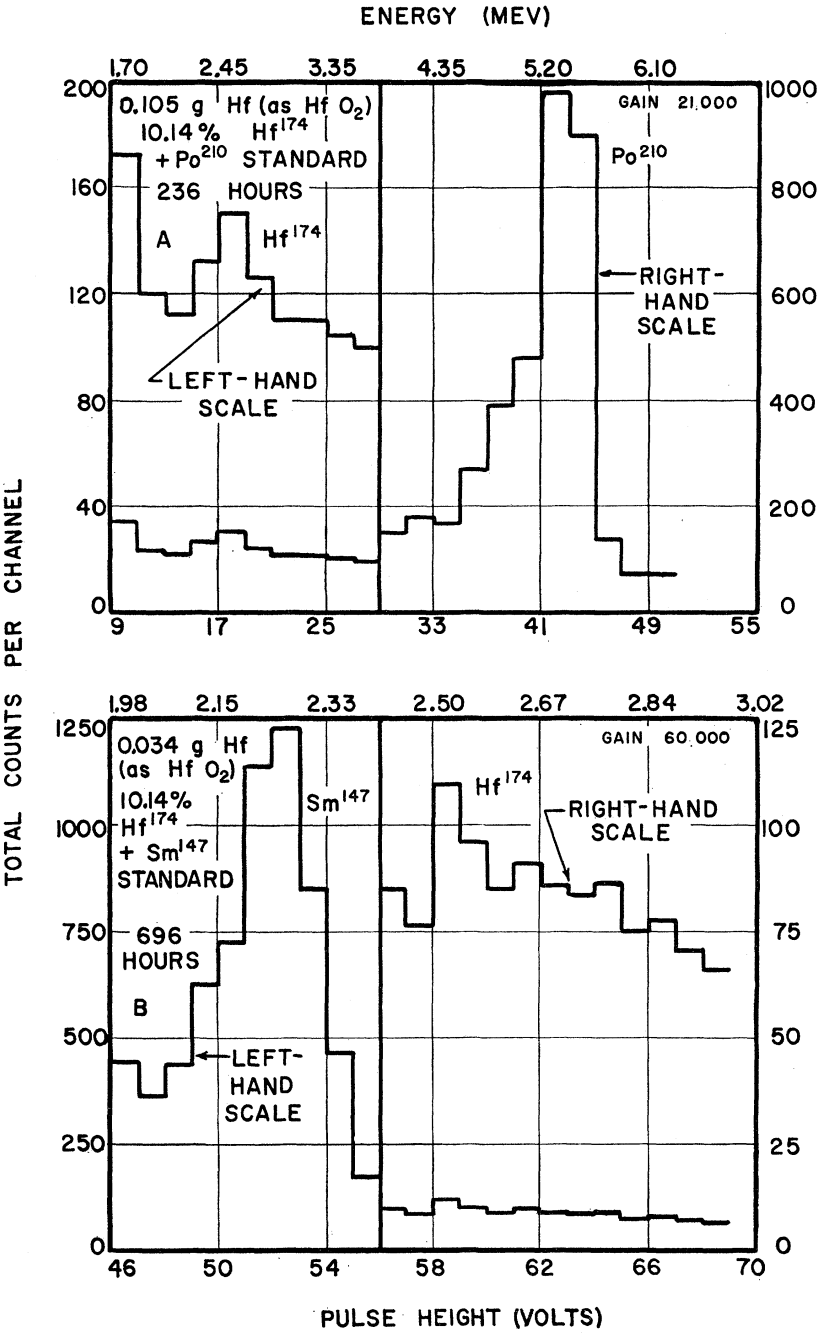

FIG. 6. Alpha spectra of enriched Hf^{174} sample: A, with added Po^{210} source; B, with admixture of Sm_2O_3.

Hg^{196}

A 121.4-mg sample of HgS enriched in Hg^{196} to 1.46% was counted for 23.9 hours. No activity was observed which could be attributed to Hg^{196}.

VIII. DISCUSSION OF RESULTS

The alpha energies determined in this work have been included in a calculation by Rasmussen[39,40] of theoretical alpha half-lives and reduced-width probabilities of a number of medium-heavy alpha emitters. The theoretical half-lives are generally in good agreement with these obtained experimentally in this work.

In Table IV are summarized the results of other experimenters[1,4,9–13,15,41–68] on the elements which we have studied.

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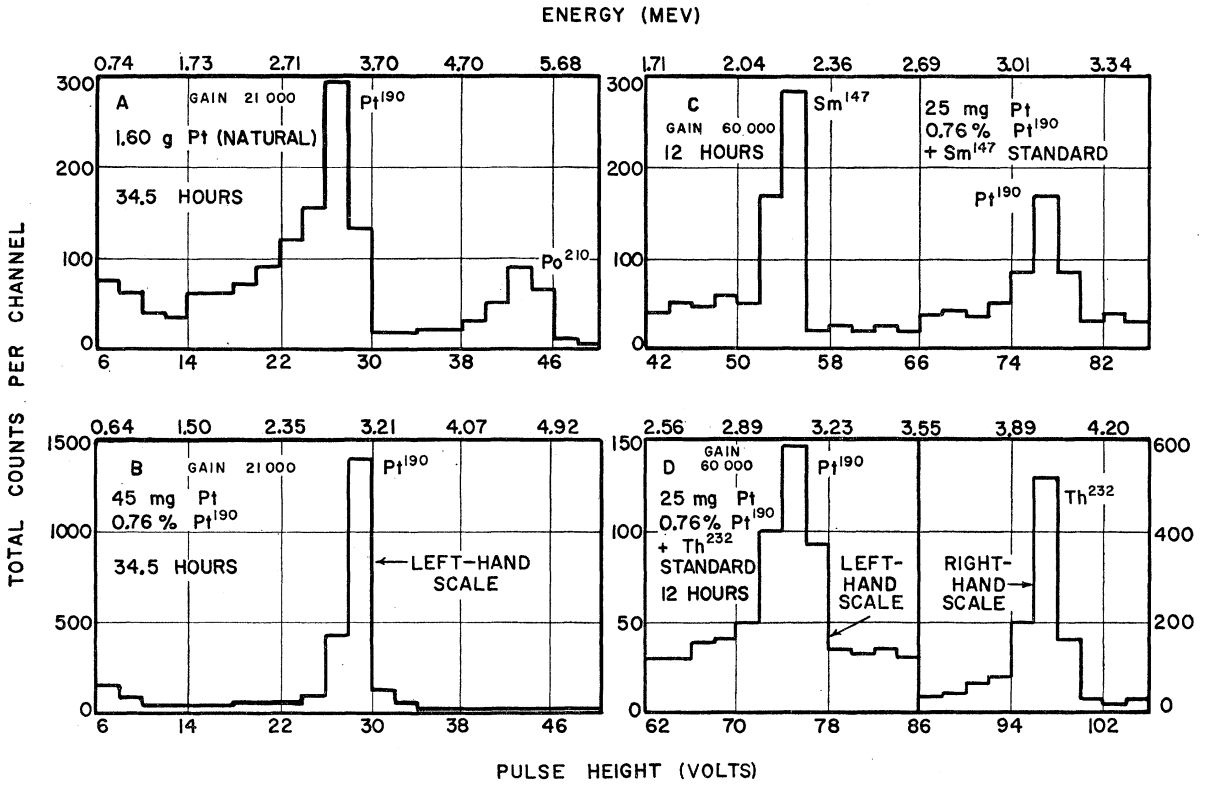

FIG. 7. Alpha spectra of platinum: A, natural platinum; B, enriched Pt^{190}; C and D, enriched Pt^{190} with energy standards.

Our Nd^{144} results are in general agreement with the nuclear emulsion results and with a recent determination using a slow ionization chamber-electrometer counter.[44] However, the value of the half-life reported by Kelly and Beard[43] using a liquid scintillator is lower by a factor of ~ 100, suggesting the likelihood of Sm^{147} contamination.

The Pt^{190} and Hf^{174} results are in reasonable agreement with those obtained by Hoffman,[1] Porschen and Riezler,[12,9] and Riezler and Kauw,[15] though the new values are considerably more precise.

The observed alpha-particle energy of Sm^{147} is somewhat higher than the most precise previous ionization chamber measurement, that of Jesse and Sadauskis,[61] 2.18 Mev, which was corrected to 2.21 by Rasmussen, Thompson, and Ghiorso[4] on the basis of nonproportionality of ionization. Very recently, Vorob'ev, Komar, Korolev, and Solyakin[66] have reported an ionization chamber value of 2.19 ± 0.01 Mev, assuming ionization proportionality. In both of these cases only relatively high-energy alphas (Po^{210} and U^{234}, respectively) were used as standards. Very recently, however, Karras and Nurmia[45] have described an ionization chamber deter-

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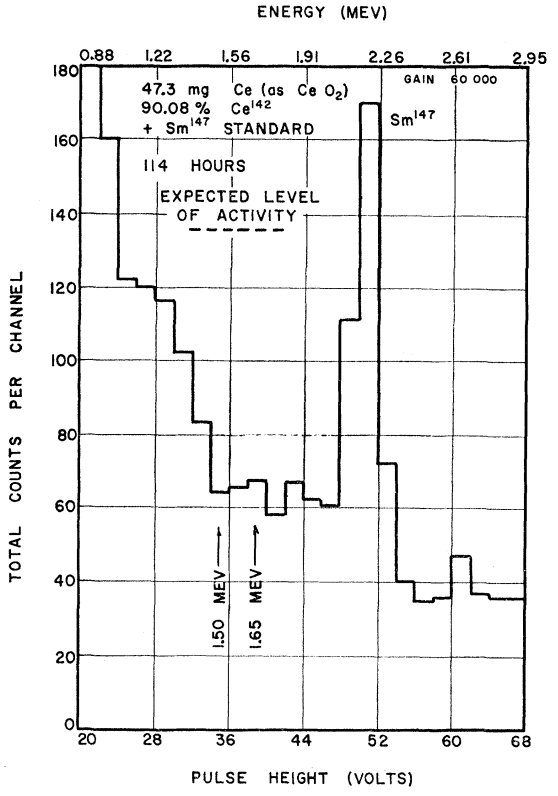

FIG. 8. Alpha spectrum of enriched Ce^{142}.

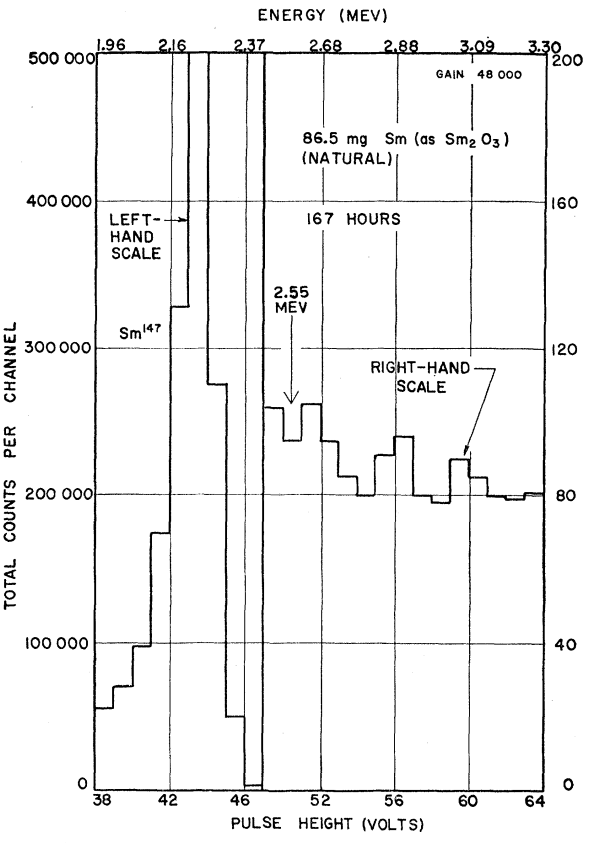

FIG. 9. Alpha spectrum of natural Sm (1.91–3.30 Mev) obtained in search for Sm^{148} activity.

mination using B+n and natural uranium alphas for calibration. They observed the weak 1.80-Mev group from B^{11*} as well as the dominant 1.47-Mev group, and derived for Sm^{147} an alpha energy of 2.20±0.03 Mev. Our value, 2.23±0.02 Mev, was obtained by use of an even closer standard (2.05 Mev) and an experimental ionization-energy relationship. This higher alpha energy removes the anomaly of an excessively long theoretical alpha half-life compared to the actual.[69] The same applies to Eu^{147}, for which the energy calibration depended on Jesse and Sadauskis value for Sm^{147}. The Eu^{147} alpha disintegration energy (electron-screening corrected) is changed from 2.98 to 3.01 Mev, resulting in better agreement between the experimental and theoretical alpha half-life.

The Gd^{152} results are significantly different from those reported by Riezler and Kauw.[15] They deduced the alpha energy to be 1.7 Mev and computed a half-life 9.5×10^{14} years, assuming about one-third of their alphas to be due to Sm^{147}. Their alpha energy is definitely low.[70] Even their own high value of the half-life requires theoretically a particle energy of at least 2.1 Mev; this (as confirmed by our ionization measurement) indicates that the Gd^{152} alpha particles could hardly be resolved or distinguished from those of Sm^{147} in nuclear emulsions. The enriched Gd^{152} samples used by Riezler and

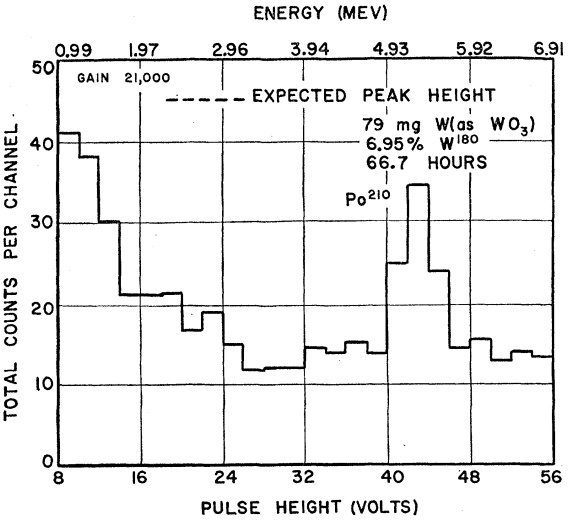

FIG. 10. Alpha spectrum of enriched W^{180} sample.

[69] J. O. Rasmussen, Ph.D. thesis, University of California Radiation Laboratory Report UCRL-1473 Rev., 1952 (unpublished).

[70] *Note added in proof.* Support for our value of the Gd^{152} α-particle energy comes from recently determined atomic masses in the rare-earth region [V. B. Bhanot, W. H. Johnson, Jr., and A. D. Nier, Phys. Rev. **120**, 235 (1960)], from which can be calculated a value of 2.18±0.18 Mev.

TABLE IV. Other results on elements studied in this work.[a]

Nuclide	Reference	Method	E_α (Mev)	Specific activity of natural element (dis sec^{-1} g^{-1})	Half-life or limit for nuclide (year)
Ce^{142}	9	N	...	[<0.002]	[>4×10^{15}]
	13	N, E	1.5	[0.002]	5.1×10^{15}
	41	N, E	...	[<0.001]	>[1×]10^{16}
Nd^{144}	42	C	...	<0.02	[>1×10^{15}]
	11	N	1.9 ±0.1	0.015	1.5×10^{15}
	12, 9	N	1.8 ±0.1	[0.005]	5×10^{15}
	9	N, E	...	[0.012]	2.2×10^{15}
	43	L	...	[1]	2×10^{13}
	44	P, E	...	[0.011)	2.1×10^{15}
	45	P	2.0 ±0.1	...	...
Sm^{147}	46	C, G	[2.3(V)], [2.23±0.07(R)]	75	[1.8×10^{11}]
	47	W, G	[$\geqslant$2.8(R)]	...	...
	48	I, G	[2.7], [3.0(R)]	67	[2.0×10^{11}]
	49	P	[2.18(R)]	...	...
	50	P	α:[2.28(R)]	$89\alpha+33p$	...
	51	P	2.46	[~70]	[~1.9×10^{11}]
	52	C, G	[2.39±0.08(R)]	[145±9]	[(0.92±0.06)×10^{11}]
	53	N	[2.23±0.03(R)]	...	...
	54	C	...	88[±5]	...
	55	C, G	[2.23±0.03(R)]	89±5	[(1.49±0.08)×10^{11}]
	56	I, G	[2.26(R)]	...	...
	57	C, G	[2.25±0.07(R)]	...	...
	58	N	[2.21±0.05(R)]	[76±6]	[(1.7 ±0.14)×10^{11}]
	59	N	...	133±6	[(1.00±0.05)×10^{11}]
	60	N	[2.23±0.07(R)]	...	...
	61	P	2.18[±0.02]	...	...
	4	X	2.21[±0.02]	...	...
	62	N	2.12 ±0.03	...	...
	9	N	2.12 ±0.03	...	...
	63	C, E	...	[106±5]	(1.25±0.06)×10^{11}
	64	N	2.18 ±0.14	[114±3]	(1.15±0.03)×10^{11}
	65	L	...	[105±3]	(1.28±0.04)×10^{11}
	66	P	2.19±0.01	...	...
	45	P	2.20±0.03	[117±5]	(1.14±0.05)×10^{11}
Gd^{152}	67	N	...	[<0.011]	[>1.6×10^{13}]
	9	N	...	[<0.002]	[>8×10^{13}]
	15	N, E	1.7	[0.00018]	9.5×10^{14}
Hf^{174}	9	N	...	[<0.0002]	[>5×10^{14}]
	15	N, E	2.5	[0.00003]	4.3×10^{15}
W^{180}	10	N	3.2 ±0.2	[0.0003]	[3×10^{14}]
	9	N	3.0 ±0.2	[0.0003]	[3×10^{14}]
	68	S	...	[<0.00011]	[>1.1×10^{15}]
Pt^{190}	1	P	[~3]	[~0.02]	[~5×10^{11}]
	12, 9	N	3.3 ±0.2	[0.009]	[1.0×10^{12}]
Hg^{196}	9	N	...	[<10]	[>1×10^{10}]

[a] [ ]=enclosed number not given in publication cited, but derived from information given therein; C=Geiger counter, or proportional counter used only for counting; E=use of isotopically enriched sample; G=range in gas measured; I=current ionization chamber; L=liquid scintillation counter with internal sample; N=nuclear emulsion; P=ionization chamber with pulse amplitude analysis; R=range in air given by authors converted to energy using range-energy relation of Bethe [H. A. Bethe, Revs. Modern Phys. **22**, 213 (1960)]; S=solid scintillation counter with internal sample; V=velocity of particles cited by authors used to calculate energy; W=Wilson cloud chamber; X=correction of measurement of preceding entry for nonproportionality in ionization versus energy.

Kauw and by us were doubtless portions of the same Oak Ridge batch. The samarium content was so low that *no* Sm^{147} peak was visible in our spectrum. Further, their background plates showed no significant activity in the region between 1 and 2.5 Mev. Probably, then, *all* of the tracks in this region in their gadolinium spectrum were due to Gd^{152}. The spectrum broadening and low apparent specific activity might both have resulted from excessive fading in this plate, so that their claim to have distinguished the gadolinium alphas from those of samarium was unjustified. The ability of the ionization counter to resolve the Gd^{152} and Sm^{147} alphas simultaneously present was thus essential to the positive identification of the Gd^{152} activity.

Our inability to find alpha activity in Ce^{142} is more difficult to reconcile with the positive result of Riezler and Kauw.[13] Recently, Senftle, Stern, and Alekna[41] published a negative result for Ce^{142} alpha activity, but their lower limit for the half-life is within the uncertainty of Riezler and Kauw's measurement. Our limit is a factor of 10 higher than the latter's value.

Concerning natural alpha activity of tungsten, it would appear from our negative result using enriched W^{180} that the activity observed by Porschen and

Riezler[10] would have to be due to some other isotope.[71] However, Beard and Kelly[68] have recently attempted a confirmation using natural $CdWO_4$ crystals as an alpha-sensitive scintillator. They were unable to find any activity, and set an upper limit to the specific activity corresponding to one third of the level reported by Porschen and Riezler. This limit is the same as that calculated for natural tungsten from our enriched W^{180} result.

Vorob'ev *et al.*[66] observed in a spectrum of natural samarium a slight excess at $\sim$2.55 Mev, the energy of Sm^{146} alphas, but did not consider it statistically significant. They considered that the Sm^{146} alpha activity could be at most 3×10^{-4} that of Sm^{147}. Nurmia and Karras[72] have reinterpreted the results of Vorob'ev *et al.* to constitute a positive discovery of Sm^{146} with an alpha activity of 5×10^{-4} that of Sm^{147} in the natural element. However, our results indicate that the Sm^{146} activity cannot be more than 1.2×10^{-4} that of Sm^{147}, so that interpretation must be rejected.[73] Recent results[36] on artificially produced Sm^{146} tend to confirm the half-life value initially obtained by Dunlavey and Seaborg,[19] which is definitely too short for survival from the cessation of nucleosynthesis to the present.

The ionization counter technique as developed by us does not have as high a sensitivity as the nuclear emulsion technique. This is especially true when only limited amounts of isotopically enriched samples are available. However, large counters can accommodate $\sim$1-g samples as compared with $\sim$1-mg samples in emulsions, so that better counting statistics result and more accurate specific activities can be obtained. The counting times are much shorter, and the analysis of the results can be made more quickly and with less tedium. The chief advantage of the ionization technique, however, is its considerably ($\sim$5-fold) greater energy resolution. This means that energies can be obtained with considerably greater precision and that alpha groups of similar energies can be more easily resolved from each other.

ACKNOWLEDGMENTS

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[71] *Note added in proof.* Recent atomic masses of W^{180} and Hf^{178} [V. B. Bhanot, W. H. Johnson, Jr., and A. O. Nier, Phys. Rev. **120**, 235 (1960)] yield for W^{180} a very low α-particle energy, 2.06 ± 0.15 Mev, corresponding to a half-life greater than 10^{22} years.

[72] M. Nurmia and M. Karras, Geophysica **7**, 83 (1960).

[73] *Note added in proof.* An even lower limit for Sm^{146} activity in natural samarium has subsequently been obtained [R. D. Macfarlane, Nature **188**, 1180 (1960)].