Ionization Yields for Fission Fragments*

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The ratio of fission fragment-to-alpha particle ionization produced in total stopping has been measured in the gases helium, argon, and a helium plus 0.25% argon mixture. A parallel-plate gridded electron collection chamber was used. Gas purity and electron loss were checked by making absolute average energy per ion pair measurements for alpha particles. These agreed with the values reported for total ion collection. The difference in ionization defects between helium and argon is consistent with the value predicted by Knipp and Ling on the basis of the greater importance of elastic atomic collisions in argon. Because adding a small amount of argon to helium enables the metastable helium atoms to produce ionization, the difference in the fission fragment-to-alpha particle ionization ratios between helium and the helium plus argon mixture shows that the ionization-to-excitation ratio in helium is lower for fission fragments than for alpha particles. Arguments are given which indicate that very little "defect" is present for fission fragments in the helium plus argon mixture.

I. INTRODUCTION

EARLY measurements of the fission mass and energy distributions were made using ionization chambers under the assumption that the average energy lost in producing an ion pair in the inert gases is the same for fission fragments as for alpha particles. These measurements have been shown to be inconsistent with results obtained by radiochemical, calorimetric, and direct velocity measurements. The discrepancies are discussed in detail elsewhere.[1,2] The results indicate that the ionization chamber values are in error, and specifically that an "ionization defect" for fission fragments is present due to a decreased ionizing efficiency for fission fragments relative to alpha particles. Because the average energy per ion pair for fast ($v \gg e^2/\hbar$) alpha particles, protons, and electrons has been shown[3–5] to be the same, it is of interest to explore this "defect" for fission fragments.

A theoretical treatment[6] was successful in predicting the order of magnitude of the defect, Δ, which was defined by

$$E_f = w^e I_f + \Delta, \tag{1}$$

where E_f is the fission fragment energy, w^e the differential energy per ion pair for a fast fission fragment, and I_f the ionization produced in completely stopping the fragment. This treatment assumed that w^e is the same for fast fission fragments as for fast alpha particles. The value for Δ in argon was then calculated on the basis of elastic atomic collisions, which become important as the fragment slows down. Direct measurement[7] of the energy-ionization relation for fission fragments stopped in argon has indeed verified a linear relationship in the high-energy region. The slope w^e was found to be close to that for alpha particles, and Δ is consistent with that calculated.

This theoretical treatment[6] also suggested that the magnitude of Δ for helium should be much smaller than for argon and, therefore, that the difference in defects between helium and argon should be a good measure of the argon defect. However, an attempt[2] to experimentally show a difference in ionization defects between helium and argon gave negative results. It was suggested that this might be explained if there were an unexpected decrease in the ionization-to-excitation ratio for fission fragments relative to alpha particles in helium. Such a decrease could be shown by employing the metastable electronic states of helium. It has been shown[8] that the amount of ionization produced in stopping a fast particle in helium to which a few tenths percent argon have been added is 40% greater than the amount produced in pure helium. This is due to the discharge of the $2\,^1S$ and $2\,^3S$ helium metastable atoms in ionizing collisions with argon atoms. If the ionization-to-excitation ratio in helium is lower for fission fragments than for alpha particles, the ratio of fission-to-alpha ionization, R, will be greater in a helium plus argon mixture than in pure helium, because proportionately more ionization will be added to the fragment ionization than to the alpha ionization by the discharge of the metastable atoms. The present experiment was undertaken to measure R in helium and a helium plus argon mixture and in pure argon to check the previous results.[2]

II. EQUIPMENT AND PROCEDURE

The basic quantity measured was the ratio of fission fragment ionization-to-alpha particle ionization produced in completely stopping a particle ($R = I_f/I_\alpha$). The difference in ionization defects between two gases

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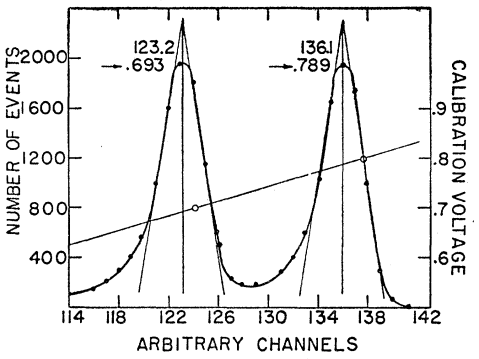

FIG. 1. Alpha particle pulse-height spectrum.

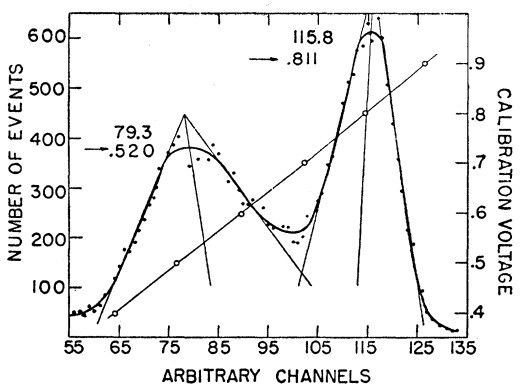

FIG. 2. Fission fragment pulse-height spectrum.

is simply the difference in R multiplied by the alpha energy E_α, because E_f is the same in each gas, and the differential energy per ion pair is very nearly equal to the average energy per ion pair in the case of alpha particles.[3] The alpha-particle energy is known from independent measurements.[9]

The ratio R was measured in the gases helium, argon, and a helium plus 0.25% argon mixture. Standard grade helium and argon having 99.99% and 99.9% purity, respectively, were used. The gas fillings were continuously purified by a calcium (90%)-magnesium purifier maintained at 470°C. The best indication of purity is the value obtained for the absolute energy per ion pair in helium. It has been shown[8] that this value is exceedingly dependent on the purity of the helium, and the value obtained here indicates a purity better than three parts in 10^5. The pressures employed were 2260, 1000, and 2260 mm Hg for the He, A, and He+A.

The fission fragments were obtained by fissioning natural uranium by neutrons from the reaction $D(d,n)He^3$ (approximate energy 3.2 Mev) with the Iowa State University kevatron. The fission rate was about 1 per second. The source was uranium oxide 17 micrograms uranium per cm^2 thick. No collimation of particles from the source was employed.[10] It should be pointed out that energy loss due to source thickness may be neglected in the present experiment because the losses were small and the source was the same for all the gases. Alpha particles from the U^{234} and U^{238} in the source were used to determine the alpha ionization.

A gridded parallel-plate electron collection chamber was used for these measurements. Except for minor electrode modifications made to improve the breakdown characteristics and field uniformity, the chamber was the same as previously described.[11] The electronic circuits and pulse-sorting equipment are also discussed there. Equal integrating and differentiating time constants were employed in the amplifier to minimize risetime effects.[12] Pulse sorting was done as before with channel widths about 1%. Figures 1 and 2 show typical pulse-height distributions obtained. The fission distribution is very similar to that reported[13] for U^{238}. The relatively poor resolution of the alpha particles was caused by chamber grid microphonics and high input capacitance, both due to the large chamber dimensions, as well as grid-shielding inefficiency effects to be discussed later. These figures also show the calibration points used in reducing the arbitrary channels to voltage pulse heights. The calibration pulses were applied to the source electrode and appeared on the collector because of the interelectrode capacitance. In this way pulses similar to the particle pulses were produced, with the advantage that the amplifier level and time constants were not disturbed. Fission pulses were a factor of 20 greater than the alpha pulses, and therefore, it was necessary to carefully verify the linearity of the pulse generator.

The maxima of the pulse-height distributions, corresponding to the most probable light and heavy fragments, were found by two methods. One consisted of drawing tangents to the peaks through the inflection points. A geometric median was then drawn and the maximum of the peak taken as the intersection of the median with the cap of the peak. This construction is shown in Fig. 2. The second method consisted of finding a mathematical median for each peak, using a consistent method for dividing the peaks. Results obtained by these two methods are equal to within the random statistical uncertainties, and the final results are an average of the two methods. The larger of the uncertainties in the two methods is retained. The first method was used to determine the alpha-particle ionization as shown in Fig. 1.

Sometimes in ionization measurements it is not necessary to be concerned with errors of a few percent. In such cases with gridded electron-collection chambers it is permissible to ignore the effects of inefficiency of the

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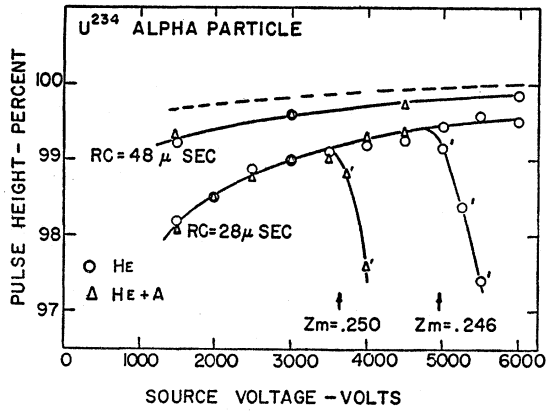

FIG. 3. Helium and helium plus argon saturation curves.

grid shielding, pulse risetime, and multiplication near the grid wires. However, in the present experiment the values for R vary only a few percent between gases, and it is the difference in these values that are of interest. It is therefore necessary to be concerned with errors as small as 0.1%, and these effects must be considered.

1. Grid Shielding

It was calculated from the geometry of the electrodes that the grid-shielding inefficiency[14] was 5.1%. Using this value and the assumptions that the field in the source region was uniform and that the electrons followed the lines of force, one calculates that the grid should have begun to collect electrons when the ratio of collector voltage-to-source voltage, z, dropped below 0.241 (grid at zero potential). Failure of either assumption would cause the experimental value to be higher. Figures 3 and 4 show the occurrence of this effect by the sudden drop in the curves. The primed points were taken at lower collector voltage. The point marked z_m is the point at which the pulse height had dropped 0.2%. The average of z_m for many such curves is given in Table I. Values obtained in argon plus 3% carbon dioxide are also included. It may be seen that z_m is definitely dependent on the gas used, an effect previously described[11] and attributed to electron diffusion. It is known that in A+CO_2 the electron diffusion is low, while in argon it is quite high. Table I shows that z_m in A+CO_2 is only slightly higher than the calculated value, confirming that both assumptions are sufficiently valid. Measurements were always made with $z>z_m$ to ensure that grid collection was not a factor.

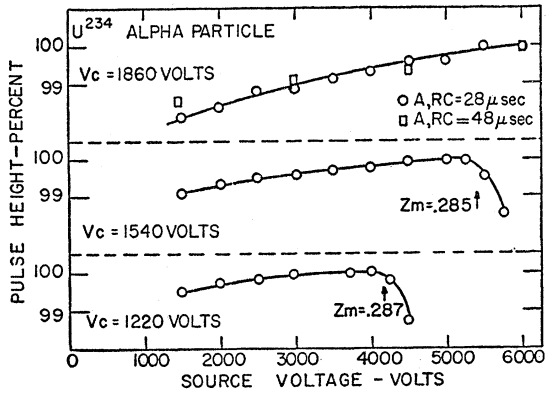

FIG. 4. Argon alpha saturation curves.

[14] Bunemann, Cranshaw, and Harvey, Can. J. Research A27, 191 (1949).

The value of the shielding inefficiency[14] was then used in computing the corrections to be applied because of the incomplete shielding of the positive ions from the collector. This correction involves the first power of the distance of the positive ions from the source electrode and the assumption of uniform field which was verified above. The corrections were found by averaging over all emission angles, and taking into consideration the nonuniformity of ionization along the fission and alpha tracks. Table II gives these corrections. Averaging over all angles is not quite valid without including source energy losses sustained at large angles, but it was shown that this error in the correction may be neglected in the present case. It was also shown that the small positive-ion motion occurring during the electron collection time may be neglected.

TABLE I. Average values for z_m.

Theoretical	0.241
A+CO_2	0.248±0.001
He+A	0.252±0.001
He	0.253±0.002
A	0.297±0.004

2. Pulse Risetime

Pulse risetime effects were minimized by a method[12] which uses equal integrating and differentiating time constants in the amplifier system. The risetime dependence of the present amplifier was found with a variable risetime pulser which produced pulses similar to the particle pulses. It was found that the risetime dependence followed the theoretical curve for an RC integrating-differentiating circuit with RC corresponding to 28 microseconds. The pulser was checked by showing that an actual 28-microsecond RC integrating-differentiating circuit produced the same dependence. The risetime of the particle pulses was computed using track length and drift velocity considerations. The risetime correction for an amplifier with 28-microsecond time constants was then averaged over all emission angles. Table II shows these corrections.

The risetime correction was checked by examining a pulse height *vs* source voltage curve as shown in Fig. 3. The upper curve was taken with an amplifier having 48-microsecond time constants, and the lower with 28

microseconds. The ordinates for the right end of the curves were fixed from the above calculations for the corresponding risetimes and time constants. The risetime was a function of the source field, and by applying the computed corrections to the experimental curves, the hatched line representing no risetime effect was determined. This hatched line fits both the 48- and 28-microsecond cases to within 0.1%. Because of the second power dependence in the risetime correction, it was possible to show that an error of 0.25% in fixing the right end ordinates would lead to an inconsistency in the left end. Furthermore, the congruence of the helium and helium plus argon curves indicates that the risetime was nearly equal for the two gases. This was confirmed by direct oscilloscope observation. Because the risetime in argon was much shorter, the difference between the 48- and 28-microsecond cases shown in Fig. 4 is not so marked.

3. Gas Multiplication

Some gas multiplication was present in the argon measurements. Although it is not expected that multiplication would occur at the average field-to-pressure ratios used here, it is not surprising in view of the strong fields existing around the grid wires. The pulse-height rose rapidly with increasing collector voltage after multiplication set in. Even for fixed collector voltage, the multiplication increased slightly with source voltage as seen in Fig. 4. The top curve was taken at 2.8% multiplication and the bottom curve with almost none, and a definite tendency for the curves to flatten with decreasing multiplication can be seen.

TABLE II. Corrections to the differences in Δ(%).

	Grid shielding	Risetime
$\Delta A-\Delta He$ (light)	−0.4	−0.1
$\Delta A-\Delta He$ (heavy)	−0.4	−0.2
$\Delta He-\Delta He+A$ (light)	0.0	0.0
$\Delta He-\Delta He+A$ (heavy)	0.0	0.0

The R measurements in argon were made with a multiplication of 2.8% so that high source field-to-pressure ratios could be attained without grid collection. In view of the strict proportionality obtained in proportional counters using high multiplication factors, it is not likely that this very small multiplication of 2.8% could be significantly different for fission fragments and alpha particles.

4. Saturation

As follows from the above discussion, the hatched line of Fig. 3 and the bottom curve of Fig. 4 show the saturation characteristics of the gases for alpha particles. Figures 5 and 6 show similar curves for fission fragments. These curves extend to field-to-pressure ratios about twice those previously used.[2] (A source voltage of 2000 here corresponds to x/p of 0.14 volt/cm mm Hg for He and He+A, and 0.31 for A.) The final results were determined from the averages indicated by the horizontal bars.

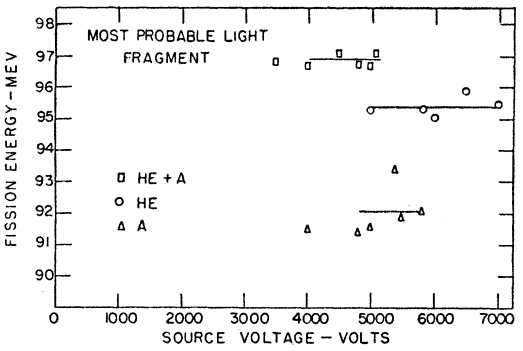

FIG. 5. Light fragment saturation curves, method II.

III. ABSOLUTE ENERGY PER ION PAIR MEASUREMENT

The assumption that the average energy per ion pair for fast particles is independent of the particle type is based for the most part on experiments done using total ion collection, while fission experiments have been done using electron collection chambers. Because of the possibility of a basic difference between the two methods, or simply a lack of complete electron collection in the second, the absolute average energy per ion pair for the alpha particles was measured for each gas used. This measurement entails basically the capacitance of the collector system and the voltage change produced by the collected electrons. In the present case the capacitance was found by a condenser substitution method where pulse heights were compared with and without a small shunt capacitance on the collector. The pulse-height ratio and the value of the added capacitance are sufficient to determine the capacitance. This method has the advantage that the capacitance is found under actual operating conditions since the pulses arise from the particles themselves. The effect of the added capacitance on the amplifier time constants

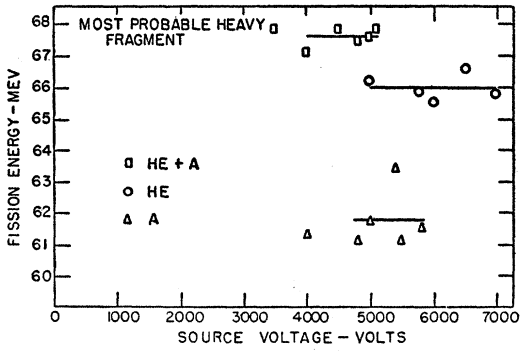

FIG. 6. Heavy fragment saturation curves, method II.

was considered. The potential change produced on the collector was found by comparison with known pulses from both the calibration pulser described before and an independent pulser used as a check. Care was taken to eliminate amplifier level changes and consider pulser coupling impedance effects. Table III shows estimates of the uncertainties (random plus systematic) as well as the magnitudes of the grid shielding, risetime, and multiplication corrections. The corrections and uncertainties for helium plus argon are essentially the same as for pure helium.

Previous work[15] indicated a difference between total ion collection and electron collection measurements but the present experiment shows satisfactory agreement (Table IV).

IV. DISCUSSION AND CONCLUSIONS

The agreement of the absolute average energy per ion pair for alpha particles in argon with that measured by total ion collection indicates that there is no basic difference in results obtained by electron collection as compared to total ion collection. The small difference in the values for helium is easily explained on the basis of the extreme sensitivity to gas purity in the helium case.

The difference in ionization defects between helium and argon is consistent with the theory of Knipp and Ling (Table V). The reason for the previous negative results[2] probably lies in saturation problems. Early measurements in the present experiment indicated that at the field-to-pressure ratios previously used the fission fragment saturation in argon is more complete than in helium.

The difference in ionization defects between helium and the helium plus argon mixture verifies that a small "defect" exists in helium due to an increase in the proportion of metastable states produced by fission fragments relative to alpha particles. This is equivalent to stating that the ionization-to-excitation ratio in helium is less for fission fragments than for alpha particles.

It is possible that this difference in the ionization-to-excitation ratio occurs in the low-velocity region. So long as the effect is confined to this region, it leads to a defect as defined in the introduction. The possibility is open that the ionization-to-excitation ratio is less for fission fragments than alpha particles over the entire range since the present experiment does not specify in which region the effect occurs.

It is not unlikely that part of the total defect in argon also is due to a decrease in the ionization-to-excitation ratio for fission fragments. This cannot be verified by the method discharging metastable states with added inert gases because their ionization potentials lie above the metastable states in argon. The close agreement of the difference in defects between helium and argon with that calculated on the basis of atomic collisions alone (Table V) suggests that a difference in the ionization-to-excitation ratio also exists in argon. The accuracy of the experiment and calculation do not justify such a conclusion however.

Finally, it appears that if the early measurements on fission fragments were repeated using the helium plus argon mixture as the stopping gas, the discrepancies discussed in the introduction would be eliminated. This may be seen if one considers the difference in defects between argon and the helium plus argon mixture. If there is no defect present in the mixture, the difference between argon and the mixture represents the defect in argon. It is seen from Table VI that this difference is indeed close to the value reported[7] for the defect in argon.

TABLE III. Corrections and uncertainties in the absolute energy per ion pair (%).

	Argon	Helium
Grid shielding	−0.5±0.2	−1.2±0.2
Risetime	−0.2±0.2	−0.5±0.3
Multiplication	0.5±0.2	0.0
Pulser coupling	0.2±0.2	0.2±0.2
Capacitance	±0.4	±0.4
Potential change	±0.7	±0.7
Total	0.0±0.9	−1.5±0.9

TABLE IV. Average energy per ion pair (ev/ion pair).

	This experiment	Jesse and Sadauskis[a,b]
A	26.7±0.3	26.4±0.1
He+A	29.5±0.3	29.6
He	41.0±0.4	42.7±0.2

[a] See reference 8.
[b] W. P. Jesse and J. Sadauskis, Phys. Rev. 90, 1120 (1953).

[15] L. O. Herwig and G. H. Miller, Phys. Rev. 94, 1183 (1954).

TABLE V. Differences in ionization defects, Mev (corrected).

	Light fragment	Heavy fragment
ΔA−ΔHe	2.5±0.6	3.9±0.5
ΔHe−ΔHe+A	1.6±0.3	1.3±0.5
ΔA (Knipp and Ling[6])	2.5	4.2

TABLE VI. Comparison of defects in argon, in Mev.

	This experiment (ΔA−ΔHe+A)	Schmitt[a] (Δ′A)
Light fragment (Mev)	4.1±0.7	5.1±0.8
Heavy fragment (Mev)	5.2±0.7	5.5±0.5

[a] See reference 7.

V. ACKNOWLEDGMENTS

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