Relative Ionization Yields for Fission Fragments in Various Gases*

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The ionizations resulting from U^{238} fission have been measured in He, Ne, A, Kr, N_2, A+3 percent CO_2, and A+5 percent N_2. Comparison of these values with those obtained for alpha particles in the same chamber fillings permit the calculation of ionization defect differences. It was found that about half of the 7-Mev defect previously ascribed to argon-CO_2 mixtures was due to CO_2. Nitrogen and krypton exhibited relatively large defects. No significant difference was observed between argon and helium. Several possible explanations for this are suggested and discussed.

I. INTRODUCTION

IN the experiment herein reported, the ionizations produced by U^{238} fission products in various gases and gas mixtures have been measured in a gridded ionization chamber. In each case, the ionizations from alpha particles were also measured. The data which are presented are the fission-alpha ionization ratios, which may be used as a means of comparing the fission fragment ionization efficiencies in the various gases. It might be expected that these ratios would depend on the stopping gas for reasons which follow.

It has been possible to obtain the mass distribution of fission products by several independent methods. Radiochemical analysis of these products has yielded such a distribution.[1] On the other hand, this distribution has been deduced from data taken with a double ionization chamber, in which simultaneous measurements were made on both fragments resulting from a single fission.[2,3] These two methods yielded curves which are significantly different. From the same double ionization chamber data it has been possible to infer a distribution in velocity. This is in significant disagreement with that obtained by direct measurement.[4] Furthermore, the total energy of fission has been measured calorimetrically[5] and is somewhat higher than the values obtained from the double ionization chamber data.

In the measurement of fission fragment energies the assumption had been made that the average energy required to make an ion pair in a gas would be the same for a fission fragment as for an alpha particle. On this basis, the value for the fission fragment energy was obtained by multiplying the energy of an alpha particle by the ratio of the ionizations produced by the fission and alpha particles. It was suggested[3,6] and later shown[4,7] that the discrepancies just pointed out could be at least partially removed if the average energy per ion pair were assumed to depend on the mass and velocity of the particle. In particular, the assumption was made that in argon about 6 and 7 Mev of the average light and heavy fragment energies respectively did not show up as ionization due to the changing ionization efficiency. A theoretical treatment was successful in predicting the order of magnitude of the undetected energies.[8] The basis of this prediction was the atomic collision, in which the ionizing particle interacts with the entire stopping atom, rather than with a single atomic electron. The fraction of the energy of the incident particle which can be transferred in such a collision depends on the ratio of the masses of the two particles. If the ratio is near unity, the incident particle can lose most of its energy in such a process. In the case of fission products, heavy gases such as argon and krypton are expected to be favorable for larger energy transfer, while light gases such as helium and hydrogen are not favorable. For an incident particle of high velocity in any gas, the probability of an atomic collision is small compared to that for electronic interaction, and atomic collisions are only expected to become important when the penetrating particle has a velocity of the order of v_0 or less.

$$(v_0=e^2/\hbar\cong 2\times 10^8 \text{ cm-sec}^{-1}).$$

The recoil atoms will also have velocities of this order or less, and so are expected to have only a low ionization efficiency.

It is convenient to discuss the change in the energy per ion pair in terms of an ionization defect.[8] The ionization defect is added to the energy calculated from the ionization to give the true energy. Thus

$$E_f=w_\alpha I_f+\Delta E_f. \quad (1)$$

E_f is the initial fission fragment energy, I_f is the ionization resulting when it is stopped completely in the gas, w_α is the energy per ion pair for the alpha particles,

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and ΔE_f is the ionization defect. It has already been pointed out that ionization defects of about 6 and 7 Mev were expected for the most probable light and heavy fission fragments in argon.

The differences in the ionization defects resulting from measurements using two different gases may be found even though the defect for either gas alone may not be measured readily. Initially, it was thought that the defect for helium should be negligible compared to that in argon, and so the difference for these two gases should be a good measure of the argon defect.

II. THE METHOD

It is necessary to have some measure of w_α in each gas used. Therefore, in each case the ionizations resulting from the alpha particles from natural uranium were measured. The energies of these are 4.18 and 4.76 Mev for U^{238} and U^{234}, respectively.[9] For each of two gases, 1 and 2, Eq. (1) may be written and E_f eliminated. This gives:

$$(\Delta E_f)_1-(\Delta E_f)_2=(w_\alpha I_f)_2-(w_\alpha I_f)_1. \quad (2)$$

Since $w_\alpha=E_\alpha/I_\alpha$, the quantities on the right are either known or can be measured. The method then consists of measuring the ratio of the most probable ionizations for fission and alpha particles in various gases. Since only the ratios are important, it is not necessary to make an absolute measurement, and the pulse heights corresponding to the maxima in the distributions may be inserted in the expression.

III. EQUIPMENT

A. The Ionization Chamber

The chamber has been described in detail elsewhere,[10] but a brief description is appropriate here. It used electron collection and employed a Frisch grid.[11] The shielding inefficiency was 6 percent and the theoretical minimum ratio of the grid-collector field to the grid-source field for complete electron collection was 1.32. The gas was purified continuously by circulation through Ca-Mg alloy chips maintained at about 470°C. This type of purifier is thought to be quite effective in the removal of molecular impurities.[12] Previous to filling, the chamber was outgassed by pumping for a period of 20 hours or more at a temperature of about 100°C, with the purifier about 40°C above its normal operating temperature. The pressure in the chamber at the end of the pumping period was always of the order of 10^{-5} mm Hg, and was limited by the vacuum attainable in the pumping system. The filling process always included the use of a cold trap.

The grid-source and grid-collector spacings were 8 and 1.25 cm, respectively. The electrode diameters were about 25 cm. The large dimensions were necessary in order to stop the alpha particles in the grid-source region for reasonable pressures in helium. The natural uranium source was in the form of UO_2, and contained about 60 μg-cm^{-2} of active material. It was prepared by a special spraying process[13] and no significant nonuniformity could be detected by either radioautograph or macroscopic alpha scanning. The source diameter was 12 cm. A collimator was placed in contact with the source which restricted the maximum angle made by an emerging particle with the normal to about 77 deg. This source has since been replaced by an uncollimated source made by the same technique and having a thickness of about 18 μg-cm^{-2}.

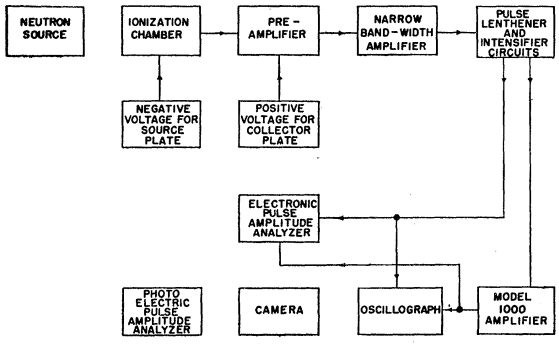

FIG. 1. Block diagram of apparatus.

B. The Electronic Equipment

The block diagram of the measuring equipment is shown in Fig. 1. The conditions of the problem placed several rather unusual restrictions on the electronic circuits. The large length of the grid wires resulted in vibrations which produced sizable microphonics signals. These had, for the most part, frequencies below 750 cps. The large electrode spacings, along with the low electron drift velocities in the purified inert gases resulted in pulse rise times as great as 15 μsec, with about 6-μsec variation due to the angular distribution of particles from the source. It was then necessary to compromise in the amplifier between the long clipping time needed to minimize rise-time effects and the short clipping time necessary to remove microphonics resulting from the grid wires. The amplifier used had equal rise and clipping times of 30 μsec, with several rejection filters tuned to frequencies below the lower half-power point. It has been shown that for such an amplifier the output pulse height has only a small

[9] British Atomic Energy Project Reports, BR-521 and BR-522, 1944 (unpublished).
[10] Herwig, Miller, and Utterback, Rev. Sci. Instr. (to be published).
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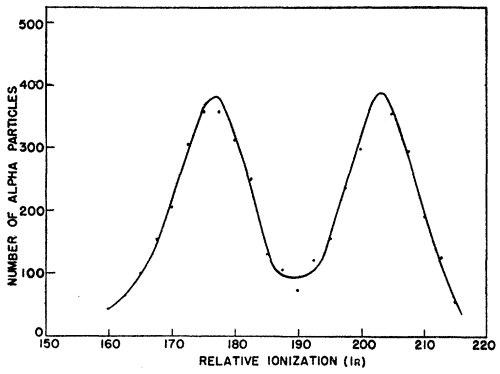

FIG. 2. Typical alpha-particle pulse-height distribution.

dependence on the pulse rise-time.[14] The pulse from this amplifier was shaped to have a flat top about 135 μsec in duration. After further amplification the signal was applied to the vertical deflection plates of a cathode ray tube. The beam in this tube was intensified for a few microseconds during the flat-topped portion, and the corresponding dot was photographed on a continuously moving film with a Dumont-Fairchild 314-A camera. The pulse-height distribution was obtained by analyzing the film in a photoscanning device.[15] In order to avoid the delay involved in this system of analysis, a 20-channel electronic analyzer was also employed.[16] This made possible the rapid examination of alpha saturation curves. The data reported in the present experiment were obtained by the photographic method.

Both alpha and fission pulse heights were obtained in terms of the pulse voltages from a standard pulse generator. These pulses were obtained by charging a condenser from an electronically regulated power supply and discharging it through a network by means of a WE 275B relay. These pulses were applied to the source electrode, and appeared on the collector due to the interelectrode capacitance. The resulting pulses were recorded on the film in the same manner as the ionization pulses.

C. The Neutron Source

The fast neutrons needed for U^{238} fission were obtained from the Ames Laboratory kevatron using the *d-d* reaction. The neutron yield was estimated to be of the order of 10^8 neutrons per second. With the chamber as close to the target as possible, a fission rate of about one per second was obtained.

IV. EXPERIMENTAL RESULTS

A typical distribution of the alpha-particle pulse heights is shown in Fig. 2. The relatively poor resolution of the peaks was due largely to the higher frequency microphonics from the grid wires in the chamber. In spite of this it was possible to locate the peaks in a systematic manner, so that the values agreed within about 0.5 percent for different runs. The selection of the peaks was accomplished using the method of diameters.[17] About 8000 events were recorded for each distribution, requiring a time of about 10 minutes.

In Fig. 3 is shown a typical fission distribution. It is believed that the relatively large width of the distribution was due to source thickness. Chamber microphonics could have no effect in this case, due to the fact that the pulses were about a factor of 20 larger than those resulting from alpha particles. The maxima of these distributions were also located by the method of diameters. In addition, a median calculation was used as a check. In this method the extrapolated value of the upper edge of the light peak was used as a reference in each case. Pulses having heights greater than 74 percent of the extrapolated pulse height were counted in the upper peak. With this quite arbitrary division, the median pulse height of each group in argon agreed with the maximum obtained by the method of diameters. It was then found, using the same criterion for the division of the pulses, that the results of the two methods were in good agreement for the other gases. For the light peak the maximum difference between values found by the two methods was about one percent, while for the heavy peak it was as high as two percent. It is to be expected that the uncertainty in locating the broader heavy peak would be greater.

Saturation curves for fission fragments in several gases are shown in Fig. 4. Alpha-particle saturation curves are not shown, but are quite similar. They are shown and discussed in an earlier reference.[10] The pressures were chosen so that the alpha-particle range was nearly the same in all of the gases. It has been pointed out[18] elsewhere that the values obtained for the

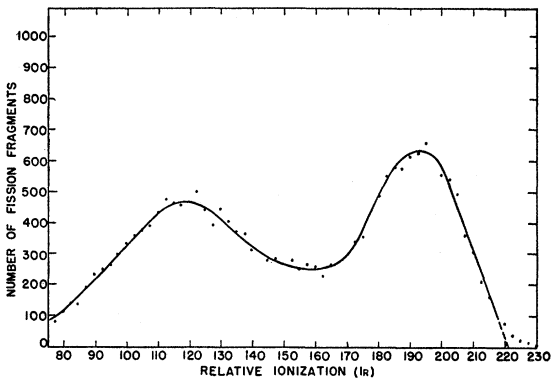

FIG. 3. Typical fission fragment pulse height distribution.

[14] Reference 11, p. 103.
[15] Hunt, Rhinehart, Weber, and Zaffarano, Rev. Sci. Instr. (to be published).
[16] W. E. Glenn, Nucleonics 9, No. 6, 24 (1951).
[17] T. E. Cranshaw and J. A. Harvey, Can. J. Research **A26**, 250 (1948).
[18] L. O. Herwig and G. H. Miller, Phys. Rev. **94**, 1183 (1954).

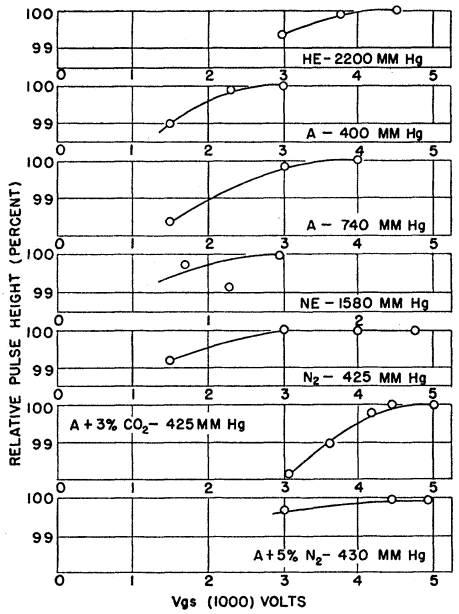

FIG. 4. Saturation curves for fission fragments in various gases.

energy per ion pair for alpha particles are quite satisfactory, an indication of saturation and also of good gas purity.

For each gas filling used, at least two fission runs were made. Usually a run consisted of about 20 000 fissions and required about seven hours. The amplifier gain was checked periodically during this time. There was always a period of at least 24 hours between two fission runs using the same chamber filling. The alpha saturation curve and distribution were checked at intervals for each filling. In the case of the inert gases, the chamber filling was allowed to purify for at least 48 hours before any data were taken. In no case was any significant change in the behavior of a filling noted. For argon, several fillings spaced about a month apart were used. The results of these were in good agreement. Alpha data were taken on several different fillings for all of the gases and mixtures except neon and krypton, where only one filling was used.

Fission runs were made in argon at two different pressures, one-half and one atmosphere. No significant difference was found in the ratio of fission to alpha ionizations for these two pressures. This is important for two reasons: (a) If saturation were not attained, the ratio of ionizations would be expected to decrease with increasing pressure. The increased ionization densities would be more serious for fission particles. (b) If rise time effects were present, they should also decrease the ionization ratio at higher pressure. This would occur because the worst case would be that of an alpha particle at one-half atmosphere. At one atmosphere, the alpha-particle pulse height would be increased relative to the fission pulse height with a consequent decrease in the ionization ratio.

In every case the film was run through the photoscanning device at least twice. The agreement obtained indicated that this part of the process contributed very little to the observational error.

The experimental results are shown in Table I. The value of $w_\alpha I_f$ is the fission fragment energy calculated on the basis of constant energy per ion pair. The difference between these values for any two gases is the right side of Eq. (2), and thus is the difference between the ionization defects in the two gases. In this case the defects have been compared to those in argon+CO_2, since this mixture was used in the results previously cited.[3,4] The fission ionization energies have not been corrected for source absorption which may be as high as four Mev. Source absorption results in a systematic error which is independent of the chamber filling and does not enter into the differences of ionization defects. The main contribution to the error is the uncertainty in the location of the peaks.

V. DISCUSSION

A. Significance of the Results

It had been expected that the fission ionization defects in helium would be as much as five Mev less than those in argon. It may be seen from Table I that no such differences were observed in the present experiment. This will be discussed later.

It is interesting to note that in nitrogen rather large defects resulted. This was not anticipated but is perhaps not surprising. The molecular energy levels offer a rather good mechanism for energy loss by atomic interaction. This may be particularly important for a low velocity particle, where the ionization probability is very much reduced.

This leads to the most important point, which is the rather large difference resulting from the addition of three percent CO_2 to argon. This is particularly significant since the ion chamber measurements mentioned in the introduction, which form the basis for the ionization defect calculations, were made with argon-CO_2 mixtures. It is rather difficult to understand how a small amount of CO_2 can produce such a large effect. Other peculiar effects have been noted in argon-CO_2 mixtures and may have some bearing on this result.[10] It is interesting to note that the addition of five percent of N_2 to argon does not produce a very marked effect.

In view of the argon-CO_2 mixture behavior, the expected difference between argon and helium would be reduced to about three Mev. There seem to be several possible reasons for not observing such a large effect. 1. The effective charge of fission fragments is believed to be lower in helium than in other gases.[19,20]

[19] N. O. Lassen, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **26**, No. 5 (1951).
[20] G. I. Bell, Phys. Rev. **90**, 548 (1953).

Since the ionization probability depends very strongly on the effective charge while the atomic collision probability does not, atomic collisions would play a somewhat greater role than otherwise expected. It has been shown by Jesse[21] that about 25 percent of the energy of an alpha particle stopped in helium is expended in excitation of the metastable states. If there really is an ionization defect for fission fragments in helium, then this percentage may be somewhat higher for fission than for alpha particles. This would offer a means of demonstrating the existence of a defect, since these metastables can be made to produce ionization by adding a small amount ($\sim$0.1 percent) of an impurity, such as argon. If the above hypothesis is correct, then the addition of an impurity should result in an increase in the ratio of fission to alpha ionizations.

2. In the comparison of the results herein contained with the results which would be expected on the basis of the atomic collision mechanism, the effect of such collisions on the stopping of alpha particles has been neglected. If these collisions are important, then it is necessary to see what effect they could have. The energy of an alpha particle may be related to its ionization in the following way.

$$E_\alpha = w^e I_\alpha + \Delta' E_\alpha. \tag{3}$$

Here w^e is the high-velocity asymptotic energy per ion pair, which is believed to be independent of the charge, mass, and energy of the ionizing particle (subject only to the condition that the velocity is high; $v \gg v_0$). The quantity w^e does depend, however, on the stopping gas. It is then necessary to replace w_α in (2) by w^e, and ΔE_f by $\Delta' E_f$. The last term, $\Delta' E_f$, is the true ionization defect, since it is based on the ideal value, w^e. By the use of Eq. (3), w^e may be eliminated, and the following result is then obtained:

$$(\Delta' E_f - \frac{I_f}{I_\alpha}\Delta' E_\alpha)_1 - (\Delta' E_f - \frac{I_f}{I_\alpha}\Delta' E_\alpha)_2 = (w_\alpha I_f)_2 - (w_\alpha I_f)_1. \tag{4}$$

TABLE I. Fission ionization energies and relative ionization defects.

	$(w_\alpha I_f)_x$ Mev		$(w_\alpha I_f)_x - (w_\alpha I_f)_{A+3\%\ CO_2}$ Mev	
gas	light	heavy	light	heavy
Helium	92.9±0.5	61.8±0.7	3.4±0.7	5.4±1.0
Neon	92.0±0.5	60.0±0.7	2.5±0.7	3.6±1.0
Argon	92.5±0.5	60.9±0.7	3.0±0.7	4.5±1.0
Krypton	89.6±0.7	56.0±1.0	0.1±0.9	−0.4±1.2
N_2	88.9±0.5	58.0±0.7	−0.6±0.7	1.6±1.0
A+3% CO_2	89.5±0.5	56.4±0.7	0	0
A+5% N_2	91.5±0.5	59.9±0.7	2.0±0.7	3.5±1.0

[21] Wm. P. Jesse, Atomic Energy Commission Report No. ANL-4944, 1952 (unpublished), p. 2.

Thus, the difference in ionization defects given by Eq. (2), which comes directly from experimentally determined quantities, is in reality different from the true value by the alpha-particle defect terms.

It is possible, but improbable, that the alpha-particle terms could account for failure to find a difference between argon and helium. It would be necessary for the alpha defect in argon to be about 0.15 Mev greater than the corresponding defect in helium. In the following section, arguments are given against the existence of defects of such a magnitude.

The results for neon indicate that it is very little different from argon or helium. Since no great difference is noted for these two gases, it would be rather suprising if neon exhibited a marked defect. On the other hand, krypton showed a rather large effect. This would be expected on the basis of the atomic collision theory, since krypton is very close to the peak of the light fragment distribution.

Unfortunately, very few data are available for the energy dependence of energy per ion for alpha particles. Measurements have been made in argon by Jesse *et al.*[22] and by Cranshaw and Harvey.[17] These results are not in good agreement. In the first case heavy ion collection was used and no significant ionization defect was found. ($\Delta' E_\alpha$ probably less than 25 kev.) In the second experiment, electron collection was employed and a defect of about 85 kev was observed.

Several measurements have been made on particles coming from the $B^{10}(n,\alpha)Li^7$ reaction in argon-CO_2 mixtures.[23–25] The first two of these indicate an ionization defect for the mixture of the order of 50 kev. The result of the third experiment is in agreement with the first two. However, it was shown in addition that as the pressure was reduced, the ratio of the ionizations of the alpha particle and the lithium recoil approached the ratio of the energies of these two particles. This is an indication, but not proof, that there is negligible alpha-particle defect in argon. If there were such a defect, a relatively greater one would be expected for the slower lithium recoil. The ratio of ionizations would then be expected to be in poor agreement with the energy ratio. On the basis of the fission results it would be rather surprising if there were as large an ionization defect for alpha particles in argon as in argon-CO_2. This laboratory is presently engaged in an investigation of the ionization defects for alpha particles in various gases and mixtures.

ACKNOWLEDGMENT

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[23] G. C. Hanna, Phys. Rev. **80**, 530 (1950).
[24] Franzen, Rhodes, and Stephens, Phys. Rev. **87**, 141 (1952).
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