Alpha-Particle Ionization in Argon-Methane Mixtures and the Energy Dependence of the Ion Pair-Formation Energy*

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Experiments have been performed to investigate a long-standing discrepancy in the measurement of α-particle energies by the ionization method. The work of Jesse and his collaborators, carried out in pure argon with a total collection of ions, indicates a linear relation between ionization and α-particle energy for energies between 1 and 9 MeV. A different relation between ionization and α energy is observed in a large number of experiments with gridded pulse chambers; here, with collection of only the electronic component of the ionization in argon-methane (or argon-carbon dioxide) mixtures, an appreciable relative decrease of ionization is observed with decreasing α energy. Discussions in the literature have often attributed this difference between the two methods to the mode of collection of ions, whether total or electronic collection. The present experiments have investigated the possibility that the discrepancy in the two types of experiment is due to the different media used – whether pure argon or argon-methane mixtures. To this end, a series of measurements was carried out for four α-particle energies, ranging from 1.58 to 5.3 MeV. For each of these particles, the ionization, relative to that in pure argon, was measured by a total collection method for a wide range of argon-methane mixtures. The results indicate a dependence with α energy in argon-methane mixtures considerably in excess of any possible dependence which may exist in pure argon. The variation with energy, moreover, in a mixture of argon with 6% methane is in good accord with past results obtained in gridded pulse chambers for this mixture. The present experiments would suggest therefore that the observed difference between the two methods would be the result, not of the method of ion collection, but of the different gaseous media used.

INTRODUCTION

In the measurement of α-particle energies by the ionization method, a very curious discrepancy has existed for a number of years in the experimental results obtained for the ionization of α particles in argon and argon-methane mixtures as a function of the initial energy of the α particle.

On the one hand, the work of Jesse and his collaborators[1–3] would indicate that in very pure argon for α particles of energies between 1 and 9 MeV a linear relation exists between the initial energy of the α particle and the ionization produced by it. Such a relation is found to be valid within the limits of experimental error – about 0.5%. In these experiments it is the practice to collect in the chamber both electrons and positive ions resulting from the ionization process.

A markedly different relation between ionization and α-particle energy has been obtained by a large number of experimenters[4–6] using gridded pulse chambers for the measurement of ionization by single α particles. In such chambers, the transient ionization pulse, produced by each α particle, is amplified and its magnitude suitably recorded. This method differs markedly from the one of Jesse in two particulars:

(1) In this method only the electron component of the ionization produced is collected.

(2) In order to achieve more satisfactory collection, a mixture of argon and methane (or argon and carbon dioxide) is generally substituted for pure argon in the chamber – with about 6% of the contaminant gas being added.

Under the above conditions the relation between ionization and α-particle energy is no longer found to be linear, but the relative ionization observed is found to decrease as the magnitude of the α-particle energy decreases. Experiments of this sort are perhaps best typified by the precision measurements carried out by the group at Chalk River.[4,5] In a resumé of this work,[7] it has been shown for the argon-methane mixtures used that the value of W, the average energy to form an ion pair, is not constant but varies according to an empirical relation, first suggested by Cranshaw and Harvey.[4]

$$W = W_\infty(1 + 0.069E^{-1/2}). \tag{1}$$

Here E denotes the initial energy of the α particle in MeV, and W_∞ is a limiting value of W obtained for α particles of very high energy. Such a formula indicates relatively large deviations of W from constancy. For instance, for an α particle of an initial energy of 1 MeV, the W value is 3.7% higher than that for a polonium α particle of 5.3 MeV.

The marked difference in the results obtained by these two methods of measurement has been the subject of much discussion in the literature during the past fifteen years.[8,9] It has been suggested that the two different methods of collection – total collection of both positive and negative ions in the one case and only the electron component in the other – cause the difference observed, on account of effects produced by the metastable atoms residing in the chamber volume.

To investigate the possible effect of the two different modes of collection, Bay and McLernon[10] have carried out parallel experiments with 5.3-MeV α particles by both methods. They come to the conclusion that, under the same carefully controlled experimental conditions, both methods yield essentially the same absolute W values. So far, therefore, no really valid reason has been ad-

vanced to explain the differences obtained from the two experiments.

Although in previous discussions there has been some casual reference to the possible effects of impurities, no theory seems to have been advanced that perhaps the differences in α ionization, observed as a function of α-particle energy, may possibly be the result of the contaminant gas usually added in the electron collection method. Perhaps the variation of ionization as a function of α energy is not the same in argon-methane mixtures as in highly purified argon.

To investigate this point, a series of experiments has therefore been carried out. Following the method of total collection, ionization experiments from a series of α particles of different energies were made, first in pure argon and then in various mixtures of methane and argon. The results of such a series of experiments are here presented.

APPARATUS AND EXPERIMENTAL METHOD

The present measurements of ionization followed the procedure already employed in previous research.[11, 12] A well-collimated beam of α particles was directed parallel to the axis of a cylindrical brass chamber of length 22 cm and internal diameter 8 cm. This chamber has been described elsewhere.[11]

The increment of ionization from each α particle was fed into a vibrating-reed electrometer and produced a jump of the recording pen of a Brown strip chart recorder. The lengths of such individual jumps were measured and averaged according to methods already described.[12] For each α particle of a given energy, the average value thus obtained gave a good measure of the relative ionization produced in the gas mixture present within the chamber.

The source of α particles was a thin film of ^{210}Po deposited from solution on a silver plate. The energy of the collimated beam of α particles employed could be varied in steps by interposing thin mica sheets between the source and the collimator. Such sheets were mounted as windows in a perforated disk, which could be rotated from without the chamber without disturbing the effective electrical capacitance of the system.

The argon used was, according to the manufacturer's estimate, of 99.99% purity. As in previous experiments, after being introduced into the chamber it was circulated through a trap of cocoanut charcoal, immersed in a suitable freezing mixture. For the measurements in pure argon, such circulation was repeated at intervals during the periods of measurement.

The methane used in the argon-methane mixtures was, according to the manufacturer's estimate, of purity higher than 99.9%. In addition, the methane fillings employed were refractionated from time to time by freezing and pumping in order to minimize impurities arising from the walls of the apparatus. The gas pressures used in the chamber varied from 3.5 to 18 cm of mercury. For each mixture, the pressures were so adjusted that the length of track for each particular α-particle energy was a maximum within the chamber, that is of the order of 20 cm.

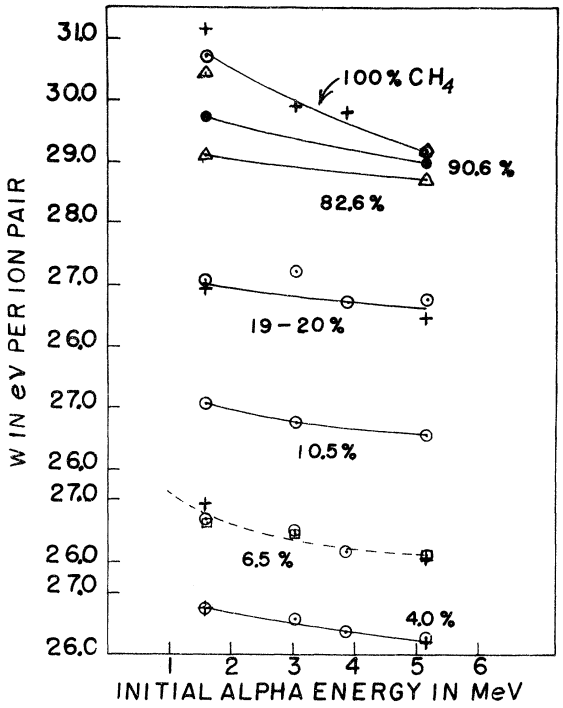

FIG. 1. W values from Table I for argon-methane mixtures are plotted against the initial energy of the ionizing α particles employed. Several individual runs are indicated. In the dashed curve the ordinate values are calculated from the Chalk River formula [Eq. (1)] for comparison with the present experimental values.

RESULTS

This series of α-particle ionization measurements was carried out for argon-methane mixtures with concentrations varying from 0 to 100% methane. Such measurements are shown in Table I and in the plots of Fig. 1 for each of four α energies employed. Thus aperture A with no window gave an estimated energy of 5.3 MeV; and for the mica windows B, C, and D, the estimated energies were those indicated at the top of Table I. Such estimates were made by comparison of the measured ionization produced in pure argon by each of the reduced α particles relative to that produced by the ^{210}Po α particle of 5.3 MeV. It should be noted that such energy estimates need be only approximate, since they are employed only in plotting the graphs in Fig. 1.

In the horizontal lines of Table I are listed the experimental determinations of the W values for each mixture, with methane concentration designated in the first vertical column. Such W values were derived from the experimentally determined ratio of the ionization in pure argon to that in the mixture under the same conditions. To obtain the W value for the mixture, this ratio was then mul-

TABLE I. Summary of W values for argon-methane mixtures as a function of α-particle energy.

Concentration of CH_4 (%)	Run	Window A. W in eV/(ion pair) for energy 5.30 MeV	Window B. W in eV/(ion pair) for energy 3.87 MeV	Window C. W in eV/(ion pair) for energy 3.04 MeV	Window D. W in eV/(ion pair) for energy 1.58 MeV	$\frac{W_{red\alpha}}{W_{Po\alpha}}$
4.0	1	26.18			26.74	
4.0	2	26.28	26.37	26.58	26.75	
	Mean	26.23			26.75	1.020
6.8	1	26.07			26.93	
6.5	2	26.10	26.16	26.52	26.67	
6.5	3	26.12			26.60	
	Mean	26.10			26.73	1.024
Calculated from Chalk River formula		(26.10)	26.23	26.34	26.73	1.024
10.5		26.54		26.74	27.03	1.018
19.0	1	26.45			26.92	
20.2	2	26.79	26.72	27.20	27.06	
	Mean	26.62			26.99	1.014
34.5		27.42			27.67	1.009
51.7		27.94			28.18	1.009
82.6		28.71			29.10	1.014
90.6		28.99			29.80	1.028
100	1	29.16			30.68	
100	2	29.19			30.42	
100	3	29.11	29.81	29.89	31.14	
	Mean	29.15			30.75	1.055

tiplied by the W value for pure argon, 26.38 eV/(ion pair),[13] which was assumed to be the same for each of the α energies employed.

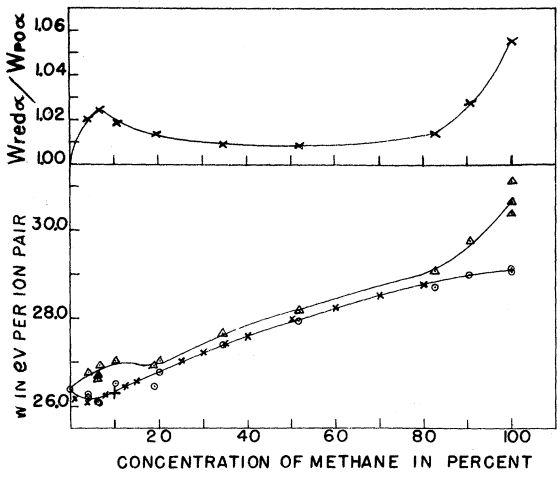

FIG. 2. In the lower diagram, the W values for 5.3 MeV α particles (indicated by ⊙) and for α particles of reduced energy 1.58 MeV (indicated by ▲) are plotted against the corresponding concentration of methane in percent. For comparison, results from Ref. 17 are indicated by × and from Ref. 10 by +. In the upper graph, the ratio of W values for 1.58-MeV α particles to W values for 5.3-MeV α particles (Table I) is plotted against the corresponding methane concentration.

A number of individual runs have been included for the W values in Table I to indicate the over-all consistency of the measurements, and such individual runs have been combined to obtain the mean values indicated. From past experience, one might estimate the standard error of the mean value to be of the order of 0.2% for the unabsorbed Po α (See Ref. 13) and 0.3 to 0.5% in the measurements for the α-particle group of lowest energy, 1.58 MeV.

In Table I it will be noted that the W values listed tend to increase as the α-particle energy decreases, and that the extent of this increase varies with the concentration of methane in the mixture. Such a combined variation of W is perhaps best seen in the last column of Table I, where the ratios of the W values for the α particles of reduced energy 1.58 MeV (column 6) to the W values for the Po α particle (column 3) are assembled. The variation of this ratio as function of methane concentration is shown graphically in the top curve of Fig. 2.

Other aspects of the above relation are shown in the lower diagram of Fig. 2. Here, as ordinates, are plotted the W values, corresponding to each of the terminal energies 5.3 and 1.58 MeV, against the concentration of methane in the mixture. Here again the data for all the individual runs in Table I are included.

In the lower curve, with the present data for polonium α particles indicated by ⊙, a sharp drop from the initial W value of 26.38 eV/(ion pair) is

first observed. Here the curve drops to a minimum value of about 26.10 eV/(ion pair) for a concentration of about 6% methane, and then rises with increasing methane concentration to a final W value of 29.15 eV/(ion pair) for pure methane. This type of curve is well known for argon-methane mixtures for α particles in the neighborhood of 5 MeV.[14-16] Points taken from previous measurements[17] are indicated by × on the diagram. As may be seen, the agreement with the present work is good. The agreement of the present work with that of Bay and McLernon[10] is also shown in Fig. 2, where their absolute value of W for a 10% mixture of methane in argon is indicated by the symbol +.

For the W values corresponding to the smaller α energy of 1.58 MeV, denoted by △ in Fig. 2, a different curve is observed as a function of methane concentration. With increasing concentration, the W value rises from an initial 26.38 eV/(ion pair) to an ill-defined maximum of about 27 eV/(ion pair), corresponding to about 10% of methane. After a slight dip, the curve continues to rise almost linearly up to a concentration of about 82%. A rapid rise then takes place to the maximum value of W of about 30.8 eV/(ion pair), corresponding to 100% methane.

DISCUSSION

The experimental results shown above would seem to indicate that the addition of methane to pure argon introduces into the mixture a greater dependence of W upon the energy of the ionizing α particle than is present in the pure gas. Furthermore the extent of this dependence of W upon α energy seems to be a function of the concentration of methane in the mixture.

Such behavior would seem not too astonishing when one considers the essential difference which has already been shown to exist between the noble gases and hydrogen on the one hand and most polyatomic gases on the other. In the first group, the variation of W with α energy has been found experimentally to be very small over a wide range of α energies, while for the polyatomic gases there is found to be a definite increase of W with diminishing α energy. The reason for such behavior in polyatomic gases is not known. A full discussion of this matter is found in Ref. 3. It will only be noted here that the curve for pure methane, shown at the top of Fig. 1, is quite typical of curves found previously for other pure hydrocarbons.

When, therefore, one adds to pure argon, with a minimum dependence of W upon α energy, a quantity of methane with a very high dependence, it does not seem unreasonable to find that the degree of energy-dependence for the mixture lies somewhere between those found for the two pure gases. This is, of course, what is found in the curves of Figs. 1 and 2.

The reactions observed in argon-methane mixtures are further complicated by the phenomenon already mentioned: small additions of methane (or carbon dioxide) to argon tend to depress the measured value of W for the mixture. Such a phenomenon is well established experimentally[14-16] for α particles of an energy of about 5 MeV. The W values for Poα in Fig. 2 and Table I are here in accord with this rule, since for 6% methane the W value is reduced from 26.38 eV/(ion pair) in pure argon to a value of 26.10 eV/(ion pair) for the mixture.

The same tendency, causing a depression of the W values, undoubtedly must exist in such mixtures for the W values corresponding to α particles of reduced energy. In such cases, however, this depression seems more than compensated by the tendency for W values in pure polyatomic gases to rise with decreasing α energy. The resulting effect is, therefore, a net rise in the W values with small additions of methane. This is shown in the upper curve of the lower diagram in Fig. 2, which indicates the variation of W for α particles of 1.58 MeV.

It is of interest to compare the results from the present experiments with the results calculated from the empirical Chalk River formula – Eq. (1) above. This formula would seem to apply to experimental determinations by the pulse-chamber method for mixtures with a concentration of approximately 6% of methane in argon. Values of W calculated from this formula are listed in Table I, immediately following the present experimental results for methane concentrations of approximately 6%.

Here the calculated and experimental values were arbitrarily made to coincide for the experimental mean W value of 26.10 eV/(ion pair), corresponding to the Poα energy of 5.3 MeV in Table I. The constant W_∞ was then calculated for this set of conditions. Further W values could then be computed from the formula, corresponding to the other particle energies used. A comparison of the experimental and calculated values is shown in Table I and in the corresponding plot in Fig. 1 for a 6.5% methane concentration. In Fig. 1, the dashed curve is that derived by calculation from the formula; the points indicated are experimental determinations from Table I. The general agreement is seen to be well within the limits of the experimental error.

Thus there is a satisfactory agreement obtained between the present measurements and those carried out in the Chalk River Laboratory, each for a mixture of argon with approximately 6% of methane. The marked difference previously obtained between measurements made in this mixture and those in pure argon would suggest an explanation of the discrepancies obtained in the past between the results from the two laboratories. Such differences would seem to come not from the method of ion collection, whether total or electronic, as has been suggested; but rather because the two experiments have been carried out in two different gaseous media.

Ionization-Energy Relation in Pure Argon

It should be pointed out that the present experiments were designed to determine the effect upon the ionization observed of the addition of increments of methane to pure argon. The results are not as precise as the earlier ones[1] for higher α-particle energies in pure argon, and hence add

little to our knowledge of the relation between ionization and particle energy for the pure gas.

A critical review of the results of our former experiments in pure argon suggest the following points:

1. In the region of α-particle energies from 5 to 9 MeV, the former data are within experimental error – about 0.5% – in support of a linear relation between ionization and α-particle energy. A substitution of more precise recent values for the α-particle energies used in Table I of Ref. 1 yields an even better agreement with an assumed linear relation than is indicated there.

2. In the region from about 1 to 5 MeV, the scarcity of natural α emitters limits the number of available measurements. The less direct results from a limited number of induced nuclear reactions, however, seem quite consistent in this region with the above assumed relation, although the reliability is not as good as in the region of high α-particle energies.

3. Such a linear relation between ionization and α-particle energy, implying a constant value of W, does not extend to indefinitely small particle energies in argon. It is well known that heavy particles with velocities comparable with v_0 ionize with reduced efficiency and hence with correspondingly increased W values. The velocity, $v_0 = e^2/\hbar$, is characteristic of an electron in the first Bohr orbit of hydrogen. For an α particle, such a velocity corresponds to an energy of the order of 100 keV. He^+ ions accelerated to such initial velocities have been shown to ionize in argon with W values in the neighborhood of 29 eV/(ion pair).[18] According to the authors cited, however, this W value decreases with increasing velocity and would seem to approach, as an extrapolation, the value of 26.4 eV/(ion pair) characteristic of higher α-particle energies in argon.

Such deficiencies in ionization at very low α-particle energies may possibly influence the linearity of the relation between ionization and α-particle energy in the vicinity of 1 MeV to a greater extent than our own measurements might indicate. Some suggestions as to the possible shape of such a curve in this low-energy region have been advanced by Platzman[19]; until, however, more extended experimental results are available for this region, such suggestions must be regarded as in the realm of conjecture.

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