

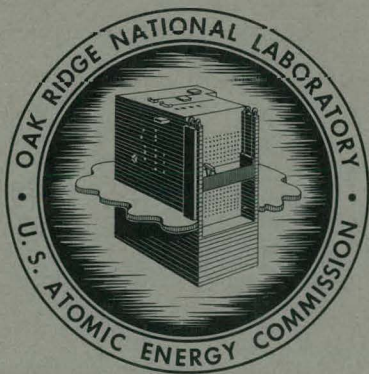
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ALPHA PARTICLE IONIZATION OF ARGON  
MIXTURES - FURTHER STUDY OF THE  
ROLE OF EXCITED STATES

T. E. Bortner  
G. S. Hurst  
Mildred Edmundson  
J. E. Parks



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ALPHA PARTICLE IONIZATION OF ARGON MIXTURES  
FURTHER STUDY OF THE ROLE OF EXCITED STATES

T. E. Bortner, G. S. Hurst, Mildred Edmundson,  
and J. E. Parks

(Submitted by M.E. as a thesis to the Faculty of the Graduate School  
of Vanderbilt University in partial fulfillment of the requirements  
for the degree of Master of Science in Physics)

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## ABSTRACT

This work is concerned with a study of the average energy required to form an ion pair,  $W$ , when alpha particles are absorbed in mixtures of argon with other gases. The other gases have been selected on the basis of their ionization potentials. One group consists of representative gases that have ionization potentials below that of argon (15.77 eV) and above the doublet metastable state of argon (11.49 and 11.66 eV). This list includes methane, carbon dioxide, nitrous oxide, xenon, and krypton. The other group of special interest consists of some gases with ionization potentials below the metastable state of argon. All gases tested in these two groups cause an increase in ionization, i.e., a decrease in  $W$ , when added to argon. As an illustration of this effect, the addition of 1/2% of acetylene to argon will increase the ionization by 23%.

The experimental data, i.e.,  $W$  as a function of relative concentration of the two gases, were fitted to a model in which it is assumed that energy is transferred from two excited levels in argon to the additive gases in collision processes. Good agreement between the experimental data and calculations based on the model is taken as additional evidence that the excited state notion is correct. A quantity of particular interest, i.e., the ionization efficiencies for the additive gases, was derived from the data fitting procedure and compared with those found in photoionization studies in a few of the cases.

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1. INTRODUCTION

When swiftly moving charged particles, e.g., alpha particles, interact with gases, they expend most of their energy by exciting and ionizing the atoms or molecules making up the gas. The ratio  $E_0/N_i$ , where  $E_0$  is the total energy of one ionizing particle and  $N_i$  is the number of ion pairs which it produces, is amenable to accurate experimental determination and is called  $W$ . It is found that  $W$  generally ranges between 20 and 40 electron volts per ion pair, its precise value depending on the type of particle, its energy, and the kind of gas. The appreciable difference in  $W$  and the ionization potential of a particular atomic or molecular species is an indication that a considerable fraction of the energy of the charged particle is expended in non-ionizing excitations of the atomic or molecular system.

In the present investigation we are concerned with the amount of ionization produced in atomic argon by alpha particles with a fixed initial energy, namely 5.14 Mev. In certain atomic gases, e.g., argon, excited states are produced which have lifetimes comparable to atomic collision times. These excited atoms may ionize additive gases by transferring their energy in collision processes. Hence, a study of the amount of ionization produced as a function of relative concentration of the mixtures may provide information on the amount of energy initially deposited in excited states, the cross section for its transfer to the additive gas, and other interesting parameters of these systems. Also,

Platzman<sup>1</sup> has shown that the analysis of experimental data obtained for mixtures of various molecules with the noble gases can give the "ionization efficiency" for highly excited molecules. Recently<sup>2</sup> the significance of these "super excited" states to radiation chemistry has been pointed out. Thus, in the present work particular attention will be given to the production not only of direction ionization but also to the production of excited atomic states and to their role in effecting ionization of additive gases.

In order to analyze experimental ionization data to obtain the interesting parameters associated with excited states, a means is needed to account for the usual or "regular" behavior found when excited states are not involved. For these cases it has been found<sup>3-5</sup> that the  $W$  value for a mixture,  $W_m$ , can be expressed by

$$\frac{1}{W_m} = \left(\frac{1}{W_1} - \frac{1}{W_2}\right)Z + \frac{1}{W_2} \quad (1)$$

where

$$Z = \frac{f_1 P}{f_1 P + a f_2 P} \quad (2)$$

---

<sup>1</sup>R. L. Platzman, J. de Physique et le Radium 21, 853 (1960).

<sup>2</sup>R. L. Platzman, Rad. Res. 17, 419 (1962).

<sup>3</sup>T. E. Bortner and G. S. Hurst, Phys. Rev. 93, 1236 (1954).

<sup>4</sup>H. J. Moe, T. E. Bortner, and G. S. Hurst, J. Phys. Chem. 61, 422 (1957).

<sup>5</sup>G. S. Hurst and T. D. Strickler, "Alpha Particle Ionization of the Binary Gas Mixtures," National Research Council Publication 752, p. 134 (1960).

The quantities  $f_1P$  and  $f_2P$  are the partial pressures for gases whose  $W$  values are  $W_1$  and  $W_2$ , respectively, and "a" is an empirical parameter depending on the particular pair of gases.

Where excited states are involved in producing additional ionization, for example in the mixtures which have a noble gas as one or both of the components,  $W_m$  does not bear this simple relationship to the composition of the mixture. The addition of very small percentages of a second gas, e.g., less than 1%, to a noble gas results in a sharp decrease in  $W_m$ . This effect has been named the "Jesse effect"<sup>6</sup> in honor of W. P. Jesse who called attention to its importance in his careful studies of alpha particle ionization of helium.<sup>7-9</sup> As the percentage of the additive gas in the mixture is increased, the value for  $W_m$  passes through a minimum and then rises smoothly to the  $W$  value of the additive gas. An equation for the  $W$  of mixtures in which excited states are taken into explicit account will be given in a later section.

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<sup>6</sup>R. L. Platzman, "The Physical and Chemical Basis of Mechanisms in Radiation Biology," in Radiation Biology and Medicine, W. D. Claus, Ed. (Addison-Wesley Publishing Co., Inc., Reading, Mass., 1958), pp. 15-72.

<sup>7</sup>W. P. Jesse and J. Sadauskis, Phys. Rev. **88**, 417 (1952).

<sup>8</sup>Ibid., **90**, 1120 (1953).

<sup>9</sup>Ibid., **100**, 1755 (1955).

## II. APPARATUS

The apparatus for the determination of the ionization in the gas mixture consisted of a parallel plate ionization chamber and its accompanying vacuum system, two lines for introducing the gases into the chamber, a high voltage supply, an electrometer, a capacitor, and a potentiometer necessary for rate-of-drift readings.

The parallel plate ionization chamber (see Fig. 1) was enclosed in a stainless steel cylinder, 26 cm in diameter, 14 cm high, and sealed at both ends. The stainless steel plates were supported by Teflon insulators at a separation of 6.85 cm. The lower plate was 18 cm in diameter and was connected to the high voltage supply. On it was flush mounted the plain disk  $\text{Pu}^{239}$ , 5.14-Mev alpha source. The collecting plate, also 18 cm in diameter, was surrounded by a guard ring mounted at a distance of 0.08 cm from the plate to assure a uniform field in the ionizing region. The collecting plate contained the lead to a "Fast" capacitor, used for "rate-of-drift" measurements. The potentiometer, designed by the Instrument Department of the Oak Ridge National Laboratory, was equipped with a helipot by which the voltage could be varied from 0 to 5000 millivolts. It also contained a standard cell and an adjustment potentiometer for calibration. The electrometer, Oak Ridge National Laboratory model Q826B, was used as the null indicator. A decade scaler counting on a 60-cycle line and equipped with a fingertip switch was used to measure the collection time.

The components of the vacuum system were connected by one-fourth in. copper tubing. A Wallace and Tiernan pressure gauge with a range of

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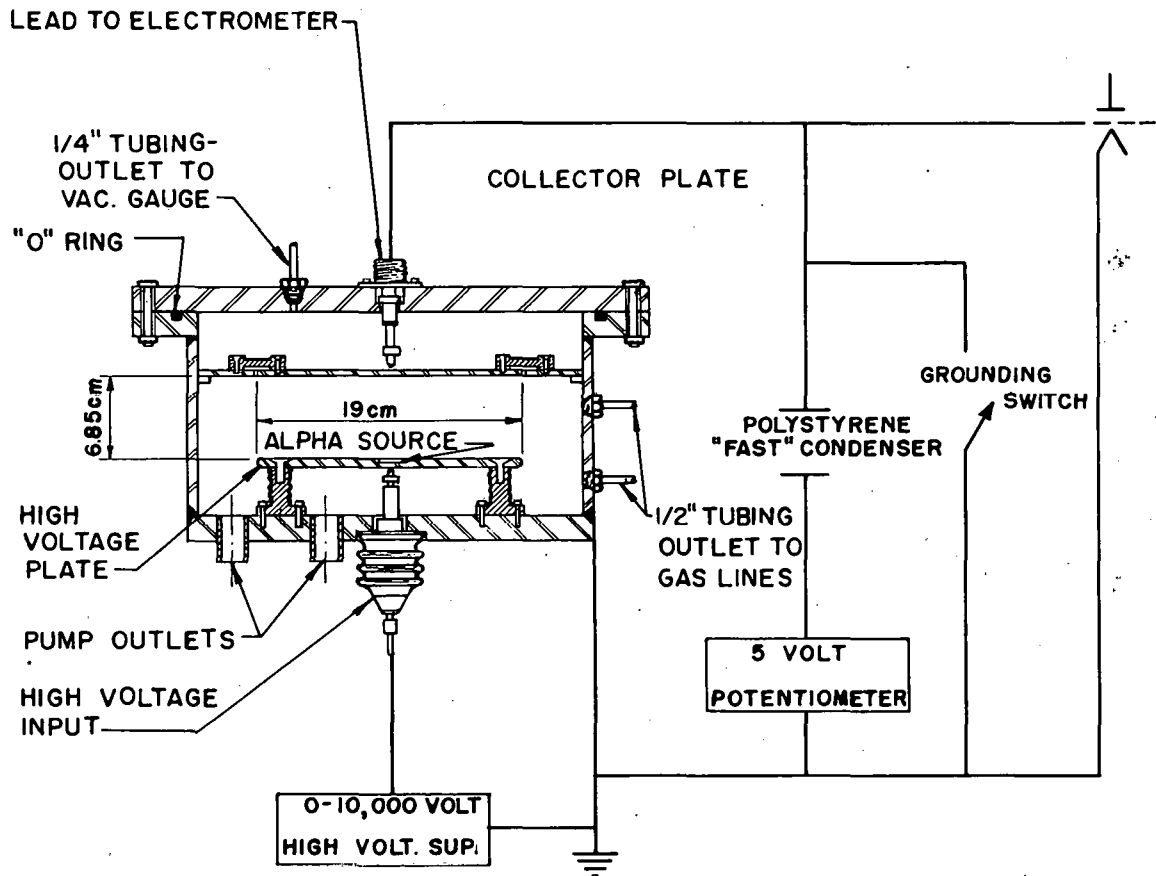


Fig. 1. Diagram of the Ionization Chamber and the Electrical Circuit for Measuring W of Gas Mixtures



0 to 228 mm Hg was mounted on the chamber (see Fig. 2). A Hastings gauge was used to indicate the vacuum achieved in the chamber during pumpdown. Each of the gas lines was equipped with a vacuum pressure gauge. The cold traps mounted on the gas lines were made of 4.5-cm copper tubing, 16.5 cm long.

Two Welch Duo-Seal mechanical pumps were incorporated in the system. One was used to rough pump the chamber, and could also be used as a by-pass to pump either or both of the gas lines. The other was connected directly to the chamber with a Veeco 3/4-in. angle valve. An Ultek MT-100 molecular sieve foreline trap was placed in the line from the valve to the pump. This pump was capable of pumping the chamber down to about  $10^{-3}$  Torr. An Ultek SR-100 sorption roughing pump was also connected to the chamber with a 3/4-in. Veeco angle valve.

A model 400BD power supply manufactured by the John Fluke Manufacturing Company was used to provide voltages from 500 to 5000 volts. A model H-30, 10-KV high voltage power supply, NJE Corporation, was used for voltages ranging from 5000 to 10,000 volts.

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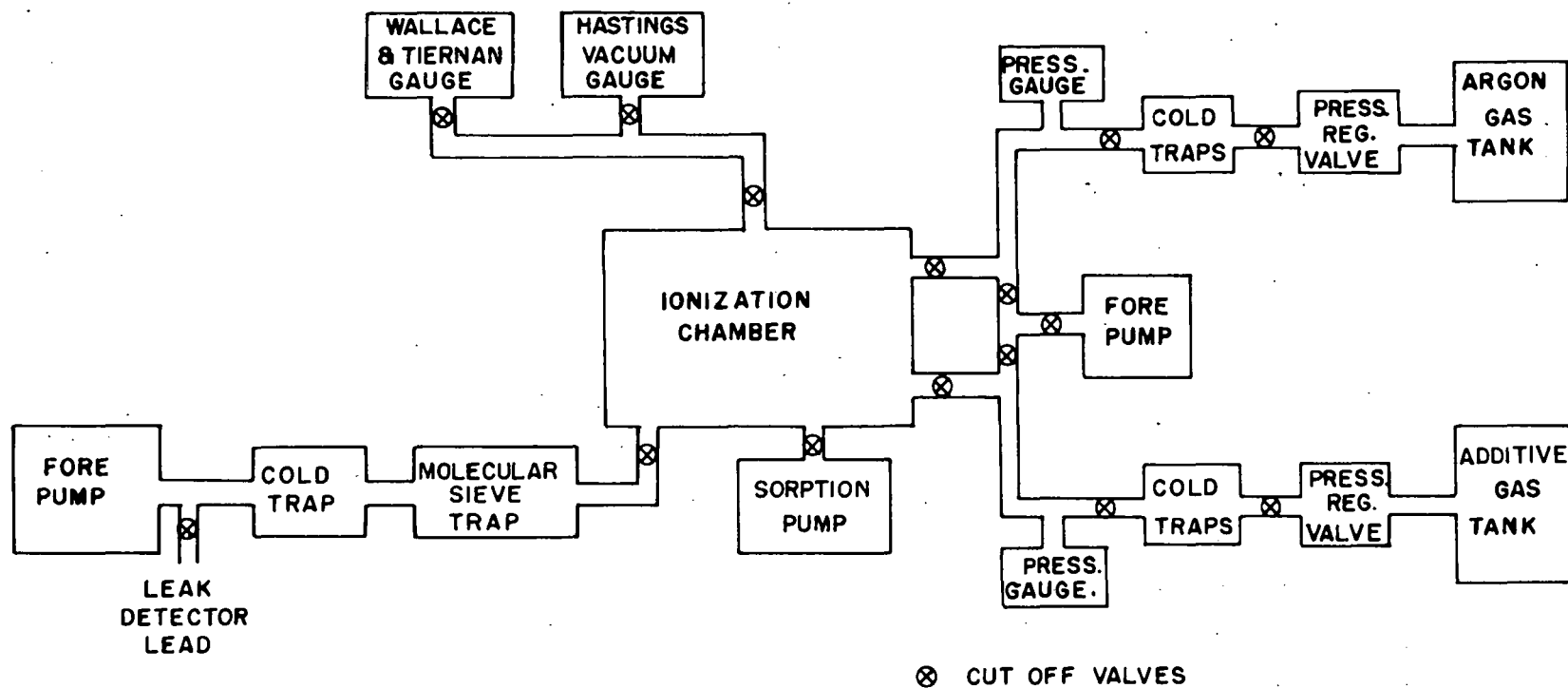


Fig. 2. Block Diagram of the Gas and Vacuum System Used for Measuring W Values of Gas Mixtures

## III. EXPERIMENTAL PROCEDURE

The system was pumped to a pressure of about  $10^{-3}$  Torr before the gases were introduced into the ionization chamber. The argon was run through liquid nitrogen cold traps and was flushed through the chamber before filling. The additive gases were run through cold traps with the appropriate coolant (liquid nitrogen or dry ice), and were flushed through the delivery system by the by-pass pump before being introduced into the chamber. Total pressure of 50 cm Hg was used for all mixtures, so that the alpha particles expended all their energy within the volume of the chamber. Mixtures were made by monitoring the partial pressures on the Wallace and Tiernan gauge. Mixtures below 1% were made by successively reducing the total pressure to 25 cm Hg and then adding argon to 50 cm, thus successively reducing the percentage of the additive gas by one-half. At least two series with alternating percentages were measured and checked against each other for the mixtures with less than 1% of the additive gas.

Voltages used were well within the saturation range such that complete collection of the electrons produced occurred with no recombination. The W values obtained were calculated by comparison to the well-established W value for argon, 26.4 electron volts (ev) per ion pair, and also with  $N_2$  at 36.6 ev as a double check. The time to make a 5000-mv collection was measured. The unknown W value is then determined by the ratio,

$$\frac{W_{Ar}}{(\text{Time})_{Ar}} = \frac{W_m}{(\text{Time})_m}$$

or

$$W_m = \frac{(\text{Time})_m}{(\text{Time})_{Ar}} W_{Ar} = (\text{Time})_m \times C_{Ar}$$

where  $C_{Ar}$  is the constant multiplying factor based on argon. This factor was carefully redetermined before each new gas series was begun.

## IV. EXPERIMENTAL DATA

Tables I through XIV and the graphs given in Figs. 3 through 17 show the experimental results. The absolute values of  $W_m$  are not more accurate than 1 part in 200. However, relative values are somewhat more accurate, and four significant figures are reported in the tables for data fitting purposes.

TABLE I  
EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND KRYPTON

Per Cent of Krypton	$W_m$ in ev
100	24.04
90.0	24.03
80.0	24.00
70.0	23.98
60.0	23.98
50.0	23.96
40.0	23.93
30.0	23.96
25.0	24.00
20.0	24.05
15.0	24.17
12.5	24.27
10.0	24.39
7.50	24.59
6.50	24.73
5.00	24.92
3.75	25.13
2.50	25.44
1.88	25.59
1.25	25.81
1.00	25.88
0.938	25.92
0.625	26.06
0.469	26.14
0.234	26.23
0.117	26.31
0.059	26.33
0	26.40

TABLE II  
 EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND XENON

Per Cent of Xenon	$W_m$ in ev	Per Cent of Xenon	$W_m$ in ev
100	21.50	2.00	23.12
90.0	21.54	1.56	23.45
80.0	21.58	1.13	23.75
70.0	21.60	1.00	24.03
64.0	21.63	0.781	24.38
50.0	21.68	0.563	24.62
40.0	21.68	0.500	24.80
32.0	21.69	0.391	25.11
25.0	21.69	0.281	25.32
20.0	21.71	0.195	25.64
16.0	21.73	0.141	25.75
12.5	21.76	0.098	25.96
10.0	21.81	0.070	26.04
8.0	21.92	0.0488	26.16
6.25	22.05	0.0352	26.20
5.00	22.20	0.0244	26.25
4.00	22.38	0.0176	26.30
3.13	22.62	0.00879	26.34
2.50	22.87	0	26.40

TABLE III

EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND CARBON DIOXIDE

Per Cent of Carbon Dioxide	$W_m$ in ev	Per Cent of Carbon Dioxide	$W_m$ in ev
100	34.45	6.00	26.11
90.0	33.68	5.00	26.06
80.0	32.76	4.50	26.05
70.0	31.87	4.00	26.03
60.0	30.98	3.50	26.03
50.0	30.04	3.00	26.03
40.0	29.13	2.50	26.05
35.0	28.67	2.00	26.07
30.0	28.20	1.75	26.09
25.0	27.72	1.50	26.12
20.0	27.27	1.25	26.15
15.0	26.80	1.00	26.18
12.5	26.59	0.800	26.22
12.0	26.53	0.750	26.25
10.0	26.39	0.400	26.31
9.00	26.30	0.375	26.32
8.00	26.22	0.200	26.35
7.50	26.18	0.100	26.39
7.00	26.17	0	26.40



TABLE IV  
EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND METHANE

Per Cent of Methane	$W_m$ in ev	Per Cent of Methane	$W_m$ in ev
100	29.26	5.00	26.09
90.0	29.03	4.00	26.08
80.0	28.79	3.00	26.04
70.0	28.53	2.50	26.05
60.0	28.26	2.00	26.10
50.0	27.98	1.50	26.11
40.0	27.62	1.25	26.12
35.0	27.43	1.00	26.17
30.0	27.23	0.75	26.21
25.0	27.03	0.50	26.25
20.0	26.83	0.375	26.29
15.0	26.56	0.250	26.32
12.5	26.45	0.188	26.34
10.0	26.30	0.125	26.35
8.00	26.26	0.0938	26.37
7.50	26.20	0.0625	26.39
6.00	26.12	0.0469	26.39
		0	26.40

TABLE V  
 EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND NITROUS OXIDE

Per Cent of Nitrous Oxide	$W_m$ in ev	Per Cent of Nitrous Oxide	$W_m$ in ev
100	34.43	2.00	25.65
90.0	33.69	1.25	25.68
80.0	32.97	1.00	25.74
70.0	32.20	0.800	25.84
60.0	31.41	0.625	25.83
50.0	30.52	0.500	25.91
40.0	29.64	0.400	26.01
30.0	28.67	0.313	26.02
20.0	27.63	0.250	26.08
12.0	26.70	0.200	26.15
10.0	26.46	0.156	26.14
8.00	26.23	0.125	26.18
6.00	25.98	0.0781	26.20
5.00	25.85	0.0625	26.25
4.00	25.74	0.0313	26.29
3.00	25.69	0.0156	26.30
2.50	25.64	0	26.40

TABLE VI

EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND ETHANE

Per Cent of Ethane	$W_m$ in ev	Per Cent of Ethane	$W_m$ in ev
100	26.70	1.75	24.58
90.0	26.64	1.50	24.61
80.0	26.54	1.00	24.81
70.0	26.41	0.750	24.97
60.0	26.30	0.600	25.06
50.0	26.18	0.500	25.14
40.0	25.98	0.375	25.29
30.0	25.70	0.300	25.35
25.0	26.53	0.250	25.46
20.0	25.30	0.188	25.56
18.0	25.27	0.125	25.66
15.0	25.09	0.0938	25.73
14.0	25.06	0.0625	25.83
13.0	24.96	0.0469	25.90
12.0	24.89	0.0313	25.92
11.0	24.85	0.0234	26.01
10.0	24.81	0.0156	26.07
9.00	24.74	0.0117	26.10
8.00	26.65	0.00781	26.18
7.00	24.56	0.00586	26.23
6.00	24.50	0.00391	26.28
5.00	24.43	0.00293	26.31
4.00	24.43	0.00195	26.33
3.50	24.44	0.00146	26.35
3.00	24.43	0.000977	26.37
2.50	24.46	0.00732	26.40
2.00	24.52	0	26.40

TABLE VII

EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND ACETYLENE

Per Cent of Acetylene	$W_m$ in ev	Per Cent of Acetylene	$W_m$ in ev
100	27.64	0.375	20.41
90.0	27.24	0.300	20.43
80.0	26.86	0.250	20.43
70.0	26.43	0.200	20.48
60.0	25.96	0.188	20.51
50.0	25.45	0.150	20.57
40.0	24.88	0.125	20.62
30.0	24.21	0.100	20.67
20.0	23.45	0.0938	20.79
10.0	22.41	0.0625	21.08
9.00	22.29	0.0469	21.28
8.00	22.14	0.0313	21.68
6.00	21.85	0.0300	21.67
5.00	21.67	0.0234	21.92
4.00	21.49	0.0156	22.39
3.20	21.31	0.0150	22.38
3.00	21.27	0.0117	22.65
2.50	21.15	0.00900	22.99
2.40	21.12	0.00781	23.20
2.00	20.98	0.00586	23.44
1.60	20.86	0.00450	23.79
1.50	20.83	0.00391	24.02
1.20	20.72	0.00293	24.25
1.00	20.63	0.00225	24.54
0.800	20.54	0.00195	24.78
0.750	20.54	0.00146	24.95
0.700	20.49	0.000977	25.38
0.600	20.49	0.000732	25.47
0.500	20.44	0.000488	25.79
0.400	20.40	0.000244	26.05
		0	26.40

TABLE VIII

EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND PROPANE

Per Cent of Propane	$W_m$ in ev	Per Cent of Propane	$W_m$ in ev	Per Cent of Propane	$W_m$ in ev
100	26.30	1.80	23.52	0.00703	25.20
90.0	26.29	1.75	23.52	0.00684	25.20
80.0	26.25	1.50	23.57	0.00391	25.63
70.0	26.10	1.25	23.53	0.00352	25.60
60.0	26.03	1.00	23.60	0.00342	25.53
50.0	25.85	0.900	23.60	0.00195	25.88
40.0	25.65	0.875	23.58	0.00176	25.85
30.0	25.26	0.500	23.70	0.00171	25.80
20.0	24.92	0.450	23.75	0.000977	26.11
18.0	24.71	0.438	23.83	0.000854	26.02
15.0	24.62	0.250	23.92	0.000488	26.28
14.0	24.51	0.225	23.93	0.000427	26.17
12.0	24.39	0.125	24.01	0.000379	26.07
10.0	24.22	0.112	24.16	0.000214	26.26
8.00	23.98	0.109	24.16	0.000189	26.22
7.00	23.95	0.0625	24.30	0.000107	26.36
6.00	23.84	0.0563	24.37	0.0000947	26.28
5.00	23.79	0.0547	24.36	0.0000533	26.40
4.50	23.69	0.0313	24.64	0.0000244	26.40
4.00	23.65	0.0273	24.63	0.0000474	26.35
3.50	23.60	0.0156	24.91	0.0000237	26.40
3.00	23.60	0.0141	24.92	0	26.40
2.25	23.53	0.0137	24.92		
2.00	23.52	0.00781	25.30		

TABLE IX

EXPERIMENTAL VALUES OF  $w_m$  FOR MIXTURES OF ARGON AND N-BUTANE

Per Cent of N-Butane	$w_m$ in ev	Per Cent of N-Butane	$w_m$ in ev
100	26.18	0.250	23.17
97.5	26.13	0.188	23.22
95.0	26.10	0.156	23.26
90.0	26.04	0.125	23.34
80.0	25.91	0.100	23.41
70.0	25.79	0.0938	23.41
60.0	25.63	0.0800	23.46
50.0	25.47	0.0781	23.44
40.0	25.26	0.0625	23.55
30.0	24.99	0.0469	23.65
20.0	24.55	0.0400	23.72
10.0	23.88	0.0313	23.84
9.00	23.80	0.0234	23.96
8.00	23.69	0.0200	24.04
7.00	23.60	0.0156	24.18
6.00	23.49	0.0117	24.31
5.00	23.38	0.0100	24.44
4.00	23.25	0.00781	24.56
3.00	23.12	0.00586	24.69
2.50	23.07	0.00500	24.82
2.00	23.01	0.00391	24.98
1.50	22.96	0.00250	25.26
1.25	22.97	0.00200	25.40
1.00	22.97	0.00125	25.59
0.750	22.98	0.00100	25.73
0.625	23.02	0.000625	25.86
0.500	23.03	0.000500	25.98
0.400	23.08	0.000250	26.13
0.375	23.10	0.000125	26.21
0.313	23.12	0.0000625	26.28
		0	26.40

TABLE X  
 EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND ETHYLENE

Per Cent of Ethylene	$W_m$ in ev	Per Cent of Ethylene	$W_m$ in ev
100	28.00	1.00	23.90
90.0	27.97	0.500	24.00
80.0	27.75	0.250	24.07
70.0	27.52	0.125	24.18
60.0	27.32	0.0800	24.48
50.0	26.96	0.0400	24.70
40.0	26.59	0.0200	24.94
30.0	26.14	0.0100	25.21
20.0	25.60	0.00500	25.53
10.0	24.85	0.00250	25.80
5.00	24.25	0.00125	26.10
2.50	24.02	0.000625	26.28
2.00	23.92	0	26.40

TABLE XI  
EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND ISOBUTANE

Per Cent of Isobutane	$W_m$ in ev	Per Cent of Isobutane	$W_m$ in ev
100	26.52	0.500	23.24
90.0	26.40	0.450	23.26
81.0	26.28	0.400	23.30
80.0	26.27	0.350	23.30
64.8	26.03	0.310	23.32
60.0	25.98	0.270	23.35
50.0	25.80	0.250	23.35
40.0	25.59	0.240	23.38
32.4	25.32	0.200	23.46
30.0	25.25	0.162	23.47
20.0	24.79	0.140	23.51
16.2	24.57	0.100	23.63
10.0	24.06	0.923	23.63
9.00	23.97	0.0840	23.61
8.10	23.90	0.0810	23.69
8.00	23.88	0.080	23.71
6.00	23.64	0.0743	23.73
5.00	23.52	0.0700	23.76
4.00	23.42	0.0594	23.84
3.00	23.28	0.0500	23.83
2.00	23.18	0.0405	23.98
1.40	23.19	0.0400	23.97
1.20	23.18	0.0297	24.18
1.00	23.19	0.0202	24.47
0.900	23.20	0.0175	24.55
0.860	23.18	0.0149	24.59
0.800	23.20	0.0101	24.92
0.700	23.22	0.00506	25.34
0.600	23.25	0.00253	25.70
0.516	23.24	0	26.40



TABLE XII

EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND CYCLOPROPANE

Per Cent of Cyclopropane	$W_m$ in ev	Per Cent of Cyclopropane	$W_m$ in ev
100	26.11	1.75	23.03
90.0	26.03	1.25	23.02
80.0	25.82	1.00	23.05
70.0	25.77	0.600	23.12
60.0	25.62	0.500	23.18
50.0	25.52	0.300	23.22
40.0	25.19	0.250	23.34
30.0	24.96	0.150	23.42
20.0	24.46	0.125	23.49
15.0	24.09	0.0750	23.63
14.0	24.07	0.0625	23.70
13.0	23.97	0.0375	23.90
12.0	23.93	0.0313	24.00
11.0	23.82	0.0188	24.28
10.0	23.79	0.0156	24.43
9.00	23.65	0.00938	24.71
8.00	23.59	0.00781	24.82
7.00	23.49	0.00469	25.14
6.00	23.33	0.00391	25.28
5.00	23.30	0.00234	25.49
4.00	23.18	0.00195	25.60
3.50	23.11	0.00117	25.83
3.00	23.07	0.000586	26.00
2.50	23.05	0.000293	26.21
2.00	23.03	0	26.40

TABLE XIII

EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND PROPYLENE

Per Cent of Propylene	$W_m$ in ev	Per Cent of Propylene	$W_m$ in ev
100	27.28	1.00	23.79
90.0	27.22	0.800	23.81
80.0	27.04	0.600	23.85
70.0	26.85	0.400	23.90
60.0	26.62	0.300	23.94
50.0	26.42	0.200	24.00
45.0	26.28	0.150	24.05
40.0	26.14	0.100	24.13
35.0	25.95	0.075	24.17
30.0	25.79	0.050	24.27
25.0	25.56	0.038	24.35
20.0	25.32	0.025	24.45
17.5	25.14	0.0188	24.55
15.0	24.99	0.0125	24.73
12.5	24.80	0.00938	24.87
10.0	24.61	0.00625	25.02
9.00	24.50	0.00469	25.12
8.00	24.42	0.00313	25.28
7.00	24.32	0.00234	25.39
6.00	24.22	0.00156	25.56
5.00	24.14	0.000781	25.77
4.00	24.01	0.000579	25.86
3.00	23.90	0.000289	26.00
2.00	23.83	0.000145	26.12
1.50	23.79	0.0000723	26.18
1.20	23.79	0	26.40

TABLE XIV

EXPERIMENTAL VALUES OF  $W_m$  FOR MIXTURES OF ARGON AND BUTENE I

Per Cent of Butene I	$W_m$ in ev	Per Cent of Butene I	$W_m$ in ev
100	27.09	1.25	23.28
90.0	26.91	1.00	23.22
80.0	26.69	0.500	23.27
70.0	26.52	0.300	23.40
60.0	26.37	0.250	23.41
50.0	26.13	0.150	23.53
40.0	25.87	0.125	23.57
30.0	25.53	0.0750	23.68
20.0	25.06	0.0625	23.80
15.0	24.74	0.0375	23.90
14.0	24.64	0.0313	24.00
13.0	24.57	0.0188	24.12
12.0	24.48	0.0156	24.28
11.0	24.40	0.00938	24.44
10.0	24.31	0.00781	24.65
9.00	24.20	0.00469	25.00
8.00	24.10	0.00391	24.01
7.50	24.09	0.00234	25.19
7.00	23.95	0.00195	25.40
6.00	23.84	0.00117	25.53
5.00	23.72	0.000977	25.75
4.00	23.62	0.000586	25.77
3.75	23.58	0.000488	25.95
3.00	23.45	0.000293	26.03
2.50	23.41	0.000244	26.07
2.00	23.32	0.00146	26.18
1.88	23.30	0	26.40

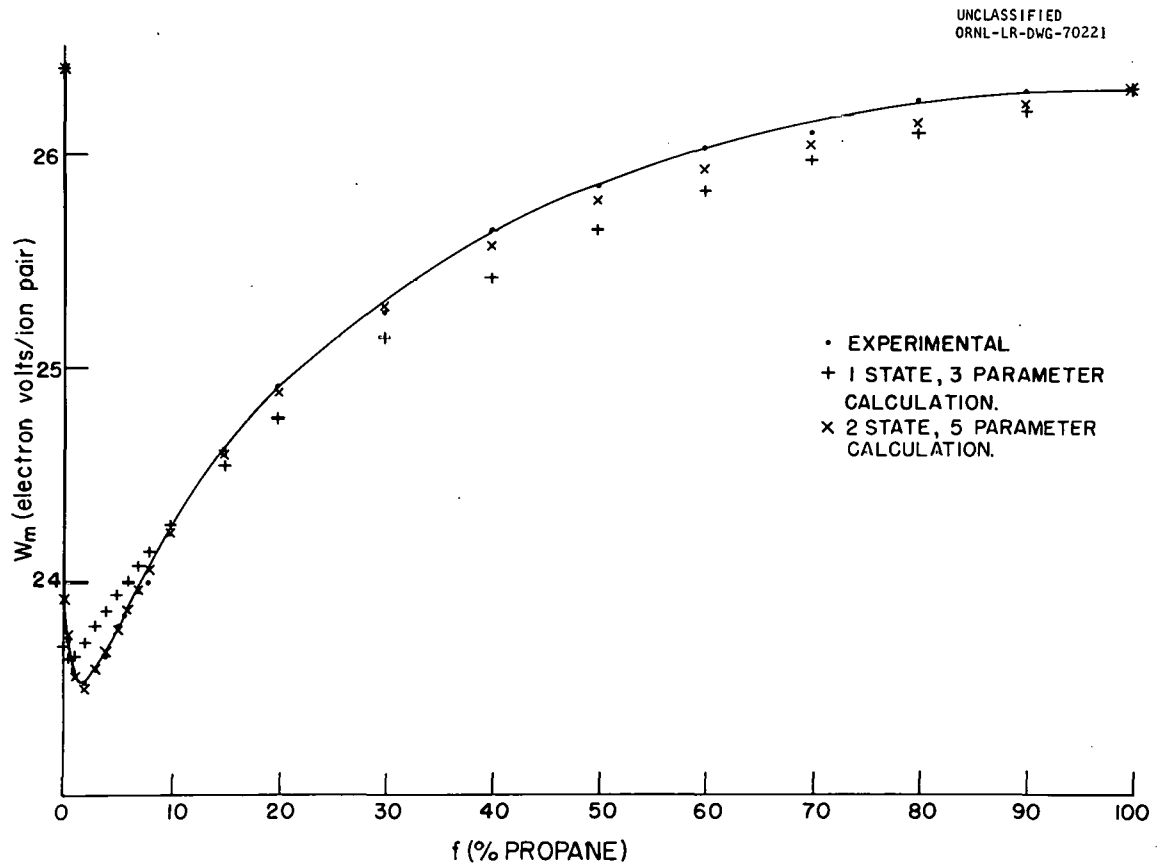


Fig. 3. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Propane

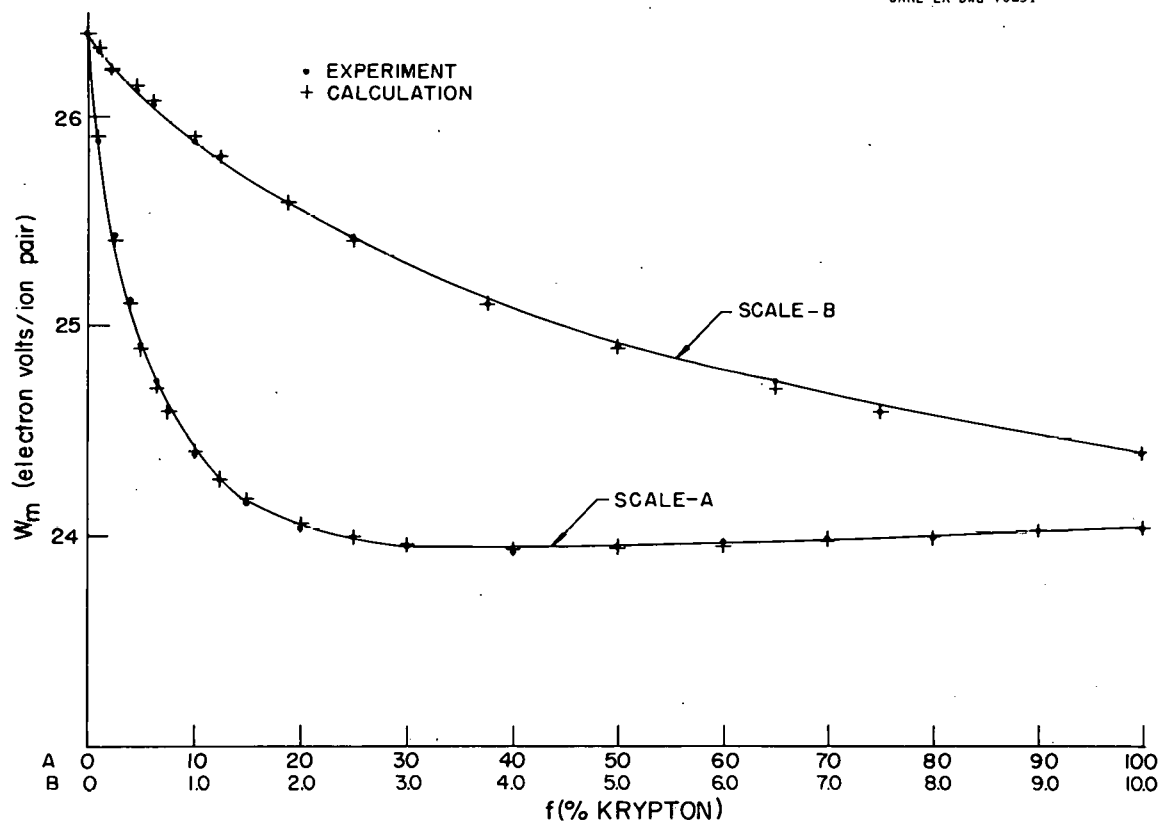
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Fig. 4. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Krypton

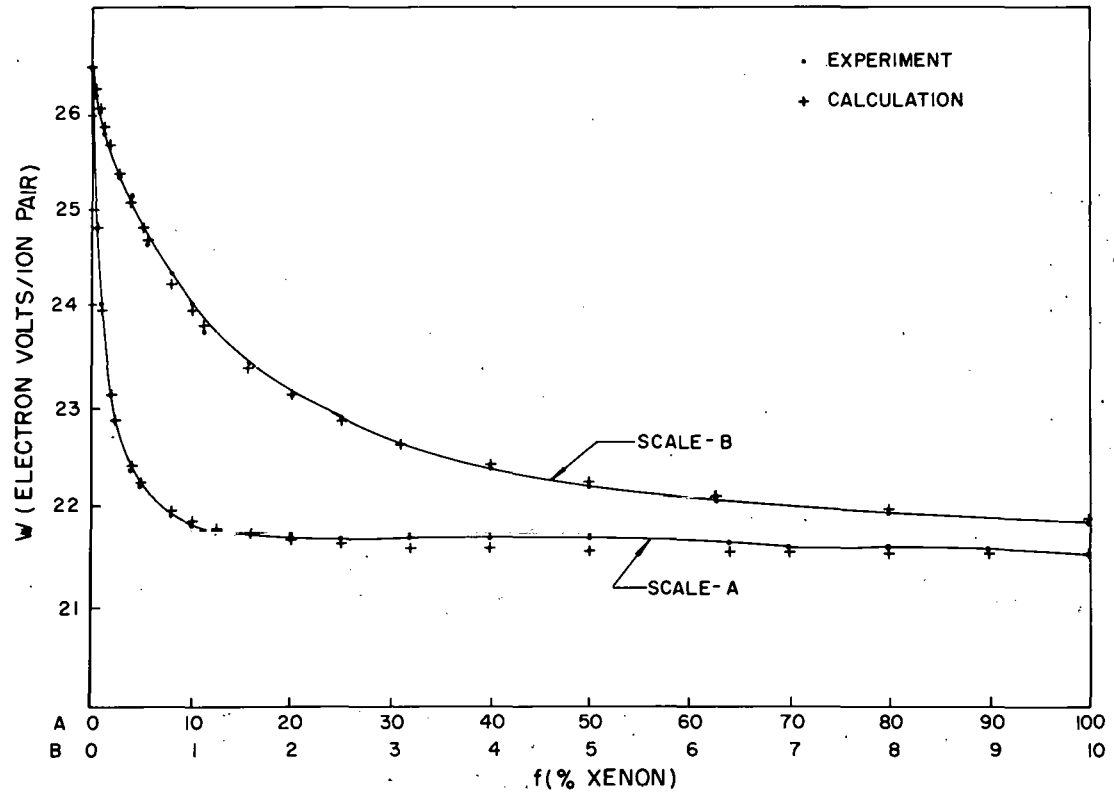
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Fig. 5. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Xenon

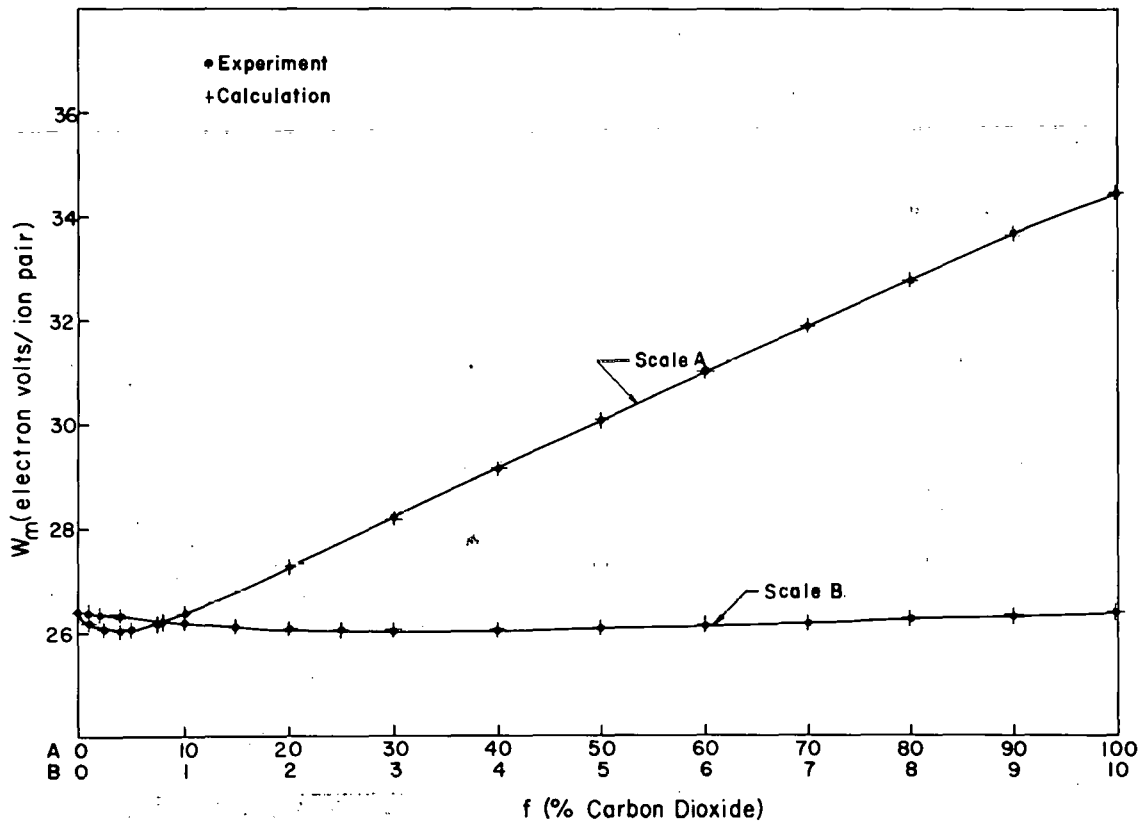
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Fig. 6. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Carbon Dioxide

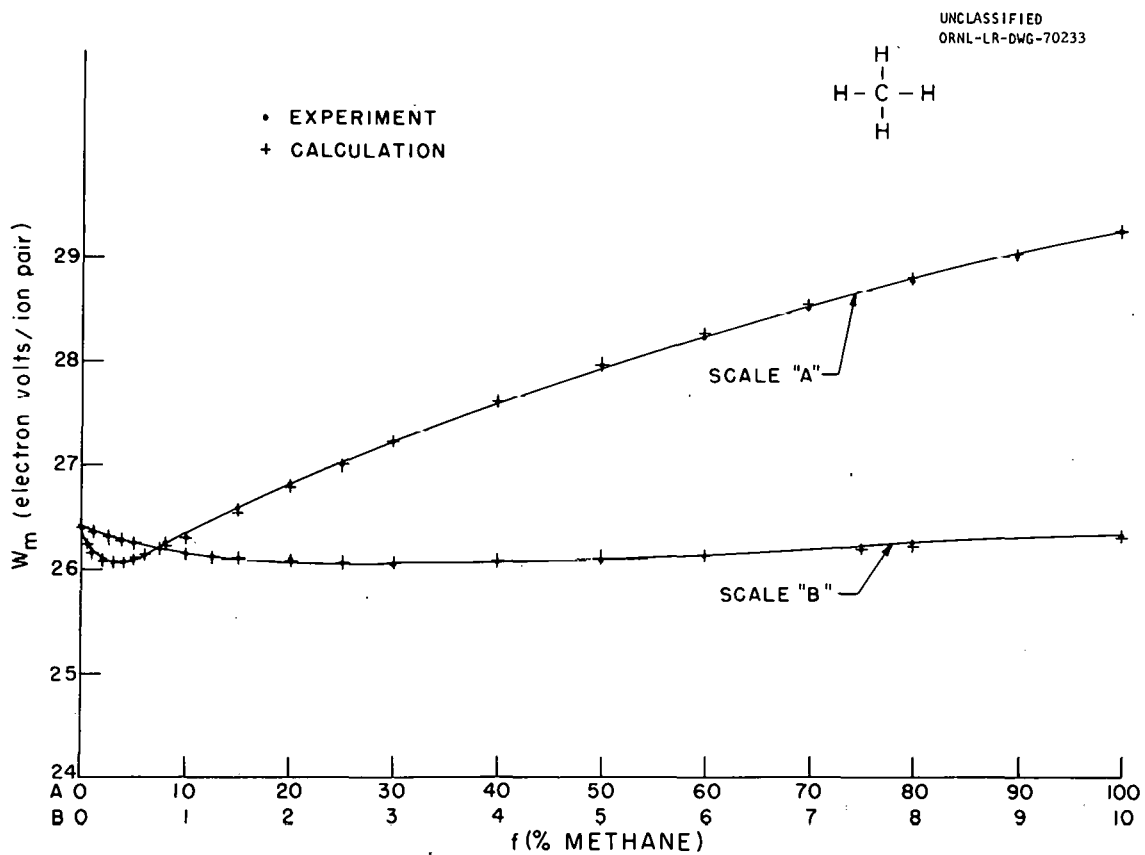


Fig. 7. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Methane



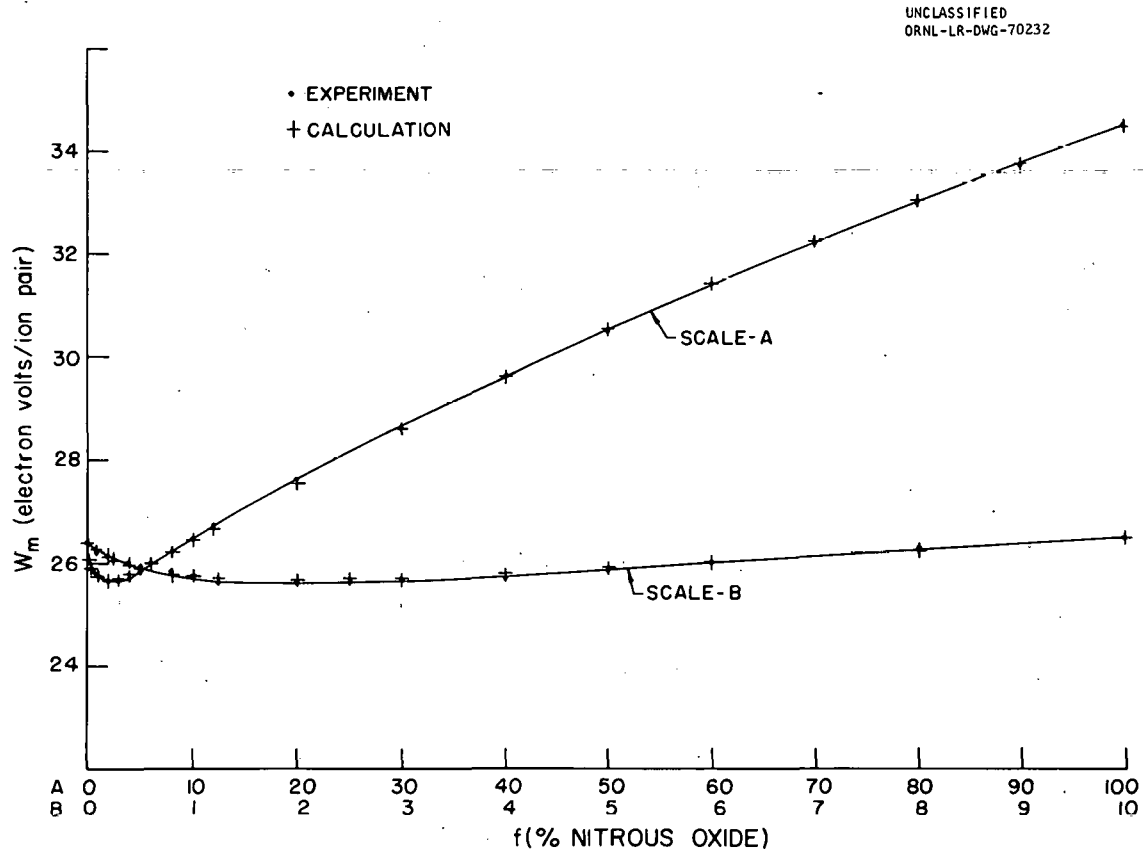


Fig. 8. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Nitrous Oxide

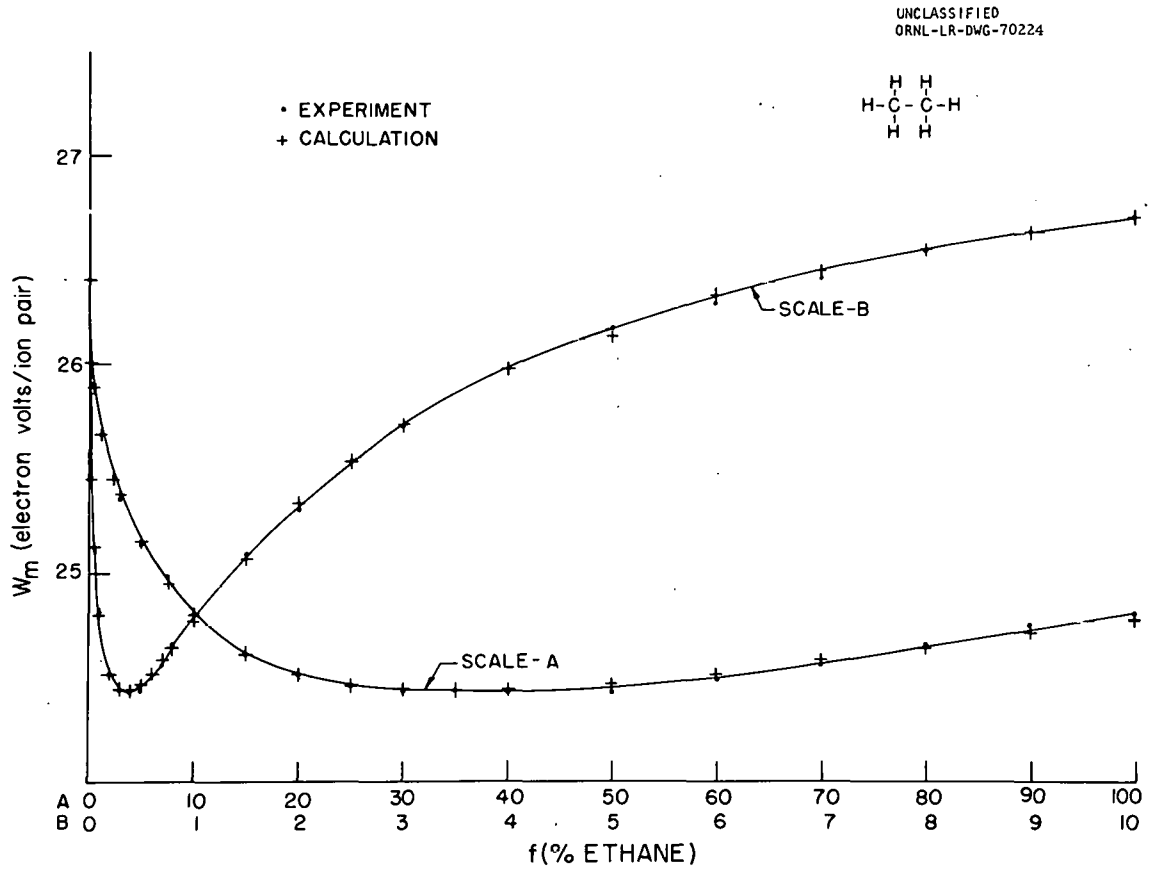


Fig. 9. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Ethane

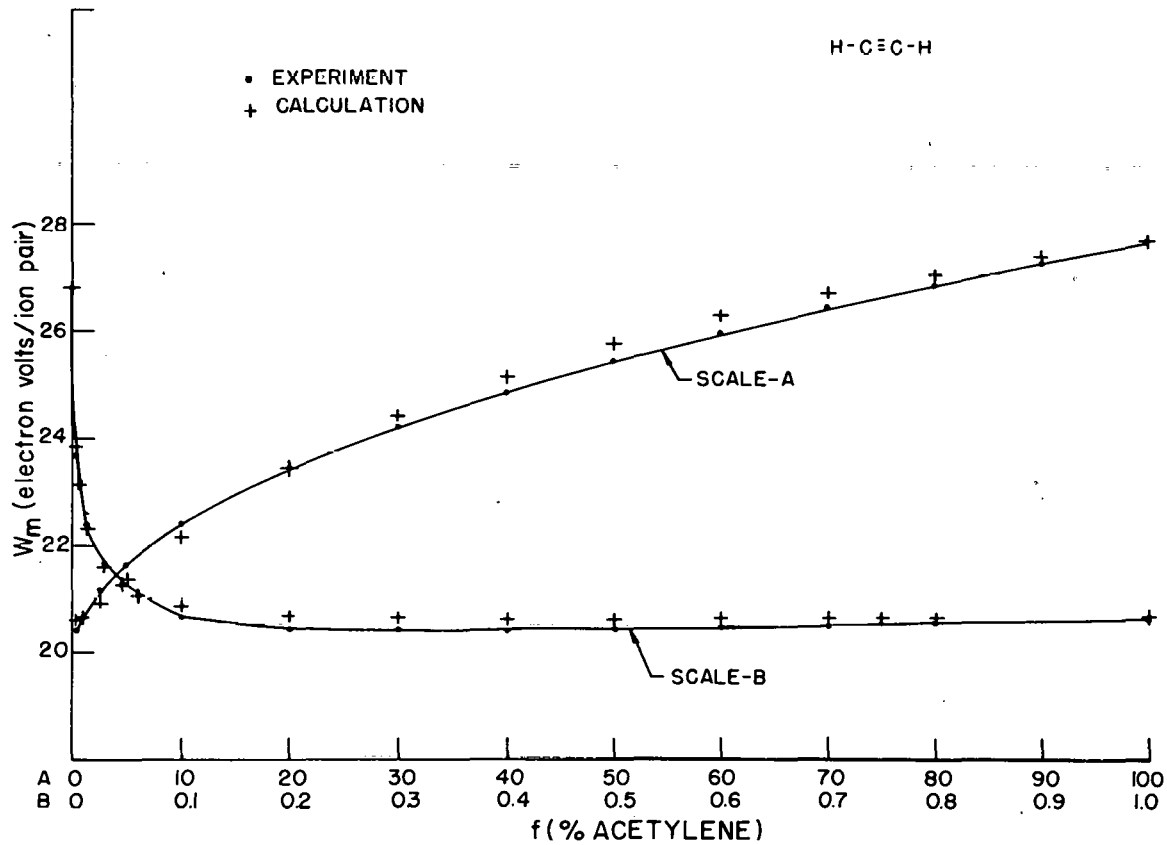
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Fig. 10. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Acetylene

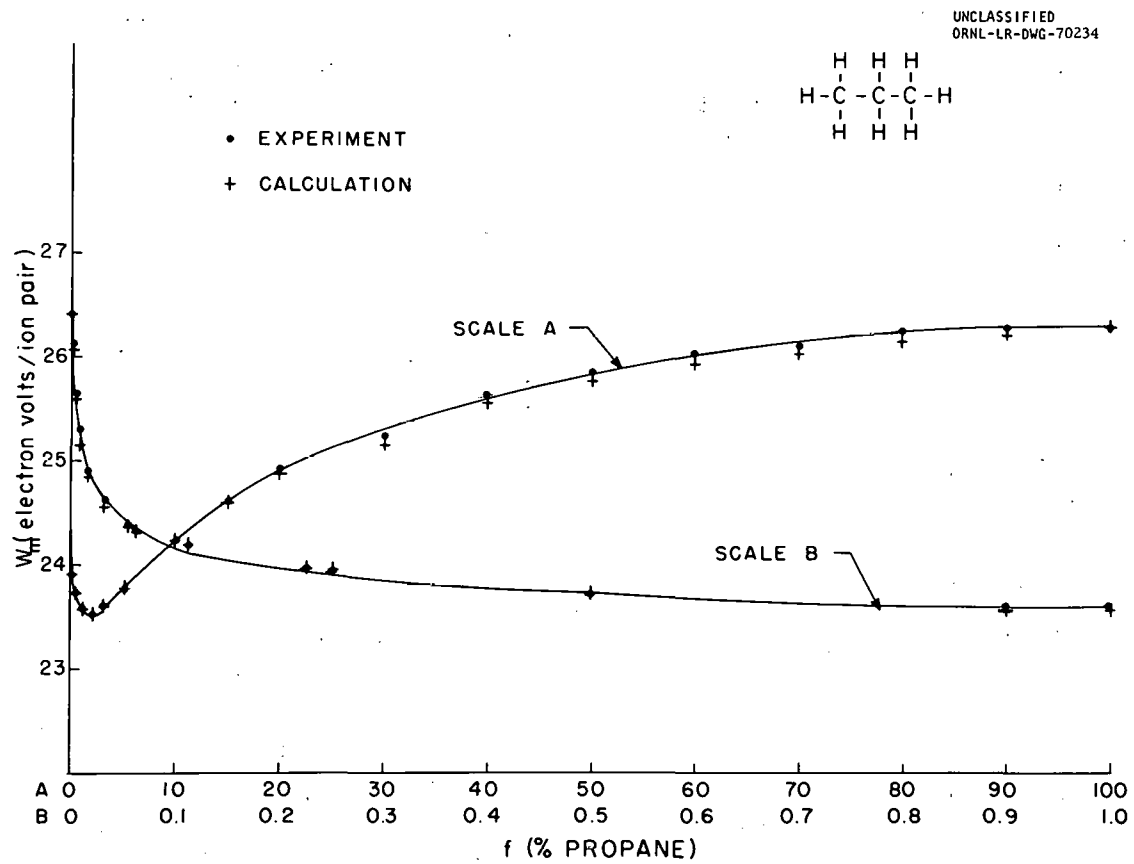


Fig. 11. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Propane

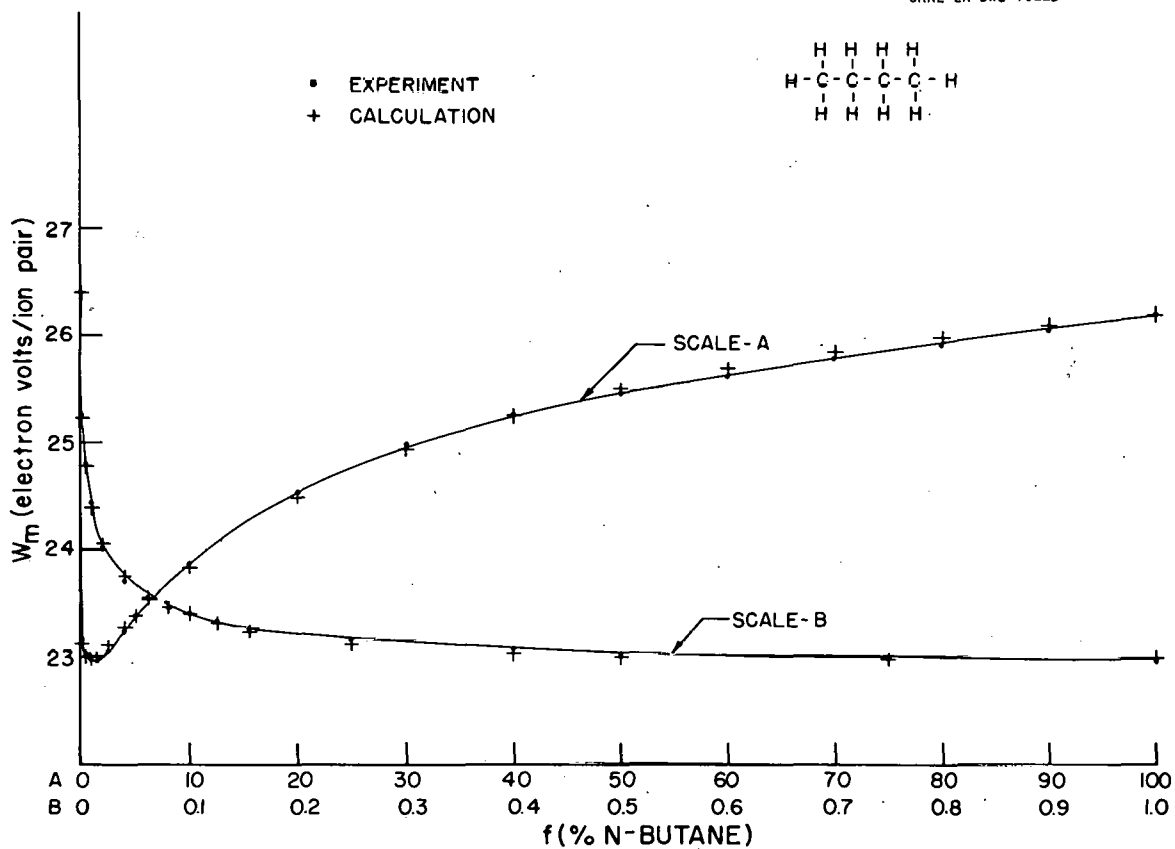
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Fig. 12. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and N-Butane

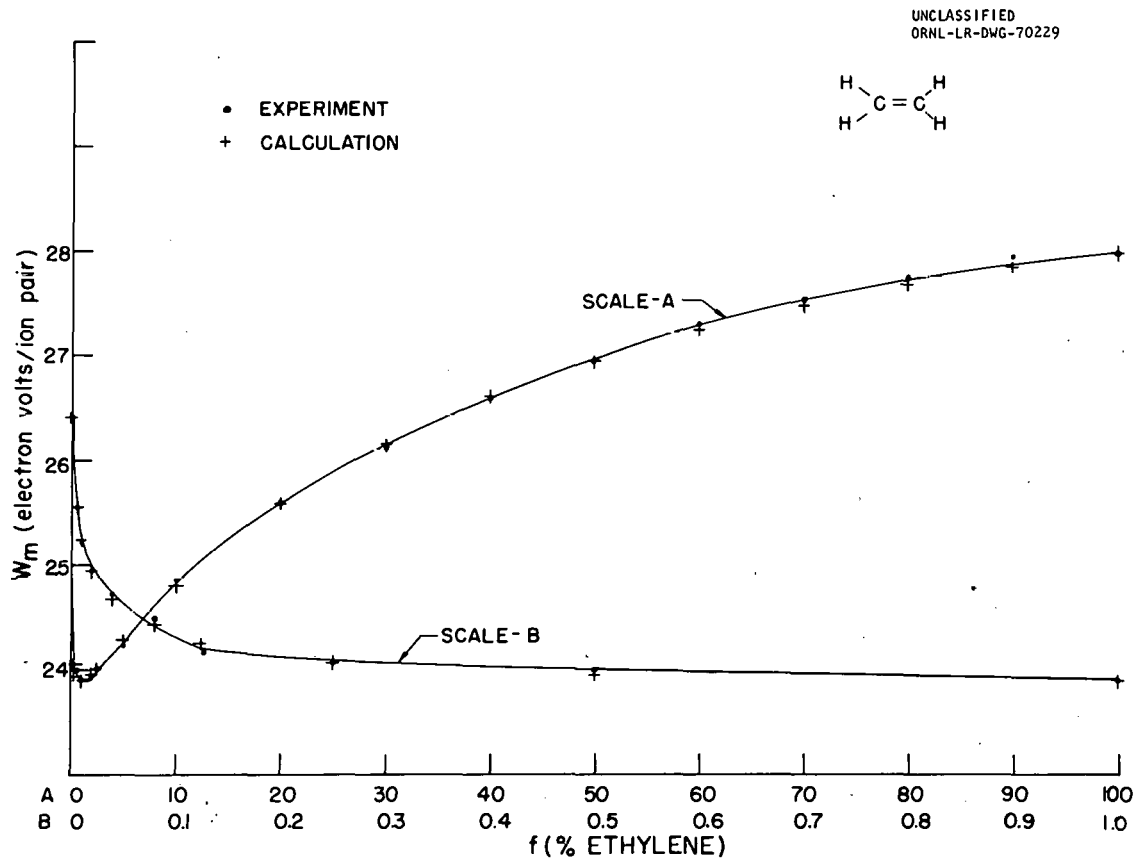


Fig. 13. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Ethylene

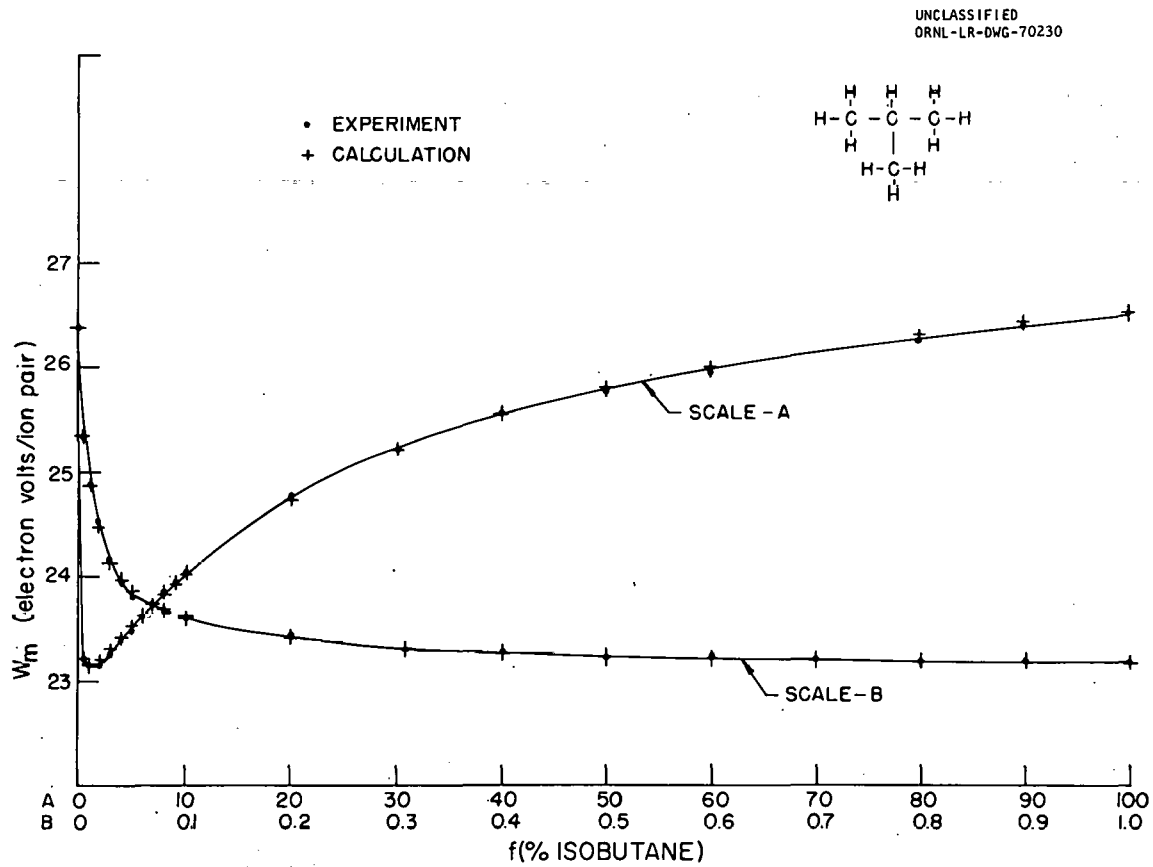


Fig. 14. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Isobutane

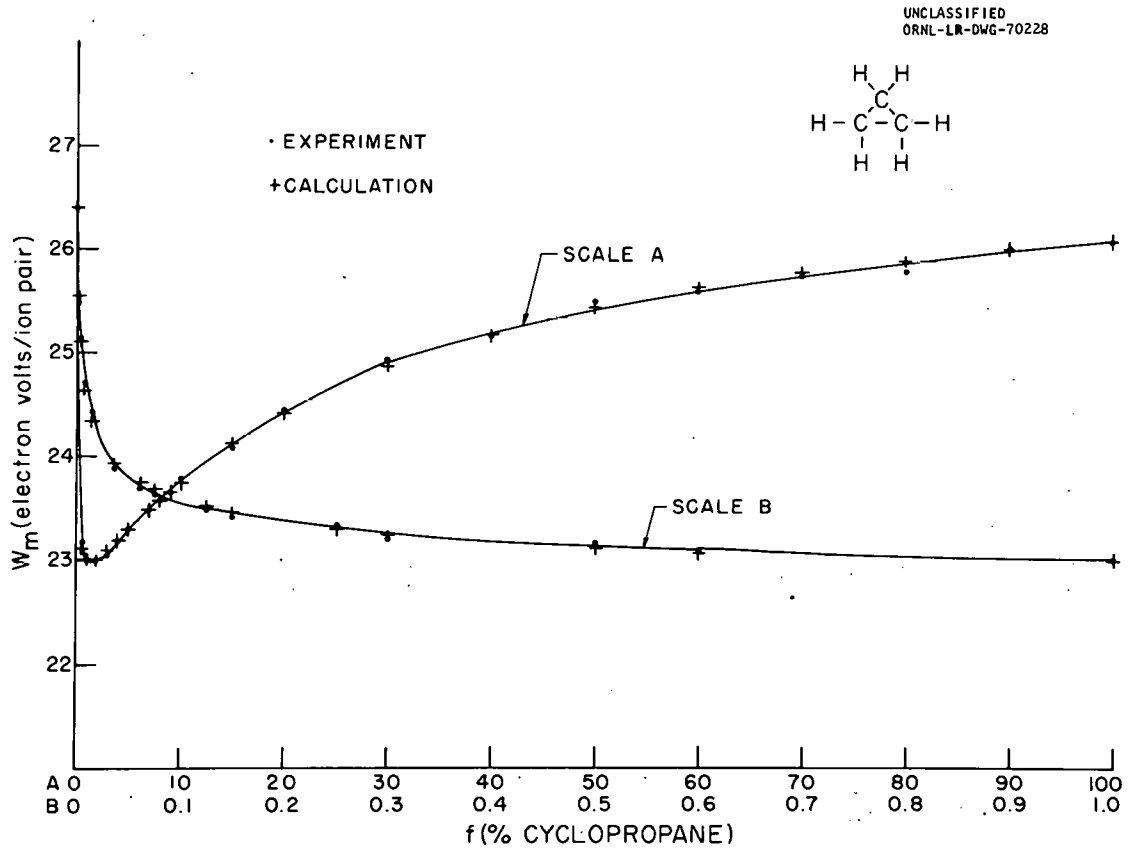


Fig. 15. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Cyclopropane



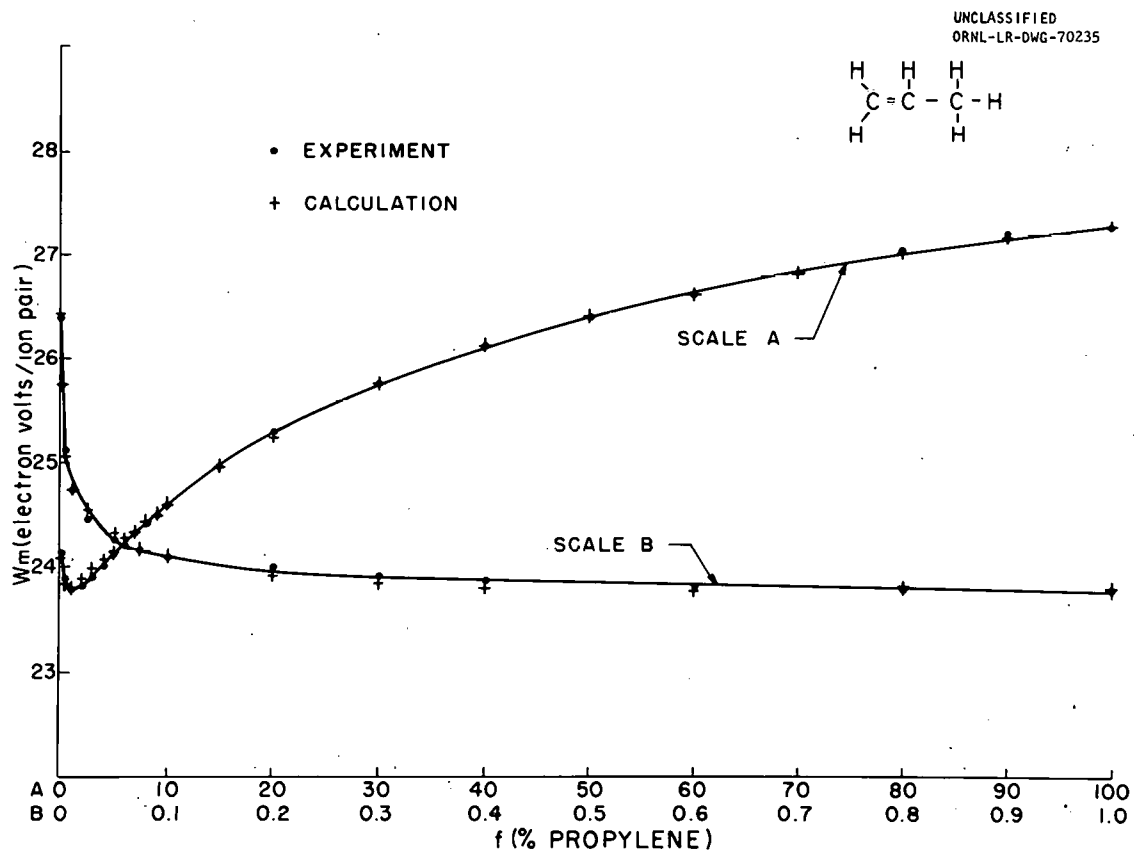


Fig. 16. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Propylene

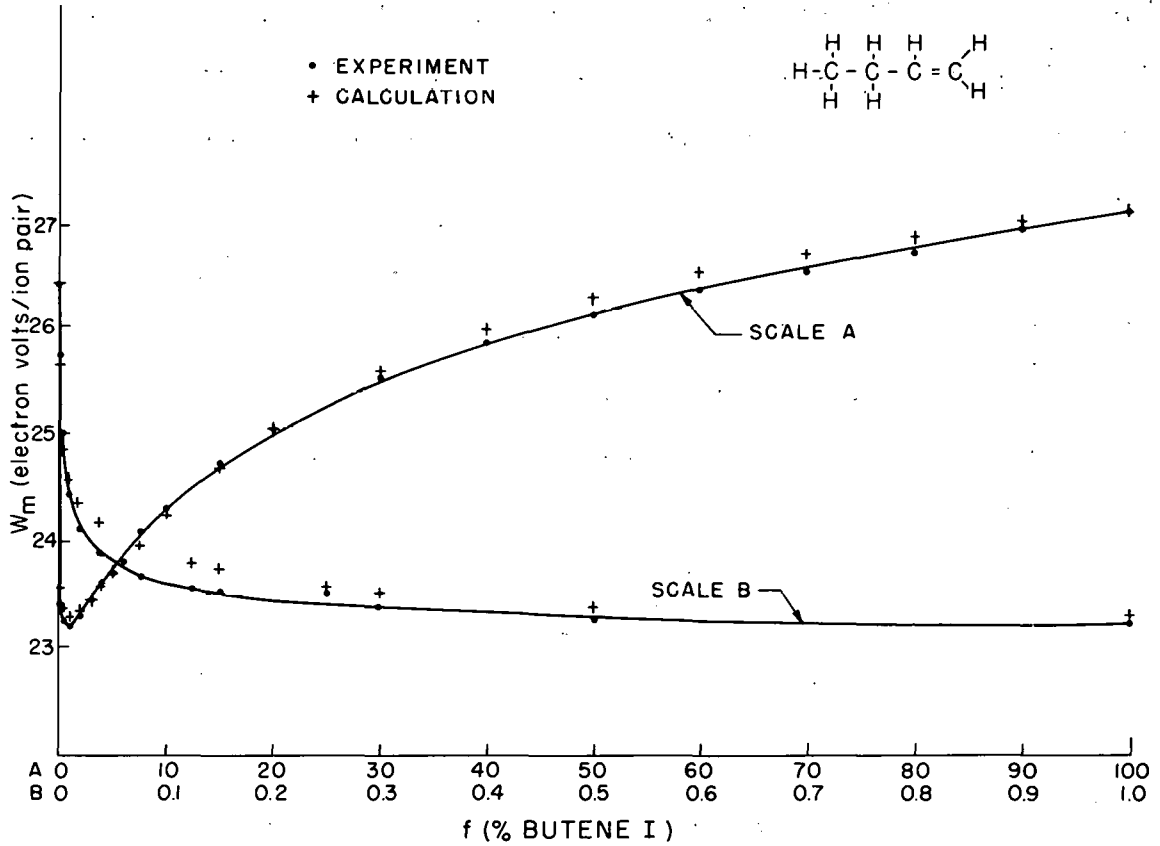
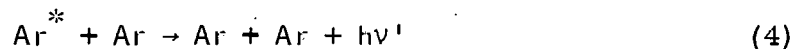
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Fig. 17. Graphical Comparison of the  $W_m$  Values, both Experimental and Calculated, for Mixtures of Argon and Butene I

## V. INTERPRETATION OF RESULTS

In previous work<sup>10,11</sup> it was suggested that the passage of alpha particles through argon leaves argon atoms in two states of excitation, each of which lasts long enough to transfer energy in collision processes. Let us suppose that an alpha particle of energy  $E_0$  produces  $\beta_1 E_0$  states excited at energy  $\epsilon_1$  (about 11.6 ev) and  $\beta_2 E_0$  states excited at energy  $\epsilon_2$  (about 15 ev). Further, we shall assume that either of these states may be depleted by three mechanisms: 1) spontaneous radiation from the initial state, 2) transfer of energy from the initial state due to radiative or radiationless collisions with argon atoms, and 3) transfer of energy in a collision with other kinds of atoms or molecules of general designation X.

According to the assumed model,



where the \* indicates electronic excitation in a general sense, e.g.,  $\text{Ar}^*$  may be either excitation of the level  $\epsilon_1$ , or the level  $\epsilon_2$ , and  $\text{X}^*$  is an unspecified level of X.

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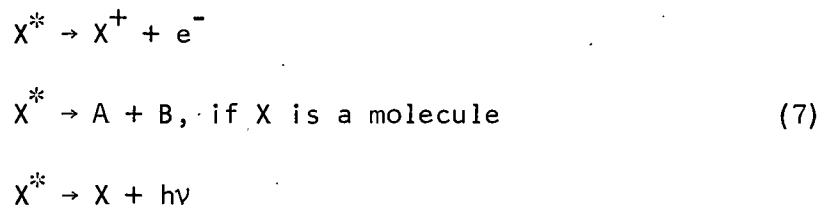
<sup>10</sup>C. E. Melton, G. S. Hurst, and T. E. Bortner, Phys. Rev. 93, 643 (1954).

<sup>11</sup>F. W. Sanders, G. S. Hurst, and T. E. Bortner, "A Study of Alpha Particle Ionization in Argon Mixtures," ORNL-2807 (October 9, 1959).

We are interested in the contribution to the total ionization due to energy transfer to X. In order to obtain this we must first calculate  $N(X^*)$ , the number of energy transfers to X as a function of gas concentration. When  $f_X P$ , the partial pressure of X, is sufficiently small compared to  $f_{Ar} P$ , the partial pressure of Ar, it is easily seen that

$$N^{1,2}(X^*) = \frac{\bar{v}_X \sigma_X^{1,2} f_X}{\bar{v}_X \sigma_X^{1,2} f_X + \bar{v}_{Ar} \sigma_{Ar}^{1,2} f_{Ar} + \lambda / N_0 P} \beta_{1,2} E_0 \quad (6)$$

where the superscripts 1,2 are used to show the validity of the expression for each of the energy levels  $\epsilon_1$  and  $\epsilon_2$ . In Eq. (6)  $\bar{v}_X$  is the average relative velocity of  $Ar^*$  and X,  $\bar{v}_{Ar}$  is the average relative velocity of  $Ar^*$  and Ar,  $\sigma_X$  is the cross section for energy transfer to X (Eq. 5),  $\sigma_{Ar}$  is the cross section for energy transfer to Ar (Eq. 4),  $\lambda$  is the decay constant for spontaneous emission (Eq. 3),  $N_0$  is the number of gas atoms per  $cm^3$  at 1 mm Hg, and P is the pressure in mm Hg. The fraction of  $N(X^*)$  which ionizes is determined by competition of ionization with dissociation and radiation, i.e.,



Thus, if we let  $\eta$  be the probability that  $X^*$  will ionize, the contribution to ionization due to an excited state is  $\eta N(X^*)$ .

Inasmuch as it has been demonstrated that Eqs. (1) and (2) give the number of ion pairs produced per electron volt of energy expended for binary mixtures where excited states are not formed, we may add the contribution due to two excited states to obtain

$$\frac{1}{W_m} = \left[ \left( \frac{1}{W_X} - \frac{1}{W_{Ar}} \right) Z + \frac{1}{W_{Ar}} \right] + \eta_1 \beta_1 Y_1 (1 - Z) + \eta_2 \beta_2 Y_2 (1 - Z) \quad (8)$$

where

$$Z = \frac{f_X}{f_X + a f_{Ar}}$$

$$Y_1 = \frac{f_X}{f_X + \left[ \bar{v}_{Ar} \sigma_{Ar}^1 / v_X \sigma_X^1 \right] f_{Ar}} = \frac{f_X}{f_X + C_1 f_{Ar}}$$

$$Y_2 = \frac{f_X}{f_X + \left[ \bar{v}_{Ar} \sigma_{Ar}^2 / v_X \sigma_X^2 \right] f_{Ar}} = \frac{f_X}{f_X + C_2 f_{Ar}}$$

In Eq. (8) the factor  $(1 - Z)$  is included to remove the restriction in Eq. (6) that  $f_X \ll f_{Ar}$ . This factor may be regarded as a correction for the decrease in the number of excited states formed in argon by the alpha particle in the same sense as "a" is regarded as "effective stopping power ratio" in the empirical Eq. (1). Also, in Eq. (8) the  $\lambda \sqrt{N_0 P}$  term in the denominator of Eq. (6) is omitted since it has been shown<sup>11</sup> that the experimental data are pressure independent. This is equivalent to the condition that

$$\lambda \ll \left( \bar{v}_X \sigma_X f_X + \bar{v}_{Ar} \sigma_{Ar} f_{Ar} \right) N_0 P \quad (9)$$

where  $P \geq 100$  mm Hg.

When experimental data are fitted to Eq. (8), the number of terms retained depends on the ionization potential of X,  $I_X$ .

If  $I_X > \epsilon_2$ , the first term on the right of Eq. (8) is retained, and one parameter,  $\underline{a}$ , is involved.

If  $\epsilon_1 < I_X < \epsilon_2$ , the first and third terms are retained, and three parameters,  $\underline{a}$ ,  $\underline{\alpha}_2 = \eta_2 \beta_2$ , and  $\underline{C}_2 = (\bar{v}_{Ar} \sigma_{Ar}^2 / \bar{v}_X \sigma_X^2)$  are involved.

If  $I_X < \epsilon_1$ , all three terms are retained, and five parameters,  $\underline{a}$ ,  $\underline{\alpha}_2$ ,  $\underline{C}_2$ ,  $\underline{\alpha}_1$ , and  $\underline{C}_1$  are involved. Since the equation for  $W_m$  is symmetric in  $\alpha$  and  $C$ , the sets of values  $\alpha_1$ ,  $C_1$  and  $\alpha_2$ ,  $C_2$  cannot be uniquely associated with the energy levels  $\epsilon_1$  and  $\epsilon_2$ , respectively.

## VI. FITTING THE EXPERIMENTAL DATA

The experimental data points were fed into a computer that was programmed to make a least squares fit and yield, upon convergence, values for the various parameters listed above. The need for a 2-state model is illustrated in Fig. 3. Data for argon-propane were first fitted to Eq. (8) where only one excited state was considered. As seen in Fig. 3, the 1-state, 3-parameter calculation is rather poor. When both states are considered, as they should be since the ionization potential of propane is less than 11.7 ev, the calculated curve agrees quite well with experiment. Accordingly, when  $I_X < 11.7$  ev, the 2-state model was used, and when  $I_X > 11.7$  ev, the 1-state model was used. Figures 4 through 17 show plots of the experimental data and the calculated curves. Table XV lists the values of the parameters obtained in the calculation.

The calculation of the curves for the appropriate number of parameters proceeded in a straightforward manner in all cases except xenon. For xenon, the standard calculation would not converge to a best least squares fit, and the difficulty is probably due to the absence of a minimum in the experimental curve (Fig. 5). Since xenon is an atomic gas (like krypton), it follows from the discussion of ionization efficiencies (see next section) that we would expect the mixtures with xenon and with krypton to have the same values for the parameter  $\alpha_2$ . In this case, the value of  $Z$  in the term of the equation giving the contribution due to energy transfer was allowed to take on a value differing from its value in the first term of the equation,

TABLE XV  
 IONIZATION POTENTIAL AND PARAMETERS USED IN  
 CALCULATING  $w_m$  OF THE GAS MIXTURES

	$I_X$	a	$c_1$	$\alpha_1$	$c_2$	$\alpha_2$
Krypton	13.9	0.412			0.0617	0.00456
Xenon	12.1	0.285 a=0.361			0.0121	0.00456*
Carbon Dioxide (CO <sub>2</sub> )	14.4	0.637			0.0351	0.00209
Methane (CH <sub>4</sub> )	13.1	0.487			0.0160	0.00114
Nitrous Oxide (N <sub>2</sub> O)	12.3	0.521			0.00656	0.00193
Ethane (C <sub>2</sub> H <sub>6</sub> )	11.6	0.167	0.00138	0.00804	0.0122	0.00401
Acetylene (C <sub>2</sub> H <sub>2</sub> )	11.4	0.256	0.0000101	0.00185	0.000117	0.00924
Propane (C <sub>3</sub> H <sub>8</sub> )	11.3	0.158	0.0000611	0.00324	0.00556	0.00255
n-Butane (C <sub>4</sub> H <sub>10</sub> )	10.3	0.210	0.0000296	0.00373	0.000852	0.00234
Ethylene (C <sub>2</sub> H <sub>4</sub> )	10.8	0.255	0.0000552	0.00254	0.00143	0.00193
Isobutane (C <sub>4</sub> H <sub>10</sub> )	10.3	0.195	0.000108	0.00469	0.00437	0.00129
Cyclopropane (C <sub>3</sub> H <sub>6</sub> )	10.2	0.197	0.0000584	0.00429	0.00351	0.00213
Propylene (C <sub>3</sub> H <sub>6</sub> )	9.8	0.262	0.0000137	0.00242	0.000645	0.00206
Butene-1 (C <sub>4</sub> H <sub>8</sub> )	9.7	0.203	0.0000197	0.00329	0.00209	0.00248

\* Fixed parameter.



i.e.,

$$\frac{1}{W_m} = \left[ \left( \frac{1}{W_{Xe}} - \frac{1}{W_{Ar}} \right) Z + \frac{1}{W_{Ar}} \right] + \alpha_{2Kr} \frac{f_{Xe}}{f_{Xe} + C_2 f_{Ar}} (1 - Z')$$

where

$$Z = \frac{f_{Xe}}{f_{Xe} + a f_{Ar}}$$

and

$$Z' = \frac{f_{Xe}}{f_{Xe} + a' f_{Ar}}$$

The calculation for xenon with fixed  $\alpha_2 = 0.00456$  and variable parameters  $a$ ,  $a'$ , and  $C_2$  produced satisfactory results. Thus, the assumption that  $\alpha_2$  has the same value for xenon and krypton is reasonable, provided that some adjustment is permitted of the parameter  $a$ .

## VII. IONIZATION EFFICIENCIES

It is to be recalled that  $\eta$  is the fraction of ionizations produced in the additive gas molecules which are excited by the transfer process. Krypton, being a monatomic gas, is not subject to dissociation. On this basis the value of  $\eta$  for krypton can be taken as 1 if one assumes that the rate of photon emission induced by krypton is negligible. Thus, krypton may be used as a reference for calculating the ionization efficiencies of gases with  $I$  greater than 11.5 ev, i.e., if  $\alpha_{2Kr} = \eta_{Kr} \beta_2 = 0.00456$ , and  $\eta_{Kr} = 1$ , then  $\beta_2 = 0.00456$ . Hence,  $\alpha_{2X} = 0.00456 \eta_{2X}$ , and  $\eta_{2X} = \alpha_{2X}/0.00456$ . The values thus obtained are tabulated in Table XVI.

The values for ionization efficiencies,  $\eta$ , are compared in Table XVI to Weissler's<sup>12</sup> values obtained for gas ionization produced by photons with the same energy as that of the transferring excited state of argon. The corresponding absolute values for  $\eta$  (Table XVI) all fall within a factor of 2, and the relative values compare very favorably.

Since it is not possible to assign uniquely the values for  $\alpha_1$  and  $\alpha_2$  with energy levels  $\epsilon_1$  and  $\epsilon_2$ , we cannot obtain ionization efficiencies from the 2-state calculation for comparison with photoionization efficiencies for many hydrocarbons of interest.<sup>13</sup>

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<sup>12</sup>G. L. Weissler, "Photoionization in Gases and Photoelectric Emission from Solids," in *Handbuch der Physik*, Vol. XXI, S. Flügge, Ed. (Springer-Verlag, Berlin, 1956), pp. 304-382.

<sup>13</sup>R. I. Schoen, "Absorption, Ionization, and Ion Fragmentation Cross Sections of Hydrocarbon Vapors for Vacuum Ultraviolet Radiation," Tech. Report No. 1, Dept. of Phys. U. of Calif. (June 15, 1960).

TABLE XVI  
CALCULATION OF IONIZATION EFFICIENCIES FOR Kr-CH<sub>4</sub>,  
Kr-CO<sub>2</sub>, AND Kr-N<sub>2</sub>O WITH KRYPTON AS 1.0

	Present Results	Weissler's Summary
$\eta_{\text{Kr}}$	1	
$\eta_{\text{CH}_4}$	0.25	0.5
$\eta_{\text{CO}_2}$	0.46	0.9
$\eta_{\text{N}_2\text{O}}$	0.42	0.7

## VIII. DISCUSSION

The amount of ionization produced in mixtures of argon with other atomic and molecular gases is very accurately described by an equation which is based on the assumption that two excited states are produced by the passage of alpha particles through argon. Furthermore, the constants derived from fitting the experimental data to the equation based on the model are quite reasonable in magnitude. Therefore, the indication is rather strong that the notion of two excited states in argon is correct. However, the sets of constants cannot be uniquely associated with the respective energy levels, and the constants  $C_1$  and  $C_2$  involved the ratios of cross sections. Both of these limitations could be removed in properly executed spectroscopic experiments. The study of the emission spectrum due to charged particle irradiation of argon and argon mixtures may also identify the particular excited states involved.

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