

Using the theory of Jones and Furry,<sup>1</sup> the Clusius-Dickel columns have a calculated performance of 0.036 gram per day concentrated 700 times above normal. The Hertz pumps have a calculated separation factor of approximately  $1.8 \times 10^6$ . The product of these is of the order of  $10^9$ . This is high enough that the final percentage in the Hertz is a linear function of time.

Working with well helium on the basis of continuous operation and using the calculated characteristic time of 7 days, a 0.25-g product was taken from the first stage and transferred to the second stage where it was processed in 7 days. The first sample of 30 cc at a pressure of 10 mm had the following analysis:

Helium 3	0.5 percent,
Helium 4	59.5 percent,
Hydrogen	40.0 percent,

as analyzed by Consolidated Engineering Corporation of Pasadena on one of their mass spectrometers.

This initial run indicates that nearly a year would be required to produce 50 percent helium 3; however, theoretical considerations predict that this time can be materially shortened.

The helium was furnished by the U. S. Navy through Contract N6onr-102, Task Order V.

<sup>1</sup> R. C. Jones and W. H. Furry, *Rev. Mod. Phys.* **18**, 151 (1946).

### Fluctuations of Ionization and Low Energy Beta-Spectra

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USING an ( $n, \gamma$ ) reaction in the Chalk River heavy water pile, we have prepared the nuclide  $A^{37}$ , which is known to decay by  $K$ -capture without emission of positrons.<sup>1</sup>  $A^{37}$  was introduced inside various proportional counters filled with a mixture of argon and methane; usually, though not always, 39 cm  $A + 12$  cm  $CH_4$  were used. The cathode diameters, wire diameters, and effective lengths of two typical counters, 1 and 2, were, respectively, 4.67 cm, 0.0047 cm, 19.7 cm for No. 1 and 1.75 cm, 0.0047 cm, 6.9 cm for No. 2. The counters were connected to a linear amplifier, the output of which was fed either into a discriminator or into a 30-channel pulse analyzer. A typical differential curve is shown in Fig. 1, obtained with counter 1 working at 2520 volts. The noise was equivalent to 4500 pair of ions and the multiplication factor in this run was about  $10^4$ . The curve is shown by a broken line below channel 2, because the effective width of channel 1 is not accurately known. The main peak corresponds to an energy of about 2800 ev, the binding energy of a  $K$  electron in the  $Cl^{37}$  atom. Since the Auger electron yield<sup>2</sup> for the  $K$  shell is about 90 percent, about 90 percent of the  $K$ -capture processes result in the emission of soft radiations dissipating the full 2800 ev in the counter.

The low energy peak corresponds to about 200 ev, as was

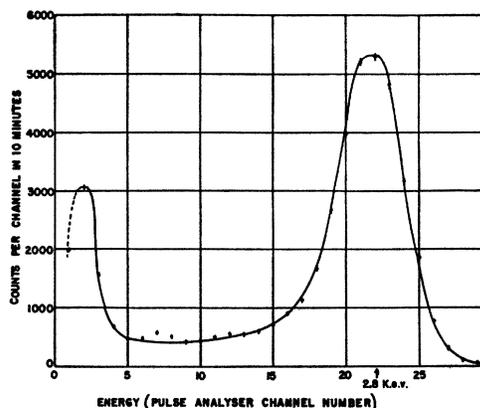


FIG. 1.

determined by runs at higher over-all gain. This is approximately the energy of ionization of the  $L$  shell. We believe that the following two mechanisms contribute in a comparable extent to the low energy peak: (a) Nuclear  $K$ -capture processes resulting in the  $K-\alpha$ -x-radiation being emitted by the  $Cl^{37}$  atom and not being absorbed in the counter. In such processes a photon of about 2600 ev would escape from the counter, while the remaining energy corresponding to the ionization of the  $L$  shell would be dissipated in the counter. (b) Nuclear absorption of an  $L$  electron, the so-called  $L$ -capture, which has been discussed in the literature but not yet observed experimentally.<sup>3</sup> While we plan to discuss in the near future the quantitative aspects of this question, we wish to turn now to the main peak.

1. While the main peak cut-off is very sharp on the high energy side, there is a "tail" on the other side. That this is partly due to the end effects of the counter, resulting in a small value of the multiplication factor near the end, has been shown by a few runs recently obtained with a counter (3) having a ratio of length to diameter three times as great as the one used in the run illustrated in Fig. 1. With counter 3 the cut-off on the low energy side is sharper.
2. That the mean energy necessary for an electron to produce a pair of ions is approximately independent of the electron energy was confirmed by experiments in which the  $K-\alpha$ -x-ray lines of copper were made to fall upon the counter. Although there was a wide distribution of pulses as a result of wall effects, the maximum size, caused by the full energy content of the copper  $K-\alpha$ -quantum ( $\approx 8000$  ev) could be measured on an oscilloscope. The ratio of this maximum pulse size to the main  $A^{37}$  pulse size was  $2.8 \approx 8000/2800$ . It is clear that  $A^{37}$  can provide an ideal "calibration line" for the proportional counter investigation of low energy beta-spectra such as that of  $H^3$ .
3. The width at 60.7 percent of the peak for the 2800-ev line is about 18 percent as determined in various runs. This corresponds to an observed standard deviation of the pulse size of  $\pm 9$  percent. Yet the mean number of pairs of ions produced by a 2800-ev electron is about 100, and if we assume that the variance of the number of

ionizations is equal to the number of ion pairs produced, then we would expect to observe a standard deviation for the pulse size equal to  $\pm(2 \times 100)^{1/2}/100 = \pm 14$  percent,\* i.e., definitely greater than the observed value. In fact, according to a theoretical estimate<sup>4</sup> of Fano, the variance of the number of ionizations is expected to be substantially smaller than that governed by a Poisson distribution. Our measurements show that in our  $A + CH_4$  mixture such a variance must be at least 2.4 times smaller; this value is obtained on the assumption that the only cause of spread in the peak is of a statistical nature. Such an assumption, under certain conditions, is justified in a proportional counter because of the negligible value of the noise in comparison with a signal produced by only a few pair of ions. It is this feature which has made possible the study of the fluctuations in the number of ionizations; in a pulse chamber without gas amplification, the minimum detectable pulse corresponds to such a great number of ion pairs that the spread in pulse size caused by fluctuations of ionization would be masked by instrumental spread.

In conclusion we wish to point out that the technique of the proportional counter at high multiplication factor should be useful not only in beta-ray problems such as orbital electron capture and continuous beta-spectra at low energy, but also in x-ray applications such as measurements of fluorescence yields.

<sup>1</sup> P. K. Weimer, J. D. Kurbatov, and M. L. Pool, Phys. Rev. **66**, 209 (1944).

<sup>2</sup> A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935), pp. 477-92.

<sup>3</sup> R. E. Marshak, Phys. Rev. **61**, 431 (1942); E. Segré, Phys. Rev. **71**, 274 (1947); R. Bouché, R. Daudel, P. Daudel, and R. Muxart, J. de Phys. et Rad. **8**, 336 (1947).

\* The factor 2 arises from the variance introduced by the multiplication process in the counter (H. S. Snyder, Phys. Rev. **72**, 181 (1947); also "Statistics of multiplicative processes," an as yet unpublished paper by O. R. Frisch.

<sup>4</sup> U. Fano, Phys. Rev. **72**, 26 (1947).

### The Magnetic Susceptibility of Uranium Tetrafluoride, $UF_4$ \*

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THE magnetic susceptibility of uranium tetrafluoride,  $UF_4$  has been measured over the temperature range 74.5°K–294.5°K. The  $UF_4$  was prepared by the conversion of Merck's "Reagent" Grade  $UO_2(NO_3)_2 \cdot 6H_2O$  to  $UO_3$ , reduction of the  $UO_3$  to  $UO_2$ , and subsequent hydrofluorination to form  $UF_4$ . These reactions were followed by measuring the changes in weight with each process. The  $UF_4$  was analyzed gravimetrically for U. The result of three analyses was  $76.08 \pm 0.43$  percent U. The theoretical value is 75.80 percent, based on a molecular weight of 314.07. The magnetic measurements were made on a Gouy balance with the salt in the form of a powder. The results, corrected for diamagnetism, are shown in Fig. 1. A Weiss-Curie law is obeyed over the whole temperature range in which observations were made.

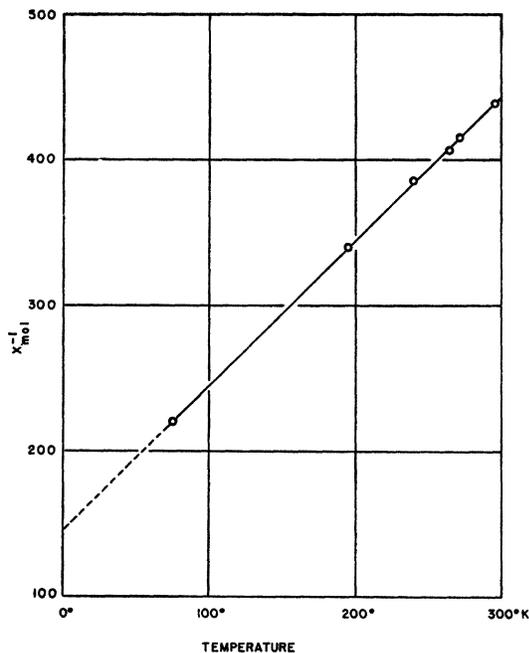


FIG. 1. Magnetic susceptibility of  $UF_4$  as a function of temperature (corrected for diamagnetism).

Writing  $\chi = C/(T + \Delta)$ ,  $\Delta$  is calculated to be 146°K, and  $C$  to be 1.00. Using the equation  $\mu = 2.83(\chi_M(T + \Delta))^{1/2}$ , the magnetic moment of the  $U^{4+}$  ion is found to be 2.83 Bohr magnetons, with an average deviation of 1 percent as determined by measurements on three samples.

If it is assumed that the  $U^{4+}$  ion has Russell-Saunders coupling, and evidence for this is furnished by the work of Kiess, Humphreys, and Laun,<sup>1</sup> one concludes from the experimental data that the crystalline field has broken down the spin-orbit coupling, and that a non-magnetic state lies considerably below all other states. The spin,  $S$  of two unpaired electrons remains free to contribute to the magnetic moment with a value  $\mu = 2(S(S+1))^{1/2}\beta = 2.828\beta$ , in good agreement with the observed moment.

The experimental results obtained for uranium tetrafluoride are similar to those found for compounds of the first row transition elements, and suggest that in this case the  $U^{4+}$  ion is in a  $^3F_2$  state with two  $6d$  electrons whose orbital contribution to the magnetic moment has been quenched by the crystalline field.

Basic states for the  $U^{4+}$  ion probably belong to the configuration  $5f^2$ ,  $5f6d$ , or  $6d^2$ . Assuming the orbital moment to make its full contribution, no magnetic moment could be calculated for these states which was compatible with the above data. This is true for  $jj$  as well as for  $LS$  coupling.

The magnetic susceptibility of  $UF_4$  does not provide a direct answer to the question of the spectroscopic state of the  $U^{4+}$  ion in this compound. The quenching of the orbital contribution to the moment is easily accounted for if the basic state is  $^3F_2$ , arising from the configuration  $6d^2$ , but not if it is  $^3H_4$ , coming from the  $5f^2$  configuration. The investigations of Wu and Goudsmit<sup>2</sup> and of M. Goepfert Mayer<sup>3</sup>