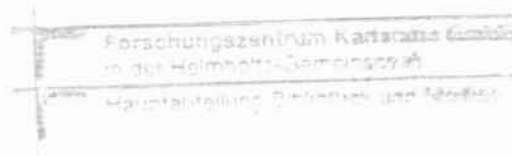


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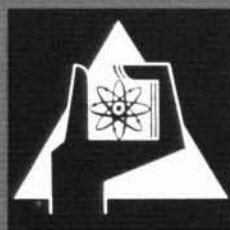


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Measurement of Thermal Diffusion and Determination of the Intermolecular Potential of Gaseous Uranium Hexafluoride

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THERMAL diffusion in liquid UF_6 has been well known for about 20 years,¹ but as far as we know the existence of thermal diffusion in gaseous UF_6 has not yet been proved. For example, Nier² and Groth³ did not obtain any measurable isotope separation using thermal-diffusion columns of the Clusius-Dickel type on gaseous UF_6 . On account of the high precision of modern special mass spectrometers for UF_6 , it seemed promising to take up the problem once more.

The experiments were carried out at various temperatures and gas pressures in a 3-m-long thermal-diffusion column made of concentric nickel tubes having a distance of 4 mm between the hot and the cold wall. The separation effects given by

$$q-1 = [n_t(1-n_b)/n_b(1-n_t)] - 1$$

(n_t , n_b are the mole fractions of $^{235}UF_6$ at the top and the bottom of the column, respectively) were found to be in the range of 5×10^{-4} to 4×10^{-3} and have been measured with a relative accuracy of about $\pm 5\%$.

The thermal-diffusion factor α has been calculated from the measured isotopic separations by using the theory for the plane thermal diffusion arrangement.⁴ For this purpose the pressure dependence of the stationary separation factor q on UF_6 pressure was measured for fixed wall temperatures. Within the limits of experimental error, this measured pressure dependence was in agreement with the expected theoretical relationship for all experiments, and therefore it was possible to calculate the thermal-diffusion factor α without using any other transport quantity of the gas. The values of α obtained by this method were taken to be at the arithmetic mean temperature. Jones and Furry⁴ showed this temperature attribution to be a good approximation, particularly because in the investigated temperature range the viscosity and thermal conductivity of UF_6 are practically linear functions of the absolute temperature.

The physical performance of the column and the

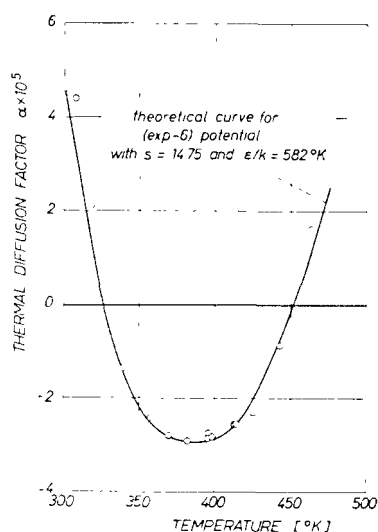


FIG. 1. The thermal diffusion factor α of gaseous UF_6 vs the absolute temperature

validity of the applied evaluation method have been checked in experiments with argon of natural isotopic composition. In these experiments the pressure dependence of the stationary separation factor practically coincided with the theoretical function, and the thermal-diffusion factor of argon determined by the above method reproduced within the limit of error a value which was measured by Stier⁵ using a two-bulb-apparatus.

In Fig. 1 the experimental values for the thermal-diffusion factor α of gaseous UF_6 are plotted vs the absolute temperature T . This observed temperature dependence of α can be represented by the kinetic theory of gases if a modified Buckingham potential (exp -6) is assumed for the molecular interaction.⁶ Using the numerical calculations of Saxena and Mason⁷ and fixing the parameters $s = 14.75$ and $\epsilon/k = 582^\circ K$ for

this potential, the theoretical curve presented by the full line in Fig. 1 is obtained.

It can be seen that within the temperature range investigated an excellent fit with the experimental values is possible.⁸ The third parameter r_m of the potential used has no influence on thermal diffusion and has to be determined from measurements of a different transport coefficient or from measurements of the second virial coefficient. By fitting the theoretical viscosity function⁹ to an experimental viscosity value of gaseous UF_6 $\eta_{UF_6}(T=407^\circ K)=227 \mu P$ ¹⁰ and using the former values of s and ϵ/κ , one obtains $r_m=5.44 \text{ \AA}$.

Now, for the completely specified intermolecular potential all transport coefficients⁹ and the second virial coefficient¹¹ of gaseous UF_6 have been calculated as functions of temperature. Within the available experimental data given by different authors,¹⁰ the results show complete agreement between theoretical and measured values of viscosity, thermal conductivity, self-diffusion, and also with experimental values of the nonideality parameter of gaseous UF_6 . This parameter is related to the second virial coefficient. Its values have been derived by Magnuson¹⁰ from his measurements of the dielectric constant of UF_6 .

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¹ P. H. Abelson, "Liquid Thermal Diffusion," U. S. Atomic Energy Commission Report TID 5229 (1958).

² A. O. Nier, University of Minneapolis Report A-41 (1941).

³ W. Groth, Z. Elektrochem. **54**, 5 (1950).

⁴ R. C. Jones and W. H. Furry, Rev. Mod. Phys. **18**, 152 (1946). R. Fleischmann and H. Jensen, Ergeb. Exakt. Naturw. **20**, 121 (1942). As the actual ratio of the radii was 1.42, any corrections for cylindrical geometry are negligible.

⁵ L. G. Stier, Phys. Rev. **62**, 548 (1942).

⁶ See for example: L. Waldmann, *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Heidelberg, Germany, 1958), Vol. 12, p. 433.

⁷ S. C. Saxena and E. A. Mason, J. Chem. Phys. **28**, 623 (1958).

⁸ Using a Lennard-Jones (12-6) potential (for which numerical computations of the transport coefficients also exist)⁷ the measured temperature dependence cannot be described even approximately. Although this model also predicts a minimum of α in the range of negative values, the absolute value of α in this minimum would differ by about one order of magnitude.

⁹ E. A. Mason, J. Chem. Phys. **22**, 169 (1954).

¹⁰ All experimental data for gaseous UF_6 have been taken from R. DeWitt, "Uranium Hexafluoride, A Survey of the Physico-Chemical Properties," Goodyear Atomic Corporation, Portsmouth, Ohio, USAEC-Report GAT 280 (1960).

¹¹ W. E. Rice and I. O. Hirschfelder, J. Chem. Phys. **22**, 187 (1954).