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SUMMARY

RADIOELEMENTS

Alpha-Beta Branching Ratio of Actinium-227 Preliminary results, based on the relative alpha activity of actinium-227 in secular equilibrium with polonium-215, indicate that the alpha-beta branching ratio of actinium-227 is 1.8%

ISOTOPE SEPARATION AND PURIFICATION

Carbon Isotope Separation

Chemical Exchange Exchange reactions conducted using dimethyl formamide as a solvent resulted in poor separation and excessive decomposition of the amine

The effects of solvent changes on the carbon-13-oxygen-18 ratios were compared. As solvent polarity decreases, the oxygen-18 depletion tends to decrease at the carbon-13 product end of the column. Carbon dioxide analyses show no conclusive evidence regarding the erratic separation factors obtained using isopropanol.

Neon The three-column (36-ft) neon tube bundle was put into continuous operation and an enrichment of 22.9% neon-21 was obtained. A gas transfer system was installed between the neon-21 product stream of the 96-ft cascade and the feed line to the tube bundle to assure immediate use of the neon-21 enriched feed gas for the neon tube bundle, thus avoiding a costly gas inventory.

Argon The argon-36 product rate was decreased from 1.6 ml/hr to 1.0 ml/hr. This change caused the argon-38 at the top stream of the five-stage cascade to decrease to 1.93%. A faster build-up of the argon-38 peak in the cascade was achieved, and it was lowered in the cascade as well. A maximum concentration of 29.8% argon-38 was obtained.

Krypton Using an operating pressure closer to that for maximum separation for final batch operation, krypton-86 was enriched on the fifth pass in a two-stage cascade to 95%.

Xenon

Thermal Diffusion Approximately 400 ml of xenon enriched to 2.62% xenon-124 and 1.05% xenon-126 were drawn off the six-stage tube bundle cascade. Upon subsequent equilibration, the highest concentration of xenon-124 achieved was 5.04%.

Using 71.75% xenon-136 as second-pass fission-product feed, the eight-stage swing unit has enriched xenon-136 to 88.37% and has depleted xenon-131 to 0.12% after three weeks of operation.

Chromatography The experimental krypton-xenon chromatograph is now being used to purify krypton having xenon as a contaminant. The purified krypton was trapped on charcoal at 77°K in one test run.

Thermal Diffusion Research

Molecular Beam Scattering Computer programs have been written for the calculation of theoretical quantum mechanical scattering intensities for experimental correlation. The inclusion of the finite velocity spread obtained in a real experiment is treated by velocity averaging the monoenergetic calculations. The observation of quantum effects is discussed in connection with the proposed velocity selector design.

Collision Integrals Collision integrals were calculated for six helium-helium intermolecular potentials which have been proposed in the literature. The integrals obtained were, in turn, used to calculate helium viscosities and thermal conductivities from 200-2500°K. A comparison with experimental data shows that none of the potentials are entirely satisfactory over the entire temperature range. However, a very encouraging result was demonstrated by the sensitivity of the calculated transport properties to the parameters in the potential function.

Transport Coefficients New data for the $\text{Ne}^{20}\text{-Ne}^{22}$ separation in the 6-ft (183-cm) experimental thermal diffusion column were in good agreement with theory. These experiments, designed to eliminate end effects, showed that end effects were not important in earlier experiments with this same system.

Transport Properties A series of experimental determinations of the thermal diffusion factor of neon-xenon mixtures was started. This mass range can be analyzed better on the mass spectrograph than the systems studied previously. The preliminary results are within the scatter of the literature values for the thermal diffusion factor.

Cascade Theory A computer program to calculate the performance of multicomponent isotope separation systems was written and checked out. Trial calculations were made for neon and argon separation cascades.

RADIOELEMENTS

Basic and applied research is being conducted on a number of radioelements to determine their physical properties, to develop analytical techniques, and to study the basic radiochemistry involved. Of particular interest are alpha emitters, their decay chains, their isotopes, and their chemical homologs. Research quantities of some of these isotopes are being isolated.

ALPHA-BETA BRANCHING RATIO OF ACTINIUM-227

Actinium-227 (half-life = 21 7714 yr¹) decays primarily by beta emission to thorium-227 (half-life = 18 719 days²), an alpha emitter. Approximately 1% of the decay of actinium-227 is by alpha emission, yielding francium-223 (half-life = 22 02 min³), a beta emitter.

The alpha branching ratio of actinium-227 was first reported by Peterson and Ghiorso⁴ to be 1.25%. A more recent value is 1.45%.⁵ Both of these results were obtained by measuring the alpha activity of purified actinium in gridded ionization chambers and comparing it with the alpha activity of the actinium decay products during the early growth period.

A more direct method consists of comparing the relative alpha activity of actinium-227 and polonium-215 in secular equilibrium. Figure 1 indicates that the branching ratio may be as high as 2%. The principal difficulty in arriving at an exact value is that the low-energy "tail" of the radium-223 alpha spectrum extends under the actinium-227 peak. The radium-223 can be almost completely eliminated by the method of residue adsorption,⁶ the actinium equilibrium mixture is dried on a platinum disk, converted to a fluoride with 1N HF and leached with 0.01N HF. However, when the radium-223 is removed, the polonium-215 also disappears (Figure 2).

To resolve this dilemma, the radium-223 was first separated from the actinium-227 and thorium-227 to permit definition of the actinium-227 peak. The hydrofluoric acid leachings were retained and, after the actinium-227 spectrum was obtained, the radium-223 was returned to the sample plate and a second spectrum was obtained, representing the synthetic equilibrium mixture. A preliminary run using this technique yielded a branching ratio of 1.8%.

¹MLM-1155 p. 12

²MLM-1254 p. 10

³MLM-1266 p. 9

⁴S. Peterson and A. Ghiorso, Paper 1910 in "The Transuranium Elements," Nat. Nuclear Energy Ser. IV-14B, Part II, New York: McGraw-Hill Book Co., 1949, p. 1395.

⁵M. Nurmi, D. Giessing, and W. Sievers, Ann. Acad. Sci. Fennicae Ser. A VI No. 167, 1-11 (1965).

⁶H. W. Kirby, J. Inorg. & Nuclear Chem. 25, 483 (1963), and 27, 1701 (1965).

Figure 1. Alpha Spectrum of Actinium-227 and Decay Products.

Figure 2. Alpha Spectrum of Actinium-227 and Thorium-227 (0.01N HF Residue).

ISOTOPE SEPARATION AND PURIFICATION

Under the sponsorship of the Division of Research, Mound is responsible for furnishing material to be used in the physical sciences to further the progress of science and technology in the public interest. Purified helium-3 and isotopes of all other noble gases as well as carbon-13 are available for distribution. After recovery, purification and concentration, these isotopes are furnished to installations for research programs in the physical sciences. Methods of separation and purification are being studied.

CARBON ISOTOPE SEPARATION

Chemical Exchange Previous attempts to improve the separation factor of carbon-13 in the chemical exchange system by changing the carrier solvent for the amine indicated that, where improvement was achieved, either the boiling point of the solvent or the character of the solvent itself was responsible for the improvement. In view of this, several reaction runs were conducted with monoethanolamine using the aprotic solvent dimethyl formamide. This solvent is dipolar but has a higher boiling point than does isopropanol.

There was very little evidence of any isotopic separation in these reaction runs and, upon analysis of the amine solution, it was learned that some decomposition had occurred. This decomposition was due to the excessive heating of the amine solution in the boiler and also to the added chemical reactivity of the solvent. Therefore, further experiments with this solvent were abandoned.

The use of isopropanol as the solvent in the separation experiments provided some information regarding the exchange reaction in addition to increasing the separation factor of carbon-13. As noted in all earlier work, the enrichment of carbon-13 occurring during the carbon dioxide-carbamate exchange was always accompanied by a depletion of oxygen-18 at the same reflux end of the column. In the experiments where methanol was used as a solvent, the increase in the separation factor of carbon-13 was accompanied by a predictable corresponding decrease in the oxygen-18. When the solvent was changed to isopropanol, the carbon-13 separation factor was increased considerably, even though the results were somewhat sporadic. The oxygen-18 depletion, however, did not show any similar decreasing trend as the carbon-13 separation factor increased. Regardless of the carbon-13 separation factor, the oxygen-18 depletion did not fall below 0.25% while values as low as 0.15% oxygen-18 were obtained with methanol (the oxygen-18 content of natural carbon dioxide is 0.37%).

When a mixed solvent of equal parts of isopropanol and methanol was used, the results were intermediate between those obtained using either solvent alone. It appears that the presence of isopropanol allows the carbon-13 separation factor to increase while the oxygen-18 separation factor remains nearly constant.

The rather sketchy evidence provided by these experiments suggests that the increase in the carbon-13 separation factor resulted not simply from an improvement in carbamate decomposition, but also, and probably more importantly, from a change in the exchange reaction mechanism. A more complete decomposition of carbamate would not, of itself, affect the oxygen-18 separation factor in the direction indicated. A change in the reaction mechanism caused by the less polar isopropanol solvent could very easily affect the separation characteristics of both isotopic species. Because of this evidence, therefore, further experimentation with less polar solvents will be conducted to gain a better insight into the causes of these changes.

During a reaction run using isopropanol, amine samples were withdrawn from the boiler each time a carbon dioxide sample was taken. These amine samples were analyzed for carbon dioxide to determine if the erratic results obtained using this solvent were caused by intermittent lapses in carbamate decomposition. The percentage of carbon dioxide, while still higher than desired, showed no dramatic day-to-day change. When compared with the separation factors obtained for the carbon dioxide samples for each corresponding analysis, no definite trend appeared.

NEON

Neon-21 enriched to 10.0% in the 96-ft cascade (Figure 3) was used as feed to the three tube bundle columns. With the tubular heaters at 750°C the operating pressure was set at 1000 torr, which was verified by static experiments to be the maximum separation pressure. Neon-20 and neon-22 product rates were set at a flow of 0.6 ml/hr. The product flows were determined by the rate of pressure rise in calibrated vessels. The feed gas for the continuous feed system on the bundle was obtained directly from the 96-ft cascade by using the new transfer system shown in Figure 4.

Figure 3. The Complete Neon System Including the Present Four-Column System and the New Three-Column Bundle

Figure 4. Gas Transfer System Used between the 96-ft Cascade and the 36-ft Tube Bundle.

Neon-21 product gas from the 96-ft cascade is stored in a compressor which is a sealed refrigeration unit and has a dead volume of approximately 1.7 liters. It would require a pressure greater than 1000 torr or a volume of 1.9 liters of gas with a compressor at a temperature of 50°C to feed the tube bundle directly from this compressor. At the present product rate of 1.20 ml/hr it would take several months to fill the compressor to 1000 torr and a large inventory of gas would be idle in the compressor. Thus the transfer system was built to eliminate this large gas inventory and time loss. When pressure in the compressor reaches 300 torr the gas is pumped into a 75-ml tank using a Randolph pump which has negligible hold-up volume. The tank is immersed in liquid nitrogen which lowers the tank pressure by a factor of 4.0, increasing the pumping speed. The compressor is pumped down to a pressure of 20 torr. When pumping is stopped the tank is warmed to room temperature. Since the tank volume is small relative to the amount of gas transferred, the pressure reaches 20 to 30 psig, which is sufficient to feed the bundle for several days. This procedure is performed twice weekly to assure a sufficient amount of gas in the 75-ml tank. In this manner, essentially all of the 10% neon-21 is put directly in the bundle where it can be further enriched.

After one week of continuous operation, the neon-21 profile at the middle of the 36-ft bundle was 22.9%. The top and bottom product streams contain essentially 100% neon-20 and 100% neon-22, respectively.

ARGON

To reduce the concentration of argon-38 at the top of the five-stage argon cascade, the product flow rate was reduced from 1.60 ml/hr to 1.0 ml/hr. This reduction in flow rate caused a build-up of argon-36 in the top, which pushed the argon-38 down the column. The present concentration of argon-38 at the top of the column is 1.93%. Changes in the argon-38 profile are shown in Figure 5.

Figure 5. Argon-38 Profile in the 11-Column, 5-Stage Cascade

The peak is expected to be further shifted down the column as the cascade is operated under present conditions until a steady state is obtained. At that time the argon-38 concentration at the bottom of the cascade will increase to a value at which the isotope will be removed in the argon-40 product stream at the same rate that it is transported into the cascade from the feed. At this point, a side stream of enriched argon-38 can be withdrawn or all of the gas in the cascade between 72 and 96 ft can be removed.

KRYPTON

A two-column, two-stage thermal diffusion cascade is being used for enriching krypton-84 and krypton-86. With continuous draw-off in the previous passes for enriching krypton-86, the columns were operated at an average pressure of 310 torr, the calculated pressure for 90% maximum separation work. On the present pass, which will be essentially batch operation, the pressure was reduced to about 200 torr, which approaches the pressure at which the separation factor is a maximum. After equilibration, the concentration at the bottom of the cascade reached 95.9% krypton-86.

XENON

Thermal Diffusion Natural xenon is being used as feed in a 24-column, 6-stage thermal diffusion concentric tube cascade to enrich the light mass fraction. Draw-off was started after the xenon-124 concentration reached 2.8%, and approximately 400 ml of product gas were obtained. This material had a concentration of 2.62% xenon-124 and 1.05% xenon-126. After removal of this product, the feed was permitted to circulate with no draw-off from the top. Table I shows the isotopic composition of the equilibrated product as well as of natural xenon for comparison. Figure 6 shows the change in the xenon-124 enrichment without draw-off

Table I
COMPARISON OF ENRICHED LIGHT FRACTION WITH NATURAL XENON

Isotope	Natural Xenon (mole %)	Enriched Xenon (mole %)
124	0.096	5.04
126	0.090	1.70
128	1.92	9.79
129	26.44	64.41
130	4.08	4.31
131	21.18	9.47
132	26.89	4.91
134	10.44	0.32
136	8.87	0.05

Figure 6. Xenon Tube Bundle Operation with No Draw-Off.

Draw-off of 5% xenon-124 was started at an initial rate of 1.7 ml/hr. This is the draw-off rate for 7.35% xenon-124 based on theoretical calculations. When analytical data become available, the draw-off rate will be adjusted accordingly to maintain at least 5% xenon-124. After approximately 0.5 liter has been withdrawn, further enrichment of the xenon-124 to higher concentrations with no draw-off is planned.

The eight-column, eight-stage, hot-wire thermal diffusion cascade with a modified Trennschaukel gas circulating system has been operating for about one month using enriched fission-product feed gas containing 71.75% xenon-136. This second pass is expected to enrich the xenon-136 to 90%. After three weeks the xenon-136 was enriched to 88.73% and the xenon-131 was depleted to 0.12%. The enrichment and depletion of the isotopes are shown in Figure 7.

Figure 7. Eight-Stage Hot-Wire Thermal Diffusion Cascade Using Enriched Feed in the Second Pass

Chromatography Xenon obtained as a reactor off-gas contains the radioisotope krypton-85 as an impurity. This impurity is removed chromatographically. Continuing research in this area is directed to further reduction of the krypton-85 level while at the same time increasing the xenon yield

The experimental chromatograph being used to investigate the factors that influence xenon purification can also be used to purify thermal diffusion column product xenon containing a krypton impurity and to purify thermal diffusion column product krypton having a xenon impurity. Work was done on the purification of krypton having a xenon impurity. The techniques of separating xenon from krypton have been developed, however, in previous work attention was directed toward recovering xenon, and krypton was treated as a waste material. Krypton has a non-negligible vapor pressure at liquid nitrogen temperature; hence, quantitative recovery of separated krypton requires the use of a charcoal trap. Pilot runs were planned to test the krypton recovery system before use on product material of high value.

One test run was completed. A mixture of commercially available xenon and krypton was used as a sample. The mixture was approximately 8% xenon and 92% krypton. An internally finned column containing Molecular Sieve 13X, 50/60 mesh, with a cross-sectional area of 4 cm² and 12-in. length was used. The column was operated at 166°K, and the sample volume was 120 ml. The krypton was trapped on 0.5 g of charcoal at 77°K. The excess nitrogen carrier gas trapped with the krypton was pumped off, and the gas was passed through a calcium trap into a storage cylinder. The calcium trap, maintained at a temperature of 700°C, removes the residual carrier gas. About 105 ml of gas were recovered in the krypton trap. Analytical results for this run have not yet been received.

THERMAL DIFFUSION RESEARCH

Molecular Beam Scattering A computer program has been written to calculate the theoretical quantum mechanical scattering intensity $I(v, \theta_v)$ as a function of the scattering angle θ in a molecular beam scattering experiment. This calculation is given by the equation

$$I(v, \theta_v) = \frac{\lambda^2}{16\pi^2} \left\{ \left[\sum_l (2l+1) (\sin 2\eta_l) P_l(\cos \theta_v) \right]^2 + \left[\sum_l (2l+1) (\cos 2\eta_l - 1) P_l(\cos \theta_v) \right]^2 \right\} \quad (1)$$

where λ is the de Broglie wavelength, l is the angular momentum quantum number, and $P_l(\cos \theta)$ is the Legendre function. The quantum mechanical phase shift η_l is obtained by interpolation from previously tabulated values.⁷

The observed scattering intensity $I(\theta_v)$ in a molecular beam experiment is influenced by any finite velocity spread such as that transmitted by a mechanical velocity selector.

Consider a gas in equilibrium at an average temperature T . The Maxwellian velocity distribution is given by

$$f(v)_g = \frac{4v^2}{\sqrt{\pi} a^3} e^{-v^2/a^2}$$

where $f(v)_g$ is the fraction of the total number of molecules in the gas that have velocities between v and $v+dv$. Since the probability of a molecule effusing through an orifice in the side of a gas "oven" is proportional to v , the velocity distribution for a beam of molecules is given by

$$f(v)_b = \frac{2}{a^4} v^3 e^{-v^2/a^2}$$

where $a = \sqrt{2kT/m}$ in both cases above.⁸

⁷R. B. Bernstein, *J. Chem. Phys.*, **33**, 795 (1960).

⁸N. F. Ramsey, *Molecular Beams*, London: Oxford Press, 1956.

The average value of the scattering intensity from two beams which include a finite velocity spread is given by

$$I(\theta_i)_{AV} = \frac{\int_{v_l}^{v_u} I(v, \theta_i) f(v)_b dv}{\int_{v_l}^{v_u} f(v)_b dv} \quad (2)$$

where v_u and v_l are, respectively, the upper and lower limits of the velocity spread

The denominator in Equation 2 may be calculated explicitly as follows:

$$\int_{v_l}^{v_u} f(v)_b dv = \frac{2}{\alpha^4} \int_{v_l}^{v_u} v^3 e^{-v^2/\alpha^2} dv$$

or, upon integrating:

$$\int_{v_l}^{v_u} f(v)_b dv = [e^{-v_l^2/\alpha^2} (v_l^2/\alpha^2 + 1)] - [e^{-v_u^2/\alpha^2} (v_u^2/\alpha^2 + 1)]$$

The numerator in Equation 2 may be numerically integrated using Simpson's rule as follows:

Let the integrand be designated by y , and

$$\begin{aligned} v_l &= v_1 \\ v_u &= v_n \end{aligned}$$

where: $n = 3, 5, 7, 9,$

Then:

$$\int_{v_l}^{v_u} I(v, \theta_i) f(v)_b dv = \frac{2\Delta v}{3\alpha^4} [y_1 + 4y_2 + 2y_3 + 4y_4 + 2y_5 + \dots + 4y_{n-1} + y_n]$$

The velocity-averaged scattering intensity $I(\theta_i)$ can now be written:

$$I(\theta_i)_{AV} = \frac{(2\Delta v/3\alpha^4) [y_1 + 4y_2 + 2y_3 + 4y_4 + 2y_5 + \dots + 4y_{n-1} + y_n]}{[e^{-v_l^2/\alpha^2} (v_l^2/\alpha^2 + 1)] - [e^{-v_n^2/\alpha^2} (v_n^2/\alpha^2 + 1)]} \quad (3)$$

The output data from Equation 1 were used as inputs for a second computer program to calculate $I(\theta_i)_{AV}$ from Equation 3 for a given velocity spread. A punched tape output permitted a direct plot of $I(\theta_i)_{AV}$ for a large number of closely spaced points.

Figure 8 shows the quantum scattering curve obtained from Equation 1 for scattering angles from 0 to 90° in argon-argon (distinguishable) scattering using a source temperature of 300°K.

Figure 8. Quantum Scattering Curve for Argon-Argon Scattering at a Source Temperature of 300°K.

The smoothing effect caused by a finite velocity spread is shown enlarged in Figure 9. The velocity-averaged scattering curves correspond to Figure 8 with velocity spreads from 0% to 10%. The most probable beam velocity was used as the median selected velocity [i.e., $\beta = \sqrt{3/2} \alpha$ such that the velocity spread is given by $\beta \pm (n-1)\Delta v/2$]. As the spread becomes larger the data approach a smooth classical curve, thus causing the observation of quantum undulations to become increasingly more difficult.

Figure 9. Scattering Curve with Various Velocity Spreads.

Our present velocity selector design gives a theoretical transmitted velocity spread of less than 2.5%.⁹ It can be seen from the figure that this spread should give a minimal deterioration of the undulatory properties of the curve. Because of this evidence, these quantum oscillations should be observable experimentally. The quantum properties of this curve have been observed to a minor degree as reported in the literature with a hydrogen-mercury scattering system, using a total velocity spread of 9.4%.¹⁰

⁹MLM-1287 p. 21

¹⁰H. U. Hostettler and R. B. Bernstein, *Phys. Rev. Letters*, **5**, 318 (1960)

Collision Integrals A theoretical and computational procedure for calculating the collision integrals which appear in the Chapman-Enskog theory of the transport properties has been outlined in previous reports.^{11,12} The collision integrals express the dependence of the transport properties on the intermolecular forces between the gas molecules.

Previously, these integrals have been calculated for only a few types of potential functions such as the Lennard-Jones potential and the exponential-six potential. In all these calculations, a potential function containing three or less parameters has been used. The procedure has been to calculate the collision integrals as a function of these parameters and then to determine the parameters for a specific system by fitting the calculated transport properties to the experimental values. It is difficult to correlate information about the intermolecular potential obtained by this procedure with information obtained from other sources.

A more informative procedure is to calculate collision integrals for a potential derived from other sources. By comparing the resulting transport properties with experimental values an indication of the validity of the potential can be obtained. In this procedure no restriction is placed on the functional form of the potential; it can be defined by different functions in different regions or it can be defined by a table of numerical values.

Helium was selected for initial investigation since its intermolecular potential has been studied extensively. One type of potential function, which was originally proposed by Margenau, is

$$\phi(r) = Ae^{-ar} - Be^{-br} - \frac{c_1}{r^6} - \frac{c_2}{r^8} - \frac{c_3}{r^{10}} \quad (4)$$

Some or all of the parameters for this potential function have been evaluated by a number of investigators. Collision integrals have been calculated for the six sets of parameters listed in Table 2 which were obtained as follows:

- (A) Slater-Kirkwood¹³ energies which were calculated by a variational procedure.
- (B) Detailed calculations of Margenau¹⁴ including exchange integrals.
- (C) Results of an examination by Margenau¹⁵ of the errors involved in adding together the valence, second-order exchange and the dispersion energies calculated by himself, Rosen¹⁶ and Page.¹⁷
- (D) Kestner's calculation¹⁸ based on a recent theory¹⁹ which allows cancellation of many electronic energy terms before the difference energy calculations are made

¹¹MLM-1274, p. 23.

¹²MLM-1287, p. 17.

¹³J. C. Slater and J. G. Kirkwood, *Phys. Rev.*, **37**, 682 (1931).

¹⁴H. Margenau, *Phys. Rev.*, **56**, 1000 (1939).

¹⁵H. Margenau, *Revs. Modern Phys.*, **11**, 1 (1939).

¹⁶N. Rosen, *Phys. Rev.*, **38**, 255 (1931).

¹⁷C. H. Page, *Phys. Rev.*, **53**, 426 (1938).

¹⁸Kestner, private communication to W. L. Taylor.

¹⁹O. Sinanoglu, *J. Chem. Phys.*, **36**, 706, 3198 (1963).

Table 2

INTERMOLECULAR POTENTIALS

Parameter	Slater-Kirkwood	Margenau	Rosen Margenau, and Page	Kestner	Yntema and Schneider	Yntema, Schneider and Fontana
A (ergs)	770×10^{-12}	770×10^{-12}	925×10^{-12}	1013×10^{-12}	1200×10^{-12}	1200×10^{-12}
a (A ⁻¹)	4 60	4 60	4 40	4 57	4 72	4 72
B (ergs)	- -	560×10^{-12}	560×10^{-12}	959×10^{-12}	- -	- -
b (A ⁻¹)	- -	5 33	5 33	5 10	- -	- -
c ₁ (ergs-A ⁶)	1.49×10^{-12}	1.39×10^{-12}	1.39×10^{-12}	1.30×10^{-12}	1.24×10^{-12}	1.26×10^{-12}
c ₂ (ergs-A ⁸)	- -	3.0×10^{-12}	3.00×10^{-12}	3.55×10^{-12}	1.89×10^{-12}	2.02×10^{-12}
c ₃ (ergs-A ¹⁰)	- -	- -	- -	3.94×10^{-12}	- -	3.96×10^{-12}
σ (A)	2 609	2 443	2 833	2 751	2 658	2 624
ϵ/k (°K)	9 248	13 814	6 375	7 898	8 258	9 176

(E) Results obtained by comparison of experimental data on the second virial coefficient with theoretical values calculated using the cited potential function. These parameters represented the best fit.²⁰

(F) Detailed dispersion energy coefficients calculated by Fontana²¹ were substituted for the trial and error values in (E).

The collision integrals for these potentials are tabulated in Table 3. In this table,

$$T^* = T/(\epsilon/k) \quad (5)$$

and

$$\Omega^{*(l,s)}(T^*) = \Omega^{(l,s)}(T^*)/\pi\sigma^2 \quad (6)$$

Below approximately 200°K, quantum effects start becoming important for helium, so calculations based on classical formulas must be limited to temperatures above this. An analysis of the equations for the collision integrals shows that for temperatures above 200°K the major contribution to the collision integrals comes from the repulsive portion of the potential having an energy greater than 10^{-15} erg.

Since the potential (A) demonstrated a spurious maximum at a very low temperature and (B) may be transformed to a Modified-Buckingham potential, potential curves (C), (D), (E), and (F) were investigated in more detail. The repulsive portions of these curves are plotted in Figure 10. The first approximations to the viscosity

²⁰J. L. Yntema and W. G. Schneider, *J. Chem. Phys.*, **18**, 646 (1950).

²¹P. Fontana *Phys. Rev.*, **123**, 1865 (1961).

Table 3

COLLISION INTEGRALS

	T^*	$\Omega^{*(1,1)}$	$\Omega^{*(2,2)}$	$\Omega^{*(3,3)}$	$\Omega^{*(4,4)}$	$\Omega^{*(1,2)}$	$\Omega^{*(1,3)}$
(A)	1.00	1.43000	1.58658	1.30110	1.37891	1.19351	1.06310
	5.00	0.81748	0.91203	0.80905	0.87060	0.75475	0.71681
	10.00	0.70604	0.80038	0.71169	0.77276	0.65920	0.62725
	20.00	0.61561	0.70803	0.62595	0.68484	0.57467	0.54538
	30.00	0.56708	0.65738	0.57828	0.63528	0.52809	0.49992
	40.00	0.53394	0.62234	0.54530	0.60075	0.49610	0.46866
	50.00	0.50889	0.59561	0.52019	0.57433	0.47186	0.44499
	60.00	0.48881	0.57405	0.49998	0.55300	0.45243	0.42602
	70.00	0.47211	0.55602	0.48311	0.53515	0.43626	0.41024
	80.00	0.45784	0.54055	0.46867	0.51984	0.42246	0.39678
	90.00	0.44541	0.52702	0.45606	0.50645	0.41043	0.38506
	100.00	0.43441	0.51502	0.44489	0.49456	0.39980	0.37470
	120.00	0.41565	0.49446	0.42581	0.47422	0.38167	0.35706
	140.00	0.40006	0.47731	0.40993	0.45725	0.36662	0.34242
	160.00	0.37518	0.44981	0.38453	0.43006	0.34264	0.31912
	200.00	0.36496	0.43846	0.37408	0.41884	0.33279	0.30956
	220.00	0.35581	0.42828	0.36472	0.40878	0.32398	0.30102
	240.00	0.34754	0.41907	0.35625	0.39968	0.31603	0.29332
	260.00	0.34001	0.41066	0.34854	0.39138	0.30880	0.28630
	300.00	0.32672	0.39578	0.33492	0.37670	0.29604	0.27394
(B)	0.10	3.90635	4.01973	3.68267	3.71979	3.47806	3.18705
	0.50	2.05663	2.29797	1.85501	2.00554	1.70381	1.46530
	1.00	1.43434	1.59830	1.30295	1.38512	1.19080	1.05591
	2.00	1.05352	1.16795	0.99424	1.06001	0.92109	0.85210
	3.00	0.91713	1.02298	0.88790	0.95303	0.82501	0.77393
	3.50	0.87690	0.98048	0.85549	0.92075	0.79485	0.74802
	4.00	0.84593	0.94782	0.82984	0.89526	0.77066	0.72671
	4.50	0.82068	0.92161	0.80871	0.87425	0.75050	0.70861
	5.00	0.79925	0.89992	0.79077	0.85639	0.73320	0.69288
	10.00	0.68102	0.78159	0.68661	0.75181	0.63091	0.59645
	15.00	0.62261	0.72233	0.63142	0.69554	0.57639	0.54350
	20.00	0.58352	0.68218	0.59342	0.65651	0.53901	0.50691
	25.00	0.55410	0.65169	0.56440	0.62657	0.51058	0.47897
	30.00	0.53049	0.62709	0.54093	0.60236	0.48766	0.45641
(C)	1.00	1.41834	1.57278	1.29560	1.37249	1.19096	1.06389
	5.00	0.82233	0.91525	0.81398	0.87459	0.76071	0.72339
	10.00	0.71276	0.80587	0.71835	0.77883	0.66665	0.63515
	20.00	0.62357	0.71531	0.63389	0.69260	0.58311	0.55409
	30.00	0.57554	0.66550	0.58675	0.64381	0.53687	0.50885
	40.00	0.54263	0.63096	0.55402	0.60971	0.50500	0.47762
	50.00	0.51768	0.60455	0.52904	0.58356	0.48078	0.45389
	60.00	0.49765	0.58320	0.50889	0.56241	0.46131	0.43482
	70.00	0.48096	0.56532	0.49203	0.54467	0.44508	0.41893
	80.00	0.46667	0.54996	0.47757	0.52943	0.43119	0.40533
	90.00	0.45419	0.53650	0.46492	0.51608	0.41907	0.39346
	100.00	0.44314	0.52455	0.45370	0.50421	0.40833	0.38296
	120.00	0.42425	0.50405	0.43448	0.48387	0.38997	0.36501
	140.00	0.40850	0.48690	0.41844	0.46686	0.37469	0.35006
	160.00	0.39503	0.47219	0.40469	0.45227	0.36161	0.33729
	180.00	0.38329	0.45933	0.39269	0.43951	0.35022	0.32615
	200.00	0.37288	0.44792	0.38204	0.42820	0.34012	0.31629
	220.00	0.36356	0.43768	0.37249	0.41805	0.33108	0.30745
	240.00	0.35511	0.42840	0.36385	0.40889	0.32289	0.29946
	260.00	0.34740	0.41991	0.35595	0.40052	0.31541	0.29216

Table 3 (Continued)

	T^*	$\Omega^{*(1,1)}$	$\Omega^{*(2,2)}$	$\Omega^{*(3,3)}$	$\Omega^{*(4,4)}$	$\Omega^{*(1,2)}$	$\Omega^{*(1,3)}$
(D)	1.00	1.40829	1.55945	1.29101	1.36584	1.18967	1.06681
	5.00	0.83228	0.92242	0.82422	0.88303	0.77254	0.73640
	10.00	0.72617	0.81659	0.73181	0.79060	0.68157	0.65115
	20.00	0.64001	0.72928	0.65043	0.70764	0.60097	0.57299
	30.00	0.59368	0.68137	0.60506	0.66077	0.55641	0.52941
	40.00	0.56197	0.64818	0.57360	0.62805	0.52571	0.49934
	50.00	0.53794	0.62281	0.54958	0.60296	0.50239	0.47650
	60.00	0.51864	0.60232	0.53021	0.58267	0.48365	0.45814
	70.00	0.50257	0.58515	0.51402	0.56567	0.46802	0.44285
	80.00	0.48881	0.57041	0.50012	0.55105	0.45465	0.42976
	90.00	0.47680	0.55750	0.48798	0.53826	0.44298	0.41834
	100.00	0.46616	0.54604	0.47720	0.52689	0.43265	0.40823
	120.00	0.44797	0.52638	0.45874	0.50740	0.41498	0.39096
	140.00	0.43282	0.50994	0.44334	0.49110	0.40028	0.37659
	160.00	0.41986	0.49584	0.43015	0.47713	0.38770	0.36430
	180.00	0.40856	0.48351	0.41863	0.46492	0.37674	0.35360
	200.00	0.39855	0.47258	0.40842	0.45408	0.36703	0.34412
	220.00	0.38958	0.46276	0.39925	0.44436	0.35834	0.33563
	240.00	0.38145	0.45386	0.39096	0.43555	0.35047	0.32794
	260.00	0.37404	0.44573	0.38337	0.42750	0.34328	0.32093
(E)	1.00	1.41668	1.56929	1.29474	1.37060	1.19113	1.06538
	2.00	1.06296	1.16789	1.00671	1.06668	0.93945	0.87516
	3.00	0.93589	1.03269	0.90784	0.96720	0.85021	0.80296
	4.00	0.86954	0.96284	0.85427	0.91394	0.80008	0.75971
	5.00	0.82665	0.91854	0.81847	0.87839	0.76576	0.72892
	10.00	0.71846	0.81048	0.72412	0.78387	0.67297	0.64194
	20.00	0.63058	0.72119	0.64098	0.69893	0.59076	0.56224
	30.00	0.58335	0.67218	0.59467	0.65098	0.54536	0.51787
	35.00	0.56592	0.65391	0.57740	0.63299	0.52852	0.50139
	40.00	0.55103	0.63823	0.56257	0.61751	0.51411	0.48728
	45.00	0.53806	0.62450	0.54961	0.60394	0.50153	0.47497
	50.00	0.52657	0.61230	0.53810	0.59187	0.49039	0.46407
	60.00	0.50694	0.59137	0.51837	0.57114	0.47135	0.44544
	70.00	0.49059	0.57384	0.50188	0.55378	0.45548	0.42993
	80.00	0.47661	0.55879	0.48775	0.53887	0.44192	0.41667
	90.00	0.46442	0.54562	0.47541	0.52582	0.43009	0.40512
	100.00	0.45362	0.53392	0.46446	0.51423	0.41961	0.39489
	120.00	0.43517	0.51388	0.44573	0.49436	0.40174	0.37744
	140.00	0.41982	0.49713	0.43011	0.47777	0.38687	0.36294
	180.00	0.39528	0.47023	0.40509	0.45113	0.36312	0.33980
	200.00	0.38517	0.45911	0.39476	0.44012	0.35334	0.33028
(F)	1.00	1.41802	1.57216	1.29563	1.37230	1.19128	1.06449
	2.00	1.06192	1.16763	1.00515	1.06551	0.93736	0.87248
	3.00	0.93376	1.03122	0.90535	0.96508	0.84733	0.79967
	4.00	0.86678	0.96073	0.85131	0.91135	0.79676	0.75606
	5.00	0.82357	0.91604	0.81519	0.87550	0.76216	0.72501
	10.00	0.71446	0.80710	0.72003	0.78020	0.66857	0.63725
	15.00	0.66118	0.75328	0.67018	0.72951	0.61919	0.58951
	20.00	0.62576	0.71704	0.63608	0.69451	0.58554	0.55670
	23.00	0.60906	0.69981	0.61979	0.67770	0.56952	0.54104
	30.00	0.57801	0.66756	0.58923	0.64606	0.53959	0.51174
	40.00	0.54531	0.63326	0.55673	0.61221	0.50792	0.48071
	50.00	0.52052	0.60704	0.53190	0.58627	0.48384	0.45711
	60.00	0.50061	0.58586	0.51188	0.56527	0.46449	0.43815
	70.00	0.48401	0.56811	0.49512	0.54768	0.44835	0.42232
	80.00	0.46980	0.55287	0.48074	0.53255	0.43452	0.40878
	90.00	0.45740	0.53952	0.46816	0.51931	0.42245	0.39695

Table 3 (Continued)

<u>T*</u>	<u>$\Omega^{*(1,1)}$</u>	<u>$\Omega^{*(2,2)}$</u>	<u>$\Omega^{*(3,3)}$</u>	<u>$\Omega^{*(4,4)}$</u>	<u>$\Omega^{*(1,2)}$</u>	<u>$\Omega^{*(1,3)}$</u>
(F) 100.00	0.44640	0.52766	0.45699	0.50754	0.41175	0.38647
120.00	0.42759	0.50732	0.43785	0.48736	0.39345	0.36854
140.00	0.41190	0.49031	0.42185	0.47049	0.37819	0.35358
160.00	0.39847	0.47572	0.40815	0.45608	0.36512	0.34078
180.00	0.38674	0.46298	0.39620	0.44361	0.35371	0.32963
200.00	0.37634	0.45170	0.38565	0.43272	0.34361	0.31979
240.00	0.35860	0.43256	0.36802	0.41505	0.32647	0.30334
260.00	0.35092	0.42438	0.36065	0.40796	0.31913	0.29645

$$\eta \times 10^7 = 266.93 \frac{\sqrt{MT}}{\sigma^2 \Omega^{*(2,2)}(T^*)} \quad (7)$$

and thermal conductivity

$$\lambda \times 10^7 = 1989.1 \sqrt{T/M} / \sigma^2 \Omega^{*(2,2)}(T^*) \quad (8)$$

were calculated and are compared with experimental data in Figures 11 and 12. None of the potentials represents the experimental data very well over the entire temperature range. In Figure 12, it may be seen that the high temperature thermal conductivity reported by Collins and that of Blais and Mann differ considerably. Weissman²² has shown that the data of Blais and Mann are inconsistent with experimental high temperature viscosity data. If this is so, then the slope of the theoretical transport properties appears to approach the experimental value. Although a "true" potential cannot be selected on the basis of the foregoing comparisons, the sensitivity of these calculations to relatively small changes in the potential should provide a rapid means of testing.

Transport Coefficients The recent discovery of a diffusion barrier in the tops of some of the other experimental columns²³ has prompted a reassessment of experimental flow data for Ne²⁰-Ne²² separation taken previously for column E5.²⁴ It appeared quite likely that a similar transport barrier was present in this column, therefore a complete new series of static and flow experiments is being run. To eliminate end effects the flow experiments are being run with the apparatus arranged as shown in Figure 13 to give an effective column length of 152 cm.

A complete set of data was obtained for a wire temperature of 815°C. The static separation data were reported last month.²⁵ Results of flow experiments at two pressures, 1250 and 800 torr, are depicted in Figure 14. Transport coefficients derived from the data are given in Table 4. Also given in Table 4 are transport coefficients calculated from the old data taken with the full 183-cm column length. The theoretical values in Table 4 were calculated from the Jones and Furry theory with Lennard-Jones shape factors. The thermal diffusion factor, α , at the cold wall temperature was assumed to be 0.0227 in these calculations.

The results show that there were essentially no end effects in the old experimental data; however, the precision of the new data is substantially better than that of the old data.

²²S. Weissman, private communication to W. L. Taylor.

²³MLM-1299 p. 29.

²⁴MLM-1238 p. 31.

²⁵MLM-1311, p. 16.

Figure 10. Repulsive Energy of Interaction.

Figure 11. Experimental and Calculated Viscosities of Helium.

Table 4

THERMAL DIFFUSION COLUMN TRANSPORT COEFFICIENTS FOR NEON IN COLUMN E5

	H' (g/sec-atm ²)	K'_C (g-cm/sec-atm ⁴)	K_d (g-cm/sec)
Theory	4.44	1.46	1.68
Experiment (from 1250-torr data)	4.30	1.35	1.72
Experiment (from 800-torr data)	4.41	1.38	1.77
Experiment (old data)	4.38	1.23	1.73

²⁶J. Kestin and W. Leidenfrost, *Physica*, **25**, 537-55 (1959).

²⁷F. A. Guevara and W. E. Wageman, *Measurement of He and H₂ Viscosities to 2340°K*, LA-3319, 1965.

²⁸J. Hilsenrath and Y. S. Touloukian, *Trans. ASME*, **76**, 967 (1954).

²⁹R. A. Makavetskas, V. N. Popov and N. V. Tsederberg, *High Temperature*, **1**, 169 (1963).

³⁰A. V. Brancker, *Ind. Chemist*, **30**, 307-12 (1954).

Figure 12. Experimental and Calculated Thermal Conductivities of Helium.

Figure 13. Experimental Arrangement for Thermal Diffusion Flow Experiments.

³¹H. Chuang, UCRL-8230, 1958, p. 146.

³²H. von Ubisch, *Arkiv Fysik*, 16, No. 7, 93-100 (1959).

³³W. G. Kannuluik and E. H. Carman, *Proc. Phys. Soc.*, 65B, 701-9 (1952).

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³⁷D. J. Collins, R. Greif and A. E. Bryson, *Intern. J. Heat Mass Transfer*, 8, 1209 (1965).

³⁸J. R. Peterson and C. F. Bonilla, "Advances in Thermophysical Properties at Extreme Temperatures and Pressures," 3rd Symposium on Thermophysical Properties, New York, ASME, 1965.

Figure 14. Separation Factors for $\text{Ne}^{20}\text{-Ne}^{22}$ in Column E5.

Transport Properties A Trennschaukel or "swing separator" is being used to measure the thermal diffusion factor of gaseous mixtures as a function of composition and temperature.

It has been pointed out³⁹ that the large scatter of data for helium-argon and helium-xenon systems was probably due to a lack of sufficiently accurate mass spectrographic analysis. Discussions revealed that neon-xenon mixtures would have optimum masses for the present equipment. A series of runs using various compositions is presently being done.

Only rather rough results have been obtained at present because considerable air has been found in several of the samples. This air was traced to leaks in three valves in the gas handling manifold whose bellows had apparently developed fatigue cracks.

In Figure 15 the reciprocal of α_T , the thermal diffusion factor, is plotted as a function of mole fraction of neon; all the results refer to an average temperature of about 327°K. This reciprocal plot should be a straight line.^{40,41} The points are: filled circles, Atkins et al.;⁴² open circle, Grew;⁴³ open square, Heymann and Kistemaker;⁴⁴ filled triangles, the present results. It is evident that the current rough data are almost within the scatter of the literature data. It is expected that, once a series of air-free runs is completed, a choice between the various literature values could be made since the Mound Laboratory data should be more precise.

³⁹MLM-1300 p. 23

⁴⁰M. F. Laranjeira, *Physica*, **26**, 417 (1960).

⁴¹E. A. Mason, S. Weissman and R. P. Wendt, *Phys. Fluids*, **7**, 174 (1964).

⁴²B. E. Atkins, R. A. Bastick and T. L. Ibbs, *Proc. Roy. Soc., (London)*, **A172**, 142 (1939).

⁴³K. E. Grew, *Proc. Roy. Soc. (London)*, **A189**, 402 (1947).

⁴⁴D. Heymann and J. Kistemaker, *Physica*, **25**, 556 (1959).

Figure 15. Thermal Diffusion Factors for Xenon-Neon Separation.

Cascade Theory A computer program to calculate the performance of multicomponent isotope separation cascades was written and checked out. The program calculates the composition of two or more product streams for specified flow rates. Systems with as many as ten components can be calculated, although trial calculations have been made for two, three and five-component systems only.

A basic part of the program is a subroutine to solve the system of $n-1$ differential equations which describe isotope transport in an n -component system:

$$P(C_{P_i} - C_i) = H_o C_i (d_i - \sum_{l=1}^{n-1} d_l C_l) - K \frac{dC_i}{dZ} \quad (9)$$

where P is the product flow rate, C_i is the fraction of component i , C_{P_i} is the fraction of component i in the product, H_o is the initial transport coefficient for one unit difference in mass, d_i is the difference in mass between component i and the key component, K is the remixing coefficient, and Z is the length

along the cascade. An $(n-1)$ -dimensional Runge-Kutta scheme was used in the subroutine for the numerical solution of Equation 9. Given the ratio P/H_o , the quantities d_i , and the composition at one point in the cascade, C_{i_o} , the subroutine calculates the composition at a second point in the cascade at a dimensionless distance $\Delta y = H_o \Delta Z / K$ from the first point.

The main program was written to handle cascades of the general form given in Figure 16 [i.e., a cascade of j stages ($j \leq 20$) with two end products and as many as $j-1$ intermediate products]. The inclusion of intermediate product streams in the computer program required that the iteration procedure for solving the cascade be quite different from schemes used in the binary cascade programs. In the binary cascade programs computation starts from one end of the cascade and proceeds to the feed point. Then the material balance is applied and a computation is made from the other end to the feed point. Iteration proceeds until a match in composition is obtained at the feed point. In the multicomponent cascade with sidestreams, however, this approach is not possible unless multiple iterations are used. The calculation scheme adopted for the new multicomponent program is based, therefore, on a computation which starts at one end of the cascade and which proceeds all the way to the other end of the cascade.

The iterative procedure for solution of the cascade is an $(n-1)$ -dimensional, finite difference version of Newton's method. The iterated variables are logarithms of the isotope ratios at one end of the cascade (i.e., $\ln R_P$ as in Equation 10):

$$\ln R_P = \ln C_{P_i} / C_{P_k} \quad (10)$$

Figure 16. General Form of Multicomponent Isotope Separation Cascade

where C_{p_k} is the fraction of the key component in the end product. Typically, convergence has been obtained in five to ten iterations, although the program has failed for certain unusual combinations of flow rates and cascade configurations.

Results of a test run to compare results of the new program with those obtained with one of the existing binary cascade programs are given in Figure 17. Good agreement was obtained, running time was longer, however, because in the binary program the closed form solution of Equation 9 can be used instead of the Runge-Kutta integration

A second test was run for neon in the 96-ft thermal diffusion cascade. Results for this three-component system are depicted in Figure 18 along with experimental results obtained recently. In connection with these results it should be emphasized that calculated compositions are extremely sensitive to small flow rate changes; hence, valid comparisons between theory and experiment are difficult to make.

Figure 17. Comparison of Binary Cascade Solutions Obtained with Binary and with Multicomponent Programs

Calculated				Experimental	
P = 0.2 g/day		P = 0.22 g/day		P = 0.24 g/day	
% Ne ²²	% Ne ²¹	% Ne ²²	% Ne ²¹	% Ne ²²	% Ne ²¹
0.0009	0.075	0.0006	0.024	0.00	0.03
61.06	13.34	14.19	11.65	49.19	12.24
99.75	0.241	98.93	1.01	99.68	0.29

Figure 18. Performance of the Neon Cascade.

More extensive calculations were made for the 11-column, 5-stage argon cascade shown in Figure 19. Partial results of this work are shown in Figure 20. Additional calculations are now being made in the flow rate range below 0.09 g/day. When these are completed, an argon-38 sidestream will be considered.

Column Properties

$H = 1.3211 \times 10^{-5} \text{ g/sec (1 Mass Unit)}$
 $HL/K = 6.164 \quad (1 \text{ Mass Unit})$

Figure 19. Argon Isotope Separation Cascade.

Figure 20. Performance of the Argon Cascade.