Separation of Gas Mixtures in Free Jets

JAMES B. ANDERSON
Princeton University, Princeton, New Jersey

Spatial distributions of species flux in axisymmetric free jets of helium-argon mixtures were determined experimentally. The experiments encompass the transition range between inviscid and free molecular flow at the exits of the nozzles forming the jets. Comparison is made with existing theories of separation. The results confirm the validity of Sherman's predictions and Rothe's measurements of diffusive separation for nearly inviscid flow. In the transition range departures from the nearly inviscid theory occur. The maximum degree of separation is observed in this range.

Diffusive separation of a gas mixture under the influence of a pressure gradient as in a gas centrifuge also occurs in flow systems. The first attempts to devise a practical separation process based on diffusion of species under the pressure gradients of a rapidly expanding gas jet appear to be those of Tahourdin and Dirac (*1*) at Oxford University reported in 1946. They observed an increased concentration of the heavy species of a binary mixture at the center of a jet formed by passing the mixture through a slit into a low-pressure chamber. Similar effects were observed independently by Becker et al. (*2*) at the University of Marburg, Germany, in 1954. Subsequently, Becker and his associates, now at Karlsruhe, Germany, have been developing a process for the separation of mixtures of uranium isotopes using jets containing gaseous uranium hexafluorides. This process may be economically competitive with the gaseous diffusion process used at Oak Ridge and elsewhere (*3*, *4*). While pressure diffusion may be neglected in the engineering of many systems, it is often important. It has major effects in the performance of low-pressure gas sampling systems and the operation of nozzle-source molecular beams (*5*).

BACKGROUND

In their early experiments Becker and co-workers passed a gas mixture through a nozzle to form a jet in supersonic flow in a low-pressure chamber. Samples of gas withdrawn by a sampling tube placed on the axis of the jet were found to be enriched in the heavier species of the mixture, while the gas flowing at the periphery of the jet was depleted in the heavier species. Two mechanisms were proposed for the separation effect: (I) pressure diffusion driven by the high pressure gradients in the expanding jet and (II) migration of the light species with respect to the heavy species due to the higher thermal velocities of the light species. Waterman and Stern (*6*) contributed further experimental measurements of the separation effect and proposed a quantitative theory for mechanism II appropriate to the free molecular portion of the jet.

Further experiments were reported by Chow (*7*), who traversed the entire flow field of a jet with a sampling tube. He observed an apparent increase in concentration of the heavy species throughout the flow field.

Reis and Fenn (*8*) explained this apparent violation of species conservation by demonstrating experimentally that separation of species could occur at the entrance of a sampling probe. They proposed a third mechanism to explain the separation observed; (III) deceleration of the gas by a bow shock on the sampling tube with the heavy gas entering the tube at a greater rate than the light because of its higher persistence of velocity. These results raised some questions on the previous interpretations of the separation phenomenon.

Zigan (*9*) of the Karlsruhe group published an analysis of the separation to be expected by the pressure diffusion mechanism for several configurations which had been investigated experimentally. The basis of the calculations was the binary diffusion equation of the Chapman-Enskog theory. Zigan calculated the properties of the supersonic flow fields involved by the method-of-characteristics procedure applicable to inviscid flow and reasoned that for small degrees of separation the departures from inviscid flow caused by separation could be neglected. The results were in qualitative agreement with many of the experimental observations made at Karlsruhe.

Analyses similar to Zigan's were later developed by Mikanic and Takashima (*10*) and by Sherman (*11*). For the conditions of the experiments by Reis and Fenn, Sherman showed that the extent of separation on the jet axis due to thermal and pressure diffusion in the jet itself could be

expected to be small in comparison to that actually measured.

Further experiments at Karlsruhe (*12*) lent support to the pressure diffusion mechanism for separation under conditions of lower nozzle Reynolds number than used by Reis and Fenn. Kogan (*13*) attempted to measure separation in a Prandtl-Meyer fan but separation at the probe obscured separation in the fan itself. Rothe (*14*) measured separation in a helium-argon jet from an orifice by an electron beam fluorescence method. His results gave quantitative verification of the theoretical predictions of Sherman for diffusive separation on the axis of the jet with Reynolds number N_{Re} greater than 100.

From previous theoretical and experimental work it has become clear that separation can occur by pressure (and thermal) diffusion in the jet (I) and by a probe effect (III). Separation by radial migration of one species with respect to the other due to differing thermal velocities (II) is seemingly possible but this mechanism has not been isolated in any experiments on the free jet itself.*

The object of the work reported here was the experimental determination of separation by mechanisms I and II over the entire transition range in axisymmetric free jets. Separation by mechanism III was eliminated in the present experiments. The limits of applicability of the theories for separation in the jet could thus be examined.

FREE JET PROPERTIES

The properties of the flow field of an inviscid, isentropic free jet expanding into a zero pressure region may be calculated by the method of characteristics applicable to supersonic flow if the transonic flow properties are known. Owen and Thornhill (*15*) and Wolff (*16*) made such calculations for diatomic and monatomic gases using an assumed sonic surface at the nozzle exit. Experiments (*11*, *15*) have shown that the flow field several diameters downstream is unaffected by minor variations in nozzle configuration and that these method-of-characteristics solutions are applicable near the axis at distances greater than two diameters from the nozzle exit. In the transonic region at the nozzle exit experimental measurements of flow properties are required.

The properties of free jets of pure monatomic gases have been studied extensively by a variety of techniques (*17* to *19*). The Wolff calculations show that beyond two- to three-nozzle diameters from the nozzle exit, the flow approaches a radial flow from a point source. The density continues to decrease with increasing distance. Eventually a transition from continuum to nearly free molecular flow occurs. The axial velocity distribution approaches a constant, nearly Maxwellian distribution, while the transverse velocity distribution becomes increasingly non-Maxwellian and continues to become narrower with increasing distance.

THEORY OF DIFFUSIVE SEPARATION (MECHANISM I)

Since it is the most complete of the available treatments, Sherman's analysis (*11*) of diffusive separation for nearly inviscid flow in a free jet is followed here. The quantities involved in the equations of change (including the Chapman-Enskog binary diffusion equation) are expanded in ascending powers of inverse Reynolds number.† Retaining only zero- and first-order terms, Sherman obtained a solution for a concentration perturbation as a function of inviscid flow properties and position in the jet. On

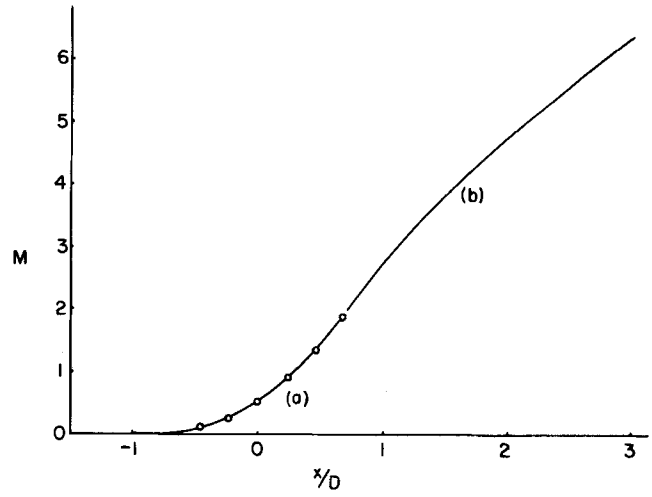

Fig. 1. Mach number variation on the axis of a free jet from an orifice ($\gamma = 5/3$). (*a*) Static probe measurements. (*b*) Method-of-characteristics calculation.

the axis of the jet, the molecular flux fraction ϕ differs from the stagnation mole fraction f_0 by

$$\phi - f_0 = \frac{1}{N_{Re}}(\gamma - 1)\,EC\left\{(M^2-1)\left(1+\frac{\gamma-1}{2}M^2\right)^{\frac{5-3\gamma}{2(\gamma-1)}} \times \frac{dM}{d(x/D)} - \frac{1}{2}\int_{-\infty}^{x}\frac{(M^2-1)(3+M^2)}{M}\left(1+\frac{\gamma-1}{2}M^2\right)^{\frac{7-5\gamma}{2(\gamma-1)}} \times \left(\frac{dM}{d(x/D)}\right)^2 d(x/D)\right\} \quad (1)$$

where

$$E = f_0\frac{(1-f_0)}{N_{Sc}}\left[\frac{m_1 - m_2}{m_0}\left(\frac{\gamma}{\gamma-1}\right) - \alpha_0\right] \quad (2)$$

and C is introduced as a constant for simplified treatment of viscosity with temperature

$$\frac{\mu}{\mu_0} = C\frac{T}{T_0} \quad (3)$$

Evaluation of Equation (1) requires a knowledge of the variation of M with x/D obtained from the properties of the inviscid flow field. For the sharp-edged orifice geometry we obtained data for argon with a static pressure probe on the axis in the transonic region one diameter upstream to one diameter downstream of the exit plane. Dimensions used were: orifice diameter, 4.3 mm.; probe diameter, 0.45 mm.; probe length, 25 mm.; static pressure hole diameters, 0.17 mm.; hole location, 15 mm. downstream of probe nose. Pressures were measured with an NRC Alphatron gauge. Data taken in the Reynolds number range of 4,400 to 5,600 are shown in Figure 1. Near an x/D of about unity the curve merges with the curve calculated by Wolff for $\gamma = 5/3$.

Equation (1) was integrated numerically from -1 to $+10$ diameters to give the curve shown in Figure 2. Beyond $+10$ diameters the analytical expression, $M = 3.20\,(x/D)^{2/3} - 0.625\,(x/D)^{-2/3}$, was used for the integration. Most of the predicted separation occurs in the region $0 < x/D < 2$ with only 20% of the total separation occurring for $x/D > 2$. The integration for the mole fraction change $f - f_0$ is also shown in Figure 2.

The major limitations in the above analysis are the requirements that the properties of the flow be closely approximated by the inviscid solution and that ordinary dif-

*Mechanism II is important (and has been observed) aft of the collimating orifice or skimmer in molecular beam systems.

†Reynolds number is based on nozzle exit diameter rather than radius as in Sherman's treatment.

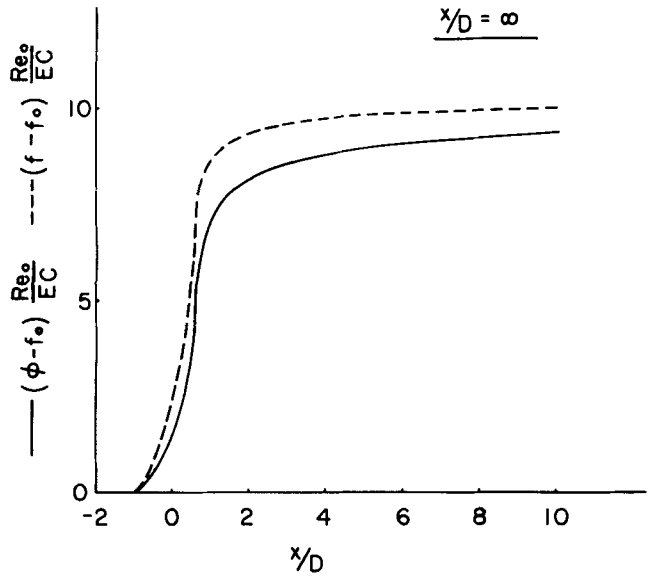

Fig. 2. Separation predicted by nearly inviscid theory on the axis of a free jet from an orifice ($\gamma = 5/3$).

fusion can be neglected. This implies a small degree of separation and negligible viscous effects.

THEORY OF SEPARATION IN FREE MOLECULAR FLOW (MECHANISM II)

The quantitative theories of Waterman and Stern (6) and others (*20, 21*) for separation by a free molecular mechanism were developed from a model for the jet in which the gas issues from the source with an *axial* velocity on which is superimposed a Maxwellian velocity distribution for each species. The two species are assumed to have the same static temperature. The lighter species then has higher thermal velocities than the heavier. The molecules crossing a "cut-off plane" are assumed to have no more collisions and to follow trajectories corresponding to their velocities at the cut-off plane. Because its velocity distribution is wider, the lighter species is spatially distributed farther from the axis than the heavier species and separation is predicted. For a zero mass velocity at the cut-off plane, the theory is identical with other treatments of free molecular or effusive flow through an orifice.

The theory can be improved for free jets by eliminating several of the assumptions previously used. By taking into account the divergence of the jet flow downstream of the exit plane and allowing for a curved cut-off surface, application may be extended into the nearly inviscid regime in which the assumption of equal species mass velocities is more reasonable. We approximate each species flux as proportional to $\cos^4\theta$ (as suggested by the method-of-characteristics predictions) at a hemispherical cut-off surface of radius z with its origin at the center of the nozzle exit plane. In terms of nozzle flow, the flux at the cut-off surface is then

$$j_s = \frac{5}{2} F_N \frac{1}{\pi z^2} \cos^4\theta \tag{4}$$

The mass velocities are taken as normal to the surface (angle θ to the axis). For each species the contribution of the flux from each element dA on the surface to the flux on the axis for $x >> z$ and $M > 3$ is given by

$$dj \cong j_s dA_s \frac{1}{\pi x^2} \frac{\gamma M^2}{2} \frac{1}{\cos^3\theta} e^{-\frac{\gamma M^2}{2}\tan^2\theta} \tag{5}$$

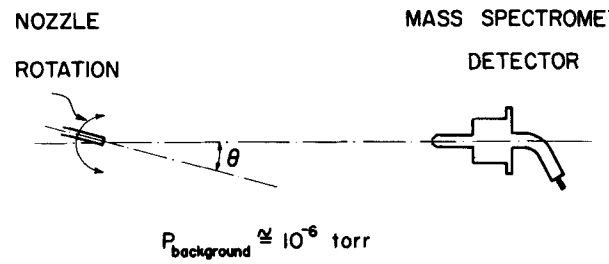

Fig. 3. Schematic diagram of apparatus for measurements of spatial distributions of species flux.

Integration over the entire surface gives the species flux on the axis at large x:

$$j \cong \frac{5}{2} F_N \frac{1}{\pi x^2} \frac{\gamma M^2}{2} e^{\frac{\gamma M^2}{2}} E_2\left(\frac{\gamma M^2}{2}\right) \tag{6}$$

where $E_2(y)$ is the exponential integral defined by

$$E_2(y) = \int_1^\infty e^{-yu} u^{-2} du \tag{7}$$

Equation (6) can be used to estimate the individual species fluxes and thus the molecular flux fraction on the axis at large x when the nozzle flow and cut-off M for each species are known.

EXPERIMENTAL EQUIPMENT AND PROCEDURE

The essential elements of the apparatus are shown schematically in Figure 3. Helium-argon mixtures were fed from storage cylinders to a small nozzle located in a high vacuum chamber. Nozzle temperature was 295° to 300°K. A mass spectrometer detector was placed 24.5 cm. from the nozzle exit. The nozzle was attached to a mechanism which allowed the angle between the nozzle axis and a line from the nozzle exit to the detector to be varied ±90 deg. by external control. The detector could thus measure species flux along any ray within 90 deg. of the nozzle axis.

The vacuum chamber was pumped by two 32-in. oil diffusion pumps backed by two 400 cu. ft./min. mechanical pumps. Capacity of the system was about 30,000 liters/sec. at 10^{-6} torr. During experiments background pressures in the chamber were maintained at 5×10^{-6} torr or less to eliminate interference with the jet by background molecules.

Two nozzles were used: a glass nozzle formed from 0.2-cm. ID. capillary tubing tapering slowly (radius of curvature at exit, 0.03 cm.) to a 0.0022-cm. exit diameter with no diverging section, and a 0.0203-cm. diameter, sharp-edged orifice cut in metal foil 0.002 cm. thick. Nozzle pressures were measured with an absolute mercury manometer (2 to 1,000 torr), a McLeod gauge (0.01 to 2 torr), or a Bourdon tube pressure gauge (1,000 to 5,000 torr). For nozzle flow near the free molecular region gas was fed to the stagnation chamber and out through an additional tube at a rate much larger than the nozzle flow to prevent a depletion of helium in the stagnation chamber due to its higher effusion rate through the nozzle.

The ion source of the detector was enclosed in a cylindrical chamber (7 cm. × 7 cm.) with a 0.2-cm. orifice at the tip of a cone on an extension 8 cm. from one end. The mass spectrometer was of the magnetic-deflection type with a 60-deg. magnetic sector (Aerovac AVA-1). Jet molecules incident on the detector passed directly into the chamber where their velocities and directions of motion were randomized. Since the molecules left the detector in free molecular flow, the net random flow out for a species was proportional to the pressure of that species inside the detector less the background pressure of that species outside the detector. At steady state the species flux incident on the detector from the jet was equal to the net random flow out and was proportional to the difference between chamber pressure and background pressure for that species. Background pressures were determined by placing a flag 2 cm. in front of the detector entrance in order to block the jet flow. The detector was calibrated as a flux detector with an effusive flow source which could be moved into position in front of the detector periodically during each experiment.

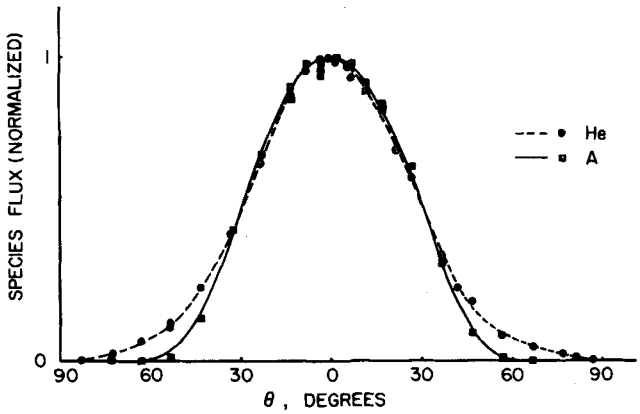

Fig. 4. Spatial distributions of helium and argon fluxes for the converging nozzle at $N_{Re} = 1{,}227$.

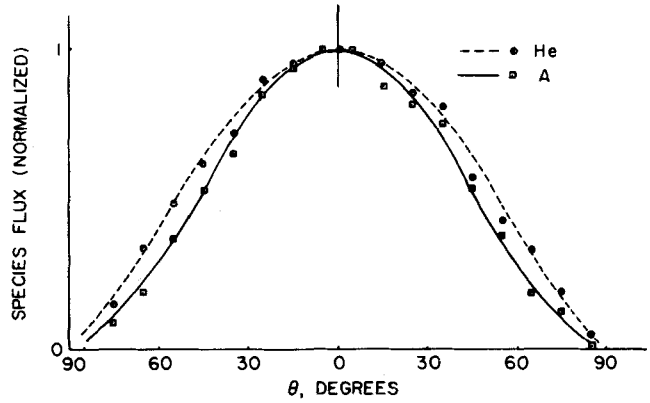

Fig. 6. Spatial distributions of helium and argon fluxes for the orifice at $N_{Re} = 1.8$.

The detector was operated in the free molecular regime for all experiments. At the highest incident flux used the Knudsen number (Lagrangian mean free path/entrance diameter) was greater than 1,000. The pressure in the detector was about 5×10^{-5} torr for this flux. In this range of operation species separation at the probe by mechanism III was negligible.

The argon-helium mixtures and the pure argon and pure helium used for calibrations were high purity grade supplied by the Matheson Company. For the results reported here a single cylinder of an argon-helium mixture was used. As analyzed by the Matheson Company, the composition was 5.27 mole % argon. Integration of the measured species fluxes over the flow field for several nozzle flows gave compositions within 0.2 mole % of the 5.27 mole % argon composition. Accuracy of individual molecular flux composition measurements is estimated to be not better than 10% for argon, for example, 7.0 ± 0.7 mole % argon.

EXPERIMENTAL RESULTS

Results of measurements of spatial distributions of the argon and helium species at several Reynolds numbers are plotted in Figures 4 to 6. For the high Reynolds number of Figure 4 ($N_{Re} = 1{,}227$) the spatial distributions of species flux, normalized to unity on the axis, are nearly the same. At the edges of the jet however a depletion of argon is seen. Measured flux composition on the axis is in this case 6.0 mole % argon.

At the lower Reynolds number for Figure 5 ($N_{Re} = 125$) the argon is distributed much nearer the axis than helium and the flux composition on the axis is 13.3 mole % argon.

At still lower Reynolds numbers the flow approaches the free molecular limit. Figure 6 shows the species distribution for orifice flow at $N_{Re} = 1.8$. Under this condition the distributions approach the cos θ distribution expected for effusive flow through an orifice, but the argon distribution is peaked on the axis slightly more than the helium distribution. Flux composition on the axis is 2.1 mole % argon. At the free molecular limit the flux composition would be uniform over the flow field and depleted in argon to 1.73 mole % argon due to the lower effusion velocity of the argon.

In Figure 7 the measured enrichment of argon $\phi - f_0$ on the axis is plotted against the inverse nozzle Reynolds number for both nozzles used. Also plotted is the prediction of $\phi - f_0$ for the nearly inviscid theory of diffusive separation. The following values were used in calculation of the theoretical line shown: $EC = 0.64$; $(\phi - f_0) \times N_{Re}/EC = 11.7$.

Figure 8 shows measured molecular flux composition on the axis in the entire range of Reynolds numbers investigated. No data were taken for the converging nozzle with $N_{Re} < 25$. Although the separation observed is similar for the two nozzles in the range $25 < N_{Re} < 1{,}200$, the behavior may be dissimilar for $N_{Re} < 25$.

DISCUSSION AND CONCLUSIONS

The experimental results for the orifice are in good agreement (within experimental error) with the predictions

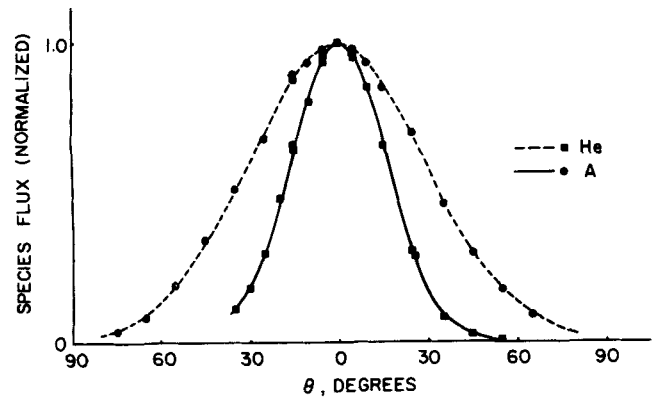

Fig. 5. Spatial distributions of helium and argon fluxes for the converging nozzle at $N_{Re} = 125$.

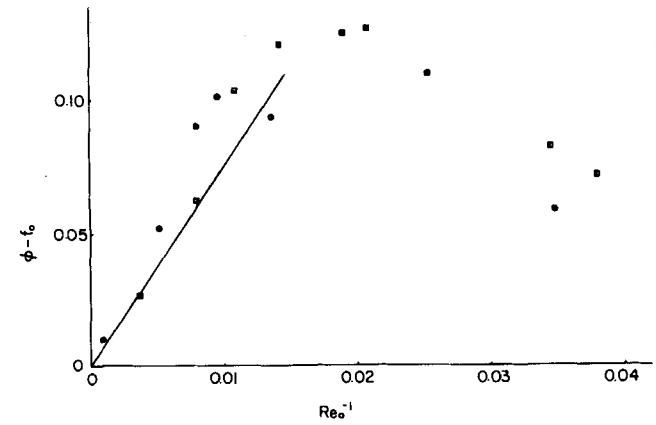

Fig. 7. Comparison of measured argon enrichment on the axis with the nearly inviscid theory of diffusive separation (for the orifice). Circles, converging nozzle; squares, orifice.

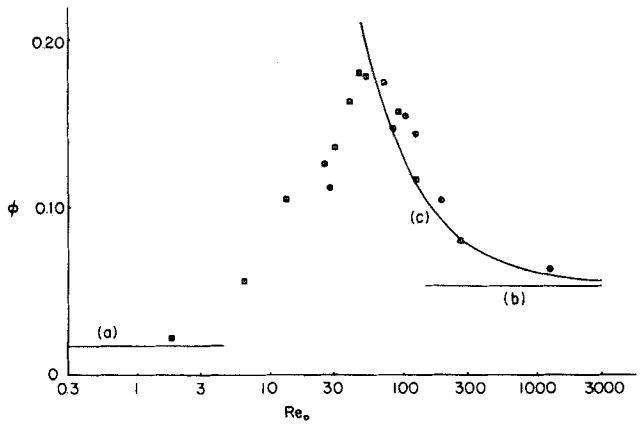

Fig. 8. Variation of flux composition on the axis in the transition between inviscid and free molecular flow. (a) Free molecular limit. (b) Inviscid limit. (c) Nearly inviscid theory for the orifice. Circles, converging nozzle, squares, orifice.

of Sherman's nearly inviscid flow theory for Reynolds numbers greater than 70 as indicated in Figure 7. The results confirm the validity of the recent detailed agreement achieved between Sherman's theory and Rothe's experiments (*14*) for $N_{Re} > 100$. Since Sherman's calculations for diatomic gases showed very little effect of nozzle shape, it is not surprising with monatomic gases to find results for the converging nozzle almost the same as those for the orifice.

A calculation of the separation to be expected by the Waterman-Stern mechanism at a Reynolds number of 100 predicts only a small fraction of the separation actually measured. With Mach number values of 6.2 and 19.6 for helium and argon, respectively, in Equation (6), the predicted flux on the axis at large x is 5.53 mole % argon (5.27 mole % argon in the stagnation chamber). For this Reynolds number the measured flux is about 13 mole % argon. The choice of Mach numbers is somewhat arbitrary since insufficient velocity distribution data are available. We base the Mach numbers on a static temperature of 15°K. at the cut-off surface which corresponds to that observed with pure gases in the transition region for $N_{Re} = 100$ (*19*).

We conclude that separation in free jets is due primarily to a pressure diffusion mechanism I and that under most conditions the free molecular flow mechanism II contributes only slightly to the effects observed. The nearly inviscid flow theory of diffusive separation is applicable when the nozzle is operated at Reynolds numbers down to 70. This verification of Sherman's theory gives added support to previous conclusions (*8, 11*) that separation at a probe or sampling tube by mechanism III can be substantial because in many cases the separation measured with a probe has been much greater than can be accounted for by diffusive separation in the jet alone.

ACKNOWLEDGMENT

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NOTATION

A_s = area of cut-off surface
C = constant as defined by Equation (3)
D = orifice or nozzle exit diameter
$\mathfrak{D}$ = binary diffusion coefficient
E = parameter as defined by Equation (2)
$E_2(y)$ = exponential integral as defined by Equation (7)
f = molar concentration fraction of heavy species
F_N = individual species nozzle flow rate
j = species flux
k = Boltzmann constant
m = molecular mass
m_0 = mean molecular mass at stagnation conditions
M = Mach number, $u/(\gamma kT/m)^{1/2}$, applying either to individual species or mixtures (for which the mean molecular mass is used)
N_{Re} = Reynolds number based on nozzle exit diameter and stagnation conditions (sonic velocity, stagnation density and viscosity)
N_{Sc} = Schmidt number, $\mu/\rho\mathfrak{D}$
s = distance along a streamline
T = static temperature, absolute
u = mean velocity (mass velocity)
x = axial distance downstream of nozzle exit plane
z = radius of cut-off hemisphere

Greek Letters

α = thermal diffusion coefficient
γ = specific heat ratio
θ = angle from axis, origin at nozzle exit
μ = viscosity
ρ = density
ϕ = molar flux fraction of heavy species

Subscripts

0 = stagnation conditions
1 = heavy species
2 = light species
S = cut-off surface

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