SAND75-0121
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Printed March 1975

GAS DYNAMIC PROBLEMS IN ISOTOPE SEPARATION[†]

(Report on a Workshop Held in Albuquerque
New Mexico, December 4 and 5, 1974)

K. J. Touryan (Principal Investigator)
Sandia Laboratories
Albuquerque, New Mexico

E. P. Muntz
University of Southern California
Los Angeles, California

L. Talbot
University of California
Berkeley, California

E. Von Halle
Union Carbide Corporation ORGDP
Oak Ridge, Tennessee

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B. Steels
Authorizing Official
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ABSTRACT

MASTER

A synopsis is given of the presentations and discussions held at a workshop on the gas dynamic problems inherent in aerodynamic isotope separation devices. Five principal aerodynamic devices are considered in some detail. These are: (1) the curved separation nozzle; (2) the opposite jet scheme; (3) the separation probe scheme; (4) the jet membrane device, and (5) the crossed-beam device.

[†]This workshop was supported by NSF, under Grant No. ENG75-05424 and by the U. S. Energy Research and Development Administration.

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SUMMARY

A synopsis is given of the presentations and discussions held at a workshop on the gas dynamic problems inherent in aerodynamic isotope separation devices. The workshop was held in Albuquerque, New Mexico, December 4 and 5, 1974.

The five principal aerodynamic devices considered in the workshop, and reported in the proceedings, are (1) the curved separation nozzle; (2) the opposing jet scheme; (3) the separation probe scheme; (4) the jet membrane device, and (5) the crossed-beam device.

The curved separation nozzle is considered to be by far the most advanced aerodynamic device and has been studied extensively at the Nuclear Research Center, Karlsruhe, Germany. Optimum operation for this device occurs at near-sonic flows, at a Reynolds number of 100 and a Knudsen number of 0.01. Detailed experiments are in progress with UF_6 gas for further optimization studies, but corresponding theoretical efforts are as yet incomplete. Much less information is available on the opposing jet scheme. Experiments are underway at the Nuclear Research Center in Karlsruhe, and preliminary results favor the curved nozzle over the opposing jet scheme. Numerical calculations are underway at Sandia Laboratories to provide a more extensive theoretical base for these measurements.

The separation probe is under investigation at Centre d'Etudes Nucleaires de Saclay, Paris, France, and Mobil Research, New Jersey. A large body of experimental data is available for various isotopic mixtures demonstrating separation (free stream $M \sim 30$, $Re \sim 100$). The critical need, at present, is to demonstrate separation of uranium isotopes, obtain the separation factor as a function of operating parameters and determine various aerodynamic losses

in the system. There is still some doubt at the precise nature of the probe separation effect itself. Monte-Carlo direct simulation calculations could settle this uncertainty.

Separation has been demonstrated in the jet membrane device using Ar, Ne and SF₆ isotopes, differentially penetrating into the structure of a free jet. The separation process is reasonably well understood in rarefied flows ($Kn \approx 1$) but recent experiments have shown separation to occur under near-continuum flow conditions thus necessitating a reevaluation of the whole process.

Series of experiments have been conducted with argon and xenon isotopes in crossed-beam devices demonstrating large separation factors. Supportive theoretical studies are at a rather primitive stage at present. As with the separation probe, a Monte-Carlo direct simulation scheme would help elucidate the separation phenomena in the crossed-beam geometry. Also, inelastic collision cross section data are needed for a more realistic evaluation of the scheme.

A special session was devoted to experimental techniques. The chemical and physical properties of the working fluid (generally UF₆), low densities, and small dimensions make diagnostic studies of aerodynamic separation devices very difficult. The commonly used techniques for the diagnostic studies of the various separation flows are of the invasive type (pitot probes, thermocouples). To obtain more reliable quantitative information, the ingenious use of optical techniques, such as uv or IR laser absorption spectroscopy and enhanced-signal laser Raman scattering, will be essential.

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SYMBOLS

A	Cross sectional area.
d,D	Probe or orifice diameter.
D^*	Nozzle diameter.
$\mathcal{D}$	Diffusion coefficient.
f	Distribution function.
F	Force. In Appendix F - Feed gas.
J	Collision terms in Boltzmann equation.
k	Boltzmann constant.
Kn	Knudsen number λ/L (λ - mean free path).
L	Characteristic length. In Appendix L - stage throughput.
m	Mass.
M	Mach number.
n	Number density. In Appendix n - number of stages.
p,P	Pressure. In Appendix P - product.
r	Turning radius or radial distance.
Re	Reynolds number.
S	Speed ratio $U/(2kT/m)^{1/2}$
t	Time.
T	Temperature.
u,U	Flow velocity.
$\bar{v}$	Molecular velocity.
V	Diffusion velocity.
W	Waste gas.
x,y	Concentrations in Eqs. 4-7 and A-1; cartesian coordinates.

SYMBOLS (cont)

Greek

α	Separation factor. See Eqs. 5-7 and A-1.
ψ or ϵ	Separation effect = $\alpha - 1$. See Eq. 4.
θ	Cut, as defined in Section IV.
μ	Viscosity; also mass ratio. See Eqs. 18-20.
ξ	See Eq. 4.
ρ	Gas density.
σ	Collision cross section.
τ	Time.
ϕ	See Eq. 11.

Subscripts

a,b	Probe arrays. See Eq. 9.
b	Beam particle. See Eqs. 18-20.
A	Auxiliary gas.
B	Background gas.
H	Heavy fraction.
j	Jet particles.
L	Light fraction.
o	Stagnation conditions.
p	Probe, except in Eqs. 12-17.
s	Source conditions (e.g., pressure).
x	Percent of total mass flux.

I. INTRODUCTION

A. Purpose and Scope of Workshop

The chief aim of the workshop held in Albuquerque, New Mexico, December 4-5, 1974, was to bring together a team of specialists in fluid mechanics, rarefied gas dynamics and related disciplines to review and critically evaluate the present level of understanding of the physical phenomena associated with isotope enrichment in aerodynamic separation processes.

Within the scope of the workshop aerodynamic separation processes were characterized as those involving diffusion of disparate masses in a binary or ternary gas mixture, driven principally by a pressure gradient. Such processes could be either equilibrium or nonequilibrium, with flow rarefaction levels varying from near free molecule, or $Kn = O(1)$, to near continuum, or $Kn = O(0.01)$.

To this end, the gas dynamics of five principal "aerodynamic devices" were discussed in detail. These were: (1) the curved separation nozzle (Trenndüse, or the Becker nozzle); (2) the opposing jet scheme; (3) the separation probe scheme; (4) the jet membrane device, and (5) the crossed-beam device.* A brief presentation was also made of a new process that involves velocity slip between disparate masses in a molecular beam. In addition, a session was devoted to gas dynamic and kinetic problems associated with laser isotope separation. The emphasis of the workshop was on the physics of the phenomena in these devices and consequently discussions concerning the economics of the processes were kept to a minimum. Briefly, the principle of operation of each of the five devices are as follows:

* Strictly speaking, only the first three of the above processes are aerodynamic. The latter two could be classified as inertial devices.

1. The separation nozzle, or the Trenndüse, pioneered by Becker and colleagues at Nuclear Research Center (NRC), Karlsruhe, Germany, uses the pressure gradient developed in a curved expanding supersonic, or near sonic, jet to effect the separation of uranium isotopes. As the expanding jet traverses the curved path, the heavier components are enriched in the vicinity of the curved wall (see Figure 1). A skimmer then divides the jet into two fractions, one enriched in light isotope, the other enriched in the heavy isotope.
2. The opposing jet scheme is a variant of the Trenndüse which uses two, or more, opposing nozzles without any kind of container vessel, to generate the pressure gradient via streamline curvature for isotope separation (Figure 2). Skimmers placed on either side of the collision plane separate the light isotope-enriched stream from the heavy isotope stream. This process too is being vigorously pursued at NRC, Karlsruhe.
3. The separation probe scheme utilizes the pressure gradient developed behind an oblique or a bow shock wave generated by tubular probe placed in a supersonic stream. Suction is applied to the probe and the heavy fraction is removed from the vicinity of the probe stagnation region (see Figure 3). The process patented by Fenn (1969) has been pursued both by Campargue and collaborators of CEN of Saclay, France, and by Mobil Research, New Jersey.

In all above three process, optimum operation is achieved in the Knudsen number range $0.01 < Kn \leq 0.1$.

4. The jet membrane device is a molecular process and operates by a differential penetration of a gas mixture into the structure of a free jet as depicted in Figure 4. Lighter molecules penetrate farther up the axis

Figure 1. Cross-section of the separation nozzle system used to date in the commercial implementation of the separation nozzle process.

Figure 2. Section through a separation nozzle system with opposed deflection guidance of uncontained gas streams.

Figure 3. Shock separation device.

Figure 4. Jet membrane process.

of the jet and an enriched fraction is withdrawn from an appropriate location. There are two versions of this process, the Campargue configuration and the Muntz-Hamel configuration. The latter is being supported by NSF and is also under study by Grumman Aerospace Corporation.

5. The crossed-beam device involves a beam, or a jet of molecules to be separated, crossed with another beam, or jet, of particles that are used to differentially deflect the molecules of interest. Skimmers separate the light isotope-rich section (see Figure 5). Crossed-beam separation schemes are being actively pursued both at NRC, Karlsruhe, Germany, and the United States.

Unlike the first three processes described above, the latter two are molecular processes and therefore are more closely related to gas effusion schemes than to bona fide aerodynamic schemes. The optimum operation for the crossed beam occurs at Knudsen numbers of order unity. For the jet-membrane there is some question regarding the condition of optimum operation (see Section V).

B. Economic Evaluation of Isotope Separation Processes

Although, as stated above, the chief aim of the workshop was the physics rather than the economics of the processes involved, one cannot divorce the two without losing sight of the ultimate objectives involved in any separation scheme. To maintain a proper perspective, and to keep the complete picture in mind, E. Von Halle of the Operation Analysis and Planning Division ORGDP, Oak Ridge, Tennessee, gave a brief review of the economic evaluation of isotope separation process. The details of his discussion are given in the Appendix.

Figure 5. Schematic view of the crossed beam device.

Von Halle, enumerated the desirable characteristics of an isotope separation process as follows: *

- 1 - high unit separation factor
- 2 - high unit throughput
- 3 - low process holdup and short plant equilibrium times
- 4 - low capital cost
- 5 - low operating cost

His discussion of the separation process included the separation stage, the separation cascade, the ideal cascade, the cascade upflow and the separative capacity.

Clearly, a new process would have to promise to produce separative work at a lower price than the present gaseous diffusion technology in order to qualify for consideration for incorporation into new plants to be contracted in the 1980's.

* See the Appendix for definition of these terms.

II. THE CURVED SEPARATION NOZZLE PROCESS

The chief contributor of this section was Dr. W. Ehrfeld of the Nuclear Research Center at Karlsruhe, Germany. A brief theoretical contribution was also made by J. N. Tunstall of ORGDP, Oak Ridge, Tennessee.

Of the five aerodynamic separation schemes discussed in the workshop, the Trenndüse or the curved separation nozzle, is by far the most advanced device and has been studied extensively from its initial conception up through a 10-stage pilot plant which has been operated in Karlsruhe today. Below, we summarize Ehrfeld's presentation of the curved separation nozzle first as it relates to the present commercial implementation of the method (mode 1) followed by the advanced concept of a double deflecting nozzle.

The standard Trenndüse: As discussed in the Introduction, Figure 1, represents the standard version of the Trenndüse. The flow Reynolds number is about 100 and the Knudsen number $Kn \approx 0.01$, which puts the flow in the continuum (slip) regime. The radius of curvature of the deflecting wall is about 0.1 mm designed to achieve optimum operating conditions at stagnation pressure P_0 near 1 atm. This requires nozzle design tolerances of order less than 1 micron, recently achieved by commercial outfits in Germany. To make flow measurements tractable, the nozzles were scaled up by a factor of 100 with a corresponding decrease in the operating pressure. Measurements were made through free molecule probes (probe diameter $d_p = 0.2-0.4$ mm) connected to a mass spectrometer for species analysis of the isotopes of the highly corrosive UF_6 (or its stand-in SF_6) with He or H_2 as the light carrier or auxiliary gas.* Measurements in the flow showed that SF_6 is supersonic ($S \approx 2.5$), He remains subsonic for an expansion pressure ratio $P_0/P = 4-5$, and the gas

* Note that the auxiliary gas, which constitutes 90 percent or more of the gas mixture, enhances the SF_6 or UF_6 flow velocity, thus increasing the centripetal separating force.

mixture is near sonic (to minimize shock induced losses). Furthermore, wall friction and low density effects produce parabolic velocity profiles throughout the curved nozzle.

Measurements have also shown that nonequilibrium conditions prevail near the outer wall, where the velocity distribution function is clearly bimodal instead of being Maxwell-Boltzmann. Low number of self-collisions among the heavy isotope molecules are chiefly responsible for this phenomenon.

Figure 6 shows a comparison of the maximum separation to be expected at equilibrium in a curved flow with the experimentally obtained values of the separation as a function of the cut. It can be seen that the experimentally obtained values for the isotopic separation of U-235 and U-238 are appreciably higher than the values calculated from equilibrium theory.*

This increase in separation can be attributed to a transient effect caused by the presence of the light auxiliary gas in the system. This can be seen from the expression for the diffusion velocity of $^{238}\text{UF}_6/^{235}\text{UF}_6$ relative to He due to the pressure gradients, viz.,

$$V_{H,L} - V_{\text{He}} = \frac{D_{HL,\text{He}}}{\rho} (m_{H,L} - m_{\text{He}}) \frac{\partial \log p}{\partial r}$$

$$= \frac{3}{8n \sigma_{HL,\text{He}}^2} \left\{ \frac{kT(m_{H,L} + m_{\text{He}})}{m_{H,L} m_{\text{He}}} \right\}^{\frac{1}{2}} \frac{(m_{H,L} - m_{\text{He}})}{kT} \frac{U^2}{r} \quad (1)$$

where H,L - heavy and light isotopes, U - flow velocity, r - turning radius.

Here $m_H > m_L$ and therefore $V_H - V_{\text{He}} > V_L - V_{\text{He}}$ or $\tau_{D(238)} < \tau_{D(235)}$.

* Equilibrium separation is characterized by the condition where each component of the mixture has assumed a density distribution that corresponds to the centrifugal force, analogous to the barometric altitude distribution of gaseous components of different weights in a gravitational field.

The highest isotopic separations are obtained in experiments in which UF_6 is the most dilute, (UF_6 concentration varied from 1.5 percent up to 10 percent).

The Advanced System: It is interesting to note from Figure 6 that the reason the isotopic separation effects for equilibrium flow cannot exceed a maximum value, even for infinitely high centrifugal forces, is the fact that the heavy isotope is concentrated into an increasingly narrow layer as the flow velocity increases. Although concentration gradients increase as the square of the velocity, the thickness of the layer decreases proportionately thus establishing a limiting value for a given cut. The auxiliary gas essentially delays the formation of a limiting narrow layer because of differences in the diffusion time of the two isotopes. To take maximum advantage of this "delay" time one can use a double deflecting system with sweep gas deflection in the second stage.

Figure 7 is a sketch of this advanced system. The heavy fraction from the first nozzle which has a high UF_6 concentration is deflected by a gas jet having low UF_6 content and high auxiliary gas concentration. By this means it is possible to make better use of the transient separation effect due to the auxiliary gas in the second stage and lower frictional losses (this design is superior to a double deflecting nozzle in which both deflections are caused by curved walls).

Figure 8 shows results of a few preliminary separation experiments in the second nozzle (Becker et al, 1974), in which the flow is deflected by a sweep gas. The plot shows the dependence of the separation effect ψ (or $\alpha - 1$) on the ratio between the sweep gas pressure and the suction pressure. In this case the sweep gas flow rate was set to 10 percent of the overall gas flow, the cut being one-third. If the suction pressure is

Figure 6. Separation effect for the standard curved nozzle showing nonequilibrium effect.

Figure 7. Advanced separation nozzle system. The heavy fraction of the first nozzle is deflected in the second nozzle by a sweep gas.

Figure 8. Results of separation experiments in the case of flow deflection by a sweep gas.

increased, the velocity of the flow will increase. However, there is another result: if this pressure ratio is increased, the UF_6 jet will be swept by the auxiliary gas more and more strongly, i.e., the UF_6 streamlines are spread farther and farther apart and the UF_6 is distributed more uniformly in the centrifugal field. It is seen that already an increase in this pressure ratio from approximately 1.3 to 2.1 will cause the effect of isotope separation to quickly rise to values of approximately 1 percent. These values are comparable with the separation effect of 1.5 percent existing in the first nozzle. In more recent experiments now under way, higher values than 1 percent have already been achieved.

Finally, measurements with D_2 as the light carrier gas instead of H_2 , showed that separation factors in the flow were higher with D_2 than with H_2 for $P_0/P \leq 5$. A cross-over occurs when the pressure ratio is greater than 5. This difference is attributed to smaller $\text{UF}_6 - \text{D}_2$ collision mean free path λ than for $\text{UF}_6 - \text{H}_2$ (Speed ratio S for H_2 remain always higher than for D_2).

Theoretical Consideration: Very little has been done theoretically with the curved nozzle flow (see for example Becker et al, 1971). The reason for it is obvious. For a proper treatment of the flow field boundary layer approximations are inadequate and the full Navier-Stokes equation must be used for a ternary gas mixture, including nonequilibrium effects. At the workshop, J. N. Tunstall (1975) presented the paper describing a mathematical model of diffusion of UF_6 and a light auxiliary gas in a curved duct. This model treated the steady state radial convection and diffusive fluxes of the ternary gas mixture of interest when the tangential convective velocity was specified and independent of turning angle. The numerical technique employed was described and results were shown of the effects of separation of varying inlet mixture composition, inlet pressure, and

the magnitude of the tangential velocity. In concluding he showed comparisons with experiment of a similar model reported by Dr. W. Ehrfeld to illustrate that this theory did not completely describe the magnitude of the separation of Becker's separation nozzle.

Another attempt (not discussed at the workshop) was made at Sandia Laboratories, Albuquerque, to simulate numerically the flow of two isotopic species (no auxiliary light gas) in a two-dimensional curved channel. Viscous stresses, heat conduction, and momentum and energy exchange between the two species were all included in the formulation via the complete conservation laws (see below). Initial results are encouraging in that the velocity field at the end of the channel (45° turn) and isotope separation effects look reasonable. Further work is needed to specify better boundary and initial conditions and reduce numerical instabilities.

Recommendations for Further Studies: It was clear from the discussion that the obvious area where additional work should be done is in the numerical simulation of the Trenndüse, both in its standard form and in the double-nozzle version. Although some skepticism was expressed at the possibility of extending the binary gas numerical calculations to ternary viscous gas mixture with nonequilibrium effects, initial studies with colliding jets have been encouraging (see Section III).

Another important area of investigation is the optimization studies. The obvious difficulty here is the plethora of parameters involved and their interdependence. Over ten governing parameters can be identified for the standard curved nozzle, alone. These include: geometric dimensions; inlet pressure; suction pressure; the cost, and the UF_6 - concentration. Parametric optimization studies are being conducted experimentally at NRC,

Karlsruhe, using an on-line digital computer for real-time evaluation of the separation efficiency (see Becker et al, 1973).

A third area of investigation could be improved flow diagnostics, especially using laser optical techniques (see Section VII, Experimental Techniques).

III. THE OPPOSING JET SCHEME

The two contributors to this discussion were Dr. W. Ehrfeld of NRC, Karlsruhe, Germany, and Dr. R. L. Fox of Sandia Laboratories, Albuquerque.

In one sense the opposing jet scheme (see Figure 2) is an extension of the curved nozzle device, especially when one considers the double deflection version of the latter. It has an intrinsic advantage over the curved nozzle in that friction losses are minimal in the opposing jet scheme. However, the latter is still in a relatively primitive stage of development.

Experimental Studies: Ehrfeld presented the results of experiments in progress at Karlsruhe with pure UF_6 , H_2/UF_6 and He/SF_6 , using subsonic and low supersonic jets. For example, in pure UF_6 with $P_0/P = 20$, and 20 percent cut, separation factors are of order 1.004 (same order as gaseous diffusion). For $P_0/P = 60$ ($P_0 = 10$ torr) $\alpha = 1.005$ with the potential of increasing it up to 1.008. With light auxiliary gas added α reaches 1.011 ($P_0/P = 3$). It is interesting to note that with P_0 varying from 5 to 20 torr, as the expansion pressure ratio increases from 1.5 to 4, there is a cross-over in the value of α for the curved wall versus opposing jets. The latter show a lower separation factor than the former at the high pressure ratio. An anomaly has been observed in the H_2/UF_6 experiments where a sudden downward jump in separation factor occurs as the stagnation pressure is increased from 5 torr to 20 torr, while holding the pressure ratio constant.

Detailed results of these experiments will be available in early 1975 from Karlsruhe.

Theoretical Studies: Fox discussed the results of some preliminary calculations of two opposing, supersonic jets for a binary mixture (UF_6

isotopes). Numerical solutions were obtained for the two-dimensional, time-dependent Navier-Stokes equation given in terms of the sum and difference of the two species, including viscous and heat conduction effects. The solutions were obtained in a quadrant bounded by the jet centerline, the jet symmetry plane, a free boundary and a backwall, allowing the development of vortices after the turning of the jet streamlines.

Global and species separation results were obtained for a range of parameters such as $Kn = 0.01 - 0.05$, $M = 2-12$, and nozzle separation distances of 1 to 1.5 diameters. A mass average separation factor α_x was defined (separation factor for x-percent of total mass flux) and for Mach 6 flow, 15 nozzle radii above the centerline, α_x was found to be as high as 0.01 (or $\alpha = 1.01$) at 10 percent of total mass flux. For 25 percent of total mass flux (roughly 1/4 cut), α_x was down to 0.005. These results are in good qualitative agreement with Ehrfelds measurements.

An animated discussion ensued concerning the validity of extending these computations to a ternary mixture where very low UF_6 concentrations (less than 5 percent) would lead to very low numbers of self-collisions among the UF_6 molecules which in turn would result in highly non-Maxwellian velocity distribution function for the UF_6 isotope gas, notably for $^{235}UF_6$.

A follow-up study [Fox and Eaton (1975)] has shown that a proper ordering of momentum and energy relaxation times in self- and mutual-collision for a 95 percent He , 5 percent UF_6 (4.96 percent $^{238}UF_6$, 0.04 percent $^{235}UF_6$) mixture would indeed result in a non-fluid behavior for the U^{235} component, i.e., $U^{235} - U^{235}$ collisions are negligible. However, the resulting set of governing conservation equations yield a coupled set for the auxiliary gas (He) and the heavy isotope (both of which are shown to behave

as proper fluids, i.e., sufficient self-collisions to yield a Boltzmann distribution), and neither of which contain momentum or energy exchange terms with the U^{235} component. Conservation equations for the two fluid components can then be solved independently of the U^{235} component and the results substituted into the governing equation for the lighter isotope. This can be seen from the governing Boltzmann equations for the species, i.e.,

$$\frac{Df_A}{Dt} = J \overset{O}{/}_{AL} + J_{AH} + J_{AA} - \text{Auxiliary gas} \quad (2a)$$

$$\frac{Df_H}{Dt} = J \overset{O}{/}_{HL} + J_{HH} + J_{HA} - \text{Heavy isotope} \quad (2b)$$

$$\frac{Df_L}{Dt} = J \overset{O}{/}_{LL} + J_{LH} + J_{LA} - \text{Light isotope} \quad (2c)$$

where

$$\frac{D}{Dt} \equiv \frac{\partial}{\partial t} + \bar{v} \cdot \frac{\partial}{\partial \bar{r}} + \frac{\bar{F}}{m} \cdot \frac{\partial}{\partial \bar{v}} \quad \text{and the } J\text{'s represent collisional terms}$$

(L,H - light and heavy isotope, A - auxiliary gas).

Recommendation for Further Studies: Unlike the curved nozzle case, the opposing jet scheme is still in its infancy both experimentally and theoretically. Extensive optimization studies are needed to permit a fair economic evaluation of the opposing jet scheme, especially vis a vis the curved nozzle.

The same comments made for the Trenndüse apply to the theoretical investigation and diagnostic studies needed for the opposing jet scheme.

IV. SEPARATION PROBE STUDIES

The contributors to this session were Professor John Fenn, Yale University; Dr. Roger Campargue, Centre d'Etudes Nucleaires de Saclay, Paris; Dr. James R. White, Mobil Research and Development Corporation, Princeton, and Professor Wesley R. Harris, M.I.T.

Development of the concept: Professor Fenn began the session by tracing the development of the separation probe concept. He pointed out that the early studies of separation of gases of different masses in jets were what led to the concept of the separation probe, where it was observed by Reis and Fenn (1963) that the separation in jet flows was actually induced by the flow field in front of the sampling probe. Since then, others have done further work on this phenomenon [(J. H. Chang, (1967); R. Campargue and M. Constant, presented at the 8th Symposium on Rarefied Gas Dynamics, Stanford, 1972)] and it is now generally agreed that the separation effect in a jet proper is small compared to that produced by a sampling probe.

Professor Fenn described a simple qualitative model of the probe separation effect, in terms of the inertia of the gas molecules. According to this model, the molecules of the heavier gas on passing through the shock wave in front of a sampling probe in a supersonic stream retain more of their axial momentum than the lighter gas molecules, and thus the gas collected from the orifice of the sampling probe is enriched in the heavier species. A lively discussion followed, in which the question was raised as to whether this conceptualization of the separation process was in fact equivalent to the description of the process which occurs in the separation nozzle; namely, baro-diffusion produced by curvature of flow streamlines, in the present instance occurring in a flow around the sampling probe downstream of the shock wave rather than in a curved nozzle flow.

Experimental Studies in France: Dr. Campargue's first remarks were concerned with the competing effects of jet-induced and invasion separation (cf. discussion of jet membrane, below) in measurements of separation occurring along the axis of a free jet expansion. Whereas jet-induced separation concentrates the heavier species on the axis, invasion toward the jet axis of the background gas is more effective for the lighter species. Thus, the two effects tend to counteract one another. Data obtained on jet-induced separation which revealed a decrease in separation effect with distance downstream from the nozzle can be explained in terms of this invasion effect.

Campargue next presented the results of a series of measurements on probe separation in Argon 36/40 isotopic mixtures. Several different types of probes were employed (see Figure 9). They consisted of 4 mm O.D. cylinder probes, 16 mm long described as:

- (a) hemispherical-cylinder probes (H.C.P.), with hemispherical noses and sampling orifices of either 1 mm, 2 mm or 2.5 mm diameters,
- (b) blunt tubular probes (B.T.P.), with end openings cut off perpendicular to the probe axis,
- (c) sharpened tubular probes (S.T.P.), with the outside diameters of the tube openings chamfered to produce sharp-edged orifices.

The probes were operated in a free jet expansion either singly or in equilateral triangular arrays of 7, 13 and 19 probes, with

$$2 < \frac{\text{Probe spacing}}{\text{Probe external diameter}} < 2.5 \quad (3)$$

HEMISPHERE CYLINDER PROBES (H. C. P.)

BLUNT TUBULAR
PROBE (B. T. P.)

SHARPENED TUBULAR
PROBE (S. T. P.)

LENGTH TO DIAMETER RATIO

$$L_{ext} / D_{ext}$$

ABSOLUTE DIMENSIONS

$$D_{ext}$$

ARRAYS OF PROBES
 $\frac{\text{ARRAY DIA}}{\text{JET DIA}}$

$$\delta / D_{ext}$$

Figure 9. Enhancement of ψ and θ by optimization of probe design.

Jet source pressures ranged from below 40 to 1340 torr, and for each probe configuration data were obtained at varying values of x/D , the position along the jet axis normalized by the free jet orifice diameter.

Several trends were exhibited in Campargue's data. Of the three types of probes employed, the sharp tubular probes (S.T.P.) produced the greatest separation effect, especially in the case of probe arrays. Data were presented in terms of the elementary separation effect $\epsilon = \xi - 1$ for the heavy species.

$$\xi = \frac{(x_{\text{HEAVY}}/x_{\text{LIGHT}})_2}{(x_{\text{HEAVY}}/x_{\text{LIGHT}})_0} \quad (4)$$

is the stage separation factor, expressed in mole fractions of the heavy and light species. Subscript 2 refers to the flow passing through the probe, and subscript 0 denotes the free stream flow ahead of the probe.

The stage separation factor for the light species would be

$$\alpha = \frac{(x_{\text{HEAVY}}/x_{\text{LIGHT}})_1}{(x_{\text{HEAVY}}/x_{\text{LIGHT}})_0} \quad (5)$$

where subscript 1 refers to the flow massing about the probes. The maximum observed values of ϵ were approximately $\epsilon_{\text{max}} \approx 0.07$. It was pointed out by Professor Muntz that these maximum values of ϵ occur at locations along the jet axis which corresponds to $Re_2 \approx 10$, where $Re_2 = \rho_2 u_2 D / \mu_2$ is the post-shock Reynolds number based on probe external diameter D . The data were in good agreement with the theory of Louvet (1973) (see Figure 10) over the range of validity of the theory, which applies to hemisphere cylinder probes in the limit of very small sampling.

Figure 10. Comparison of theory and experiment for hemispherical nose separation probe with small heavy fraction cut.

The various arrays of probes behaved in essentially the same fashion as the single probes. Values of $\epsilon_{\max} \approx 0.07$ were observed. Some of the factors which influence the maximum value of ϵ were discussed, such as the end-wall effect, alignment of the probes with the diverging streamlines of the free jet expansion, and variation of flow properties with distance from the center-line of the free jet.

Campargue also discussed the cut θ which might be achieved by a probe array in a free jet expansion. The cut θ was defined as the ratio of the heavy fraction flow passing through the probes to the total flow in the jet. He represented θ as the product of two factors, $\theta = \theta' \cdot \theta''$. The factor θ' is the cut based on swept area of a single probe. The second factor θ'' is a geometrical factor which expresses the solidity of the probe array. A value of $\theta' = \frac{1}{2}$ has been suggested by Muntz. Campargue estimated values of $1/12 < \theta'' < 1/7$ for circular orifices, and $0.3 < \theta'' < 0.4$ for an array of slits, and suggested that the final expected cut θ could be a maximum of 0.05 for the circular openings before interaction between probes became serious and would be in the range $0.15 < \theta < 0.2$ for the slits.

Several other results were reported by Campargue. One observation was that changes in source temperature from 300°K to 1750°K produced little change in the observed values of ϵ . Another was that experiments made with a mixture of 1% argon and 99% hydrogen carrier gas exhibited a decrease in separation over that for pure argon at high source pressures, but the opposite effect occurred at low source pressure (see Figure 11). This result cannot be considered definitive for two reasons, one being the difficulty in analyzing accurately a mixture with so small a concentration of argon, and the other being the lack of certainty as to whether the probe pressure was high enough to produce a well-defined bow shock wave.

Figure 11. Experimental separation curves for hemisphere cylinder probe as function of distance from nozzle exit for low and high source pressures.

The effect of increasing probe pressure (produced by throttling the flow withdrawn through the probe) was to produce initially a small increase in ϵ , but this was followed by a very rapid decrease in ϵ as the probe pressure approached the post-shock stagnation pressure.

Experimental Studies in the U.S.: The work at the Central Research Laboratories of the Mobil Research and Development Corporation is concerned with the evaluation of the Fenn patent ("Methods and Apparatus for Separation of Components from Gaseous Streams," U.S. Patent, 3, 465, 500, Sept. 9, 1969). The concept is that of shock-induced separation (see report of Professor Fenn's presentation).

Dr. White introduced two separation factors. He denoted the free stream flow as the "feed" flow (Campargue's subscript "0"), the "heads" flow as that passing around the probe (Campargue's subscript "1"), and the "tails" flow as that extracted by the probe (Campargue's subscript "2"). In terms of these flows, White's definitions are, in terms of the mole fraction of the lighter species

$$\text{Stage separation factor } \alpha = \frac{\left(\frac{x_{\text{LIGHT}}}{1-x_{\text{LIGHT}}} \right)_{\text{HEADS}}}{\left(\frac{x_{\text{LIGHT}}}{1-x_{\text{LIGHT}}} \right)_{\text{TAILS}}} \quad (6)$$

$$\text{Tails separation factor } \xi = \frac{\left(\frac{x_{\text{LIGHT}}}{1-x_{\text{LIGHT}}} \right)_{\text{FEED}}}{\left(\frac{x_{\text{LIGHT}}}{1-x_{\text{LIGHT}}} \right)_{\text{TAILS}}} \quad (7)$$

Experiments gave values of $\alpha_{H_2-D_2} \approx 2$ and $\alpha_{Ar^{36}-Ar^{40}} \approx 1.1$. White noted that these values were approximately equal to the mass ratio $\left(\frac{M_{HEAVY}}{M_{LIGHT}}\right)^N$, with $N \approx 1$, and thus suggested that if the extrapolation can be made, $\alpha_{U^{238}-U^{235}}$ should be about 1.0086.

Data presented for H_2-D_2 mixtures showed α to vary between 1.85 and 2.1 as the streamline withdrawal factor was increased from 0 to 0.5 (see Figure 12).

Next the concept of cut in the shock separation process was discussed. White used the same definition as Campargue, as applied to the light species. (Campargue's values of the cut were for the heavy species, and thus $\theta_{Campargue} = 1 - \theta_{White}$). He defined the streamline withdrawal fraction SWF as the ratio of the mass flow extracted by the probe to the mass flow contained in the probe area projection, and thus gave for the cut of an array of probes in a stream of total cross-sectional area A

$$\text{array cut} = \left(1 - SWF \cdot \frac{\Sigma A_p}{A}\right) \quad (8)$$

where ΣA_p = the total cross-sectional area occupied by the probes. If a separation stage contained, say, two arrays of probes, one downstream from the other, the stage cut would be given by

$$\text{stage cut} = \left[1 - \left(SWF_a \cdot \frac{(\Sigma A_p)_a}{A} + SWF_b \cdot \frac{(\Sigma A_p)_b}{A}\right)\right] \quad (9)$$

where subscripts a and b refer to the first and second arrays respectively.

Now since

Figure 12. Experimental separation curves for H_2/D_2 and Ar^{36}/Ar^{40} isotopic mixtures for separation probe.

$$\frac{\text{Separative Work}}{\text{Pumping Work}} \sim (\alpha-1)^2(\text{cut})(1-\text{cut}) \quad (10)$$

it was considered important to evaluate the following factors:

- (1) Relation to SWF to α for a single orifice;
- (2) How much area within a duct could be occupied by orifices in a single plane;
- (3) How much arrays of orifices could be used before aerodynamic losses reduce supersonic flow to unusably low values.

A single sampling probe shock separation apparatus was described next in which the measurements on $\text{H}_2\text{-D}_2$ and $\text{Ar}^{36}\text{-Ar}^{40}$ were made. White's measurements were reported in terms of plots of $\ln \xi / \ln \left(\frac{M_{\text{HEAVY}}}{M_{\text{LIGHT}}} \right)$ versus probe Knudsen number (see Figure 13). For pure argon mixtures ($\text{Ar}^{36}\text{-Ar}^{40}$), the maximum value of ξ to be obtained was about 1.07 at a probe Reynolds number $\text{Re}_2 \approx 10$, which agrees very well with Campargue's values. For $\text{Ar}^{36}\text{-Ar}^{40}$ diluted in helium in a 4:1 dilution ratio, his maximum value of ξ was approximately 1.16, which is higher than has been observed by Campargue in his hydrogen-argon mixture measurements. The minimum value of $p_s D_*$ (the product of source pressure and nozzle diameter) in Campargue's experiments was $p_s D_* = 20$, whereas White's values were obtained for a value $p_s D_* = 10$, so this may indicate a trend of increasing ξ with decreasing $p_s D_*$.

Five constraints were listed with respect to the shock separation process:

- (1) Ratio of mean free path to orifice dimension, which limits the maximum pressure in the duct for practical orifice openings.

Figure 13. Separation of $\text{H}_2\text{-D}_2$ and $\text{Ar}^{36}/\text{Ar}^{40}$.

- (2) Mach number in UF_6 limited to 3.5 at most because of constraint of practical expansion ratios.
- (3) Cut not greater than 0.7-0.8.
- (4) Turbulent losses along duct walls and around probes probably not greater than 10-fold, including reconvergence, to keep recompression work of heads flow and pump displacement volumes within acceptable bounds.
- (5) Stage α for UF_6 must be of the order of 1.007 or greater to compete with gaseous diffusion.

The critical investigative directions at present, according to Dr. White, were to demonstrate separation of UF_6 and the separation factor as a function of operating parameters in his system and to determine the aerodynamic losses for supersonic flow of UF_6 through a duct and system of orifices.

Theoretical Studies: The last formal presentation in the session on separation probes was given by Professor Harris. He reported on a theoretical analysis which he is currently carrying out on the probe separation effect. Thus far he has analyzed the separation produced by a curved shock wave in a supersonic flow, but he intends to continue the analysis so as to include the influence of the flow field produced by the probe itself behind the shock wave.

The basic assumption in Harris' analysis is that the shock wave is spherical. His analysis follows that of R. Chow (1959), in utilizing an expansion of the dependent variables in a power series in Re^{-1} where $Re \gg 1$. Thus, for any dependent variable ϕ ,

$$\phi = \phi^{(0)} + \frac{1}{Re} \phi^{(1)} + \dots \quad (11)$$

where the Reynolds number is based on free stream conditions and the shock radius of curvature R_0 . In a He-Ar mixture, with mole fraction varying from 0.1 to 0.5, at $M = 2$ and 8, and $Re = 100$, no separation occurs in the shock, to first order. The first order equations are similar to those employed by Sherman (1965).

Harris' analysis results in universal curves for the dependent variables which are independent of Reynolds number. No comparisons with experiment were presented because, as Professor Harris explained, the theory was not yet complete. A Monte-Carlo direct simulation method employed by Vogenitz (1967) and Bird (1969) and applied to binary or ternary mixtures would be the most suitable way of analyzing shock separation for $Re \leq 10$.

Discussion of the Probe Separation Effect & Recommendation: Two main points were brought out in the discussions which followed the formal presentations. The first, which has already been mentioned, had to do with the nature of the probe separation effect itself. Whereas most view the observed separation as being due primarily to pressure gradient diffusion (baro-diffusion) associated with the sharply curved streamlines in the post-shock merged layer flow ahead of a probe, others appear to attribute the separation to the curved shock wave itself. This may be largely a question of semantics, however, since strongly-curved streamlines would not exist were it not for the shock wave. In a subsonic flow, for example, the streamline curvature produced by a probe is so gentle that although in principle baro-diffusion would exist here also, its magnitude would be exceedingly small. However, one should note that inertial effects are the governing separation mechanism in molecular processes, such as crossed beam devices, including one scheme wherein high speed, heavy gas particles are directed into a stagnant gas of

light particules (cf crossed beam discussion below). A simple, momentum balance analysis indicates that the penetration distance in this case is proportional to the mass of the penetrating particle and the mean free path λ .

The second main point of the discussion had to do with the magnitude of the viscous losses which would inevitably be present in any separation probe system. Proponents of the shock separation approach maintained that since the expansion to supersonic flow would be carried out isentropically, losses in the expansion would be small, and only the viscous losses through the shock waves and in the flow around the probes would have to be considered, as contrasted with the separation nozzle system where the flow throughout the entire separation nozzle was in the viscous flow regime. However, others were of the opinion that the shock and probe viscous losses in the shock separation scheme would in fact be greater than those of the separation nozzle. As White pointed out in his presentation, this is one of the primary questions which has to be answered in the evaluation of the shock separation method.

V. THE JET MEMBRANE

The main contributors to this session were professors E. P. Muntz of the University of Southern California and B. B. Hamel of Drexel University, Pennsylvania.

Background and Theoretical Considerations: The invasion separation effect of the free jet shock wave structure was found in 1966 at Saclay [Campargue (1967)]. It operates by the differential penetration of a gas mixture or isotopic mixture into the structure of a free jet (Figure 14). The first mention of background gas penetration into portions of a very rarefied free jet was likely that of Fenn and Anderson (1966). Over the next several years, two independent developments were begun and reported in 1969. Campargue (1969a, 1969b) published papers during 1969 on background penetration into free jets. Muntz, Hamel and Maguire (1970) published a paper on a simple explanation of the observations made by them using electron beam fluorescence to study free jet characteristics. Campargue made the first direct observation of background penetration separation (1969a, 1970) by obtaining high elemental separation effects for a He - Ar mixture and the argon isotopes Ar^{36} and Ar^{40} , as well as $^{32}\text{SF}_6$ and $^{34}\text{SF}_6$. Muntz, Hamel and Scott (1971) published at least a semi-quantitative explanation of Campargue's measurements based on the simple model presented by Muntz et al (1969).

Further electron beam data was presented at the 7th Rarefied Gas Dynamics Symposium by Muntz and Maguire (1970). A formal theory of the phenomena was recently given by Brook and Hamel (1972) and Brook, hamel and Muntz (1972). At the 9th Rarefied Gas Dynamics Symposium, Rosengard (1974) presented an "ad hoc" Monte-Carlo analysis of separation induced by the penetration phenomena, which appears to explain Campargue's previous results in some detail.

Figure 14. Experimental configuration of Campargue's separation experiments.

Very briefly, the background penetration separation depends on a certain degree of rarefaction of the interaction between a jet and its background. The background contains the species that are to be separated. The jet gas is merely a medium to cause separation to occur. In the approximate spirit of Muntz et al (1969) background species penetrates the jet according to the relationship

$$n_{B_i} = n_{B_{i\infty}} \exp^{-r_p/r} \quad (12)$$

where $(n_{B_{i\infty}})$ is the density of the i 'th specie background gas far from the jet, r_p is the penetration distance, r the radial distance from the jet origin along the jet centerline, and n_{B_i} is the number density of species i . When the interaction between the jet and background is rarefied, the phenomena can be scaled by the plume Knudsen number, where

$$Kn_p = r_e/r_p, \quad (13)$$

r_e is the mean free path of the jet molecules in the undisturbed background gas and r_p is the distance measured from the jet origin to the point which the average background molecule of a particular specie penetrates. For two penetrating species, the jet centerline concentration ratio becomes from Equation 12

$$\frac{n_{B_1}}{n_{B_2}} \left(\frac{n_{B_2}}{n_{B_1\infty}} \right) = \exp \left(- \frac{r_{p_1} - r_{p_2}}{r} \right). \quad (14)$$

The general form of this expression has been confirmed (Muntz et al, 1971), using the electron beam fluorescence probe, for Kn_p 's greater than unity. Campargue's measurements also indicate some separation at a much smaller Kn_p , on order of 0.02.

Recent Experiments. There have been recent experimental results obtained by Deglow (1974) using the apparatus shown in Figure 15 to measure the separation of He and Ne as a function of several parameters. These include: the distance r that the extraction probe extends through the orifice; the rarefaction level, and the flux through the sampling probe compared to the flux obtained in simple diffusion through the same sampling probe exposed to the background number density with no jet. As one would expect, there is a decrease in the flux as $\alpha - 1$ is increased.

The validity of a system using the jet membrane depends on the outcome of a cascade optimization using flux and $\alpha - 1$ as parameters, as well as a knowledge of the highest pressures (or least rarefaction) for which the process will give satisfactory separation. The initial supposition was that when the flow approaches continuum the separation effect would disappear.

A summary of Deglow's (1974) experiments is shown in Figure 16. The open symbols were obtained using the apparatus of Figure 15 with N_2 as the jet gas. In these experiments the rarefaction parameter $D\sqrt{P_{B\infty}P_O}$ ranged from 20 to 200 $\mu\text{Hg cm}$, where D is the orifice diameter, $P_{B\infty}$ and P_O are the background and stagnation pressure in microns Hg, respectively. The process gas was a mixture of He and Ne in which separation was clearly observed. The pressure ratio $P_O/P_{B\infty}$ in these experiments was large and always greater than 10. Furthermore, the least rarefied flow ($D\sqrt{P_{B\infty}P_O} = 200$) corresponds to a plume Knudsen number of 0.01, which is closer to being

Figure 15. Upstream sampling configuration jet membrane.

Figure 16. Flux versus separation curves showing data from recent experiments in jet membrane device.

continuum than rarefied. The filled symbols are results obtained using the apparatus shown in Figure 17. These experiments go up to rarefaction parameters of 2600 $\mu\text{Hg cm}$, with $P_{B\infty}$ ranging up to 50 torr and $P_0/P_{B\infty} = 2$. These high pressure results have led to a re-examination of the previous high pressure operation limits imposed in the analysis of the jet membrane process.

To this end, the high pressure experiments were modeled, as a first attempt, by a uniform diameter jet of sonic speed, with diffusion from the jet boundary, or edge, where the concentration is assumed to be uniform. The one-dimensional time-dependent diffusion equation was modified to represent downstream distance, x , instead of time, viz.,

$$\underbrace{v \frac{dn}{dx}}_{\text{axial sweep}} = \underbrace{D \frac{d^2 n}{dy^2}}_{\text{transverse penetration by continuum diffusion}} \quad (15)$$

For $r_p/2x \gg 1$, Eq. 15 yields an expression analogous to the rarefied case given by Eq. 14, i.e.,

$$n = \frac{n_0}{\sqrt{\pi}} \left(e^{-r_p^2/2x} \right) \left(r_p/2x \right)^{-1/2} \quad (16)$$

Preliminary results of sampling probe flux versus $\alpha/\alpha_{\text{gas dyn}}$ look remarkably similar to the results predicted by the old MHM theory [Muntz et al (1969)]. A more accurate model is presently under study, but the general conclusion is that the jet membrane process is operative at pressure heretofore considered high (i.e., $p_{B\infty} \sim 80$ torr).

JET ORIFICE DIA. = 0.026 IN.
SAMPLING TUBE DIA. = 0.008 IN
(EXTENSION 1 ORIFICE DIA.)

Figure 17. High pressure experimental configuration.

There was much discussion concerning the similarity between the high pressure jet membrane and mass diffusion. Hamel argued that there were significant quantitative differences between the two that offer more advantageous boundary conditions for the diffusion induced by the jet membrane configuration. For example,

- (a) In the classical MHM mode, the velocity distribution function f is highly nonMaxwellian, whereas f in sweep diffusion is Maxwellian and the penetration velocity is small.
- (b) The separation factors for sweep diffusion between species 1 and 2 and the jet membrane can be given by

$$\alpha_{\text{sweep}} = \exp \left\{ \frac{vL}{D} \left[\frac{1}{D_1} - \frac{1}{D_2} \right] \right\}$$

$$\alpha_{\text{jet membrane}} = \exp \left[\frac{\Delta r_p}{r} (r_p/2x) \right] \quad (17)$$

where, for $r_p/2x \gg 1$, α can be made very large.

- (c) In the jet membrane case, the jet by preferential scattering creates, in effect, multiple molecular beams directed radially inward, i.e., background particles with increased velocity and direction.

Recommendation for Further Studies. Several points need further clarification in the jet membrane process. These are:

- (1) A better understanding of low rarefaction, or high pressure, limits of the process and the determination of the optimum configurations in the high pressure limit.

- (2) The maximum ratio of upflow to separating jet flow.
- (3) Problems of pressure drop in small diameter withdrawal tubes and the existence of a suitable jet fluid with rather stringent requirements.

VI. MOLECULAR BEAMS AND JETS

The contributors to this session were Dr. Jurgen Gspann of NRC, Karlsruhe, Germany, and Prof. James B. Anderson of Pennsylvania State University.

A. The Crossed Beam Device

Gradual deflection of the particles of a molecular beam by multiple collisions with lighter molecules of a crossed free jet leads to an angular separation of beam particles of different masses, or collision cross sections. In an equivalent scheme, a molecular beam of mass M is directed into a stagnant gas of a lighter mass (m). The penetration distance then depends on the mass of the penetrating beam particle. In fact, a simple momentum analysis shows that the penetration distance in this case depends on the ratio M/m and the collision mean free path, λ .

Although work is in progress on crossed beam devices for isotope separation both in the United States, and in Germany, very little unclassified information is available from the U.S. The most extensive work in the open literature comes from Europe, notably from Karlsruhe (Gspann, 1974). To this end, a presentation was made by J. Gspann on a crossed beam device depicted in Figure 5, where the beam consists of xenon isotopes, 129 and 132, accelerated as 1% seed gas in hydrogen and deflected through an angle λ by an ammonia jet. The velocity of the ammonia jet is 1.02×10^5 cm/sec and that of the beam, 2.01×10^5 cm/sec.* The key limiting factor for mass separation is the randomization of the beam particle velocities

* Details of this experiment are given in Gspann & Vollmar (1974).

caused by the statistical distribution of collision parameters in the multiple collisions of a beam particle with cross-jet molecules. To this end Gspann presented a central limit theorem representing the randomly distributed velocities of the beam particle, after successive collisions with the jet molecules, by a homogeneous Markoff chain (Gspann, 1974). In spite of the obvious limitations inherent in such an approach, Figure 18 shows reasonable agreement between theory and experiment. The chief drawback of the central limit theorem is the fact that it does not take into account the randomness introduced by variations in the initial velocity of the beam. Furthermore, the mean number of collisions ($= 4.91$) in the above experiment were far too small for the small deflection per collision assumption to hold. Hamel and Brook (private communication) have analyzed the collision between a monoenergetic UF_6 beam and a monoenergetic H_2 jet using the distribution function for mixtures developed by Hamel (1965) viz.,

$$f_n = (2 kT_n / m_b)^{-3/2} \exp \left\{ - \frac{(\bar{v} - \bar{v}_n)^2 m_b}{2kT_n} \right\} \quad (18)$$

where

$$\bar{v}_n = \mu_b \bar{v}_{n-1} + \mu_j \bar{v}_j \quad (19)$$

$$T_n = T_{n-1} (1 - 2\mu\mu) + \frac{(m_j + m_b)}{3k} \mu\mu^2 (\bar{v}_{n-1} - \bar{v}_j)^2 \quad (20)$$

and $\mu = \frac{m_k}{m_j + m_b}$; j - jet; b - beam; n - no. of collisions.

Figure 18. Angular profiles of the intensity of the xenon isotope Xe^{132} and of the isotope abundance ratio I^{129}/I^{132} of the isotopes Xe^{129} and Xe^{132} in an atomic beam of xenon deflected by a cross-jet of ammonia. The theoretical profile (solid lines) are fitted at the experimental points at $\alpha = 22.5^\circ$ (I^{132} - profile) and at $\alpha = 25^\circ$ (profile of I^{129}/I^{132}). The broken curve of I^{129}/I^{132} is calculated using the experimental I^{132} - profile.

Now assuming that Eqs. (2) and (3) can be used for successive collisions, f_n can be found for any collisions. Hamel (1966), shows that if one considers the deflection of a particle through 90° as a "complete collision," it will take m_b/m_j cross encounters with the light species before a heavy particle can be said to have undergone a collision. For UF_6/H_2 system this means 175-875 collisions rather than the 1-5 collisions for which the flux versus separation curve of Figure 19 has been obtained. In other words, the fluxes shown in Figure 19 must be reduced by the ratio m_b/m_j .

There are other effects that should be included before the crossed beam analysis could be considered reliable for predicting isotope separation factors and fluxes. These are:

- a. Beam and jet divergence caused by thermal effects. The monoenergetic assumption should be regarded as a first approximation only.
- b. Real gas effects - elastic versus inelastic collisions with data on inelastic collision cross sections for UF_6 .
- c. Proper geometric considerations for a practical device, i.e., two-dimensional effects.
- d. Possible shock waves in high-pressure, supersonic operation of the beam-jet scheme.
- e. Perturbations introduced by the finite thickness of the skimmer, or knife-edge.

It is clear from the above discussions that the most attractive and practical technique for analyzing beam-jet interactions would be a Monte-Carlo direct simulation scheme, similar to that used by Bird (1969). One could start with several thousand molecules in the beam with Maxwell-Boltzmann distribution, follow the molecular motions exactly and treat

Figure 19. Calculation of flux versus separation curve for monoenergetic $^{235}\text{UF}_6/\text{H}_2$ beam and monoenergetic jet.

collisions between beam molecules and the jet molecules (with thermal spread), using statistical sampling until the beam molecules pass through the collision zone.

B. The Velocity Slip Process

Unfortunately, much of what constitutes crossed-beam studies for isotope separation in the United States is unavailable in the open literature. Consequently, Prof. Anderson, Pennsylvania State University, discussed a new method of separating isotopes in a gaseous mixture which takes advantage of the differences in velocities of isotopic species in a molecular beam formed by expansion of the mixture with a light gas from a nozzle source.

In generating molecular beams by the "seeded-beam" technique [Abuaf et al, (1967), Haberland et al, (1972)] the acceleration of heavy species by the light gas is incomplete and leads to a velocity slip between the various species. When such slip occurs in the expansion of a mixture of two heavy species with a light gas, the final velocity U_i of each heavy species depends on its mass. For different average velocities of two isotopes accelerated by a light gas, their velocity distributions are separated as shown in Figure 20. The distribution for each species is approximately a Gaussian distribution centered on the average velocity $\bar{u}_1$ or $\bar{u}_2$, which differ by an amount $\Delta\bar{u}$. With a time-of-flight apparatus or a rotating velocity filter, the stream may be separated into components having velocities lower and higher than the midpoint indicated by the dashed line of Figure 20.

Anderson shows that for isotopes of nearly equal mass, the separation factor α is given by the approximation (Anderson and Davidovits, 1975).

Figure 20. Velocity distribution of two isotopes.

$$\alpha \cong 1 + \frac{4}{\sqrt{\pi}} S \frac{\Delta \bar{u}}{\bar{u}} \quad (21)$$

where $S = \frac{\bar{u}}{(2kT/m)^{1/2}}$ is the speed ratio of the isotope. The ratio $\Delta \bar{u}/\bar{u}$ is evaluated in terms of two parameters, the ratio $\bar{u}/\bar{u}_c$ and Kn_{slip}^{-1} , used in Abuaf et al, (1967) and Haberland et al, (1971) to correlate the experimental data.* Assuming these data to be representative of a 1-5 percent mole fraction UF_6 (balance He) Anderson calculates $\alpha \approx 1.1$, which corresponds to a velocity slip ratio $\Delta \bar{u}/\bar{u}$ of 0.00212. (The maximum α for gaseous diffusion process is 1.0043 and typical Trenndüse values are of the order of 1.01-1.05.)

A key consideration for the above process, of course, is the use of a skimmer which will enable one to extract the core of the expanding jet to form a molecular beam, without affecting the species velocities. The process, however, shows merit and should be pursued further to provide a better quantitative base for economic considerations.

* $\bar{u}/\bar{u}_c$ - average velocity of heavy species/velocity for complete continuum expansion. $Kn_{slip}^{-1} = \sqrt{2} n_o \sigma D m_{av} / (m_{h1,2} - m_p)$, n_o - number density of molecules in the nozzle, σ - collision cross section between heavy and light molecules; D - nozzle exit diameter.

VII. EXPERIMENTAL TECHNIQUES

The contributors to this session were G. F. Malling, Oak Ridge Gaseous Diffusion Development Division, R. A. Hill, Sandia Laboratories, Albuquerque, and Dr. W. Ehrfeld, NRC, Karlsruhe, Germany.

The chemical and physical properties of the working fluid, low densities, and small dimensions make diagnostic studies of aerodynamic separation devices very difficult. In most cases, the dimensions of the devices have to be scaled up to practical levels to allow any kind of measurement. This in turn would mean lowering the pressure proportionally. For example, the present dimensions of a prototype separation nozzle (see Figure 1) are of the order of 1 mm or less to permit operation at stagnation pressures, P_0 , close to atmospheric. To make diagnostic measurements possible, the nozzle radius has to be scaled up a factor of 100 or more thus reducing P_0 to 5 mm or less.

At pressures below 5 torr, both qualitative (flow visualization) and quantitative (optical diagnostic) measurements are difficult to make. Multipass schlieren measurements have been successfully used by the NRC-Karlsruhe team (Ehrfeld, et al) for pressures as low as 1 torr. A better technique would involve using paraboloids instead of spherical mirrors for low density. This technique is being pursued at Sandia Laboratories, Albuquerque. Gas discharge techniques, suitable for operation below 10 mm, dissociate the heavy gas (whether SF_6 , C_7F_{14} , or UF_6) and are therefore useless. Among quantitative techniques, concentration measurements through electron-beam-induced fluorescence are well suited to pressures below 1 torr. However, problems would arise in the case of fluorine compounds such as SF_6 , C_7F_{14} , or UF_6 because of the electron affinity of halogenes, and halogene compounds.

Quantitative techniques used most extensively by Ehrfeld and co-workers are of the invasive type and include pitot probes for the determination of velocity profiles in the Trenndüse through pressure measurements and chromel/alumel thermocouples for temperature measurement. In all devices, measurements of isotope separation have been by mass spectrometry. Ehrfeld and co-workers have also used infrared absorption spectroscopy with a CO_2 laser to study the flow field in the separation nozzle. However, they were unable to determine isotopic separation effects in UF_6 . A recent article on the vibrational spectrum and force field of UF_6 by McDowell et al, (1974) gives results of measurement of the $^{235}\text{UF}_6 - ^{238}\text{UF}_6$ isotope shifts in ν_3 and ν_4 . The shift in ν_3 was found to be 0.65 cm^{-1} and for ν_4 , 0.16 cm^{-1} . In principle then, one should be able to use IR laser absorption spectroscopy for in situ determination of isotope separation in an aerodynamic device.

Hill discussed laser-Raman scattering techniques for determination of species concentration and rotational temperatures. Because of the structure of the UF_6 (or SF_6) molecule one would not expect to see isotopic effect in the Raman signal. However, for other types of heavy isotope mixtures used in simulation experiments, laser-Raman spectroscopy may be a very useful tool even at pressures below 5 torr, through signal enhancement. This can be accomplished by reflecting the incident laser light through the observation region a large number of times. Such a system consists of an ellipsoidal mirror arrangement called a Light-Trapping Cell [Hill and Hartley (1974)] which has actually yielded a 100-fold increase in the Raman-scattered signal strength. A second technique for enhancing the Raman signal involves a "comb" filter developed by Barrett and Meyers (1971). The "comb" filter, a Fabry-Perot interferometer provides a method whereby all the rotational Raman lines from one species can be detected simultaneously while effectively

blocking the Rayleigh line and most of the lines from other bands by making use of the fact that the rotational Raman lines have a constant spacing in frequency (neglecting centrifugal distortion) of $4B_v$ (where B_v is the rotational constant for the vibrational state v).

Recommendations

The limitations on optical techniques imposed by low-density effects and the particular structure of the UF_6 molecule call for novel applications of these techniques. Very little work has been done in this area and the field is wide open for ingenious ideas. Some promising avenues of investigation are:

- A. For qualitative measurements: multipass schlieren or schlieren photographs with parabolic mirrors.
- B. For quantitative measurements: IR or uv absorption spectroscopy using a multipass scheme or an opto-acoustic chamber such as the spectrophone [see Kreutzer (1971)]; enhanced Raman signals using the light-trapping cell or the comb filter.

APPENDIX

by E. Von Halle

The first step in comparing processes is to define the separation task of interest. In this regard we have found it useful to introduce the concept of separative capacity and to compare processes on the basis of cost per kg USW.

Theoretical Background

As a model for an isotope separation process, consider a stage-wise enrichment process such as gaseous diffusion.

The Separation Stage. A simple isotope separation stage can be considered as a black box which divides a feed stream into two product streams, one enriched and the other depleted with respect to the desired isotope. A schematic representation of a simple stage is shown in Figure A-1.

Figure A-1

The concentration of the desired isotope in the enriched stream is designated by y and in the depleted stream by x . For every isotope separation process there exists a fundamental quantity α , independent of concentration, which

relates the concentrations of the two streams leaving the stage. This relationship is

$$\alpha = \left(\frac{y}{1-y} \right) / \left(\frac{x}{1-x} \right) . \quad (A-1)$$

In the case of a gaseous diffusion stage, where the enrichment is due to the effusion of a fraction of the feed stream through a porous membrane, α is proportional to the square root of the ratio of the molecular weights of the components of the mixture. Isotope separation technology differs from that of conventional separation processes in that the enrichment afforded by a single stage is quite small, that is, α is a number only slightly greater than unity. It is therefore both convenient and permissible to recast Equation A-1 in the appropriate form

$$y - x \approx (\alpha - 1) x (1 - x) \equiv \Psi x (1 - x) \quad (A-2)$$

where Ψ , called the stage enrichment factor, is a very small positive quantity.

The Separation Cascade. An isotope separation cascade is the name given to an assembly of separating elements, or stages connected together for the purpose of accomplishing some specific separation task. Just as a simple separation stage divides a feed stream into an enriched and a depleted stream, the simple cascade also divided a single feed stream into a product stream, enriched with respect to the desired isotope, and a waste stream depleted with respect to the desired component. Figure A-2 shows the nomenclature associated with a simple cascade.

Figure A-2

For such a cascade one has the following two material balance relationships:

the total material balance,

$$F = P + W , \quad (A-3)$$

and the desired component material balance,

$$Fx_F = Py_P + Wx_W . \quad (A-4)$$

The scheme used to form cascade from the individual stages depends to some extent on the type of equipment under consideration. In the case which will be considered here, it will be assumed that stages with any desired throughput are readily available. When this situation prevails a simple series connection of the stages, such as is employed in Figure A-3 is normally employed.

Figure A-3

Figure A-3 shows, in particular, a section of the enricher. From a combination of the material balances around the top of the enricher stage n , which can be written

$$L_n y_n = (L_n - P)x_{n+1} + Py_p \quad (\text{A-5})$$

frequently called the operating line equation, and Equation A-2 relating the concentrations leaving a stage, frequently called the equilibrium line equation, the gradient equation can be obtained. For the enriching section the resulting equation is

$$x_{n+1} - x_n = \frac{L_n}{L_n - P} \left\{ \psi_{x_n} (1 - x_n) - \frac{P}{L_n} (y_p - x_n) \right\}. \quad (\text{A-6})$$

However, since in the case of isotope separation, the stage enrichment is small, the finite difference in this equation may be replaced by the differential enrichment per stage, dx/dn , and P may be neglected with respect to L_n producing the gradient equation

$$\frac{dx}{dn} = \psi x(1 - x) - \frac{P}{L} (y_p - x) \quad (A-7)$$

which governs the enriching section. For the stripping section of the corresponding equation

$$\frac{dx}{dn} = \psi x(1 - x) - \frac{W}{L} (x - x_w) \quad (A-8)$$

is similarly obtained.

The Ideal Cascade. An ideal cascade may be defined as follows: it is a cascade in which there is no mixing of streams which differ in concentration anywhere within the cascade. Thus, in an ideal cascade, the two streams which join at each stage inlet to form the feed to that stage have the same concentration. Referring to Figure A-3, it can be seen that this condition for ideality is satisfied if

$$x_{n+2} = y_n \quad (A-9)$$

The concentration of the downflowing stream from stage $n+2$ can be replaced by $x_n + 2(dx/dn)$ while the concentration of the upflowing stream from stage n is equal to $x_n + \psi x(1 - x)$. Thus, the condition for ideality expressed by Equation A-9 yields the gradient equation

$$\left(\frac{dx}{dn}\right)_I = \frac{\psi x(1 - x)}{2} \quad (A-10)$$

for an ideal cascade. Integration of this equation gives the number of stages required in an ideal cascade to span any desired concentration range, and the combination of this equation with Equations A-7 and A-8 can be used to give the required stage upflow at any point in the cascade. One obtains the following set of equations which can be used to design an ideal enricher and stripper:

Enriching Section

$$N_I = \frac{2}{\Psi} \ln \left\{ \left(\frac{x}{1-x} \right) \middle/ \left(\frac{x_F}{1-x_F} \right) \right\} ; L_I = \frac{2P(y_p - x)}{\Psi x(1-x)}$$

Stripping Section

$$N_I = \frac{2}{\Psi} \ln \left\{ \left(\frac{x_F}{1-x_F} \right) \middle/ \left(\frac{x}{1-x} \right) \right\} ; L_I = \frac{2W(x - x_W)}{\Psi x(1-x)} \quad (A-11)$$

The Cascade Upflow. A particularly interesting quantity which can now be calculated for the case of an ideal cascade is the sum of all the stage upflows in the cascade. In the case of gaseous diffusion, since the stage upflow is the amount of gas which effuses through the porous membrane in each stage and must be recompressed between stages, the summation of stage upflows is directly proportional to the total membrane area required for the cascade and also to the power required for compression of the gas. The expression for the total upflow, ΣL , is obtained as follows

$$\Sigma L(\text{cascade}) = \Sigma L(\text{enricher}) + \Sigma L(\text{stripper})$$

$$\begin{aligned}
 &= \int_{x_F}^{y_P} L \frac{dn}{dx} dx + \int_{x_W}^{x_F} L \frac{dn}{dx} dx \\
 &= \int_{x_F}^{y_P} \frac{4P(y_P - x)}{\psi^2 x^2 (1 - x)^2} dx + \int_{x_W}^{x_F} \frac{4W(x - x_W)}{\psi^2 x^2 (1 - x)^2} dx \\
 &= \frac{4}{\psi^2} \left\{ Pv(y_P) + Wv(x_W) - Fv(x_F) \right\} \quad (\text{A-12})
 \end{aligned}$$

where $v(x)$ termed the value function is defined by $v(x) = (2x - 1) \ln \{x/(1-x)\}$.

The expression in brackets in Equation A-12 is called the separative capacity of the cascade.

The Separative Capacity. The separative capacity is therefore a quantity which is directly proportional to the sum of the cascade upflows in an ideal cascade, and hence, directly proportional both to the size of the cascade and to the power required to perform a given separation. It can be seen that the separative capacity is measured in the same units as the streams P, F, and W. However, care should be taken never to confuse the separative capacity with a stream. The separative capacity of a cascade is the rate at which it performs separative work. It is usually measured in kilograms of uranium per year (kg. USW/yr.) or in metric tons of uranium per year (MTUSW/hr.). The SW, standing for separative work, is included in the units specifically to prevent confusing the separative capacity with the process streams.

Example. The following example illustrates the use of the preceding equations. Consider a cascade to produce material containing 3.5% uranium-235 from natural feed containing 0.71% U-235 with the waste concentration equal to 0.2% U-235.

The production of a single kilogram of product material will require 6.4706 kg. of feed and be accompanied by 5.4706 kg. of waste. From the material balance Equations A-3 and A-4.

The separative work associated with the production of 1 kg. of product is given by

$$\begin{aligned}\Delta U &= P v(y_P) + W v(x_W) - F v(x_F) \\ &= 3.0846 + 5.4706 \times 6.1876 \\ &\quad - 6.4706 \times 4.8704 \\ &= 5.421 \text{ kg. USW .}\end{aligned}$$

Thus, a plant with a separative capacity of 6,000 MTUSW/yr. could produce 1,107 metric tons of product per year. [From equation A-12.]

Assuming that the stage enrichment factor is equal to 0.004, the ideal cascade would have a total stage upflow of 277×10^6 MTU/yr., would contain 812 stages in the enricher, 636 stages in the stripper, and would have the shape sketched in Figure A-4. [From equations A-6 and A-11.]

Although the word "uranium" does not appear in the title of this conference, I believe a poll of the attendees would show that the separation of uranium isotopes is the number one concern of many of us.

Figure A-4

Figure A-5 shows a recent estimate of the demand for enriched uranium based upon estimates of the nuclear power generating facilities to be installed in the U.S. and abroad. What should be seen from this graph is the following:

- (1) The present capacity of the U.S. diffusion plants when restored to full power operation (6000 MW) is approximately 17,000 metric tons uranium of separative work per year. The annual demand is expected to exceed present capacity in 1976.
- (2) The cascade improvement program and the cascade up-rating program will increase the output of the U.S. diffusion plants to about 27,000 metric tons U separative work per year. The demand is expected to exceed this figure by 1979.

Figure A-5

There has been some slippage in the demand curve since this chart was prepared and the dates at which new plants are needed have been postponed for a few years. It is now anticipated that new enrichment plants will be required starting in 1982 or 1983 with regular additions of capacity required after that date such that the capital requirement for new plants will exceed \$1 billion per year for the following decade. It is this anticipated large expenditure for uranium isotope separation that accounts at least in part for the great interest at this time in uranium isotope separation processes.

Table A-1 presents the main cost factors for both the gaseous diffusion process and the gas centrifuge process. For the case of our present gaseous diffusion technology, it can be seen that with capital charges computed at 15% per year and power costs of 10 mills/kw-hr the breakdown of the unit cost of separative work would be as follows:

Unit Capital Cost	\$24.00/kg USW
Unit Power Cost	\$24.00/kg USW
Other Operating Cost	<u>\$ 1.80/kg USW</u>
Total Cost	\$50.00/kg USW

A new process would have to be able to produce separative work at a lower price than this in order to qualify for consideration for the new plants; it would have to have a total cost of separative work less than the operating cost of the gaseous diffusion plants given above in order to replace the gaseous diffusion plants now in operation.

TABLE A-1

SUMMARY OF ENRICHMENT PLANT ESTIMATES
8.75 Million SWU/Year Plants at New Site
FY 1974 Dollars

	<u>Gaseous Diffusion Plants</u>		<u>Gas Centrifuge Plants</u>
	<u>CIP Technology</u>	<u>Advanced Technology</u>	<u>Range of Costs</u>
Capital Investment, \$MM	1400	1200	1130-1710
Specific Investment, \$/SWU/yr	160	137	129-195
Operating Cost (Excluding Power*) \$MM/yr	16	16	70-115
<p>* A gaseous diffusion plant of 8.75 million SWU/yr capacity requires about 2400 megawatts of electric power. A gas centrifuge plant of the same capacity requires perhaps only 10% as much power.</p>			

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