Reports

Isotope Separation in a "Seeded Beam"

Abstract. *A new method of separating isotopes in a gaseous mixture is described. The method 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. For the separation of the hexafluorides of uranium-235 and uranium-238 the technique has an estimated separative work factor about 500 times higher than the gaseous diffusion process and 100 times higher than the curved-jet method.*

We describe here a new method for separating gaseous mixtures of isotopes. It takes advantage of the differences in velocities of heavy species of different masses in a molecular beam formed by expansion of a mixture with a light gas from a nozzle source. When molecular beams are generated by the "seeded beam" technique (*1–5*) the acceleration of heavy species by the light gas is, in many cases, incomplete, and the phenomenon of "velocity slip" occurs. When such slip occurs in the expansion of a mixture of two heavy species with a light gas, the final velocity of each heavy species depends on its mass. Conditions of the expansion can be selected to produce appreciable velocity differences for heavy isotopic species, such as $^{235}UF_6$ and $^{238}UF_6$, in the molecular beam formed. With the relatively narrow velocity distributions obtained for the heavy species, segregation by any of several techniques can produce an isotope separation effect significantly higher than can a single stage of the conventional gaseous diffusion process (*6*) or the curved-jet process of Becker *et al.* (*7*).

The most convenient way to separate the isotopes on the basis of velocity difference in the beam is to use a vaned rotating velocity selector mounted radially around the source. This arrangement allows nearly continuous beam flow (*8*). The molecular beam or jet is directed toward the velocity selector, which consists of rotating curved vanes shaped so that molecules having desired velocities pass between the vanes while molecules with lower velocities strike the vanes. The molecules passing through the vanes enter an outer chamber while the molecules that are intercepted enter an inner chamber. The molecules are then removed by pumping from each of the chambers. The desired part of the beam is intercepted only when the narrow edge of the vane passes through the beam. For this arrangement the molecular beam or jet may be conveniently formed by orifices or nozzles of various shapes. Specifically, the use of a long narrow slit offers the advantage of increased mass flow. Many jets may be distributed radially in conjunction with a single vaned rotor.

The flow of a gas mixture through an orifice or nozzle into a low-pressure region may be characterized by the Knudsen number $Kn = \lambda/D$, where λ is the upstream mean free path and D is the orifice diameter. At high densities ($Kn \lesssim 0.001$) the collision rate between heavy and light species is sufficiently high to produce nearly equal average species velocities. At low densities ($Kn \gtrsim 1$), as in effusive flow, the species velocities are independent, each inversely proportional to the square root of the species mass. In the intermediate regime ($0.001 \lesssim Kn \lesssim 1$) the heavy species are accelerated in collisions with light species, but the collision rate is not sufficient to produce equal average species velocities. With proper precautions the core of the expanding jet may be extracted by a skimmer to form a molecular beam without affecting the species velocities (*5*).

We now estimate the extent of separation obtainable by the technique we have described. For two isotopes accelerated by a light gas and having different average velocities, the velocity distributions are separated, as shown schematically in Fig. 1. The distribution for each species is approximately Gaussian, centered on the average velocity, $\bar{u}_1$ or $\bar{u}_2$. The average velocities 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 Fig. 1.

It is customary to define a separation factor α expressed in terms of the number of molecules in each of the separated streams. For a division of isotopes 1 and 2 according to velocity left (L) or right (R) of the midpoint, the usual definition of α is

$$\alpha = \frac{n_{1L}n_{2R}}{n_{1R}n_{2L}} \qquad (1)$$

Here, for example, n_{1L} is the number of molecules of isotope 1 with velocities to the left of the midpoint. To evaluate α one must know the velocities and velocity distributions of the molecules in the beam.

Measurements of velocities and velocity distributions in molecular beams of gas mixtures have been made in several laboratories (*1–4*). For moderately high source densities the velocities of heavy species are found to be in approximate agreement with predictions (*3*) based on the Chapman-Enskog diffusion equation. Results have been successfully correlated (*2, 3*) in terms of a velocity ratio $\bar{u}/\bar{u}_c$ (the ratio of the average velocity of the heavy species to the average velocity for complete continuum expansion) and a modified Knudsen number Kn_{slip} defined as

$$Kn_{slip}^{-1} = 2^{1/2}\, n_0 \sigma D \frac{m_{avg}}{m_h - m_l} \qquad (2)$$

in which n_0 is the number density of molecules in the nozzle, σ is the cross section for collisions between heavy and light molecules, D is the nozzle exit diameter, and m_{avg} is the weighted average molecular mass for the mixture of heavy and light molecules of masses m_h and m_l.

Velocity distributions of heavy species in seeded beams have been found to be approximately those of a one-dimensional Maxwellian distribution (Gaussian) about the average velocity and may be characterized by a speed ratio S_i, the ratio of the average velocity $\bar{u}_i$ to the thermal velocity at a temperature T_i, corresponding to that of the Maxwellian distribution

$$S_i = \frac{\bar{u}_i}{(2kT_i/m_i)^{1/2}} \qquad (3)$$

Table 1. Comparison of α and δW for separation of $^{235}UF_6$ and $^{238}UF_6$. The basis for δW is $L = 1$ mole and $\theta = 0.5$ (see text).

Process	$\alpha - 1$	δW
Velocity slip	0.1	2.5×10^{-3}
Gaseous diffusion	0.0043	4.6×10^{-6}
Curved jet (*7*)	0.010	2.5×10^{-5}

where k is the Boltzmann constant and m_i is the mass of species i. Experimental measurements of S for heavy species by Haberland *et al.* (*4*) and Abuaf *et al.* (*2*) gave different results, which are at present unexplained. Haberland *et al.* (*4*) for mixtures of Xe and H_2 found S in approximate agreement with the predictions of a simplified kinetic theory for source-flow expansions (*3*). Abuaf *et al.* (*2*) found S somewhat lower than the kinetic theory predicts for Ar in mixtures with He.

For Gaussian distributions the quantities in Eq. 1 are given by

$$n_{1L} = \tfrac{1}{2}n_1\left[1 + \operatorname{erf}\frac{\Delta\bar{u}/2}{(2\mu_1)^{1/2}}\right]$$

$$n_{1R} = \tfrac{1}{2}n_1\left[1 - \operatorname{erf}\frac{\Delta\bar{u}/2}{(2\mu_1)^{1/2}}\right]$$

$$n_{2L} = \tfrac{1}{2}n_2\left[1 - \operatorname{erf}\frac{\Delta\bar{u}/2}{(2\mu_2)^{1/2}}\right]$$

$$n_{2R} = \tfrac{1}{2}n_2\left[1 + \operatorname{erf}\frac{\Delta\bar{u}/2}{(2\mu_2)^{1/2}}\right] \qquad (4)$$

Here n_1 and n_2 are the total numbers of molecules of isotopes 1 and 2, and μ_i is the second moment of the velocity distribution about the mean for each isotope. For the one-dimensional Maxwellian distribution we have

$$(2\mu_i)^{1/2} = \left(\frac{2kT_i}{m_i}\right)^{1/2} \qquad (5)$$

Combining this with Eq. 3 we have μ_i in terms of S_i and $\bar{u}_i$ as

$$(2\mu_i)^{1/2} = \frac{\bar{u}_i}{S_i} \qquad (6)$$

For isotopes of nearly equal mass the velocities and speed ratios are nearly equal, and μ_1 and μ_2 can be designated by a single term μ. For low values of the argument of the error function in Eq. 4, to be expected for isotopes of nearly equal mass, the separation factor α is given with good accuracy by the approximation

$$\alpha \cong 1 + 4 \operatorname{erf}\frac{\Delta\bar{u}/2}{(2\mu)^{1/2}} \cong 1 + \frac{4}{\pi^{1/2}} S \frac{\Delta\bar{u}}{\bar{u}} \qquad (7)$$

We now consider the specific case of a molecular beam formed from a mixture of $^{235}UF_6$ and $^{238}UF_6$ with helium as the accelerating gas. If the mole fraction of the UF_6 is low (say 5 percent or less) and the nozzle is operated in the intermediate pressure regime, one can expect a large velocity slip for each of the heavy species. To our knowledge, relevant data for UF_6 mixtures are not available. However, the conditions in the experiments described in (*2*) and (*4*) are sufficiently close to those desired that the data may be used to estimate the parameters important in UF_6 isotope separation.

In our estimate of the separation factor α we will use values of the speed ratio S presented in (*4*). [In figure 3 of (*4*) S is plotted as a function of $Kn^{-0.4}$.] The ratio $\Delta\bar{u}/\bar{u}$ in Eq. 7 is evaluated as follows. Using a plot of $\bar{u}/\bar{u}_c$ as a function of Kn_{slip}^{-1} from (*2*) or the plot of $(\bar{u}/\bar{u}_c)^2$ against $Kn_{\text{slip}}^{-0.4}$ from (*4*), we obtain the slope $d(\bar{u}/\bar{u}_c)/d(Kn_{\text{slip}}^{-1})$ or $(1/\bar{u}_c)d\bar{u}/d(Kn_{\text{slip}}^{-1})$. The desired ratio $\Delta\bar{u}/\bar{u}$ is then

$$\frac{\Delta\bar{u}}{\bar{u}} = \left[\frac{(1/\bar{u}_c)\,d\bar{u}}{d(Kn_{\text{slip}}^{-1}}\right]\frac{\bar{u}_c}{\bar{u}}\Delta Kn_{\text{slip}}^{-1} \qquad (8)$$

where $\Delta Kn_{\text{slip}}^{-1}$ is the difference in Kn_{slip}^{-1} for the two isotopes calculated from Eq. 2.

We varied Kn_{slip}^{-1} to obtain the maximum value for α. Separate computations were made with the data from the two references. From the data of (*2*) the optimum value of Kn_{slip}^{-1} is about 14 and the corresponding difference in Kn_{slip}^{-1} is 0.12. The slope $(1/\bar{u}_c)d\bar{u}/d(Kn_{\text{slip}}^{-1})$ in this region is 0.015 and the ratio $\bar{u}/\bar{u}_c$ is 0.85. Substituting these numbers into Eq. 8 we obtain $\Delta\bar{u}/\bar{u}$ as 0.00212. The value of S in Eq. 7 depends on the initial composition of the mixture. We will complete our calculation of α for two initial compositions: (A) 1.0 mole percent UF_6 and 99.0 mole percent He and (B) 5.0 mole percent UF_6 and 95.0 mole percent He.

For beam composition A, the value of m_{avg} in Eq. 2 is 7.5. The inverse Knudsen number Kn^{-1} is 654 [$Kn^{-1} = Kn_{\text{slip}}^{-1}\,(m_h - m_l)/m_{\text{avg}}$]. For a nozzle diameter of 0.1 cm and a hard-sphere collision cross section of 100 Å^2, this value of Kn^{-1} is obtained with an initial gas density of about 4.5×10^{17} cm^{-3}. The corresponding value for S is 24.4 (*4*, figure 3). With this value in Eq. 7 we obtain

$$\alpha \cong 1.12 \qquad (9)$$

For beam composition B, Kn^{-1} is 229, obtained with a pressure-diameter product of 0.5 torr cm. The corresponding value for S is 17.6, and from Eq. 7 we obtain

$$\alpha \cong 1.09 \qquad (10)$$

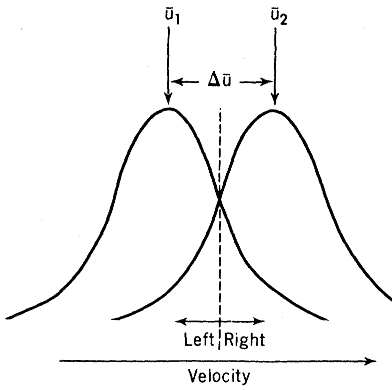

Fig. 1. The velocity distribution of two isotopes.

Since the data from (*2*) and (*4*) are not in complete agreement, they yield different values for the optimum Kn_{slip}^{-1}. Using data from (*4*) we obtained the maximum value of α with Kn_{slip}^{-1} equal to about 2.8. The maximum values for α, however, are nearly the same. In this case we have $\alpha \cong 1.10$ for composition A and 1.07 for composition B.

Averaging the values of α obtained with data from (*2*) and (*4*) and initial compositions A and B, we have $\alpha = 1.1$, which is reasonable for the new separation process. For the higher mole fraction of UF_6 (5.0 percent) the velocity difference between the two isotopes might be reduced by collisions between them. However, for the lower mole fraction of UF_6 (1.0 percent) we estimate that, on the average, each UF_6 molecule will have less than one collision with other UF_6 molecules during the expansion, and expect the effect of such collisions to be negligible.

The factor α is a measure of the degree of separation given by a process. In comparing the economics of different separation processes it is useful to consider values of the separative work factor δW (*6*) per stage

$$\delta W = L\theta(1 - \theta)(\alpha - 1)^2 \qquad (11)$$

where L is the molar throughput of separated isotopes, θ is the fraction of the separated isotopes leaving in one exit stream, and $1 - \theta$ is the fraction leaving in the other. For the gaseous diffusion process, in which the flow of each isotopic species through a porous barrier is inversely proportional to the square root of its molecular mass, the maximum value of α is 1.0043 for the separation of $^{235}UF_6$ and $^{238}UF_6$. In the curved-jet process a typical value of α is 1.010. Values of $\alpha - 1$ and δW for the seeded-beam (velocity-slip) process outlined above, the gaseous diffusion process, and the curved-jet process are listed in Table 1. The values of δW were calculated with $\theta = 0.5$, which appears to be the optimum for all three processes. As is shown in Table 1 for the separation of uranium hexafluorides, the proposed new technique may give a value of δW about 500 times higher than the gaseous diffusion process and 100 times higher than the curved-jet process.

A complete analysis of the economics of the separations must include the energy inputs and the capital and maintenance costs. In the velocity-slip process some of the gas will be lost if the beam is passed through a skimmer and a collimator. In conventional nozzle systems the fraction passed by the skimmer may be as low as 1 percent (*5*). However, with the type of radial velocity selector we described above, it may not be necessary to collimate the beam, so nearly all the molecules in the jet will enter the velocity selector. For the gaseous diffusion process the separated streams must be compressed to the initial pressure of the feed to each stage. In both the velocity-slip and curved-jet processes the separated streams of UF_6 plus light gas must be recompressed at a much higher pressure ratio. However, the much higher values of $\alpha - 1$ and δW per stage in the velocity-slip process offset the advantage of a lower pressure ratio and gas flow in the gaseous diffusion process, even if the total molar flow is 100 times the UF_6 flow and the pressure ratio required is 100 times greater than for the gaseous diffusion process. Thus, the velocity-slip process is estimated to require a lower energy input per unit of separative work obtained than either the gaseous diffusion process or the curved-jet process.

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Energy Conservation Policies of the Federal Energy Office: Economic Demand Analysis

Abstract. *Forecasts for crude petroleum consumption are presented which take into account attempts by the Federal Energy Office to regulate residential and commercial thermostats. On the basis of an economic demand model, it is found that lowering all thermostats by 6°F would lower the projected residential and commercial petroleum demand in 1973 by 12 percent. This amount is compared to forecasts made by the Federal Energy Office and to the effect of a price increase of $0.10 per gallon for heating oil.*

An important component of federal energy conservation policy is the recommendation that residential and commercial thermostat settings be lowered by 6°F (*1*). This recommendation was based primarily on engineering heat loss studies and some tersely specified statistical analyses that tended to ignore fundamental economic considerations (*1, 2*). In this report we describe an economic demand study that includes ambient indoor temperature as an explanatory variable. We then use this model to evaluate the likely impact of the 6°F policy on the demand for crude oil for space heating regionally and nationally.

Space heating is estimated to account for 77 percent of the gas, oil, and coal consumed in the residential and commercial sector, and for 82 percent of the oil alone consumed in that sector (*3*). A cross-sectional econometric model was formulated for the demand for these fuels in 1971. The following variables were used (*4*):

X_1 = total oil, natural gas, and coal consumed in the residential and commercial sector by state for the year 1971. Measured in 10^{12} British thermal units (Btu's) (1 Btu = 1.06×10^3 joules).

X_2 = total population in each state in the year 1971. Measured in thousands of people.

$X_3 = (X_1/X_2) \times 10^3$ = total oil, natural gas, and coal consumed per capita in the residential and commercial sector by state in 1971. Measured in 10^6 Btu per capita.

X_4 = mean personal income per capita by state for 1971. Measured in dollars per capita.

X_5 = weighted average residential and commercial price of oil, natural gas, and coal for each state in 1971. The weights used are consumption levels by fuel for each state in 1971. Measured in dollars per 10^6 Btu.

X_6 = average residential and commercial price of electricity in each state in 1971. Measured in dollars per 100 kwh.

X_7 = weighted average total heating degree-days by state for 1971. The weights used are 1970 population levels for all Standard Metropolitan Statistical Areas and selected other cities with reporting weather stations. Measured in degree-days per year.

We examined several different functional specifications for these variables, including linear and double-log regressions of total fuel demand (X_1) on X_2 and X_4 through X_7, and linear and double-log regressions of fuel demand per capita (X_3) on X_4 through X_7. While the various specifications assured us of the robustness of the degree-day coefficient, we report only our preferred specification, based on ordinary least squares

$$\begin{aligned}\log X_3 = & -\underset{(.5908)}{1.0771} + \underset{(.1649)}{0.2671} \log X_4 - \\ & \underset{(.0858)}{0.2800} \log X_5 + \underset{(.1079)}{0.5039} \log X_6 + \\ & \underset{(.0405)}{0.4955} \log X_7;\ R^2 = 0.847 \qquad (1)\end{aligned}$$

with R^2 = the multiple coefficient of determination, 43 degrees of freedom, an F ratio of 59.696, and the estimated standard errors of the coefficients reported parenthetically. In this model the coefficients of fuel price (X_5), electricity price (X_6), and degree-days (X_7) can be shown to be significant in a symmetric 95 percent confidence test. The coefficients in this double-log regression may be interpreted as elasticities of demand (*5*). The most important coefficients for our conservation policy evaluation are the heating