On the Separation of Gas Mixtures in a Jet

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In 1954 Becker found that the components of a gas mixture could be separated by allowing a jet of the gas mixture to expand in a low-pressure chamber. The heavier molecules would concentrate near the center of the jet and the lighter ones near the edge. Thus the jet could be split into two streams, one containing more of the heavy component and one containing more of the light component. In this paper it will be shown that this effect can be predicted from the kinetic theory of gases and that the predicted magnitude of the effect agrees favorably with the experimental data of Becker.

A diagram of the jet-separation apparatus employing an axially symmetric jet is shown in Figure 1. The axis of symmetry has been designated as the *X* axis. The gas mixture to be separated flows through the feed tube and expands into the low-pressure chamber. Here the jet is separated into two streams by the conically shaped divider *D*. The center stream will contain more of the heavy component, and the side stream will contain more of the light component. These streams may then be recompressed and sent to other jet-separation units. A typical separation process would employ many such units, arranged in a cascade.

An alternative to the circular orifice indicated in Figure 1 would be one in the form of a long slit. Although the calculated separations are somewhat lower for this geometry, it would allow a much greater throughput per unit and could effect a substantial saving in equipment cost.

THEORY

The basic assumptions in this development are

1. The gas in the feed tube has a Maxwell-Boltzmann distribution of velocities superimposed on the mean bulk-gas velocity.
2. The jet is axially symmetric.
3. Collisions between molecules in the low-pressure region (collector) may be neglected.

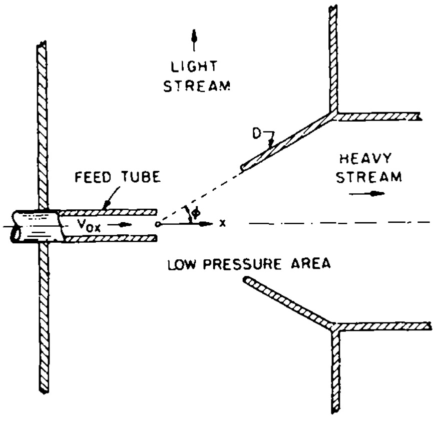

Fig. 1. Diagram of jet separation apparatus.

Allowing no collisions between molecules in the collector describes the limiting case of zero pressure in the collector. Actual pressures used by Becker were on the order of 0.01 mm. Hg. At this pressure and room temperature the mean free path of N_2 is 6 mm. As the dimensions of the apparatus were about 1 mm., it is seen that neglecting collisions is not unreasonable so long as the dimensions of the apparatus are kept small.

The Maxwell-Boltzmann velocity-distribution function for a pure gas in three dimensions may be written as

$$dn = n_o \left(\frac{m}{2\pi kT}\right)^{3/2} e^{-\frac{m}{2kT}[(V_x - V_{ox})^2 + V_y^2 + V_z^2]} \; dV_x \, dV_y \, dV_z \qquad (1)$$

Figure 2 shows that

$$V_y^2 + V_z^2 = V_*^2 = V_x^2 \tan^2\theta \qquad (2)$$

In the V_*, θ α coordinate system Equation (1) becomes

$$dn = n_o \left(\frac{m}{2\pi kT}\right)^{3/2} e^{-\frac{m}{2kT} V_{ox}^2 \sin^2\theta} \; e^{-\frac{m}{2kT}\left[\frac{V_*}{\sin\theta} - V_{ox}\cos\theta\right]^2} \qquad (3)$$
$$\frac{V_*^2}{\sin^2\theta} \, dV_* \, d\theta \, d\alpha$$

To obtain the angular distribution of molecules, Equation (3) is integrated over the range $0 \leq \alpha \leq 2\pi$ and $0 \leq V_* \leq \infty$. This gives the angular distribution of molecules:

$$dn = n_o e^{-\beta^2} \left[e^{\beta^2 \cos^2\theta} \left(\frac{1}{2} + \beta^2 \cos^2\theta \right) \operatorname{erfc}(-\beta\cos\theta) + \frac{\beta}{\sqrt{\pi}} \cos\theta \right] \sin\theta \, d\theta \qquad (4)$$

where the dimensionless parameter is defined as

$$\beta = V_{ox} \sqrt{\frac{m}{2kT}} \qquad (5)$$

and the complementary error function is

$$\operatorname{erfc}(t) = \frac{2}{\sqrt{\pi}} \int_t^\infty e^{-t_1^2} \, dt_1 \qquad (6)$$

The jet is now broken into two streams. All molecules moving at an angle greater than some particular value of θ, say ϕ, go into the side stream; the remainder go into the center stream. The number in the side stream is found by integrating (4) from $\theta = 180$ deg. to $\theta = \phi$. This yields

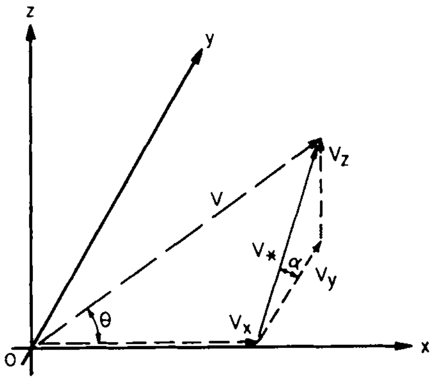

Fig. 2. Vector diagram.

$$f = \frac{1}{2} \cos\phi \, e^{-\beta^2 \sin^2\phi} \operatorname{erfc}(-\beta\cos\phi) + \frac{1}{2} \operatorname{erfc}(\beta) \qquad (7)$$

where $f = n/n_o$.

According to kinetic theory, the velocity distribution of one component of a gas mixture is unaffected by the presence of the other components. Thus for a multicomponent mixture Equation (7) will be valid for each component separately, with the appropriate value of β inserted for each component. This equation may be used to predict separations of multicomponent mixtures. For a two-component mixture the separation coefficient is defined by the ratio of light to heavy molecules in the light stream divided by the same ratio for the heavy stream:

$$A = \frac{\left[\dfrac{\%\ \text{light}}{\%\ \text{heavy}}\right]_{\text{light stream}}}{\left[\dfrac{\%\ \text{light}}{\%\ \text{heavy}}\right]_{\text{heavy stream}}} \qquad (8)$$

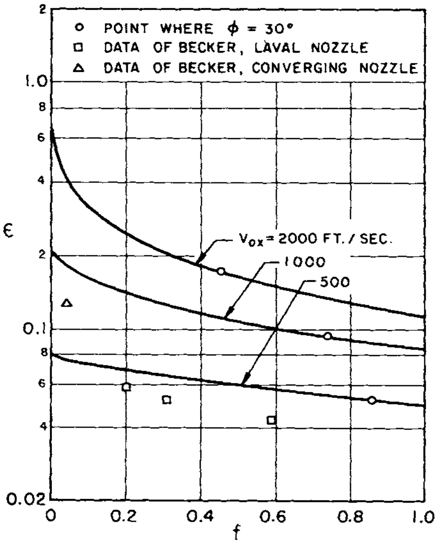

Fig. 3. Separation factors for mixtures of argon-36 and argon-40 at 300°K. For the converging nozzle, the calculated velocity and temperature was 600 ft./sec. at 226°K. This gives the same value of β as 693 ft./sec. at 300°K. For the De Laval nozzle the calculated velocity and temperature was 1,010 ft./sec. at 39°K. This gives the same value of β as 2,790 ft./sec. at 300 °K.

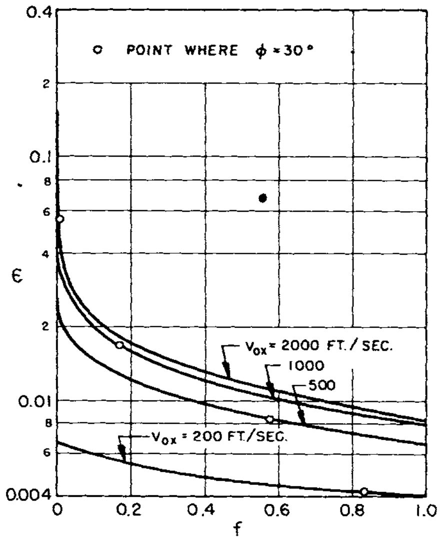

Fig. 4. Separation factors for mixtures of $U^{235}F_6$ and $U^{238}F_6$ at 300°K.

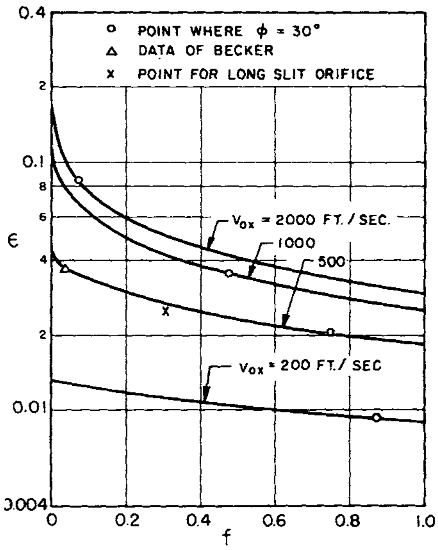

Fig. 5. Separation factors for mixtures of xenon-130 and xenon-134 at 300°K. The data of Becker were taken at a calculated velocity of 600 ft./sec. and a calculated temperature of 226°K. The point for the long slit was calculated for a velocity of 1,000 ft./sec., a temperature of 300°K., and an angle of 30 deg.

The enrichment factor is defined as $A - 1 = \epsilon$; thus $\epsilon \geq 0$. When ϵ is small, it is essentially independent of the feed composition. Substituting Equation (7) into this definition, one gets

$$\epsilon = \frac{\left[\cos\phi\, e^{-\beta_L{}^2 \sin^2\phi} \operatorname{erfc}(-\beta_L \cos\phi) + \operatorname{erfc}(\beta_L)\right] - \left[\cos\phi\, e^{-\beta_H{}^2 \sin^2\phi} \operatorname{erfc}(-\beta_H \cos\phi) + \operatorname{erfc}(\beta_H)\right]}{\left[\cos\phi\, e^{-\beta_H{}^2 \sin^2\phi} \operatorname{erfc}(-\beta_H \cos\phi) + \operatorname{erfc}(\beta_H)\right]\left[1 - \frac{1}{2}\cos\phi\, e^{-\beta_L{}^2 \sin^2\phi} \operatorname{erfc}(-\beta_L \cos\phi) - \frac{1}{2}\operatorname{erfc}(\beta_L)\right]} \tag{9}$$

For separations where the difference in β values is small, as in isotopic separations, Equation (9) involves a small difference between large numbers. When one uses digital computers capable of a large number of significant figures, this is no hindrance. However for hand calculation a more convenient form may be obtained by expanding f in a Taylor series and retaining only the first term. The result of this is

$$\epsilon = \frac{\left(\frac{\partial f}{\partial \beta}\right)_\phi (\beta_L - \beta_H)}{f(1 - f)} \tag{10}$$

Here f is found from Equation (7), and $(\partial f/\partial \beta)_\phi$ is

$$\left(\frac{\partial f}{\partial \beta}\right)_\phi = \frac{e^{-\beta^2}}{\sqrt{\pi}}\left[\cos^2\phi - 1 - \sqrt{\pi}\,\beta e^{\beta^2 \cos^2\phi} \operatorname{erfc}(-\beta\cos\phi)\sin^2\phi\cos\phi\right] \tag{11}$$

Equation (10) applies only if the relative difference between the values of β, and hence of f, for the two components is small.

TWO-DIMENSIONAL JET

For a two-dimensional jet, that is one where the orifice is a long narrow slit, expressions for f and ϵ may be derived in a manner similar to the above for the axially symmetric jet. In this case one of the integrals cannot be expressed in closed form. The result for f is

$$f = e^{-\beta^2}\left[1 - \frac{\phi}{\pi} + \int_\phi^\pi \frac{\beta}{\sqrt{\pi}} \cos\theta\, e^{\beta^2 \cos^2\theta} \operatorname{erfc}(-\beta\cos\theta)\, d\theta \right. \tag{12}$$

NUMERICAL RESULTS

Predicted enrichments have been calculated for some typical separations with the aid of an IBM-650 computer and are presented in Figures 3, 4, and 5. It was assumed here that the inlet temperature was 300°K.; inlet velocities were 200, 500 1,000 and 2,000 ft./sec. When the velocities are supersonic, the curves indicated will apply only if no shock waves are formed in the separation zone. Approximate sonic velocities at 300°K. for argon, xenon, and uranium hexafluoride are 900, 500 and 300 ft./sec. respectively. Becker's data (*1, 2*) on the separation of Ar^{36}–Ar^{40} mixtures are plotted in Figure 3 for comparison. In Becker's experiment the pressure in the inlet tube was 30 mm. Hg, and the pressure in the collector was 0.01 mm. Hg. The orifice dimensions and the approximate throughputs were given by Becker, and from these, inlet velocities and temperatures have been calculated as indicated on the figures. The DeLaval (converging-diverging) nozzle gave smaller enrichment factors than the converging nozzle, even though the calculated velocity is greater and the calculated temperature is less. This may be due to mixing caused by shock waves formed in the low-pressure region by the DeLaval nozzle. On each theoretical curve the point at which the collection angle ϕ is 30 deg. has been indicated by a circle; this gives an indication of the variation of geometry which the other parameters involved.

For the separation of the isotopes of uranium hexafluoride (Figure 4) it is seen that the predicted enrichment factor for jet separation is larger than 0.003, the value for the commonly used process of barrier effusion. This indicates that jet separation should be given serious consideration for separations of this type. One great advantage of jet separation as contrasted with the competing process of barrier effusion is the extreme simplicity of the apparatus used. This would allow construction and maintenance costs to be kept very low.

From Figure 5 it may be seen that

the values of ϵ for the long slit are lower than the values for the circular orifice. However the long slit may be more practical because of the reduced equipment cost; for example a circular orifice 1 mm. in diameter will pass only 700 g. uranium hexafluoride/hr. at a pressure of 30 mm. Hg and a velocity of 1,000 ft./sec., while a slit 1 mm. by 1 m. will pass 900 kg. under the same conditions. Thus the smaller enrichment may be justified by the greatly increased throughput made possible by the long slit.

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NOTATION

A = separation coefficient for a two-component mixture
f = fraction of feed entering the side stream
k = gas law constant
m = mass of molecule
n_o = total number of molecules
T = absolute temperature
V = molecular velocity
V_{ox} = bulk-gas velocity at orifice
V_x = molecular velocity component in the x direction
V_y = molecular velocity component in the y direction
V_z = molecular velocity component in the z direction
V_* = sum of x and y velocity components

Greek Letters

α = angle between V_* and V_y
β = dimensionless parameter, $\beta = V_{ox}\sqrt{\dfrac{m}{2kT}}$
β_H = value of β for the member of a two-component mixture having the higher molecular weight
β_L = value of β for the member of a two-component mixture having the lower molecular weight
ϵ = enrichment factor for a two-component mixture
θ = angle between V and the x axis
ϕ = angle at which jet is broken into two streams by the divider

LITERATURE CITED

1. Becker, Bier, and Buyhoff, *Z. Naturforsch.*, **10a**, 565 (1955).
2. Becker, *ibid.*, **12a**, 609 (1957).

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COMMUNICATIONS TO THE EDITOR

Dear Editor:

The policy of publishing translations of the better papers which appear in Japanese Journals is a good one. Since we have been studying heat transfer to Bingham plastics, the paper by Hirai which appeared recently in the *Journal* (*1*) was not unknown to us; however having the English translation was quite helpful. The problem of heat transfer to a Bingham plastic in laminar flow is important because of the extreme velocities required to attain turbulent flow, and consequently in many industrial applications the flow will be laminar. The same problem considered by Hirai has been solved independently by the writers (*2*) using an entirely different approach. Owing to differences in the mean temperature reported by Hirai in Figure 4 of the original paper and the mean temperatures that we calculated, we have studied the paper rather carefully. The following errors or omissions were noted.

Equations (1.18), (1.19), and (1.20) are improper because the functions $P_m(\xi)$ are not orthogonal on the interval $(a, 1)$. This can be seen by observing that $P_m(\xi)$ satisfies

$$\frac{d^2P_m}{d\xi^2} + \frac{1}{\xi}\frac{dP_m}{d\xi} + \beta^2_m\left[1 - \left(\frac{\xi-a}{1-a}\right)^2\right]P_m = 0 \qquad (1.10)$$

$$P_m(1) = 0$$

$$\frac{dP_m(0)}{dr} = 0 \text{ (not specified by Hirai)}$$

Equations (1.18), (1.19), and (1.20) are valid only if

$$\int_a^1 P_m(\xi)P_n(\xi)\xi\left[1 - \left(\frac{\xi-a}{1-a}\right)^2\right]d\xi$$

$$= 0 \text{ if } m = n$$

$$= \text{constant if } m = n$$

From Equation (1.10)

$$\int_a^1 P_m(\xi)P_n(\xi)\xi\left[1 - \left(\frac{\xi-a}{1-a}\right)^2 d\xi\right.$$

$$= \left(\frac{a}{\beta_m^2-\beta_n^2}\right)\left[\frac{dP_m(a)}{d\xi}P_n(a) - \frac{dP_n(a)}{d\xi}P_m(a)\right]$$

The right-hand side can be zero only if

$$P_m(a) = 0$$

or if

$$\frac{1}{P_m(a)}\frac{dP_m(a)}{d\xi} = \frac{1}{P_n(a)}\frac{dP_n(a)}{d\xi}$$

Referring to Figure 3 of Hirai's original paper, we see that neither of these conditions is satisfied by P_0, P_1, and P_2. Consequently the functions $P_m(\xi)$ are not orthogonal, and even if a set of A_m's existed as defined in Equation (1.18), they could not be determined by the use of Equations (1.19) and (1.20).

We wonder why Hirai has not mentioned the second solution of Equation (1.10). This solution cannot be thrown out because of its singularity at $\xi = 0$, since the solution is used only in the region from $\xi = a$ to $\xi = 1$. As a result of this omission Hirai has apparently satisfied only the condition

$$\theta(r_p) = \theta_p(r_p)$$

and assumed that the condition

$$\frac{\partial\theta(r_p)}{\partial r} = \frac{\partial\theta_p(r_p)}{\partial r}$$

is satisfied.

1. Hirai, E., *A.I.Ch.E. Journal*, **5**, 13 (1959).
2. Wissler, E. H., and R. S. Schechter, *Chem. Engr. Progr. Symposium Ser. No. 29*, **55**, 203 (1959).

Yours very truly,
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