$n(x)$ = number of collisions of a particle
$n(\Delta x)$ = number of collisions of a particle between x and $x + \Delta x$
n_o = relaxation number of collisions when $n(x) = n_o$, $q/m_p = 0.632\ \{(q/m_p)_\infty - (q/m_p)_o\}$
P = constant yield pressure
q = charge on a particle
$(q/m_p)_o$ = initial charge per unit mass of particles at $x = o$
$(q/m_p)_\infty$ = maximum possible charge per unit mass of particles at $x = \infty$
R = internal resistance
S = area of contact
Δt = duration of contact
ΔT = time required for deformation
u = air velocity
u^* = friction velocity
v = axial component of particle velocity
v_r = radial component of particle velocity
v_{ro} = radial component of particle velocity just before impact
V = deformed volume
V_c = contact potential difference
W = powder mass flow rate
W_R = work done by normal impulse
x = flow length from inlet ($x = o$)
z_o = gap between contact bodies

Greek Letters

β = constant in Equation (23)
γ = constant in Equation (23)
ϵ = dielectric constant of a particle
ϵ_0 = dielectric constant of air, 8.85×10^{-12} F/m
κ = constant in Equation (21)
λ = constant in Equation (21)
μ = viscosity of air
ρ = specific resistance of particle
ρ_a = density of air
ρ_p = density of particle
τ = relaxation time ($= RC$)

Superscript

— = mean value

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The Steady and Unsteady State Analysis of a Simple Gas Centrifuge

The operating theory of the gas centrifuge is developed under both steady and unsteady state behavior. It is shown that countercurrent flow patterns in a gas centrifuge enhance the possible separation. The effect of self-diffusion of the gaseous mixtures is shown to be significant with low molecular weight gases but of less significance with heavy gases such as uranium isotopes. The time required to develop steady state pressure and concentration profiles is in the order of minutes, even with no net flow to and from the machine.

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SCOPE

The gas centrifuge as a processing tool has not been widely studied. Its principal application to date has been in the field of isotope separation. The elementary theory of various gas centrifuge types has been reported in the literature to the point where a countercurrent centrifuge appears to be the most useful type. The analysis of flow patterns within such a separator and the evaluation of the optimal magnitude of flows has not been reported. Furthermore, the times required to achieve a fully developed flow pattern have not been considered, even though the flow characteristics and transient times become very important in the design of a gas centrifuge. These factors play an important part in the potential application of the gas centrifuge to the processing of low molecular gases, where the self-diffusion is large.

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CONCLUSIONS AND SIGNIFICANCE

From the analysis made of gas centrifuge flow characteristics, the following conclusions can be drawn regarding the production gas centrifuge with a feed and product stream.

1. To make the best use of the small fluxes, the device should have axial countercurrent flow. The effect would be to continually upset the establishment of an equilibrium profile, allowing the fullest realization of the flux by pressure diffusion. Also, separations greater than those in the simple centrifuge could be realized.

2. The magnitudes of the fluxes due to pressure diffusion are very small. Times in the order of minutes are required to approach equilibrium. In the production centrifuge, this means very large residence times will be required for a separation to take place. Although increasing the size of the centrifuge (increasing the length and/or diameter) rather than decreasing the throughput rate is also a means of increasing the residence time, certain mechanical limitations (due to vibrational problems) on the size of the centrifuge prohibit this from being a reasonable approach. Increasing the temperature, and hence increasing the diffusivity, correspondingly increases the radial flux rate and, to a certain degree, provides a means of decreasing the required residence time.

3. The magnitude of the centrifuge's rotation and the charge composition have very little effect on the time required to establish the steady state mole fraction profiles. Thus, these two parameters can be expected to have very little effect on the residence time required in the production centrifuge, but, of course, they have a substantial effect on the magnitude of the separation.

4. Although the unsteady state simple centrifuge was not analyzed with a stationary center pipe, the presence of the pipe would cause the times required for the composition profile to approach equilibrium to increase significantly. This is due to the slower (than normal) moving gas near the center pipe. More important, the presence of the center pipe and the possible turbulence created by it may rule out the possibility of applying the Zippe (1960) type of centrifuge to the separation of gases at normal pressures.

The change in composition of the atmosphere with altitude is a well-known effect of the earth's gravitational field. For example, at an altitude of 3 000 m, with a constant temperature of 5°C assumed, the ratio of the partial pressure of oxygen to that at ground level is calculated to be 0.661.

Since the ratio of the partial pressures is dependent on the molecular weight of the species, the ratios of the partial pressures of different species will decrease at different rates. This is illustrated by taking the ratio of the partial pressures of species i and j at a height h:

$$\frac{P_i(h)}{P_j(h)} = \frac{P_i(O)}{P_j(O)} \exp\left[\frac{-(MW_i - MW_j)gh}{RT}\right] \quad (1)$$

If the ratio of the partial pressure of oxygen to nitrogen is taken as 0.25 at ground level, then at 3 000 m, with a constant temperature of 5°C assumed, the ratio reduces to 0.237.

In 1919, Lindeman and Aston suggested that the gas separations caused by the earth's gravitational field could be duplicated by using a centrifugal field. That is, for separations in a centrifugal field, the gravitation energy term could be replaced by the equivalent centrifugal energy term ($\omega^2 r^2/2$).

The most practical centrifuge design involves the countercurrent flow of two concentric gas streams in opposite axial directions. As a result, the inner stream becomes richer in the lighter species near its exit, and the outer stream becomes richer in the heavier species near its exit. Since the opposing flow continually upsets the establishment of a radial equilibrium composition profile, separations greater than equilibrium can be achieved. Work on the development of the countercurrent centrifuge has been reported by Beams and Skarstrom (1939), Groth et al. (1958), Kistemaker et al. (1958), and Zippe (1960), among others.

In 1951, however, the first and only available theoretical analysis of gas centrifugation was presented by Cohen. The analysis was contained in a collection of theories made available for various processes which could be used in the large scale separation of uranium isotopes. Equations resulting from Cohen's analysis of the countercurrent centrifuge, although containing several approximations, were used to predict (with reasonable success) experimental results in future published experimental works.

Even though research on the subject began in the 1920s, still missing from the available literature is a general analysis of the gas centrifuge. An analysis in which a thorough study of the countercurrent gas centrifuge is made, detailing important operating variables, size restrictions, allowable flow rates, and any other fundamental limitations that may exist, is needed. This is the goal set for this work. Throughout the body of this analysis, important points are illustrated by using the gas pairs sulfur dioxide-nitrogen, sulfur dioxide-hydrogen, and UF_6 (235 and 238 isotopes). The sulfur dioxide-nitrogen combination was chosen reflecting the current interest in removing sulfur dioxide from stack gases. The sulfur dioxide-hydrogen combination was used as a variation to give a gas mixture with a very low density at standard conditions (at low sulfur dioxide concentrations), large diffusion coefficient, and large molecular weight difference. The gasified uranium isotopes (as UF_6), on the other hand, represent an opposite extreme having a relatively large density at standard conditions, low self-diffusion coefficient, and small molecular weight difference.

STEADY STATE SIMPLE CENTRIFUGE THEORY

A simple gas centrifuge shall be defined as a closed rotating cylinder containing an isothermal pure gas or gas mixture. The gas rotates uniformly at the same speed as the cylinder and has no radial or axial motions. To prevent radial flow, a pressure gradient is established in the radial direction balancing the centrifugal force per unit mass ($\omega^2 r$) created by the rotation of the gas. That is

$$\frac{1}{\rho}\frac{\partial P}{\partial r} = \omega^2 r \quad (2)$$

By using the ideal gas law and by assuming a pure gas with molecular weight MW, the pressure gradient equation can be written as

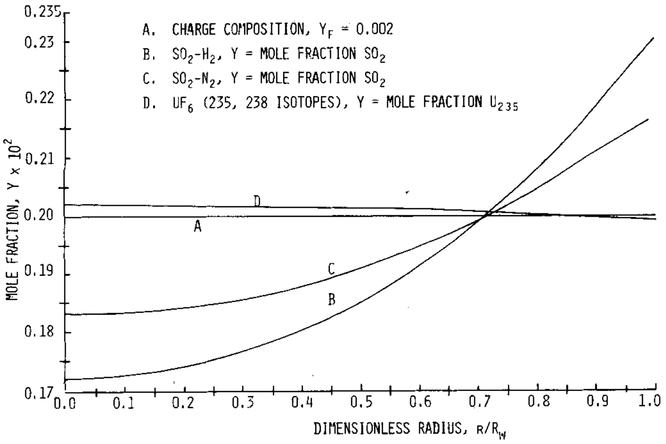

Fig. 1. Mole fraction profiles developed in a simple centrifuge with a peripheral speed of 150 m/s for the gas pairs: sulfur dioxide-hydrogen, sulfur dioxide-nitrogen, and UF₆ (235, 238 isotopes) at 20°C.

$$\frac{1}{P}\frac{\partial P}{\partial r} = \frac{\mathrm{MW}\omega^2 r}{RT} \tag{3}$$

which upon integration gives

$$P(\text{wall})/P(\text{axis}) = \exp\left(\frac{\mathrm{MW}\, s^2}{2RT}\right) \tag{4}$$

If a perfect mixture of gas species is placed in a simple gas centrifuge, a pressure gradient will be established for each specie according to the above equation. For example, for species i we have

$$\frac{P_i(r)}{P_i(O)} = \exp\left(\frac{\mathrm{MW}_i\omega^2 r^2}{2RT}\right) \tag{5}$$

The total pressure at any point r is then defined by

$$P(r) = \sum_{i=1}^{n} P_i(O) \exp\left(\frac{\mathrm{MW}_i\omega^2 r^2}{2RT}\right) \tag{6}$$

Owing to the different molecular weights of each species, the individual partial pressures increase at different rates proceeding from the axis to the wall. Thus, the ratio of species i and j at the axis will differ from their ratios at any other position:

$$\frac{P_i(r)}{P_j(r)} = \frac{P_i(O)}{P_j(O)} \exp\left[\frac{(\mathrm{MW}_i - \mathrm{MW}_j)\omega^2 r^2}{2RT}\right] \tag{7}$$

This, then, can be evaluated in terms of mole fraction as

$$y(r) = \frac{y(O)\exp\left(\frac{Ar^2}{2}\right)}{y(O)\left[\exp\left(\frac{Ar^2}{2}\right) - 1\right] + 1} \tag{8}$$

where

$$A = \frac{(\mathrm{MW}_1 - \mathrm{MW}_2)\omega^2}{RT} = \frac{\Delta\mathrm{MW}\omega^2}{RT} \tag{9}$$

A separtion factor for the simple gas centrifuge may be defined as the ratio of species 1 to 2 at the wall to the ratio of species 1 to 2 at the axis; that is

$$\alpha = \frac{y(R_w)}{1 - y(R_w)} \cdot \frac{1 - y(O)}{y(O)} \tag{10}$$

or, in terms of the mole fraction distribution given above

$$\alpha = \frac{1 - y(O)}{y(O)} \quad \text{times}$$

$$\frac{y(O)\exp\left(\frac{AR_w^2}{2}\right)}{y(O)\left[\exp\left(\frac{AR_w^2}{2}\right) - 1\right] + 1 - y(O)\exp\left(\frac{AR_w^2}{2}\right)} \tag{11}$$

which simplifies to

$$\alpha = \exp\left(\frac{AR_w^2}{2}\right) \tag{12}$$

An expression for $y(R_w)$ can now be written in terms of α; that is

$$y(R_w) = \frac{\alpha\, y(O)}{y(O)(\alpha - 1) + 1}$$

An important aspect to be noted in the expression for α is that it is a function of molecular weight difference, not the ratio of the molecular weights or the molecular weight difference divided by the sum of molecular weights as in diffusion processes. This gives the gas centrifuge an important advantage for separating heavy gases, hence the interest in the field of gas centrifugation for the separation of uranium isotopes ($\Delta\mathrm{MW} = 3$) when gasified as uranium hexafluoride ($\mathrm{MW} = 352$).

By combining these expressions with a material balance on the components charged to a centrifuge, Figure 1 was constructed showing the mole fraction profiles developed in a simple centrifuge for the following three gas pairs: sulfur dioxide-hydrogen, sulfur dioxide-nitrogen, and UF_6 (235, 238 isotopes) at 20°C. As expected (due to the large ΔMW), the sulfur dioxide-hydrogen mixture separates best as indicated by the separation factors. Also, owing to the radial pressure gradient, the point at which the mole fraction profile crosses the value of y_f does not occur at the equal area position ($s = 353.5$ ft/s). Instead, the crossing point is 358 ft/s for sulfur dioxide-nitrogen and 398 ft/s for UF_6 (235, 238 isotopes). The large molecular weight of the UF_6 (larger pressure gradient) pushes the crossing point closest to the periphery. Also worth noting is that the values of $y(O)$ and $y(R_w)$ are not equal distances on either side of y_f, and the mole fraction profile becomes flat as the axis approaches. This situation exists since as the axis is approached, the centrifugal force diminishes and the concentration decreases simply to make up for what has moved to the wall. On the other hand, near the periphery where the difference in the centrifugal forces acting on the heavy and light species is growing farther apart, the mole fraction profile increases quite rapidly.

Experimental gas centrifuge enrichment of the uranium isotopes has been accomplished by using centrifuges containing no internals and to a lesser extent by centrifuges containing a stationary pipe centered along the axis. Such work has been done by Zippe (1960) and Groth et al. (1958). The effect of the stationary center pipe on the pressure ratios and separation factors must be answered. An analysis of this effect was made by using the theory developed by Schlichting (1955). The results indicate that the turbulence produced by the stationary center pipe would reduce the pressure ratios and simple separation factors only slightly as long as the ratio of pipe to centrifuge bowl diameter is kept small (less than 0.1).

UNSTEADY STATE SIMPLE GAS CENTRIFUGE THEORY

The analysis of the unsteady state operation of the simple centrifuge will provide insight as to how fast the equilibrium mole fraction profile develops.

Consider a simple centrifuge that has been filled with a perfect binary gas mixture (total pressure = P_o) with

TABLE 1. OPERATING CONDITIONS AND DIFFUSIVITIES USED IN THE UNSTEADY STATE ANALYSIS OF THE GAS PAIRS SULFUR DIOXIDE-NITROGEN, SULFUR DIOXIDE-HYDROGEN AND UF_6 (235, 238 ISOTOPES)

Parameter	Gas pairs $SO\text{-}N_2$	 $SO_2\text{-}H_2$	 UF_6(235, 238 isotopes)
Charge pressure, atm	1.0	1.0	0.14
Charge composition, y	0.002(SO_2)	0.002(SO_2)	0.002(U_{235})
Temperatures, °C	20, 150	20, 150	27
Centrifuge radius, cm	10	10	10
Centrifuge, rev/min	10 000, 20 000	10 000, 20 000	20 000
Molecular weights	64, 28	64, 2	349, 352
Diffusivities, cm^2/s			
@ 20°C and 1 atm	0.1346	0.5282	—
@ 27°C and 0.14 atm	—	—	0.1136
@ 150°C and 1 atm	0.2590	0.9869	—

composition y_f (heavy species) and then quickly brought up to rotational speed.

The first thing to occur will be a rapid bulk radial flow of gas to the wall. When this flow essentially stops, diffusional flow will establish the equilibrium mole fraction profile. Starting from this point in time (when the gas is still of uniform composition y_f), the unsteady state problem will be solved.

By using diffusional flux equations, the equation of continuity for the heavy specie in the rotating cylinder is given by

$$\frac{\partial}{\partial t}\left(\frac{Py}{RT}\right)+\frac{1}{r}\frac{\partial}{\partial r}\left\{\frac{P}{RT}D_{12}\left[Ar^2y(1-y)-r\frac{\partial y}{\partial r}\right]\right\}=0$$

At this point it is worthwhile to note that the product PD_{12} can be taken as a constant. That is, in expressions for the diffusivity (binary mixture) resulting from kinetic theory and corresponding states arguments, at low pressures the diffusivity was found to vary inversely with pressure to the first power. Such expressions are given by, among others, Hirschfelder, Bird, and Spotz (1949). The boundary conditions are that there is no flux at the axis or the wall as expressed by the following equations:

$$\text{at} \quad r=0, \quad \frac{\partial y}{\partial r}=0$$

$$\text{at} \quad r=R_w, \quad \frac{\partial y}{\partial r}=Ar\,y(1-y)$$

The initial condition is that the mole fraction is uniform at the value y_f. As the equilibrium mole fraction profile develops, the pressure profile also changes owing to changing centrifugal forces coupled with the fact that conservation of mass must be maintained.

Owing to the coupling of P and y and hence nonlinearity of the partial differential equations which had to be solved, a numerical solution was sought. The numerical approach chosen was to solve the partial differential equations implicitly. That is, all partial derivatives were approximated with fourth-order finite difference formulas at the next increment in time, resulting in a set of simultaneous equations.

To obtain the numerical solution, a general FORTRAN program, UNSTEADY, aided by the scientific subroutine ONEDIAG to solve the simultaneous equations was written. The details of these programs may be found in Auvil's work (1974).

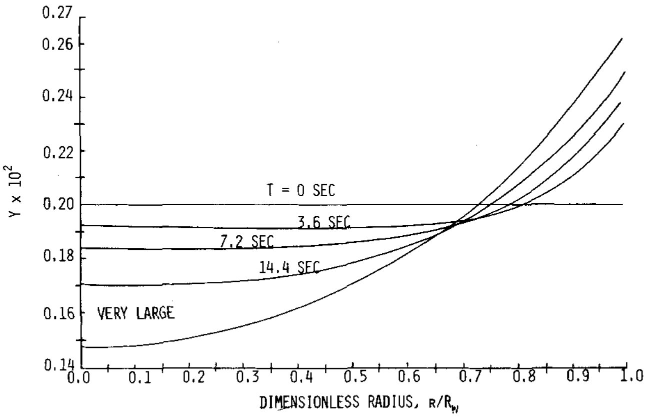

Fig. 2. Mole fraction profiles for sulfur dioxide, y, vs. dimensionless radius, r/R_w (R_w = 10 cm), at various times in the unsteady state operation of a simple centrifuge containing sulfur dioxide-hydrogen at 20°C; rev/min = 20 000.

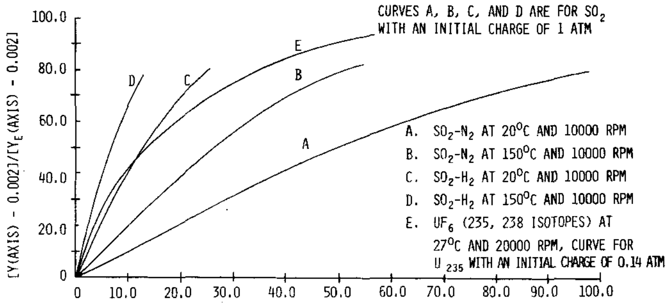

Fig. 3. [y(axis) — 0.002]/[y_e(axis) — 0.002] vs. time in the unsteady state simple centrifuge with a radius of 10 cm.

With the aid of the numerical solution, the separation of the two gas pairs, sulfur dioxide-nitrogen and sulfur dioxide-hydrogen, was analyzed in the unsteady state simple centrifuge at temperatures of 20° and 150°C with centrifuge revolutions per minute of 10 000 and 20 000. Also, a mixture of uranium isotopes (as UF_6) was analyzed at a temperature of 27°C and a centrifuge revolutions per minute of 20 000. Table 1 contains a complete tabulation of the operating conditions and diffusivities used. Values of the diffusivities were either calculated or found in the literature.

Figure 2 illustrates the changing mole fraction profiles with time for the gas pair sulfur dioxide-hydrogen at 20°C and a centrifuge rotation of 20 000 rev/min. Also included is the equilibrium mole fraction profile, that is, the profile established after a long time period. The curves in Figure 2 illustrate the expected fact that the profile develops much quicker near the wall than near the axis.

For a better comparison of the gas pairs, Figure 3 contains plots of $(y - y_f)/(y_e - y_f)$ at the axis vs. time at each of the temperatures investigated.

The results presented here are only for a centrifuge having a diameter of 20 cm. As shown earlier, varying the radius but maintaining the same peripheral speed does not affect the magnitude of the equilibrium separation. However, in the case of the unsteady state operation of the simple centrifuge, maintaining the same peripheral speed implies the same flux due to pressure diffusion at a given dimensionless radius r/R_w. Hence, since increasing the radius allows more mass to be contained in the centrifuge (all other operating conditions being equal), the time for the separation to occur must necessarily increase.

From the above information, three observations can be made regarding operating parameters which control the rate of separation in the simple gas centrifuge:

1. As illustrated with the gas pairs sulfur dioxide-nitrogen, increasing the rotational speed from 10 000 to 20 000 rev/min has very little effect on the time required for the mole fraction profile to develop. This is expected, since increasing the rotational speed increases the flux due to pressure diffusion (depends on the rotational speed squared), which correspondingly increases the magnitude of the separation as established earlier. The fact that increasing the rotational speed moves the gas contained in the centrifuge nearer the wall (steeper pressure profile) leaves less gas near the axis.

2. As illustrated in Figure 3, temperature has a substantial effect on the time required for the mole fraction profiles to develop. The effect of changing the temperature is of an indirect nature, since increasing the temperature from 20° to 150°C almost doubles the diffusivity (~1.9 times), which in turn almost halves the time for mass transfer to take place. However, increasing the temperature reduces the effective value of ΔMW (decreases flux due to pressure diffusion), hence decreasing the magnitude of the separation. Along these same lines, it should be noted that the ratio of the diffusivities times pressure for sulfur dioxide-nitrogen at 20°C to the corresponding value for UF_6 at 27°C is 8.3. This would appear to indicate that if it required 49 s in the case of sulfur dioxide-nitrogen for the dimensionless axis composition $(y - y_f)/(y_e - y_f)$ to move within 50% of its equilibrium value, it would require 407 s for the corresponding situation to occur for UF_6 owing to the much lower radial diffusion flux in the case of UF_6. However, the actual time calculated for UF_6 is only 13 s. The key to this apparent inconsistency is that not only was the charge pressure in the case of the UF_6 1/7 that of the sulfur dioxide-nitrogen (less total mass present), but at 20 000 rev/min the much greater molecular weight of UF_6 further reduces the ratio of the pressures at the axis to 0.024. Correcting for such a reduction in the mass present at the axis reduces the expected time from 407 to approximately 10 s.

3. Perhaps the most important fact is that the times required for mass transfer to take place in the gas centrifuge are relatively large when compared to times for mass transfer to take place in conventional gas separation. For example, consider the gas pair sulfur dioxide-nitrogen at 20°C, 10 000 rev/min, and $t = 0$ in the unsteady state simple centrifuge; the flux due to concentration diffusion is zero, and the flux due to pressure diffusion is 2.0×10^{-10} g moles/s/cm^2 with $y_f = 0.002$ and $r = 7.5$ cm. At future times, the net flux, of course, becomes smaller as the flux due to the concentration gradient negates a greater and greater part of the flux due to pressure diffusion. In a gas centrifuge with axial flow, the effect of the small fluxes is to require long residence times (low feed ratio) to achieve any degree of separation.

ACKNOWLEDGMENT

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NOTATION

A = $\Delta MW\omega^2/RT$
D = diffusivity
g = acceleration of gravity
h = height above sea level
MW = molecular weight
P = pressure
r = radius
R = universal gas constant
R_w = wall radius
s = peripheral speed
t = time
T = absolute temperature
y = mole fraction
$\alpha = \dfrac{y(R_w)}{1 - y(R_w)} \cdot \dfrac{1 - y(O)}{y(O)}$
ρ = density
ω = rotational speed

Subscripts

e = equilibrium
f = feed
i, j = components

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