A simplified formulation of the steady state in gaseous thermal diffusion columns

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Abstract. By introducing a flow pattern weighted average temperature as a reference temperature in a gaseous thermal diffusion column, a simplified formulation for the relevant steady quantities—the maximum separation factor and the optimum pressure—is obtained. This formulation describes within a few per cent all the cases which present in common separation practice and allows us to determine thermal diffusion factors from column steady measurements.

1. Introduction

According to the theory of Furry, Jones and Onsager (FJO; Furry *et al* 1939, Jones and Furry 1946) which describes the behaviour of a gaseous thermal diffusion column, the steady state is determined by two quantities: the maximum separation factor and the pressure at which it occurs, the so-called optimum pressure. These quantities depend in a complicated way on the geometrical characteristics of the column, the operation wall temperatures and the mixture's physical properties as well as their temperature dependences. These dependences are, in most cases, rather complex and numerical calculations are then needed.

Several attempts (Saxena and Raman 1962, Slieker 1965) have been made to obtain a simplified formulation of the column operation. In this respect, recent numerical results (Saviron *et al* 1971) indicate that the choice of a suitable mean temperature as reference temperature for the relevant physical properties could greatly reduce the standard FJO results.

That simplified formulation is required in many respects. In the design of separation installations, in particular, it would be desirable to have explicit expressions which allow the determination of the steady behaviour of a separation unit without recourse to numerical calculations. On the other hand, because of the relatively large separations, the column can be used advantageously to determine thermal diffusion factors of isotopic and non-isotopic gaseous mixtures. In this respect one of the difficulties reported in the literature (Rutherford 1970, Saviron *et al* 1971) is the involved way in which the thermal diffusion factor appears in the FJO formulation which makes it difficult to assign temperature to the measured values. Although numerical computations indicate that the maximum separation factor is proportional to the thermal diffusion factor evaluated at some mean temperature, it is important to have an explicit expression relating these two quantities.

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The purpose of this paper is to present simple and accurate expressions for the maximum separation factor and the optimum pressure. We shall start from the FJO standard equations and after introducing the adequate reference temperature we shall obtain a formulation in which the influence of the gas nature appears only through the gas properties evaluated at the reference temperature.

The obtained results allow a precise enough description of the column operation in common separation practice and can also be used in the evaluation of gaseous thermal diffusion factors from column measurements.

2. Theory

According to the FJO theory (Furry *et al* 1939, Jones and Furry 1946) the steady maximum separation factor, Q^*, and the corresponding optimum pressure, P^*, in a gaseous thermal diffusion column are given by

$$\ln Q^* = HL/2(K_c K_d)^{1/2} \qquad P^*/P = (K_d/K_c)^{1/4}. \tag{1}$$

Here P is the pressure, L is the length of the column and H, K_c and K_d are the so-called column constants defined by

$$H = \frac{2\pi}{Q_1^3} \int_{T_1}^{T_2} \frac{\alpha G(T)}{T} \, dT \qquad K_c = \frac{2\pi}{Q_1^7} \int_{T_1}^{T_2} \frac{\lambda}{\rho D} G^2(T) \, dT$$

$$K_d = \frac{2\pi}{Q_1} \int_{T_1}^{T_2} r^2 \rho D \lambda \, dT \tag{2}$$

where T_1 and T_2 are, respectively, the temperatures at the cold and hot walls, r is the radial coordinate; α is the thermal diffusion factor, λ is the conductivity coefficient, D is the ordinary diffusion coefficient, ρ is the density and $2\pi Q_1$ is the radial heat flux per unit length. This quantity and the temperature distribution in the column are given by the heat conduction equations

$$Q_1 \ln \frac{r_2}{r_1} = \int_{T_2}^{T_1} \lambda \, dT \qquad Q_1 \ln \frac{r}{r_1} = \int_{T}^{T_1} \lambda \, dT \tag{3}$$

where r_1 and r_2 are, respectively, the outer and inner cylinder radii. Finally, $G(T)$ is related to the mass circulation rate in the column and is given by the solution of the hydrodynamic equation

$$\frac{d}{dT} \frac{1}{r^2 \lambda} \frac{d}{dT} \frac{\eta}{\lambda} \frac{d}{dT} \frac{1}{r^2 \rho \lambda} \frac{dG}{dT} = -g \frac{d\rho}{dT} \tag{4}$$

with the boundary conditions

$$G(T_1) = G(T_2) = G'(T_1) = G'(T_2) = 0 \tag{5}$$

where η is the viscosity coefficient and G' stands for the temperature derivative of $G(T)$. As has been recently shown (Navarro *et al* 1982), the density composition dependence can usually be neglected in (4).

By taking the arithmetical mean temperature, $\bar{T} = (T_1 + T_2)/2$, as reference temperature for the gas physical properties in the preceding equations, (1) can be written as

$$\ln Q^* = \frac{0.418L}{r_1 - r_2}\left(\overline{\frac{\alpha\,\Delta T}{T}}\right)q \qquad \frac{P^*}{P} = \frac{24.54}{(r_1 - r_2)^{3/2}}\left(\overline{\frac{\eta DT}{\rho g \Delta T}}\right)^{1/2}p \qquad (6)$$

where the barred quantities are to be evaluated at $\bar{T}$, and p and q are correction factors which reduce to unity in the limit $T_2/T_1 \to 1$, $r_2/r_1 \to 1$. In the general case they depend on the ratios r_1/r_2, T_1/T_2 and on the temperature dependence of the physical properties relevant in (2)–(5). In terms of the corresponding correction factors to H, K_c and K_d—the so-called shape factors h, k_c and k_d, respectively—the quantities q and p are given by

$$q = h/(k_c k_d)^{1/2} \qquad p = (k_d/k_c)^{1/4}. \qquad (7)$$

These quantities can be determined by solving (1)–(5). In general, only numerical results can be obtained and for several molecular models extensive shape factor tables have been reported (McInteer and Reisfeld 1961, Saviron *et al* 1965, Greene *et al* 1966). We shall show below that an adequate choice of the reference temperature allows us to obtain simple expressions for q and p.

3. The reference temperature

We shall take as a reference temperature in a column the average mean temperature over the interval T_1 to T_2 defined by

$$T_s = \int_{T_1}^{T_2} TG(T)\,dT \bigg/ \int_{T_1}^{T_2} G(T)\,dT. \qquad (8)$$

This temperature will depend on the radii, the wall temperatures and the way in which the physical gas properties depend on temperature. An explicit expression for T_s may be found only for the case of small temperature differences in which these properties can be assumed to be constant. We shall first solve this case and then we shall show that the derived expression may be used as a good approximation in the general case.

For constant physical properties, (3)–(5) can be conveniently solved by introducing the new variable

$$t = 2[(T - T_1)/(T_2 - T_1)]\ln(r_2/r_1) \qquad (9)$$

and the auxiliary function

$$\gamma(t) = (\eta/r_1^4 \lambda^3 \rho^2 \beta g)G(T) \qquad (10)$$

where β is the thermal expansivity. In terms of these quantities, equations (3) yield for the temperature distribution

$$r = r_1 \exp(t/2) \qquad (11)$$

and (4) and (5) reduce to

$$\frac{d}{dt}e^{-t}\frac{d^2}{dt^2}e^{-t}\frac{d\gamma}{dt} = 1 \qquad \gamma(0) = \gamma'(0) = \gamma(t_2) = \gamma'(t_2) = 0 \qquad (12)$$

where t_2 stands for the value of t for $T = T_2$, i.e. $t_2 = 2 \ln(r_2/r_1)$. Denoting by t_s the value of t at $T = T_s$, we obtain from (8)

$$t_s = \int_0^{t_2} \gamma(t)t \, \mathrm{d}t \Big/ \int_0^{t_2} \gamma(t) \, \mathrm{d}t. \tag{13}$$

Introducing the solution of (12) in (13) we get for t_s after tedious but straightforward calculations the following expansion in powers of t_2:

$$t_s = \tfrac{1}{2}t_2(1 + \tfrac{3}{35}t_2 - (4 \times 35^2)^{-1}t_2^3 + \ldots). \tag{14}$$

This third-order approximation for t_s agrees to within 1% with the exact values given by (13), in all the range of r_1/r_2 of practical interest ($r_1/r_2 < 100$).

Substituting (14) in (9) we finally obtain for T_s

$$\frac{T_s}{\bar{T}} = 1 - \frac{3}{35} \frac{\Delta T}{\bar{T}} \left[\ln \frac{r_1}{r_2} - \frac{1}{3 \times 35} \left(\ln \frac{r_1}{r_2} \right)^3 \right]. \tag{15}$$

The validity of this equation for the case in which the temperature dependence of the fluid properties is taken into account can be checked against the numerical results derived from (8) for a particular gas model. We have used the Lennard-Jones (12-6) model following the numerical procedure described elsewhere (Saviron *et al* 1971). In table 1 are displayed the $T_s/\bar{T}$ values so obtained and the corresponding approximate ones given by (15) for two typical situations: a concentric type of column with $r_1/r_2 = 2$ and a hot wire type with $r_1/r_2 = 40$. The numerical values have been calculated for two far-apart reduced temperatures, $T_1^* = 0.8$ and $T_1^* = 30$, which cover well enough the current ranges of variation with temperature of the gas properties. As can be seen, the gas dependence of these values is rather low and they are in good agreement with the approximate ones. Therefore (15) can be confidently used to determine the temperature T_s.

4. Results and conclusions

By taking T_s as a reference temperature for the physical gas properties, (6) may be rewritten

$$\ln Q^* = \frac{0.418}{r_1 - r_2} L \left(\frac{\alpha \, \Delta T}{T} \right)_s q_s \tag{16}$$

$$\frac{P^*}{P} = \frac{24.54}{(r_1 - r_2)^{3/2}} \left(\frac{\eta D T}{\rho g \, \Delta T} \right)_s^{1/2} p_s \tag{17}$$

where the subscript indicates that the affected quantities are to be evaluated at T_s, and q_s and p_s are new correction factors which, respectively, tend to q and p and therefore to unity for $T_2/T_1 \to 1$, $r_2/r_1 \to 1$ because in this limit $T_s/\bar{T} \to 1$.

Explicit expressions for q_s and p_s may be easily derived only for constant physical properties. In such a case these quantities depend only on r_2/r_1 and we shall denote them by q_0 and p_0, respectively. As the expressions are complicated we shall omit them here for the sake of brevity. Numerical values of q_0 and p_0 are presented in table 2 instead. Intermediate values can be determined with an accuracy better than 1% by linear interpolation.

Table 1. Exact and approximate values for $T_s/\bar{T}$.

r_1/r_2	T_2/T_1	$T_s/\bar{T}$		
		$T_1^* = 0.8$	$T_1^* = 30$	Approx
2	1.5	0.976	0.975	0.976
	2	0.960	0.958	0.961
	2.5	0.948	0.944	0.949
	3	0.940	0.932	0.941
40	1.5	0.891	0.890	0.890
	2	0.820	0.816	0.817
	2.5	0.769	0.765	0.764
	3	0.732	0.726	0.725

Exact values of q_s and p_s can be obtained for a molecular model from (6), (7), (16) and (17) using the corresponding shape factor tables. In this respect we have used the inverse power model as well as the Lennard-Jones (12-6) model relying upon the shape factors in the literature.

Table 2. Values of q_s and p_s for constant gas properties.

r_1/r_2	q_0	p_0
1	1	1
2	0.968	1.025
4	0.887	1.092
6	0.825	1.144
8	0.781	1.184
10	0.746	1.217
15	0.683	1.276
20	0.634	1.319
30	0.591	1.379
40	0.555	1.417
50	0.530	1.447

Analysis of the values so obtained reveals that, to a good approximation, they fit the equations

$$q_s = (\bar{T}/T_s)^{1/2} q_0 \qquad p_s = (T_s/\bar{T})^{1/4} p_0. \qquad (18)$$

In figure 1 we plot the exact q_s and p_s values against the corresponding approximate ones given by (18) for the temperature ratios $T_2/T_1 = 2$ and 3. In the case of the inverse power model we have considered the ratios of the radii $r_1/r_2 = 5$ and 20, the dependences $\alpha = $ constant and $\alpha \propto T$ and the viscosity index $n = 0.6$ and $n = 1$. For the Lennard-Jones (12-6) model the displayed data correspond to $r_1/r_2 = 2$, 10 and 40 and $T_1^* = 0.8$, 1.5, 5 and 40. As can be seen (18) gives correct values of q_s and p_s within deviations smaller than 5%. At higher values for T_2/T_1, the deviations slightly increase, about 6% for $T_2/T_1 = 4$, and reduce to less than 1% for $T_2/T_1 \leqslant 1.5$.

From this result we can conclude that when the mean temperature given by (15) is adopted as a reference temperature, the maximum separation factor and the optimum
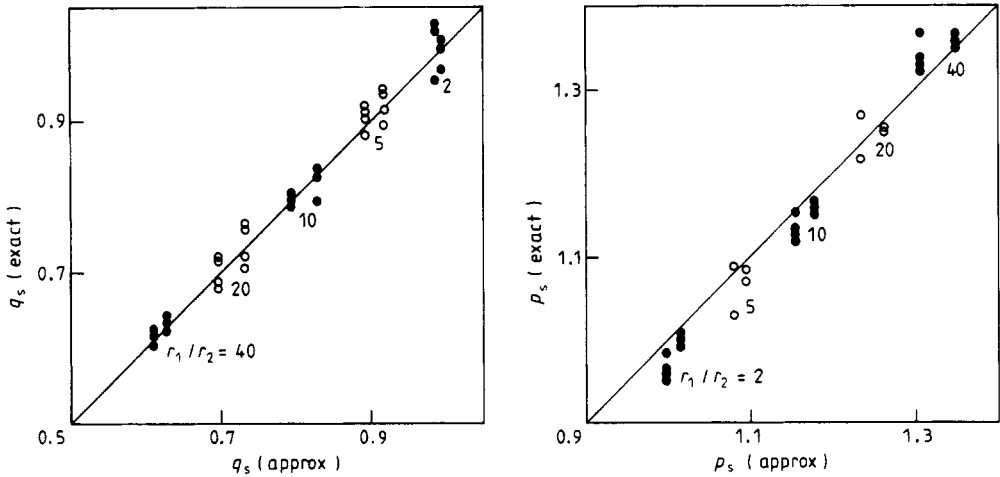
Figure 1. Correlation between exact and approximate values of q_s and p_s. ○ Inverse power model. ● Lennard-Jones (12-6) model.

pressure are accurately given in all the cases in common practice by the simple equations (16), (17) and (18).

This conclusion is particularly relevant when the column is used to determine thermal diffusion factors. In fact the obtained equation for the maximum separation factor establishes a proportionality relationship, accurate within 1% for $T_2/T_1 \leqslant 1.5$, between the logarithm of this quantity and the thermal diffusion factor. The fact that the proportionality constant appears to be independent of the gas mixture agrees with previous numerical results (Saviron *et al* 1971) and gives additional theoretical support to the method of relative measurements of thermal diffusion factors (Saviron *et al* 1969, 1971). We can mention here that according to recent experimental work (Trengove *et al* 1981) this method can be confidently used to determine thermal diffusion factors.

In the simplified formulation of Slieker (1965) the fluid properties are assumed to be constant when evaluated at the mean temperature of Fleischmann and Jensen (1942). Therefore the maximum separation factor and the optimum pressure are given by (16) and (17) if we use for T_s this reference temperature and, respectively, take for q_s and p_s, q_0 and p_0. To check the validity of this formulation we have calculated from (6), (7), (16) and (17) exact q_s and p_s values for the Lennard-Jones (12-6) model with $T_2/T_1 = 3$, $T_1^* = 5$ and $r_1/r_2 = 5$, 10 and 40. Deviations between these values and the corresponding ones for q_0 and p_0 as high as 50% are obtained, indicating that the Slieker formulation can be used only for rough estimations.

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