Publicado en
Zeitschrift für Naturforschung.
Band 11a, Heft 4, 1956

COMISION NACIONAL DE LA ENERGIA ATOMICA

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BUENOS AIRES
1956

Sonderabdruck aus der ZEITSCHRIFT FÜR NATURFORSCHUNG Band 11 a. Heft 4, 1956
Verlag der Zeitschrift für Naturforschung, Tübingen

A Countercurrent Gaseous Diffusion Process; Theory of the Separation at Equilibrium

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(Z. Naturforschg. 11 a, 284—285 [1956]; eingegangen am 21. Oktober 1955)

An experimental device for the obtention of a gaseous diffusion countercurrent process is proposed. The theory of the separation at equilibrium for a binary isotopic mixture is developed on the basis of the transport up the tube, the total and the desired molecules transverse transport. The solution is given for any value of the relative abundances.

Fig. 1 shows an experimental device in which a countercurrent gaseous diffusion process can be obtained. P is a pump, and M ist a porous barrier. The length h determines the ratio of pressures ξ for each value of the velocity v_h $(z=h)$. The notation used follows.

N : molar fraction of light molecules,
$1-N$: molar fraction of heavy molecules,
N_h : molar fraction of light molecules at $z=h$,
N_0 : molar fraction of light molecules at $z=0$,
δ : density of the gas,
p : pressure of the gas,
v : velocity of the gas,
w : gap-space of the chambers,
σ : cross-section of the chambers,
c_1 : mean thermal velocity of light molecules,
c_2 : mean thermal velocity of heavy molecules,
$c=N c_1+(1-N)\, c_2$: mean thermal velocity of the molecules of a gas of light molar fraction N,
D : self-diffusion coefficient,
$\varkappa$: specific free area of the porous wall,
h : total length of the countercurrent, and
∇ : $\partial/\partial z$.

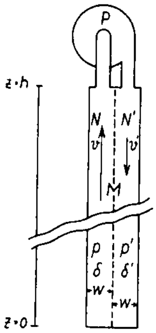

Fig. 1. Device for countercurrent gaseous diffusion.

We shall study the case of an apparatus connected to the pump at one end and the other end closed. For the sake of simplicity, we shall consider a barrier of unit y-dimension, x being the width dimension.

Transport up the Tube

At equilibrium, the transport up the tube of the desired molecule, — let us assume the lighter one of a binary mixture —, must be zero. In fact, there are two opposed transports, one due to the countercurrent (φ_v), the other to ordinary diffusion (φ_D). They are respectively

$$\varphi_v=\sigma(\delta\, v\, N+\delta'\, v'\, N')\,, \tag{1}$$

$$\varphi_D=\sigma(\delta\, D \nabla N+\delta'\, D' \nabla N')\,. \tag{2}$$

Now, taking into account that

$$\delta\, v+\delta'\, v'=0\,; \quad \delta\, p'=\delta'\, p\,; \quad \delta D=\delta' D'\,, \tag{3}$$

and that in all places $\nabla N=\nabla N'$, we arrive at

$$v\, \Delta N-2\, D \nabla N=0 \tag{4}$$

where

$$\Delta N=N-N'\,. \tag{5}$$

Total Transverse Transport

To a far greater extent than is the case with the conventional countercurrent, the gaseous diffusion countercurrent is sensitive to its length. It must be remarked that the total transverse transport is zero in a conventional countercurrent. For a gaseous diffusion one it is not. On the contrary, it is even greater than the desired molecules transport itself. Consequently, the velocity of each current is a function of z. Let us consider the velocity of the gas in the low-pressure chamber. The variation ∇v is due to the total transverse flow of material per unit length of column per second. Then, if there is no viscous flow through the porous barrier,

$$w\, \delta \nabla v=\varkappa\, c\,(\delta'-\delta) \tag{6}$$

where $\varkappa$ is a dimensionless quantity that characterizes the porous barrier.

Consequently, since $v_{z=0}=0$, the velocity results

$$v=(\varkappa c/w)(\xi-1)\,z=\nabla v\cdot z \tag{7}$$

where $\xi=p'/p$.

Making $z=h$, one obtains, obviously,

$$v_h=(\varkappa c/w)(\xi-1)\,h \tag{8}$$

which gives the relation between ξ, h and v_h.

Transverse Transport of the Desired Molecules

The effects that tend to change the value of ΔN must add to zero. There are two such effects, the change in concentration due to the countercurrent and the gaseous diffusion itself. Let ψ be the transverse transport per unit area of unit length of porous wall per second. The change in concentration per unit length per second due to the countercurrent is, for the low pressure chamber,

$$\delta w[\nabla(vN)-D\nabla^2 N]\,. \tag{9}$$

We have then

$$\psi-\delta w[\nabla(vN)-D\nabla^2 N]=0\,. \tag{10}$$

The transport ψ is the consequence of two transports, ψ_+ due to diffusion from high pressure chamber, and ψ_- due to diffusion from low pressure chamber. They are respectively

$$\psi_+=\varkappa c_1\,\delta' N'\,,\quad \psi_-=\varkappa c_1\,\delta N\,. \qquad (11),\,(12)$$

Substitution of (11) and (12) in (10) then gives, neglecting $D\nabla^2 N$,

$$(\varkappa c_1/w)\,[N(\xi-1)-\xi\,\Delta N]=v\,\nabla N+N\,\nabla v\,. \tag{13}$$

Consequences of the Transport Equations

Equation (4) may now be solved for ΔN, and substitution of this value in (13) then gives

$$N\frac{c_1-c}{c}\nabla v=\left[v+\frac{2\varkappa c_1}{w\,v}\xi D\right]\nabla N \tag{14}$$

where, for the sake of simplicity, we have supposed the term in $D\nabla^2 N$ to be null, in so far as it actually is much smaller than $\nabla(vN)$.

Now, since from the definition of c

$$c_1-c=(1-N)\,\Delta c$$

where $\Delta c=c_1-c_2$, equation (14) becomes

$$N(1-N)\frac{\Delta c}{c}\nabla v=\left[v+\frac{2\varkappa c_1}{w\,v}\xi D\right]\nabla N \tag{15}$$

which leads to

$$\mathrm{d}\ln\frac{N}{1-N}=\frac{\Delta c}{c}\,\frac{v\,\nabla v}{\Omega}\,\mathrm{d}z \tag{16}$$

where

$$\Omega=v^2+2(\varkappa c_1/w)\,\xi D\,. \tag{17}$$

The solution of equation (16) is

$$\ln\frac{N}{1-N}=\frac{\Delta c}{2c}\ln\Omega+C \tag{18}$$

which leads to the separation factor

$$q=\frac{N_h(1-N_0)}{N_0(1-N_h)}=\left[1+\frac{v_h{}^2}{B}\right]^{\Delta c/2c} \tag{19}$$

where

$$B=2(\varkappa c_1/w)\,\xi D\ll v_h\,. \tag{20}$$

Equation (19) can also be given as a function of ξ and h, $(c_1\sim c)$, as follows,

$$q=\left[1+\frac{\varkappa c(\xi-1)^2}{2w\xi D}h^2\right]^{\Delta c/2c}. \tag{21}$$

If one applies equation (19) to isotopic mixtures, the following approximation usually stands

$$\Delta c/c=\ln\alpha \tag{22}$$

where α is the separation factor of a unit process. The equivalent number of unit processes is then, from (19)

$$n=\tfrac{1}{2}\ln[1+\varkappa c(\xi-1)^2 h^2/2w\xi D]\,. \tag{23}$$

Interpretation of Results

For the interpretation of the results, we shall now consider the separation of a heavy isotopic mixture treated through a barrier of $\varkappa\sim10^{-3}$, in an apparatus of $h\sim1$ m and $w\sim1$ cm, with a ratio of pressures $\xi\sim10$. The thermal velocities are, of course, ca. 10^4 cm/s. In such a case, the value of n results ca. 7. It must be remarked that the concept of height equivalent to a theoretical unit looses its sense. In fact, further increasings of h increase slighter and slighter the separation factor. In the practical case discussed, for example, n takes the value of 8 for $h=3$ m, 9 for 10 m, and only 11 for 100 m!

A laboratory apparatus could be made 3 m-length. In such a case, when applied to, let us say, ^{36}A-^{40}A, for which $\alpha=1.05$, the total separation factor attained would be 1.48.

It obviously is possible to adjust the apparatus for continuous production. In this case, the separation factor would be smaller.