Darin ist

$$u_{\mathrm{f}} = \omega_{\mathrm{f}}\, \vartheta_{\mathrm{f}}/\Omega\,, \quad u_{\mathrm{d}} = \omega_{\mathrm{d}}\, \vartheta_{\mathrm{d}}/\Omega\,. \tag{35}$$

Nimmt man an, daß fokussierender und defokussierender Sektor von gleicher azimutaler Länge sind, also $\vartheta_{\mathrm{f}} = \vartheta_{\mathrm{d}}$, so spezialisiert sich (34) zu

$$\varrho = \frac{[1/(1+\sigma_{\mathrm{f}}) - 1/(1+\sigma_{\mathrm{d}})]\,(\sigma_{\mathrm{f}} - \sigma_{\mathrm{d}})}{\sqrt{(1+\sigma_{\mathrm{f}})\,(1+\sigma_{\mathrm{d}})}} \cdot \left[u_{\mathrm{f}} \operatorname{ctg} \frac{u_{\mathrm{d}}}{2} + u_{\mathrm{d}} \operatorname{ctg} \frac{u_{\mathrm{f}}}{2}\right]^{-1} + \frac{1}{2}\,[1/(1+\sigma_{\mathrm{f}}) + 1/(1+\sigma_{\mathrm{d}})]\,. \tag{36}$$

Dies stimmt mit der Gl. (8) der Arbeit von Courant et al.[1] bis auf das Vorzeichen des ersten Terms von (36) überein, eine Diskrepanz, welche im Prinzip davon herrührt, daß die Autoren[1] das Vorzeichen von $(-1)^{3/2} = \pm i$ nicht diskutieren und im Endresultat verkehrt angeben, wie wenn mit positivem p/P die Bahnlänge abnähme. In (34) und (36) ist die Wurzel positiv zu nehmen. Beachtet man, daß im defokussierenden Sektor $\sigma_{\mathrm{d}} < 0$, also wegen (32) und (35) u_{d}/i reell, so läßt sich (34) schreiben

$$\varrho = \frac{[1/(1+\sigma_{\mathrm{f}}) - 1/(1+\sigma_{\mathrm{d}})]\,(\sigma_{\mathrm{f}} - \sigma_{\mathrm{d}})}{\sqrt{(1+\sigma_{\mathrm{f}})\,(-1-\sigma_{\mathrm{d}})}}\, \frac{2\,\vartheta_{\mathrm{f}}\,\vartheta_{\mathrm{d}}}{\vartheta_{\mathrm{f}} + \vartheta_{\mathrm{d}}} \cdot \left[u_{\mathrm{f}}\,\vartheta_{\mathrm{d}}\, \mathfrak{Cotg}\left(\frac{u_{\mathrm{d}}}{2\,i}\right) - \left(\frac{u_{\mathrm{d}}}{i}\right) \vartheta_{\mathrm{f}} \operatorname{ctg} \frac{u_{\mathrm{f}}}{2}\right]^{-1} + [\vartheta_{\mathrm{f}}/(1+\sigma_{\mathrm{f}}) + \vartheta_{\mathrm{d}}/(1+\sigma_{\mathrm{d}})]/(\vartheta_{\mathrm{f}} + \vartheta_{\mathrm{d}})\,. \tag{37}$$

Für $\vartheta_{\mathrm{f}} = \vartheta_{\mathrm{d}}$, $\sigma_{\mathrm{f}} = -\sigma_{\mathrm{d}} = \sigma \gg 1$ und die Mitte des Stabilitätsbereiches $u_{\mathrm{f}} = u_{\mathrm{d}}/i = \pi/2$ wird aus (37)

$$\varrho = 8/\pi\, [\mathfrak{Cotg}\,(\pi/4) - \operatorname{cotg}\,(\pi/4)]\, \sigma = 4{,}85/\sigma\,. \tag{38}$$

Herrn Dr. W. Humbach (Erlangen) danke ich verbindlichst für den Hinweis auf Literatur[4].

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[4] Weitere Literatur: R. Wideröe, Schweizer Arch. angew. Wiss. Techn. **13**, 225, 299 [1947]. — N. M. Blachmann, Rev. Sci. Instr. **22**, 569 [1951]. — K. J. Le Couteur, Proc. Phys. Soc. B **64**, 1073 [1951]. — M. Hoyaux, Atomics, Lond. **2**, 310 [1951]. — D. C. de Packh, Phys. Rev. **86**, 433 [1952]. — M. Hoyaux, Atomics, Lond. **3**, 33 [1952]. — C. Simone, Elektr. Obz. **41**, 362 [1952]. — E. R. Caianiello, Nuovo Cim. **10**, 581 [1953]. — E. R. Caianiello u. A. Turrin, Nuovo Cim. **10**, 594 [1953]. — M. Sands u. B. Touschek, Nuovo Cim. **10**, 604 [1953]. — T. Kitagaki, Phys. Rev. **89**, 1161 [1953]. — E. D. Courant, M. S. Livingstone, H. S. Snyder u. J. P. Blewett, Phys. Rev. **91**, 202 [1953]. — E. D. Courant, Phys. Rev. **91**, 456 [1953]. — Stig Lundquist, Phys. Rev. **91**, 981 [1953]. — L. Good, Phys. Rev. **92**, 538 [1953]. — B. C. Carlson, Phys. Rev. **92**, 839 [1953]. — B. Cork u. E. Zajec, Phys. Rev. **92**, 853 [1953]. — J. S. Bell, Nature, Lond. **171**, 167 [1953]. — J. B. Adams, M. G. N. Hine u. J. C. Lawson, Nature, Lond. **171**, 926 [1953]. — M. S. Livingstone, Nucleonics **11**, 12 [1953]. — Chihiro Hayashi, J. Appl. Phys. **24**, 344 [1953]. — L. A. Pipes, J. Appl. Phys. **24**, 902 [1953]. — M. H. Blewett, Rev. Sci. Instr. **24**, 725 [1953]. — I. S. Blumenthal, Amer. J. Phys. **21**, 164 [1953]. — W. Paul u. H. Steinwedel, Z. Naturforschg. **8a**, 448 [1953]. — E. Bodenstedt, Z. Naturforschg. **8a**, 502 [1953]. — J. Seiden, C. R. Acad. Sci. (Paris) **236**, 1145 [1953]. — G. Lüders, Phys. Verh. **4**, 24, 149 [1953]. — Ch. Schmelzer, Physikertagung Innsbruck, Physik-Verlag, Mosbach/Baden, 1954, S. 16 bis 36.

The Chemical Separation of Isotopes as a Problem of Rectification

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In this paper the best operation of a column for separating isotopes by chemical exchange reactions and the best cascading of a plant are discussed.

The results are presented in a form directly utilizable by the plant-designer.

The method for separating the stable isotopes by means of chemical exchange reactions developed by Urey and collaborators permits the processing of large amounts of material in plants very similar to those used in the fractional distillation technique, thus making possible the production of separated or enriched isotopes on an industrial scale.

This method has been successfully applied to the separation of ^{13}C, ^{15}N, ^{18}O, ^{34}S, which are rather important as biological tracers, and of deuterium, which is very important in nuclear technology. Having in mind production on an industrial scale, great care has to be applied to proper planning of the flow sheet of the plant, remembering that the main goals are cheap product, rugged plants, and troublefree operation. The first problem is to minimize plant and operation costs, the second one to simplify the plant and to reduce component parts.

Only the first problem will be considered in our example. The second is the task of the designer who has to meet many particular situations which are not easily classificable.

In the book of Cohen[1] one could find many good suggestions to solve the problem. Unfortunately, however, his treatment of the matter is so abstract and often fragmentary, that the experimental physicist in whose hands lies the technical accomplishment of the problem is tempted to start the study of the question from the beginning in a form directly usable for his purpose, as was shown for instance in the interesting treatise on the subject by Becker and Bier[2].

The particular value function used by these authors leads to columns which are quite near to the minimum equilibrium time, but which are not so good in respect to plant and operation costs. We have laid stress on this latter point showing the general situation instead of developing a value function of limited validity of our own.

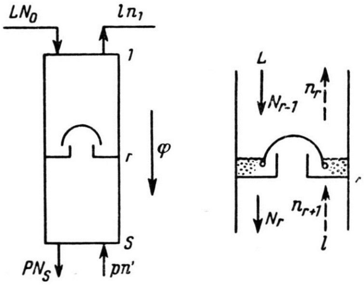

Fig. 1. Scheme for the tower and plate operation.

The Best Operation of a Column

Let us suppose to have a plate column operated as shown in fig. 1. A stream of feed material L of concentration N_0 is introduced at the top of the column, and a stream P of enriched material is withdrawn from the bottom, where a balancing feed of equal amount P, but of different concentration, is introduced. Thus the depleted stream is equal to L.

The plates are numbered starting from the feed. N_r is the concentration of the desired isotope in the liquid phase and plate r. n_r is the concentration of the desired isotope in the gas phase coming from plate r. L, l are moles/h resp. of liquid and gas flowing along the column. φ is the net transport of desired isotope along the column, i. e. the isotope withdrawn at the bottom as difference between that carried by the outcoming and that carried by the incoming streams in moles/h.

For the conservation of matter in the plate r we have

$$L N_{r-1} + l n_{r+1} = L N_r + l n_r \,, \tag{1}$$

and for the definition of φ

$$L N_r = l n_{r+1} + \varphi . \tag{2}$$

We shall suppose $N_r \ll 1$, so that the equilibrium relation

$$K = \frac{N_r}{1-N_r} \Big/ \frac{n_r}{1-n_r}$$

is simplified to

$$K = N_r / n_r \,. \tag{3}$$

K can have any value $K > 1$.

By combining (1), (2), (3) and $L = l$ we eliminate n_r, n_{r+1} and obtain with

$$b = 1/K, \ c = \varphi / L : \tag{4}$$

$$N_{r-1} = b N_r + c \quad \text{or} \quad N(r-1) = b N(r) + c \,. \tag{5}$$

This is a canonical form for plate columns.

The finite difference equation (5) is easily solved and gives, if S is the number of plates of the column and $N(0) \equiv N_0$,

$$N_S = \left(N_0 + \frac{c}{b-1} \right) b^{-S} - \frac{c}{b-1} \,. \tag{6}$$

This can be rewritten in a physically more significant form:

$$S = \ln \left[\left(\frac{N_S}{N_0} + \frac{1}{N_0} \frac{c}{b-1} \right) \Big/ \left(1 + \frac{1}{N_0} \frac{c}{b-1} \right) \right] \Big/ \ln \frac{1}{b} \,. \tag{6'}$$

The expression $- c/N_0 (b-1)$ is the isotope recovery of the column of S plates (defined as the ratio between the *isotope* withdrawn at the bottom φ and that introduced with the feed, LN_0), divided by the recovery of a column with $S = \infty$.

In fact (4):

$$1 + \frac{1}{N_0} \frac{c}{b-1} = 1 + \frac{\varphi}{L N_0} \frac{K}{1-K} \,, \tag{7}$$

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[1] K. Cohen, The theory of isotope separation as applied to the large scale production of ^{235}U, McGraw Hill, New York 1951.

[2] E. W. Becker u. K. Bier, Z. Naturforschg. **7a,** 651 [1952].

and if $S=\infty$ must be (6′):

$$1+\frac{1}{N_0}\frac{c}{b-1}=0 \text{ so that } \frac{\varphi}{LN_0}=\frac{K-1}{K}. \quad (7')$$

Thus we call $-c/N_0(b-1)=\varrho$ the relative recovery of the column and rewrite the (6′) in the form:

$$S=\ln\left[\left(\frac{N_S}{N_0}-\varrho\right)\Big/(1-\varrho)\right]\Big/\ln K. \quad (6'')$$

If we operate a column with no isotope withdrawal, its enrichment at equilibrium will be ($\varrho=0$):

$$\frac{\bar{N}_S}{N_0}=K^S \text{ or } S=\ln\frac{\bar{N}_S}{N_0}\Big/\ln K.$$

The expression $(N_S/N_0-\varrho)/(1-\varrho)$ in (6″) then coincides with the enrichment of the same column of S plates at no withdrawal.

We shall call $N_S/N_0=a$, $\bar{N}_S/N_0=A$.

Thus if S is constant:

$$\frac{a(\varrho)-\varrho}{1-\varrho}=a(0)=A \text{ and } a(\varrho)=A-\varrho(A-1). \quad (7'')$$

The product SL can be called the total throughput of the column; for a packed column hydraulically operated in a fixed way SL is proportional to its volume, in a plate column it gives the total section of plates; it is at any rate a size factor for the plant. ϱL is the product output; it is particularly significant for the function:

$$\frac{SL}{\varrho L}=\frac{S}{\varrho}, \quad (8)$$

which gives the volume of plant per unit of product.

In fig. 2 graphs of $SL/\varrho L$ vs. ϱ, taking a as parameter, are reported.

As is shown by fig. 2, the curves (8) show a marked minimum. For the columns operated at the left of the minimum, the high specific volume is due to the low isotope withdrawal, for the columns at the right to heavy loss in enrichment, which asks for very high and voluminous columns.

From the point of view of economy, a column should never be operated at the left of said minimum, for, with the same specific plant volume or throughput, the column can be operated at the right of the minimum, thus permitting a higher isotope recovery, which is always an advantage.

If the cost of the plant is the major item, so that operating costs can be neglected, or if the operating costs are proportional to the total throughput, and feed (or the possibility of its transformation at the bottom) is in unlimited supply, the minimum of the S/ϱ gives the best operating condition for the column.

In general, an optimal balance must be found between plant costs, roughly proportional to the total throughput, SL, and the operation costs, given by the difference in value between L and l, and by the cost of transformation of L at the base of the column.

To minimize the expression $\alpha S/\varrho+\beta L/\varrho$, where α and β are the cost factors for the enrichment part, and resp. for the feed and its transformation, can be, as usual, a first criterion.

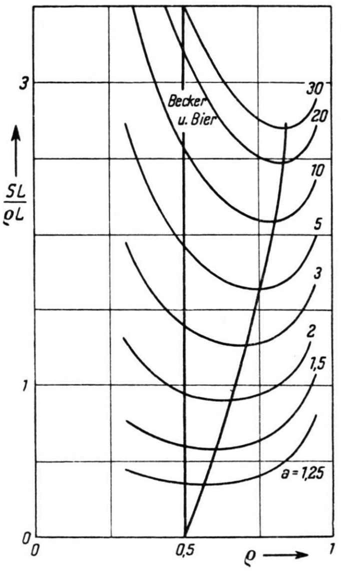

Fig. 2. Specific volume of the column as a function of the recovery. The ordinates are normalized multiplying by log K.

If one has to choose the best operation for a *given* column, and not the best column for a given separation, graphs of the type reported in fig. 3 can be useful. Here $a(\varrho)$ for a given column (7″) is set against ϱ; $a(0)=A$ is the enrichment "at total reflux" or with no isotope withdrawal; the curve at the right reproduces the locus of minimum specific volume in fig. 1, the vertical line the Becker and Bier's[2] optimum. For the hydraulical operation of a given column it can be said that, if the equilibrium time is not very important, the column must be run at LS maximum, i. e. at the maximum total throughput per unit of volume; if the equilibrium time is critical, it can be convenient to operate the column at LS/Q max., where Q is the holdup of the column thus having the maximum total throughput per unit of holdup (2).

In general, an intermediate situation will be the most convenient.

The equilibrium time is strictly proportional to Q/LS.

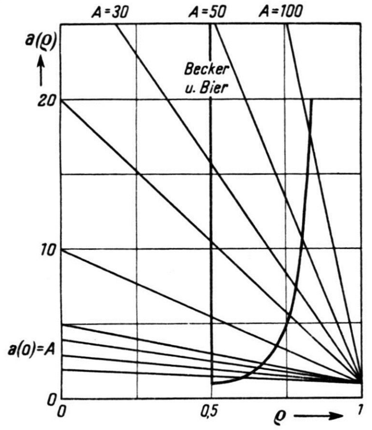

Fig. 3. Fractionation vs. recovery. The vertical line gives the best columns with the value function of Becker and Bier². The curve gives the columns of minimum specific volume.

The Cascading Problem

Except for the case of a very low enrichment, ($\lesssim 2$), it is not economical to have a single column of great length, because the transport φ being constant, L can be made inversely proportional to N, as is easily seen by applying an expression like the (7′) to the section of a column down from the point, where the concentration is N, and so, by making L constant a great volume of the plant is not properly utilized. Thus one could be led to scale the plant plate by plate; this procedure being not very practical, it is advantageous to cut up the plant into several stages, each stage being constituted by a constant L column. In this way, much can be saved in plant size and equilibrium time.

We shall solve here the problem of the best cut of a certain enrichment interval. We shall select the case where volume is the limiting factor (or proportional to all the economical factors), and single stage enrichment is $\lesssim 3$, and this only, because the mathematical results come very simple.

The locus of minimum specific volume or throughput in fig. 2 is a one valued function of ϱ; thus we have a relation between a and ϱ, that for $a \lesssim 3$ can be well approximated by the expression

$$\varrho = a/(1+a) \qquad (9)$$

which corresponds to the known „square root rule‟: for minimizing *volume* and for enrichments up to ≈ 3 must be $a = A^{1/2}$; or, if $S_{\min}$ is the minimum number of plates for a certain fractionation ($\varrho = 0$), $S_{\text{opt}} = 2\,S_{\min}$.

In fact, if $A = (a-\varrho)/(1-\varrho) = a^2$, we obtain (9) for ϱ.

Let us now suppose to have a certain enrichment interval I, and to look for the best division in two subintervals a_1, a_2. $I = a_1 \cdot a_2$, $a_1 = N_1/N_0$, $a_2 = N_2/N_1$; N_1, N_2 are the concentrations at the output ends of the two intervals. N_1 must be such, that for a certain product output, the plant volume be minimum; thus we must minimize:

$$L_1 S_1 + L_2 S_2 = V. \qquad (10)$$

By the definition of ϱ:

$$\varrho = -\frac{1}{N_0}\,\frac{c}{b-1} = \frac{\varphi}{LN_0}\,\frac{K}{K-1} = \frac{a}{a+1}, \qquad (11)$$

we obtain for L:

$$L = \varphi\,\frac{K}{K-1}\,\frac{1}{N_0}\,\frac{a}{a+1} = \varphi\,\frac{K}{K-1}\left(\frac{1}{N_S}+\frac{1}{N_0}\right) \qquad (12)$$

so that

$$L_1 = \varphi\,\frac{K}{K-1}\left(\frac{1}{N_1}+\frac{1}{N_0}\right), \qquad (12')$$

$$L_2 = \varphi\,\frac{K}{K-1}\left(\frac{1}{N_2}+\frac{1}{N_1}\right).$$

We shall have then (9), (12′):

$$L_1 S_1 + L_2 S_2 = \varphi\,\frac{K}{K-1}\left(\frac{1}{N_1}+\frac{1}{N_0}\right)\frac{\ln a_1^2}{\ln K} + \varphi\,\frac{K}{K-1}\left(\frac{1}{N_2}+\frac{1}{N_1}\right)\frac{\ln a_2^2}{\ln K}. \qquad (13)$$

φ is a constant along the plant.

The value of the intermediate point N_1 or of a_1, that minimizes the expression (13), is found by derivation to be:

$$a_1 = \frac{I \ln I}{I-1}. \qquad (14)$$

If we have m intervals so that $a_1 \cdot a_2 \cdot \ldots \cdot a_m = I$, we must minimize

$$V = L_1 S_1 + L_2 S_2 + \ldots + L_m S_m, \qquad (15)$$

and to be a minimum, (15) must be:

$$\frac{\partial V}{\partial N_1} = 0, \frac{\partial V}{\partial N_2} = 0, \ldots, \frac{\partial V}{\partial N_m} = 0. \qquad (16)$$

But N_1 appears only in L_1S_1, L_2S_2; N_2 in L_2S_2, L_3S_3 . . ., so that the problem is reduced to the minimization of a function like the (13).

This means physically that an interval is well divided when every couple of adjacent subintervals forms a well divided interval.

Thus the problem of subdivision is a very simple one. One chooses a_1, and by the (14) finds a_2, by the use of a_2 in the same way finds a_3, and so on.

It must be noted that, if one of the three quantities a_1, ϱ_1, m, is fixed, (I given) the other two are as well; small a_1 means small recovery ϱ_1 (9), and thus big L or small plant volume and high number of stages; large a_1 means high recovery ϱ_1, small L or high plant volume and small number of stages.

There also exists an economical optimum here.

It could be useful to plot a_2 vs. I as given by the (14) on a bilogarithmic chart (fig. 4), that permits rapid graphical construction of the a_r with the use of the compass only.

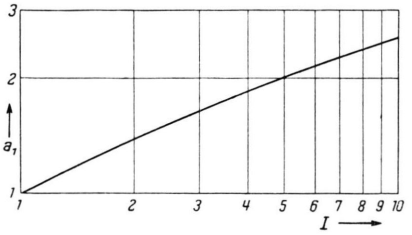

Fig. 4. Best bisection curve for $a \lesssim 3$ and minimum volume cascades.

(16) and the associated argument for the variables are not dependent on the particular form of (11) and (6′), and thus the rule about when the interval is well divided holds in general. Columns operated at $\varrho = a/(1+a)$ have another advantage, the holdup of desired isotope is minimum by respect to any column with the same concentration extremes and production, the hydraulical situation being the same, of course. Apart from constant factors the product output and the holdup per plate of desired isotope are resp. $L\varrho$ and $La(\varrho)$; the specific holdup of desired isotope for a column of S plates will be then:

$$H = [\sum_1^S La(r)]/L\varrho \text{ or } H = \int_0^S [a(\varrho)/\varrho]\,\mathrm{d}r\,. \quad (17)$$

Remembering that $A(r) = K^r$ we have $a(r) = K^r - \varrho(K^r - 1)$; substituting in (17) and integrating, $H = [(K^S - 1)(1 - \varrho)/\varrho \ln K] + S$. Substituting for S (6″) and for $K^S = A = (a - \varrho)/(1 - \varrho)$ and simplifying,

$$H = (a-1)/\varrho \ln K + [\ln (a-\varrho)/(1-\varrho)]/\ln K\,.$$

For $\varrho \to 0$ and $\varrho \to 1$, $H \to +\infty$ and the only zero of the derivative, at $\varrho = a/(1+a)$ gives a minimum.

The same holds with obvious conditions for the cascade above studied.

The above mentioned considerations have been developed while bearing in mind a plant for preenriching heavy water, and this accounts for the particular simplifications used; the concentration N being always less than some percent, $N/(1-N) \sim N$ is a safe approximation; and as the heavy water is withdrawn at a rate of 99.8% by an annex, to be attached to the plant and not considered here, the relative difference between L and l is of the order of N, i. e. of one per ten thousand for the first column and one percent for the last, and $L = l$ can be fully justified.

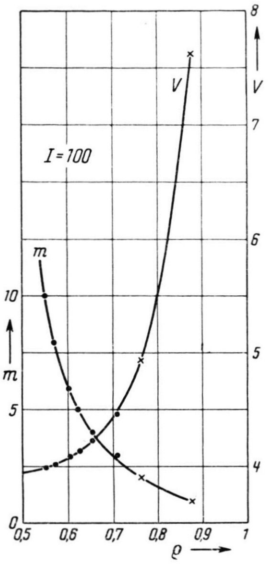

Fig. 5. Plant volume and number of stages vs. recovery for a minimum volume plant. Points ⊙ are calculated with the formulae in the text, points + have been obtained graphically. $I = 100$.

The K is not directly the equilibrium constant for the deuterium exchange between hydrogen and water because it is difficult to make the hydrogen exchange with water in liquid phase and it is costly to evaporate and condense water in every plate. So the design has been chosen that embodies a countercurrent between water and a stream of steam-hydrogen, the hydrogen transferring deuterium to steam in the gas phase over a catalyst, and afterwards the steam to the water in a bubble plate, and so on.

So the K appearing in the formulae is a function of the constant of equilibrium at the temperature of operation and of liquid, steam, and hydrogen flows, but it is independent from N and then (5) holds.

I wish to thank Dr. Ing. S. Finzi for his kind collaboration and Prof. G. Bolla for his interest and support.