673. *The Separation of Isotopes by Fractional Distillation. Part II.** *Determination of Parameters from Production Data. Value of the Unit Process Separation Factor for the* $H_2{}^{16}O$–$H_2{}^{18}O$ *System.*

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Application of the theory of fractional distillation to the data obtained in the production of enriched isotopic materials makes it possible to derive simultaneously the values for the unit process separation factor, α, and the effectiveness of the column for the system used. A graphical method is presented which facilitates the derivation of such information from experimental data. The values of the unit process separation factors for the systems $H_2{}^{16}O$–$H_2{}^{18}O$ and H_2O–HDO have been determined from the results of fractionation of water.

I. *Steady-state Operation of Fractionating Column for Isotope Separation.*—The theory of the steady-state operation of a packed column for isotope separation has been developed by Cohen (*J. Chem. Phys.*, 1940, **8**, 588; cf. N.N.E.S., Div. III, Vol. IB) and the authors (*Bull. Res. Council, Israel*, 1951, **1**, 120; Farkas Memorial Volume, in the press). The

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* Part I, preceding paper.

relevant equations, subject to assumptions and limitations discussed before by the authors (*loc. cit.*), are :

$$L\,\mathrm{d}N/\mathrm{d}z + k(N - \alpha n) = 0 \qquad (1.1)$$

$$l\,\mathrm{d}n/\mathrm{d}z + k(N - \alpha n) = 0 \qquad (1.2)$$

where z is measured from 0 at the head of the column to Z at its foot, $N(z)$, $n(z)$ are the mole fractions of the rarer isotope in the liquid and the vapour, respectively, at the distance z from the top of the column; L and l are respectively the downward rate of flow of liquid and upward rate of flow of vapour; k is a coefficient which determines the rate of transfer of isotope at the vapour–liquid interface ; and α is the unit process separation factor of the rare isotope for the liquid–vapour system.

It is assumed that the mixture is dilute and obeys Henry's law, *i.e.*, at equilibrium $n = N/\alpha$ and the distance of the system from equilibrium conditions can be represented by $N - \alpha n$. The transfer between the phases is assumed to be proportional to this distance from equilibrium. The more exact form of the transfer law is irrelevant for concentrations below 10%, and if it transpires that the Raoult-like law and not this one is the more accurate for higher concentrations then all our results remain valid provided we replace throughout N and n by $RN/(1 - N)$ and $rn/(1 - n)$ respectively.

The boundary conditions for operation with liquid feed at the top of the column at constant conentration N_0 and with product taken from a finite boiler at the bottom are :

$$N(0) = N_0 \qquad (1.3)$$

and

$$L[N(Z) - \alpha n(Z)] + (\alpha - 1)\,ln(Z) = 0 \qquad (1.4)$$

The solution of equations (1.1) and (1.2), subject to conditions (1.3) and (1.4), is immediate and leads to

$$n(Z) = N_0 \frac{\dfrac{p}{\alpha - 1} + \dfrac{1}{\alpha}}{\dfrac{\alpha p}{\alpha - 1} + \dfrac{1}{\alpha(1 + p)}\mathrm{e}^{-\frac{kZ}{L}(\alpha - 1 + \alpha p)}} \qquad (1.5)$$

where we have written

$$p = (L - l)/l \qquad (1.6)$$

Now the concentration of rarer isotope in the boiler

$$= N_{\mathrm{B}}\ (\text{say}) = \alpha n(Z)$$

and so

$$N_{\mathrm{B}} = N_0 \frac{\dfrac{\alpha p}{\alpha - 1} + 1}{\dfrac{\alpha p}{\alpha - 1} + \dfrac{1}{\alpha(1 + p)}\mathrm{e}^{-\omega Z\left(1 + \frac{\alpha p}{\alpha - 1}\right)}} \qquad (1.7)$$

where we have written

$$\omega = k(\alpha - 1)/L \qquad (1.8)$$

The enrichment $q = N_{\mathrm{B}}/N_0$

$$= \frac{\dfrac{\alpha p}{\alpha - 1} + 1}{\dfrac{\alpha p}{\alpha - 1} + \dfrac{1}{\alpha(1 + p)}\mathrm{e}^{-\omega Z\left(1 + \frac{\alpha p}{\alpha - 1}\right)}} \qquad (1.9)$$

II. *Determination of Parameters from Steady-state Operation Data.*—For a given column the equation (1.9) determines q as a function of p. When $p = 0$ we get

$$q = \alpha \mathrm{e}^{k(\alpha - 1)Z/L}$$

a result given by Cohen (*loc. cit.*). This, by analogy with Rayleigh's equation (*Phil. Mag.*, 1902, **4**, 521), suggests that we identify kZ/L with the number of transfer units.

In the case of isotope separation this number is for practical purposes equal to the number of theoretical plates in the column. The inverse proportion between this latter number and L is confirmed, for the type of packing used, by our experimental results (cf. section V) though it is known not to hold for some other types of packing.

It is worth considering what happens if we let $kZ/L \longrightarrow \infty$, *i.e.*, make the column infinitely efficient. Then (1.9) becomes

$$q = 1 + \alpha - 1/\alpha p \qquad (2.1)$$

and this is the maximum enrichment attainable for the given value of α and p. An immediate practical application is that there is in general a limit beyond which it is not economic to extend the number of theoretical plates.

If we know the values of the enrichment corresponding to two or more sets of values of flow and production rates, we can, theoretically, deduce the values of α and kZ. The best technique for making this deduction will depend on the circumstances. If the value of L is the same in both production runs we may use tabulated values of (1.9) as a function of α, p, and kZ/L (cf. Dostrovsky, Gillis, and Llewellyn, Farkas Memorial Volume).

The use of the tables depends on the fact that a given pair of (p, q) values determines a relation between α and kZ/L, *i.e.*, a curve in the co-ordinate plane, α, kZ/L. From the intersection of two such curves we obtain the actual values of α and kZ/L. It should be emphasized that this method is only approximate and that the tables themselves, in so far as one depends on linear interpolation between parameter values, may at best give only a good approximation to the true values. The refinement of the approximate values to the degree of precision required by the circumstances may be effected by a trial-and-error use of equation (1.9). The limitation of the use of the tables to the case in which the different runs are taken with the same value of L is a substantial one, particularly as it may be technically difficult to keep L constant over long periods of time. The graphical method described below in section III is not subject to this limitation.

III. *Graphical Method.*—This method is approximate and is valid only for small values of $(\alpha - 1)$ and p. However the restriction is unimportant since the most useful practical cases are still covered. We start from equation (1.9) and write

$$x = \alpha p/(\alpha - 1), \qquad y = kZ(\alpha - 1)/L, \qquad \alpha(1 + p) \sim 1$$

Then

$$q \sim \frac{x + 1}{x + e^{-y(1+x)}} \qquad (3.1)$$

For each of a range of values of q we plot, on the same sheet, $\log_{10} y$ against $-\log_{10} x$. For this purpose we write

$$Y = \log_{10} y \qquad (3.2)$$
$$X = \log_{10} x$$

and denote the curve of Y against X by Γq.

Values of (X, Y) for a set of values of q are tabulated in the Appendix. The general character of the curves Γq is shown in Fig. 1.

Suppose now that we are given two sets of values of L and p with the corresponding values of q. Let us denote these by (L_1, p_1, q_1) and (L_2, p_2, q_2). Substituting each of these sets in turn in equation (1.9) we obtain a pair of equations for kZ and α. These can be solved graphically by means of the curves Γq. We seek two points (X_1, Y_1) and (X_2, Y_2) with the following properties :

(*a*) (X_1, Y_1) lies on Γq_i $(i = 1, 2)$
(*b*) $X_2 - X_1 = \log_{10} (p_1/p_2)$
(*c*) $Y_2 - Y_1 = \log_{10} (L_1/L_2)$

When these have been found, *e.g.*, by a technique to be described below, we may write

$$(\alpha - 1)/\alpha = p_1 . \text{antilog}_{10} X_1 = p_2 . \text{antilog}_{10} X_2 \qquad (3.3)$$

thus determining α.

Then

$$kZ = L_1/(\alpha - 1) \,.\, \text{antilog}_{10} Y_1 = L_2/(\alpha - 1) \,.\, \text{antilog}_{10} Y_2 \quad . \quad (3.4)$$

The only approximation involved is $\alpha(1 + p) \sim 1$. Moreover this is not a very serious one since the term in question multiplies $\exp.\{[-kZ(\alpha - 1)/L] \,.\, [(1 + \alpha p/(\alpha - 1)]\}$ which in practice is always quite small, and particularly so when $(\alpha - 1)$ or p is large enough for $\alpha(1 + p)$ to differ significantly from 1.

A simple technique for locating the points (X_1, Y_1), (X_2, Y_2) is as follows: Take a set-square, preferably transparent, ABC, with its right angle at B. Lay this square with B at the origin of co-ordinates O, and with BA and BC along the axes OX and OY respectively. Now mark off BQ_1, along BA equal to $\log_{10} (p_1/p_2)$ in units of the X-scale. Similarly mark off BQ_2 from BC equal to $\log_{10} (L_1/L_2)$ in units of the Y-scale. Set the triangle keeping BA and BC parallel to OX and OY, with A to the left or right of B according as $p_1 >$ or $< p_2$, and with C above or below B according as $L_1 >$ or $< L_2$. Now move the triangle parallel to itself until Q_1 lies on Γq_1, and Q_2 on Γq_2. The positions of Q_1, Q_2 will be the points (X_1, Y_1), (X_2, Y_2) required.

FIG. 1.

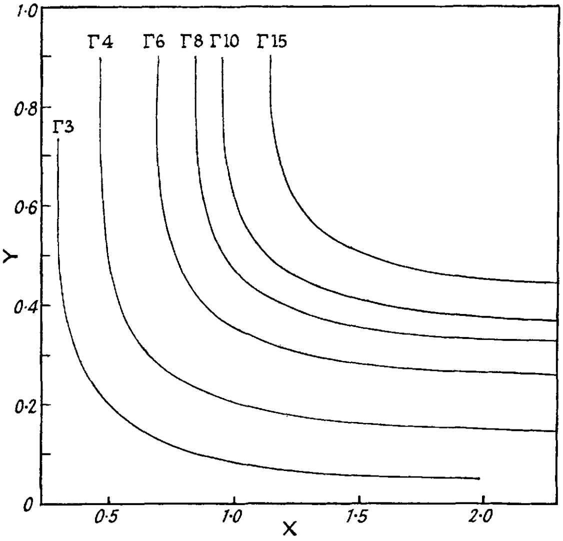
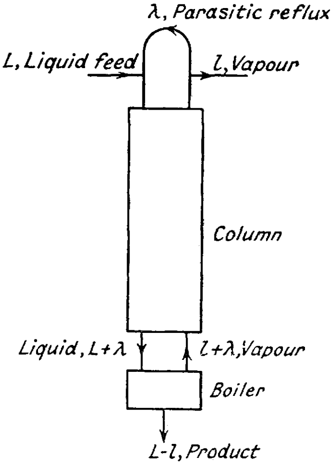


FIG. 2.



IV. *Effect of Condensation in the Column.*—In many practical systems, a part of the vapour may be unavoidably condensed in the upper part of the column, either through heat loss or by the feed liquid if the latter is below the boiling point. This effect will lead to rates of liquid and vapour flow within the column greater than feed and distillate rates respectively. Let λ be the additional flow (both liquid and vapour) in the column arising from this effect. The situation is described diagrammatically in Fig. 2.

Equations (1.1, 1.2) remain valid except that L, l are to be replaced by $L + \lambda$, $l + \lambda$ respectively. The boundary conditions are now :

(i) Material balance in boiler :

$$(L + \lambda)N(Z) - (l + \lambda)n(Z) = (L - l)\,\alpha n(Z) \quad . \quad . \quad . \quad . \quad (4.1)$$

(ii) Overall material balance :

$$LN_0 - ln(0) = (L - l)\,\alpha n(Z) \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4.2)$$

The assumption underlying (4.2) is that the outgoing vapour has the same composition as that rising from the top of the packing. Another possible assumption could be that the condensate is in equilibrium with the uncondensed vapour, *i.e.*, that there is a slight stripping of the uncondensed vapour and that therefore the concentration of heavy isotopes in the distillate leaving the column is slightly below $n(0)$. Both of these extreme cases have been worked out in detail and it transpires that there is no significant difference between the two values of q. For simplicity we shall use equation (4.2) as it stands.

The solution of the differential equations with these boundary conditions leads to :

$$q = \alpha n(Z)/N_0$$

$$= \frac{1 + \dfrac{\alpha}{\alpha - 1} \cdot \dfrac{L - l}{l + \lambda}}{\dfrac{l}{\alpha L} e^{-\omega Z} + \dfrac{\alpha}{\alpha - 1} \cdot \dfrac{L - l}{l + \lambda} \cdot \dfrac{L + [\alpha - 1)/\alpha]\lambda}{L}} \quad . \quad . \quad . \quad . \quad . \quad (4.3)$$

where

$$\omega Z = \{kZ/(L + \lambda)\}\left\{\alpha - 1 + \frac{\alpha(L - l)}{l + \lambda}\right\} \quad . \quad . \quad . \quad . \quad . \quad (4.4)$$

We now write $p = (L - l)/l$, *i.e.*, the *observed* reflux ratio. Then (4.3) becomes

$$q = \frac{1 + \dfrac{\alpha}{\alpha - 1} \cdot \dfrac{p}{1 + \lambda/l}}{\dfrac{1}{\alpha(1 + p)} e^{-\omega Z} + \left[1 + \dfrac{\alpha - 1}{\alpha(1 + p)} \cdot \dfrac{\lambda}{l}\right] \dfrac{\alpha}{\alpha - 1} \cdot \dfrac{p}{1 + \gamma/l}} \quad . \quad . \quad . \quad . \quad (4.5)$$

The effect of the parasitic reflux γ on the observed enrichment q may be in either direction depending on the position of $(L + \lambda)$ with respect to the optimal flow (cf. Dostrovsky, Gillis, and Vromen, *Bull. Israel Research Council*, Vol. II, in the press).

We also write

$$p' = (L - l)/(l + \lambda) \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4.6)$$

and

$$q' = \frac{1 + \dfrac{\alpha p'}{\alpha - 1}}{\dfrac{1}{\alpha(p' + 1)} e^{-\omega Z} + \dfrac{\alpha p'}{\alpha - 1}} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4.7)$$

In the denominators of the expressions for q, q', the term involving $e^{-\omega Z}$ is generally small and the difference between these two terms negligible. We are thus led to the relation

$$1 < q'/q < 1 + (\alpha - 1)(\lambda/l) \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4.8)$$

It follows from (4.8) that for all moderate values of λ/l, q' may be taken as an adequate approximation to q. Now the expression (4.7) for q' is simply (1.9) with L, l, replaced by $L + \lambda$, $l + \lambda$ respectively. Hence if we know the value of λ we can still use the methods of Sections II and III to determine α, kZ.

V. *The Value of* α *for the* $H_2{}^{16}O$–$H_2{}^{18}O$ *System.*—The unit process separation factor for the system $H_2{}^{16}O/H_2{}^{18}O$ was first determined by Wahl and Urey (*J. Chem. Phys.*, 1935, **3**, 411) using simple evaporation of water, and analysing the increase in ^{18}O content of the residue. A similar method was used by Riesenfeld and Chang (*Z. physikal. Chem.*, 1936, **33**, 127). Using packed fractionating columns operating with a liquid feed of normal water and with small boiler, we carried out a series of production experiments. The various runs differed in conditions of production rate, flow, and pressure. The results of some of the runs are shown in the Table.

It should be emphasized that there is necessarily a pressure difference ΔP between the top of the column and its foot and so α is not constant over the length of the column. It follows that our equations and their solutions involve the use of an average value of α as an approximation. However for the small pressure differences with which we operated the variation of α along the column is slight and it is certainly adequate to define its derived value as that for the mean pressure in the column. The values of q reported in the Table are the averages for a period at least 11 days after the steady state had been reached.

Data for the production of $H_2{}^{18}O$ *by fractionation of water.*

Mean pressure, mm. Hg	ΔP, mm. Hg	L, l./day	p	$q_{obs.}$	$q_{calc.}$
Column A.					
257	75	51·7	0·00290	3·22	3·26
259	78	50·1	0·00200	4·10	4·10
271	52	37·9	0·000849	8·26	8·18
274	48	36·7	0·000540	11·23	11·30
265	42	33·3	0·000593	11·20	11·24
Column B.					
355	130	57·5	0·00348	3·08	3·08
355	130	57·5	0·00174	5·05	5·04
355	130	57·5	0·00087	8·52	8·49
355	130	57·5	0·00261	3·74	3·75

Column A (Rehovoth, Israel) had a packed section of 90 mm. × 7 m.
Column B (London, England) had a packed section of 75 mm. × 15 m.

Great care has to be taken to ensure that q is actually the steady-state value, and to continue the production long enough for this to be beyond doubt. Otherwise transient effects arising from the initiation of production can seriously affect the value of q obtained. This error was encountered by us in earlier work before its nature was sufficiently appreciated. A physical explanation and theoretical discussion will be given in a subsequent paper of this series.

It was assumed that λ/l was constant for each column. This assumption, although inexact, is probably sufficiently accurate for practical purposes. In both columns A and B the water was fed at room temperature and it can be calculated that this would lead to a condensation of vapour equivalent to 10% of the inflow. This amount is the lower limit for λ/l and in practice a slightly higher value may be expected as a result of heat losses in the upper parts of the columns. For each column the values of λ/l, α, kZ were determined by a "least squares" method. The data of column A led to $\alpha = 1{\cdot}0065$, $kZ = 22{,}940$, $\lambda/l = 0{\cdot}10$. Column B gave $\alpha = 1{\cdot}0064$, $kZ = 40{,}200$, $\lambda/l = 0{\cdot}15$. The values of q calculated with these values of the constants are shown in the last column of the Table.

It should be remarked that the slightly lower value of α for column B may be associated with the higher pressure of operation compared with column A. Also the higher value of λ/l is readily understood if we take into account the fact that the London environment was on the average much colder than that at Rehovoth and, moreover, that column B was provided with a steam jacket to compensate for heat loss from its walls. It is interesting also to note that $(kZ)_B/(kZ)_A = 1{\cdot}75$ though $Z_B/Z_A = 2{\cdot}14$. This is in accordance with the known physical fact that the "number of theoretical plates" does not increase proportionally with an increase in the length of the column.

The satisfactory agreement between the observed and the calculated values of q confirms the validity of the model described by the differential equations and, incidentally, the inverse proportion between L and the number of theoretical plates.

The value of α which we have derived for the $H_2{}^{16}O$–$H_2{}^{18}O$ system is seen to be in substantial agreement with that obtained by Wahl and Urey (*loc. cit.*) ($\alpha = 1{\cdot}0066$ at 300 mm.), but somewhat higher than that given by Riesenfeld and Chang (*loc. cit.*) ($\alpha = 1{\cdot}0052$ at 300 mm.).

VI. *The Value of* α *for the* H_2O–HDO *System*.—For this system, with its comparatively high value of α and correspondingly low value of $\exp\{(-kZ/L) \,.\, (\alpha - 1)\}$, the enrichment is determined by the rate of production and depends hardly at all on the length of our column. This is equivalent to saying that our column is effectively infinite for the system and so q is given quite simply by (2.1), but using p' instead of p (cf. equation 4.6). The enrichment of deuterium was measured for the last two runs of column A reported in the Table. The values were 78 ± 4 and 70 ± 5 respectively. Applying the modified equation (2.1) and using $\lambda/l = 0{\cdot}1$ we obtain for α the respective values 1·038 ± 0·003, 1·038 ± 0·002. This result is in agreement with the values given by Riesenfeld and Chang (*op. cit.*) for the pressures used.

We remark that the column was designed and operated for the production of $H_2{}^{18}O$ and in consequence the conditions with respect to HDO production were far from optimal and could be regarded as wasteful.

In conclusion the authors express their appreciation to Drs. C. A. Bunton and F. Klein for the numerous mass-spectrometric analyses performed, and to Professor E. D. Hughes, F.R.S., for his continued interest in this work. One of the authors (D. R. Llewellyn) acknowledges with gratitude the tenure of an I.C.I. Fellowship at University College, London.

APPENDIX

Tables of Γ *functions for various values of* q.

$q = 3$		$q = 4$		$q = 5$		$q = 6$	
X	Y	X	Y	X	Y	X	Y
0·305	0·590	0·48	0·682	0·605	0·723	0·700	0·813
0·310	0·525	0·485	0·609	0·61	0·655	0·710	0·661
0·33	0·418	0·49	0·568	0·62	0·590	0·720	0·609
0·35	0·363	0·50	0·520	0·63	0·550	0·730	0·576
0·37	0·325	0·51	0·486	0·64	0·522	0·740	0·551
0·39	0·296	0·52	0·460	0·65	0·500	0·75	0·530
0·42	0·263	0·54	0·420	0·66	0·481	0·76	0·513
0·44	0·245	0·56	0·391	0·68	0·451	0·77	0·498
0·46	0·229	0·58	0·367	0·70	0·427	0·79	0·474
0·50	0·203	0·60	0·348	0·72	0·408	0·80	0·463
0·55	0·177	0·64	0·316	0·76	0·377	0·82	0·445
0·60	0·157	0·68	0·293	0·80	0·354	0·84	0·429
0·65	0·140	0·725	0·271	0·84	0·335	0·86	0·415
0·70	0·127	0·775	0·252	0·86	0·326	0·90	0·393
0·75	0·116	0·85	0·230	0·90	0·312	0·95	0·371
0·80	0·106	0·90	0·218	0·95	0·297	1·00	0·354
0·90	0·091	1·00	0·200	1·00	0·285	1·1	0·328
1·00	0·080	1·1	0·186	1·1	0·266	1·2	0·310
1·25	0·062	1·3	0·169	1·2	0·252	1·3	0·297
1·50	0·052	1·5	0·158	1·3	0·242	1·5	0·280
1·75	0·047	1·7	0·152	1·5	0·228	1·8	0·266
2·00	0·045	2·0	0·147	1·75	0·219	2·25	0·258
3·40	0·041	2·5	0·144	2·0	0·213	2·5	0·256
4·00	0·041	3·0	0·142	3·0	0·207	3·0	0·254

$q = 7$		$q = 8$		$q = 9$		$q = 10$	
X	Y	X	Y	X	Y	X	Y
0·78	0·806	0·85	0·762	0·905	0·838	0·955	0·888
0·785	0·721	0·855	0·711	0·91	0·751	0·96	0·774
0·79	0·680	0·86	0·681	0·915	0·714	0·965	0·735
0·80	0·632	0·87	0·641	0·92	0·688	0·97	0·708
0·81	0·600	0·88	0·613	0·925	0·669	0·975	0·686
0·82	0·576	0·89	0·591	0·93	0·652	0·98	0·670
0·83	0·556	0·90	0·573	0·94	0·625	0·99	0·646
0·84	0·540	0·92	0·544	0·95	0·605	1·00	0·622
0·85	0·526	0·94	0·522	0·96	0·587	1·02	0·590
0·86	0·513	0·96	0·504	0·98	0·560	1·04	0·567
0·87	0·502	0·98	0·488	1·00	0·539	1·06	0·550
0·88	0·491	1·00	0·475	1·25	0·517	1·1	0·518
0·89	0·482	1·1	0·428	1·05	0·500	1·2	0·471
0·925	0·456	1·2	0·399	1·75	0·485	1·3	0·442
0·95	0·440	1·3	0·379	1·1	0·473	1·4	0·422
1·0	0·415	1·4	0·364	1·2	0·436	1·5	0·408
1·1	0·381	1·5	0·354	1·3	0·412	1·6	0·397
1·2	0·358	1·6	0·346	1·4	0·395	1·7	0·386
1·3	0·341	1·7	0·340	1·5	0·382	1·8	0·383
1·5	0·320	1·8	0·335	1·6	0·373	1·9	0·379
1·7	0·308	1·9	0·331	1·8	0·361	2·0	0·375
2·0	0·298	2·0	0·329	2·0	0·354	2·2	0·370
2·5	0·292	2·5	0·321	2·5	0·346	2·6	0·366
3·0	0·290	3·0	0·319	3·0	0·343	3·0	0·363

APPENDIX—*continued.*

$q = 11$		$q = 12$		$q = 15$		$q = 20$	
X	Y	X	Y	X	Y	X	Y
1·005	0·796	1·045	0·825	1·15	0·841	1·285	0·838
1·01	0·753	1·05	0·770	1·155	0·791	1·29	0·802
1·015	0·722	1·06	0·716	1·16	0·761	1·295	0·778
1·02	0·701	1·07	0·684	1·165	0·740	1·30	0·760
1·025	0·684	1·075	0·671	1·17	0·723	1·31	0·732
1·03	0·669	1·08	0·660	1·175	0·708	1·32	0·712
1·04	0·646	1·09	0·641	1·18	0·696	1·34	0·682
1·05	0·627	1·10	0·625	1·19	0·676	1·35	0·670
1·06	0·612	1·125	0·600	1·20	0·660	1·375	0·646
1·08	0·587	1·15	0·572	1·22	0·634	1·4	0·627
1·1	0·567	1·175	0·553	1·24	0·614	1·425	0·612
1·12	0·550	1·2	0·538	1·26	0·600	1·45	0·560
1·14	0·536	1·25	0·514	1·28	0·584	1·475	0·589
1·16	0·524	1·3	0·495	1·35	0·548	1·5	0·579
1·18	0·514	1·35	0·481	1·4	0·530	1·55	0·563
1·2	0·504	1·4	0·469	1·45	0·516	1·6	0·551
1·25	0·485	1·5	0·451	1·5	0·504	1·65	0·541
1·3	0·469	1·6	0·439	1·6	0·486	1·7	0·532
1·4	0·446	1·7	0·428	1·7	0·474	1·8	0·519
1·5	0·430	1·8	0·421	1·8	0·464	1·9	0·509
1·7	0·410	2·0	0·411	2·0	0·452	2·0	0·502
2·0	0·394	2·2	0·405	2·2	0·445	2·2	0·492
2·4	0·385	2·6	0·399	2·6	0·437	2·6	0·483
3·0	0·381	3·0	0·397	3·0	0·434	3·0	0·479

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