PURIFICATION FACTOR CHARACTERIZATION OF ZONE REFINING

BY

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ABSTRACT

A quantity π_T, termed the purification factor, defines the potential state of purity realizable in a specimen that has undergone zone-refinement treatment. An exact analysis appears intractable, but neglect of the terminal zone perturbation (which becomes vanishingly small for the infinite ingot) allows a lower bound to be established on the remanent solute impurity. Such numerical values of π_T are evaluated for some sensible distribution coefficients and zone dimensions involving a maximum of ten passes. An original expression for the solute impurity distribution is deduced and numerically appraised for a range of operational conditions not presently tabulated; the relation has been applied to the indicated π_T determinations.

INTRODUCTION

The purpose here is to describe in some detail a useful parameter for characterizing the progress made at any stage of the zone-refining process. In doing so, it may also be of value to present still another manner for expressing the impurity distribution which evidently lends itself more readily to numerical calculation. Thus a quantity identified as a purification factor is explicitly evaluated for a range of conditions considered to be representative of zone-refining practice.

Actually, the present treatment is restricted to an upper bound identification of the degree of purification attained during the refinement precedure; analytic treatment of the terminal zone effect does not appear to be tractable, although the correction could be numerically evaluated. The technique of terminal zone cropping tends to moderate the departure from the ideal purification factor since it operates to offset the approach to an equilibrium impurity distribution. A brief description of this approach was offered earlier (1).[3]

IMPURITY BALANCE AND THE PURIFICATION FACTOR

A simple impurity balance may be posed ignoring the terminal zone perturbation. Conservation of solute in an ingot of length L having an initial distribution $C_0(x)$ may be represented as follows:

$$\sum S = C_0 L = S_T = S_{I,n} + S_{II,n} + S_{III,n-1} + \sum_{n-1} S_{f,n-1} \qquad (1)$$

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[3] The boldface numbers in parentheses refer to the references appended to this paper.

where the total impurity solute S_T is expressed in terms of the components $S_{I,n}$, the solute in the purified zone after the n^{th} pass, $S_{II,n}$, the solute in the liquid zone, $S_{III,n-1}$, the solute in the remaining ingot from the previous pass, exclusive of the terminal zone, which must be summed over $S_{f,n-1}$ for the maximum accumulation of impurity after $n - 1$ passes.

The compartmental distribution of impurity then is prescribed by the relations

$$S_{I,n} = \int_0^x C_n(x)dx \tag{2a}$$

$$S_{II,n} = \frac{l}{k} C_n(x) \tag{2b}$$

$$S_{III,n-1} = \int_{x+l}^{L-l} C_{n-1}(x)dx \tag{2c}$$

$$S_{f,n-1} = lC_{n-1}(x)\big|_{x=L-l} \tag{2d}$$

where l is the zone length and k is the solute distribution coefficient.

Thus for the fraction of solute impurity removed after the n^{th} pass P_n there derives

$$P_n = \frac{1}{C_0L}\int_0^{L-l} [C_{n-1}(x) - C_n(x)]dx \tag{3}$$

whereas for the cumulative removal of impurity it follows that

$$P_T = \sum_n P_n = \left(C_0L - \int_0^{L-l} C_n(x)dx\right)(C_0L)^{-1}. \tag{4}$$

The fraction of impurity yet to be transferred to the terminal zone may be defined as the purification factor π_T and is clearly the result

$$\pi_T = 1 - P_T. \tag{5}$$

Explicit calculation of the purification factor demands identification of $C_n(x)$ and the subsequent evaluation of Eq. 4; the next section deals with this aspect.

ELABORATION OF THE P_T RELATION[4]: A NEW FORM FOR $C_n(X)$

The detailed calculations of the purification factors are based upon a form of $C_n(x)$ not previously described in the literature (2). Starting with the underlying equation (3) which specifies the spatial distribution

[4] The usual idealizations are embodied as, for example, perfect mixing of components in the liquidus, limited diffusion in the solidus, etc.

of solute

$$\frac{dS_n(x)}{dx} = C_{(n-1)}(x) - \frac{k}{l} S_n(x) \tag{6}$$

the solution may readily be shown to be of the form

$$S_n(x) = e^{-(k/l)x} \left\{ \int_0^x C_{(n-1)} e^{(k/l)x} dx + K_{(n-1)} \right\} \tag{7}$$

wherein the $K_{(n-1)}$ are integration constants prescribed by the initial condition

$$C_n(x)\,|_{x=0} = k\bar{c}, \qquad \bar{c} = \frac{1}{l}\int_0^l C_{(n-1)}(x)dx \tag{8}$$

when utilized in conjunction with

$$C_n(x) = \frac{k}{l} S_n(x). \tag{9}$$

In the appendix are provided details for the inductive prescription of $Q_n(x)$ in the generalized form of $C_n(x)$ which resembles that of the single pass result:

$$C_n(x)/C_0 = 1 - Q_n(1 - k)e^{-(k/l)x} \tag{10a}$$

with

$$Q_n = \sum_{p=n}^{1} A_p \frac{\left(\frac{k}{l}x\right)^{n-p}}{(n-p)!} \tag{10b}$$

and the diverse A_p in turn given by

$$A_j = 1 - \frac{A_{j-1}S_1}{0!} - \frac{A_{j-2}S_2}{1!} - \frac{A_{j-3}S_3}{2!} - \cdots$$
$$- \frac{A_2 S_{j-2}}{(j-3)!} + \frac{1}{(j-2)!}\left(\frac{k}{l}\right)^{j-1} \int_0^l x^{j-2} e^{-(k/l)x} dx. \tag{10c}$$

In (10c) the S_m takes on values determined from

$$S_m = -\int_0^k x^{j-1}e^{-x}dx = -\left[\int k^{j-1}e^{-k}dk + (j-1)!\right]. \tag{10d}$$

Typically, for $j = 1$, $A_1 = 1$ and $Q_1 = 1$.

In Table I are collected the solute distributions $C_n(x)/C_0$ as a function of x/l for $n = 1$ to 10 with k values selected as 0.001, 0.007, 0.05, 0.07 and 0.12. The over-all behavior of the solute distribution is exhibited in Figs. 1–6; two different ranges are displayed in Figs. 1 and 2, with the latter appropriate for small x/l and $k = 0.001$. Other values

TABLE I.—$C_n(x)/C_0$ *vs* x/l *Data for* k = 0.001, 0.007, 0.05, 0.07, 0.12.

$k = .001$

$\frac{x}{l}$	$\frac{C_1(x)}{C_0}$	$\frac{C_2(x)}{C_0}$	$\frac{C_3(x)}{C_0}$	$\frac{C_4(x)}{C_0}$	$\frac{C_5(x)}{C_0}$	$\frac{C_6(x)}{C_0}$	$\frac{C_7(x)}{C_0}$	$\frac{C_8(x)}{C_0}$	$\frac{C_9(x)}{C_0}$	$\frac{C_{10}(x)}{C_0}$
0	.001	$.150 \cdot 10^{-5}$	0	0	0	0	0	0	0	0
1	.001985	$.300 \cdot 10^{-5}$	0	0	0	0	0	0	0	0
10	.01094	$.610 \cdot 10^{-4}$	$.23 \cdot 10^{-6}$	0	0	0	0	0	0	0
100	.09607	.004771	$.1593 \cdot 10^{-3}$	$.400 \cdot 10^{-5}$	$.8 \cdot 10^{-7}$	0	0	0	0	0
500	.3935	.09051	.01446	.001764	$.1757 \cdot 10^{-3}$	$.143 \cdot 10^{-4}$	$.102 \cdot 10^{-5}$	$.6 \cdot 10^{-7}$	0	0
1000	.6321	.2646	.08048	.01574	.003675	$.5973 \cdot 10^{-3}$	$.838 \cdot 10^{-4}$	$.103 \cdot 10^{-4}$	$.113 \cdot 10^{-5}$	$.11 \cdot 10^{-6}$
2000	.8647	.5943	.3236	.1418	.05274	.01660	.005050	.001100	$.2382 \cdot 10^{-3}$	$.470 \cdot 10^{-4}$
3000	.9502	.8010	.5770	.3530	.1849	.08402	.03356	.01193	.003811	.001105
4000	.9817	.9085	.7620	.5667	.3714	.2150	.1108	.05119	.02139	.008146
5000	.9913	.9596	.8754	.7351	.5597	.3842	.2380	.1335	.06816	.03186
6000	.9975	.9827	.9381	.8489	.7151	.5545	.3938	.2526	.1529	.08399
7000	.9991	.9927	.9704	.9183	.8271	.6994	.5504	.4014	.2710	.1696
8000	.9997	.9970	.9862	.9576	.9004	.8088	.6867	.5472	.4076	.2835
9000	.9999	.9988	.9938	.9788	.9451	.8845	.7933	.6763	.5445	.4127
10,000	1.0	.9995	.9972	.9897	.9708	.9330	.8699	.7799	.6670	.5422
12,000								.9105	.8450	.7577
15,000	1.0	1.0	1.0	.9998	.9991	.9972	.9924	.9820	.9626	.9302
20,000				1.0	1.0	.9999	.9997	.9992	.9979	.9950
22,000						1.0	1.0	.9998	.9994	.9988

$k = .007$

$\frac{x}{l}$	$\frac{C_1(x)}{C_0}$	$\frac{C_2(x)}{C_0}$	$\frac{C_3(x)}{C_0}$	$\frac{C_4(x)}{C_0}$	$\frac{C_5(x)}{C_0}$	$\frac{C_6(x)}{C_0}$	$\frac{C_7(x)}{C_0}$	$\frac{C_8(x)}{C_0}$	$\frac{C_9(x)}{C_0}$	$\frac{C_{10}(x)}{C_0}$
0	.007	.0000733	.00000074							
1	.01393	.0001458	.000003	0						
3	.02764	.000434		0						
10	.07413	.002864	.000073	.0000018	0	0				
30	.1951	.02044	.001461		0	0	0			
50				.000512	.0000366	.0000025	0	0	0	0
100	.5069	.1583	.03502	.00599	.00090	.000074	.0000083	0	0	0
200	.7551	.4106	.1682	.05456	.01432	.00343	.000703	.00016	.0000172	.00000264
300	.8784	.6222	.3523	.1627	.06217	.02078	.0060	.00156	.000346	.0000718
500	.9700	.8648	.6705	.4649	.2746	.1434	.0659	.02707	.01004	.00336
600						.2480	.1334	.06448	.02825	.01132
800			.9180	.8101	.6579	.4893	.3309	.2040	.1150	.05956
1000	.9991	.9928	.9710	.9186	.8206	.7002	.5514	.4024	.2718	.1703
1300						.8907	.8030	.6885	.5584	.4267
1500	1.0	.9997		.9929	.9789	.9498	.8987	.8220	.7213	.6037
2000		1.0	.9999	.9995	.9982	.9945	.9858	.9685	.9382	.8909
2500			1.0					.9960	.9906	.980
3000				1.0	1.0	1.0	.9999	.9996	.999	.9972
4000										1.0

TABLE I.—*Continued.*

$\frac{x}{l}$	$\frac{C_1(x)}{C_0}$	$\frac{C_2(x)}{C_0}$	$\frac{C_3(x)}{C_0}$	$\frac{C_4(x)}{C_0}$	$\frac{C_5(x)}{C_0}$	$\frac{C_6(x)}{C_0}$	$\frac{C_7(x)}{C_0}$	$\frac{C_8(x)}{C_0}$	$\frac{C_9(x)}{C_0}$	$\frac{C_{10}(x)}{C_0}$
					$k = .05$					
0	.05	.003668	.000260	.0000184	.0000013	.0000001	0			
1	.0963	.007076	.000501	.0000354	.0000025	.0000002	0			
4	.2222	.02853	.002780	.000229	.0000172	.0000013	.0000001	0		
10	.4238	.1076	.01945	.002751	.000323	.0000330	.0000030	.0000002	0	
15	.5512	.1928	.04857	.009541	.001539	.000212	.0000258	.0000027	.0000003	0
20	.6505	.2840	.09094	.02283	.004706	.000824	.000125	.0000169	.0000021	.0000002
30	.7880	.4597	.2050	.07293	.02146	.005372	.001169	.000225	.0000388	.0000061
40	.8714	.6080	.3379	.1530	.05790	.01875	.005295	.001324	.000297	.0000604
50	.9220	.7233	.4698	.2541	.1164	.04591	.01586	.004866	.001341	.000335
60	.9527	.8085	.5886	.3648	.1940	.08964	.03645	.01320	.004302	.001274
80	.9826	.9122	.7695	.5768	.3817	.2234	.1165	.0545	.0233	.00892
100	.9936	.9613	.8797	.7423	.5688	.3935	.2458	.1392	.0718	.0339
120	.9976	.9834	.9403	.8534	.7220	.5629	.4024	.2635	.1585	.0878
140	.9991	.9930	.9715	.9209	.8318	.7060	.5582	.4093	.2780	.1751
160	.9997	.9971	.9868	.9591	.9033	.8136	.6931	.5545	.4150	.2901
180	.9999	.9988	.9940	.9795	.9468	.8875	.79800	.6823	.5514	.4197
200	1.0	.9995	.9973	.9900	.9717	.9349	.8732	.7845	.6732	.5488
220	1.0	.9998	.9988	.9953	.9854	.9636	.9235	.8602	.7727	.6653
240		.9999	.9995	.9978	.9927	.9803	.9555	.9128	.8484	.7622

TABLE I—*Continued.*

$\frac{x}{l}$	$\frac{C_1(x)}{C_0}$	$\frac{C_2(x)}{C_0}$	$\frac{C_3(x)}{C_0}$	$\frac{C_4(x)}{C_0}$	$\frac{C_5(x)}{C_0}$	$\frac{C_6(x)}{C_0}$	$\frac{C_7(x)}{C_0}$	$\frac{C_8(x)}{C_0}$	$\frac{C_9(x)}{C_0}$	$\frac{C_{10}(x)}{C_0}$	$\frac{C_{11}(x)}{C_0}$
						$k = .07$					
0	.07	.007126	.0007007	.00006879	.0000068	.0000007	0	0	0	0	0
1	.1329	.01355	.001333	.0001308	.00001285	.0000013	.0000001	0	0	0	0
3	.2462	.03689	.004352	.0004585	.00004608	.0000046	.0000004	0	0	0	0
5	.3446	.07096	.01078	.001355	.0001514	.00001582	.0000016	0	0	0	0
8	.4688	.1354	.02830	.004701	.0006603	.00008202	.0000093	0	0	0	0
10	.5382	.1837	.04548	.008885	.001448	.0002051	.00002604	.0000030	.0000004	0	0
15	.6746	.3108	.106091	.02859	.006370	.001214	.0002029	.00003041	.0000042	0	0
20	.7707	.4341	.1861	.06360	.01801	.004354	.0009189	.0001724	.00002921	.00000452	.00000065
30	.8861	.6393	.3712	.1767	.07063	.02423	.007265	.001934	.0004628	.0001006	.00002007
40	.9434	.7813	.5485	.3255	.1650	.07250	.02801	.009634	.002983	.0008397	.0002166
50	.9719	.8717	.6929	.4799	.2894	.1532	.07178	.03009	.01139	.003928	.0012419
60	.9860	.9265	.7995	.6185	.4252	.2598	.1418	.06969	.03103	.01262	.004713
70						.3802	.2348	.1313	.06689	.03120	.01341
80	.9966	.9771	.9218	.8174	.6693	.5012	.3422	.2132	.1216	.06373	.03085
100	.9992	.9932	.9720	.9220	.8338	.7089	.5617	.4129	.2812	.1776	.1043
110					.8868	.7874	.6586	.5155	.3765	.2563	.1628
120	.9998	.9980	.9905	.9694	.9245	.8486	.7413	.6114	.4738	.3442	.2343
130	.9999	.9989	.9946	.9812	.9507	.8946	.8088	.6964	.5674	.4360	.3154
140						.9280	.8618	.7684	.6531	.5266	.4017
150					.9799	.9518	.9022	.8273	.7282	.6119	.4888
160							.9320	.8737	.7916	.6888	.5728
180	1.0	1.0	.9997	.9986	.9953	.9868	.9687	.9361	.8845	.8117	.7198
200		1.0	.9999	.9996	.9983	.9947	.9864	.9697	.9402	.8941	.8293
220		1.0		.9999	.9994	.9980	.9944	.9863	.9708	.9442	.9029
250						.9996	.9986	.9962	.9909	.9807	.9626

TABLE I.—*Continued.*

$\frac{x}{l}$	$\frac{C_1(x)}{C_0}$	$\frac{C_2(x)}{C_0}$	$\frac{C_3(x)}{C_0}$	$\frac{C_4(x)}{C_0}$	$\frac{C_5(x)}{C_0}$	$\frac{C_6(x)}{C_0}$	$\frac{C_7(x)}{C_0}$	$\frac{C_8(x)}{C_0}$	$\frac{C_9(x)}{C_0}$	$\frac{C_{10}(x)}{C_0}$
					$k = .12$					
0	.12	.02049	.003386	.0005588	.00009223	.00001522	.0000025	0	0	0
1	.3132	.03759	.006214	.001025	.0001692	.00002794	.0000046	.0000008	0	0
2	.3908	.06335	.01118	.001875	.0003104	.00005125	.0000085	.0000014	0	0
3	.4597	.09560	.01888	.003343	.0005649	.00009387	.00001552	.0000026	0	0
4	.5208	.1325	.02965	.005692	.001005	.0001703	.00002513	.0000047	.0000008	0
5	.5750	.1727	.04358	.009175	.001724	.0003031	.00005142	.0000086	.0000014	0
6	.6230	.2148	.06059	.01402	.002832	.0005234	.00009159	.00001554	.0000026	0
7	.6657	.2580	.08051	.02041	.004450	.0007711	.0001591	.00002771	.0000047	.0000008
8	.7035	.3015	.1031	.02848	.006704	.001400	.0002683	.00004839	.0000084	.0000014
9	.7370	.3446	.1280	.03834	.009718	.002166	.0004379	.00008237	.00001472	.0000025
10	.7668	.3869	.1550	.05001	.01361	.003236	.0006918	.0001363	.00002526	.0000045
20	.9202	.7195	.4664	.2525	.1162	.04634	.01629	.005120	.001459	.0003811
30	.9788	.8867	.7206	.5143	.3215	.1771	.08679	.03818	.01521	.005540
40	.9936	.9572	.8697	.7261	.5490	.3743	.2303	.1284	.06527	.03042
50	.9981	.9845	.9437	.8605	.7330	.5767	.4168	.2765	.1686	.09487
75	.9999	.9989	.9944	.9807	.9494	.8923	.8055	.6922	.5629	.4316
100	1.0	.9999	.9995	.9979	.9931	.9813	.9575	.9163	.8538	.7696
125		1.0	1.0	.9998	.9992	.9975	.9930	.9834	.9651	.9345
150				1.0	.9999	.9997	.9990	.9973	.9935	.9857
175					1.0	1.0	.9999	.9996	.9990	.9974

of k, ranging from 0.007, 0.05, 0.07 to 0.12 are displayed successively in Figs. 3, 4, 5, and 6.

These results of $C_n(x)/C_0$ are restricted to the infinite bar (that is, terminal zone freezing neglected) and supplement data now incorporated as standard matter (2). The validity of Eqs. 10 is thus established.

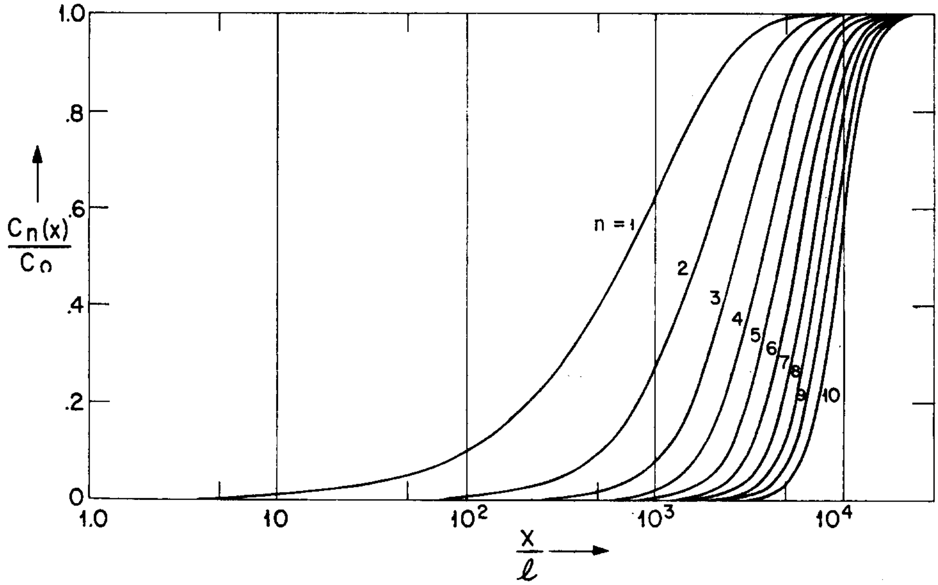

FIG. 1. Transformation of the solute distribution $C_n(x)/C_0$ by cascade zone refining. The parameter n indicates stage of purification. The case $k = 0.001$ showing approach to a limiting distribution as $n \to \infty$.

Incorporating the new findings in Eq. 4 produces

$$P_T = \frac{l}{L} + \frac{(1-k)}{L}\int_0^{L-l} e^{-(k/l)x}\cdot \sum_{p=n}^{1} A_p \frac{\left(\frac{k}{l}x\right)^{n-p}}{(n-p)!}dx \tag{11}$$

whence the purification factor may be written in the form

$$\pi_T = 1 - \frac{l}{L} + \frac{(1-k)}{L}\cdot\frac{l}{k}\left[\frac{A_nB_1}{0!} + \frac{A_{n-1}B_2}{1!} + \frac{A_{n-2}B_3}{2!} + \cdots \frac{A_1B_n}{(n-1)!}\right] \tag{12a}$$

wherein the A_n are as stated in Eq. 10c and now

$$B_n = \left(\frac{k}{l}\right)^n \int_0^{L-l} x^{n-1}e^{-(k/l)x}dx. \tag{12b}$$

CONCLUSION

The paramount objective of characterizing the zone-refinement operation by introduction of a purification factor has now been fulfilled. The investigation may be appreciated more fully by numerical

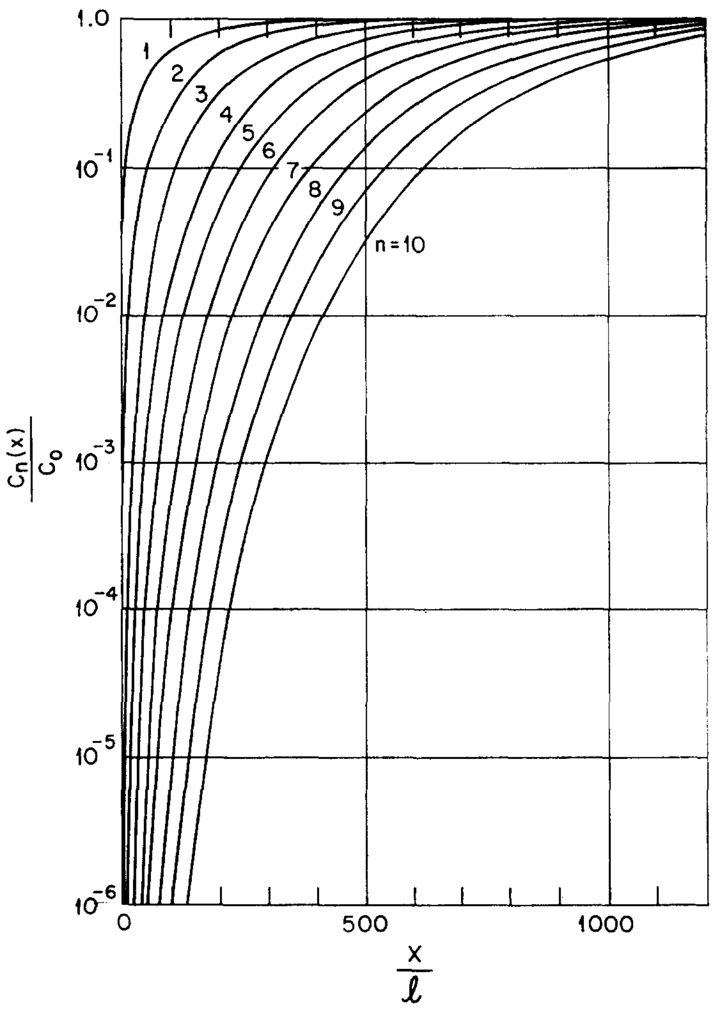

FIG. 2. The case $k = 0.001$ in the region of $\frac{x}{l} \rightarrow 0$. (Cf. Fig. 1.)

description of π_T for the values of k already embodied in the delineation of the solute distribution. Table II contains the π_T behavior for three selected values of l/L, *viz.* 1/2, 1/4 and 1/10. Finally, Figs. 7, 8 and 9 depict the expected fall-off of the purification factor with increasing passes for the gamut of k and l/L indicated.

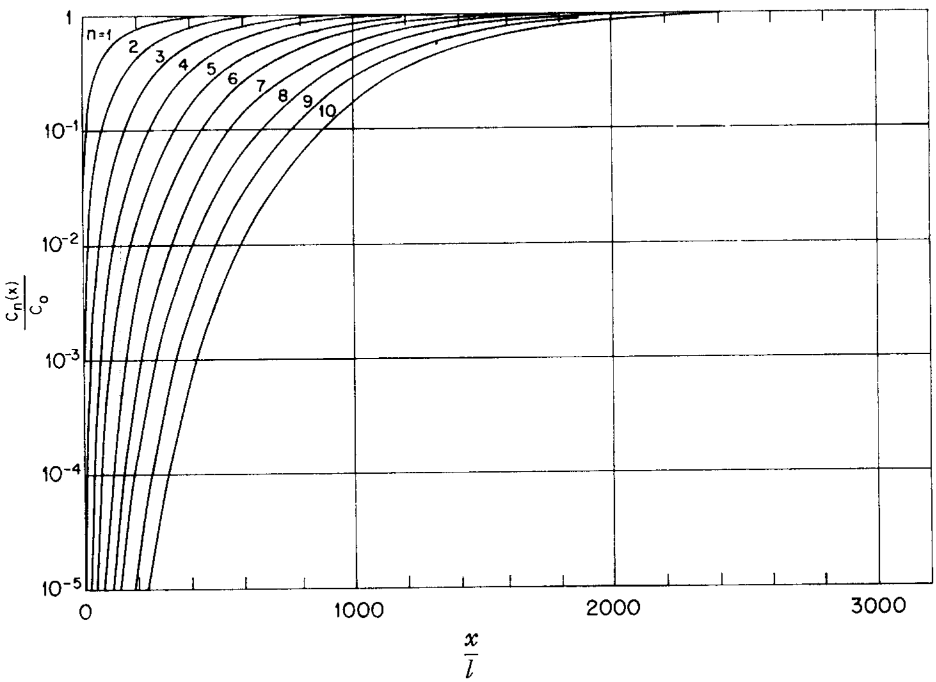

FIG. 3. The case $k = 0.007$. (Cf. Fig. 1.)

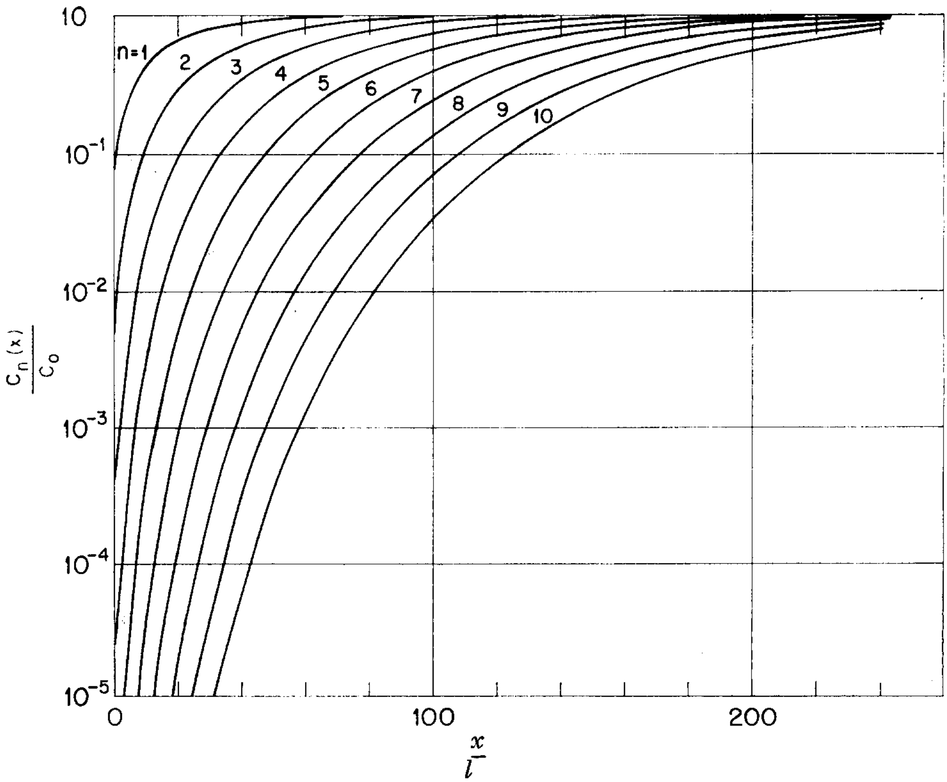

FIG. 4. The case $k = 0.05$. (Cf. Fig. 1.)

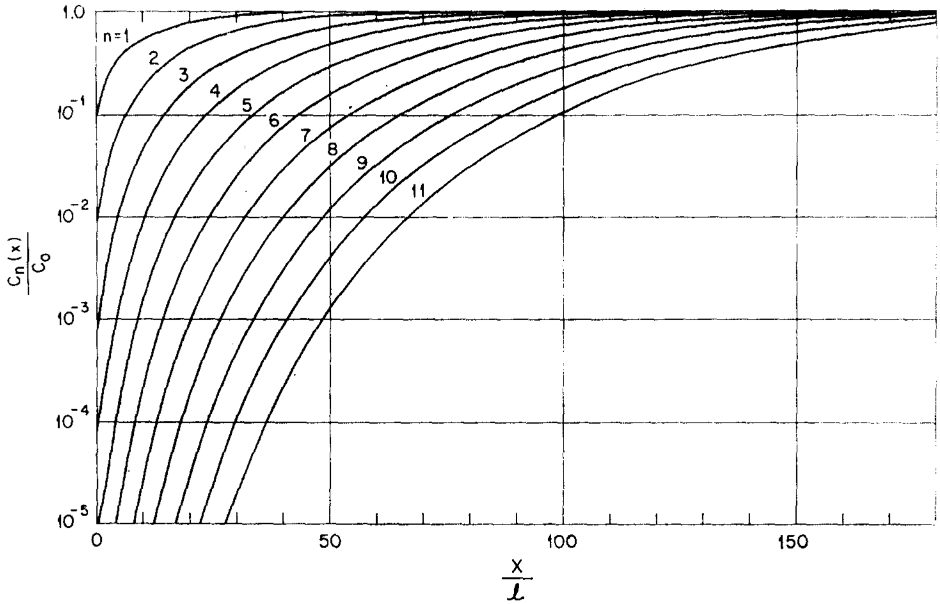

FIG. 5. The case $k = 0.07$. (Cf. Fig. 1.)

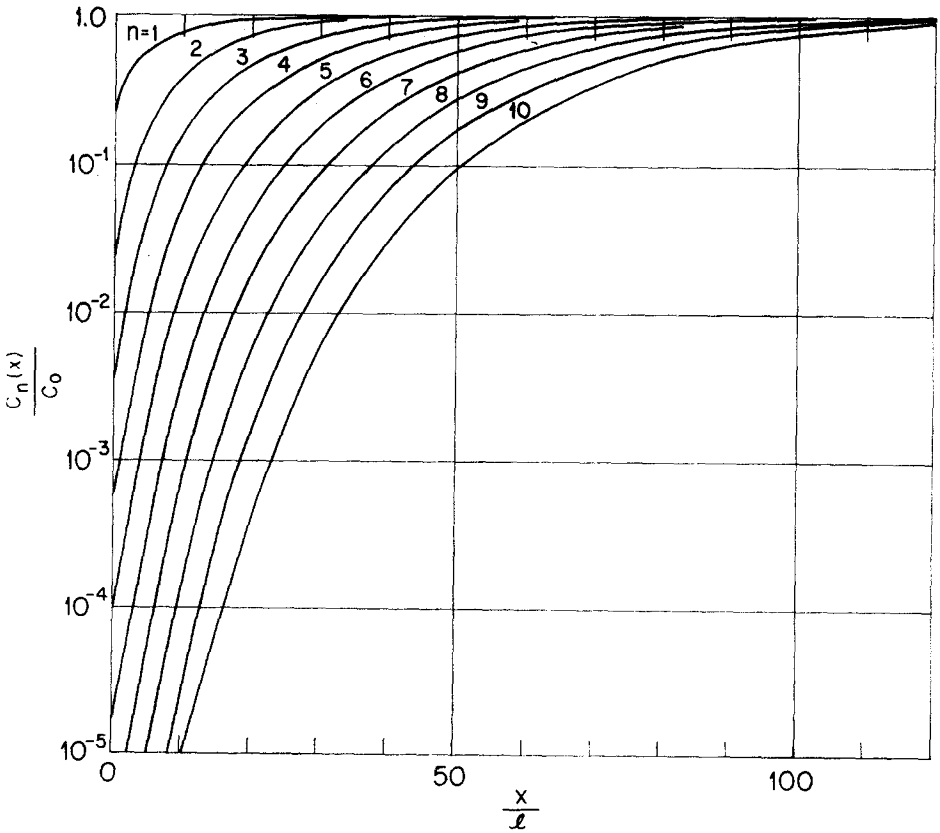

FIG. 6. The case $k = 0.12$. (Cf. Fig. 1.)

TABLE II.—π_T *vs* n *Data for* l/L = 0.5, 0.25, 0.10.

n	l/L = 0.5	l/L = 0.25	l/L = 0.10
		k = .001	
1	.0007496	.0018728	.0058298
2	.0000010	.0000034	.0000174
3	0	0	0
		k = .007	
1	.0052337	.0130154	.0338696
2	.0000528	.0001633	.00083117
3	.0000005	.0000018	.0000156
4	0	0	.0000002
5			0
		k = .05	
1	.0366795	.0883629	.2114935
2	.0025966	.0077118	.0345802
3	.0001836	.0005898	.0044488
4	.0000130	.0000428	.0004804
5	.0000009	.0000030	.0000455
6	0	.0000002	.0000039
7		0	.0000003
8			0
		k = .07	
1	.0509018	.1208691	.2790148
2	.0050049	.0145876	.0618266
3	.0004914	.9015478	.0109057
4	.0000483	.0001559	.0016245
5	.0000047	.0000154	.0002129
6	.0000005	.0000015	.0000254
7	0	.0000001	.0000028
8		0	.0000003
9			0
		k = .12	
1	.0853749	.1957399	.4157034
2	.0141092	.0392708	.1456024
3	.0023283	.0069822	.0417406
4	.0003843	.0011810	.0102595
5	.0000634	.0001963	.0022383
6	.0000105	.0000324	.0004459
7	.0000017	.0000054	.0000830
8	.0000003	.0000009	.0000148
9	0	0	.0000025
10		0	.0000004

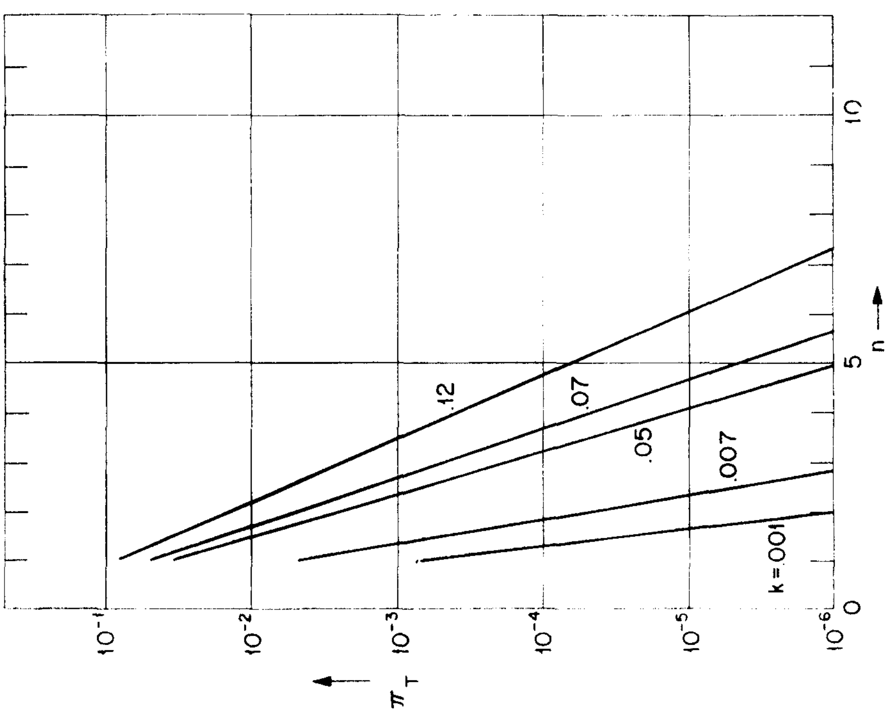

FIG. 7. The purification factor π_T after the n^{th} pass with indicated k values as a parameter. The case for $l/L = 0.5$.

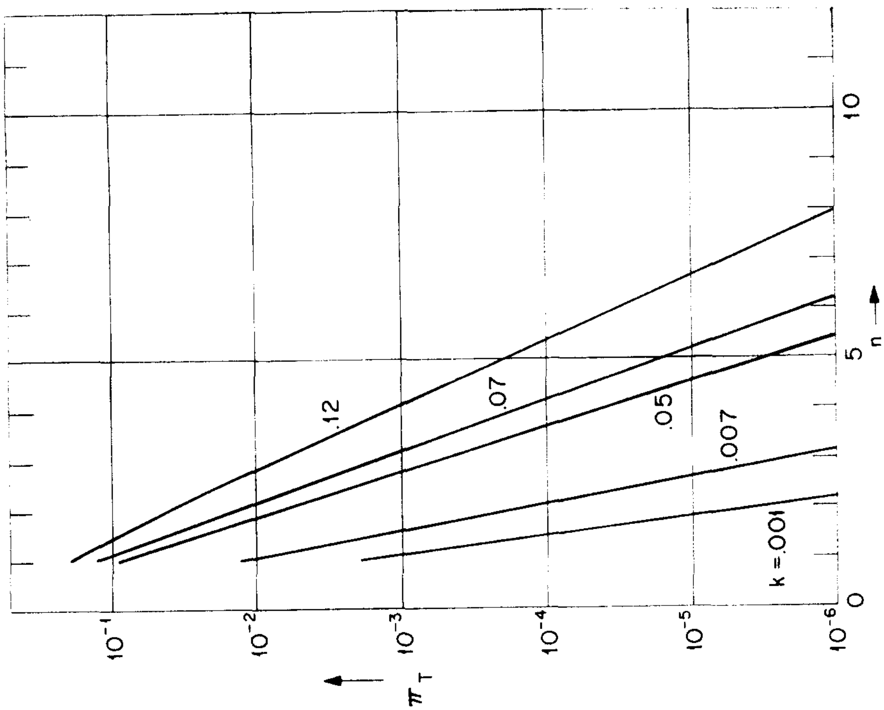

FIG. 8. The case for $l/L = 0.25$. (Cf. Fig. 7.)

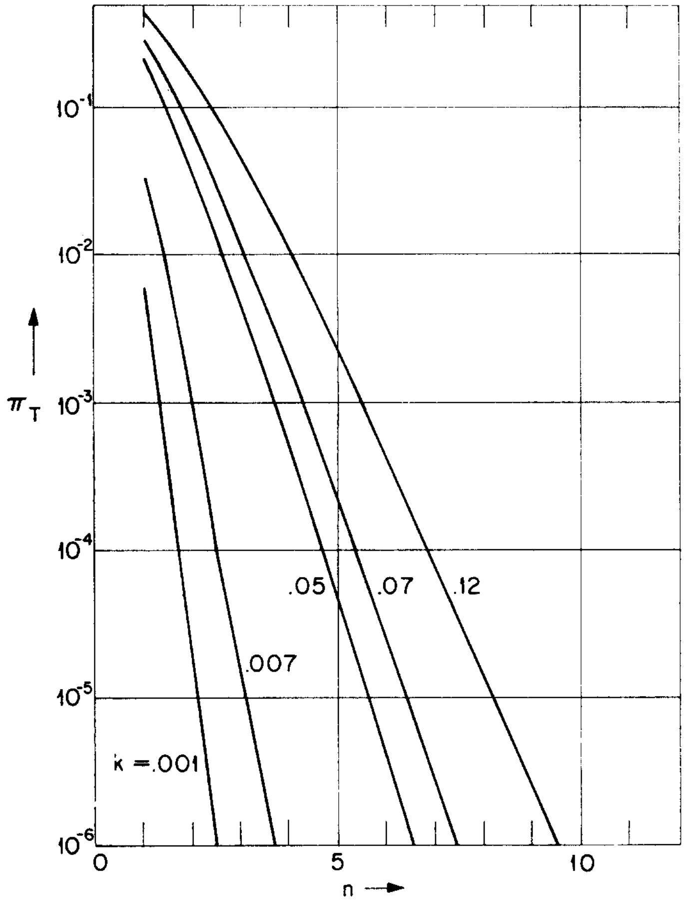

FIG. 9. The case for $l/L = 0.10$. (Cf. Fig. 7.)

APPENDIX

The $Q_n(x)$ coefficients of Eq. 10a assume successively the values:

$$Q_1 = 1$$

$$Q_2 = 2 - e^{-k} + \left(\frac{k}{l}x\right)$$

$$Q_3 = (2 - e^{-k})^2 - ke^{-k} + (2 - e^{-k})\left(\frac{k}{l}x\right) + \left(\frac{k}{l}x\right)^2 \Big/ 2!$$

$$Q_4 = (2 - e^{-k})^3 - 2ke^{-k}(2 - e^{-k})\,\frac{k^2}{2}e^{-k} + [(2 - e^{-k})^2 - ke^{-k}]\frac{k}{l}x$$
$$+ (2 - e^{-k})\left(\frac{k}{l}x\right)^2 + \left(\frac{k}{l}x\right)^3 \Big/ 3!$$

$$Q_5 = \left[(2 - e^{-k})^4 - 3ke^{-k}(2 - e^{-k})^2 - 2k^2e^{-k}(1 - e^{-k}) - \frac{k^3}{6}e^{-k}\right]$$
$$+ \left[(2 - e^{-k})^3 - 2ke^{-k}(2 - e^{-k}) - \frac{k^2}{2}e^{-k}\right]\frac{k}{l}x$$
$$+ [(2 - e^{-k})^2 - ke^{-k}]\left[\left(\frac{k}{l}x\right)^2\Big/2!\right] + (2 - e^{-k})\left(\frac{k}{l}x\right)^3\Big/3! + \left(\frac{k}{l}x\right)^4\Big/4!$$

$$Q_6 = \Big[(2 - e^{-k})^5 - 4ke^{-k}(2 - e^{-k})^3 - 6k^2e^{-k}(1 - e^{-k})^2$$
$$+ \tfrac{3}{2}k^2e^{-3k} - \tfrac{2}{3}k^3e^{-k}(1 - 2e^{-k}) - \frac{k^4}{24}e^{-k}\Big]$$
$$+ \left[(2 - e^{-k})^4 - 3ke^{-k}(2 - e^{-k})^2 - 2k^2e^{-k}(1 - e^{-k}) - \frac{k^3}{6}e^{-k}\right]\left(\frac{k}{l}x\right)$$
$$+ \left[(2 - e^{-k})^3 - 2ke^{-k}(2 - e^{-k}) - \frac{k^2}{2}e^{-k}\right]\left(\frac{k}{l}x\right)^2\Big/2!$$
$$+ [(2 - e^{-k})^2 - ke^{-k}]\left(\frac{k}{l}x\right)^3\Big/3! + (2 - e^{-k})\left(\frac{k}{l}x\right)^4\Big/4! + \left(\frac{k}{l}x\right)^5\Big/5! \qquad \text{(A-1)}$$

whence the recursion formula (10*b*) is deduced.

Applying the boundary condition at $x = 0$, there obtains from Eqs. 7 and 9

$$\frac{C_n(0)}{C_0} = \frac{k}{l}\int_0^l \frac{C_{n-1}(x)dx}{C_0} = 1 - (1 - k)A_n$$

or

$$A_n = (1 - k)^{-1}\left(1 - \frac{C_n(0)}{C_0}\right). \qquad \text{(A-2)}$$

In this manner Eqs. 10*c* and 10*d* follow with the first few coefficients identified as

$$A_1 = 1, \qquad A_2 = 2 - e^{-k}, \qquad A_3 = (2 - e^{-k})^2 - ke^{-k}$$

and

$$S_1 = e^{-k} - 1, \qquad S_2 = e^{-k}(k + 1) - 1, \qquad S_3 = e^{-k}(k^2 + 2k + 2) - 2.$$

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