ON THE CHROMATOGRAPHY OF RECIPROCAL SALT SYSTEMS*

JOHN E. GORDON

Woods Hole Oceanographic Institution, Woods Hole, Mass. (U.S.A.)

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Chromatography of a reciprocal salt system composed of monovalent cations C^1 and C^2, and anions A^1 and A^2, can produce up to four bands or spots. It is not immediately apparent without a detailed analysis of the problem, however, how many bands will be observed and what will be their relative intensities. Four spots have been reported in paper chromatography[1] and in thin-layer chromatography on silica gel G[2], the inner spots being less intense than the extreme spots in the latter case.

This report describes the results of application of distribution theory to this and a related system, and comparison of the theoretical predictions with available experimental results.

METHOD

The pertinent equilibrium expressions are

$$C^1A^1 + C^2A^2 \underset{}{\overset{K^M}{\rightleftharpoons}} C^1A^2 + C^2A^1 \qquad K^M = \frac{(C^1A^2)^M (C^2A^1)^M}{(C^1A^1)^M (C^2A^2)^M} \tag{1}$$

$$(C^1A^1)^F = K_D^{11} (C^1A^1)^M \tag{2}$$

$$(C^1A^2)^F = K_D^{12} (C^1A^2)^M \tag{3}$$

$$(C^2A^1)^F = K_D^{21} (C^2A^1)^M \tag{4}$$

$$(C^2A^2)^F = K_D^{22} (C^2A^2)^M \tag{5}$$

$$(C^1A^1)^F + (C^1A^1)^M + (C^1A^2)^F + (C^1A^2)^M = T^{11} + T^{12} \tag{6}$$

$$(C^1A^2)^F + (C^1A^2)^M + (C^2A^2)^F + (C^2A^2)^M = T^{12} + T^{22} \tag{7}$$

$$(C^2A^1)^F + (C^2A^1)^M + (C^2A^2)^F + (C^2A^2)^M = T^{21} + T^{22} \tag{8}$$

$$(C^1A^1)^F + (C^1A^1)^M + (C^2A^1)^F + (C^2A^1)^M = T^{11} + T^{21} \tag{9}$$

where $(\ )^F$ and $(\ )^M$ denote concentrations in the fixed and mobile phases and T^{ij} represents the total concentration of C^iA^j charged into a given plate before equilibration. Simultaneous solution of (1)–(5) and any three of (6)–(9) affords straightforward though involved analytical expressions for the eight variables $(C^iA^j)^F$ and $(C^iA^j)^M$.

Plate theory computations for this system were carried out using a General Electric 225 computer. The first cells of banks of equal volume cells of the two phases were opposed, one mole each of C^1A^1 and C^2A^2 or C^1A^2 and C^2A^1 was charged into the first cell of the fixed phase, the equilibrium values of $(C^iA^j)^F$ and $(C^iA^j)^M$ were computed, the mobile bank of cells was advanced to the next plate, new T^{ij} and then new (C^iA^j) were computed for each cell and so on until the requisite number of plates had been equilibrated.

This model corresponds to a partition chromatogram in which the rate of attainment of equilibrium is large relative to the rate of flow of the mobile phase. The above formulation of the distribution equilibria assumes that ion pairs are the only important species in both phases.

The computation routine described, when applied to pure, noninteracting components, produced highly symmetrical peaks in 50-plate chromatograms. The reciprocal salt system results obtained invariably satisfied the symmetry and mass balance requirements of the system ($\Sigma(C^1A^1) = \Sigma(C^2A^2)$, $\Sigma(C^1A^2) = \Sigma(C^2A^1)$, $\Sigma(C^1A^1) + \Sigma(C^1A^2) = 1$, the summation, here and elsewhere in this paper, extending over all plates of the chromatogram) and the output peak maxima appeared at the R_F values required by the values of K_D^{ij} read into each run.

RESULTS

The results for 23 runs are given in Table I. Fig. 1 illustrates the course of development of a typical chromatogram. Fortunately, a 200-plate computation (the practical limit in the present work) is sufficient to resolve completely the left and right halves of the chromatogram, though not the individual components of
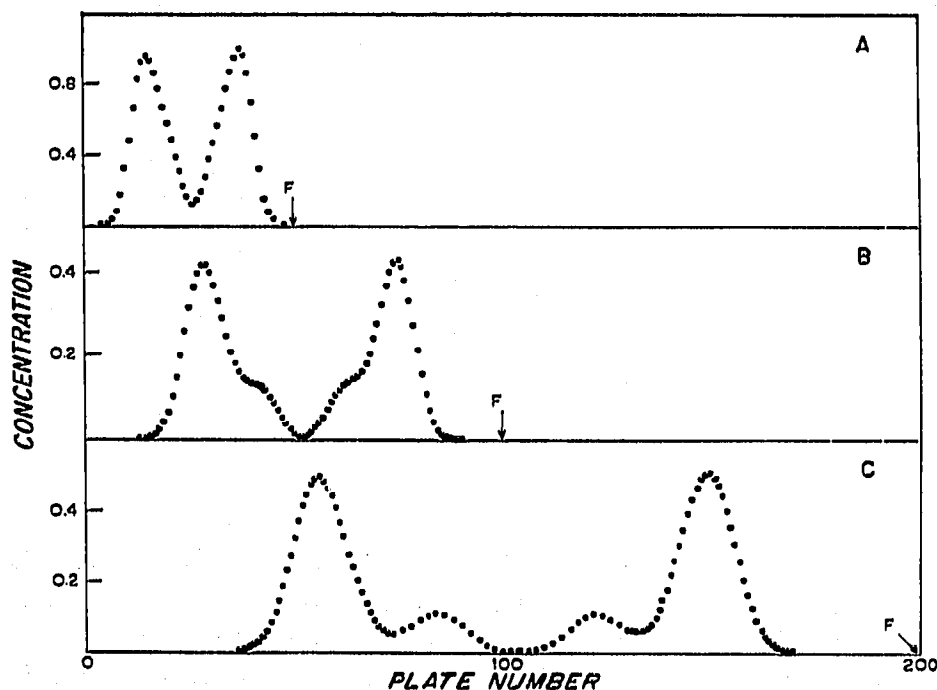

Fig. 1. Partition chromatogram of the system $C^1A^1 + C^2A^2 = C^1A^2 + C^2A^1$, with $R_F^{11} = 0.2727$, $R_F^{12} = 0.4286$, $R_F^{21} = 0.6000$, $R_F^{22} = 0.7500$ and $K^M = K^F = 1$. After (A) 50; (B) 100; (C) 200 plates (runs 8, 10, 11 of Table I). F marks the solvent front.

TABLE I

CHROMATOGRAMS OF THE SYSTEM C^1A^1–C^1A^2–C^2A^1–C^2A^2 COMPUTED BY DISTRIBUTION THEORY

Run	N^*	KM	R_F^{11}	R_F^{12}	R_F^{21}	R_F^{22}	$\dfrac{K^{12}K^{21}}{K^{11}K^{22}}$	$\dfrac{R_F^{21}-R_F^{12}}{R_F^{22}-R_F^{11}}$	$(X^{12}+X^{21})$ cor.	$X^{12}+X^{21}$
1	50	1	0.2500	0.2625	0.7375	0.7500	1.000	0.950	0.4778	0.4779
2	50	1	0.2500	0.2750	0.7250	0.7500	1.000	0.900	0.4552	0.4554
3	50	1	0.2500	0.3000	0.7000	0.7500	1.000	0.800	0.4082	0.4085
4	50	1	0.2500	0.3500	0.6500	0.7500	1.000	0.600	0.3073	0.3084
5	50	1	0.2500	0.4500	0.7500	0.7500	0.407	0.600	0.2762	0.2777
6	50	1	0.2500	0.3845	0.6000	0.7500	1.068	0.431	0.2193	0.2226
7	25	1	0.2727	0.4286	0.6000	0.7500	1.000	0.359	0.1812	0.2275
8	50	1	0.2727	0.4286	0.6000	0.7500	1.000	0.359	0.1802	0.1876
9	75	1	0.2727	0.4286	0.6000	0.7500	1.000	0.359	0.1774	0.1787
10	100	1	0.2727	0.4286	0.6000	0.7500	1.000	0.359	0.1756	0.1759
11	200	1	0.2727	0.4286	0.6000	0.7500	1.000	0.359	0.1738	0.1738
12	50	1	0.4286	0.6000	0.7500	0.8571	1.000	0.350	0.1815	0.1921
13	50	1	0.2727	0.4286	0.6000	0.8333	1.667	0.306	0.1564	0.1595
14	50	1	0.2727	0.4545	0.5555	0.7500	1.079	0.212	0.1053	0.1221
15	50	1	0.2500	0.4545	0.5555	0.8000	1.278	0.184	0.0867	0.0944
16	100	1	0.2500	0.4545	0.5555	0.8000	1.278	0.184	0.0785	0.0789
17	50	1	0.2500	0.7143	0.7692	0.8333	0.200	0.094	0.0361	0.0483
18	50	1	0.2727	0.5000	0.5000	0.7500	1.122	0.000	0.0000	0.0737
19	100	1	0.2727	0.5000	0.5000	0.7500	1.122	0.000	0.0000	0.0251
20	50	1	0.2500	0.2500	0.7500	0.7500	1.000	1.000	1.0000	1.0000
21	50	2	0.2727	0.4286	0.6000	0.7500	1.000	0.359	0.2376	0.2517
22	50	5	0.2727	0.4286	0.6000	0.7500	1.000	0.359	0.3255	0.3574
23	50	50	0.2727	0.4286	0.6000	0.7500	1.000	0.359	0.5503	0.6601

* N = Number of plates.

each wing. Such four-peak chromatograms are produced in each case where the four salts possess different K_D values. Fig. 1C bears a close resemblance to the thin-layer chromatograms previously observed[2].

Table I reveals that the intensity distribution between outer and inner peaks, measured conveniently by $X^{12} + X^{21} = [\Sigma(C^1A^2) + \Sigma(C^2A^1)]/2$, is a function of the set of K_D values of the four salts. In working out this dependence (and that on K^M) it was necessary for the sake of economy to work principally with 50-plate runs. Fig. 2A shows that complete resolution about the center of symmetry of the chromatogram is achieved at about 200 plates, and that $X^{12} + X^{21}$ does not become constant
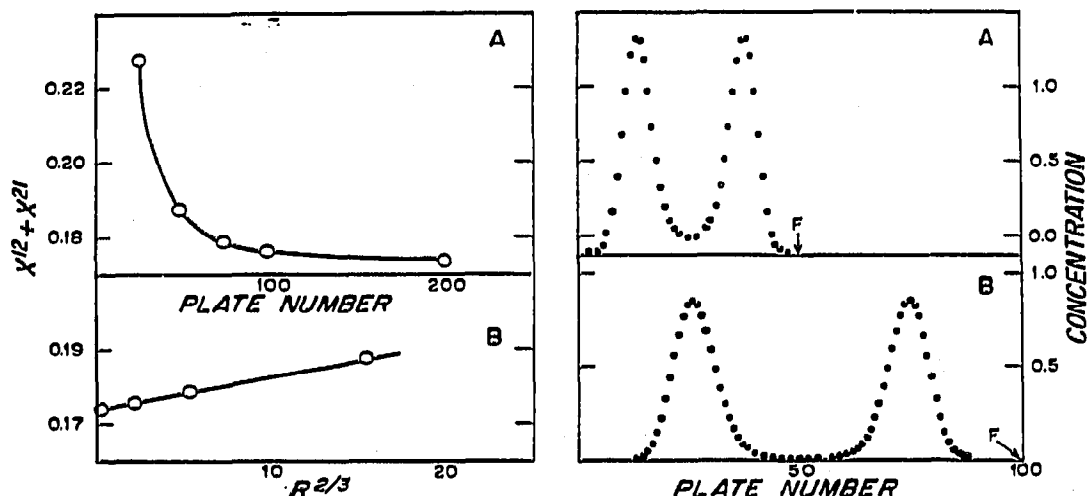

Fig. 2. Characteristics of the chromatograms of Fig. 1. $X^{12} + X^{21}$ (A) vs. number of plates; (B) vs. a function of the cross-hatched area of Fig. 4.

Fig. 3. Partition chromatogram of the system $CA^1_2 + CA^2_2 = CA^1A^2$, with $R_F^{11} = 0.25$, $R_F^{12} = 0.50$, $R_F^{22} = 0.75$ after (A) 50, (B) 100 plates.

until this point is reached. (Redistribution of material *within* the right and left halves of the chromatogram according to eqn. (1) can take place only through a region of overlap *between* the right and left halves.) Fig. 2B uses a convenient extrapolation function to show that $X^{12} + X^{21}$ for the 200-plate chromatogram differs negligibly from that for complete resolution; the form of Fig. 1C is thus quantitatively correct. The 50-plate chromatograms were corrected approximately for the incomplete resolution by attributing the overlap area R (see Fig. 4A where the composition of a 50-plate peak is illustrated and R is shown as the cross hatched area) to the outer maxima, *i.e.* $(X^{12} + X^{21})$cor. $= 0.5\ \Sigma\ |(C^1A^2)^M + (C^1A^2)^F - (C^2A^1)^M - (C^2A^1)^F|$. This gave $X^{12} + X^{21}$ within about 3 % of the 200-plate value and this was considered acceptable. Material redistribution in the developing band substantially reduces the rate of resolution in these chromatograms in comparison with the separation of noninteracting substrates with the same R_F difference, as is shown in Fig. 4B.

$X^{12} + X^{21}$ proves to be, to a close approximation, proportional to the quantity $(R_F^{21} - R_F^{12})/(R_F^{22} - R_F^{11})$ as illustrated in Fig. 5*. There is a further minor de-

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* Here and elsewhere in this article the ions are so labelled that $R_F^{11} < R_F^{12} < R_F^{21} < R_F^{22}$; experimentally, of course, systems with $R_F^{21} < R_F^{12}$ are also observed[2], but the form of the chromatogram is not changed by this inversion.
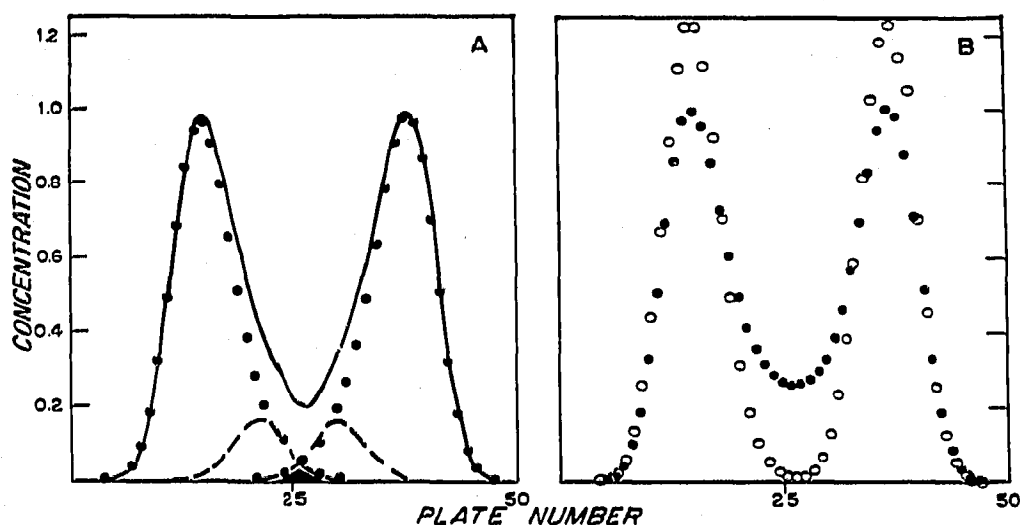
Fig. 4. Characteristics of chromatograms of the type of Fig. 1. (A) Breakdown of individual components for run 8 of Table I: ●, C^1A^1 and C^2A^2; – – –, C^1A^2 and C^2A^1; ———, total. (B) Comparison of run 18 (solid circles) with a chromatogram computed for two pure, noninteracting substances (open circles) having R_F values equal to those of C^1A^1 and C^2A^2 in run 18. (Note that run 18 is the situation in which the reciprocal system eventually resolves into only two peaks).

pendence on the symmetry of the K_D^{ij} values about unity, with $K_D^{12}K_D^{21}/K_D^{11}K_D^{22} > 1$ producing positive deviations in $X^{12} + X^{21}$ and *vice versa*. This is considered in a later section. In any case the principal intensity distribution is given by

$$X^{12} + X^{21} = 0.50\ (R_F^{21} - R_F^{12})/(R_F^{22} - R_F^{11})$$ (10)

with an average deviation of 0.014 in $X^{12} + X^{21}$. Thus the chromatogram collapses to two spots only in the limit of $K_D^{12} = K_D^{21}$, and it approaches four spots of equal intensity only in the limit $K_D^{12} \longrightarrow K_D^{11}$, $K_D^{21} \longrightarrow K_D^{22}$. The point at $R_F^{11} \equiv R_F^{12}$, $R_F^{21} \equiv R_F^{22}$ is idiosyncratic, computation predicting (run 20) two peaks of pure C^1A^2 and C^2A^1, *i.e.* $X^{12} + X^{21} = 1$ rather than 0.5 as predicted by eqn. (10). The value $X^{12} + X^{21} = 0$ at $R_F^{12} = R_F^{21}$ was difficult to demonstrate directly with precision. Since this point represents the minimum rate of resolution (Fig. 4B) a 100-plate run still shows an appreciable $X^{12} + X^{21}$ value (Table I). However, extra-
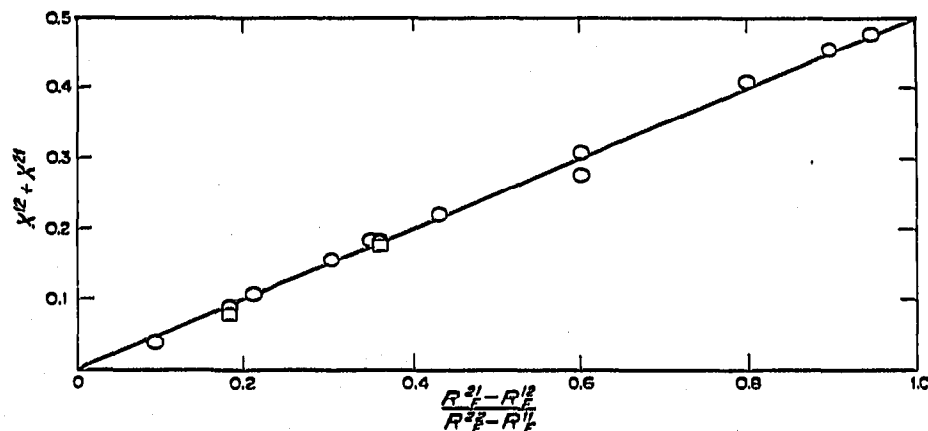

Fig. 5. Plot of $X^{12} + X^{21}$ *vs.* $(R_F^{21} - R_F^{12})/(R_F^{22} - R_F^{11})$ according to eqn. (10).

polation of the 50- and 100-plate results, the general form of the $X^{12} + X^{21}$ vs. $(R_F{}^{21} - R_F{}^{12})/(R_F{}^{22} - R_F{}^{11})$ dependence, and the analogy with the $CA^1{}_2$-$CA^2{}_2$-CA^1A^2 system discussed in the next section together leave little doubt regarding this point.

The system $CA^1{}_2$-$CA^2{}_2$-CA^1A^2

It is instructive to consider next the system of a bivalent cation with two mono-valent anions for which eqn. (11) together with five equations analogous to (2)–(9) have been employed

$$CA^1{}_2 + CA^2{}_2 \rightleftharpoons 2\ CA^1A^2 \tag{11}$$

in computations analogous to those of Table I. As Fig. 3 and Table II illustrate, this system does not give three spots, but only those due to $CA^1{}_2$ and $CA^2{}_2$. Clearly this occurs, just as in the case of C^1A^1-C^1A^2-C^2A^1-C^2A^2 with $K_D{}^{12} = K_D{}^{21}$, because the two central ion pairs ($2\ CA^1A^2$ or $C^1A^2 + C^2A^1$) remain physically superposed and are thus progressively converted via eqn. (1) or eqn. (11) to $CA^1{}_2 + CA^2{}_2$ or $C^1A^1 + C^2A^2$. With the rest of the systems of Table I on the other hand, C^1A^2 and C^2A^1, having different R_F values, disengage physically before they are wholly consumed.

TABLE II

CHROMATOGRAMS OF THE SYSTEM $CA^1{}_2$-$CA^2{}_2$-CA^1A^2 COMPUTED ACCORDING TO DISTRIBUTION THEORY

N^*	K^M	$R_F{}^{11}$	$R_F{}^{12}$	$R_F{}^{22}$	X^{12} **
50	I	0.25	0.50	0.75	0.0176
100	I	0.25	0.50	0.75	0.0074

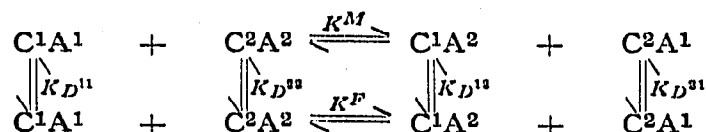* N = number of plates.
** X^{12} = mole fraction of CA^1A^2.

The quotient $K_D{}^{12}K_D{}^{21}/K_D{}^{11}K_D{}^{22}$

If one writes the analog of eqn. (1) for the fixed phase

$$K^F = \frac{(C^1A^2)^F\ (C^2A^1)^F}{(C^1A^1)^F\ (C^2A^2)^F} \tag{12}$$

it is clear that the partition equilibrium

$$C^1A^1 \quad + \quad C^2A^2 \underset{K^M}{\rightleftharpoons} C^1A^2 \quad + \quad C^2A^1$$

$$\updownarrow K_D{}^{11} \qquad\quad \updownarrow K_D{}^{22} \quad\ \underset{K^F}{\rightleftharpoons} \quad \updownarrow K_D{}^{12} \qquad\quad \updownarrow K_D{}^{21}$$

$$C^1A^1 \quad + \quad C^2A^2 \rightleftharpoons C^1A^2 \quad + \quad C^2A^1$$

is overdetermined and that

$$\frac{K^F}{K^M} = \frac{K_D{}^{12}K_D{}^{21}}{K_D{}^{11}K_D{}^{22}} \tag{13}$$

The situation $K_D{}^{12}K_D{}^{21}/K_D{}^{11}K_D{}^{22} = 1$ occupies a special position as the only case where K^F and K^M can both the unity, as would occur in the absence of ion association. This is the case corresponding to additivity of ionic contributions to the free energy of transport of C^iA^j from one phase to the other[3].

This is why eqn. (10) is strictly valid only for $K_D{}^{12}K_D{}^{21}/K_D{}^{11}K_D{}^{22} = K^F = K^M = 1$, since otherwise, eqn. (1) and eqn. (12) will in general bias the material distribution of the chromatogram somewhat. If the residual error due to incomplete resolution is kept in mind, this bias is in the right direction to qualitatively account for the deviations in Fig. 5; quantitative corrections would be difficult to compute.

In most chromatographic systems ion association will be present, and $K_D{}^{12}K_D{}^{21}/K_D{}^{11}K_D{}^{22}$ values different from unity are expected. Data for thin-layer chromatography of quaternary ammonium iodides, bromides, nitrates and perchlorates[2] on silica gel G in chloroform–ethanol (24:1) provide 31 reciprocal salt systems for which the distribution of $K_D{}^{12}K_D{}^{21}/K_D{}^{11}K_D{}^{22}$ values is given in Table III.

TABLE III

DISTRIBUTION OF EXPERIMENTAL $K_D{}^{12}K_D{}^{21}/K_D{}^{11}K_D{}^{22}$ VALUES

$K_D{}^{12}K_D{}^{21}/K_D{}^{11}K_D{}^{22}$ *	Number of cases **
0.90–0.94	4
0.95–0.99	5
1.00–1.04	7
1.05–1.09	7
1.10–1.14	3
1.15–1.20	5

* Assigned such that $K_D{}^{11} > K_D{}^{12} > K_D{}^{21} > K_D{}^{22}$.
** Data from ref. 2.

It is of interest to attempt to account for the values of $K_D{}^{12}K_D{}^{21}/K_D{}^{11}K_D{}^{22}$ for these reasonably simple, near-spherical ions on the basis of simple electrostatic considerations. For a reciprocal ion pair equilibrium of the type

$$C^{large}A^{large} + C^{small}A^{small} \overset{K}{\rightleftharpoons} C^{large}A^{small} + C^{small}A^{large} \tag{14}$$
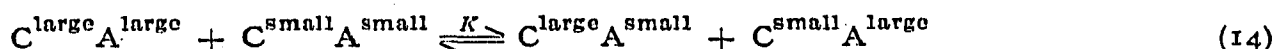
where the superscripts refer to the ionic radii, it is possible to demonstrate, assuming only that the distances of closest approach are sums of constant ionic contributions, that the electrostatic contribution to eqn. (14) is such as to favor $C^{large}A^{large} + C^{small}A^{small}$, i.e. make $K < 1$*. Increase in the dielectric constant of the medium will then reduce in magnitude the electrostatic energies and make K approach unity, i.e. increase K. If one makes the reasonable assumption that the fixed phase in these thin-layer chromatograms is the more polar phase, this requires that

$$\frac{K^F}{K^M} = \frac{K_D{}^{12}K_D{}^{21}}{K_D{}^{11}K_D{}^{22}} > 1$$

* This problem reduces to the following relations among the distances of closest approach: $a < b < c < d$ and $a + d = b + c$, from which it is readily shown that $(1/a + 1/d) > (1/b + 1/c)$. Since the electrostatic energy of the ion pair is proportional to $1/a$ etc., this is a sufficient condition for the above rule.

when $C^1A^1 = C^{large}A^{large}$, $C^1A^2 = C^{large}A^{small}$, $C^2A^1 = C^{small}A^{large}$ and $C^2A^2 = C^{small}A^{small}$. Thus, if the ion pairs are labelled (as throughout this paper) with C^1A^1 and C^2A^2 the slowest and fastest species, respectively, the following should hold

$$K_D{}^{12}K_D{}^{21}/K_D{}^{11}K_D{}^{22} > 1$$

if the slowest- and fastest-migrating ion pairs are $C^{large}A^{large}$ and $C^{small}A^{small}$ and

$$K_D{}^{12}K_D{}^{21}/K_D{}^{11}K_D{}^{22} < 1$$

if the slowest and fastest ion pairs are $C^{large}A^{small}$ and $C^{small}A^{large}$.

The test of this proposition is shown in Table IV.

In 24 of 29 cases (83 %) the prediction is correct. Furthermore, four of the five contrary cases involve systems where the two quaternary ammonium cations differ by only one CH_2 group in a 25-carbon atom ion, and the difference in R_F values is in fact not greater than their experimental uncertainties. The apparent success of this

TABLE IV

TEST OF THE ELECTROSTATIC MODEL FOR ESTIMATION OF $K_D{}^{12}K_D{}^{21}/K_D{}^{11}K_D{}^{22}$

	Number of cases*	
	R_F extremes are C^{large} A^{large} and $C^{small}A^{small}$**	R_F extremes are $C^{large}A^{small}$ and $C^{small}A^{large}$**
$K > 1$	18	2
$K < 1$	3	6

   * Of the 31 systems, two had $K_D{}^{12}K_D{}^{21}/K_D{}^{11}K_D{}^{22} = 1.0$.
   ** Ionic radii used (Å): I-, 2.16; ClO$_4$-, 2.00; Br-, 1.95 (GRUNWALD, BAUGHMAN AND KOHN-STAM[4]); NO$_3$-, 1.75–1.85 (PAULING[5] and MESCHEL AND KLEPPA[6]).

simple model implies that the forces determining K^F and K^M are predominantly electrostatic and that the extrapolation of distribution phenomena to treat the thin-layer chromatograms (which are in large part adsorption chromatograms) is a usable one.

It is not surprising that (Tables III and IV) the cases of coincidence of ion pair radius–sum–and chromatographic mobility–extremes outnumber the cases of non-coincidence nearly three fold (21:8). In every one of the coincident cases, moreover, $C^{large}A^{large}$ is the fastest, $C^{small}A^{small}$ the slowest species. Thus the fastest species are distributed among the types $C^{large}A^{large}$, $C^{large}A^{small}$, $C^{small}A^{large}$ and $C^{small}A^{small}$ in the ratio 21:8:0:0, reflecting the expected preference of the larger ions for the less polar phase[7], as well as a predominance of the influence of the cation.

The magnitude of K^M

Table I (runs 8, 21–23) gives the variation in $X^{12} + X^{21}$ produced by variation of the input value of K^M. The generalizations of the preceding paragraph imply that the most common deviation from eqn. (10) will be that in which the extreme peaks are enriched at the expense of the inner peaks. Quite sizeable values of K^M can arise

from purely electrostatic contributions. For instance a system with ionic radii of 1, 1, 5 and 5 Å (perhaps the most extreme case involving relatively simple ions) in a medium of dielectric constant $= 10$ generates $K^M = 2 \cdot 10^{-7}$. On the other hand, the systems of Tables III and IV probably have $0.8 \leqslant K^M \leqslant 1.0$. It is interesting to note that values of K (eqn. 14) obtained by GRISWOLD, JONES AND BIRDWHISTELL[8] from salt effects on salt solubilities in acetic acid can be accounted for rather well by the electrostatic model. For $Li^+-K^+-Br^--acetate^-$ (or formate$^-$) they report 0.136 (0.178) vs. 0.13 calculated taking the effective radius of RCO_2^- equal to that of hydroxide ion; similarly for $Na^+-K^+-Br^--acetate^-$ (or formate$^-$), 0.621 (0.556) vs. 0.42 calculated on the same basis.

With more complicated, perhaps polyfunctional ions, other forces can influence K^M, for example electron or proton donor–acceptor interaction. Representing acceptor and donor properties by the subscripts a and d, the system

$$C^1{}_a A^1{}_d + C^2{}_d A^2{}_a \underset{\xrightarrow{K^M}}{\rightleftharpoons} C^1{}_a A^2{}_a + C^2{}_d A^1{}_d$$
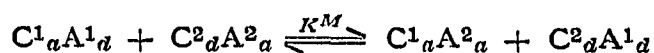
could clearly have $K^M \simeq 10^{-4}$ from this source alone.

SUMMARY

A theoretical plate treatment of the partition chromatography of a reciprocal salt system has been carried out with the aid of a high-speed digital computer, and the results have been compared with the available experimental information. The following points were established.

1. The system of univalent cations C^1 and C^2 and univalent anions A^1 and A^2 produces, in general, four bands whose relative magnitudes are a function of the R_F values of the four possible ion pairs. The distribution of material when $K_D{}^{12} K_D{}^{21} / K_D{}^{11} K_D{}^{22} = 1$ is given to a good approximation by

$$X^{12} = X^{21} = 0.25 \; (R_F{}^{21} - R_F{}^{12})/(R_F{}^{22} - R_F{}^{11})$$

where X^{11}, $R_F{}^{11}$ and $K_D{}^{11}$ are the mole fraction, R_F value and partition coefficient of the slowest species $C^1 A^1$ etc.

2. The 2:1 electrolyte system $C^{++}-(A^1)^--(A^2)^-$ produces no band corresponding to CA^1A^2.

3. For simple ions in media in which the only important species are ion pairs, the magnitudes of the equilibrium constant K for $C^1 A^1 + C^2 A^2 = C^1 A^2 + C^2 A^1$ can be estimated reliably from simple electrostatic considerations. The ratio of K values for the fixed and mobile phases of a partition chromatogram gives the important quotient $K_D{}^{12} K_D{}^{21} / K_D{}^{11} K_D{}^{22}$ whose deviation from unity can be qualitatively predicted from a knowledge of the ionic radii and the relative polarity of the two phases.

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