

PAPER-ELECTROPHORETIC STUDIES OF THE SOLUTION  
CHEMISTRY OF RaD ( $^{210}\text{Pb}$ ), RaE ( $^{210}\text{Bi}$ ) AND RaF ( $^{210}\text{Po}$ )

## I. IN HYDROCHLORIC ACID

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In general, the principal types of methods for isolation and purification for obtaining carrier-free tracers are<sup>1</sup>: (1) coprecipitation with nonisotopic carrier; (2) ion-exchange adsorption and elution; (3) solvent extraction; and (4) volatilization or distillation. Paper chromatography was relatively recently added to this list<sup>2-5</sup> and it has been often recommended as especially suitable for purposes of purification when it is desired to obtain the element free from other radioactive species for the study of its nuclear properties<sup>3-5</sup>. The decomposition products formed by the action of  $\alpha$ -rays on the organic solvents often used in paper chromatographic methods make the latter unsuitable for the purification of  $\alpha$ -emitting elements<sup>6</sup>. Although the outstanding advantages of paper electrophoresis over the ordinary wet-processes of separation have already been emphasized<sup>7-14</sup>, this technique has found still less application as a means of separating and studying the solution chemistry of an element at a trace level. Thus, for example, to the knowledge of the authors the only electrophoretic separation of RaD, RaE and RaF that has been mentioned is that of STRAIN and coworkers<sup>1,16</sup>, who reported that the separation of the two last mentioned elements is incomplete in the electrolyte 0.1 *N* lactic acid. LEDERER<sup>4</sup> studied the movement of RaF in 0.1, 0.05 and 0.025 *N* hydrochloric acid. Although the great tendency of polonium to form complexes, even with anions that do not readily form complexes, has been pointed out<sup>17</sup>, there is no *a priori* reason why this property has not been utilized with advantage for the separation of polonium from its parent elements using inorganic solvents. The present scheme of investigation was undertaken principally to study the solution chemistry of RaD, RaE and RaF in different mineral acids by paper electrophoresis, in order to establish conditions which favour the separation of these elements. The present paper reports the results obtained with hydrochloric acid in the concentration range 0.0025 *N* to 8 *N*.

## EXPERIMENTAL

The glass-plate technique, as described by LEDERER AND WARD<sup>18</sup>, was used with Arches No. 302 paper strips ( $2.7 \times 40$  cm). Since the paper strip was attacked by the acid at higher concentrations and became fragile, it could not be used beyond 3 *N*. The displacement in electrolytes of higher concentrations was therefore investigated

on Whatman glass-paper (GF/A) strips ( $2 \times 40$  cm). The paper strip was soaked with the electrolyte by drawing the strip through it, after which the excess of liquid was removed by blotting the strip between filter papers. The solution of RaD, RaE and RaF in radioactive equilibrium (stored in  $2.5 N$   $HNO_3$  and diluted ten times) was applied by a micropipette at a previously marked point on the paper. Then the paper was sandwiched between two 5 mm thick glass plates ( $8 \times 30$  cm) pressed against each other by four clips and the assembly was placed on two similar electrode vessels containing the same volume of the electrolyte. The free ends of the paper dipped well into the electrolyte. The electrophoretic movement was studied in hydrochloric acid the concentration of which was varied in the range  $0.0025 N$  to  $8 N$ . A potential difference of 300 V D.C. was applied to the two platinum wire electrodes dipping in the electrode vessels. After an electrophoretic run of 45 min the voltage was switched off, the paper taken out and dried by a current of warm air from a hair-dryer. When relatively concentrated acid was used as the electrolyte, the acid on the paper was neutralized before counting by exposing the paper to ammonia in a closed vessel.

RaE and RaF could be located on the paper just after the electrophoresis by scanning the electrophoregram using the automatic scanner Frieske-Hoepfner FH 452. The paper passed at a speed of 3 cm/min under a 0.5 mm wide slit below the thin window counter. The position of polonium was distinguished from that of bismuth by enclosing the band between two cellophane bands before passing it under the counter, by which means all the  $\alpha$ -rays from RaF were cut off by the cellophane and only the position of RaE was recorded. On the fresh electrophoregram, RaD was not observed. It could be detected very well by scanning the electrophoregram after about five days, owing to the growth of its  $\beta$ -emitting daughter RaE.

#### RESULTS AND DISCUSSION

Table I summarises the paper electrophoretic displacement of RaD, RaE and RaF in hydrochloric acid of various concentrations in the range  $0.0025 N$  to  $8.0 N$ . Since no attempt was made to run the electrophoresis at a given temperature, which is an important factor in the determination of the migration rate of an ion, the results reported here are chiefly of qualitative and analytical interest. Fig. 1 gives a typical electrophoregram. All three elements are cationic in dilute hydrochloric acid and become anionic, due to complexation with  $Cl^-$ , in relatively concentrated acid. This change from cation to anion takes place for polonium at a lower concentration of HCl than that for bismuth, while the concentration of HCl required to make lead anionic is higher than  $0.75 N$ . Thus, the isoelectric concentration lies between  $0.01 N$  and  $0.02 N$  for RaF, between  $0.3 N$  and  $0.4 N$  for RaE, and between  $0.75 N$  and  $0.85 N$  for RaD. This is in agreement with the tendency of these elements to form complexes and the charge carried by the central metal. RaD exists as cation in the concentration range  $0.0025 N$  to  $0.75 N$ , RaE in the concentration range  $0.0025 N$  to  $0.03 N$  and RaF in the concentration range  $0.0025 N$  to  $0.01 N$ . The maximum cationic displacement observed was only 2.4 mm for RaF, while for RaE it was 5.4 mm and for RaD as high as 56.4 mm. Although RaD exists as anion in hydrochloric acid at concentrations higher than  $0.85 N$ , RaE and RaF begin to move anionically from  $0.4 N$  and  $0.02 N$  respectively. It should be noted here that according to the results of PANETH AND BENJAMIN<sup>19</sup>, the anionic migration of polonium in hydrochloric acid is

TABLE I  
PAPER ELECTROPHORETIC DISPLACEMENT OF RaD, RaE AND RaF  
IN HYDROCHLORIC ACID MEDIUM

Normality of the acid	Displacement (mm)		
	RaD	RaE	RaF
0.0025	-12.0	-2.1	-2.1
0.005	-30.0	-2.2	-2.2
0.0075	-33.0	-2.4	-2.4
0.01	-43.2	-2.4	-2.4
0.02	-51.9	-3.75	+1.2
0.03	-50.0	-5.4	+3.0
0.04	-56.4	+1.5	+12.6
0.05	-53.4	+3.6	+15.9
0.10	-52.0	+9.6	+43.8
0.15	-48.1	+19.2	+62.4
0.20	-37.5	+31.2	+70.5
0.25	-35.4	+40.8	+80.4
0.30	-33.3	+42.0	+83.4
0.35	-23.4	+54.0	+91.2
0.40	-16.8	+63.6	+100.8
0.45	-16.2	+60.9	+94.8
0.50	-15.6	+58.5	+89.1
0.60	-8.4	+58.1	+85.5
0.75	-1.0	+63.4	+85.0
0.85	+4.8	+65.5	+84.4
1.00	+16.4	+66.6	+79.7
1.25	+20.4	+64.5	+64.5
1.50	+25.5	+60.6	+60.6
1.75	+21.8	+55.1	+55.1
2.00	+22.2	+51.6	+51.6
3.00*	+39.5	+55.0	+55.0
4.00*	+28.8	+53.1	+53.1
5.00*	+36.0	+46.9	+46.9
6.00*	+40.5	+42.0	+42.0
7.00*	Trailing (from +18 to +57 mm) containing the three elements.		
8.00*	Trailing (from +24 to +60 mm) containing the three elements.		

The + sign represents displacement towards the anode while the - sign represents displacement towards the cathode.

\* Whatman glass-fibre paper (GF/A) strips were used for electrophoresis in these media.

observed only in concentrations of this acid higher than 0.2 *N*. The maximum anionic displacement attained by RaD, RaE and RaF are 40.5 (in 6 *N* HCl), 66.6 (in 1 *N* HCl) and 100.8 mm (in 0.4 *N* HCl) respectively. As shown by the values of the displacement of the three elements at different concentrations of HCl (Table I), their separation can be realized by electrophoresis over a wide concentration range (from 0.02 *N* to 1 *N*) of HCl. Although RaD can be separated from RaE and RaF using hydrochloric acid as dilute as 0.0025 *N*, the two last-mentioned elements have the same electrophoretic mobility in HCl more dilute than 0.02 *N* and thus do not separate from each other. This is presumably due to the hydrolytic effect of the dilute acid on highly charged RaE and RaF. Similarly in HCl more concentrated than 1 *N*, the mobility of RaE and RaF are the same and their separation becomes impracticable by electro-

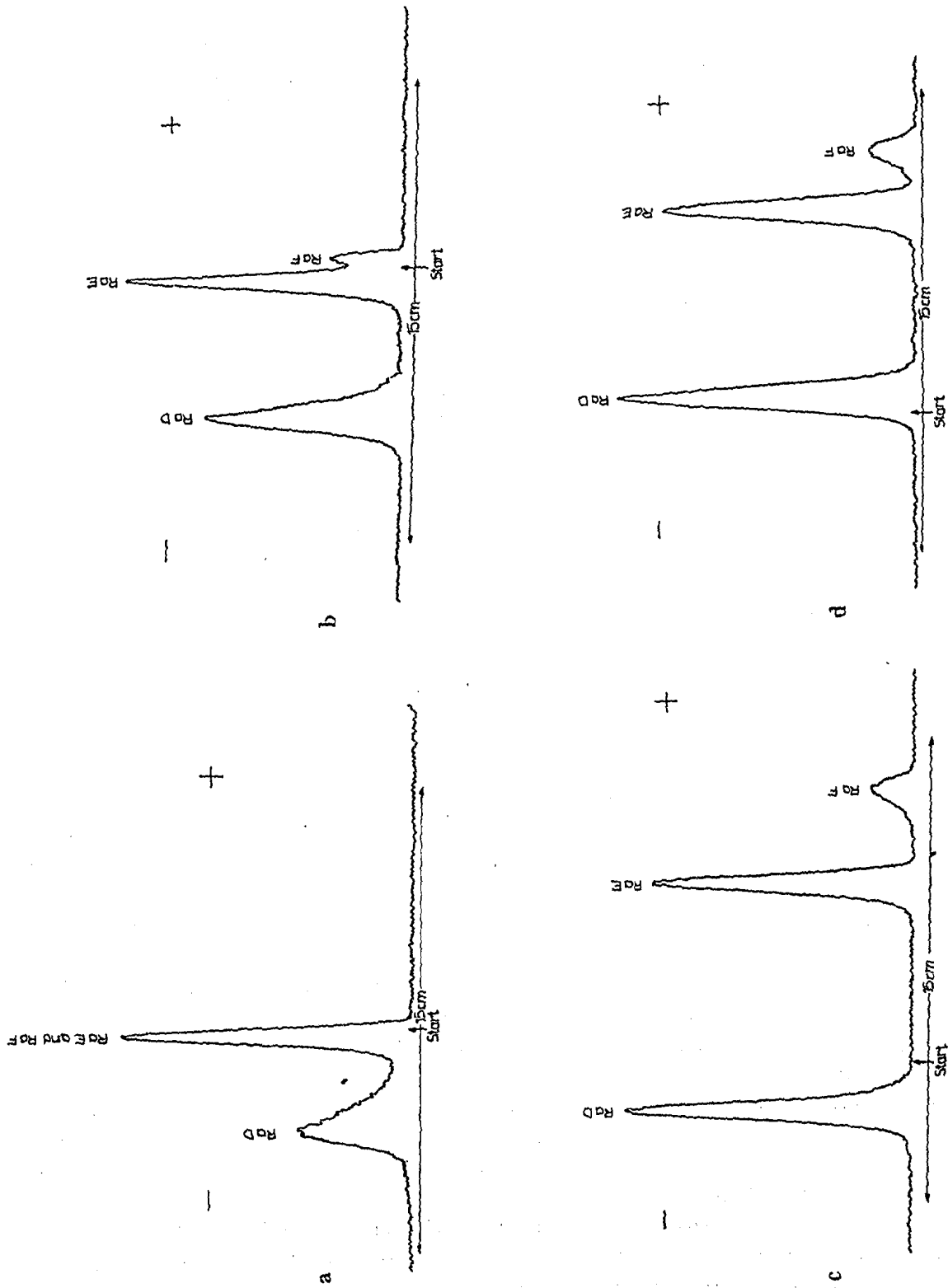
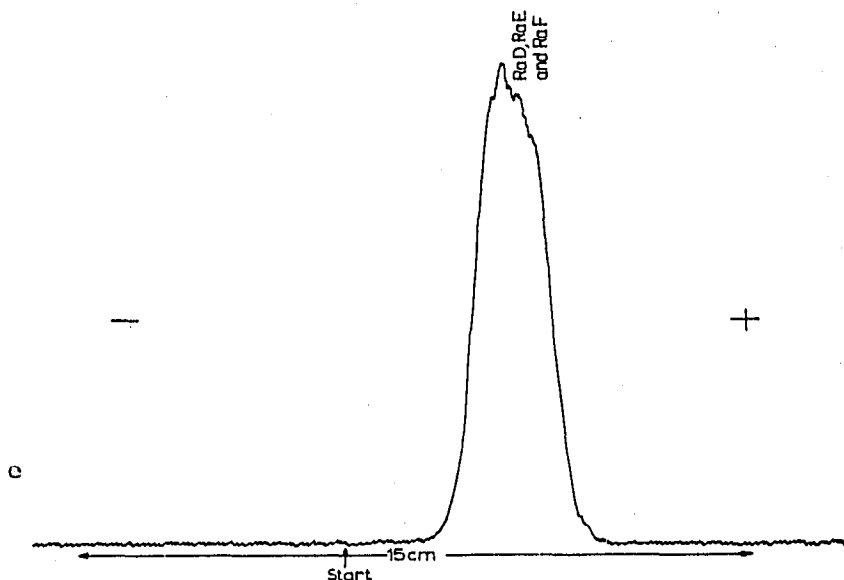


Fig. 1. Electrophoregrams (300 V; 45 min) of RaD, RaE and RaF in hydrochloric acid as electrolyte. (a) 0.0075 N; (b) 0.03 N; (c) 0.5 N; (d) 0.85 N.

(Continued on p. 505)

Fig. 1c. Electrophoregram (300 V; 45 min) of RaD, RaE and RaF in 7.0 *N* hydrochloric acid as electrolyte.



phoresis, while RaD, which has less tendency to form complexes moves more slowly and thus can be separated from the other two, to a greater or lesser extent, up to 6 *N* HCl. At concentrations of HCl higher than 6 *N*, only a wide band is obtained, because the electrophoretic bands of RaD, RaE and RaF overlap each other. As shown in Table I and Fig. 1, the favourable concentration range of HCl for the electrophoretic separation of RaD, RaE and RaF lies between 0.05 *N* and 1.0 *N*. The best separation, however, is obtained with HCl as the electrolyte in the concentration range 0.35–0.6 *N*, where the three elements give compact electrophoretic bands, widely separated from each other.

#### SUMMARY

The paper electrophoretic mobilities of RaD, RaE and RaF have been measured with hydrochloric acid as the back-ground electrolyte in the concentration range 0.0025 *N* to 8 *N*. The isoelectric concentration, where the change from cationic to anionic movement takes place, has been determined for the three elements; this was found to be the lowest for RaF and the highest for RaD. This is in agreement with the tendency of these elements to form complexes and also with the number of the positive charge carried by the ions. The concentration ranges in which RaD, RaE and RaF exist as cation and as anion have been investigated. An attempt has been made to define the concentration range of HCl in which excellent separations of these elements can be obtained by paper electrophoresis.

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