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Fractionation of Barium Isotopes in Cation-Exchange Chromatography

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ABSTRACT

Barium isotope fractionation has been observed in cation-exchange chromatography of barium salts operated in a breakthrough manner. The lighter isotopes are depleted in the front end fractions of the barium adsorption zones. Values of the single-stage separation factor minus one per unit mass difference between isotopes are 5.3×10^{-6} for the barium chloride system, 6.1×10^{-6} for the barium acetate system, and 4.8×10^{-6} for the barium lactate system at 25°C. No significant influence of barium–carboxylic acid complex formations on barium isotope effects has thus been observed.

INTRODUCTION

Investigation of isotope fractionation in ion-exchange systems contributes to stimulating the basic isotope effect theory as well as to actual isotope separation practice; for example, it may provide information on forces acting on an ion in a solution and solvent–solute interactions. In previous papers we reported isotope effects observed in the cation-exchange chromatography of salts of magnesium (1), calcium (2), and stron-

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tium (3) in aqueous media. The lighter isotopes were found to be preferentially fractionated into the resin phase. In this paper we describe barium isotope fractionation in cation-exchange chromatography, completing an investigation of isotope effects of alkaline earth elements with more than one stable isotope.

Naturally occurring barium consists of seven stable isotopes (^{130}Ba, 0.106%; ^{132}Ba, 0.101%; ^{134}Ba, 2.417%; ^{135}Ba, 6.592%; ^{136}Ba, 7.854%; ^{137}Ba, 11.23%; ^{138}Ba, 71.70%) (4). Enriched or separated barium isotopes do not seem to have large-scale applications in science and industry at present, but variations in barium isotopic composition of natural substances may be used as a tracer in geochemical studies (5).

EXPERIMENTAL

Reagents

The ion-exchange resin used was a highly porous, strongly acidic cation-exchange resin, Asahi LS-6, in the H^+ form, 100–200 mesh, with the sulfo groups (—$SO_3^- H^+$) as the exchange group, and an exchange capacity of 2.75 meq/g (the resin, in the H^+ form, was dried at 80°C and 5 torr for 24 hours). Reagents were all special-grade chemicals from Wako Chemical Ltd. and were used without further purification. All barium compounds used were of natural isotopic composition.

Chromatographic Process

Three experiments were carried out in the breakthrough operation. Experimental conditions are tabulated in Table 1. They are basically similar except for the barium salts in the feed solutions. This is to examine the effect of counterions of barium ions on barium isotope effects. Acetate and lactate ions are expected to form complexes with barium ions while chloride ions are not, and complex formations increase, decrease, or have little affect on the overall (observed) isotope effects of alkaline earth metals in aqueous systems (1–3). The chromatographic columns used were Pyrex glass columns (210 cm in length × 1.0 cm in inner diameter) with water jackets. They were connected in series with a Teflon tube (1 mm inner diameter) to obtain the resin bed heights specified in Table 1.

In every experiment a barium feed solution was fed to the first column, packed with the resin in the H^+ form, at a constant flow rate by a peristaltic pump, and the effluent from the last column was collected and portioned into fractions of 5.0 cm^3. The temperature of the columns was kept constant at 25.0 ± 0.2°C throughout the experiment by passing temperature-controlled water through the water jackets.

TABLE 1
Experimental Conditions[a]

	Run		
	Ba-1	Ba-2	Ba-3
Ba feed solution	0.11 M $BaCl_2$ (pH 6.50)	0.11 M Ba $(CH_3COO)_2$ + 0.3 M CH_3COOH (pH 4.29)	0.098 M $Ba(OH)_2$ + 0.50 M $CH_3CHOHCOOH$ (pH 3.34)
Resin bed height (cm)	1089	1096	955
Flow rate ($cm^3 \cdot cm^{-2} \cdot h^{-1}$)	14.5	16.9	10.3
Zone velocity ($cm \cdot h^{-1}$)	2.13	2.37	1.43
Breakthrough point (cm^3)	5795	6120	5390

[a] Temperature = 25.0 ± 0.2°C; operating manner = breakthrough.

Analysis

For every fraction of the effluents, the pH and barium concentration were measured with a pH meter and an inductively coupled plasma-atomic emission spectrometer, respectively. For the feed solutions and several fractions in the front end parts of the barium adsorption zones, the isotopic ratios $^{135}Ba/^{130}Ba$, $^{135}Ba/^{132}Ba$, $^{135}Ba/^{134}Ba$, $^{136}Ba/^{135}Ba$, $^{137}Ba/^{135}Ba$, and $^{138}Ba/^{135}Ba$, were measured with a Finnigan MAT 261 mass spectrometer. The chemical form of barium for mass spectrometry was barium chloride. The procedures for the preparation of samples for mass spectrometry (mass samples) and isotopic ratio measurements are described below.

In Run Ba-1 (barium chloride system), an aliquot of a fraction containing about 50 μmol barium was taken, and after its barium concentration was adjusted to 0.1 M (1 M = 1 mol/dm^3), it was subjected to isotopic ratio measurement. In Run Ba-2 (barium acetate system), 0.5 cm^3 concentrated hydrochloric acid was added to an aliquot of a fraction containing about 50 μmol barium, and the resultant solution was evaporated to dryness. This treatment was repeated twice to remove acetate ions completely. The evaporated residue was heated at 900°C for 7 hours in an electric furnace. The barium oxide thus obtained was dissolved with hydrochloric acid, and after its concentration was adjusted to 0.1 M, it was subjected to isotopic ratio measurement. In Run Ba-3 (barium lactate system), a procedure similar to that of Run Ba-2 was followed except that concentrated nitric acid instead of hydrochloric acid was used to remove lactate ions.

The barium isotopic ratios of a mass sample were measured with the double-filament thermal ionization technique. The filament unit had two filaments, one a vaporizing filament and the other an ionization filament, both made of rhenium ribbon. An aliquot of the mass sample (barium chloride) containing 2 μg barium was loaded on the surface of the vaporizing filament. Ionization was performed by passing an electric current through the ionization filament and an electric current through the vaporizing filament which was about one-tenth of the current through the ionization filament. When the ion beam intensities of $^{130}Ba^+$, $^{132}Ba^+$, $^{134}Ba^+$, $^{135}Ba^+$, $^{136}Ba^+$, $^{137}Ba^+$, and $^{138}Ba^+$ became sufficiently high about 1 hour after initiation of heating, the $^{130}Ba^+$–$^{138}Ba^+$ mass peaks were repeatedly recorded (output full scales: 10 V for $^{138}Ba^+$; 3 V for $^{137}Ba^+$; 1 V for $^{135}Ba^+$, $^{136}Ba^+$; 0.3 V for $^{134}Ba^+$; 0.03 V for $^{130}Ba^+$, $^{132}Ba^+$). The mass scanning was usually repeated seven times in a block, and 10 blocks were recorded as one measurement in most cases. The measuring time was about 2 hours, and hence the total time of one measurement was about 3 hours. The barium isotopic ratios of a block were calculated by averaging

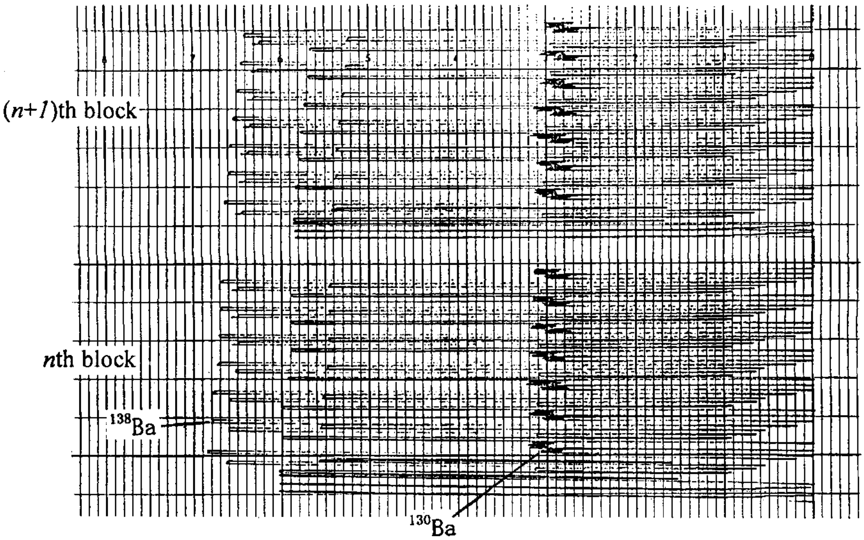

FIG. 1 An example of the mass spectra of the barium isotopes drawn on a recorder chart.

TABLE 2
Examples (printout) of Barium Isotopic Ratios and Relative Standard Deviations Measured

Block	Isotopic pair	Mean	Relative standard deviation (%)
1	^{138}Ba/^{135}Ba	10.823038	0.0094
	^{137}Ba/^{135}Ba	1.696910	0.0085
	^{136}Ba/^{135}Ba	1.189419	0.0072
	^{135}Ba/^{134}Ba	2.723002	0.0102
	^{135}Ba/^{132}Ba	65.296031	0.1184
	^{135}Ba/^{130}Ba	61.837408	0.0598
2	^{138}Ba/^{135}Ba	10.823265	0.0043
	^{137}Ba/^{135}Ba	1.696962	0.0079
	^{136}Ba/^{135}Ba	1.189361	0.0071
	^{135}Ba/^{134}Ba	2.722819	0.0131
	^{135}Ba/^{132}Ba	65.209832	0.1914
	^{135}Ba/^{130}Ba	61.602423	0.1328
3	^{138}Ba/^{135}Ba	10.822649	0.0091
	^{137}Ba/^{135}Ba	1.696742	0.0052
	^{136}Ba/^{135}Ba	1.189373	0.0023
	^{135}Ba/^{134}Ba	2.722286	0.0037
	^{135}Ba/^{132}Ba	65.171138	0.0707
	^{135}Ba/^{130}Ba	61.630913	0.1096

all the peak-height ratios in the blocks, and those of the mass sample were calculated as the averages of the ratios of the blocks (the peak height of ^{135}Ba^{+} was used as the basis of all the isotopic ratios). In these calculations the ratios whose deviations from the averages were large were excluded. The relative standard deviation of a measurement was typically 0.34% for the ^{135}Ba/^{130}Ba isotopic pair, 0.32% for ^{135}Ba/^{132}Ba, 0.04% for ^{135}Ba/^{134}Ba, 0.02% for ^{136}Ba/^{135}Ba, 0.06% for ^{137}Ba/^{135}Ba, and 0.10% for ^{138}Ba/^{135}Ba. An example of the mass spectra of the barium isotopes outputted on a recorder is shown in Fig. 1. In Table 2, examples of the barium isotopic ratios and accompanying relative standard deviations obtained in blocks of a measurement are tabulated.

RESULTS AND DISCUSSION

As an example of barium chromatograms and isotopic ratios obtained, those of Run Ba-1 are shown in Fig. 2. In the lower half of the figure is drawn the barium concentration profile. In the upper part are shown local

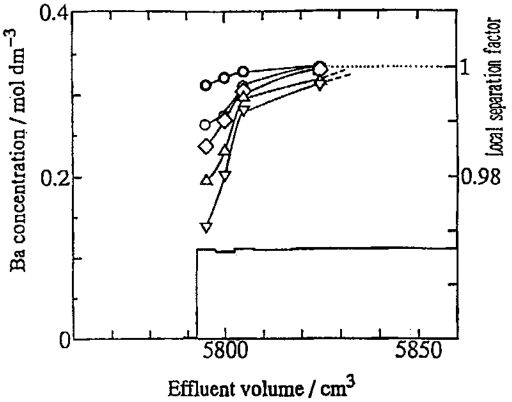

FIG. 2 The chromatogram and local separation factors for the barium–chloride system (Ba-1). ○ for the ^{137}Ba/^{138}Ba isotopic pair; ○ for the ^{135}Ba/^{138}Ba pair; ◇ for the ^{134}Ba/^{138}Ba pair; △ for the ^{132}Ba/^{138}Ba pair; ▽ for the ^{130}Ba/^{138}Ba pair.

separation factors, i.e., the isotopic ratios ^{137}Ba/^{138}Ba, ^{136}Ba/^{138}Ba, ^{135}Ba/^{138}Ba, ^{134}Ba/^{138}Ba, ^{132}Ba/^{138}Ba, and ^{130}Ba/^{138}Ba in the fractions divided by the corresponding ratios in the feed solution. The ^{x}Ba/^{138}Ba (x = 130, 132, 134, 135, 136, or 137) ratio was calculated by (^{x}Ba/^{135}Ba)(^{135}Ba/^{138}Ba). It is clearly seen that the lighter isotopes are depleted in the front end of the barium adsorption zone. This seems to be the first observation of barium isotope fractionation in ion-exchange systems. Depletion of the lighter isotopes in the front end of the barium zone means that they are preferentially fractionated into the resin phase. This tendency is the same as those of the other alkaline earth metals so far studied (1–3). It is also seen in Fig. 2 that the larger the mass difference between the two isotopes is, the larger the deviation of the local separation factor from unity is in a given fraction.

Similar observations can be made for Runs Ba-2 and Ba-3.

The single-stage separation factors, $S(x/138)$ for the ^{x}Ba/^{138}Ba isotopic pair (x = 130, 132, 134, 135, 136, and 137), were calculated by using the equation (6)

$$S = 1 + \sum |R_i - R_o| f_i/[QR_o(1 - R_o)] \qquad (1)$$

where, assuming a two-isotope system (^{138}Ba and ^{x}Ba with x = 130, 132, 134, 135, 136, or 137), R_o is the isotopic molar fraction of ^{x}Ba in the feed solution, R_i that in the ith fraction of the effluent, f_i the amount of barium in the ith fraction, Q the total exchange capacity of the resin bed for the barium ion, and the summation is taken over all fractions depleted in the lighter isotopes. The calculated results are summarized in Table 3. The data are given in two forms, i.e., ϵ ($= S - 1$) and $\epsilon/\Delta M$ (ϵ per unit mass difference between the two isotopes).

From Table 3 the following may be extracted concerning barium isotope effects in aqueous ion-exchange systems:

1. In each run the ϵ values of the pairs with larger mass differences are generally larger than those of the pairs with smaller mass differences. The $\epsilon/\Delta M$ values, which are on the order of 10^{-6}, are, however, about the same for all the pairs within experimental errors. Thus, anomalous odd–even isotope effects in equilibria, which were first observed for uranium isotopes in redox systems in aqueous media (7, 8) and later found for isotopes of other elements in solvent extraction systems (9), are not in evidence.
2. The average $\epsilon/\Delta M$ values of the three experiments are very similar to each other. Against our expectation, changing counterions of the barium ion does not make a substantial contribution to barium isotope effects in ion-exchange systems within the experimental conditions examined.

TABLE 3
Separation Factors Obtained

Run	System	Isotopic pair	ϵ	$\epsilon/\Delta M$	Average $\epsilon/\Delta M$
Ba-1	Barium chloride	$^{130}Ba/^{138}Ba$	4.1×10^{-5}	5.2×10^{-6}	
		$^{132}Ba/^{138}Ba$	3.1×10^{-5}	5.1×10^{-6}	
		$^{134}Ba/^{138}Ba$	2.2×10^{-5}	5.6×10^{-6}	5.3×10^{-6}
		$^{135}Ba/^{138}Ba$	1.6×10^{-5}	5.5×10^{-6}	
		$^{136}Ba/^{138}Ba$	—	—	
		$^{137}Ba/^{138}Ba$	5.3×10^{-6}	5.3×10^{-6}	
Ba-2	Barium acetate	$^{130}Ba/^{138}Ba$	3.2×10^{-5}	4.0×10^{-6}	
		$^{132}Ba/^{138}Ba$	2.9×10^{-5}	4.9×10^{-6}	
		$^{134}Ba/^{138}Ba$	2.3×10^{-5}	5.6×10^{-6}	6.1×10^{-6}
		$^{135}Ba/^{138}Ba$	1.7×10^{-5}	5.7×10^{-6}	
		$^{136}Ba/^{138}Ba$	1.8×10^{-5}	9.2×10^{-6}	
		$^{137}Ba/^{138}Ba$	7.1×10^{-6}	7.1×10^{-6}	
Ba-3	Barium lactate	$^{130}Ba/^{138}Ba$	3.3×10^{-5}	4.2×10^{-6}	
		$^{132}Ba/^{138}Ba$	3.3×10^{-5}	5.6×10^{-6}	
		$^{134}Ba/^{138}Ba$	1.8×10^{-5}	4.6×10^{-6}	4.8×10^{-6}
		$^{135}Ba/^{138}Ba$	1.2×10^{-5}	3.9×10^{-6}	
		$^{136}Ba/^{138}Ba$	1.1×10^{-5}	5.7×10^{-6}	
		$^{137}Ba/^{138}Ba$	4.7×10^{-6}	4.7×10^{-6}	

Analysis of *S* Data Based on a Theory of Isotope Distribution between Two Phases (10)

For the barium acetate and barium lactate systems, $S(S_{\text{Ba-Ac}}$ or $S_{\text{Ba-Lc}})$ can be expressed in terms of the isotopic reduced partition function ratios (RPFRs) (11) and molar fractions of the barium species involved in the systems as

$$\ln S = \ln(x_{\text{Ba}} f_{\text{Ba}} + x_{\text{BaL}} f_{\text{BaL}}) - \ln(y_{\text{Ba}} g_{\text{Ba}} + y_{\text{BaL}} g_{\text{BaL}}) \tag{2}$$

where x_{Ba} and f_{Ba} are the molar fraction and the RPFR of the simple hydrated barium ion in the solution phase, x_{BaL} and f_{BaL} are those of complex species (BaL^+; L = acetate ion or lactate ion) in the solution phase, and y and g are the quantities in the resin phase corresponding to x and f, respectively. Equation (2) is rearranged into

$$\begin{aligned}\ln S = {} & \ln(f_{\text{Ba}}/g_{\text{Ba}}) + \ln[x_{\text{Ba}} + (1 - x_{\text{Ba}}) f_{\text{BaL}}/f_{\text{Ba}}] \\ & - \ln[(y_{\text{Ba}} + (1 - y_{\text{Ba}}) g_{\text{BaL}}/g_{\text{Ba}}]\end{aligned} \tag{3}$$

The ratio $f_{\text{Ba}}/g_{\text{Ba}}$ in Eq. (3) is the equilibrium constant of the barium isotope exchange reaction between simple hydrated barium ions in the solution phase and those in the resin phase. It represents the barium isotope effect accompanying the distribution of simple hydrated barium ions between the two phases and can be approximated as being equal to the separation factor in the barium chloride system ($S_{\text{Ba-Cl}}$), since no appreciable complex formation between the barium ions and the chloride ions is expected under the present experimental conditions in this system. The quantities $f_{\text{BaL}}/f_{\text{Ba}}$ and $g_{\text{BaL}}/g_{\text{Ba}}$ represent the isotope effects accompanying complex formations between Ba^{2+} and L^- in the solution and the resin phases, respectively. Assuming reasonably that $f_{\text{BaL}}/f_{\text{Ba}} = g_{\text{BaL}}/g_{\text{Ba}}$, Eq. (3) becomes

$$\begin{aligned}\ln S = {} & \ln S_{\text{Ba-Cl}} + \ln[x_{\text{Ba}} + (1 - x_{\text{Ba}}) f_{\text{BaL}}/f_{\text{Ba}}] \\ & - \ln[(y_{\text{Ba}} + (1 - y_{\text{Ba}}) f_{\text{BaL}}/f_{\text{Ba}}]\end{aligned} \tag{4}$$

Equation (4) expresses $S(S_{\text{Ba-Ac}}$ or $S_{\text{Ba-Lc}})$ as a function of $f_{\text{BaL}}/f_{\text{Ba}}$ for given x_{Ba} and y_{Ba}. The sign of $\ln(S/S_{\text{Ba-Cl}})$ depends on the signs of $\ln(x_{\text{Ba}}/y_{\text{Ba}})$ and $\ln(f_{\text{BaL}}/f_{\text{Ba}})$. $\ln(S/S_{\text{Ba-Cl}})$ is positive when $x_{\text{Ba}} > y_{\text{Ba}}$ and $f_{\text{BaL}} < f_{\text{Ba}}$ or when $x_{\text{Ba}} < y_{\text{Ba}}$ and $f_{\text{BaL}} > f_{\text{Ba}}$.

The x_{Ba} values of barium acetate and barium lactate systems can be estimated by using the required stability constant and dissociation constant data (12) on the chemical species involved in those systems. The y_{Ba} values can be calculated from the x_{Ba} values and zone velocities of the chromatographic experiments as was done in the previous papers (1–3). Those estimations show that $x_{\text{Ba}} < y_{\text{Ba}}$ in both the barium acetate and barium lactate systems. Thus, the sign of $\ln(S/S_{\text{Ba-Cl}})$ ($S = S_{\text{Ba-Ac}}$ or $S_{\text{Ba-Lc}}$) is the same as that of $\ln(f_{\text{BaL}}/f_{\text{Ba}})$. Experimentally, S is nearly equal to $S_{\text{Ba-Cl}}$, showing that f_{BaL} is almost the same as f_{Ba}, that is, complex formations between barium ions and acetate or lactate ions make only immaterial contributions to S. In other words, the sum of forces acting on a barium ion is little changed by such complex formations.

Estimation of the Reduced Partition Function Ratios

The RPFR, f_{Ba}, of the simple hydrated barium ion in the solution phase for any barium isotopic pair can be estimated by using the Bigeleisen–Mayer simplifying formula for the RPFR (11) with omission of the symmetry numbers and with neglect of second and higher hydration spheres,

$$\ln f_{\text{Ba}} = [nm\Delta M/(24MM')](hc\omega/kT)^2 \tag{5}$$

where n is the hydration number in the first hydration sphere, m is the mass of the water molecule, M and M' are the masses of the heavier and lighter barium isotopes in concern, $\Delta M = M - M'$, h is Planck's constant, c is the velocity of light, ω is the Ba^{2+}–H_2O totally symmetric stretching frequency in cm^{-1}, k is Boltzmann's constant, and T is the temperature. The values of the quantities in Eq. (5), except for n and ω, are readily known.

Much is not known about n and ω values for the barium ion in aqueous solutions. An x-ray diffraction study (13) of an aqueous barium chloride solution indicated that $n = 9.5$ [the author (13) said that $n = 8$ was reasonable]. No experimental nor theoretical value of ω is found in the literature. Kanno (14) found a linear correlation between ω and the reciprocal of the cation-hydrating water molecule distance. By assuming that the peak found at 290 pm in the electron radial distribution function for a barium chloride solution (13) corresponds to this distance and using Kanno's correlation (14), ω is estimated to be 217 cm^{-1}. Using the values of $n = 9.5$ and $\omega = 217$, f_{Ba} can then be calculated; its value for the $^{136}Ba/^{138}Ba$ isotopic pair is 1.0008345 at 25°C. This should be compared with corresponding values for isotopic pairs of other alkaline earth metals with $\Delta M = 2$; 1.044545 for the $^{24}Mg/^{26}Mg$ pair (1), 1.017681 for the $^{40}Ca/^{42}Ca$ pair (2), and 1.0028341 for the $^{86}Sr/^{88}Sr$ (3). Correlation of fs with atomic masses is apparent.

The f_{Ba} values of the other isotopic pairs can easily calculated with Eq. (5), and the g_{Ba} (RPFRs in the solution phase) values are obtained using the f_{Ba} values and the relation, $S_{\text{Ba-Cl}} = f_{\text{Ba}}/g_{\text{Ba}}$.

Approximate calculations of RPFRs shown above will be improved by using, instead of Eq. (5), the finite orthogonal polynomial method (15) as well as the WINIMAX method (16).

CONCLUSION

Major findings of the present study are as follows.

1. The lighter isotopes of barium were preferentially fractionated into the resin phase rather than into the solution phase in cation-exchange chromatography of barium salts operated in the breakthrough manner at 25°C. This tendency was the same as those observed for other alkaline earth metals.
2. For a given counterion of the barium ion, the ϵ value is, in general, larger for the isotopic pair with a larger mass difference, but the $\epsilon/\Delta M$ value is the same for all the isotopic pairs examined within experimental errors. Thus, no indication of anomalous odd–even isotope effects is evidenced.
3. The kind of counterion of the barium ion has no significant effect on barium isotope effect within experimental conditions studied. This corresponds to the fact that the sum of forces acting on a barium ion is little altered by complex formations.

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