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**Equilibrium Constants of
Liquid-Liquid Distribution Reactions:
Part IV**

CHELATING EXTRACTANTS



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Equilibrium Constants of Liquid—Liquid Distribution Reactions

PART IV: CHELATING EXTRACTANTS

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CHELATING EXTRACTANTS

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PREFACE

This volume has been prepared in partial response to the decision of the Commission on Equilibrium Data (V. 6) of the Analytical Chemistry Division of the International Union of Pure and Applied Chemistry to make a compilation of liquid-liquid distribution equilibrium data. Although at the outset it was planned to include this in the second edition of the Stability Constants*, the somewhat different organization of the material as well as the time required to prepare the compilation, suggested the wisdom of separate publication. Although this part is specifically concerned with extractants that form chelates, it should be recognized that some of the extractants dealt with in previous parts under certain conditions may also form chelates.

This tabulation includes all data published up to the end of 1972 for the major chelating extractants. All papers were read in the original. Wherever, in the compilers' judgment the data presented in the paper warranted it, calculation (or recalculation) from experimental results (Tables, Figures) was used to supplement the given equilibrium parameters.

We are greatly indebted to many chemists throughout the world who have kept us informed about recent or pending publications and for other valuable suggestions. Especially helpful comments were obtained from Professors E. I. Astakhova-Ivanova, D. Dyrssen, T. V. Healy, D. N. Hume, C. Keller, A. S. Kertes, N. M. Kuzmin, J. O. Liljenzin, Y. Marcus, O. Navratil, S. Oki, F. J. C. Rossotti, J. Rydberg, G. Schweitzer, T. Sekine, and Yu. A. Zolotov. We also thank Mrs. T. Vrbska for technical help in arranging Tables.

* Stability Constants of Metal-Ion Complexes, Second Edition, compiled by L. G. Sillén (inorganic ligands) and A. E. Martell (organic ligands), The Chemical Society, London, Special Publication No. 17 (1964).

HOW TO USE THE TABLES

1. General arrangement

The table summarizes the data up to the end of 1972. For each extractant and its extractable metal complexes, distribution equilibrium constants, as well as appropriate extraction constants, are recorded. In addition, the homogeneous equilibria involving the acid dissociation of the extractant in the aqueous phase and adduct or mixed ligand complex formation in the organic phase are characterized. Aqueous phase metal complex formation constants however, are not included inasmuch as these are already covered in Stability Constants.

The information given for each system will be the chemical reactions, the equilibrium constant, the temperature, the composition of the aqueous and organic phases, as well as the reference to the original literature.

We have followed the practice of previous parts of this series of not listing the method of measurement. In most cases, these involved standard radiochemical or spectrophotometric analytical techniques.

2. Order of ligands and metal ions

Although the previous parts dealing with liquid-liquid distribution followed in the main the arrangement found in Stability Constants, the authors felt that the sufficiently great convenience in listing chelating extractants in family groupings to be an overriding factor. Wherever the extractant is one that is listed in Stability Constants, the order of reagents follows the Beilstein system of arrangement in the Stability Constants. Metal ions are listed in the same order as found in Stability Constants.

3. Temperature

Under this heading are given the temperatures in °C at which the distribution reactions were studied. Room temperature is denoted by "rt".

4. Conditions

Under the heading "aq" appears the composition of the aqueous phase, as well as the ionic strength wherever listed. The term "I=0" signifies values extrapolated to zero ionic strength. The figures given for composition of aqueous phase pertain, in every case, to the initial composition of the aqueous phase, rather than to equilibrium composition.

The entries under the heading "org" give the relevant information for the composition of the organic phase. The diluent used, the molar concentration of the extractant and, wherever appropriate, the auxiliary ligand are specified. Here again, the initial rather than the equilibrium composition of the organic phase is given. The diluents have been arranged in the following order: aliphatic hydrocarbons, aromatic hydrocarbons, halogenated hydrocarbons, other hydrocarbons, ethers, ketones, esters, alcohols, other solvents; within each group the order is according to increasing molecular weight.

5. Results

The third column records the logarithm to the base 10 of the equilibrium constants ($\log K$) with the convention that "log" and "=" are always omitted. Concentrations are expressed in mole liter⁻¹.

A formula of the type H_pA is assigned to each extractant listed in the table, in order to define the entity A in terms of which the metal complexes are described. In the case of mixed ligand chelates, the second ligand will be designated as B. The symbol M^{n+} designates either a simple metal ion, such as Fe^{3+} or Zn^{2+} , or an oxygen-containing ion such as UO_2^{2+} or $PaO(OH)^{2+}$. Additional ligands (adduct reagents) are represented by S.

6. Equilibrium constants

Because of the variety of reactions involved in distribution equilibria, the practice used in previous parts of this series of listing the reaction will be continued here. A line drawn above any formula indicates that the species is in the organic phase. With chelating extractants a greater degree of uniformity is seen, however, and it would be useful to summarize the various reaction equilibria that are generally involved.

(a) Distribution constant of the extractant



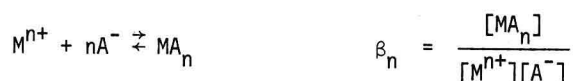
(b) Distribution constant of the extractable complex



(c) Aqueous phase acid dissociation constant of the extractant



(d) Aqueous phase metal complex formation constants (overall)

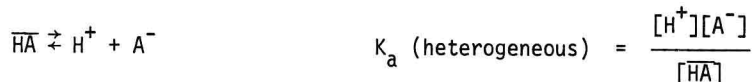


(e) Adduct formation constant



Examples of all the above types of constants with the exception of β values (see d) which are tabulated in Stability Constants will be listed here along with those of equilibria involving both distribution and ligand dissociation of metal complex formation.

Thus, a heterogeneous dissociation equilibrium for the extractant



which by comparison with (a) and (c) shows this constant to be equivalent to $K_a/K_D(\text{HA})$.

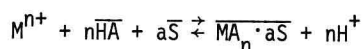
Similarly, the overall extraction constant involving metal complex formation and its distribution



can be described in terms of contributing constants (a) to (d)

$$K_{\text{ex}} = \frac{K_D(\text{MA})_n \beta_n K_a^n}{K_D^n(\text{HA})}$$

For chelate extraction systems involving additional ligands characterized by



the overall extraction constant, K_{ex} , is

$$K_{\text{ex}} (\text{adduct}) = \frac{[\overline{\text{MA}}_n \cdot a\overline{\text{S}}][\text{H}^+]^n}{[\text{M}^{n+}][\overline{\text{HA}}]^n[\overline{\text{S}}]^a} = \frac{K_D(\text{MA})_n \beta_n K_a^n \overline{K}}{K_D^n(\text{HA})} = \overline{K} \cdot K_{\text{ex}}$$

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I. DIALKYLDITHIOCARBAMIC ACIDS

Extractant: C₅H₉NS₂ 1-Pyrrolidinedithiocarbamic acid, HA

Distribution constant:

HA = \overline{HA} 3.04 24 0.1 HA in CHCl₃ 71L

Dissociation:

HA = H⁺ + A⁻ -3.09 24 0.1 71L
 $\overline{HA} = \overline{H^+} + \overline{A^-}$ -6.13 24 0.1 HA in CHCl₃

Extraction constants:

Co²⁺ M²⁺ + 2 \overline{HA} = $\overline{MA_2}$ + 2H⁺ -0.8 24 0.1 71L
Cu²⁺ M²⁺ + 2 \overline{HA} = $\overline{MA_2}$ + 2H⁺ 11.4 24 0.1 HA in CHCl₃
Zn²⁺ M²⁺ + 2 \overline{HA} = $\overline{MA_2}$ + 2H⁺ 0.4 24 0.1 HA in CHCl₃
Cd²⁺ M²⁺ + 2 \overline{HA} = $\overline{MA_2}$ + 2H⁺ 1.0 24 0.1 HA in CHCl₃
Ga³⁺ M³⁺ + 3 \overline{HA} = $\overline{MA_3}$ + 3H⁺ 4.5 24 0.1 HA in CHCl₃
Bi³⁺ M³⁺ + 3 \overline{HA} = $\overline{MA_3}$ + 3H⁺ 15.5 24 0.1 HA in CHCl₃

Reference:

71L Likussar W., Boltz D. F., Anal. Chem., 43, 1273 (1971)

Extractant: 332 C₅H₁₁NS₂ Diethyldithiocarbamic acid, HA

Distribution constants:

H⁺ HA = \overline{HA} 3.37 20 0.1 60B
HA in CHCl₃
2.38 20 0.1 60B
HA in CCl₄
2.6 20 64S
HA in CCl₄

Distribution constants (cont.)

| | | | | |
|-----------|-------------------|------|---------------|-----|
| Mn^{2+} | $\overline{MA_2}$ | 3.11 | C_6H_6 | 70U |
| | | 3.40 | $CHCl_3$ | |
| | | 3.15 | CCl_4 | |
| | | 3.45 | Butyl acetate | |
| Co^{2+} | $\overline{MA_2}$ | 5.98 | C_6H_6 | |
| | | 6.60 | $CHCl_3$ | |
| | | 5.38 | CCl_4 | |
| | | 6.04 | Butyl acetate | |
| Ni^{2+} | $\overline{MA_2}$ | 5.83 | C_6H_6 | |
| | | 6.36 | $CHCl_3$ | |
| | | 5.67 | CCl_4 | |
| | | 5.95 | Butyl acetate | |
| Cu^{2+} | $\overline{MA_2}$ | 8.11 | C_6H_6 | 70U |
| | | 9.34 | $CHCl_3$ | |
| | | 8.34 | CCl_4 | |
| | | 8.88 | Butyl acetate | |
| Zn^{2+} | $\overline{MA_2}$ | 5.11 | C_6H_6 | |
| | | 5.28 | $CHCl_3$ | |
| | | 4.81 | CCl_4 | |
| | | 5.11 | Butyl acetate | |

Distribution constants (cont.)

| | | | | |
|------------------|--|------|------------------------|-----|
| Cd^{2+} | $\text{MA}_2 = \overline{\text{MA}}_2$ | 5.81 | C_6H_6 | 70U |
| | | 5.88 | CHCl_3 | |
| | | 5.81 | CCl_4 | |
| | | 5.51 | Butyl acetate | |
| | | 6.72 | C_6H_6 | |
| Pb^{2+} | $\text{MA}_2 = \overline{\text{MA}}_2$ | 7.41 | CHCl_3 | |
| | | 6.48 | CCl_4 | |
| | | 6.48 | Butyl acetate | |
| | | 8.26 | C_6H_6 | |
| | | 9.00 | CHCl_3 | |
| Bi^{3+} | $\text{BiA}_3 = \overline{\text{BiA}}_3$ | 7.65 | CCl_4 | |
| | | 7.65 | Butyl acetate | |

Dissociation:

| | | | |
|--|-------|---|-----------------------|
| $\text{HA} = \text{H}^+ + \text{A}^-$ | 0 | $(\text{Na}^+, \text{H}^+) \text{CH}_3\text{COO}^-$ | 57B |
| | -3.95 | | 59Z |
| | -3.6 | 20 0.1 | 64S |
| | -3.4 | 25 0 | 70A |
| | -6.21 | $(\text{Na}^+, \text{H}^+) (\text{CH}_3\text{COO}^-, \text{PO}_4^{3-})$ | 54B |
| $\overline{\text{HA}} = \text{H}^+ + \text{A}^-$ | -6.21 | 20 $(\text{Na}^+, \text{H}^+) \text{ClO}_4^-$ 0.1 | 68S |
| | -7.0 | 25 $(\text{Na}^+, \text{H}^+) \text{PO}_4^{3-}$ 0.1 | 71V |
| | | | HA in CCl_4 |
| | | | HA in CCl_4 |
| | | | HA in CHCl_3 |

Extraction constants:

| | | | | | | |
|-----------|--|--|----|--------------------------|--|-------------------|
| Mn^{2+} | $M^{2+} + 2\overline{HA} = \overline{MA}_2 + 2H^+$ | -3.5 ^a | 20 | $(Na^+, H^+)ClO_4^-$ 0.1 | CHCl ₃ HA in CCl ₄ CCl ₄ C ₆ H ₆ CHCl ₃ CCl ₄ Butyl acetate | 70U 68S 70U |
| | $M^{2+} + 2A^- = \overline{MA}_2$ | -4.42 -2.2 ^a 10.20 10.48 10.23 10.52 | | | | |
| Fe^{2+} | $M^{2+} + 2\overline{HA} = \overline{MA}_2 + 2H^+$ | 1.20 | 20 | $(Na^+, H^+)ClO_4^-$ 0.1 | HA in CCl ₄ | 68S |
| Co^{2+} | $M^{2+} + 2\overline{HA} = \overline{MA}_2 + 2H^+$ | 1.28 5.7 ^a 2.33 6.1 ^a 19.15 19.76 18.53 19.20 | 20 | $(Na^+, H^+)ClO_4^-$ | CCl ₄ CHCl ₃ HA in CCl ₄ CCl ₄ C ₆ H ₆ CHCl ₃ CCl ₄ Butyl acetate | 70U 68S 70U |
| Ni^{2+} | $M^{2+} + 2\overline{HA} = \overline{MA}_2 + 2H^+$ | 5.0 ^a 11.6(?) 5.9 ^a | 20 | $(Na^+, H^+)ClO_4^-$ | CHCl ₃ CCl ₄ CCl ₄ | 68S 70U |

Extraction constants (cont.)

| | | | | | | |
|------------------|--|-------------------|----|--|-------------------------------|------|
| Ni ²⁺ | $M^{2+} + 2A^- = \overline{MA}_2$ | 18.46 | | | C ₆ H ₆ | 70U |
| | | 19.00 | | | CHCl ₃ | |
| | | 18.30 | | | CCl ₄ | |
| | | 18.59 | | | Butyl acetate | |
| Cu ²⁺ | $M^{2+} + 2\overline{HA} = \overline{MA}_2 + 2H^+$ | 12.5 ^a | 20 | (Na ⁺ , H ⁺)ClO ₄ ⁻ | CHCl ₃ | 64S |
| | | 13.7 ^a | | | CHCl ₃ | 70U |
| | | 13.70 | 20 | (Na ⁺ , H ⁺)ClO ₄ ⁻ 0.1 | HA in CCl ₄ | 68S |
| | | 14.1 | 20 | | CCl ₄ | 68Sa |
| | | 14.3 ^a | | | CCl ₄ | 70U |
| | | 14.0 | | | CCl ₄ | 71S |
| | $M^{2+} + 2A^- = \overline{MA}_2$ | 26.52 | | | C ₆ H ₆ | 70U |
| | | 27.76 | | | CHCl ₃ | |
| | | 26.76 | | | CCl ₄ | |
| | | 27.30 | | | Butyl acetate | |
| | | 12.2 ^a | 20 | | CHCl ₃ | 64S |
| | | 11.90 | 20 | (Na ⁺ , H ⁺)ClO ₄ ⁻ 0.1 | HA in CCl ₄ | 68S |
| Ag ⁺ | $M^+ + \overline{HA} = \overline{MA} + H^+$ | 11.52 | 20 | | CCl ₄ | 68Sa |
| | | 11.9 | 20 | | CCl ₄ | 71S |
| | | 2.1 ^a | 20 | | CHCl ₃ | 64S |
| | | 1.9 ^a | | | CHCl ₃ | 70U |
| Zn ²⁺ | $M^{2+} + 2\overline{HA} = \overline{MA}_2 + 2H^+$ | | | | | |
| | | | | | | |

Extraction constants (cont.)

| | | | | | | | |
|------------------|--|-------------------|----|----------------------|-----|-------------------------------|------|
| Zn ²⁺ | $M^{2+} + 2\overline{HA} = \overline{MA}_2 + 2H^+$ | 2.96 | 20 | $(Na^+, H^+)ClO_4^-$ | 0.1 | HA in CCl ₄ | 68S |
| | | 2.54 | 20 | | | CCl ₄ | 68Sa |
| | | 2.85 | 25 | $(Na^+, H^+)ClO_4^-$ | | HA in CCl ₄ | 70Z |
| | | 3.0 ^a | | | | CCl ₄ | 70U |
| | | 2.8 | 20 | 0.1 | | CCl ₄ | 71S |
| | | 15.71 | | | | C ₆ H ₆ | 70U |
| | | 15.88 | | | | CHCl ₃ | |
| | | 15.40 | | | | CCl ₄ | |
| | | 15.71 | | | | Butyl acetate | |
| | | 4.4 ^a | 20 | | | CHCl ₃ | 64S |
| Cd ²⁺ | $M^{2+} + 2\overline{HA} = \overline{MA}_2 + 2H^+$ | 4.75 ^a | | | | CHCl ₃ | 70U |
| | | 5.41 | 20 | $(Na^+, H^+)ClO_4^-$ | 0.1 | HA in CCl ₄ | 68S |
| | | 5.13 | 20 | | | CCl ₄ | 68Sa |
| | | 5.81 | 25 | $(Na^+, H^+)ClO_4^-$ | | -HA in CCl ₄ | 70Z |
| | | 5.94 | 25 | | | CCl ₄ | |
| | | 6.26 ^a | | | | CCl ₄ | 70U |
| | | 5.8 | 20 | 0.1 | | CCl ₄ | 71S |
| | | 18.68 | | | | C ₆ H ₆ | 70U |
| | | 18.75 | | | | CHCl ₃ | |
| | | 18.68 | | | | CCl ₄ | |
| | $M^{2+} + 2A^- = \overline{MA}_2$ | 18.38 | | | | Butyl acetate | |