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Data of Inorganic Solvent Extraction (1)

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Data of Inorganic Solvent Extraction (1)

Summary

Distribution behavior of more than sixty elements was studied in the following solvent extraction systems:

(1) 100% TBP–HCl
(2) 50% TBP toluene–HCl
(3) 25% TBP toluene–HCl
(4) 10% TBP toluene–HCl
(5) 100% TBP–HNO_3
(6) 25% TBP CCl_4–HNO_3
(7) 1% TBPO toluene–HCl
(8) 5% TOPO toluene–HCl
(9) 5% TOPO toluene–HNO_3
(10) 10% TBMDP xylene–HNO_3
(11) 10% TBEDP xylene–HNO_3
(12) 5% TIOA xylene–HCl
(13) 5% TIOA xylene–HNO_3
(14) 10% Amberlite LA-1 xylene–HCl
(15) 10% Amberlite LA-1–HNO_3
(16) 10% Primene JM-T xylene–HCl
(17) 50% HDEHP toluene–HCl
(18) 3% DBSA–HCl
(19) 0.1M tetraethyl-benzyl-ammonium chloride $CHCl_3$–ammonium thiocyanate
(20) 0.1M dimethyl-benzyl-phenyl ammonium chloride $CHCl_3$–HCl
(21) tetraphenylarsonium chloride $CHCl_3$–HCl

Results are summarized in the acid dependence curves arranged according to the sequence of the periodic table.

Jan. 1963

Editors Tomitaro Ishimori and Eiko Nakamura
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無機溶媒抽出データ集 (1)

要　旨

約60種の化学元素について，次にあげるような21種の溶媒抽出系における抽出行動をしらべた.

(1) 100% TBP–HCl
(2) 50% TBP トルエン–HCl
(3) 25% TBP トルエン–HCl
(4) 10% TBP トルエン–HCl
(5) 100% TBP–HNO_3
(6) 25% TBP CCl_4–HNO_3
(7) 1% TBPO トルエン–HCl
(8) 5% TOPO トルエン–HCl
(9) 5% TOPO トルエン–HNO_3
(10) 10% TBMDP キシレン–HNO_3
(11) 10% TBEDP キシレン–HNO_3
(12) 5% TIOA キシレン–HCl
(13) 5% TIOA キシレン–HNO_3
(14) 10% アンバーライト LA-1 キシレン–HCl
(15) 10% アンバーライト LA-1–HNO_3
(16) 10% プライメン JM-T キシレン–HCl
(17) 50% HDEHP トルエン–HCl
(18) 3% ドデシルベンゼンスルホン酸ナトリウム–HCl
(19) 0.1M テトラブチルベンジルアンモニウムクロリド $CHCl_3$–チオシアン酸アンモニウム
(20) 0.1M ジメチルベンジルフェニルアンモニウムクロリド $CHCl_3$–HCl
(21) テトラフェニルアルソニウムクロリド $CHCl_3$–HCl

各元素の分配比の酸濃度に対する dependence をしらべ，その結果を周期律表の形の図にまとめて配列した.

1963 年 1 月

編集者：日本原子力研究所 化学部　石森富太郎，中村　永子

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1 INTRODUCTION

Since 1958, the solvent extraction of inorganic ions had been studied both in Division of Chemistry and in Division of Health Physics, JAERI, aiming the concrete elucidation of differences in the extraction behavior of various ions.

In these surveys, diluted or undiluted organic extractants are taken as the organic phase, whereas nitric or hydrochloric acid of known concentrations are chosen as the aqueous. Determination of K_d values were performed radiometrically.

The present paper gives the accumulated basic data thus obtained. They are shown as a series of 21 figures where acid dependence curves for most of chemical elements are compiled in the periodic table presentation.

2 EXPERIMENTAL

2.1 Extracting agents

The solvent extraction systems studied are given in TABLE 1 containing the name of the extracting agents, diluents used and the aqueous phase applied.

2.2 Radioisotopes used

TABLE 2 includes a list of radioisotopes used together with their target materials. Some radioisotopes were imported from Oak Ridge National Laboratory, USA or Radiochemical Centre, Amersham, UK. In some cases the chemical species of tracers could not be clearely defined and the most common species were chosen as far as possible. However, in other cases the definite oxidation states were attained as is given in TABLE 3. The scrubbing techniques were often used in these cases in order to get definite K_d values. The oxidation states defined are shown in Figs. 1.1~6.3. Many other details concerning radioactive tracers are read in the references.[1)-16)]

2.3 Determination of K_d values

The distribution ratios, K_d values, were determined radiometrically in the ordinary way[1)]. The organic phase was pre-equilibrated with the corresponding aqueous acid solutions except for tributyl phosphine oxide, dodecylbenzensulfonic acid, tetraethyl-benzyl-ammonium chloride, dimethyl-benzyl-phenyl-ammonium chloride and tetraphenylarsonium chloride extraction systems, in which the aqueous phase was saturated with the organic extractant before determination of K_d values.

3 TABLES OF ACID DEPENDENCE CURVES

Results obtained are summarized in Figs. 1.1~6.3 as a series of graphs of $\log K_d$ vs. N HCl or N HNO_3, where K_d is the distribution ratio and N the normality of acids.

Figures 7~13 show the exact K_d values for some solvent extraction systems.

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TABLE 1 Extracting agents

	Reagents	Sources	Concentration	Diluent	Aq. phase	References
1-1	Tri-n-butyl phosphate (TBP)	Commercial TBP purified	100%	—	HCl	1
2	〃	〃	50	Toluene	〃	2
3	〃	〃	25	〃	〃	2
4	〃	〃	10	〃	〃	2
5	〃	〃	100	—	HNO_3	3, 4
6	〃	〃	25	CCl_4	〃	
2-1	Tri-n-butyl phosphine oxide (TBPO)	Tama Chem. Co.,	1	Toluene	HCl	5
2	Tri-n-octyl phosphine oxide (TOPO)	Eastman Chemicals Co.,	5	〃	〃	6, 4
3	〃	〃	5	〃	HNO_3	6, 4, 7
3-1	Tetrabutyl methylene diphosphonate (TBMDP)	Tama Chem. Co.,	10	Xylene	〃	8
2	Tetrabutyl ethylene diphosphonate (TBEDP)	〃	10	〃	〃	8
4-1	Tri-iso-octyl amine (TIOA)	Union Carbide & Chem. Corp.,	5	〃	HCl	9
2	〃	〃	5	〃	HNO_3	10
3	Amberlite LA-1	Commercial	10	〃	HCl	9
4	〃	〃	10	〃	HNO_3	10
5	Primene JM-T	〃	10	〃	HCl	11
5-1	Bis- (2-ethyl hexyl) phosphate (HDEHP)	Virginia Chemicals Co.,	50	Toluene	〃	12
2	Dodecylbenzenesulfonic acid (DBSA)	Commercial	3	Ether+ethyl acetate (1 : 1)	〃	13
6-1	Triethyl-benzyl-ammonium chloride	Prepared	0.1M	$CHCl_3$	Ammonium thiocyanate	14
2	Dimethyl-benzyl-phenyl-ammonium chloride	Prepared	0.1M	$CHCl_3$	HCl	15
3	Tetraphenylarsonium chloride	Commercial reagents recrystalized	0.05M	$CHCl_3$	〃	16

TABLE 2 Radioisotopes used

Tracer	Target (irradiated in) and chemical treatment
^{24}Na	Na_2CO_3 (JRR-1) *1
^{27}Mg	Mg metal (JRR-1)
^{28}Al	Al metal (JRR-1)
^{32}P	Interim Facilities for Radioisotope Production, JAERI
^{35}S	〃
^{38}Cl	NH_4Cl (JRR-1)
^{42}K	KNO_3 (JRR-1)
^{45}Ca	Imported*2
^{46}Sc	Imported ; Sc_2O_3 (JRR-2) *3
^{51}Ti	Ti hydroxide (JRR-1)
^{52}V	NH_4VO_3 (JRR-1)
^{51}Cr	Interim Facilities for Radioisotope Production, JAERI
^{56}Mn	MnO_2 (JRR-1)
$^{55,59}Fe$	Imported
^{60}Co	Imported
^{65}Ni	Ni sponge (JRR-1), separated from Co
^{64}Cu	Cu metal
^{65}Zn	Imported ; Zn metal (JRR-2)
^{72}Ga	Ga_2O_3 (JRR-1)
^{71}Ge	GeO_2 (JRR-2)
^{76}As	As_2O_3 (JRR-1)
^{75}Se	Imported
$^{80m,82}Br$	NH_4Br (JRR-1)
^{86}Rb	Imported
^{85}Sr	Imported
^{90}Sr	Imported
^{88}Y	Y_2O_3 (LINAC) *4
^{90}Y	Milked from ^{90}Sr-Y
^{91}Y	Imported
^{95}Zr-Nb	Imported
^{95}Nb	Imported ; milked from ^{95}Zr-Nb
^{99}Mo	Ammonium molybdate (JRR-1)
^{99m}Tc	Milked from ^{99}Mo-^{99m}Tc
^{99}Tc	Imported
^{106}Ru	Imported
^{109}Pd	Pd metal (JRR-1), purified from ^{111}Ag
^{108}Ag	$AgNO_3$ or Ag metal
^{110m}Ag	$AgNO_3$ (JRR-1)
^{115m}Cd	Imported
^{114m}In	Imported
^{113}Sn	Imported
^{124}Sb	Imported
$^{127,129}Te$	TeO_2 (JRR-2)
^{131}I	Interim Facilities for Radioisotope Production, JAERI

(Continued from the previous page)

Tracer	Target (irradiated in) and chemical treatment
^{137}Cs	Imported
^{139}Ba	$Ba(OH)_2 \cdot 2H_2O$ (JRR-1)
^{140}Ba	Imported
^{140}La	La_2O_3 (JRR-1)
^{144}Ce	Imported
^{147}Pm	Imported
^{152,154}Eu	Imported
^{177}Lu	Lu_2O_3 (JRR-1; JRR-2)
^{181}Hf	Imported
^{182}Ta	Imported
^{185}W	Imported
^{186,188}Re	Re metal (JRR-1)
^{191}Os	Imported
^{192}Ir	Imported
^{194}Ir	Ir sponge (JRR-1)
^{193,197,199}Pt	Pt sponge (JRR-1), separated from Au
^{198}Au	Au metal (JRR-1)
^{203}Hg	Imported
^{204}Tl	Imported
^{212}Pb	Radioactive deposit of Th
^{210}Bi	Milked from Ra DEF
^{244}Ra	Milked from ^{228}Th
^{231}Th	ThO_2 (LINAC), separated by TBP extraction
^{234}Th	Milked from uranyl nitrate
^{233}Pa	$Th(NO_3)_4$ (JRR-1), separated by TBP extraction
^{233}U	ThO_2 (JRR-1)
^{237}U	UO_2 (LINAC), separated by TBP extraction
^{239}Np	Uranyl nitrate, separated by TBP extraction
^{239}Pu	UO_2, separated by TBP extraction
^{241}Am	Imported
^{242}Cm	^{241}Am (JRR-1), separated by ion exchange

*1 neutron flux $= 10^{11} n/cm^2/sec$

*2 from ORNL, USA or from Radio Chemical Centre, Amersham, England

*3 neutron flux $= 10^{13} n/cm^2/sec$

*4 (γ, n) reaction was used

TABLE 3 Oxidation state and chemical treatment

Element and oxidation state	Treatment	
	in HCl	in HNO_3
Fe^{3+}	Oxidized with ceric ammonium nitrate	〃
As^{3+}	Reduced with ascorbic acid	〃
As^{5+}	Oxidized with $KBrO_3$ or $KClO_4$ in 6N HCl	〃
Se^{4+}	Warmed in 2∼4N HCl	〃
Sn^{4+}	Oxidized with $KClO_3$	〃
Sb^{3+}	Reduced with ascorbic acid	〃
Sb^{5+}	Oxidized with $KBrO_3$	〃
Hg^{2+}	Heated in conc. HNO_3	〃
Tl^{1+}	Reduced with hydroxylamine hydrochloride	〃
Tl^{3+}	Oxidized with $KBrO_3$	Ceric ammonium nitrate
Ce^{3+}	——	Reduced with hydroxylamine hydrochloride
Ce^{4+}	——	Ceric ammonium nitrate
Np^{4+}	Reduced with hydroquinone & KI	Ferrous sulfamate, or processed by TTA extraction
Np^{5+}	Warmed in dil. perchloric acid	〃
Np^{6+}	Oxidized with Cl_2 in water bath	Ceric ammonium nitrate

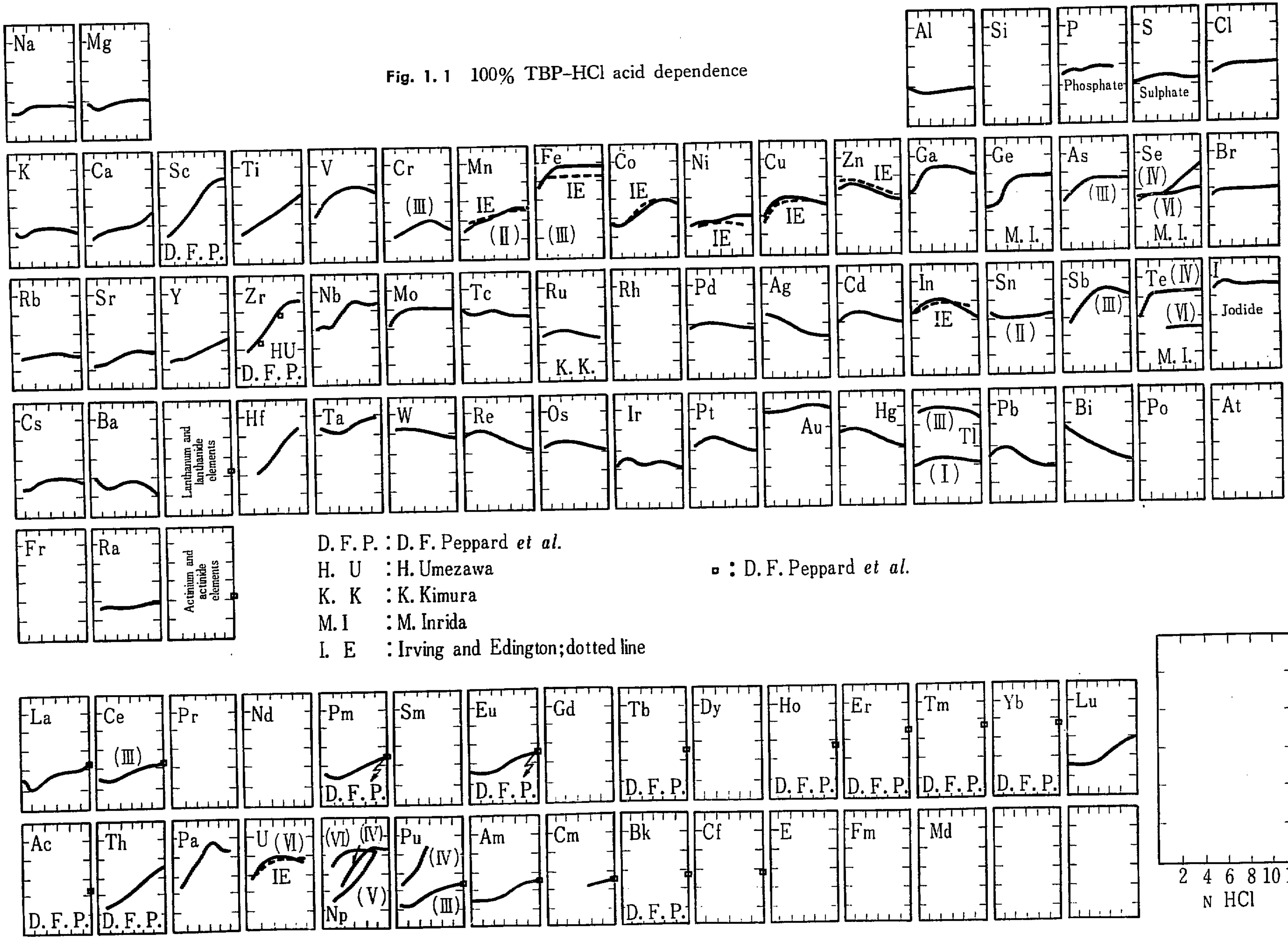

Fig. 1. 1 100% TBP–HCl acid dependence

D. F. P. : D. F. Peppard *et al.*
H. U : H. Umezawa
K. K : K. Kimura
M. I : M. Inrida
I. E : Irving and Edington; dotted line

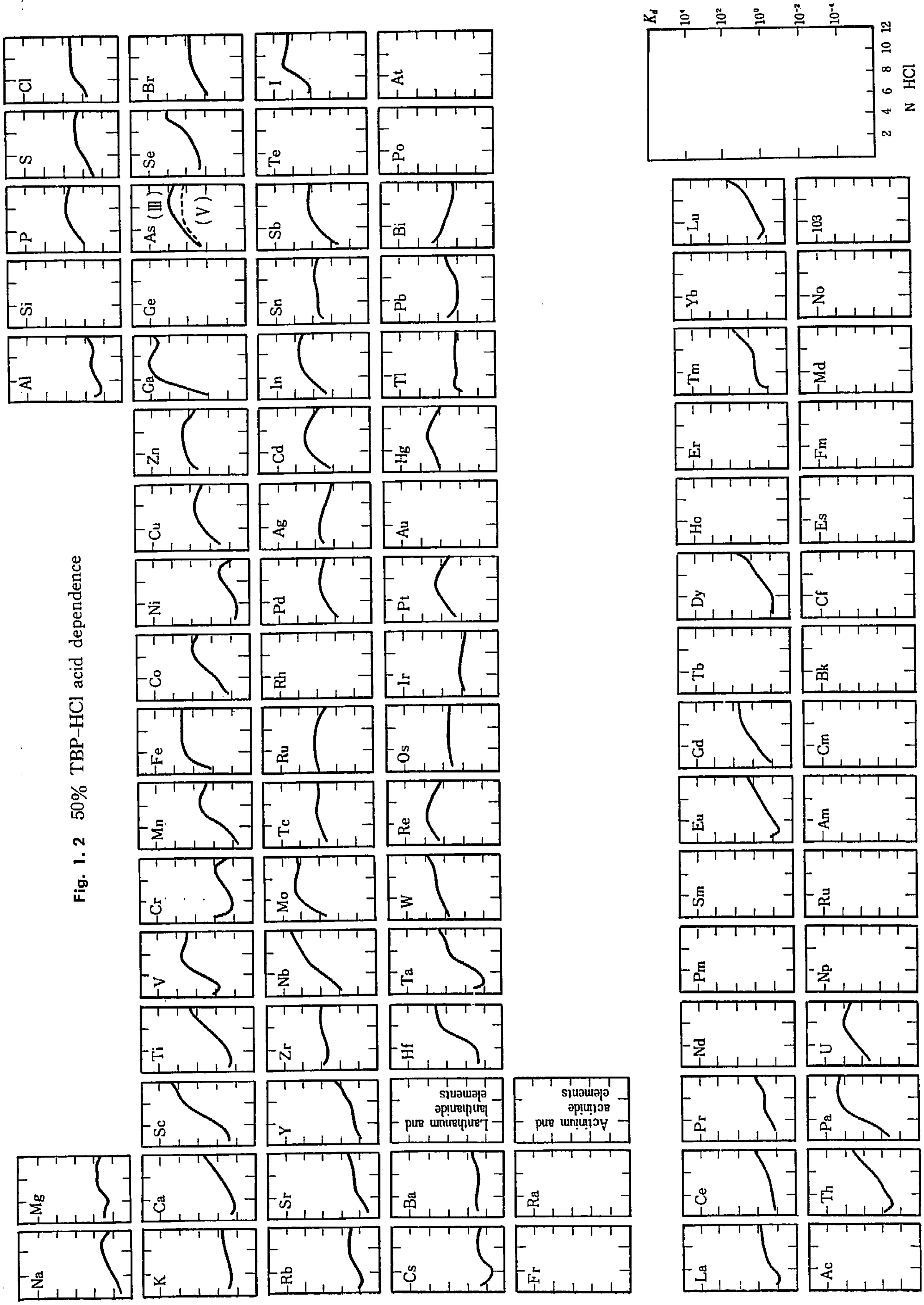

Fig. 1.2 50% TBP-HCl acid dependence

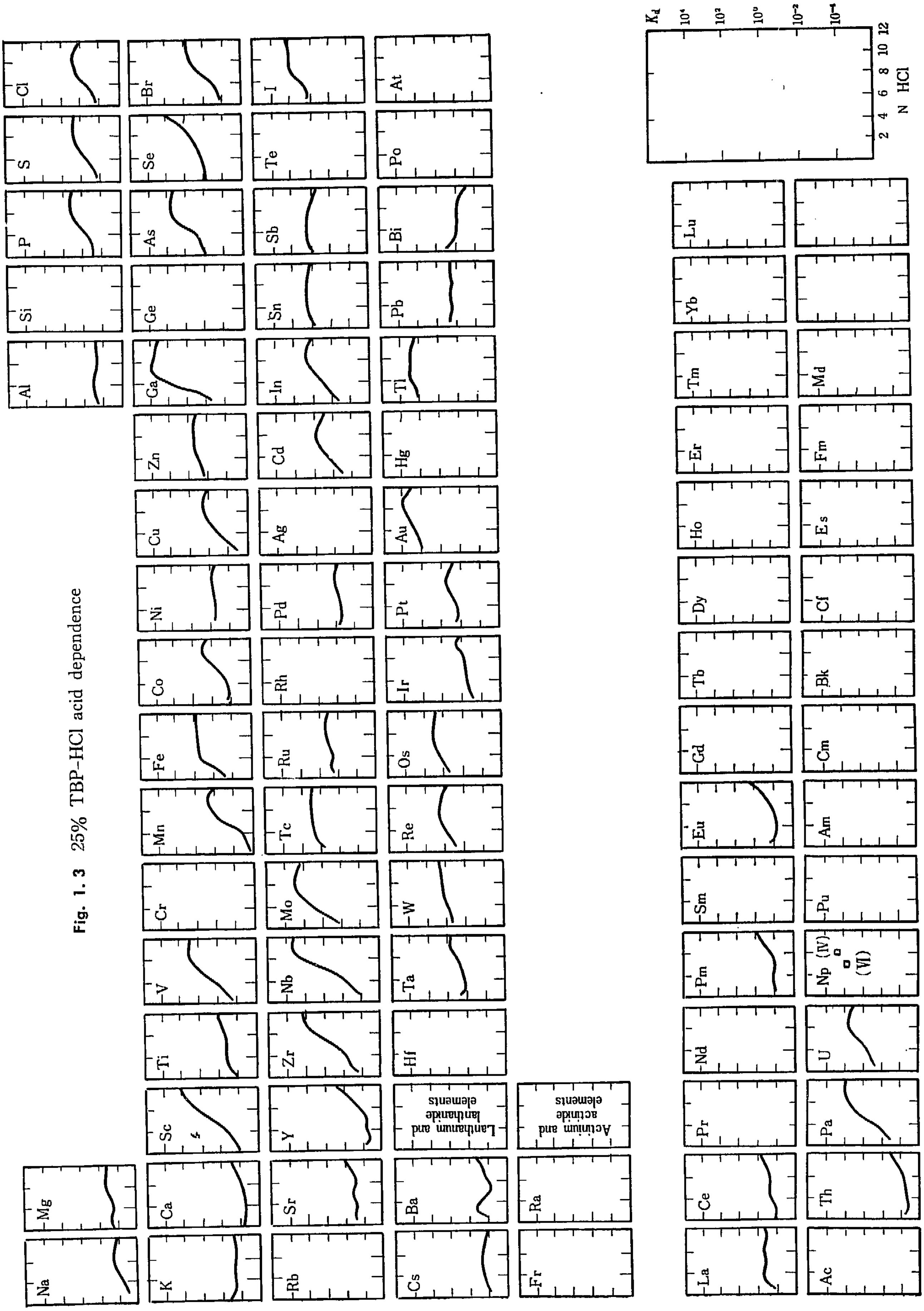

Fig. 1.3 25% TBP-HCl acid dependence

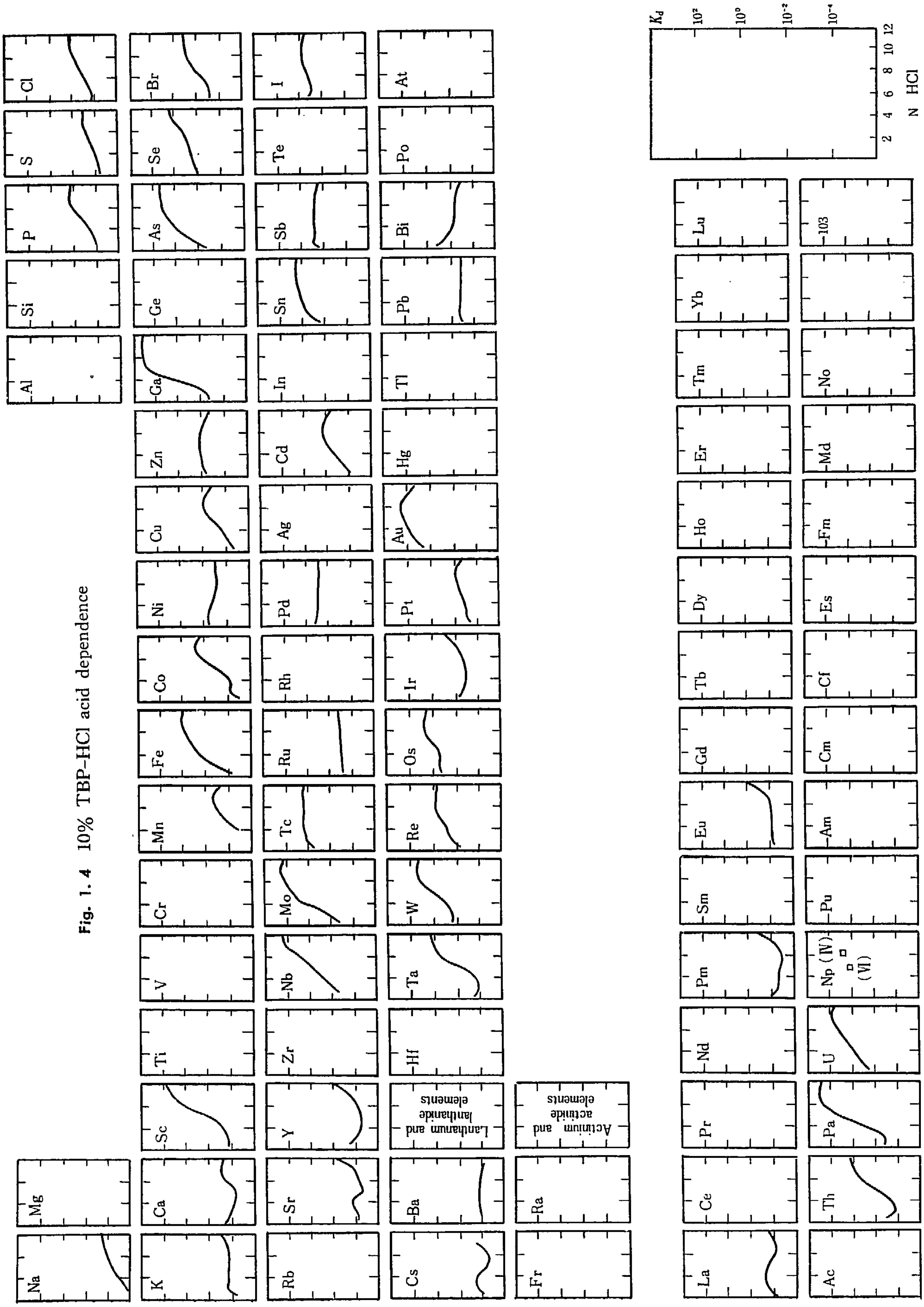

Fig. 1. 4 10% TBP-HCl acid dependence

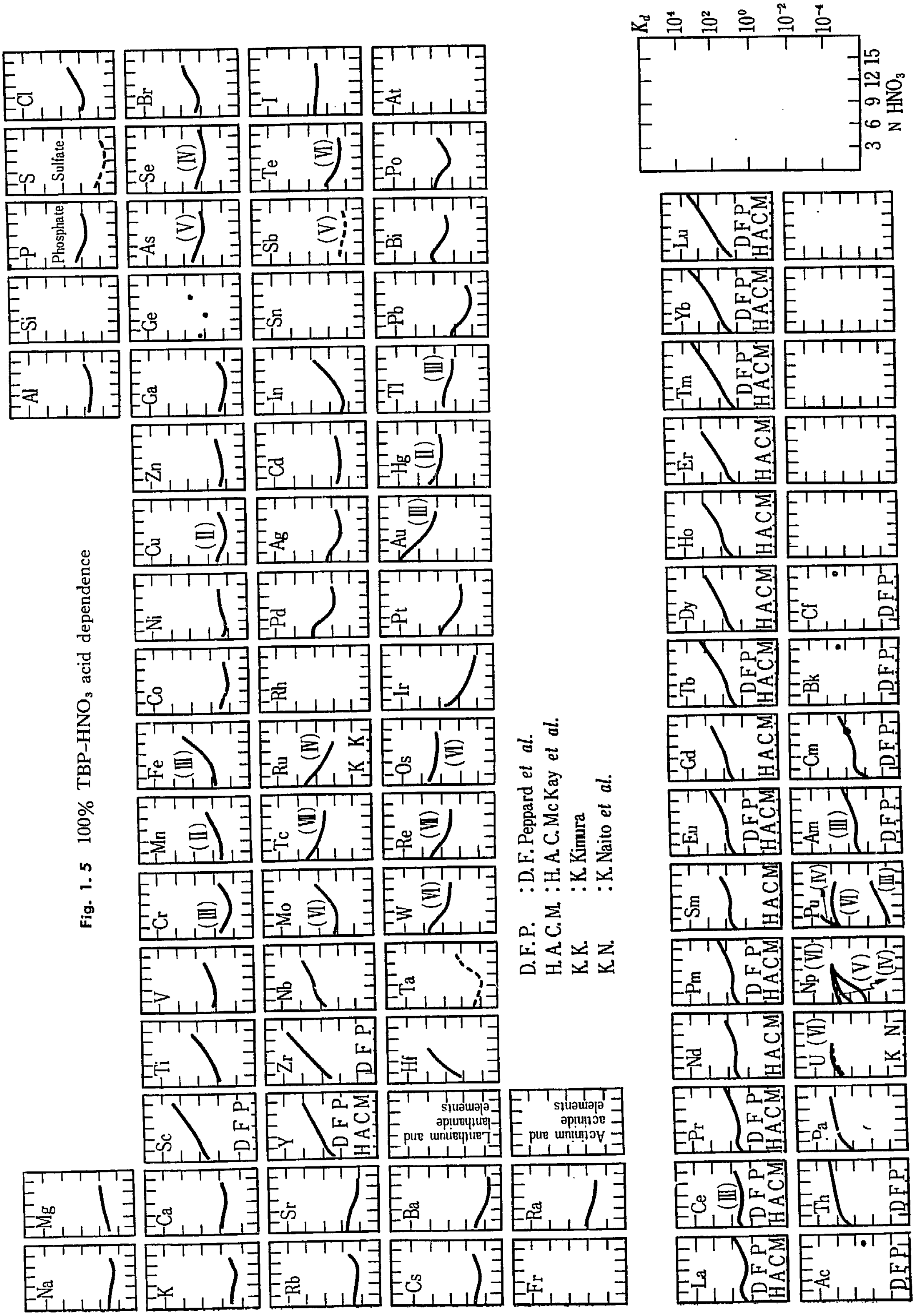

Fig. 1.5 100% TBP-HNO$_3$ acid dependence

D.F.P. : D.F. Peppard *et al.*
H.A.C.M. : H.A.C. McKay *et al.*
K.K. : K. Kimura
K.N. : K. Naito *et al.*

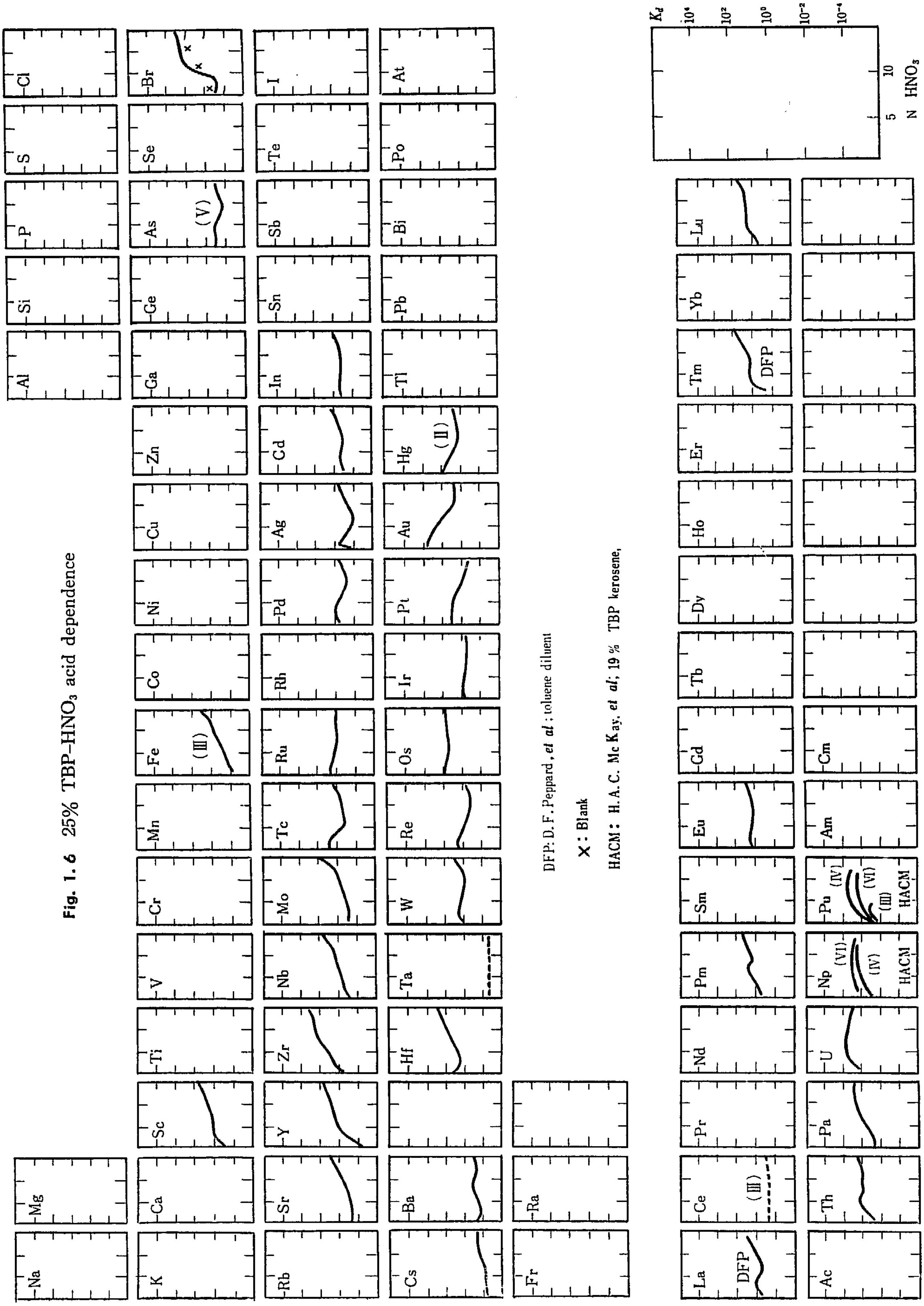

Fig. 1.6 25% TBP-HNO_3 acid dependence

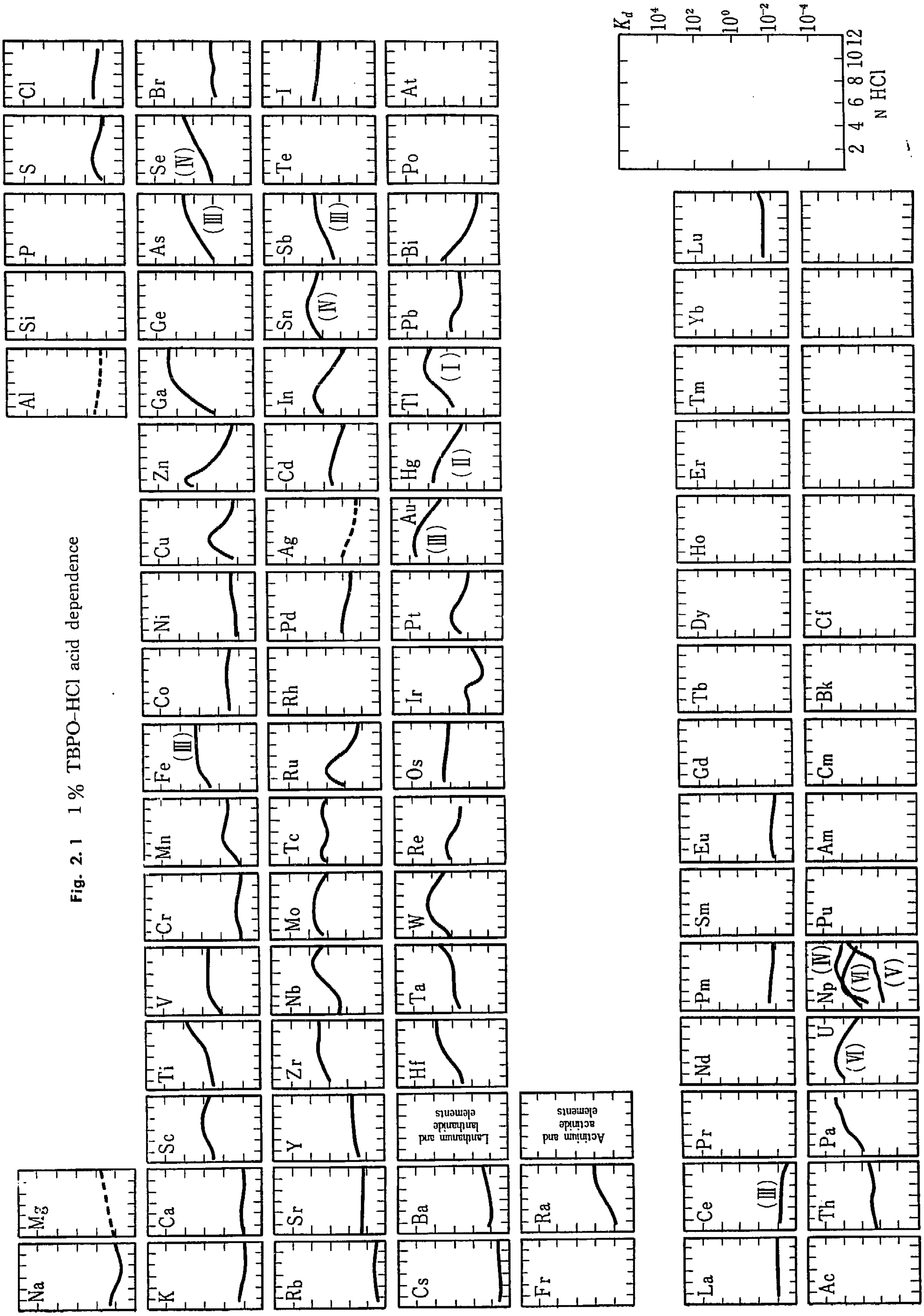

Fig. 2.1 1 % TBPO-HCl acid dependence

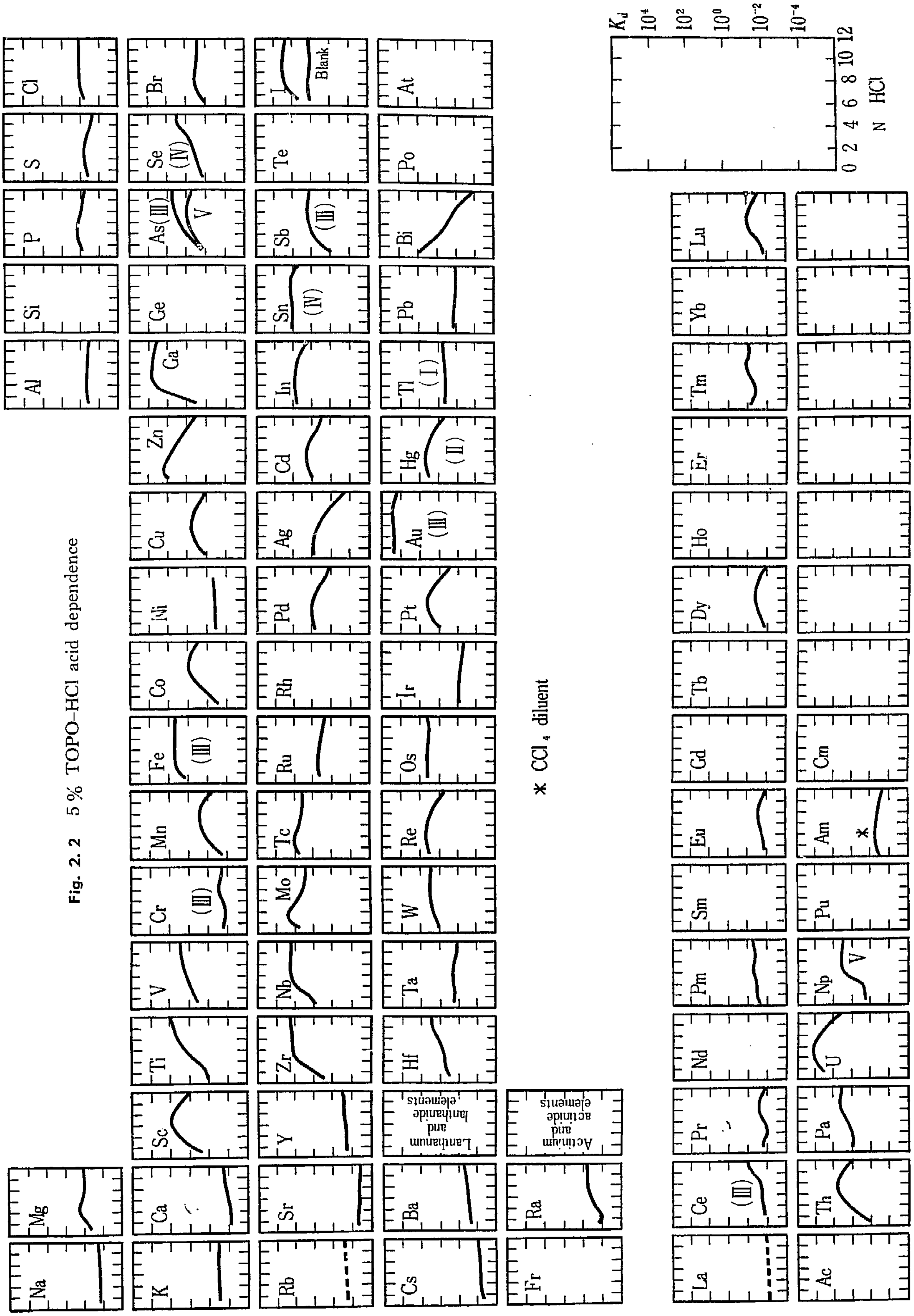

Fig. 2. 2 5 % TOPO-HCl acid dependence

* CCl_4 diluent

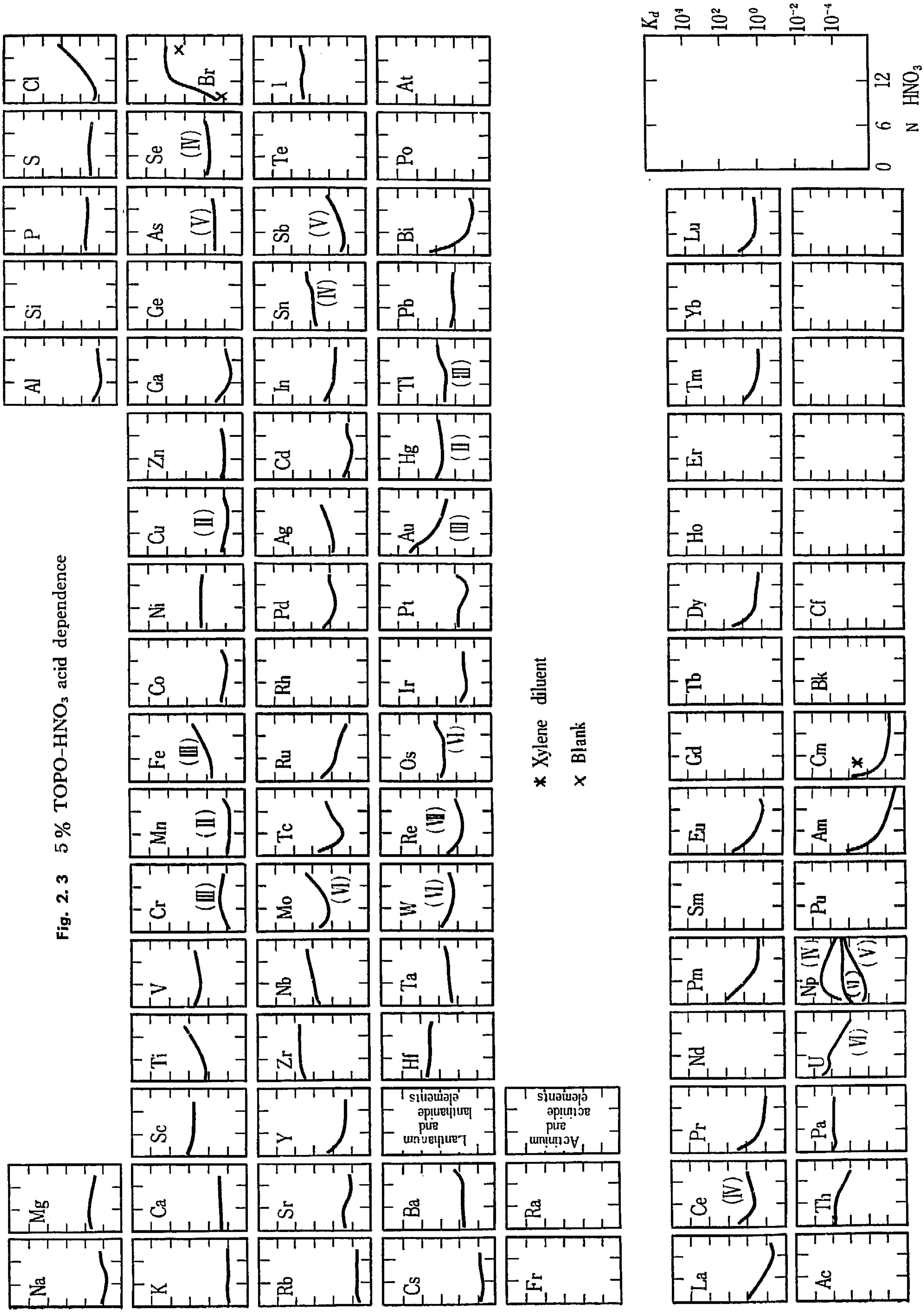

Fig. 2.3 5 % TOPO-HNO_3 acid dependence

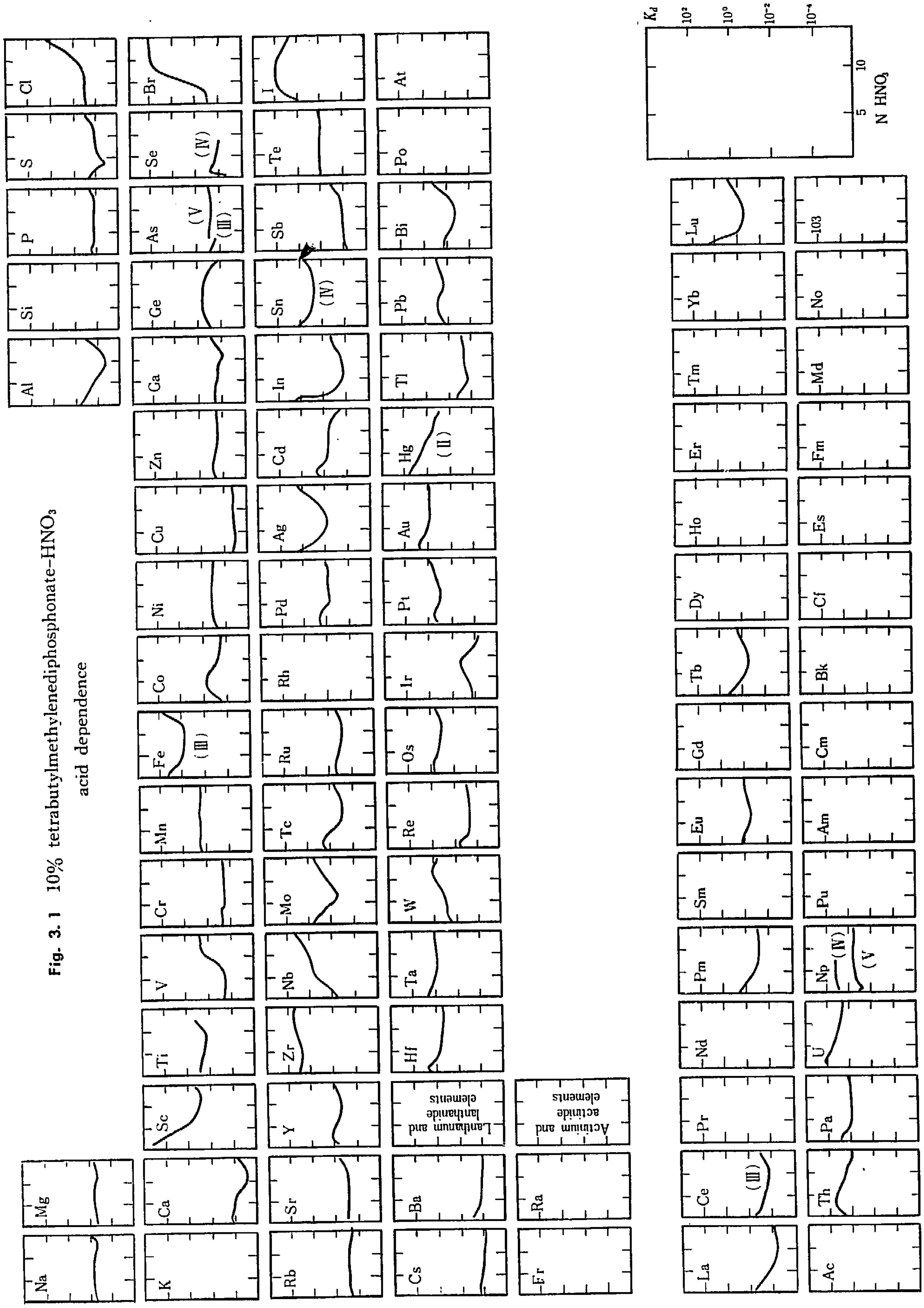

Fig. 3. 1 10% tetrabutylmethylenediphosphonate-HNO_3 acid dependence

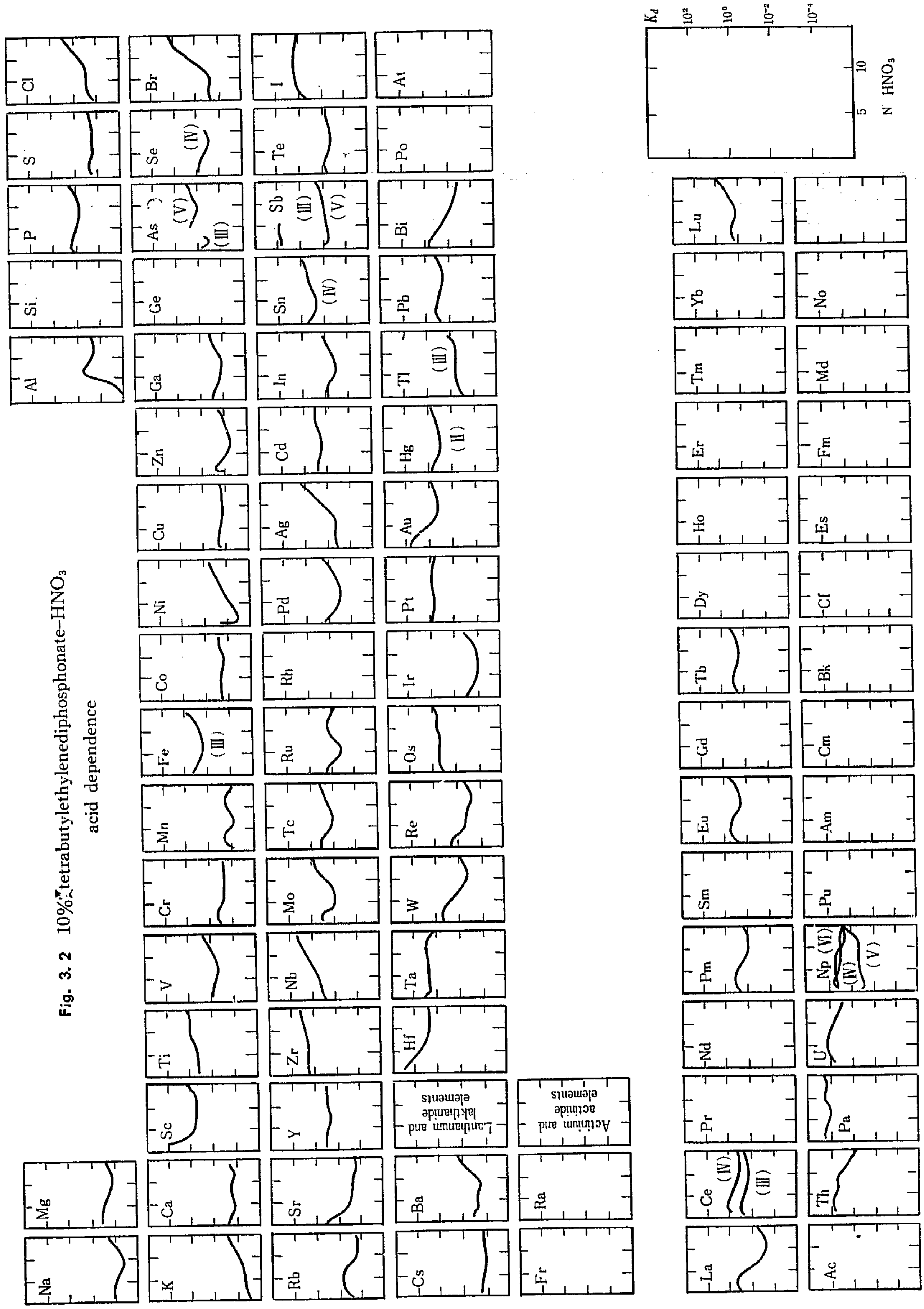

Fig. 3. 2 10% tetrabutylethylenediphosphonate–HNO_3 acid dependence

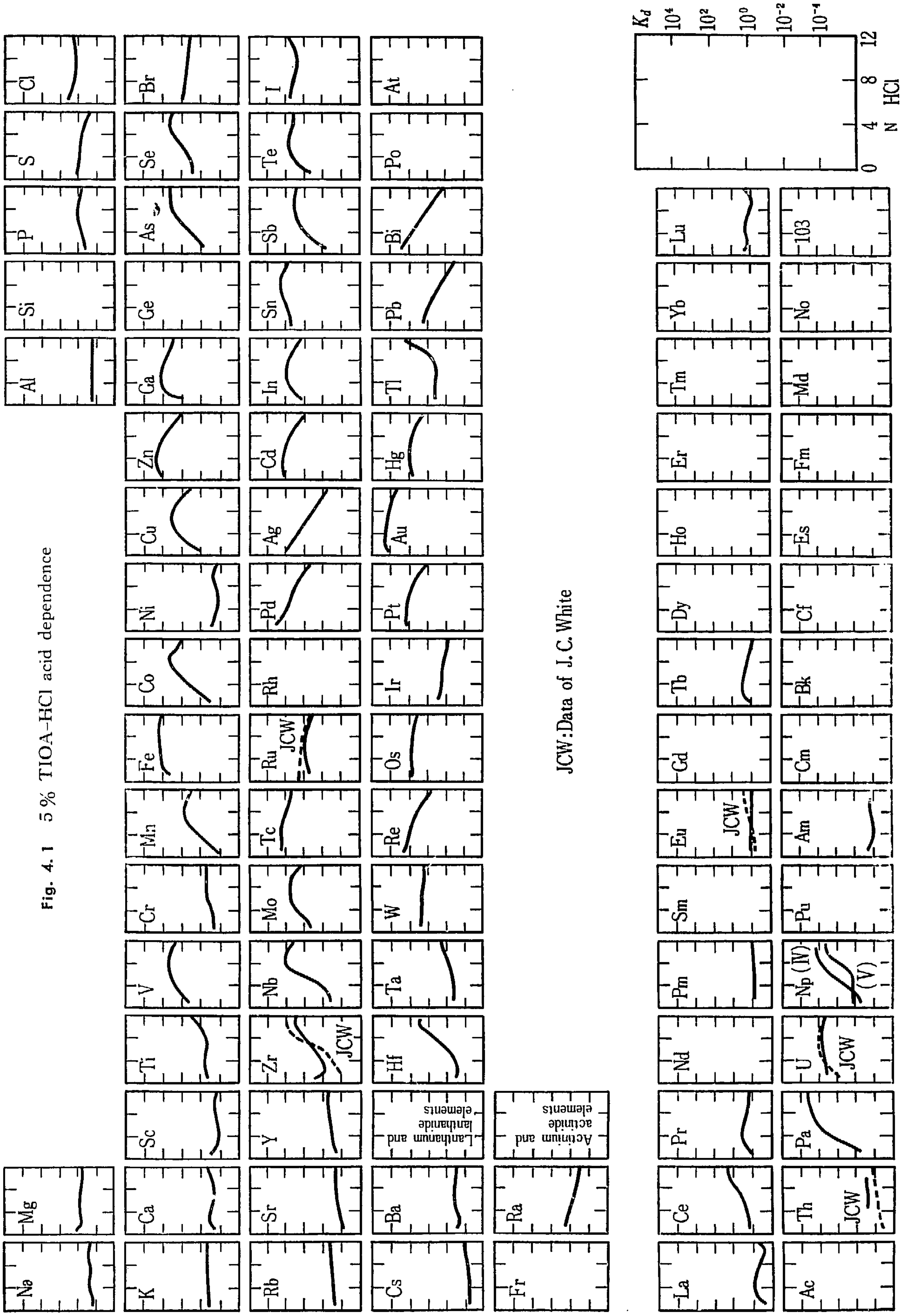

Fig. 4.1 5% TIOA-HCl acid dependence

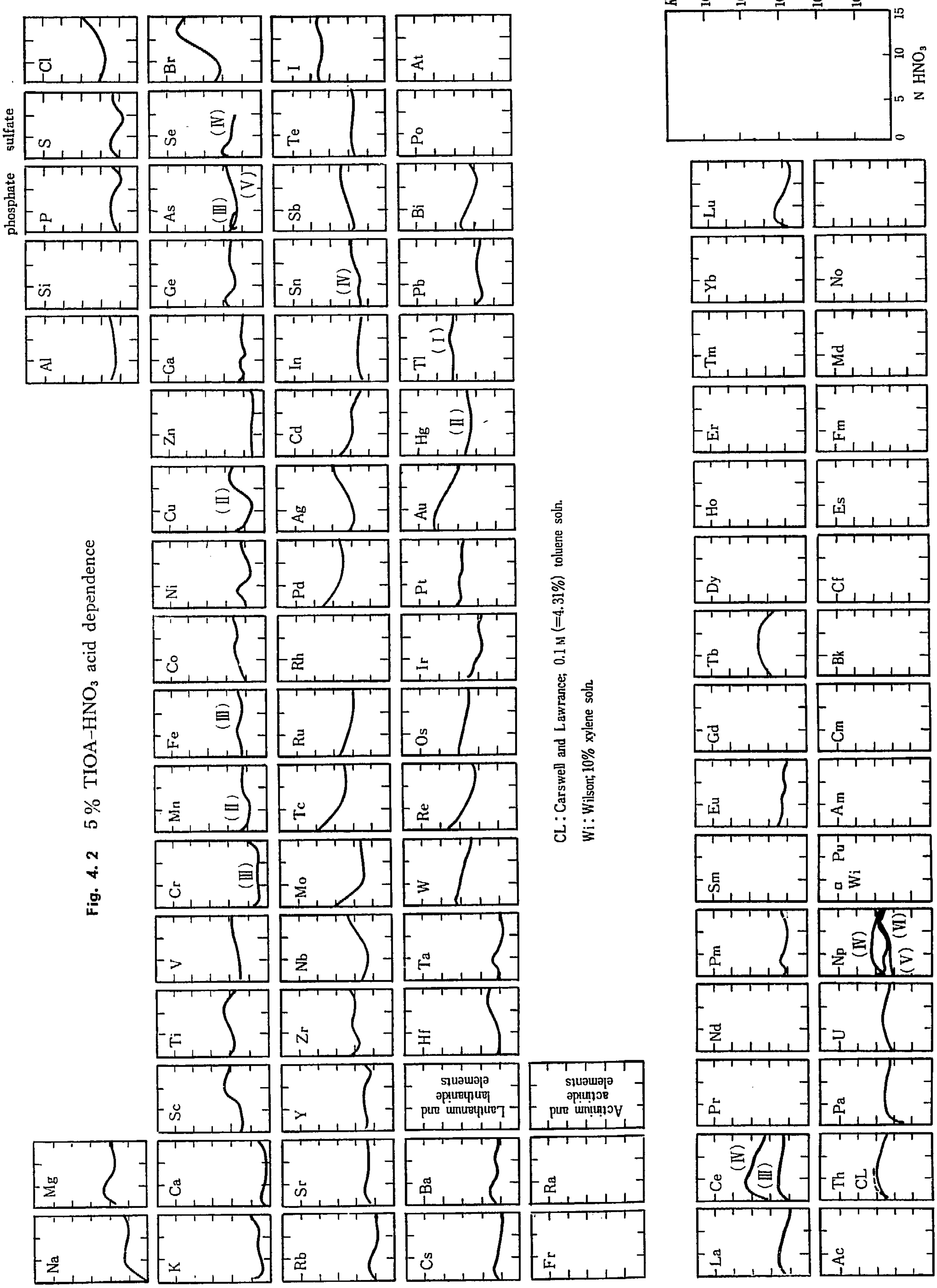

Fig. 4.2 5% TIOA-HNO_3 acid dependence

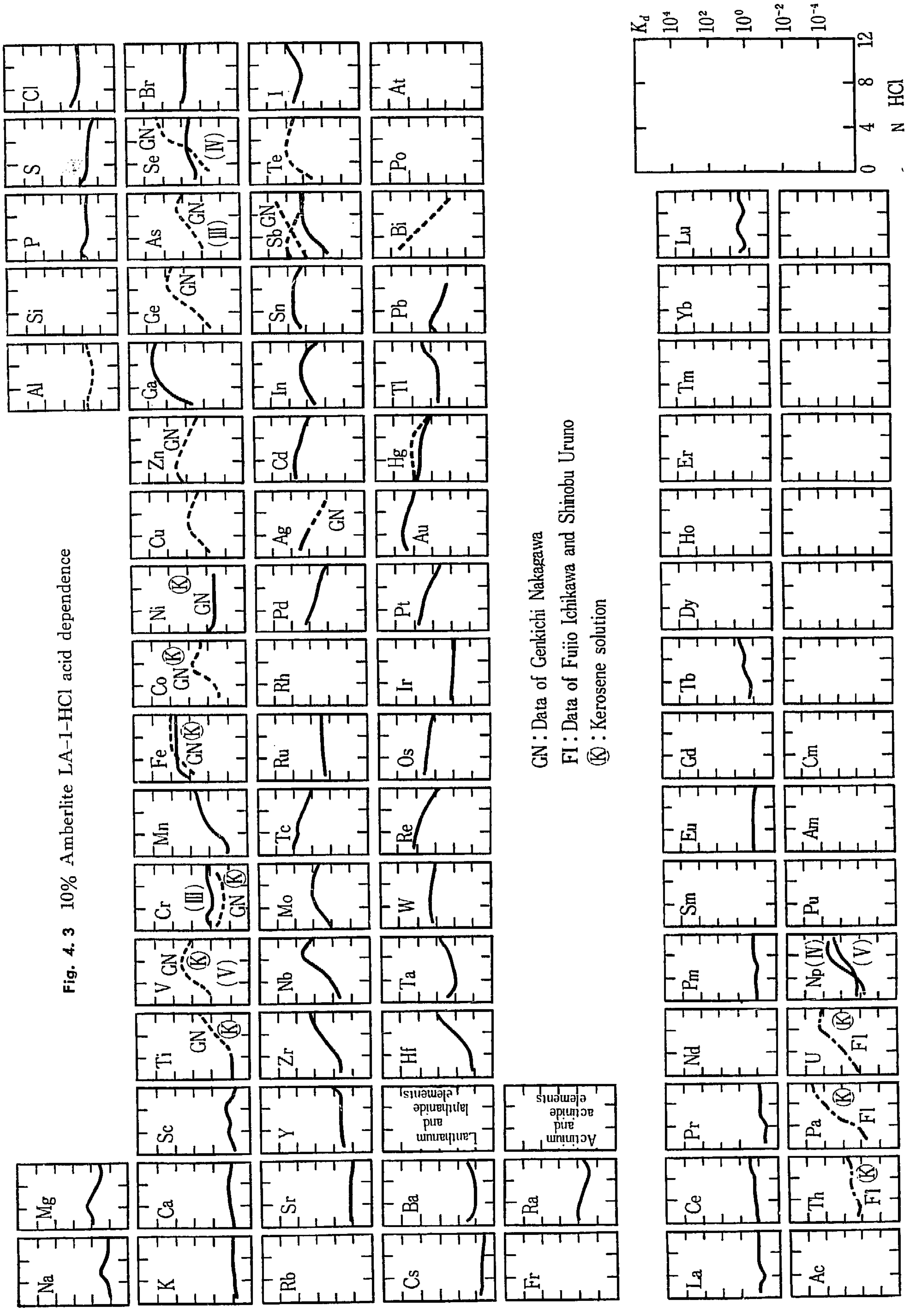

Fig. 4.3 10% Amberlite LA-1-HCl acid dependence

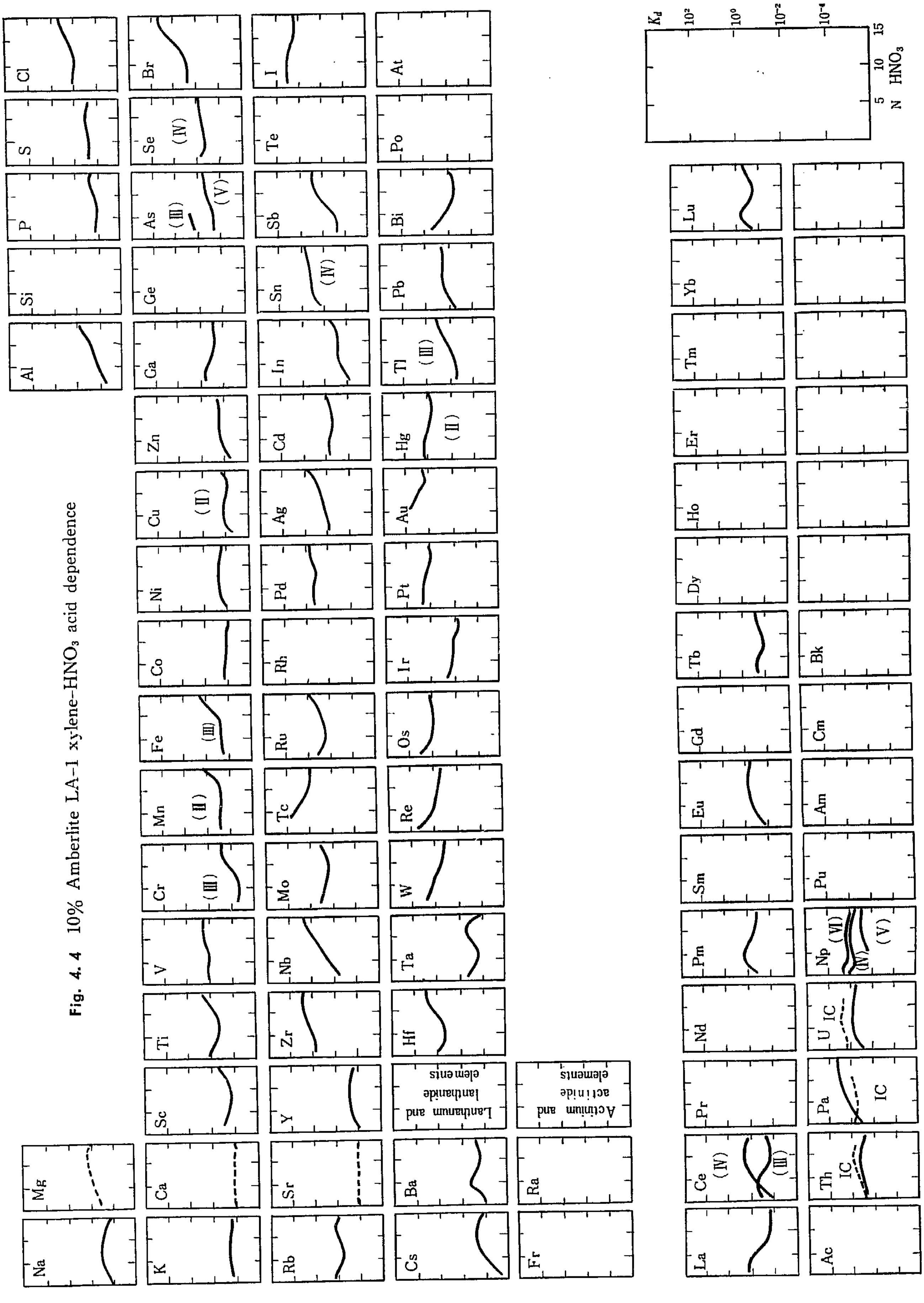

Fig. 4.4 10% Amberlite LA-1 xylene-HNO_3 acid dependence

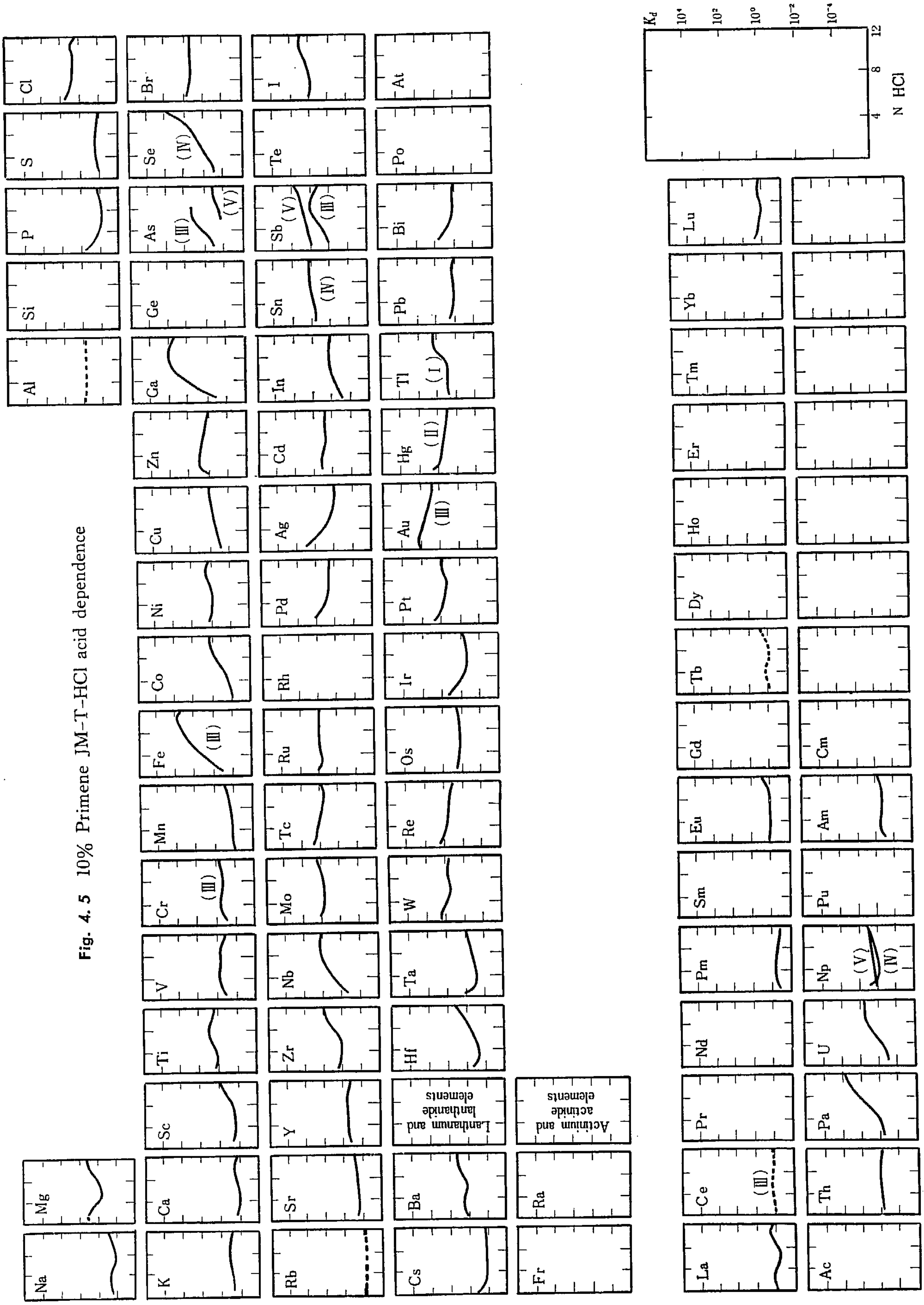

Fig. 4.5 10% Primene JM-T-HCl acid dependence

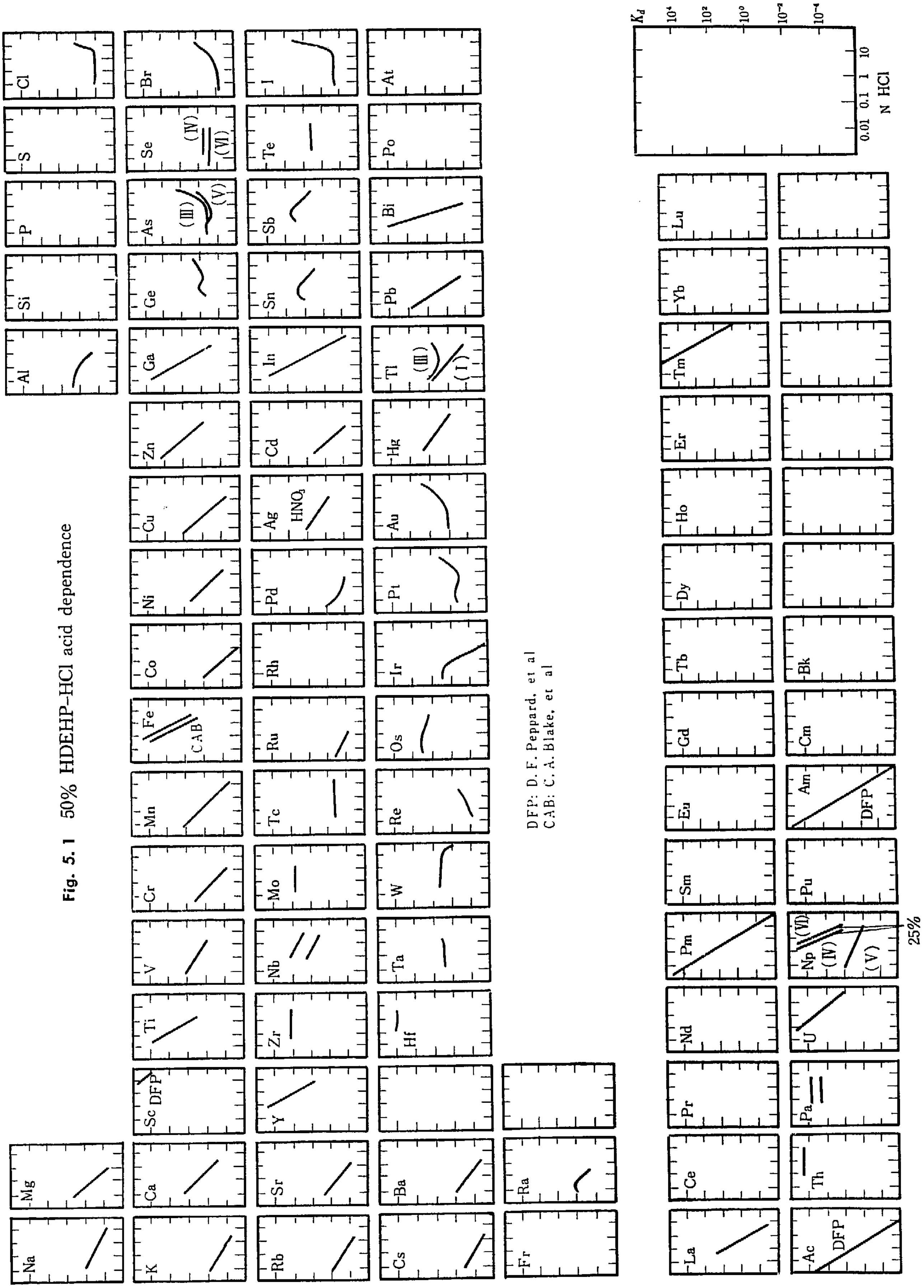

Fig. 5.1 50% HDEHP-HCl acid dependence

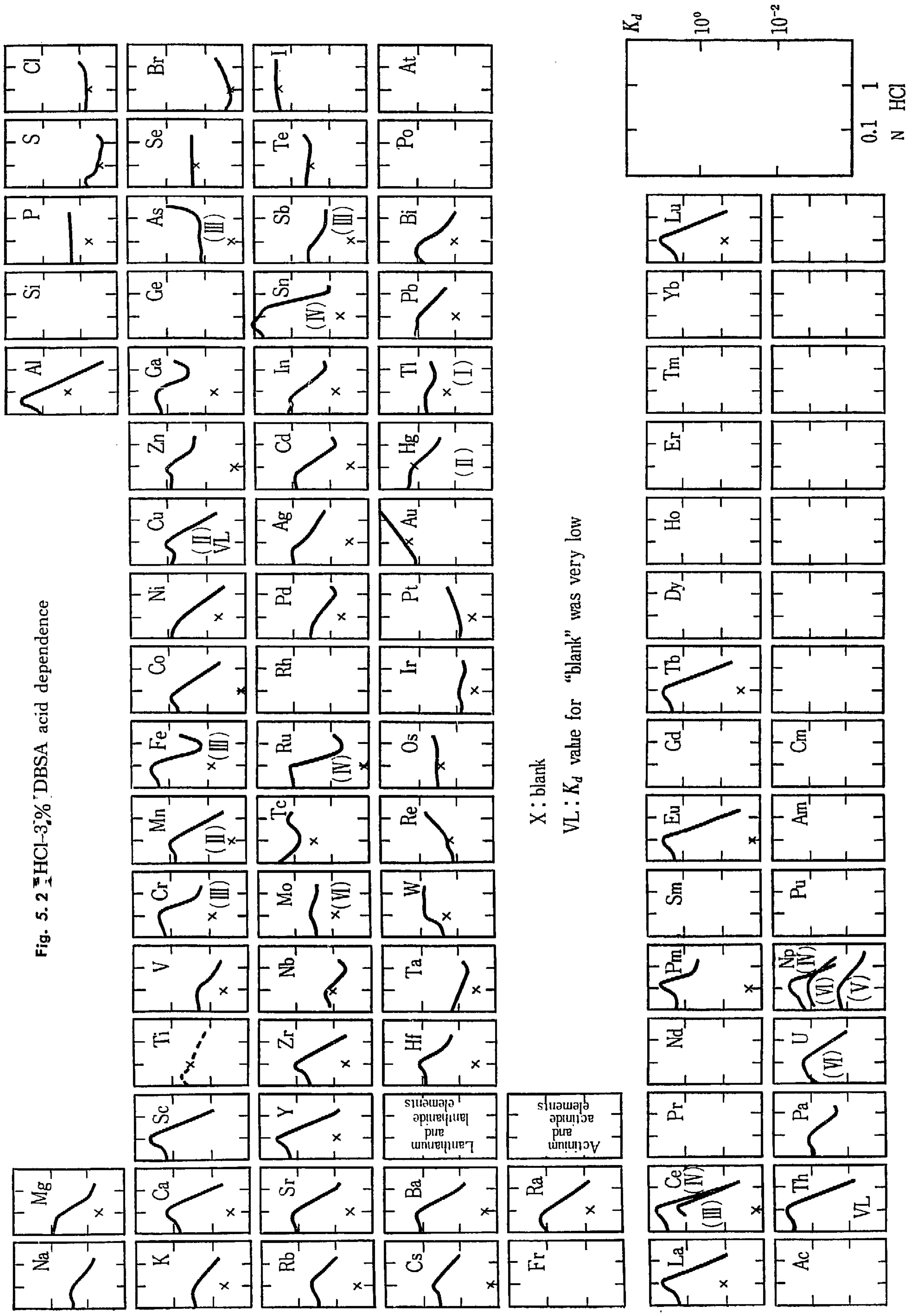

Fig. 5.2 HCl-3% DBSA acid dependence

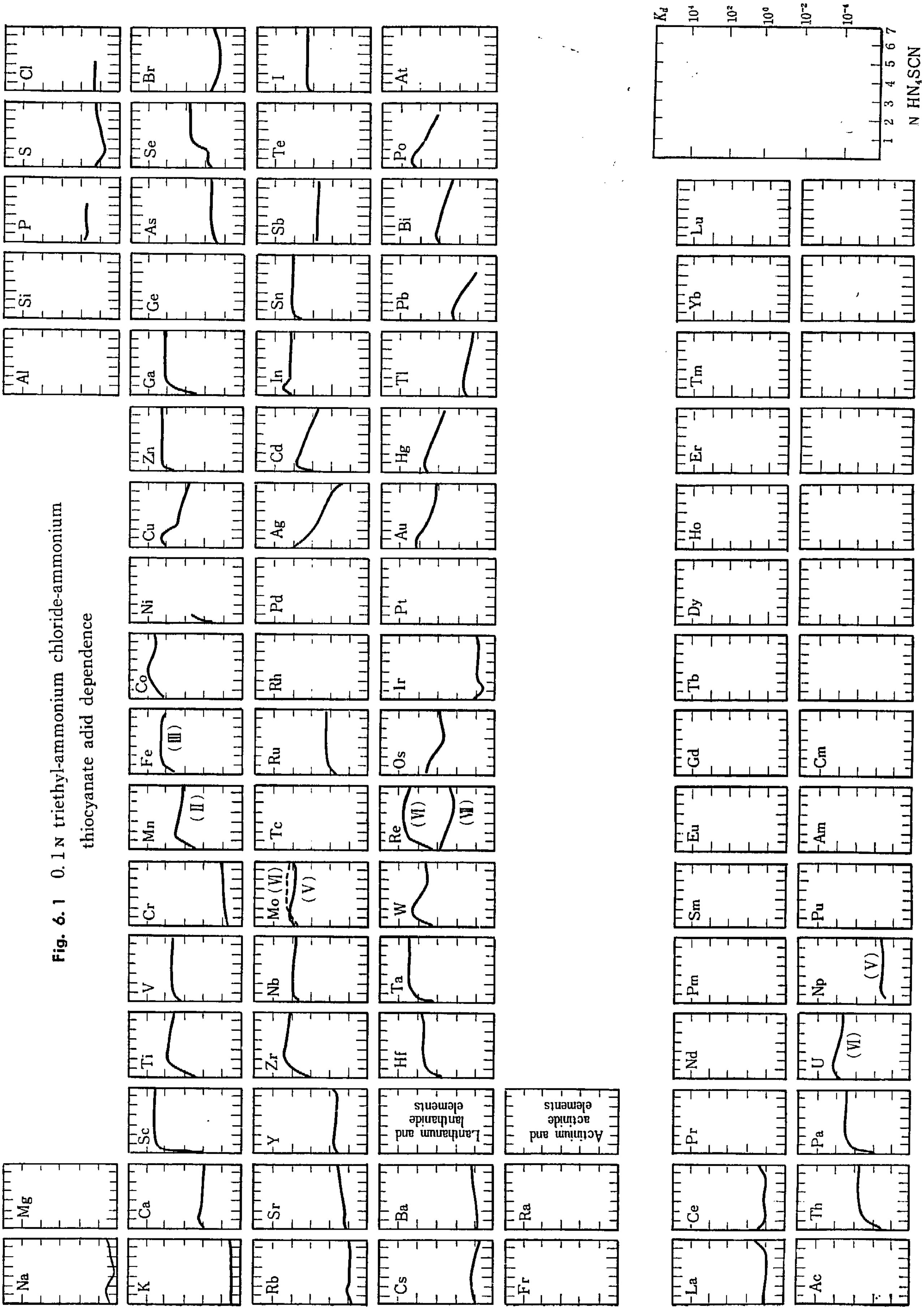

Fig. 6.1 0.1 N triethyl-ammonium chloride-ammonium thiocyanate adid dependence

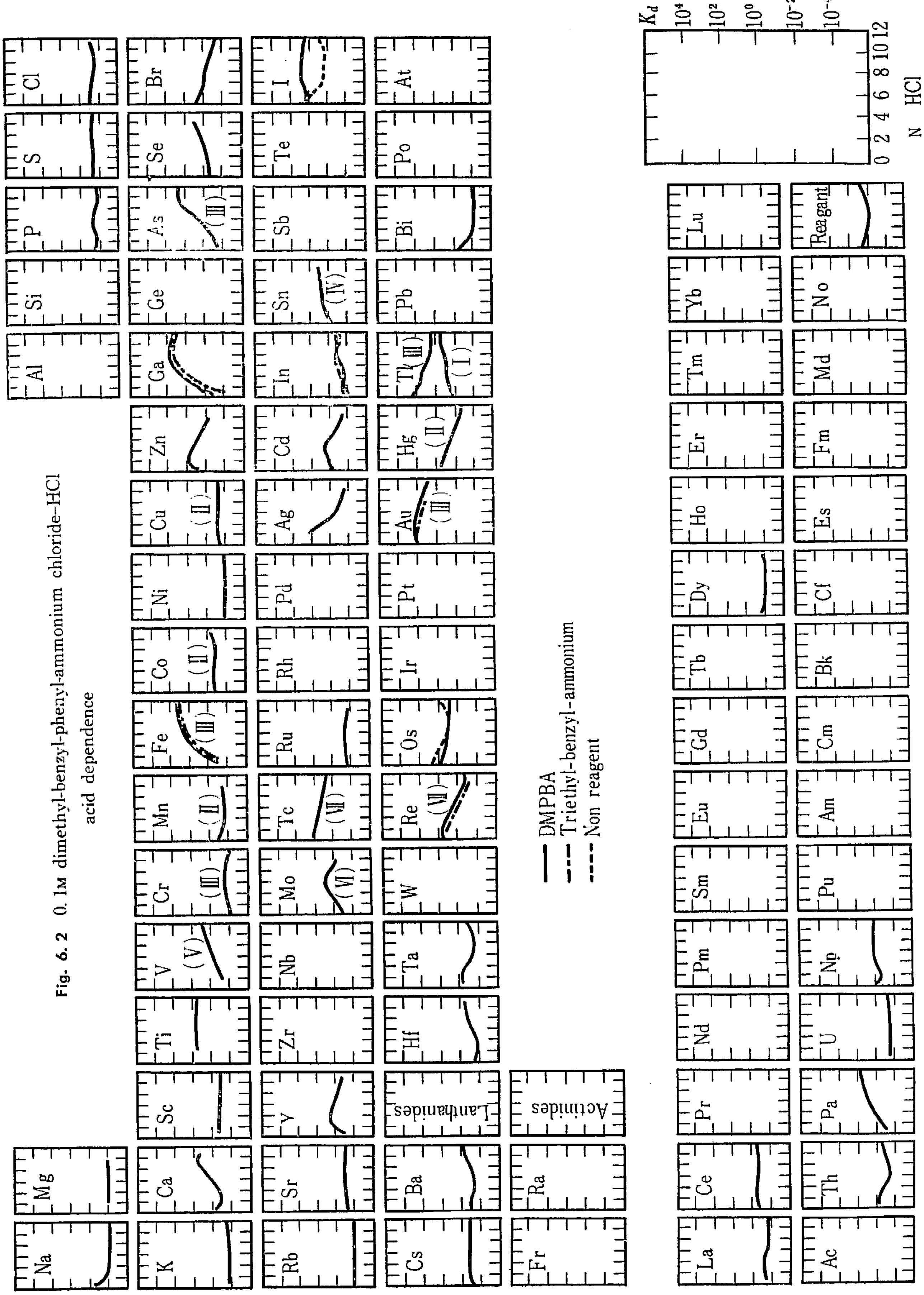

Fig. 6.2 0.1M dimethyl-benzyl-phenyl-ammonium chloride-HCl acid dependence

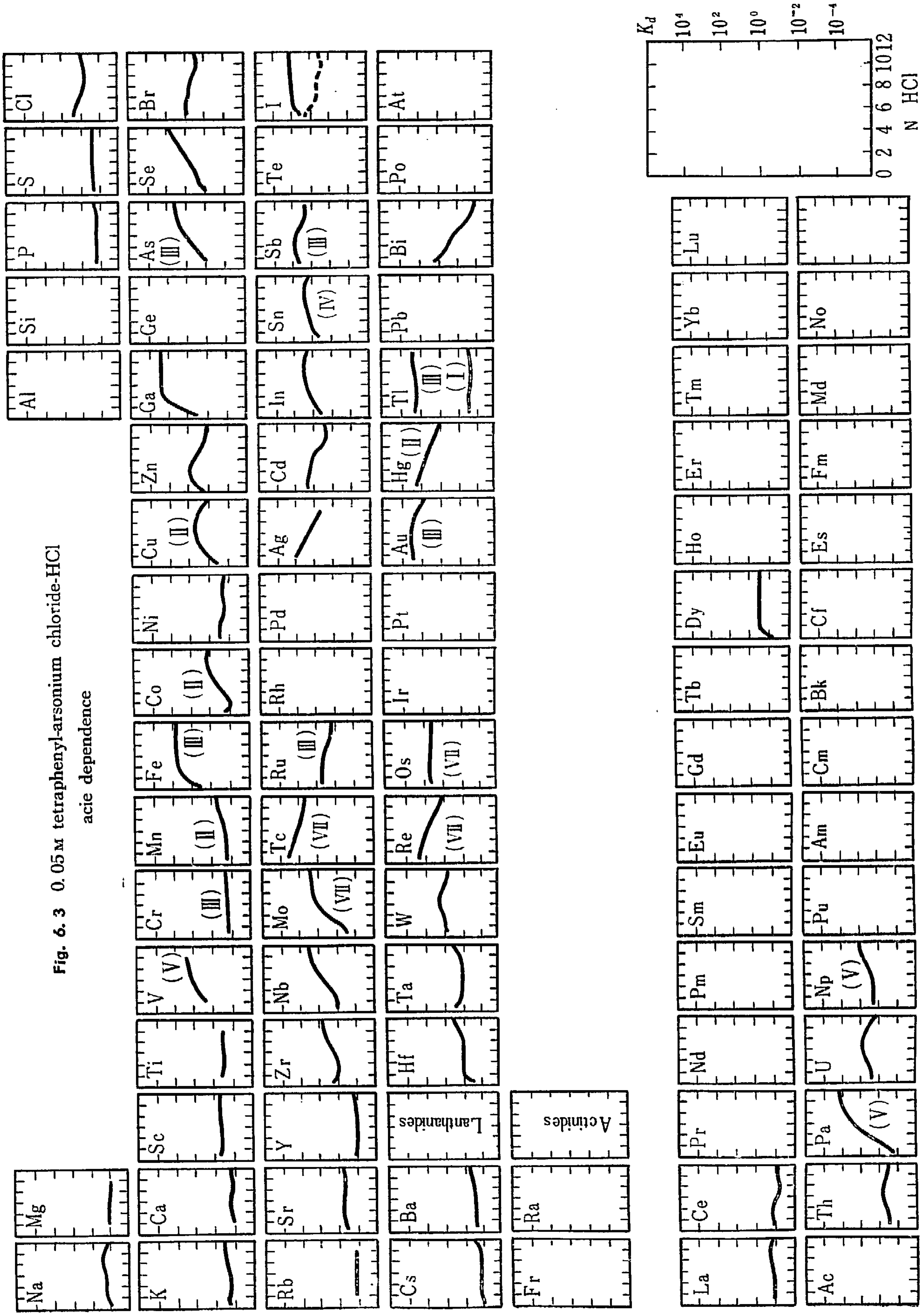

Fig. 6.3 0.05 M tetraphenyl-arsonium chloride-HCl acie dependence

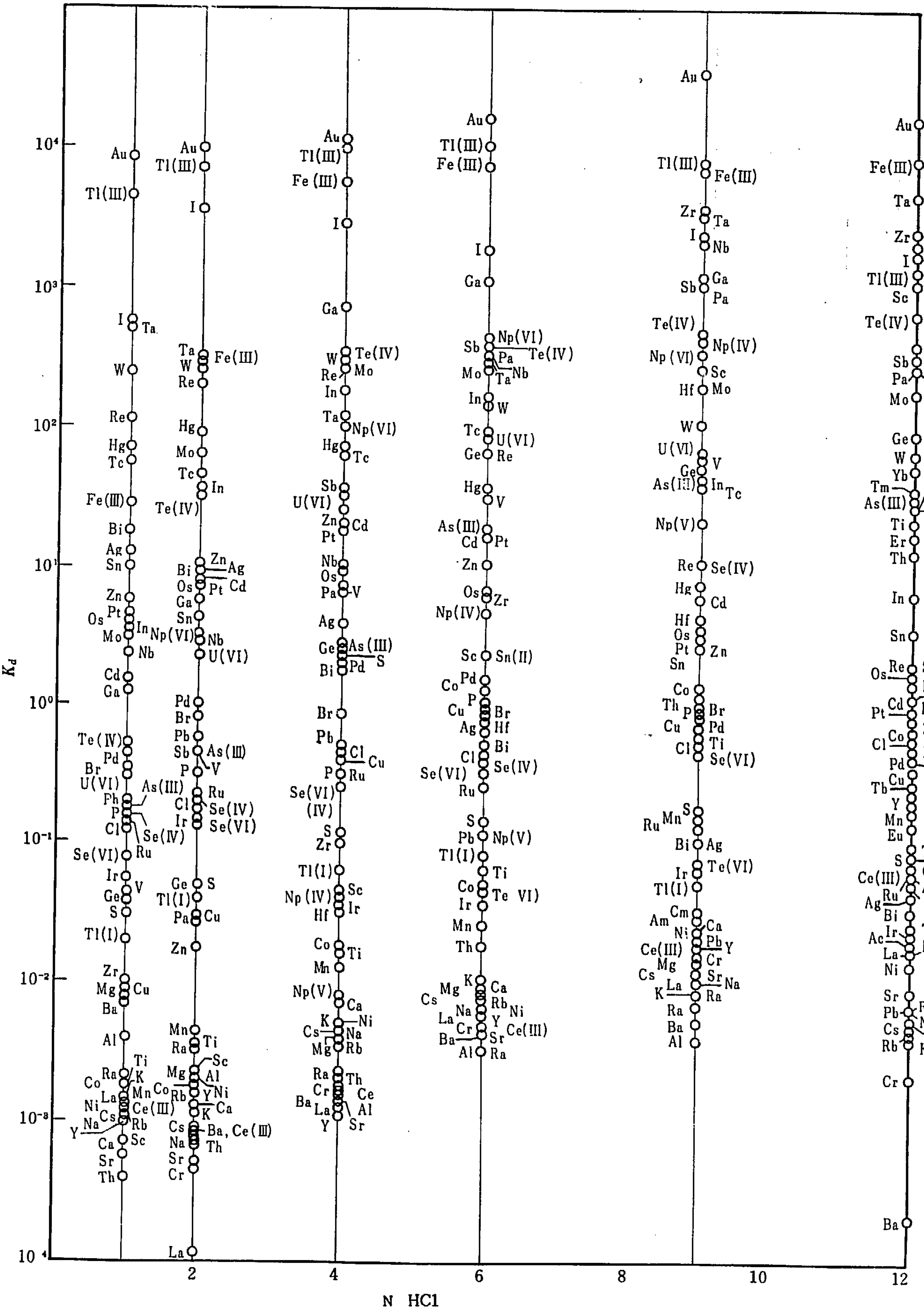

Fig. 7 K_d values in 100% TBP-HCl system

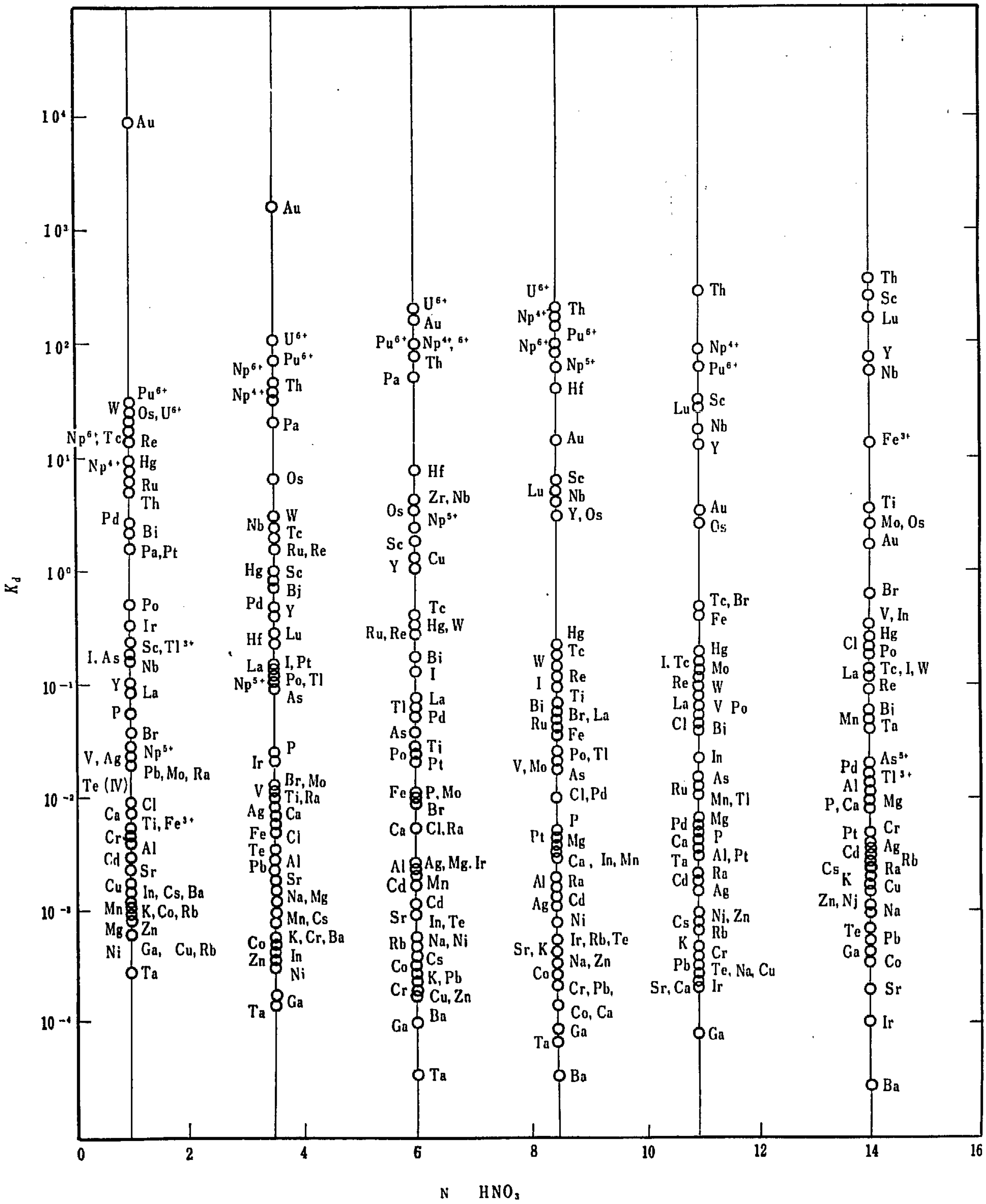

Fig. 8 Kd values in 100% TBP-HNO_3 system

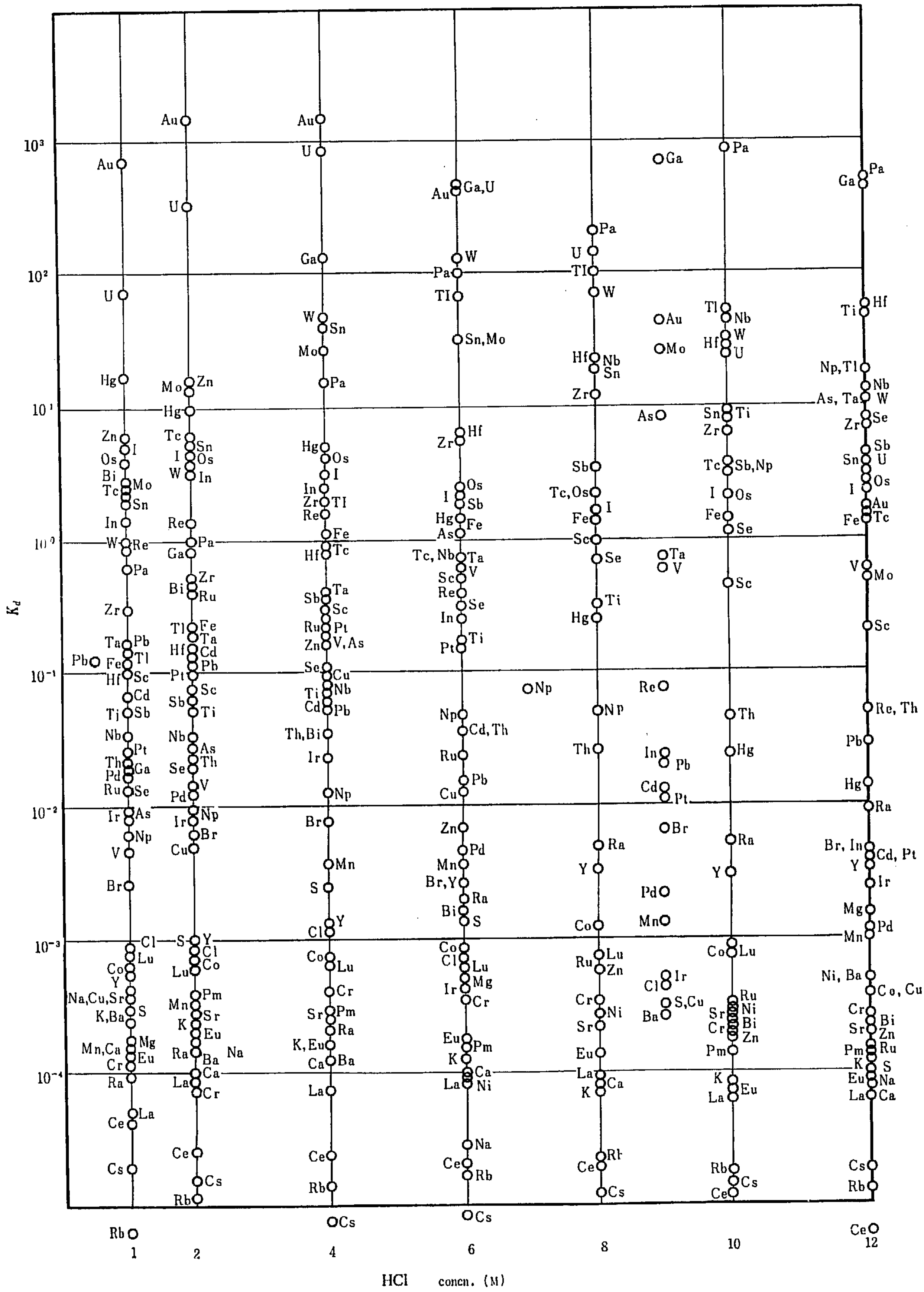

Fig. 9 K_d values in 1 % TBPO–HCl system

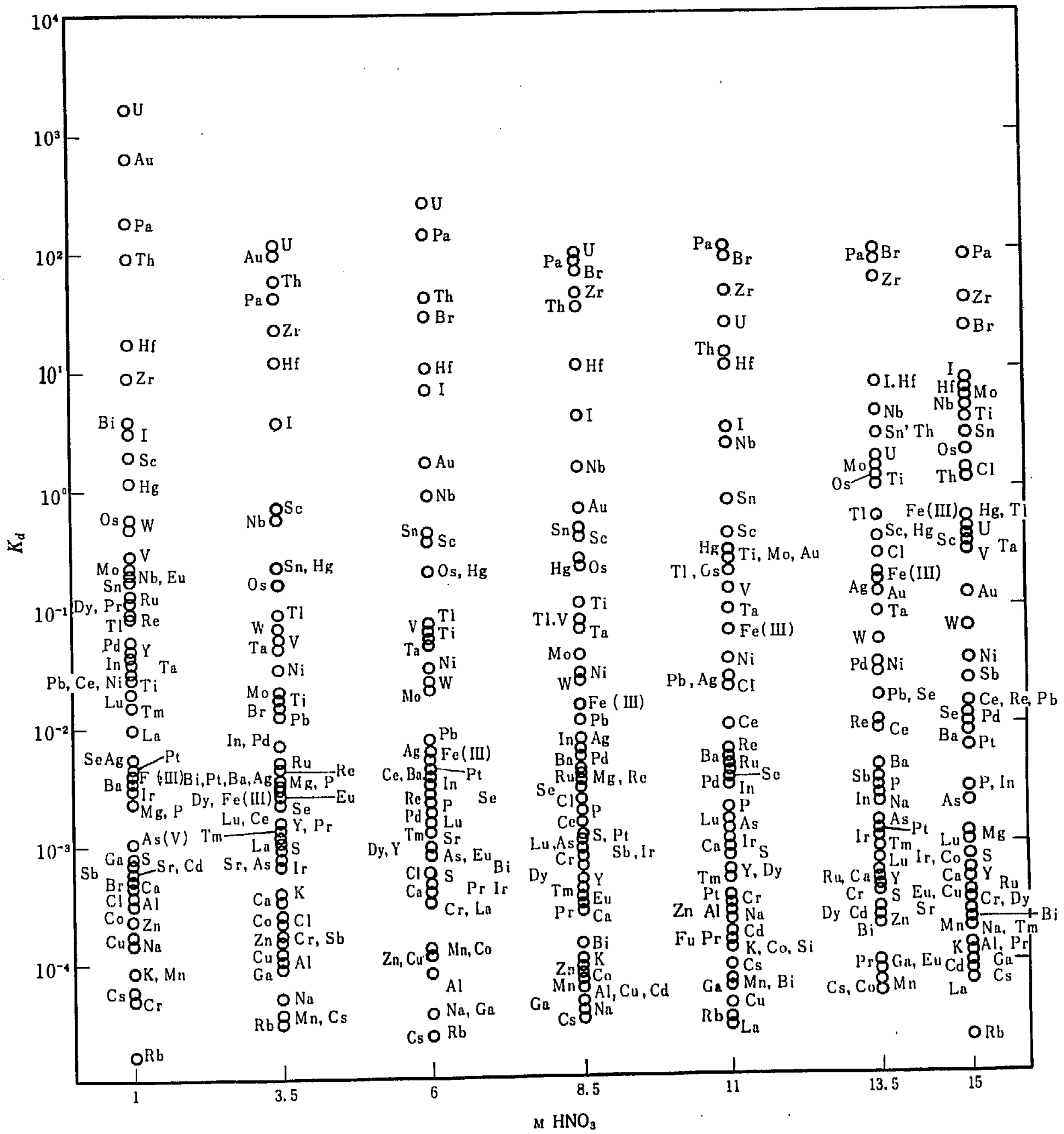

Fig. 10 K_d values in 5% TOPO-HNO_3 system

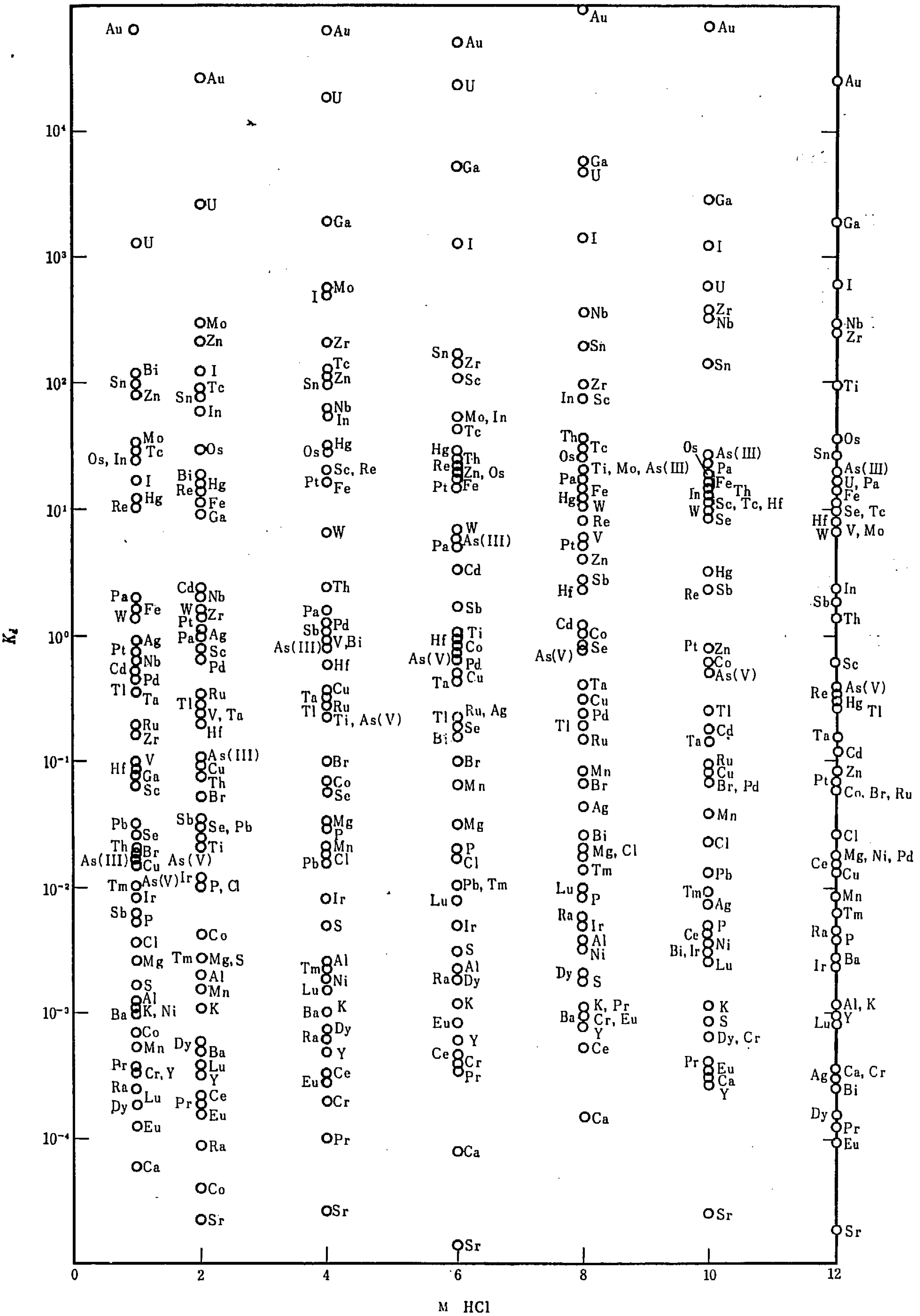

Fig. 11 K_d values in 5 % TOPO-1~12 M HCl system

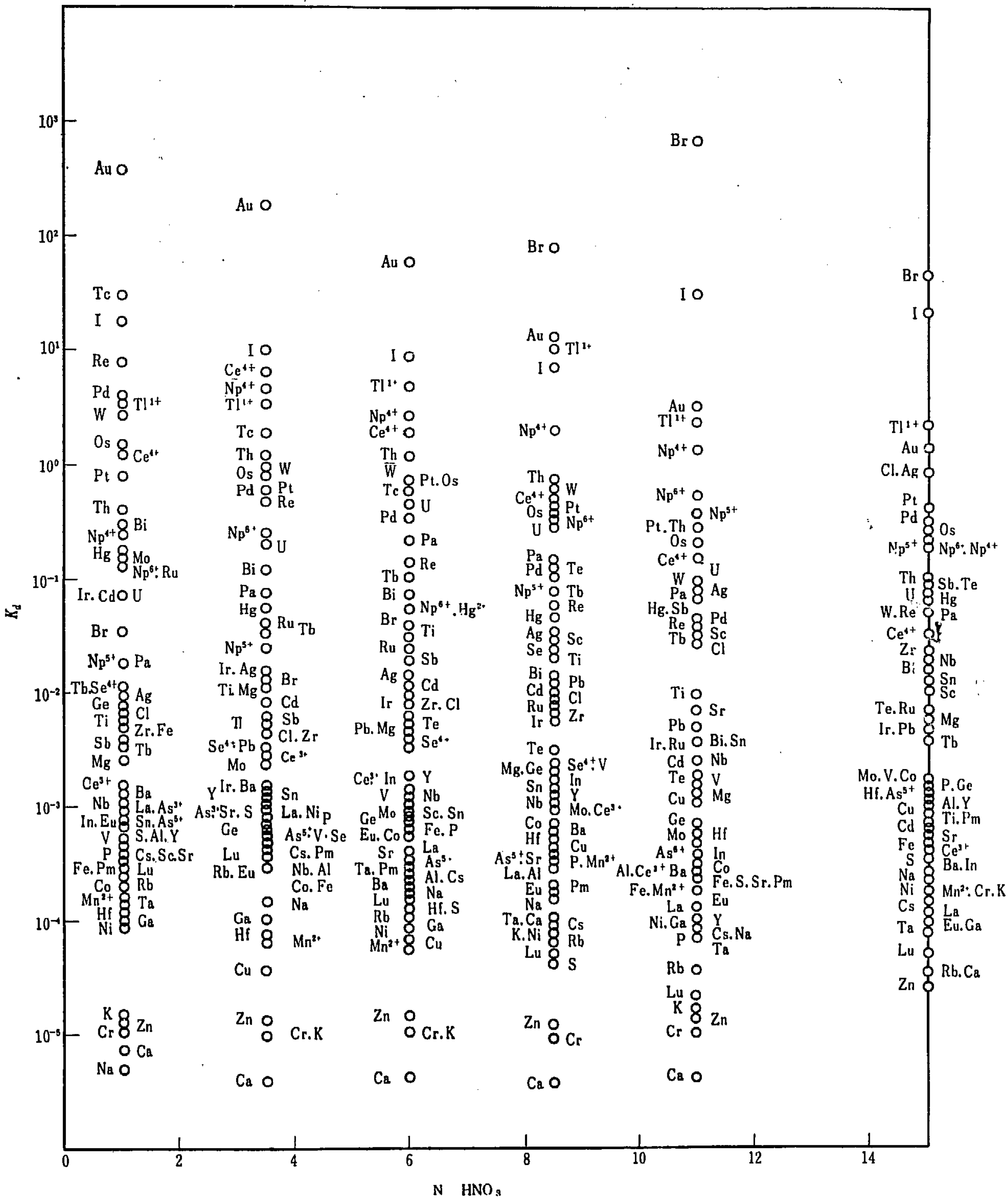

Fig. 12 K_d values in 5 % TIOA-HNO_3 system

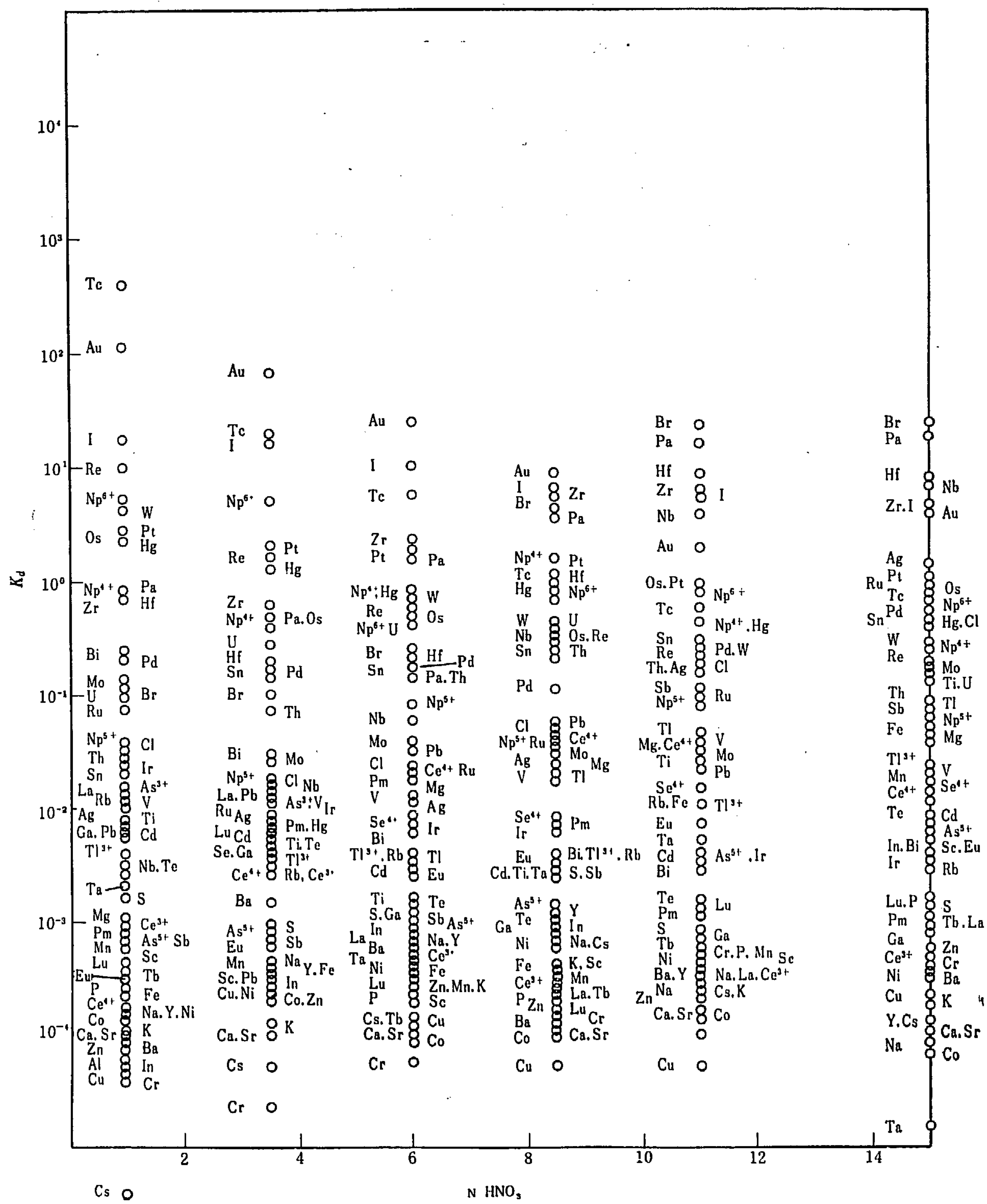

Fig. 13 K_d values in 10% LA-1-HNO_3 system

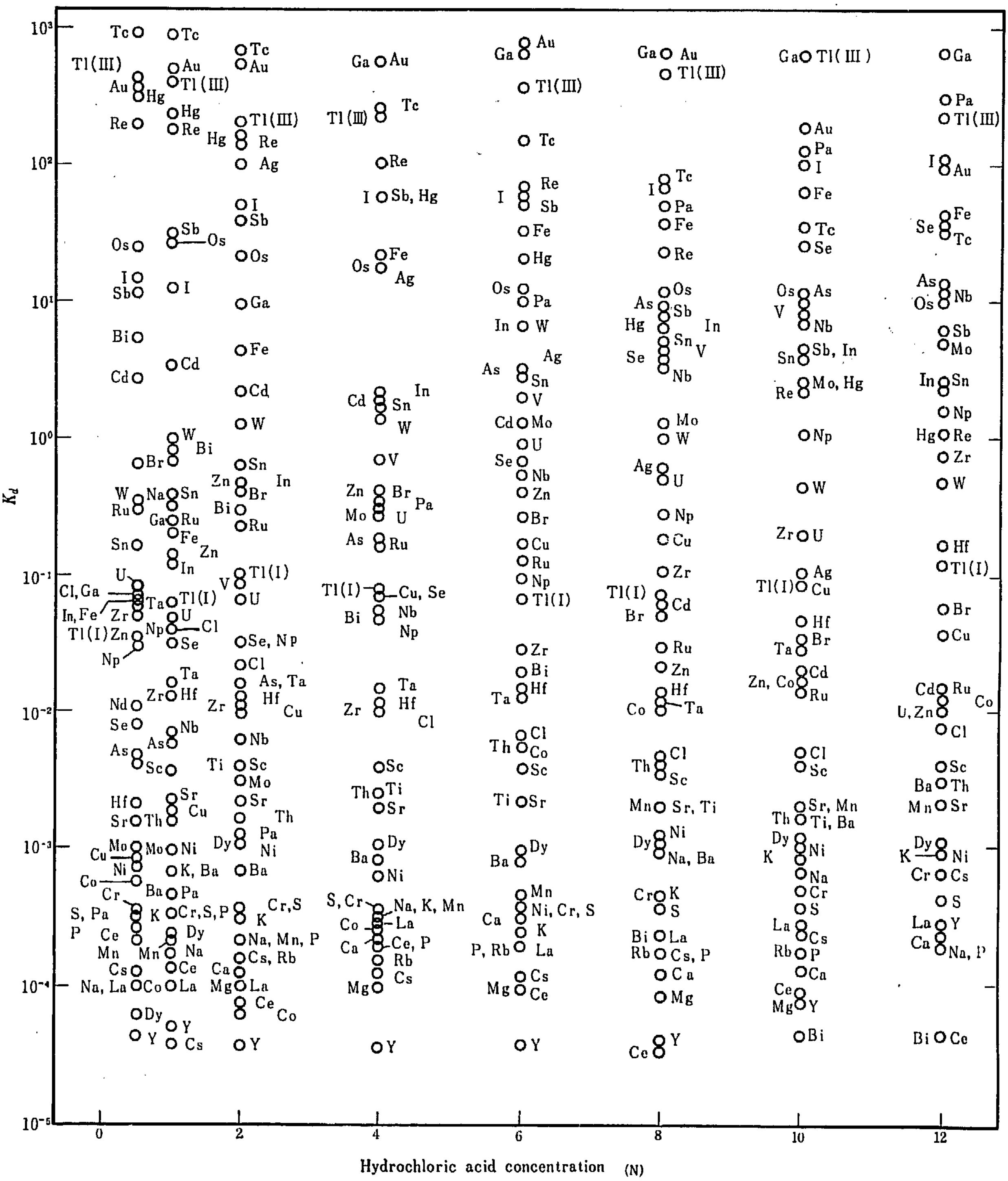

Fig. 14 K_d values in 0.05 M tetraphenyl-arsonium chloride–hydrochloric acid system