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CRITICAL EVALUATION OF STABILITY CONSTANTS OF METAL COMPLEXES OF COMPLEXONES FOR BIOMEDICAL AND ENVIRONMENTAL APPLICATIONS**

(IUPAC Technical Report)

Prepared for publication by
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**Series Title: Critical Evaluation of Stability Constants of Metal Complexes in Solution

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Critical evaluation of stability constants of metal complexes of complexones for biomedical and environmental applications

(IUPAC Technical Report)

Abstract: Available experimental data on stability constants of proton (hydron) and metal complexes for seven complexones of particular biomedical and environmental interest: iminodiacetic acid (2,2'-azanediyldiacetic acid, IDA); (methylimino)diacetic acid (2,2'-(methylazanediyl)diacetic acid, MIDA); 2,2',2'',2'''-{[(carboxymethyl)azanediyl]bis[(ethane-1,2-diyl)nitrilo]}tetraacetic acid (DTPA); 3,6,9,12-tetrakis(carboxymethyl)-3,6,9,12-tetraazatetradecanedioic acid (TTHA); 2,2',2''-(1,4,7-triazanonane-1,4,7-triyl)triacetic acid (NOTA); 2,2',2'',2'''-(1,4,7,10-tetraazacyclododecane-1,4,7,10-tetrayl)tetraacetic acid (DOTA); 2,2',2'',2'''-(1,4,8,11-tetraazacyclotetradecane-1,4,8,11-tetrayl)tetraacetic acid (TETA), published in 1945–2000, have been critically evaluated. Some typical errors in stability constant measurements for particular complexones are summarized. Higher quality data are selected and presented as **"Recommended"** or **"Provisional"**.

Keywords: Complexones; proton complexes; metal complexes; stability constants; biomedical; environmental; Division V.

CONTENTS

1. INTRODUCTION

The name complexone was introduced in 1945 by G. Schwarzenbach [45SKa] for a series of artificial amino acids, containing normally at least one iminodiacetic acid group, $N(CH_2COOH)_2$, or two aminoacetic acid groups, $NHCH_2COOH$. Two of these substances were already known at that time under the names Trilon A and B, and were used to eliminate hardness of water arising from calcium and magnesium ions without separating them from the water. Schwarzenbach demonstrated that, in solution, aminopolycarboxylate anions are able to bind calcium and other cations so strongly that they sometimes cannot be detected by the usual classical precipitation or colorimetric reagents. Further research indicated the capability of complexones to form stable highly soluble complexes with almost all known metal ions [05BC, 03IU, 88DT, 87AN].

The high values for the stability constants of the complexes formed by these ligands are due to the cumulative effect of basic amino groups and the high negative charge of several carboxylate groups, as well as the formation of numerous stable five-membered chelate rings with the metal ions. As the importance of critical assessment of stability constant data for complexones is widely recognized [87SM], two ligands [nitrilotriacetic acid (NTA) and 2,2',2'',2'''-(ethane-1,2-diyldinitrilo)tetraacetic acid, better known as ethylenediaminetetraacetic acid (EDTA, H_4edta)] have been reviewed previously within IUPAC Projects [82ANa, 77AA].

The present study involves evaluation of all reported proton (hydrogen ion, hydron; see first footnote, p. 1450) and metal ion binding constants for the remaining commonly used complexones, and the identification of recommended values for use in chemical speciation calculations. Within these objectives, a priority was given to compounds of strong medical and environmental importance and to those (IDA and MIDA) that represent complex-forming fragments and decomposition products of higher denticity complexones.

Within a broad variety of applications, complexones have in common the regulation of metal concentrations in widely differing systems. Uses of complexones span fields such as detergents, textile and paper processing, photographic developing solutions, scale solubilization in processing tanks, electroplating, control of the activity of metal-dependent polymerization, etc. [92HE, 88DT, 87AN]. The high solubility and stability of complexes formed by complexones make them useful as components of agricultural micro-fertilizers [88DT, 87AN]. Annual industrial output of EDTA and other complexones is in the thousands of tons [92HE].

Ligands considered

IUPAC ligand name	Acronym	Other names[a]
Iminodiacetic acid[b]	IDA, H_2ida	2,2'-Azanediyldiacetic acid[b] Iminodiethanoic acid[c]
(Methylimino)diacetic acid[b]	MIDA, H_2mida	2,2'-(Methylazanediyl)diacetic acid[b] (Methylimino)diethanoic acid[c]
2,2',2'',2'''-{[(Carboxymethyl)azanediyl]bis [(ethane-1,2-diyl)nitrilo]}tetraacetic acid[b]	DTPA, H_5dtpa	Diethylenetriamine-*N,N,N',N'',N''*-pentaacetic acid[e]; Pentenic acid[e]; Diethylenetriaminepentaethanoic acid[d], *N,N*-bis[2-(bis[Carboxymethyl]amino)ethyl]glycine[e]
3,6,9,12-Tetrakis(carboxymethyl)-3,6,9,12-tetraazatetradecanedioic acid[b]	TTHA, H_6ttha	Triethylenetetramine-*N,N,N',N'',N''',N'''*-hexaacetic acid[e] Triethylenetetraminehexaethanoic acid[d]
2,2',2''-(1,4,7-Triazanonane-1,4,7-triyl)triacetic acid[b]	NOTA, H_3nota	1,4,7-Triazacyclononane-1,4,7-triacetic acid[e] 1,4,7-Triaazacyclononane-1,4,7-triethanoic acid[d]
2,2',2'',2'''-(1,4,7,10-Tetraazacyclododecane-1,4,7,10-tetrayl)tetraacetic acid[b]	DOTA, H_4dota	1,4,7,10-Tetraazacyclododecane-1,4,7,10-tetraethanoic acid[d]
2,2',2'',2'''-(1,4,8,11-Tetraazacyclotetradecane-1,4,8,11-tetrayl)tetraacetic acid[b]	TETA, H_4teta	1,4,8,11-Tetraazacyclotetradecane-1,4,8,11-tetraethanoic acid[d]

[a] In this report, the names that have been most frequently encountered in the literature are used.
[b] Names in accordance with IUPAC recommendations.
[c] Names used in IUPAC SC-Database [03IU] and in accordance with IUPAC nomenclature.
[d] Names used in IUPAC SC-Database [03IU] and not in accordance with IUPAC nomenclature.
[e] Name not recommended by IUPAC.

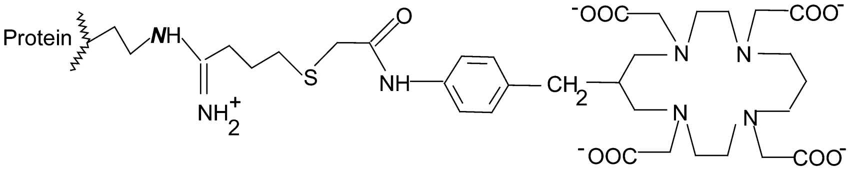
The highly stable complexes formed with polyaminopolycarboxylate ligands are of particular importance in biology and medicine [05BC, 01LE, 00BCa, 00HF, 98AB, 97BH, 84DMb]. The initial field of complexone biomedical application was chelation therapy [99CT]. Since the advent of nuclear fission during the 1940s, mankind has been increasingly exposed to the possibility of radionuclide intoxication from nuclear weapons testing and the expanding use of nuclear fission as a source of power. Only a small proportion of transuranic elements assimilated by humans is excreted, with the remaining part being retained, and the danger that continuing emission of radiation will eventually lead to tumor formation. Accordingly, efficient removal of radionuclides from the body by DTPA calcium complexes was proposed [88DT, 83MB]. The therapeutic antitumor use of radionuclides is based mostly on chelating agents. At the present time DTPA, as well as DOTA, NOTA, and their derivatives, are the agents of choice [01LE, 99AN, 99VH, 96SA, 90LT, 84DMb].

Another medical application where complexones are particularly important is magnetic resonance imaging (MRI). In MRI, paramagnetic metal complexes are used to increase image contrast [00BC, 99AW, 99CE, 98AB, 95AL]. The first clinically utilized contrast enhancement agent (MAGNEVIST) was based on the $[Gd(dtpa)]^{2-}$ complex, which distributes in extracellular space and significantly increases proton relaxation rates [98AB]. $[Gd(dota)]^{-}$ has also entered into clinical practice. Both of these complexes are now reference compounds for development and evaluation of new agents. Other paramagnetic lanthanoid(III) complexes endowed with shift reagent capabilities are used to distinguish NMR resonances of species present in inner and outer cellular compartments and for measurement of pH and temperature. Among these are $[Dy(ttha)]^{3-}$ and $[Tm(ttha)]^{3-}$ [98AB]. NOTA complexes with lanthanoids have been proposed as aqueous shift reagents for biology [87GM, 85GS]. $[Gd(cdta)]^{-}$* is proposed for selective NMR line broadening for complex structure investigation in aqueous solutions [91PV].

***trans*-2,2',2'',2'''-(cyclohexane-1,2-diyldinitrilo)tetraacetic acid, *trans*-1,2-cyclohexanediamine-*N,N,N',N'*-tetraacetic acid, CDTA, H_4cdta.

Complexones have also been incorporated in complexes of ^{99m}Tc and other radionuclides, to obtain scintigraphic imaging of human organs in diagnostic nuclear medicine [99VH, 98DP, 95AL, 88DT, 87AN]. The complex ^{99m}Tc-DTPA has been approved for use as a kidney imaging agent [98DP].

Recently a new generation of compounds—bifunctional complexones—has been intensively studied for MRI and photometric or radioactive imaging and therapy [01LE, 00HF, 99CE, 99VH, 96SA, 95PT, 90SW, 86BG, 86ME, 84MW]. These bifunctional complexones assemble in the same molecule a chelating group (fragment of MIDA, EDTA, DTPA, TTHA, NOTA, DOTA, or TETA) and the chemically reactive functional group, which can be covalently attached to biological macromolecules. The structure illustrates a conjugate of TETA, linked through a C_4S spacer group to a protein lysine *N* nitrogen [86ME]:



Complexones are widely studied and used for mobilization of heavy metals and radionuclides from contaminated soils [05BC, 99BS, 97PK, 95AH, 93HB]. Complexone properties can be advantageous in soil-washing decontamination technologies [97PK], but also disadvantageous by contributing to migration of low-level radionuclides from liquid-waste disposal pits [78MC]. Moreover, strong chelators such as DTPA can influence uptake of radionuclides by plants [81RW, 81WR, 81WW].

The broad and intensive applications of complexones require reliable stability constant data in order to allow equilibrium modeling and prediction of important technological, environmental, and pharmacokinetic equilibria [99BS, 96GM, 95GDa]. A direct relationship between stability constants and the toxicity of gadolinium and some other metals has been observed [00BC, 90CQa]. As complexones are resistant to biodegradation, chemical speciation calculations based on numerical equilibrium data are of extreme importance in environmental science, waste management, agriculture, magnetic resonance imaging, behavior of radiopharmaceuticals in blood plasma, and many other applications [05BC, 99BS, 95AH, 90CQa, 87SM, 84DMb].

The data of interest are partly accumulated in a number of monographs [88DT, 71AN, 70PE], reviews [00BC, 87AN, 87SM], and compilations of stability constants [95IP, 91IP, 89MS, 87SM, 85IB]. Recently, three new computer databases have become commercially available [03IU, 97CS, 91MM]*. The IUPAC Stability Constants Database [03IU] is the more comprehensive of these three and is taken here as the major source of data.

This review is based on data published in SC-Database [03IU]. The period 1965–1998 is covered exhaustively, but selected earlier and later publications are also included. References citing publications which include data not entered in [03IU] have the format “99AN”, etc. References taken from SC-Database use the database short reference format, e.g., “88THc”. In some cases, the SC-Database [03IU] gives one collective reference for several publications. For example, [72PRc] embraces data from six independent publications by the same authors. In this review, these publications are presented in a format “72PA, 72PB…72PZ” followed by the SC-Database short reference in the reference list. The names of journals are presented in original transcription followed by English version references (if any).

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*The latest versions available are: *Stability Constants Database and Mini-SCDatabase*. IUPAC and Academic Software, Version 5.3, 2003, Sourby Old Farm, Timble, Otley, Yorks, UK; <scdbase@acadsoft.co.uk>; NIST Standard Reference Database 46. *Critically Selected Stability Constants of Metal Complexes Database*, compiled by R. M. Smith, A. E. Martell, R. J. Motekaitis, Version 7.0 for Windows, 2003, U.S. National Institute of Standards and Technology Standard Reference Data Program, Gaithersburg, MD 20899; JESS: ‘jess.murdoch.edu.au’.

2. PRESENTATION OF EQUILIBRIUM DATA AND ABBREVIATIONS USED

The stepwise protonation constants* of ligands are presented as K_n for equilibrium

$$H_{n-1}L + H \rightleftharpoons H_nL \text{ with } K_n = [H_nL]c^o[H_{n-1}L]^{-1}[H]^{-1} \ (c^o = 1 \text{ mol L}^{-1})$$

In tables, the equilibrium $H_{n-1}L + H \rightleftharpoons H_nL$ is indicated in a brief form: $H_{n-1}L + H$ and the corresponding constant as $K(H_{n-1}L + H)$. In all cases, L indicates species with all –COOH and ammonium groups deprotonated. M represents the cation and I symbolizes the ionic strength**. Equilibria for metal-complexes are self-explanatory: $M + L \rightleftharpoons ML$ is presented as M + L and $K = [ML]c^o[M]^{-1}[L]^{-1}$ as K_{ML} or $K(M + L)$; $M + H + L \rightleftharpoons MHL$ as M + H + L and $K = [MHL](c^o)^2[M]^{-1}[H]^{-1}[L]^{-1}$ as $K(M + H + L)$ or as β_{MHL}; $M + HL \rightleftharpoons MHL$ as $K(M + HL)$, etc.

In potentiometric titrations with a glass electrode, the calibration technique governs the type of constant calculated. Concentration quotients (stability constants) are obtained when the electrode system is calibrated with solutions of known hydrogen ion concentration (e.g., a monoprotic strong acid) or by the conversion of pH values using the hydrogen ion activity coefficient. In the text, these are designated by "Conc.". Mixed constants [91SMa, 84PE] are obtained when standard buffer solutions of known hydrogen ion activity are used for electrode calibration (e.g., potassium hydrogen phthalate buffer with pH 4.008 at 25 °C). Such constants include both activity (hydrogen ions) and concentration (all other participants of the complexation equilibrium) terms. Following the reasons described elsewhere [91KSa, 91SMa, 84PE], priority is given to concentration constants.

Methods used in the papers selected for evaluation are denoted by the following symbols:

gl	glass electrode (pH-metry)
EMF	metal electrode (e.m.f. measurement)
red	redox electrode (e.m.f. measurement)
ix	ion-exchange
sp	spectrophotometry
NMR	nuclear magnetic resonance
cal	calorimetry
dis	distribution
pol	polarography
kin	kinetic measurements
sol	solubility

3. GENERAL PHYSICOCHEMICAL PROPERTIES OF COMPLEXONES

Complexones (H_nL) are generally poorly soluble in water (pH 2–3), while their alkali metal salts have a high solubility at pH > 4. At pH < 2, all complexones are capable of forming positively charged highly soluble species, such as H_3ida^+, H_6dtpa^+, H_7dtpa^{2+}, H_7ttha^+, etc. The solubility of all compexones therefore attains a minimum between pH 1 and 4 [88DT, 83KDb, 67ANb]. This causes considerable accuracy problems in the measurement of protonation constants (lg K_n) of the neutral species H_nL and the ion $H_{n+1}L^+$.

The use of an incorrect or incomplete set of lg K_n values may result in appreciable errors in calculated stability constants for highly stable complexes, for which measurements at low pH are required. The errors increase with the number of protonation constants that contribute significantly to the magnitude of [L] [77AN]. A very common error involves the neglect of positively charged ligand species

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*By common usage in solution chemistry, the term "protonation" refers to the natural isotopic mixture of hydrogen, not isotopically pure ^1H. Strictly speaking, the reaction is "hydronation"; electric charges are omitted.

**All the data (values) refer to amount concentrations (I_c; mol dm^{-3}) unless otherwise stated. For reasons of brevity, mol L^{-1} instead of mol dm^{-3} is used.

that exist between pH 0 and 2; this is pertinent to spectrophotometric, electromigration, and other methodologies for the measurement of stability constants [73CC, 71BRa, 71EPb, 69DBa, 65BAc]. Some research groups recognized this problem, but did not have the required lg K_n values to overcome it. For example, the formation of $[Bi(H_4ttha)]^+$, $[Bi(H_3ttha)]$, $[BiH(ttha)]^{2-}$, and $[Bi(ttha)]^{3-}$ was established by spectrophotometry at pH < 2, but the authors failed to obtain the corresponding stability constants because “only six” of the required nine TTHA protonation constants were quantified [79NPa].

Discrepancies in lg K(L + H) values for all complexones, and lg K(HL + H) for DTPA, TTHA, DOTA, and NOTA are due to the binding of complexone anions to alkali metal ions of the background electrolytes used. The structure of $[Co(NH_3)_6]_2[Na_2(edta)_2H_2O]\cdot 6H_2O$ [88DT, 84PP] reveals that sodium coordinates to 2 nitrogen atoms and 4 oxygen atoms of EDTA. Thus, at appropriate pH alkali cations can efficiently compete with protons for nitrogen donor atoms of complexones. The most “neutral” (indifferent) cations for the system would seem to be tetramethylammonium or ammonium ions. Even though their ability to bind polyaminopolyacetates is not yet known, complexation of these cations is expected to be negligible in comparison to that of the alkali metal ions.

The interaction of alkali metal ions with macrocyclic complexones is rather strong. The protonation constant values for NOTA and DOTA were chosen, therefore, from those determined in $(CH_3)_4NCl$ or $(CH_3)_4NNO_3$ as supporting electrolytes. Since DOTA forms stable complexes with Na^+, and also with K^+, media containing these cations lead to anomalously low values for K(L + H) and K(HL + H).

Another problem with DOTA measurements is the high value for the first protonation constant, K(L + H). In such cases, lg K is difficult to determine by the usual potentiometric methods. NMR titration is the preferred technique, but current applications of this technique are unacceptable. Usually, lg K(L + H) values determined by NMR data are not accurate, owing to poor control of the ionic strength, to difficulties in preventing contact of the solution with the atmosphere, and the need to compare data obtained in D_2O and H_2O media [91DSa]*.

Complexones appear to interact with all known metal ions except possibly Fr^+ [59ML]. For some metals, the highest reported stability constants have been observed with complexones. Strongly hydrolyzed cations, such as Al^{III}, In^{III}, Tl^{III}, Bi^{III}, Zr^{IV}, Th^{IV}, Pd^{II}, etc., are especially strongly complexed by complexones [03IU]. K_{ML} values for many of these metals range between 10^{30} and 10^{40}. In these cases, direct potentiometric or spectrophotometric titrations cannot readily be used. Reaction with a competing ligand: 2,2',2''-triaminotriethylamine, (tren), 2,2'-{ethane-1,2-diylbis[(2-hydroxybenzyl)azanediyl]}diacetic acid, (*N,N'*-di-(2-hydroxybenzyl)-diaminoethane-*N,N'*-diacetic acid, HBED) [99DLa, 76AMa, 76HMd, 59CFc] or competing cation (Hg^{2+} [59CFc], Ga^{3+} [88THa], Cu^{2+} [72BCb]) is required. This in turn introduces additional systematic errors and problems. One such problem can arise owing to fairly slow kinetics of ligand–ligand displacement reactions, e.g., for Fe^{III} complexonates, equilibrium in most cases is established in a few days [90ADb].

K_{ML} values have generally been determined in KCl or KNO_3 media. The calculations have used ligand protonation constants obtained in solutions of potassium salts with no correction for K^+ complexation. Such an approach can be accepted for IDA and MIDA, but not for DTPA, TTHA, and DOTA. Unfortunately, the stability constants of $[K(dtpa)]^{4-}$, $[Na(dtpa)]^{4-}$, and $[K(ttha)]^{5-}$ are not yet published. It has been demonstrated that if published data need to be corrected for the formation of $[K(edta)]^{3-}$ in 0.1 mol L^{-1} KNO_3, then the stability constant requires corrections by a factor of $1 + [K^+]K_{Kedta}$ [77AN]. For lg K_{Medta} the correction is +0.21. Taking into account the higher negative charge and denticity of DTPA and TTHA relative to EDTA, the corresponding lg K_{KL} corrections are expected to be even higher.

The determination of stability constants for metal complexes of DOTA is highly dependent on the values used for K(L + H) and K(HL + H). One of the reasons for the spread of values found in the literature for stability constants of complexes with this ligand is the variety of K(L + H) values used by

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*IUPAC recommendations for NMR lg K(L + H) measurements are in preparation by K. Popov, H. Rönkkömäki, and L. H. J. Lajunen.

different authors. Those working with supporting Na^+ electrolytes always report lower values of K_{ML}. The same situation occurs with those working with K^+ electrolytes, but the lower stability constant for the K^+-DOTA complex can increase the comparative significance of other experimental errors.

The high potential denticity of complexones creates high (sometimes anomalously high) cation coordination numbers. For example, in $[Mg(H_2O)_6][Mg(H_2O)edta]{\cdot}2H_2O$ the magnesium ion chelated by EDTA has coordination number (CN) 7, while the other has CN 6 [84PP]. With the hexadentate ligand EDTA, In^{III} has CN 7 and forms five chelate rings in $Na[In(edta)(H_2O)]{\cdot}2H_2O$ [95IM], while with the octadentate DTPA it reveals CN 8 in $Na_2[In(dtpa)]{\cdot}7H_2O$ and forms 7 chelate rings [89MR]. Thus, additional chelate rings are commonly formed relative to other complexes of the same cation with lower denticity ligands. Owing to high denticity, the most common species for DTPA, DOTA, TETA, and NOTA are ML and MH_nL, while complexes such as M(OH)L are less common. ML complexes normally have mononuclear structures both in the solid state and in aqueous solution [88DT]. Thus, there are no special problems with association or polymerization of complexes.

TTHA is known to form M_2L/ML mixtures at 1:1 metal/ligand total concentration ratios. Ignorance of this fact has led to erroneous constants for Al^{3+} and Ga^{3+} in [80KHb, 80MMd]. The same situation occurs for Tl^+, Tl^{3+}, and many other cations. In contrast, MIDA and IDA are likely to form ML and ML_2 species at 1:1 metal/ligand total concentration ratios, while M_2L is less common. ML complexes formed by MIDA and IDA have a higher tendency for hydrolysis, polymerization, and coagulation [88DT].

When all numerical values listed in [03IU] for complexones are compared, it appears that the relative internal consistency of data reported by one research group is much better than that observed between different groups. This causes serious problems for critical evaluation of lanthanoid lg K(M + L) data. For this particular case, the difference in lg K(M + L) between neighboring cations reported by one research group is much smaller than the difference between results obtained by different groups for the same element. Any attempt to present averaged values for a pair of neighboring lanthanoids leads to significant distortion of the trend in stability constants with increasing atomic number. In such cases, publications providing reliable stability constant trends for lanthanoids are identified in table footnotes, while the tables themselves present data for only one lanthanoid.

Protonation of ML (DTPA, TTHA, DOTA, TETA, NOTA) complexes does not necessarily lead to their decomposition [88DT]. For example, in $LaHedta{\cdot}7H_2O$ and $CoH_2edta{\cdot}3H_2O$ the carboxylate groups of EDTA are simultaneously protonated and coordinated [88DT]. In complexes formed by EDTA, DTPA, and TTHA the proton is normally localized on a carboxylate group, which either remains coordinated or leaves the metal ion coordination sphere. In the case of MIDA and IDA, protonation frequently occurs on the nitrogen atom. In this case, protonation leads to a complete decomposition of complex [88DT].

Kinetics of formation of ML is generally comparable with the rate of exchange of water molecules of aqueous-cations [78MA, 74MPa]. An exception arises with macrocyclic ligands (DOTA, TETA, and NOTA), and special precautions have to be taken to ensure an equilibrium state in stability constant measurements. For certain cations, such as Ni^{2+} and the lanthanoids, complex formation rates are very slow. In these cases, it is not possible to obtain reliable values by continuous titration methods, and the discontinuous or potentially less accurate batch process is required.

Most complexones are commercially available at high purity, with the exception of DOTA, TETA, and NOTA [82WB, 80DE, 77TT]. These macrocyclic compounds are prepared by several synthetic steps, and the purification of the final compound is difficult. Thus, the purity of the compounds used by some authors is not sufficient for accurate equilibrium constant determinations. For this reason, all the data obtained by [91CMa] and [91CMb] for DOTA were discarded. The separation of inorganic salts resulting from the synthesis of NOTA or DOTA can be made by ionic exchange chromatography, however, several authors do not perform this step. TETA is much less soluble in aqueous solutions and can be easily obtained without contamination by inorganic salts or other organic impurities.

At pH < 2, complexones are readily oxidized by cations such as Ce^{IV} or Mn^{III} even at room temperature [72YPa, 71BPh, 71MA, 71PMc, 71MAn, 70MMb]. Although this process is considerably slower at higher pH [88DT], all equilibrium data for these cations have been treated as doubtful.

Some special comments are needed with respect to TTHA. Because of the very large electrical charge of the ligand anion, maintenance of a constant ionic strength I (e.g., $I = 0.1$ mol L^{-1}) is very difficult. A total TTHA concentration larger than 1 millimol L^{-1} should be avoided in strongly basic solutions. For instance, a 1 millimol L^{-1} solution of K_6ttha gives $I = 0.021$ mol L^{-1}, i.e., more than 20 % of the ionic strength. This can be compensated in a titration only partially through the volume increase associated with strong base addition.

The calculation of equilibrium constants from pH measurements in solutions containing a metal ion and a complexone normally requires solutions free of complicated metal hydrolysis products. Some caution is therefore needed in the case of metal ions such as Bi^{3+}, which forms $Bi_6(OH)_{12}{}^{6+}$ below pH 1.5. Apparently, the authors of [69YMa] used data for the determination of the different constants of Bi^{3+} with TTHA without considering this fact.

4. DATA EVALUATION CRITERIA

Each recommended value is normally based on a comparison of at least two independent high-quality publications. Data published on complexones have been evaluated by applying the following general criteria [01PRa, 97LP, 96YOa, 95SM, 91KSa, 91SMa, 75NB]:

(a) Clear definition of constants reported (i.e., unambiguous specification of complex stoichiometry MHL, M(OH)L, etc., and of corresponding stability constants)*.

(b) The extent to which essential reaction conditions (the purity of the ligand, temperature, ionic strength, nature of the supporting electrolyte, account of metal–ligand reaction kinetics, ligand:metal ratio, etc.) have been specified**.

(c) The soundness of calibration of the apparatus used (e.g., calibration of the electrode system for potentiometric measurements) and specification whether concentration or mixed constants were calculated†.

(d) The maintenance of constant temperature and ionic strength during titrations††.

(e) Reliable treatment of the experimental data (e.g., careful consideration of all possible species formed: parent and mixed hydroxo-complexes of readily hydrolyzable metal ions, formation of dimers and polymers, cationic forms of a ligand, stability constants of competing ligand, etc.)‡.

(f) Correct selection of auxiliary data from the literature (e.g., selection of the concentration constants for ligand protonation required for the evaluation of spectrophotometric, magnetic relaxation or polarographic and pH measurements carried out on metal–ligand systems)‡‡.

(g) Details of the calculation method used#.

On the basis of these criteria, experimental data have been examined and initially grouped into two categories: "accepted" and "rejected". Among those data that passed the preliminary acceptance

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*Most papers meet this requirement; the few exceptions [85MBb, 74TPa, 66LPa] were rejected.

**This requirement was not met by [00BMa, 99SBd, 97YSa, 82VNa, 81DSa, 80KJa, 80OOb, 79MMf, 72KNb, 70KMe, 70MSd, 69HGa, 66KRa] and only for some of the studied ligands in [91CMa] and [91CMb], namely for DOTA.

†Such important information is missing in [85GAb, 80BTa, 75LBa, 72YPa, 71OBb, 56FRa] etc.

††This was not specified in [66STb].

‡References [90RNc, 88THa, 84HKa, 81DSa, 80MMd, 80KHb, 80KJa, 73CTa, 71GGa, 71GKb, 70HAa, 70KMe, 69YMa, 66EMd] have poor information in this field or errors (for which correction was sometimes possible).

‡‡Because of incorrect use of auxiliary data from the literature the following papers have been rejected or the obtained data have been corrected: [99DLa, 90CBc, 84HKa, 82OLa, 81DSa, 78RSa, 75NWa, 73CCc, 71BRa, 71EPb, 71LUa, 70HAa, 69KTc, 69YMa, 65BMf, 65KKa].

#Most papers published in 1945-1990 do not report on calculation methods used.

criteria, those that exhibited the best agreement were selected for further treatment. These were averaged, rounded and, depending on the standard deviations (s.d.), the mean values were regarded as **Recommended** (**R**): s.d. ≤ 0.05 for H-complexes (e.g., H + L) and ≤ 0.1 for metal complexes (M + L, H + ML, or M + HL) or **Provisional** (**P**): 0.05 < s.d. ≤ 0.2 for H-complexes and 0.1 < s.d. ≤ 0.2 for metal-complexes. The s.d. indicates therefore an agreement among the selected data and is included in Tables in parentheses. In those cases where the experimental uncertainty for each single value was much less than that derived from the mean value, then the latter is given. When the agreement of published data was better than experimental uncertainty, then the largest rounded s.d. from the original publication is listed. Although complexones have been studied intensively since 1945 and the total number of publications devoted to ligands of interest is about 480, data of sufficiently high quality have been found only in 99 papers. Therefore, for most metal–ligand combinations the group of "accepted" values did not allow comparison of stability constants measured by independent research group(s) under the same conditions. The amount of reliable data represented by a single group constituted 84 % (IDA), 95 % MIDA, 85 % (DTPA), 66 % (TTHA), 100 % (NOTA), 42 % (TETA), and 18 % (DOTA). Therefore, comparison between two or more independent research groups was impossible for the majority of values "accepted" in the preliminary selection.

In this situation, we nominated the data presented by a single research group as **Recommended 1 (R1)** if (a) we had no doubt as to the adequacy of applied experimental or calculation procedures and the research group has (b) either **R**-level agreement with Recommended values for other cations with similar properties (e.g., within lanthanoid or alkaline earth series), or revealed equally good agreement with independent researchers that measured the same constant for the same cation, but under slightly different experimental conditions (temperature, ionic strength). The former case can be illustrated by K^+ and Ba^{2+} complexes of DOTA (Table 6), while the latter by K(HL + H) values for IDA, Table 1 (**R1** for 20 °C [76AMa] and R for 25 °C [86ANb, 71GKb]; both for 1 mol L^{-1} $NaClO_4$). In a few exceptional cases, the most reliable (and widely accepted) values are also nominated **R1**, for example, for Pd^{2+} complexes of DTPA, Table 3. The s.d. then reflects either the original value reported by the author or the one rounded by the reviewer, taking into account the level of agreement of this author's other data with independent research.

In a similar way, category **P** was given to some results from single papers if the corresponding research group reveals **P**-level agreement with other researchers for at least one different cation. Provisional category was also given to those data having good agreement among several groups, but where the reviewers observed some deviations from the necessary rigor. In a few cases, values from a single publication that fit the general trend within **P**-level results has also been treated as Provisional, e.g., Ra^{II} complexes with DTPA, Table 3.

It should be stressed that the formulation of uniform criteria for ligands of different denticity is not possible. For IDA and MIDA, the precision of stability constants should be as high as that found for Ni^{II}-glycine [87BOa]; in contrast for DTPA, DOTA, TETA, and TTHA, it is significantly lower. For example, an evaluation of uncertainty in the constants for $[(UO_2)_2dtpa]^-$ measured by direct potentiometric titration, gave a value of 0.36 lg units [82OLa]. In the case of competition reactions between complexones and another ligand (e.g., tren) for the metal ion, the uncertainty should be even larger owing to additional contribution of uncertainties associated with tren protonation and complexation.

In a few specific cases when evaluation identified some mistake in the determination of the constants, but these are nevertheless of semi-quantitative value, the criteria 0.2 < s.d. ≤ 0.3 (lg K_{HL}) and 0.2 < s.d. ≤ 1.0 (lg K_{ML}) were used to indicate values that the present authors assess as being reliable. Such data are not included in tables, but are given in footnotes. The same treatment has been used for some papers that do not have evident errors but reveal gaps in the description of some important experimental details.

Some papers with data that are rejected contain important supplementary information (normally spectroscopic) that could be helpful in future research. Thus, all the references including rejected (or partly rejected) data are listed at the beginning of any section devoted to the particular ligand, and the cations studied are indicated.

The rejected data are, however, not listed in the Tables. References that are cited, but not included in the Tables could also be:

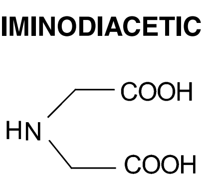
- communications with possibly correct data, but inadequate or poor description of experimental conditions, e.g., [80KJa, 80BTa];
- communications that reviewers could not access in the original version (in this case, an original reference is normally followed by *Chem. Abstr.*, IUPAC SC-Database, or some other secondary citation, indicating the place from which data are taken and the fact that they are not critically evaluated in the present Report);
- publications of the same research group with stability constant data that completely duplicate the cited one;
- publications that need further independent evaluation (this situation includes the cases where two independent research groups offer data that formally meet the requirements stated above, but owing to some hidden systematic errors reveal very large numerical discrepancies);
- publications that provide data for conditions that contrast with those for other data (e.g., high or low temperatures [86LDb, 81DMa, 67TMf], "unusual" ionic strengths [79ZLa, 76GAa], mixed solvents [95LBb], ternary or mixed ligand complexes [98LVa, 97BH, 93BNb, 92RKb, 91NBa, 90SSc, 90UBc, 87FZa, 79BCa, 79KNa, 78KNc, 76PAa, 76PTb], effective or conditional stability constants [75BUb, 75HTa], etc.).
- publications that present only enthalpy values.

The ligands are considered in the order of increasing complexity. The stability constants of metal complexes are surveyed in the following groups of the periodic table: hydrogen ion, groups 1, 2, 13, 14, 15, 3*d*-group; 4*d*-5*d*-groups, group 3, and a group of 4*f*-5*f* metal ions.

Complete information on the experimental conditions used in papers selected for evaluation is given before each Table. The averaged stability constants (with standard deviation in parentheses) and their evaluation category are tabulated, together with the most important experimental information (medium, temperature) and the references that contributed to the mean value listed in the table. When the average value is derived from data obtained in different media, then symbols like Na/KCl or KCl/NO_3 are used.

The reviewers did not recalculate stability constants to a uniform ionic strength. With the exception of a few values that were recalculated for TTHA complexes using more reliable ligand protonation constants, the data listed represent an average of those reported in the accepted publications. Critically evaluated data are presented in Tables 1–7.

5. 2,2'-AZANEDIYLDIACETIC ACID (IMINODIACETIC ACID), IDA, H_2ida



Cations studied[a–f]: **H^+**: 00BMa, 99SBb, 99SEb, 95MAa, 94TSa, 92ANa, 92CGa, 92GLa, 92RAc, 89MIa, 88THa, 87FZa, 87MDa, 86ANb, 85HAc, 83DBb, 83FSa, 83SVa, 82NBa, 82VNa, 81DSa, 81MOa, 80OOb, 79ZLa, 78JSb, 78MGa, 78RSa, 77PTb, 76AMa, 76GMb, 75MRb, 73CBc, 73CTa, 73SKb, 73STc, 72NAa, 71BB, 71GGa, 71GKb, 71LNb, 71TSh, 70KMe, 70NPc, 69PMd, 68KSa, 67MY, 67SKg, 67TMg, 66KRa, 66MAb, 64ANa, 64RMc, 62THa, 52CMa, 45SKa

Be$^{2+\,b}$: 87MDa, 81DSa
Mg$^{2+\,e}$: 95LBb, 69ASb, 64ANa, 45SKa
Ca$^{2+\,e}$: 75MRb, 69ASb, 68KSa, 64ANa, 57TBb, 45SKa
Sr^{2+}: 69ASb, 64ANa
Ba^{2+}: 64ANa, 45SKa
Al$^{3+\,b}$: 97YSa, 81DSa, 71LNb
Ga^{3+}: 97YSa , 85SAa, 76HMd
In^{3+}: 97YSa, 85MMa, 85SAa, 84PGa, 66MAb
Tl^{+}: 70FUb
Sn^{IV}: 92CGa
Pb^{2+}: 83FSa, 81MOa, 80NWa, 76KIa, 76NCa, 72NAa, 71KTd, 64ANa
Bi^{3+}: 76KIa
V^{V}: 79ZLa
VO^{2+}: 99SBb, 84FVa, 83FSa, 78JSb, 73STc, 66KFc
Cr^{2+}: 83MDb
Cr^{3+}: 82VNa, 81DSa, 70MSd
Fe^{2+}: 00BMa, 72NAc, 64ANa
Fe^{3+}: 00BMa, 99SEb, 97YSa, 86ANb, 72NAb
Co^{2+}: 87FZa, 84VRa, 83DBb, 80OOb, 64ANa, 52CMa
Co^{3+}: 76BCb, 69BHb
Ni^{2+}: 92GLa, 90UBc, 84VKb, 83FSa, 81ACa, 71TSh, 71TSj, 70CMa, 70NPc, 69FDa, 64ANa, 57TBb, 52CMa
Cu^{2+}: 95LBb, 92GLa, 92NAa, 92RAc, 92RKb, 90UBc, 87NDa, 85KVa, 85SNa, 85SRc, 84HKa, 83FSa, 83SVa, 82VRa, 81FMb, 80NWa, 79BCa, 78WIa, 76KIa, 75NWa, 73YBa, 71TSh, 71TSj, 70STf, 69LAa, 69STb, 67TMg, 64ANa, 57SYb, 57TBb, 52CMa
Zn^{2+}: 92GLa, 92NAa, 92RAc, 85SNa, 83VRa, 81FMb, 78KCa, 73HAb, 71TSh, 71TSj, 70FDa, 70STf, 64ANa, 57SYb, 52CMa
Cd^{2+}: 95LBb, 92GLa, 84MRa, 83SVa, 83YWa, 81GKa, 78KCa, 71TSh, 71TSj, 70STf, 64ANa, 57SYb, 52CMa
Hg^{2+}: 75LBa, 67SKg
Hg$_2^{2+}$: 67SKg
Ag^{+}: 92ANa, 89MIa, 81SCa
Zr^{4+}: 64PVc
Hf^{4+}: 78RSa
Mo^{VI}: 79ZLa, 66KRa
W^{VI}: 79ZLa, 66KRa
Pd^{2+}: 76AMa, 75CGc, 75VCa
Ru^{3+}: 88THa
Sc^{3+}: 97YSa, 85SAa, 80SKc, 74KPd, 72GGa
Y^{3+}: 97YSa, 71GKb, 62THa
La^{3+}: 97YSa, 88VSc, 84KTb, 80KTb, 76TBb, 74KPd, 71GKb, 64ANa, 62THa
Ce^{3+}: 88VSc, 79TKb, 76TBb, 71GKb, 62THa
Pr^{3+}: 88VSc, 84KTb, 80KTb, 72GGa, 71GKb, 62THa
Nd^{3+}: 88VSc, 84KTb, 80KTb, 74PLa, 74TDa, 73TEb, 71GKb, 70KMe, 69PMd, 68KRc, 67TKa, 66KTa, 62THa
Sm^{3+}: 88VSc, 71GKb, 62THa
Eu^{2+}: 73CTa
Eu^{3+}: 88VSc, 76TBb, 73CTa, 72GGa, 71GGa, 71GKb, 71TKf, 66MAb, 62THa
Gd^{3+}: 88VCc, 84KTb, 80KTb, 71GKb, 62THa
Tb^{3+}: 88VSc, 76TBb, 71GKb, 62THa
Dy^{3+}: 88VSc, 84KTb, 80KTb, 71GKa, 62THa
Ho^{3+}: 74PLa, 72GGa, 71GKa, 62THa
Er^{3+}: 74PLa, 71GKb, 71TKf, 71TSh, 62THa
Tm^{3+}: 71GKa, 62THa
Yb^{3+}: 71GKb, 69PMd, 62THa
Lu^{3+}: 72GGa, 71GKb, 62THa
UO$_2^{2+}$: 84BLb, 82NBa, 80BTa, 73CBc, 67LCa, 64RMc
NpO$_2^{+\,f}$: 94TSa, 90RNc, 83ITa, 73CBc, 70EWa, 70KC
Am^{3+}: 89RSa, 71BB, 69DBa
PuO$_2^{+}$: 73CBc, 70EWa
Th^{4+}: 85SAa, 83BCa, 82NBa, 77PTb, 74KPd, 73SKb

Experimental conditions of papers selected for critical evaluation:

I = 0.1 mol L^{-1} KNO$_3$, 20 °C, Conc., gl: 64ANa
I = 0.1 mol L^{-1} NaClO$_4$, 25 °C, Conc., gl: 81DSa
I = 0.1 mol L^{-1} KCl, 25 °C, Conc., gl: 88THa
I = 0.1 mol L^{-1} KNO$_3$, 25 °C, Conc., gl: 92CGa, 84FVa, 83FSa, 82NBa, 78JSb, 62THa; EMF: 83SVa
I = 0.1 mol L^{-1} Me$_4$NBr, 25 °C, Conc., sp: 85HAc
I = 0.1 mol L^{-1} KCl, 30 °C, Conc., gl: 52CMa
I = 0.1 mol L^{-1} NaClO$_4$, 35 °C, Conc., gl: 81DSa
I = 0.2 mol L^{-1} KCl, 25 °C, Conc., gl: 99SBb

I = 0.5 mol L^{-1} $NaClO_4$, 25 °C, Conc., gl: 92GLa, 87MDa, 73CTa, 72NAa, 72NAc, 71LNb
I = 0.5 mol L^{-1} KNO_3, 25 °C, Conc., gl.: 99SEb
I = 1.0 mol L^{-1} $NaClO_4$, 20 °C, Conc., gl: 76AMa, 73CBc
I = 1.0 mol L^{-1} $NaNO_3$, 25 °C, Conc., gl: 95MAa
I = 1.0 mol L^{-1} $NaClO_4$, 25 °C, Conc., gl: 86ANb, 71GGa, 71GKb
I = 1.0 mol L^{-1} KNO_3, 25 °C, Conc., gl: 92ANa, 81MOa, 64RMc
I = 1.0 mol L^{-1} KCl, 25 °C, Conc., gl: 78MGa, 76GMb

Table 1 Recommended and provisional data for IDA.

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
H^+	H + L	0.1; Me_4NBr	25	9.45 (0.01)	P	85HAc
		0.1; KNO_3	20	9.45 (0.02)	P	64ANa
		0.1; KNO_3/Cl	25	9.32 (0.02)	R	92CGa, 88THa, 83FSa, 83SVa, 82NBa, 78JSb, 62THa
		0.2; KCl	25	9.29 (0.03)	P	99SBb
		0.5; KNO_3	25	9.25 (0.06)	P	99SEb
		1.0; KNO_3/Cl	25	9.27 (0.03)	R	92ANa, 81MOa, 78MGa
		0.1; $NaClO_4$	35	9.25 (0.06)	P	81DSa
		0.5; $NaClO_4$	25	9.22 (0.05)	R	87MDa, 72NAa, 71LNa
		1.0; $NaClO_4$	20	9.33 (0.08)	P	76AMa, 73CBc
		1.0; $NaClO_4$	25	9.29 (0.05)	R1	86ANb
	HL + H	0.1; KNO_3/Cl	25	2.60 (0.03)	R	92CGa, 88THa, 83FSa, 62THa
		0.2; KCl	25	2.54 (0.04)	P	99SBb
		0.5; KNO_3	25	2.53 (0.06)	P	99SEb
		1.0; KNO_3/Cl	25	2.60 (0.03)	R	81MOa, 78MGa
		0.1; $NaClO_4$	25	2.70 (0.07)	P	81DSa
		0.1; $NaClO_4$	35	2.66 (0.03)	P	81DSa
		0.5; $NaClO_4$	25	2.58 (0.02)	R	92CGa, 87MDa, 73CTa, 72NAa, 71LNa
		1.0; $NaClO_4$	20	2.64 (0.03)	R1	76AMa
		1.0; $NaNO_3$	25	2.65 (0.04)	R1	95MAa
		1.0; $NaClO_4$	25	2.58 (0.03)	R	71GKb, 86ANb
	H_2L + H	1.0; KNO_3/Cl	25	1.82 (0.06)	P	81MOa, 76GMb
		0.5; $NaClO_4$	20	1.8 (0.1)	P	76AMa
		0.5; $NaClO_4$	25	1.79 (0.03)	P	87MDa
		1.0; $NaClO_4$	25	1.87 (0.02)	R	86ANb, 71GGa
$Mg^{2+\ e}$	M + L	0.1; KNO_3	20	2.94 (0.05)	P	64ANa
$Ca^{2+\ e}$	M + L	0.1; KNO_3	20	2.59 (0.05)	P	64ANa
Sr^{2+}	M + L	0.1; KNO_3	20	2.23 (0.05)	P	64ANa
Ba^{2+}	M + L	0.1; KNO_3	20	1.67 (0.05)	P	64ANa
Sn^{IV}	$(SnMe_2)^{2+}$ + L	0.1; KNO_3	25	9.4 (0.1)	P	92CGa

(*continues on next page*)

Table 1 (*Continued*).

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
Pb^{2+}	M + L	0.1; KNO_3	20	7.45 (0.03)	P	64ANa
		0.1; KNO_3	25	7.41 (0.01)	P	83FSa
		0.5; $NaClO_4$	25	7.3 (0.1)	P	72NAa
	M + H + L	0.5; $NaClO_4$	25	10.4 (0.1)	P	72NAa
	M + 2H + L	0.5; $NaClO_4$	25	12.7 (0.1)	P	72NAa
VO^{2+}	M + L	0.1; KNO_3	25	9.00 (0.01)	P	84FVa, 83FSa
		0.2; KCl	25	8.84 (0.02)	P	99SBb
	M + 2L	0.2; KCl	25	15.32 (0.06)	P	99SBb
Fe^{2+}	M + L	0.1; KNO_3	20	5.8 (0.1)	P	64ANa
		0.5; $NaClO_4$	25	5.1 (0.1)	P	72NAc
	M + 2L	0.1; KNO_3	20	10.0 (0.1)	P	64ANa
		0.5; $NaClO_4$	25	9.8 (0.2)	P	72NAc
Fe^{3+}	M + L	0.5; KNO_3	25	10.90 (0.02)	P	99SEb
		1.0; $NaClO_4$	25	11.1 (0.2)	P	86ANb
	M + 2L	0.5; KNO_3	25	19.33 (0.03)	P	99SEb
Co^{2+}	M + L	0.1; KNO_3	20	6.97 (0.05)	P	64ANa
		0.1; KCl	30	6.95 (0.02)	P	52CMa
	M + 2L	0.1; KNO_3	20	12.3 (0.2)	P	64ANa
		0.1; KCl	30	12.3 (0.1)	P	52CMa
Ni^{2+}	M + L	0.1; KNO_3	20	8.19 (0.03)	P	64ANa
		0.1; KNO_3	25	8.13 (0.03)	P	83FSa
	M + 2L	0.1; KNO_3	20	14.3 (0.1)	P	64ANa
Cu^{2+}	M + L	0.1; KNO_3	20	10.6 (0.2)	P	64ANa
		0.1; KNO_3	25	10.6 (0.1)	P	83FSa
		0.1; KCl	30	10.55 (0.08)	P	52CMa
	M + 2L	0.1; KNO_3	25	16.3 (0.1)	P	83SVa
		0.1; KCl	30	16.20 (0.05)	P	52CMa
Zn^{2+}	M + L	0.1; KCl	30	7.03 (0.02)	P	52CMa
		0.5; $NaClO_4$	25	7.0 (0.1)	P	92GLa
	M + 2L	0.5; $NaClO_4$	25	12.4 (0.1)	P	92GLa
Cd^{2+}	M + L	0.1; KNO_3	20	5.7 (0.2)	P	64ANa
		0.5; $NaClO_4$	25	5.55 (0.02)	P	92GLa
		0.1; KNO_3	25	5.48 (0.05)	P	83SVa
		0.1; KCl	30	5.4 (0.2)	P	52CMa
	M + 2L	0.5; $NaClO_4$	25	9.99 (0.02)	P	92GLa
Ag^{+}	M + L	1.0; KNO_3	25	3.27 (0.02)	P	92ANa
	M + 2L	1.0; KNO_3	25	5.90 (0.05)	P	92ANa
Pd^{2+}	M + L	1.0; $NaClO_4$	20	17.5 (0.1)	R1	76AMa
	ML + L	1.0; $NaClO_4$	20	9.3 (0.1)	R1	76AMa
Y^{3+}	M + L	0.1; KNO_3	25	6.8 (0.1)	P	62THa
	M + 2L	0.1; KNO_3	25	12.0 (0.1)	P	62THa

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Table 1 (*Continued*).

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
La^{3+}	M + L	0.1; KNO$_3$	25	5.9 (0.1)	P	62THa
	M + 2L	0.1; KNO$_3$	20	9.7 (0.2)	P	64ANa
		0.1; KNO$_3$	25	10.0 (0.2)	P	62THa
Ce^{3+}	M + L	0.1; KNO$_3$	25	6.2 (0.1)	P	62THa
	M + 2L	0.1; KNO$_3$	25	10.7 (0.1)	P	62THa
Pr^{3+}	M + L	0.1; KNO$_3$	25	6.4 (0.1)	P	62THa
	M + 2L	0.1; KNO$_3$	25	11.2 (0.1)	P	62THa
Nd^{3+}	M + L	0.1; KNO$_3$	25	6.54 (0.04)	P	62THa
Sm^{3+}	M + L	0.1; KNO$_3$	25	6.6 (0.1)	P	62THa
Eu^{2+}	M + L	0.5; NaClO$_4$	25	4.9 (0.1)	P	73CTa
	M + 2L	0.5; NaClO$_4$	25	7.5 (0.1)	P	73CTa
Eu^{3+}	M + L	0.1; KNO$_3$	25	6.7 (0.1)	P	62THa
		0.5; NaClO$_4$	25	6.62 (0.06)	P	73CTa
		1.0; NaClO$_4$	25	6.48 (0.08)	R	71GGa, 71GKb
	M + 2L	0.1; KNO$_3$	25	12.1 (0.1)	P	62THa
		1.0; NaClO$_4$	25	11.65 (0.05)	R	71GGa, 71GKb
	M + 3L	0.5; NaClO$_4$	25	15.5 (0.2)	P	73CT
		1.0; NaClO$_4$	25	15.70 (0.03)	R	71GGa, 71GKb
Gd^{3+}	M + L	0.1; KNO$_3$	25	6.68 (0.05)	P	62THa
	M + 2L	0.1; KNO$_3$	25	12.1 (0.1)	P	62THa
Tb^{3+}	M + L	0.1; KNO$_3$	25	6.78 (0.15)	P	62THa
Dy^{3+}	M + L	0.1; KNO$_3$	25	6.9 (0.1)	P	62THa
Ho^{3+}	M + L	0.1; KNO$_3$	25	7.0 (0.2)	P	62THa
Er^{3+}	M + L	0.1; KNO$_3$	25	7.1 (0.1)	P	62THa
	M + 2L	0.1; KNO$_3$	25	12.7 (0.1)	P	62THa
Tm^{3+}	M + L	0.1; KNO$_3$	25	7.2 (0.1)	P	62THa
	M + 2L	0.1; KNO$_3$	25	12.9 (0.1)	P	62THa
Yb^{3+}	M + L	0.1; KNO$_3$	25	7.4 (0.1)	P	62THa
	M + 2L	0.1; KNO$_3$	25	13.3 (0.1)	P	62THa
Lu^{3+}	M + L	0.1; KNO$_3$	25	7.6 (0.1)	P	62THa
	M + 2L	0.1; KNO$_3$	25	13.7 (0.1)	P	62THa
UO$_2$$^{2+}$	M + L	0.1; KNO$_3$	25	8.73 (0.04)	P	82NBa
		1.0; NaClO$_4$	20	8.7 (0.1)	P	73CBc
		1.0; KNO$_3$	25	8.73 (0.04)	P	64RMc
	M + 2L	0.1; KNO$_3$	25	17.28 (0.05)	P	82NBa

[a] Owing to the fact that IDA has been studied since 1945 [45SKa], there are many results, especially for its protonation, at temperatures of 20 or 25 °C. At the same time, for physiological conditions (I = 0.15 mol L^{-1} NaClO$_4$, 37 °C) only a single paper [92RAc] (Conc., gl) seems to present reliable data, although even these do not meet all the requirements for recommendation. The data presented by [92RAc] are also in disagreement with two other papers for similar conditions (I = 0.10 mol L^{-1} KNO$_3$, 35 °C) [78RSa, 77PTb].

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Table 1 (*Continued*).

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[b]The values for the formation constants of the complexes with some ions demonstrate a very large disparity. It was not possible to recommend values for Al^{3+}: the two publications on Al-IDA complexes [81DSa, 71LNb] reveal a large disagreement, likely because of different metal ion hydrolysis models and values being assumed. The results for Cr^{3+} show a large disparity [82VNa, 81DSa, 70MSd] associated with an inappropriate account of slow formation of the complexed species of this ion.

The Be^{2+} species are represented in two publications. One paper [81DSa] does not take account of the hydrolyzed species of the metal ion and of the complexes. The other paper has better-quality data [87MDa], but further independent research is needed for reliable evaluation.

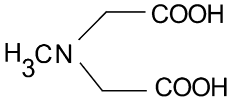
[c]There are also many cases where the formation constants of the complexes are in good agreement, but the data were not recommended because the protonation constant values for the ligand were assessed to be incorrect when compared with the **Recommended** values (e.g., [84HKa] and [78RSa]).

[d]For Ln^{3+}, the generally reliable reference [62THa] does not take into account the possible formation of LnL_3 complexes in 1:2 metal/ligand total molar ratio mixture [70KMe, 71GGa, 71GKb, 73CTa].

[e]The trend found in [64ANa] for Ca^{2+} and Mg^{2+} IDA complexes ($\lg K_{MgL} > \lg K_{CaL}$) is the inverse of that for MIDA [68NPb, 55SAa], Table 2. Thus, the corresponding data need further support by an independent study of both IDA and MIDA complexes with Mg^{2+} and Ca^{2+}.

[f]Values of $\lg K(M + L)$ reported for $NpO_2{}^+$-IDA complexes range from 5.64 to 8.72 [94TSa, 90RNc, 83ITa, 73CBc, 70EWa]. The value $\lg K(M + L) = 6.4$ (0.3) [94TSa], can be treated as reliable (1.0 mol L^{-1} $NaClO_4$, 25 °C, Conc., dis) although outside of the "**Provisional**" range.

6. 2,2'-(METHYLAZANEDIYL)DIACETIC ACID, ((METHYLIMINO)DIACETIC ACID), MIDA, H_2mida



Cations studied [a–e]: H^+ [b]: 99SEb, 96ANb, 92GLa, 87MDa, 86MDa, 83FSa, 79MMd, 77MGb, 77NAa, 77TIa, 76YNa, 75MRb, 70FSa, 68NPb, 66KRa, 66KUa, 65ANa, 56OMa, 55SAa, 45SKa

Be^{2+}: 87MDa
Mg^{2+} [e]: 77TIa, 69VPa, 68NPb, 65ANa, 56MAa, 55SAa, 45SKa
Ca^{2+} [e]: 75MRb, 69VPa, 68NPb, 65ANa, 55SAa, 45SKa
Sr^{2+}: 68NPb, 65ANa, 56MAa, 55SAa
Ba^{2+}: 68NPb, 65ANa, 55SAa, 45SKa
Al^{3+}: 84NAa
Pb^{2+}: 85NAa, 83FSa, 69VPa, 65ANa, 45SKa
Sn^{IV}: 97TNa, 96ANb
V^{V}: 76YNa
VO^{2+}: 83FSa, 77NAa
Cr^{2+}: 86MNa, 82CGa, 76BDa
Mn^{2+}: 69VPa, 65ANa, 56MAa, 55SAa
Fe^{2+}: 86MDa, 77TIa, 55SAa
Fe^{3+}: 99SEb
Co^{2+}: 77TIa, 73IVa, 72IVa, 65ANa, 55SAa
Ni^{2+}: 92GLa, 83FSa, 77TIa, 73IVa, 72IVa, 70CMa, 69VPa, 65ANa, 55SAa
Cu^{2+}: 92GLa, 83FSa, 77TIa, 73IVa, 72IVa, 69VPa, 65ANa, 55SAa
Zn^{2+}: 92GLa, 77MGb, 73HAb, 65ANa, 45SKa
Cd^{2+}: 92GLa, 73IVa, 72IVa, 65ANa, 55SAa
Hg^{2+}: 55SAa
Mo^{VI}: 66KRa, 66KUa
W^{VI}: 66KRa
Pd^{2+}: 87KUa
Y^{3+}: 80MGc
La^{3+}: 80MGc, 72KNb
Pr^{3+}: 80MGc
Nd^{3+} [d]: 80MGc, 79MMf
Sm^{3+}: 80MGc, 77TIa
Eu^{3+}: 80MGc
Gd^{3+}: 80MGc
Tb^{3+}: 80MGc
Dy^{3+}: 80MGc
Ho^{3+}: 80MGc
Er^{3+}: 80MGc
Tm^{3+}: 80MGc
Yb^{3+}: 80MGc
Lu^{3+}: 80MGc, 72KNb
$UO_2{}^{2+}$: 70FSa
NpO^{2+}: 90RNc, 70EWa

Experimental conditions of papers selected for critical evaluation:

$I = 0.1$ mol L^{-1} KCl, 20 °C, Conc., gl: 55SAa
$I = 0.1$ mol L^{-1} $NaClO_4$, 25 °C, Conc., gl: 68NPb
$I = 0.1$ mol L^{-1} KNO_3, 25 °C, Conc., gl: 83FSa, 70FSa
$I = 0.1$ mol L^{-1} KCl, 25 °C, Conc., gl: 86MDa, 68NPb
$I = 0.5$ mol L^{-1} $NaClO_4$, 25 °C, Conc., gl: 92GLa, 87MDa, 84NAa, 77NAa
$I = 0.5$ mol L^{-1} KNO_3, 25 °C, Conc., gl.: 99SEb
$I = 1.0$ mol L^{-1} $NaClO_4$, 25 °C, Conc., gl: 76YNa

Table 2 Recommended and provisional data for MIDA.

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
H^+ [b]	H + L	0.1; KCl	20	9.65 (0.07)	P	55SAa
		0.1; KCl/NO_3	25	9.59 (0.02)	R	86MDa, 68NPb, 83FSa
		0.5; $NaClO_4$	25	9.43 (0.03)	R	92GLa, 77NAa
		0.5; KNO_3	25	9.46 (0.03)	R1	99SEb
		1.0; $NaClO_4$	25	9.48 (0.06)	P	76YNa
	HL + H	0.1; KCl	20	2.12 (0.09)	P	55SAa
		0.5; $NaClO_4$	25	2.28 (0.02)	R	92GLa, 87MDa
		0.5; KNO_3	25	2.32 (0.03)	R1	99SEb
		1.0; $NaClO_4$	25	2.4 (0.1)	P	76YNa
	H_2L + H	0.5; KNO_3	25	1.4 (0.1)	P	99SEb
		1.0; $NaClO_4$	25	1.6 (0.1)	P	76YNa
Mg^{2+} [e]	M + L	0.1; KCl	20	3.4 (0.1)	P	55SAa
		0.1; $NaClO_4$	25	3.5 (0.1)	P	68NPb
	M + 2L	0.1; $NaClO_4$	25	5.83 (0.05)	P	68NPb
Ca^{2+} [e]	M + L	0.1; KCl	20	3.8 (0.1)	P	55SAa
		0.1; $NaClO_4$	25	3.9 (0.1)	P	68NPb
	M + 2L	0.1; $NaClO_4$	25	6.6 (0.1)	P	68NPb
Sr^{2+}	M + L	0.1; KCl	20	2.9 (0.1)	P	55SAa
		0.1; $NaClO_4$	25	3.0 (0.1)	P	68NPb
	M + 2L	0.1; $NaClO_4$	25	4.8 (0.1)	P	68NPb
Ba^{2+}	M + L	0.1; KCl	20	2.6 (0.1)	P	55SAa
		0.1; $NaClO_4$	25	2.6 (0.1)	P	68NPb
	M + 2L	0.1; $NaClO_4$	25	4.9 (0.1)	P	68NPb
Al^{3+}	M + L	0.5; $NaClO_4$	25	7.6 (0.1)	P	84NAa
Pb^{2+}	M + L	0.1; KNO_3	25	7.94 (0.05)	P	83FSa
		0.1; KCl	20	8.0 (0.1)	P	55SAa
	M + 2L	0.1; KCl	20	12.1 (0.1)	P	55SAa
VO_2^+	M + L	1.0; $NaClO_4$	25	10.2 (0.1)	P	76YNa
VO^{2+}	M + L	0.1; KNO_3	25	9.56 (0.06)	P	83FSa
		0.5; $NaClO_4$	25	9.44 (0.06)	P	77NAa
Mn^{2+}	M + L	0.1; KCl	20	5.4 (0.1)	P	55SAa
	M + 2L	0.1; KCl	20	9.6 (0.1)	P	55SAa

(*continues on next page*)

Table 2 (*Continued*).

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
Fe^{2+}	M + L	0.1; KCl	20	6.7 (0.1)	P	55SAa
		0.1; KCl	25	6.71 (0.04)	P	86MDa
	M + 2L	0.1; KCl	20	12.0 (0.1)	P	55SAa
		0.1; KCl	25	11.8 (0.1)	P	86MDa
Fe^{3+}	M + L	0.5; KNO_3	25	10.99 (0.03)	P	99SEb
	M + 2L	0.5; KNO_3	25	20.72 (0.03)	P	99SEb
Co^{2+}	M + L	0.1; KCl	20	7.6 (0.1)	P	55SAa
	M + 2L	0.1; KCl	20	13.9 (0.1)	P	55SAa
Ni^{2+}	M + L	0.1; KCl	20	8.7 (0.1)	P	55SAa
		0.1; KNO_3	25	8.7 (0.1)	P	83FSa
		0.5; $NaClO_4$	25	8.5 (0.1)	P	92GLa
	M + 2L	0.1; KCl	20	15.9 (0.1)	P	55SAa
Cu^{2+}	M + L	0.1; KCl	20	11.1 (0.1)	R1	55SAa
		0.1; KNO_3	25	11.04 (0.02)	R1	83FSa
		0.5; $NaClO_4$	25	10.9 (0.1)	P	92GLa
	M + 2L	0.1; KCl	20	17.9 (0.1)	P	55SAa
Zn^{2+}	M + L	0.1; KCl	20	7.7 (0.1)	P	55SAa
		0.5; $NaClO_4$	25	7.5 (0.1)	P	92GLa
	M + 2L	0.1; KCl	20	14.1 (0.2)	P	55SAa
		0.5; $NaClO_4$	25	13.7 (0.2)	P	92GLa
Cd^{2+}	M + L	0.1; KNO_3	20	6.8 (0.2)	P	55SAa
		0.5; $NaClO_4$	25	6.4 (0.2)	P	92GLa
	M + 2L	0.5; $NaClO_4$	25	11.8 (0.2)	P	92GLa
Hg^{2+}	M + L	0.1; KCl	25	5.5 (0.1)	P	55SAa
	M + 2L	0.1; KCl	25	9.2 (0.1)	P	55SAa
UO_2^{2+}	M + L	0.1; KNO_3	25	9.7 (0.1)	P	70FSa

[a] Although MIDA has been studied since 1945 [45SKa], no values have been reported for physiological conditions (I = 0.15 mol L^{-1} $NaClO_4$, 37 °C). Of the two values reported for 35 °C, one was measured at I = 2.0 mol L^{-1} KNO_3 [77MGb] and the other ([66KRa]) did not report experimental conditions.

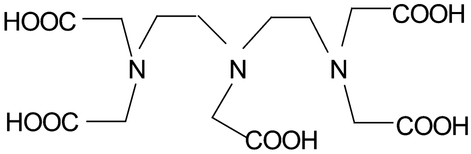
[b] The lg K(H + L) value for MIDA is higher than that for IDA by ca. 0.3 lg units.

[c] Generally, the observed trend for MIDA complexes is that they have higher stability constants than those for IDA.

[d] Reference [79MMf] gives reliable spectrophotometric value for NdL_3 complex formation owing to a good resolution of individual absorption bands for all species: lg $K(NdL_2 + L)$ = 3.4 (0.1) (I = 0.4 mol L^{-1} KCl and "room temperature").

[e] See comment for IDA complexes with Ca^{2+} and Mg^{2+}.

7. 2,2',2'',2'''-{[(CARBOXYMETHYL)AZANEDIYL]BIS[(ETHANE-1,2-DIYL)NITRILO]} TETRAACETIC ACID (DIETHYLENETRIAMINEPENTAACETIC ACID), DTPA, H_5dtpa



Cations studied [a–l]: **H^+** [a,b]: 01CCa, 99SBd, 97DFa, 96GMa, 94KCa, 92DHb, 91DMc, 90ADb, 88SC, 87ZGa, 84DMb, 84ZGa, 83KDb, 82OLa, 81MPa, 80KHb, 80MIa, 79LMa, 78MGa, 77GGb, 76AMa, 76HMd, 75NAb, 74MPb, 74BAa, 74MPa, 72KIa, 72MP, 70AMa, 68CL, 68KNa, 68WRa, 67ANb, 67NKb, 65AA, 65BAc, 65RVb, 62MTc, 62SKa, 60WAa, 59ANd, 59VCa, 58DRa, 55WAa

Li^{+}: 60WAa
Mg^{2+}: 84DMb, 68CW, 68WRa, 65AA, 65WHa, 60WAa, 58DRa
Ca^{2+}: 99SBd, 87ZGa, 84DMb, 84ZGa, 70AMa, 68CW, 68WRa, 65AA, 65WHa, 60HRa, 59ANd, 59CFc, 58DRa, 55WAa
Sr^{2+}: 68CW, 65AA, 65WHa, 62TIa, 60WAa, 58DRa, 55WAa
Ba^{2+}: 68CW, 65WHa, 58DRa, 60WAa, 55WAa
Ra^{2+}: 68SKa
Al^{3+}[c]: 96YHa, 80KHb, 68CA, 67ABb, 66MCa
Ga^{3+}: 97DFa, 80KHb, 76HMd, 73NKa, 70CAa, 67BAc, 66MCa
In^{3+}[d]: 99DLa, 97DFa, 80KHb, 74LKc, 72NKa, 67BAc, 66ZAc, 63RMb
Tl^{+}: 79ABa, 68KKa, 67ABc
Tl^{3+}[e]: 78VP, 67ABc, 67KAb, 67KA
Pb^{2+}: 93BNb, 72LWa, 69NKa, 65AA, 65WHa, 60HRa, 59ANd
Ti^{IV}: 70KB
Sb^{3+}: 71OBb, 70AMa
Bi^{3+}[f]: 87KTa, 67BAc, 67NKb
V^{3+}: 74TPa, 70KB
VO^{2+}: 75NAb
Cr^{3+}: 93BNb, 91BMa, 91NBa, 69KC
Mn^{2+}: 84DMb, 65AA, 65WHa, 60HRa, 60WAa, 59ANd, 59CFc, 58DRa
Mn^{3+}: 71BPh, 71MAn
Fe^{2+}: 00BMa, 85PLb, 65WHa, 59ANd, 59CFc, 59VCa, 58DRa
Fe^{3+}: 00BMa, 97DFa, 90ADb, 85PLb, 74MBa, 73KBc, 67BAc, 59ANd, 59VCa
Co^{2+}: 74MBa, 68KAb, 65AA, 65WHa, 60WAa, 59ANd, 59CFc, 58DRa
Co^{3+}: 72BCb
Ni^{2+}: 79KNa, 78KNc, 74MBa, 65AA, 65WHa, 60WAa, 59ANd, 59CFc, 58DRa
Cu^{2+}[g]: 91NBa, 85KLb, 84HKa, 74BAa, 74MBa, 73KBc, 69KTc, 65AA, 65WHa, 60WAa, 59ANd, 59CFc, 58DRa, 57HLa
Zn^{2+}: 98LVa, 84DMb, 74DTa, 68KA, 65AA, 65WHa, 60HRa, 60WAa, 59ANd, 59CFc, 58DRa
Cd^{2+}: 84DMb, 83YWa, 75LWa, 74DTa, 68KA, 65AA, 65WHa, 60HRa, 60WAa, 59ANd, 59CFc, 58DRa
Hg^{2+}: 77GGb, 75LBa, 67KAb, 65AA, 65WHa, 62MTc , 62SKa, 60HRa, 60WAa, 59ANd
Ag^{+}: 68WRa, 67OA
Zr^{4+}[h]: 67BAc, 67TIa, 66EMd, 66LPa, 64EMd, 64PVb
Hf^{4+}: 66EMd, 64EMd, 64PVb
Mo^{VI}: 71LUa
Pd^{2+}: 76AMa, 72KIa
Ru^{3+}: 88THa
Sc^{3+}: 69KA, 68CA, 67BAc
Y^{3+}: 94KCa, 87YJa, 77CGc 77GGb, 69KA, 68CA, 62MTc, 62STd, 59HCa
La^{3+}[i]: 00CVa, 98LVa, 88MIa, 87CN, 87YJa, 77CGc, 77GGb, 76GAa, 69KA, 68CA, 65AA, 62MTc, 60HRa, 59HCa
Ce^{3+}: 88MIa, 87CN, 87YJa, 87ZGa, 83HPb, 82CP, 77CGc, 77GGb, 71PVb, 71SHb, 68CL, 68LFb 62MTc, 59ANd
Ce^{4+}: 72YPa, 71PMc, 70MMb
Pr^{3+}: 88MIa, 87CN, 87YJa, 82CP, 77CGc, 77GGb, 71PRa, 70KA, 68CL, 62MTc, 59HCa
Nd^{3+}: 88MIa, 87CN, 87YJa, 84NMa, 82CP, 77CGc, 77GGb, 70KTd, 68CL, 63GB, 62MTc, 59HCa, 57HLa
Sm^{3+}: 88MIa, 87CN, 87YJa, 87ZGa, 84ZGa, 77CGc, 77GGb, 76GAa, 68CL, 63GA, 62MTc, 59HCa
Eu^{2+}: 73TKd
Eu^{3+}: 97WHb, 96WHa, 88MIa, 87CN, 87YJa, 87ZGa, 77CGc, 77GGb, 76GAa, 68CL, 65BAc, 63GA, 62MTc, 59HCa
Gd^{3+}: 00SBc, 99SBd, 97BH, 96GMa, 94KCa, 88MIa, 88SC, 87CN, 87YJa, 77CGc, 77GGb, 69KB, 68CL, 62MTc, 59HCa
Tb^{3+}: 88MIa, 87CN, 77CGc, 77GGb, 68CL, 62MTc, 59HCa
Dy^{3+}: 88MIa, 87CN, 87YJa, 77CGc, 77GGb, 68CL, 62MTc, 59HCa
Ho^{3+}: 88MIa, 87CN, 77CGc, 77GGb, 68CL, 62MTc, 59HCa

Er^{3+}: 88MIa, 87CN, 77CGc, 77GGb, 68CL, 62MTc, 59HCa
Tm^{3+}: 88MIa, 87CN, 77CGc, 77GGb, 68CL, 62MTc, 59HCa
Yb^{3+}: 89MIa, 87CN, 87YJa, 77CGc, 77GGb, 76GAa, 68CL, 62MTc, 59HCa
Lu^{3+}: 88MIa, 87CN, 77CGc, 77GGb, 69KB, 68CL, 62MTc
Th^{4+}: 90SS, 89KGa, 83HPb, 76PTb, 76PAa, 75PTb, 72PA, 67BAc
UO$_2^{2+}$[j]: 98BMa, 90SS, 82OLa, 80KJa
U^{4+}[k]: 72PR, 68CMb
U^{3+}: 69MOc
AmO$_2^+$: 74NSa
Am^{3+}[l]: 89RSa, 72PZ, 71BRa, 71MB, 71SHb, 69MOc, 69DBa, 68LFb, 66STb, 65BAc
Bk^{3+}: 66STb, 65BAc
Cf^{3+}: 71B, 66STb, 65BAc
Cm^{3+}: 72PW, 71BRa, 71MB, 71SHb, 69MOc, 68LFb, 66STb, 65BAc
Es^{3+}: 65BAc
Fm^{3+}: 65BAc
Pm^{3+}: 68LFb
Np^{3+}: 74KMd, 69MOc
Np^{4+}: 73CCc, 72PB, 71EPb, 71MA, 69MOc
NpO$_2^+$: 71MA
Pu^{3+}: 78MGa, 71MB, 69MOc
Pu^{4+}: 72PE, 71MA, 69MOc

Experimental conditions of papers selected for critical evaluation:

I = 0.1 mol L^{-1} KNO$_3$, 20 °C, Conc., gl: 79ABa, 67ANb, 67ABc, 66MCa, 62MTc
I = 0.1 mol L^{-1} KCl, 20 °C, Conc., gl, red: 59ANd, 68CMb (also, data for I = 0.1 mol L^{-1} KNO$_3$)
I = 0.1 mol L^{-1} KCl, 25 °C, Conc., luminescence: 96WHa
I = 0.1 mol L^{-1} KNO$_3$, 25 °C, Conc., gl: 97DFa, 82OLa, 76HMd, 74BAa, 66MCa, 62MTc, 60WAa, 59CFc, 55WAa; EMF: 68WRa, 60HRa; dis: 97DFa, 59VCa
I = 0.1 mol L^{-1} Na/HClO$_4$, 20 °C, Conc., red: 67BAc; gl: 70AMa
I = 0.1 mol L^{-1} NaClO$_4$, 25 °C, Conc., ext: 68SKa
I = 0.15 mol L^{-1} NaClO$_4$, 25 °C, Conc., gl, sp: 96GMa
I = 0.15 mol L^{-1} NaCl, 37 °C, Conc., gl: 91DMc, 84DMb
I = 0.5 mol L^{-1} NaClO$_4$, 25 °C, Conc., gl: 75NAb
I = 1.0 mol L^{-1} (CH$_3$)$_4$NCl, 20 °C, Conc., EMF: 67ANb
I = 1.0 mol L^{-1} KCl, 25 °C, Conc., gl: 80MIa, 78MGa
I = 1.0 mol L^{-1} Na/HClO$_4$, 20 °C, Conc., gl, sp: 91BMa, 76AMa, 67ANb; sol: 83KDb; red: 67ABc, 67BAc

Table 3 Recommended and provisional data for DTPA.

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
H$^+$[a,b]	H + L	0.1; KNO$_3$	20	10.58 (0.03)	R	67ANb, 62MTc
		0.1; KNO$_3$	25	10.54 (0.03)	R	97DFa, 76HMd, 74BAa
		0.15; NaClO$_4$	25	9.76 (0.02)	P	96GMa
		0.15; NaCl	37	9.67 (0.02)	R	91DMc, 84DMb
		1.0; (CH$_3$)$_4$NCl	20	10.46 (0.03)	P	67ANb
		1.0; KCl	25	10.06 (0.03)	P	80MIa
		1.0; NaCl	20	9.48 (0.03)	P	67ANb
	HL + H	0.1; KNO$_3$	20	8.60 (0.05)	R	74BAa, 67ANb, 62MTc
		0.1; KNO$_3$	25	8.56 (0.01)	P	97DFa
		0.15; NaClO$_4$	25	8.33 (0.03)	P	96GMa
		0.15; NaCl	37	8.27 (0.03)	P	84DMb
		1.0; (CH$_3$)$_4$NCl	20	8.41 (0.03)	P	67ANb
		1.0; KCl	25	8.32 (0.03)	P	80MIa
		1.0; NaCl	20	8.26 (0.03)	P	67ANb

(*continues on next page*)

Table 3 (*Continued*).

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
	$H_2L + H$	0.1; KNO_3	20	4.30 (0.03)	R	67ANb, 62MTc
		0.1; KNO_3	25	4.30 (0.03)	R	97DFa, 76HMd, 74BAa, 82OLa
		0.15; $NaClO_4$	25	4.18 (0.03)	P	96GMa
		0.15; $NaClO_4$	37	4.15 (0.03)	P	84DMb
		1.0; $(CH_3)_4NCl$	20	4.14 (0.03)	P	67ANb
		1.0; KCl	25	4.13 (0.03)	P	80MIa
		1.0; NaCl	20	4.19 (0.03)	P	67ANb
	$H_3L + H$	0.1; KNO_3	20	2.58 (0.03)	R	67ANb, 62MTc
		0.1; KNO_3	25	2.77 (0.05)	R	97DFa, 76HMd
		0.15; $NaClO_4$	25	2.68 (0.03)	P	96GMa
		0.15; $NaClO_4$	37	2.68 (0.03)	P	84DMb
		1.0; $(CH_3)_4NCl$	20	2.7 (0.1)	P	67ANb
		1.0; KCl	25	2.5 (0.1)	P	80MIa
		1.0; NaCl	20	2.5 (0.1)	P	67ANb
	$H_4L + H$	0.1; KNO_3	20	1.8 (0.1)	P	62MTc
		0.1; KNO_3	25	2.0 (0.1)	R	76HMd, 68WRa
		0.15; $NaClO_4$	25	2.0 (0.1)	P	96GMa
		0.15; NaCl	37	2.1 (0.1)	P	84DMb
		1.0; $(CH_3)_4NCl$	20	2.2 (0.1)	P	67ANb
		1.0; KCl	25	2.3 (0.1)	P	80MIa
		1.0; NaCl	20	1.9 (0.1)	P	83KDb
	$H_5L + H$	1.0; Na/$HClO_4$	20	1.2 (0.2)	P	83KDb
		1.0; K/HCl	25	1.7 (0.2)	P	80MIa
	$H_6L + H$	1.0; Na/$HClO_4$	20	0.8 (0.2)	P	83KDb
		1.0; K/HCl	25	0.9 (0.2)	P	80MIa
Li^+	M + L	0.1; KNO_3	25	3.1 (0.2)	P	60WAa
Mg^{2+}	M + L	0.1; KNO_3	25	9.3 (0.1)	R	68WRa, 60WAa
		0.15; NaCl	37	8.56 (0.05)	P	84DMb
	ML + H	0.1; KNO_3	25	6.9 (0.1)	P	60WAa
		0.15; NaCl	37	6.96 (0.05)	P	84DMb
	MHL + H	0.15; NaCl	37	4.68 (0.05)	P	84DMb
	$MH_2L + H$	0.15; NaCl	37	3.74 (0.05)	P	84DMb
	ML + M	0.15; NaCl	37	2.07 (0.05)	P	84DMb
Ca^{2+}	M + L	0.1; KNO_3	25	10.7 (0.1)	R	68WRa, 6OHRa, 59CFc, 55WAa
		0.1; $NaClO_4$	20	10.8 (0.1)	P	70AMa, 59ANd
		0.15; NaCl	37	9.8 (0.1)	P	84DMb
	ML + H	0.1; $NaNO_3$	20	6.10 (0.05)	P	59ANd
		0.1; KNO_3	25	6.10 (0.05)	P	59CFc
		0.15; NaCl	37	6.00 (0.05)	P	84DMb
	MHL + H	0.15; NaCl	37	4.40 (0.05)	P	84DMb
	$MH_2L + H$	0.15; NaCl	37	3.70 (0.05)	P	84DMb
	ML + M	0.1; $NaNO_3$	20	2.0 (0.1)	P	59ANd
		0.1; $NaNO_3$	37	2.1 (0.1)	P	84DMb

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Table 3 (*Continued*).

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
Sr^{2+}	M + L	0.1; KNO_3	25	9.7 (0.1)	P	60WAa, 55WAa
Ba^{2+}	M + L	0.1; KNO_3	25	8.8 (0.1)	P	60WAa, 55WAa
Ra^{2+}	M + L	0.1; $NaClO_4$	25	8.5 (0.1)	P	68SKa
Al^{3+} [c]	M + L	0.1; KNO_3	20	18.4 (0.1)	P	66MCa
		0.1; KNO_3	25	18.5 (0.1)	P	66MCa
	ML + H	0.1; KNO_3	20	4.6 (0.1)	P	66MCa
Ga^{3+}	M + L	0.1; $NaClO_4$	20	25.5 (0.1)	P	67BAc
		0.1; KNO_3	25	25.1 (0.1)	P	97DFa
	ML + H	0.1; $NaClO_4$	20	4.35 (0.05)	P	67BAc
		0.1; KNO_3	25	4.10 (0.05)	R	97DFa, 76HMd
	MHL + H	0.1; KNO_3	25	1.8 (0.1)	P	97DFa
	ML + OH	0.1; $NaClO_4$	20	6.52 (0.05)	P	67BAc
In^{3+} [d]	ML + OH	0.1; Na/$KClO_4$	20	2.06 (0.05)	P	67BAc
Tl^{+}	M + L	0.1; KNO_3	20	5.97 (0.05)	R1	79ABa, 67ABc
	ML + H	0.1; KNO_3	20	8.8 (0.1)	P	79ABa
Tl^{3+} [e]	M(OH)L + H	1.0; $NaClO_4$	20	10.9 (0.1)	P	67ABc
Pb^{2+}	M + L	0.1; $NaNO_3$	20	18.9 (0.1)	P	59ANd
	ML + H	0.1; $NaNO_3$	20	4.52 (0.05)	P	59ANd
	ML + M	0.1; $NaNO_3$	20	3.41 (0.05)	P	59ANd
Sb^{3+}	ML + H	0.1; $NaClO_4$	20	3.57 (0.05)	P	70AMa
	ML + 2OH	0.1; $NaClO_4$	20	9.82 (0.05)	P	70AMa
Bi^{3+} [f]	ML + H	1.0; $NaClO_4$	20	2.6 (0.1)	P	67BAc
	ML + OH	1.0; $NaClO_4$	20	2.7 (0.1)	P	67BAc
VO^{2+}	M + L	0.5; $NaClO_4$	25	16.3 (0.2)	P	75NAb
	ML + H	0.5; $NaClO_4$	25	7.0 (0.1)	P	75NAb
	2M + L	0.5; $NaClO_4$	25	23.3 (0.2)	P	75NAb
Cr^{3+}	M + L	1.0; Na/$HClO_4$	20	22.1 (0.1)	P	91BMa
	ML + H	1.0; Na/$HClO_4$	20	7.65 (0.05)	P	91BMa
	MHL + H	1.0; Na/$HClO_4$	20	6.15 (0.05)	P	91BMa
	MH_2L + H	1.0; Na/$HClO_4$	20	2.85 (0.05)	P	91BMa
	MH_3L + H	1.0; Na/$HClO_4$	20	1.50 (0.1)	P	91BMa
Mn^{2+}	M + L	0.1; $NaNO_3$	20	15.6 (0.1)	P	59ANd
		0.1; KNO_3	25	15.5 (0.1)	R	60HRa, 60WAa
		0.15; NaCl	37	14.3 (0.1)	P	84DMb
	ML + H	0.1; $NaNO_3$	20	4.03 (0.05)	P	59ANd
	ML + M	0.1; $NaNO_3$	20	2.1 (0.1)	P	59ANd
Fe^{2+}	M + L	0.1; $NaNO_3$	20	16.0 (0.1)	P	59ANd
	ML + H	0.1; $NaNO_3$	20	5.32 (0.05)	R1	59CFc
		0.1; KNO_3	25	5.30 (0.05)	R1	59ANd
	ML + M	0.1; $NaNO_3$	20	3.0 (0.1)	P	59ANd

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Table 3 (*Continued*).

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
Fe^{3+}	M + L	0.1; $NaClO_4$	20	27.3 (0.2)	P	67BAc
		0.1; KNO_3	25	27.8 (0.1)	P	97DFa
	ML + H	0.1; $NaClO_4$	20	3.58 (0.05)	R1	67BAc
		0.1; KNO_3	25	3.56 (0.05)	R1	59VCa
	ML + OH	0.1; $NaClO_4$	20	3.9 (0.1)	R1	67BAc
		0.1; KNO_3	25	4.1 (0.1)	R1	59VCa
Co^{2+}	M + L	0.1; $NaNO_3$	20	19.3 (0.1)	P	59ANd
	ML + H	0.1; $NaNO_3$	20	4.72 (0.05)	P	59ANd
	ML + M	0.1; $NaNO_3$	20	3.5 (0.1)	P	59ANd
Ni^{2+}	M + L	0.1; $NaNO_3$	20	20.2 (0.1)	P	59ANd
		0.1; KNO_3	25	20.1 (0.1)	P	60WAa, 59CFc
	ML + H	0.1; $NaNO_3$	20	5.62 (0.05)	P	59ANd
		0.1; KNO_3	25	5.6 (0.1)	P	60WAa, 59CFc
	ML + M	0.1; $NaNO_3$	20	5.4 (0.1)	P	59ANd
		0.1; KNO_3	25	5.6 (0.1)	P	60WAa, 59CFc
Cu^{2+}[g]	M + L	0.1; $NaNO_3$	20	21.5 (0.1)	P	59ANd
		0.1; $KClO_4$	25	21.5 (0.1)	P	74BAa
	ML + H	0.1; $NaNO_3$	20	4.74 (0.02)	R1	59ANd
		0.1; KNO_3/ClO_4	25	4.79 (0.02)	R	74BAa, 59CFc
	MHL + H	0.1; $KClO_4$	25	2.88 (0.05)	P	74BAa
	ML + M	0.1; $NaNO_3$	20	5.5 (0.1)	P	59ANd
Zn^{2+}	M + L	0.1; $NaNO_3$	20	18.6 (0.1)	P	59ANd
		0.15; NaCl	37	17.45 (0.05)	P	84DMb
	ML + H	0.1; $NaNO_3$	20	5.43 (0.05)	P	59ANd
		0.15; NaCl	37	5.08 (0.05)	P	84DMb
	MHL + H	0.15; NaCl	37	2.35 (0.05)	P	84DMb
	ML + M	0.1; $NaNO_3$	20	4.4 (0.1)	P	59ANd
Cd^{2+}	M + L	0.1; $NaNO_3$	20	19.3 (0.1)	P	59ANd
		0.15; NaCl	37	17.76 (0.05)	P	84DMb
	ML + H	0.1; $NaNO_3$	20	4.06 (0.05)	P	59ANd
		0.15; NaCl	37	3.77 (0.05)	P	84DMb
	MHL + H	0.15; NaCl	37	2.79 (0.05)	P	84DMb
	ML + M	0.1; $NaNO_3$	20	3.0 (0.1)	P	59ANd
Hg^{2+}	M + L	0.1; $NaNO_3$	20	26.7 (0.1)	R	59ANd, 62MTc
	ML + H	0.1; $NaNO_3$	20	4.2 (0.1)	R	59ANd, 62MTc
Ag^{+}	M + L	0.1; KNO_3	25	8.7 (0.1)	P	68WRa
Zr^{4+}[h]	ML + OH	1.0; $NaClO_4$	20	8.1 (0.1)	P	67BAc
Pd^{2+}	M + L	1.0; $HClO_4$	20	29.7 (0.1)	R1	76AMa
	ML + H	1.0; $NaClO_4$	20	3.49 (0.05)	R1	76AMa
	MHL + H	1.0; $NaClO_4$	20	2.9 (0.1)	P	76AMa
	MH_2L + H	1.0; $NaClO_4$	20	2.6 (0.1)	P	76AMa
	MH_3L + H	1.0; $NaClO_4$	20	1.9 (0.1)	P	76AMa
Eu^{3+}[i]	M + L	0.1; KNO_3	25	22.4 (0.1)	P	96WHa, 62MTc
Gd^{3+}[i]	M + L	0.1; KNO_3	25	22.5 (0.1)	P	62MTc

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Table 3 (*Continued*).

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
Th^{4+}	M + L	0.1; $NaClO_4$	20	28.8 (0.1)	P	67BAc
	ML + H	0.1; $NaClO_4$	20	2.16 (0.05)	P	67BAc
	ML + OH	0.1; $NaClO_4$	20	4.9 (0.2)	P	67BAc
UO_2^{2+} [j]	M + HL	0.1; KNO_3	25	8.8 (0.2)	P	82OLa
U^{4+} [k]	ML(OH) + H	0.1; KCl	25	7.8 (0.1)	P	68CMb

[a] The reaction enthalpy for DTPA protonation $\Delta_r H_c$ (H_nL + H) is exothermic for $n = 0$ or 1 and endothermic for $n = 3$ or 4 [74MPb, 62MTc]. The general trend is therefore a decrease in lg K(L + H) and lg K(HL + H) with an increase of temperature, with the opposite trend for lg K(H_3L + H) and lg K(H_4L + H).

[b] Values for DTPA protonation L + nH (n = 1 and 2) are strongly dependent on the concentration and nature of the background cation owing to complex formation with Na^+ and K^+. For $n > 2$, as well as for lg K(ML + H), the influence of the background cation's nature is negligible. Although [97CS] gives a stability constant for K^+ (lg K_{KL} = 3.1) the original reference was not found by the reviewers. But the same value is reported for the DTPA complex with Li^+ (lg K_{LiL} = 3.1) [60WAa].

[c] Unlike most other cations the formation of the Al^{III}-DTPA complex has a positive $\Delta_r H_c$. The values given by [66MCa] for 25 °C and 20 °C are consistent with this. The value lg K(AlL + OH) = 6.6 (0.5) [67ABb] is considered reliable (0.1 mol L^{-1} KNO_3, 25 °C).

[d] A critical evaluation of In^{III} stability constants [83TU] reports the mean lg K(In + L) = 28.4 (0.8) from three determinations: 29.0, 27.65 and 28.42 [67BAc, 63RMb, 66ZAc], and evaluates this value as doubtful. Later research [99DLa] based on spectrophotometric titration of the In^{III}-HBED-DTPA system in 0.5 mol L^{-1} Me_4NCl at 25 °C reported lg K(In + L) = 31.17 (0.02). This value is higher than lg K_{ML} for In^{III}-HBED and is assigned an unrealistic precision as some important experimental details are missing (e.g., lg K(L + nH) values of DTPA for 0.5 mol L^{-1} Me_4NCl at 25 °C). Nevertheless it supports indirectly the stability constant obtained in [67BAc] (lg K_{InL} = 29.0 for 0.1 mol L^{-1} $NaClO_4$, 25 °C), which is lower owing to electrolyte cation (Na/$KClO_4$) competition. Another recent publication based on the Fe^{3+}/Fe^{2+} redox equilibrium gives lg K(In + L) = 29.48 (0.04) [97DFa] for 0.1 mol L^{-1} KNO_3 and 25 °C . Thus, the value lg K(M + L) = 29.3 (0.5) (0.1 mol L^{-1} Na/$KClO_4$, 25 °C) [97DFa, 67BAc] is considered the most reliable to date.

[e] For Tl^{3+} [67ABc] gives an estimate of lg K(M + L) = 46 (1) (1.0 mol L^{-1} $NaClO_4$, 20 °C), which is considered the most reliable among data published, although far outside the "**Provisional**" range.

[f] For Bi^{3+} [67BAc] gives an estimate of lg K(M + L) = 36 (1) (1.0 mol L^{-1} $NaClO_4$, 20 °C), which is considered the most reliable among data published.

[g] lg K(CuH_2L + H) = 2.1(0.3) [85KLb] at I = 0.1 mol L^{-1} NaCl and 25 °C is is considered to be reliable.

[h] For Zr^{4+}, [67BAc] gives an estimate of lg K(M + L) = 37 (1) (1.0 mol L^{-1} $NaClO_4$, 20 °C), which is considered the most reliable among data published.

[i] For the rare earths, the sequence reported by [62MTc] (0.1 mol L^{-1} KNO_3, 25 °C, Conc.), is recommended as **Provisional**:

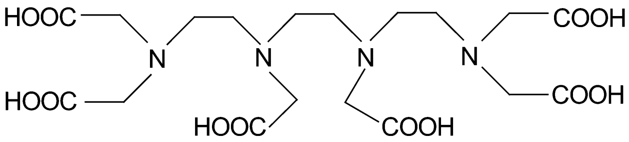
Ln^{3+}	lg K(M + L)	Ln^{3+}	lg K(M + L)
La^{3+}	19.48 (0.08)	Tb^{3+}	22.71 (0.08)
Ce^{3+}	20.5 (0.2)	Dy^{3+}	22.82 (0.08)
Pr^{3+}	21.07 (0.08)	Ho^{3+}	22.78 (0.08)
Nd^{3+}	21.6 (0.08)	Er^{3+}	22.74 (0.08)
Sm^{3+}	22.34 (0.08)	Tm^{3+}	22.72 (0.08)
Eu^{3+}	22.39 (0.08)	Yb^{3+}	22.62 (0.08)
Gd^{3+}	22.46 (0.08)	Lu^{3+}	22.44 (0.08)

[j] In the report for UO_2^{2+} [82OLa] lg K(H + L) and lg K(HL + H) values used were both 0.1 log units lower than the recommended ones, thus the originally reported stability constants lg K(2M + L) = 19.0 (0.4); lg K(2M + HL) = 13.4 (0.3) will be an underestimate by ca. 0.2 log units. Within this precision, these data are treated as reliable: lg K(2M + L) = 19.2 (0.6); lg K(2M + HL) = 13.5 (0.5).

[k] lg K(U^{IV} + L) can be estimated as ca. 30.9 [72PR] if one uses the data of [67BAc] for Th^{IV}, and the relative stability for U^{IV} and Th^{IV} complexes as reported in [72PR] for I = 0.1 – 0.5 mol L^{-1} and 20 °C.

[l] lg K(Am^{III} + L) = 23.0 (0.5) reported by [65BAc] (0.1 mol L^{-1} NH_4ClO_4, 25 °C, ix) is accepted as realistic, although it will be an underestimate owing to oversight of H_6dtpa^+ formation within the pH range 2.2–2.7.

8. 3,6,9,12-TETRAKIS(CARBOXYMETHYL)-3,6,9,12-TETRAAZATETRADECANEDIOIC ACID (TRIETHYLENETETRAMINEHEXAACETIC ACID), TTHA, H$_6$ttha



Cations studied: **H^{+}**[a]: 00CLa, 98ACc, 98AKa, 97DFa, 82OLa, 80KHb, 80MMd, 79LMa, 76HMd, 76NAa, 71LUa, 70LAd, 69ALb, 65BMf, 63GHa, 56FRa

Na^{+}[a]: 80KNa
Mg^{2+}[b]: 70HAa , 67BMd, 65BMf, 63GHa
Ca^{2+}: 69ALb, 65BMf, 63GHa
Sr^{2+}: 63GHa
Ba^{2+}: 63GHa
Al^{3+}[c]: 98ACa, 80KHb, 70HAa
Ga^{3+}: 98ACa, 98AKa, 97DFa, 80KHb, 80MMd, 76HMd, 69YMa
In^{3+}[d]: 97DFa, 84TMc
Tl^{+}: 77CNa
Tl^{3+}[e]: 00CLa
Pb^{2+}[f]: 88HPa, 71YMb, 70HAa, 69LUa, 68SCa
Bi^{3+}[g]: 79NPa, 69YMa
VO^{2+}[h]: 76NAa
Mn^{2+}: 98AKa, 70HAa
Fe^{2+}[i]: 65SCb
Fe^{3+}[j]: 97DFa, 70HAa, 67BMa, 65SCb
Co^{2+}[k,l]: 70HAa, 68SCa, 65BMf
Co^{3+}[m]: 69BHb
Ni^{2+}[k,n]: 70HAa, 68SCa, 67BMd, 65BMf
Cu^{2+}[k,o]: 70HAa, 69ALb, 67BMd, 67BMa, 65BMf, 65KKa
Zn^{2+}: 71NK, 71YMb, 70HAa, 70LAd, 69ALb, 69LUa, 68SCa
Cd^{2+}[p]: 88HPa, 81MNa, 71YMb, 70HAa, 69LUa, 68SCa, 65CKa
Hg^{2+}[q,r]: 90AC, 70HAa, 70LAd, 69YMa, 66SCb
Ag^{+}: 72RHb, 68WRa
Zr^{4+}[s]: 96YHa, 66ENc
Hf^{4+}: 96YHa, 66ENc
Mo^{VI}[t]: 71LUa
La^{3+}: 00CVa, 98AKa, 76GAa, 75AA, 70HAa, 69YMa, 65BMf
Ce^{3+}[u]: 69HGa
Pr^{3+}: 75AA
Nd^{3+}: 75AA, 70HAa, 69YMa
Sm^{3+}: 75AA, 69LUa, 68SCa
Eu^{3+}: 75AA, 76GAa,
Gd^{3+}: 75AA
Tb^{3+}: 75AA
Dy^{3+}: 75AA
Ho^{3+}: 75AA, 69YMa
Er^{3+}: 70HAa
Tm^{3+}: 75AA
Yb^{3+}: 76GAa, 75AA
Th^{4+}[v]: 85MSc, 70HAa, 65BMf
U^{4+}: 68CMb
UO$_2^{2+}$: 82NAc, 82OLa
Am^{3+}[w]: 69DBa

Experimental conditions of papers selected for critical evaluation:

I = 0.1 mol L^{-1} KNO$_3$, 20 °C, Conc., gl: 70LAd, 69ALb
I = 0.1 mol L^{-1} KNO$_3$, 25 °C, Conc., gl: 98AKa, 97DFa, 90AC, 82OLa, 79LMa, 75AA, 76HMd, 70HAa, 69BHb, 69LUa, 69YMa, 68CMb, 68WRa, 65BMf
I = 0.1 mol L^{-1} KCl, 30 °C, Conc., gl: 63GHa
I = 0.1 mol L^{-1} Me$_4$NNO$_3$, 25 °C, Conc., red: 98ACc
I = 0.5 mol L^{-1} NaClO$_4$, 25 °C, Conc., gl: 76NAa, 77CNa
I = 0.5 mol L^{-1} Na/KNO$_3$, 25 °C, Conc., kin: 80KNa
I = 1.0 mol L^{-1} NaClO$_4$, 25 °C, Conc., gl: 00CLa
I = 1.0 mol L^{-1} NaNO$_3$, 25 °C, Conc., gl: 00CLa

Table 4 Recommended and provisional data for TTHA.

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
H+ [a,b]	H + L	0.1; KNO_3	20	10.65 (0.03)	R1	69ALb
		0.1; KNO_3	25	10.62 (0.02)	R	98AKa, 97DFa, 79LMa, 76HMd
		0.1; Me_4NNO_3	25	10.63 (0.05)	R1	98ACc
		0.5; $NaClO_4$	25	9.73 (0.05)	P	76NAa
		1.0; $NaClO_4$	25	9.43 (0.03)	R1	00CLa
		1.0; $NaNO_3$	25	9.39 (0.02)	R1	00CLa
	HL + H	0.1; KNO_3	20	9.54 (0.03)	R1	69ALb
		0.1; KNO_3	25	9.54 (0.03)	R	98AKa, 97DFa, 79LMa, 76HMd
		0.1; Me_4NNO_3	25	9.46 (0.02)	R1	98ACc
		0.5; $NaClO_4$	25	8.76 (0.05)	P	76NAa
		1.0; $NaClO_4$	25	8.69 (0.03)	R1	00CLa
		1.0; $NaNO_3$	25	8.74 (0.03)	R1	00CLa
	H_2L + H	0.1; KNO_3	20	6.10 (0.02)	R1	69ALb
		0.1; KNO_3	25	6.15 (0.03)	R	98AKa, 97DFa, 79LMa, 76HMd
		0.1; Me_4NNO_3	25	6.11 (0.03)	R1	98ACc
		0.5; $NaClO_4$	25	5.92 (0.05)	P	76NAa
		1.0; $NaClO_4$	25	6.00 (0.03)	R1	00CLa
		1.0; $NaNO_3$	25	5.88 (0.03)	R1	00CLa
	H_3L + H	0.1; KNO_3	20	4.03 (0.04)	P	69ALb
		0.1; KNO_3	25	4.07 (0.03)	R	98AKa, 97DFa, 79LMa, 76HMd
		0.1; Me_4NNO_3	25	4.04 (0.04)	R1	98ACc
		0.5; $NaClO_4$	25	3.94 (0.05)	P	76NAa
		1.0; $NaClO_4$	25	3.99 (0.05)	P	00CLa
		1.0; $NaNO_3$	25	3.97 (0.03)	P	00CLa
	H_4L + H	0.1; KNO_3	20	2.7 (0.1)	P	69ALb
		0.1; KNO_3	25	2.79 (0.07)	P	98AKa, 97DFa, 79LMa, 76HMd
		0.1; Me_4NNO_3	25	2.75 (0.04)	R1	98ACc
		0.5; $NaClO_4$	25	2.8 (0.1)	P	76NAa
		1.0; $NaClO_4$	25	2.7 (0.1)	P	00CLa
		1.0; $NaNO_3$	25	2.6 (0.1)	P	00CLa
	H_5L + H	0.1; KNO_3	20	2.3 (0.1)	P	69ALb
		0.1; KNO_3	25	2.2 (0.1)	P	98AKa, 97DFa, 79LMa, 76HMd
		0.1; Me_4NNO_3	25	2.34 (0.07)	R1	98ACc
		0.5; $NaClO_4$	25	2.3 (0.1)	P	76NAa
		1.0; $NaClO_4$	25	2.3 (0.1)	P	00CLa
		1.0; $NaNO_3$	25	2.2 (0.1)	P	00CLa
	H_6L + H	0.1; KNO_3	25	1.8 (0.1)	P	98AKa, 79LMa
	H_7L + H	0.1; KNO_3	25	1.5 (0.1)	P	98AKa, 79LMa

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Table 4 (*Continued*).

Cation	Equilibrium	*I*/mol L^{-1}	*t*/°C	lg *K*	Category	References
Na^+	M + L	0.5; Na/KNO_3	25	1.0 (0.2)	P	80KNa
Mg^{2+}	M + L	0.1; KNO_3	25	8.5 (0.1)	P	70HAa, 65BMf
	ML + H	0.1; KNO_3	25	9.3 (0.1)	P	70HAa, 65BMf
	MHL + H	0.1; KNO_3	25	4.7 (0.1)	P	65BMf
	ML + M	0.1; KNO_3	25	5.9 (0.1)	P	70HAa
Ca^{2+}	M + L	0.1; KNO_3	20	10.5 (0.1)	P	69ALb
	ML + H	0.1; KNO_3	20	8.4 (0.1)	P	69ALb
	MHL + H	0.1; KNO_3	20	4.8 (0.1)	P	69ALb
	ML + M	0.1; KNO_3	20	4.2 (0.1)	P	69ALb
Sr^{2+}	M + L	0.1; KCl	30	9.3 (0.1)	P	63GHa
	ML + H	0.1; KCl	30	7.8 (0.1)	P	63GHa
	MHL + H	0.1; KCl	30	2.6 (0.1)	P	63GHa
	ML + M	0.1; KCl	30	3.5 (0.1)	P	63GHa
Ba^{2+}	M + L	0.1; KCl	30	8.2 (0.1)	P	63GHa
	ML + H	0.1; KCl	30	7.7 (0.1)	P	63GHa
	MHL + H	0.1; KCl	30	3.7 (0.1)	P	63GHa
	ML + M	0.1; KCl	30	3.4 (0.1)	P	63GHa
Al^{3+}	ML + H	0.1; K/Me_4NNO_3	25	5.9 (0.1)	P	98ACa,70HAa
Ga^{3+}	M + L	0.1; K/Me_4NNO_3	25	27.7 (0.2)	P	98ACa, 98AKa, 97DFa
	ML + H	0.1; K/Me_4NNO_3	25	5.1 (0.2)	P	98ACa, 98AKa
	MHL + H	0.1; K/Me_4NNO_3	25	4.0 (0.2)	P	98ACa, 98AKa
	M(OH)L + H	0.1; K/Me_4NNO_3	25	9.4 (0.2)	P	98ACa, 98AKa, 97DFa
In^{3+}	ML + H	0.1; KNO_3	25	7.3 (0.2)	P	97DFa
Tl^+	M + L	0.5; $NaClO_4$	25	4.9 (0.1)	P	77CNa
	ML + H	0.5; $NaClO_4$	25	9.7 (0.1)	P	77CNa
Tl^{3+}	ML + H	1.0; $NaClO_4$	25	5.1 (0.2)	P	00CLa
Pb^{2+}	M + L	0.1; KNO_3	25	18.0 (0.2)	P	70HAa, 69LUa
	ML + H	0.1; KNO_3	25	6.1 (0.2)	P	70HAa, 69LUa
	ML + M	0.1; KNO_3	25	11.0 (0.2)	P	70HAa, 69LUa
Mn^{2+}	M + L	0.1; KNO_3	25	14.7 (0.2)	P	98AKa, 70HAa
	ML + H	0.1; KNO_3	25	9.0 (0.2)	P	98AKa, 70HAa
	MHL + H	0.1; KNO_3	25	3.5 (0.2)	P	98AKa, 70HAa
	MH_2L + H	0.1; KNO_3	25	2.8 (0.2)	P	98AKa, 70HAa
	ML + M	0.1; KNO_3	25	6.3 (0.2)	P	98AKa, 70HAa
Fe^{3+}	ML + H	0.1; KNO_3	25	7.5 (0.2)	P	97DFa, 70HAa
Co^{2+}	ML + H	0.1; KNO_3	25	8.2 (0.2)	P	70HAa
	M_2L + H	0.1; KNO_3	25	3.0 (0.2)	P	70HAa
	M_2HL + H	0.1; KNO_3	25	2.6 (0.2)	P	70HAa
Ni^{2+}	ML + H	0.1; KNO_3	25	8.0 (0.2)	P	70HAa
	M_2L + H	0.1; KNO_3	25	2.6 (0.2)	P	70HAa

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Table 4 (*Continued*).

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
Cu^{2+}	ML + H	0.1; KNO_3	25	8.00 (0.05)	R	70HAa, 69ALb
	ML + M	0.1; KNO_3	25	13.5 (0.1)	P	70HAa, 69ALb
	M_2L + H	0.1; KNO_3	25	3.0 (0.2)	P	70HAa, 69ALb
	M_2HL + H	0.1; KNO_3	25	2.7 (0.2)	P	70HAa, 69ALb
Zn^{2+}	M + L	0.1; KNO_3	25	18.1 (0.2)	P	70LAd, 69ALb
	ML + H	0.1; KNO_3	25	8.1 (0.2)	P	70LAd, 69ALb
Cd^{2+}	M + L	0.1; KNO_3	25	18.7 (0.2)	P	70HAa
	ML + H	0.1; KNO_3	25	8.3 (0.2)	P	70HAa
	MHL + H	0.1; KNO_3	25	3.2 (0.3)	P	70HAa
	ML + M	0.1; KNO_3	25	8.2 (0.3)	P	70HAa
Hg^{2+}	M+L	0.1; KNO_3	25	28.1 (0.2)	P	90AC, 70HAa
	ML+H	0.1; KNO_3	25	6.2 (0.2)	P	90AC, 70HAa
	ML+M	0.1; KNO_3	25	13.8 (0.2)	P	90AC, 70HAa
Ag^{+}	M + L	0.1; KNO_3	25	8.7 (0.2)	P	68WRa
	ML + H	0.1; KNO_3	25	9.1 (0.2)	P	68WRa
	ML + M	0.1; KNO_3	25	5.2 (0.2)	P	68WRa
La^{3+}	ML + H	0.1; KNO_3	25	3.2 (0.1)	P	98AKa, 75AA, 70HAa
Pr^{3+}	ML + H	0.1; KNO_3	25	3.8 (0.2)	P	75AA
	MHL + H	0.1; KNO_3	25	2.4 (0.2)	P	75AA
Nd^{3+}	ML + H	0.1; KNO_3	25	3.9 (0.2)	P	75AA, 70HAa, 69YMa
	MHL + H	0.1; KNO_3	25	2.6 (0.2)	P	75AA, 69YMa
	ML + M	0.1; KNO_3	25	3.9 (0.2)	P	70HAa
Sm^{3+}	ML + H	0.1; KNO_3	25	4.5 (0.2)	P	75AA, 69LUa
	MHL + H	0.1; KNO_3	25	2.6 (0.2)	P	75AA, 69LUa
Eu^{3+}	ML + H	0.1; KNO_3	25	4.7 (0.2)	P	75AA
	MHL + H	0.1; KNO_3	25	3.1 (0.2)	P	75AA
Gd^{3+}	ML + H	0.1; KNO_3	25	4.5 (0.2)	P	75AA
	MHL + H	0.1; KNO_3	25	2.4 (0.2)	P	75AA
Tb^{3+}	ML + H	0.1; KNO_3	25	4.4 (0.2)	P	75AA
	MHL + H	0.1; KNO_3	25	2.3 (0.2)	P	75AA
Dy^{3+}	ML + H	0.1; KNO_3	25	4.5 (0.2)	P	75AA
	MHL + H	0.1; KNO_3	25	2.3 (0.2)	P	75AA
Ho^{3+}	ML + H	0.1; KNO_3	25	5.0 (0.2)	P	75AA
	MHL + H	0.1; KNO_3	25	2.3 (0.2)	P	75AA
Tm^{3+}	ML + H	0.1; KNO_3	25	4.7 (0.2)	P	75AA
	MHL + H	0.1; KNO_3	25	2.7 (0.2)	P	75AA
Yb^{3+}	ML + H	0.1; KNO_3	25	4.8 (0.2)	P	75AA
	MHL + H	0.1; KNO_3	25	2.5 (0.2)	P	75AA
Lu^{3+}	ML + H	0.1; KNO_3	25	5.0 (0.2)	P	75AA
Th^{4+}	ML + H	0.1; KNO_3	25	3.1 (0.2)	P	70HAa
U^{4+}	ML + H	0.1; KNO_3	25	2.3 (0.2)	P	68CMb

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Table 4 (*Continued*).

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
UO_2^{2+}	ML + H	0.1; KNO_3	25	2.3 (0.2)	P	82OLa
	M + H_2L	0.1; KNO_3	25	7.6 (0.2)	P	82OLa
	M + H_3L	0.1; KNO_3	25	5.5 (0.2)	P	82OLa
	2M + H_2L	0.1; KNO_3	25	11.8 (0.2)	P	82OLa

[a] As far as Na^+ forms complexes with complexones, the use of NaOH instead of KOH for titrations of H_6ttha in solutions with potassium salts as inert electrolytes should be avoided. NaOH leads to pH and lg K(H + L) values for Httha^{5-} [82OLa, 71LUa, 65BMf] lower than those obtained with KOH. There are no adequate investigations on Na^+ interaction, in contrast to the case of EDTA. From the TTHA protonation constants reported, the formation of a [Na(ttha)]$^{5-}$, and probably of a [Na_2(ttha)]$^{4-}$, complex appears possible. This explains why the protonation constants obtained by [65BMf] for ttha^{6-} (10.19) and Httha^{5-} (9.40) from titration of H_6ttha with NaOH in 0.1 mol L^{-1} KNO_3 and 25 °C are lower than those shown in Table 4, for which KOH was used. In contrast for the same conditions (inert salt concentration and temperature), nearly 40 years later Letkeman et al. using KNO_3 obtained lg K(H + L) = 10.68 [79LMa]. For this reason, the data presented by [65BMf], as well as those of [71LUa] and [82OLa], were excluded from the evaluation, although these values have been used in the calculation of stability constants by a large number of researchers, particularly those from North Europe [70HAa]. The resultant constants are lower and require correction in order to allow comparison with other values. For I = 0.1 mol L^{-1} KNO_3 and 25 °C, the protonation constants measured by titration with KOH [98AKa, 97DFa, 79LMa, 76HMd] give the accepted values in Table 4.

For Na^+, the complex formation has not been studied in detail. The single published value for Na^+ lg K(Na + L) = 1.00 is from a kinetic study of the reaction of the nickel(II)-TTHA complex with cyanide [80KNa]. Here, as also in other cases with TTHA, more studies are needed based on crystalline sodium compounds as was done for EDTA, in order to have information on the sodium coordination. Similar remarks are also valid for the other alkali cations.

[b] For Mg^{2+}, the lg K_{ML} value was recalculated by the reviewer; the published value (lg K_{ML} = 8.43), for which the protonation constants of [65BMf] had been used, was increased by 0.07 to 8.5.

[c] The TTHA complexation of Al^{3+} and Ga^{3+} is characterized by formation of ML as well as M_2L complexes. Both are present in mixtures with metal:ligand total molar concentration ratio 1:1. In several papers, this fact was not considered [80KHb, 80MMd], leading to erroneous constants. For Al^{3+}, only the values from [98ACa, 70HAa] and for Ga^{3+} those from [98ACa, 98AKa, 97DFa] are accepted. Additional values from these references, despite large standard deviations, are considered as the most reliable: lg K(Al + L) = 20.0 (0.3), lg K(2Al + L) = 28.8 (0.3), lg K(Al_2L + 2OH) = 16.8 (0.9), lg K(GaH_2L + L) = 2.1 (0.3), lg K(GaL + Ga) = 13.8 (1.4), lg K(Ga_2L + 2OH) = 19.7 (0.5), lg K(Ga_2L + H) = 1.2 (0.5) (0.1 mol L^{-1} K/Me_4NNO_3, 25 °C).

[d] For In^{3+}, the data presented by [97DFa] lg K(In + L) = 26.9 (0.5) and lg K(InL + In) = 9.0 (0.5) for 0.1 mol L^{-1} KNO_3 and 25 °C are the most reliable, although the uncertainty is greater than for **Provisional** assignment. The value reported in [84TMc] (lg K(In + L) = 26.75; 0.1 mol L^{-1} KCl , 25 °C, Conc., gl) is in close agreement with that in [97DFa], but the description of experimental conditions is inadequate.

[e] For Tl^{3+}, the most reliable value is: lg K(Tl + L) = 41 (1), 1.0 mol L^{-1} $NaClO_4$, 25 °C [00CLa].

[f] The fair agreement amongst other results for Pb^{2+}, allows specification of acceptable values, but some caution is needed in evaluation of their precision: lg K(Pb_2L + H) = 2.6 (0.4), lg K(Pb_2HL + H) = 2.3 (0.4) [70HAa, 69LUa] in 0.1 mol L^{-1} KNO_3, 25 °C.

[g] The determination of equilibrium constants from pH measurements in solutions containing a metal ion and a ligand is normally done avoiding the presence of polymeric hydrolysis products. In this respect, some caution is needed for metal ions such as Bi^{3+}, which forms $Bi_6(OH)_{12}^{6+}$ below pH 1.5. Apparently, the authors of [69YMa] used data for the determination of the different constants of Bi^{3+} without considering this fact. In another paper, by use of spectrophotometric measurements, the formation of [Bi(H_4ttha)]$^+$, [Bi(H_3ttha)], [Bi(Httha)]$^{2-}$, and [Bi(ttha)]$^{3-}$ was identified, but the author was not able to calculate the stability constants correctly as "only six" of the nine TTHA protonation constants were known [79NPa]. The corresponding values are therefore rejected in the present study.

[h] The investigations done by [76NAa] with VO^{2+} are inconclusive; the stability constants of the ML and M_2L complexes with this cation were not reported as they "are not easy to calculate because of overlapping equilibria". Also, the protonation constants for [VO(ttha)]$^{4-}$ are doubtful, because of the claimed presence of M_2L species in the mixture with total molar ratio metal:ligand of 1:1 [76NAa].

[i] Complex formation equilibria between Fe^{2+} and complexones are amongst the most difficult to study. This is because of its tendency to oxidize to the more stable TTHA-complex of trivalent iron. This process strongly influences the formation of the Fe^{2+}-complex. Only one paper was published: [65SCb]. The reported results indicate formation of ML, M_2L, and protonated complexes. However, no recommendation is possible. For comparison, see Mn^{2+} and Co^{2+}.

[j] For Fe^{3+}, two papers [65SCb, 70HAa] present similar results: for a 1:1 total molar ratio of metal:ligand a protonated complex

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Table 4 (*Continued*).

MHL forms with lg K(FeL + H) close to 8. For a 2:1 molar ratio, the complexes Fe_2L and $Fe_2(OH)_2L^{2-}$ form. As for the analogous complex $Na_2[Ga_2(OH)_2ttha(H_2O)_6]\cdot 2H_2O$ [98ACc], the latter has two equivalent Fe^{III} cations, each binding five donor atoms of one half of $ttha^{6-}$ ion. The remaining coordinated H_2O molecules undergo μ-diol formation with loss of one proton in acidic solution. In [70HAa], there is insufficient experimental information and a doubtful conversion of activities into concentrations. The values lg K(Fe+L) = 27.3 (0.5) [97DFa, 70HAa] and lg K(2Fe+L) = 40.2 (0.5) for Fe^{3+} in 0.1 mol L^{-1} KNO_3 , 25 °C are therefore treated as approximate.

[k] The assumption that only homobinuclear and not heterobinuclear species are present in solution has no theoretical justification. In [70LAd], the formation of $[CaCu(ttha)]^{2-}$ and $[CaZn(ttha)]^{2-}$ was reported; further Kopanica [71NK, 73HKb] reported equilibrium constants for the formation of $[CdNi(ttha)]^{2-}$, $[ZnNi(ttha)]^{2-}$, $[ZnCd(ttha)]^{2-}$ and $[CuNi(ttha)]^{2-}$. More recently the copper(II) and nickel(II) complexes of $[Th(ttha)]^{2-}$, $[Fe(ttha)]^{3-}$ [85MSc], and $[Tl(ttha)]^{3-}$ [00CLa] were characterized.

Another important point in relation to [70HAa] is the systematic use of unbuffered (pH or pM) solutions for electrode potential measurements. The error in the calculated constant could be significantly reduced by use of buffered (pM or pH) solutions, as in the determination of K(Hg + L) (using solutions with known and similar concentrations of HgL and excess of L), and for K(2Hg + L) (using solutions with known and similar concentrations of HgL and Hg_2L). Buffering is also important to reduce the effect of any impurities in the inert salt.

[l] For Co^{2+}, the values obtained by [70HAa] are more convincing than those of [67BMd], because in solutions with a total concentration molar ratio [Cation]:[Ligand] = 1:1 the species $Co_2H_pL^{(p-2)+}$ are considered in the evaluation of the constants. This is particularly important because, as a consequence of the formation of $Co_2H_pL^{(p-2)+}$ species, a certain amount of TTHA, not bound by M, is present in solution. Calculations from these experimental data considering uniquely the presence of MH_pL species give erroneous results. The data lg K(Co + L) = 18.2 (0.3) and lg K(CoL + Co) = 11.7 (0.3) in 0.1 mol L^{-1} KNO_3, 25 °C [70HAa] are the most reliable.

[m] The value published by [69BHb] for Co^{3+} is considered approximate: lg K(M + L) = 49.5 (0.5) in 0.1 mol L^{-1} KNO_3, 25 °C.

[n] For Ni^{2+}, the values of [70HAa] lg K(Ni + L) = 19.1 (0.3) and lg K(NiL + Ni) = 14.2 (0.3), 0.1 mol L^{-1} KNO_3, 25 °C are outside the precision requirements of the present review.

[o] Three papers report values for lg K(Cu + L) in 0.1 mol L^{-1} KNO_3 that are in poor agreement: 19.2 ([70HAa], 25 °C), 21.87 ([69ALb], 20 °C) and 20.3 ([67BMd], 25 °C), but with fair agreement for lg K(CuL + H) [69ALb, 70HAa, 67BMd]: 8.03; 7.96 and 8.0, Table 4. For CuL, the mean value lg K(Cu + L) = 20.5 ± 1.3 has an uncertainty that exceeds our criteria. Unfortunately, the values from [84HKa] presented in [03IU] are the secondary citation from [89MS], and therefore cannot be considered here.

[p] For Cd^{2+}, the values presented by [70HAa]: lg K(CdHL + H) = 3.2 (0.3); lg K(CdL + Cd) = 8.2 (0.3) are considered to be the most reliable, although the uncertainties are outside the precision limits assumed for "**Provisional**" classification.

[q] In the case of Hg^{2+}, several authors tried to use a mercury electrode, but had limited success in obtaining equilibrium constants. In the case of 0.1 mol L^{-1} $NaClO_4$ as inert salt [66SCb], difficulties are expected in the measurement of potentials between a mercury electrode and the saturated calomel electrode (SCE). This is because of the insolubility of $KClO_4$, which is formed at the contact boundary between the investigated solution and that of the reference electrode (3.74 mol L^{-1} KCl). This problem is avoided by use of KNO_3 as inert salt [70HAa]. Further, preliminary measurements by [76NAa] using a mercury electrode in solution with Hg^{II}, V^{IV}, and TTHA in total molar ratio 1:1:1 showed the formation of a precipitate, even at high pH. Schröder [66SCb] noted the formation of a precipitate at a total molar ratio [Hg]:[TTHA] = 2:1, following addition of Hg^{II} to a H_6ttha solution. However, the precipitate dissolved after base addition. For this, he assumed the formation of $[Hg_2(OH)_2ttha]^{4-}$, but without any experimental confirmation.

[r] The hydrolysis of Hg_2L does not correspond to the formation of $Hg_2(OH)_2L$ [66SCb, 70HAa] but to the following reaction:

$$Hg_2L^{2-} + 2\,OH^- \rightleftharpoons HgL^{4-} + Hg(OH)_2$$

with formation of soluble monomolecular $Hg(OH)_2$ [90AC, 58ASa].

[s] According to Ermakov and coauthors, the complex formation with Zr^{4+} and Hf^{4+} in 1–2 mol L^{-1} $HClO_4$ occurs with formation of MH_2ttha: lg K(M + H_2L) = 19.7 (Zr) and 19.1 (Hf) [66ENc]. As the exact composition of acidic solutions of these cations is still unknown, it is not possible to evaluate the importance of these results. Besides, no cationic TTHA species were considered.

[t] From alkalimetric titrations of protonated TTHA in the presence of disodium molybdate, Lund [71LUa] (0.1 mol L^{-1} KNO_3, 25 °C) determined the equilibrium constants of eight different complexes: lg $K(2MoO_4 + H_6L \rightleftharpoons (MoO_3)_2H_2L + 2H_2O)$ 13.80 (0.25); lg $K(2MoO_4 + H_5L \rightleftharpoons (MoO_3)_2HL + 2H_2O)$ 11.70 (0.25); lg $K(2MoO_4 + H_4L \rightleftharpoons (MoO_3)_2L + 2H_2O)$ 8.4 (0.3); lg $K(MoO_4 + H_6L \rightleftharpoons MoO_3H_4L + H_2O)$ 7.45 (0.30); lg $K(MoO_4 + H_5L \rightleftharpoons MoO_3H_3L + H_2O)$ 6.7 (0.3); lg $K(MoO_4 + H_4L \rightleftharpoons MoO_3H_2L + H_2O)$ 5.60 (0.25); lg $K(MoO_4 + H_3L \rightleftharpoons MoO_3HL + H_2O)$ 3.2 (0.3); lg $K(MoO_4 + H_2L \rightleftharpoons MoO_3L + H_2O)$ 3.1 (0.3). The protonation constants of TTHA were determined under the same conditions. "The $(MoO_3)_2L^{6-}$ complex was, however, just on the limit of detection, and the stability constant of that complex should consequently be regarded with suspicion", Lund [71LUa]. An examination of the complex distribution diagram shows that similar conclusions are valid also for MoO_3HL^{5-}, $MoO_3H_3L^{3-}$, and $MoO_3H_4L^{2-}$. This remark was considered in our assignment of uncertainty.

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Table 4 (*Continued*).

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[u] Among the rare earth trivalent cations, the information given in [69HGa] on the Ce^{3+} complexes cannot be considered because the temperature and the use of an inert salt are not specified.

The most important studies on trivalent lanthanide complexes are those of [70HAa, 69LUa] and [75AA]. The latter represents a systematic investigation of 12 different cations excluding Er and Pm. These cations have a coordination number larger than six and form very stable ML complexes with TTHA. In $[Ln(ttha)]^{3-}$, few donor atoms remain free for a second cation, thus, the stability constants for formation of $[M_2(ttha)]$ from $[M(ttha)]^{3-}$ are small, their logarithmic values being lower than 4. For lg K(M + L), the following approximate values are reported: La^{3+}: 23.4 (1) [98AKa, 75AA, 70HAa, 65BMf], Pr^{3+}: 23.7 (1) [75AA], Nd^{3+}: 23.8 (1) [75AA, 70HAa], Sm^{3+}: 23.7 (1) [75AA], Eu^{3+}: 23.5 (1) [75AA], Gd^{3+}: 23.5 (1) [75AA], Tb^{3+}: 23.6 (1) [75AA], Dy^{3+}: 23.7 (1) [75AA], Ho^{3+}: 23.6 (1) [75AA], Er^{3+}: 23.2 (1) [70HAa], Tm^{3+}: 23.2 (1) [75AA], Yb^{3+}: 23.0 (1) [75AA] and Lu^{3+}: 23.0 (1) [75AA].

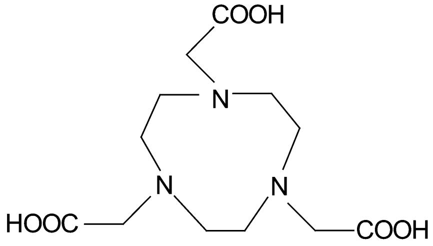
The large standard deviations are a consequence of the difficulty in determining values of lg K(M + L) > 20. The s.d. assigned above corresponds to that for a pH shift of only 0.04. Only in the case of Hg^{2+}, but by use of a very sensitive and stable electrode, can the s.d. have a lower value.

The values lg K(ErL + H) = 4.5 (0.2) and lg K(ErL + Er) = 3.8 (0.2) of [70HAa] contrast with those presented in [75AA] for other lanthanide ions.

[v] The value for Th^{IV} [70HAa] is accepted as approximate: lg K(M + L) = 32 (1) (0.1 mol L^{-1} KNO_3, 25 °C).

[w] The value in [69DBa] for Am^{3+} lg K(M + L) = 27.6 (0.1 mol L^{-1} NH_4ClO_4 , 25 °C) contrasts with data published for Ln^{III} complexes (ca. 4 lg units higher). Further research is therefore needed.

9. 2,2',2''-(1,4,7-TRIAZANONANE-1,4,7-TRIYL)TRIACETIC ACID, NOTA, H_3nota



Cations studied: **H^+** [a]: 96GEb, 93KT, 91GSa, 91CMd, 87BGc, 85GS, 85MBb

Mg^{2+} [b]:	87BGc, 85MBb	**Co^{2+}**:	75HTa	**Eu^{3+}** [e]:	97WHb, 87CN
Ca^{2+} [b]:	87BGc, 85MBb	**Cu^{2+}** [d]:	87BGc, 75HTa	**Gd^{3+}** [e]:	91BCa, 90BSe, 87CN
Sr^{2+} [b]:	87BGc	**Zn^{2+}**:	75HTa	**Tb^{3+}** [e]:	87CN
Ba^{2+} [b]:	87BGc	**Cd^{2+}**:	75HTa	**Dy^{3+}** [e]:	87CN
Ga^{3+}:	91CMd	**La^{3+}** [e]:	87CN	**Ho^{3+}** [e]:	87CN
In^{3+}:	91CMd	**Ce^{3+}** [e]:	90BSe, 87CN	**Er^{3+}** [e]:	87CN, 90BSe
Pb^{2+}:	75HTa	**Pr^{3+}** [e]:	87CN	**Tm^{3+}** [e]:	87CN
Mn^{2+} [c]:	90CBc, 75HTa	**Nd^{3+}** [e]:	87CN	**Yb^{3+}** [e]:	87CN
Fe^{3+}:	91CMd	**Sm^{3+}** [e]:	87CN	**Lu^{3+}** [e]:	87CN

Experimental conditions of papers selected for critical evaluation:

I = 0.1 mol L^{-1} $NaNO_3$, 25 °C, Conc., gl: 87BGc
I = 0.1 mol L^{-1} $NaNO_3$, 35 °C, Conc., gl: 87BGc
I = 0.1 mol L^{-1} KNO_3, 25 °C, Conc., gl: 96GEb
I = 0.1 mol L^{-1} KCl, 25 °C, Conc., gl: 91CMd
I = 0.1 mol L^{-1} KCl, 25 °C, Conc., sp: 91CMd
I = 0.1 mol L^{-1} $(CH_3)_4NCl$, 25 °C, Conc., gl: 91CMd
I = 1.0 mol L^{-1} $NaClO_4$, 25 °C, Conc., gl: 87BGc
I = 1.0 mol L^{-1} $NaClO_4$, 35 °C, Conc., gl: 87BGc

$I = 1.0$ mol L^{-1} $NaClO_4$, 25 °C, Conc., sp: 87BGc
$I = 0.1$ mol L^{-1} $NaClO_4$, 35 °C, Conc., sp: 87BGc

Table 5 Recommended and provisional data for NOTA[f].

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
H^{+} [a]	H + L	0.1; $NaNO_3$	25	11.73 (0.02)	P	87BGc
		0.1; $NaNO_3$	35	11.58 (0.02)	P	87BGc
		0.1; $K/(CH_3)_4NCl$	25	11.98 (0.03)	P	91CMd
		1.0; $NaClO_4$	25	10.77 (0.01)	P	87BGc
		1.0; $NaClO_4$	35	10.54 (0.01)	P	87BGc
	HL + H	0.1; $NaNO_3$	25	5.74 (0.01)	R1	87BGc
		0.1; $NaNO_3$	35	5.67 (0.01)	R1	87BGc
		0.1; $K/(CH_3)_4NCl$	25	5.65 (0.02)	R1	91CMd
		0.1; $NaClO_4$	25	5.62 (0.04)	R1	96GEb
		0.1; KNO_3	25	5.58 (0.04)	R1	96GEb
		1.0; $NaClO_4$	25	6.03 (0.01)	R1	87BGc
		1.0; $NaClO_4$	35	5.93 (0.01)	R1	87BGc
	H_2L + H	0.1; $NaNO_3$	25	3.16 (0.01)	R1	87BGc
		0.1; $NaNO_3$	35	3.17 (0.01)	R1	87BGc
		0.1; $K/(CH_3)_4NCl$	25	3.18 (0.03)	R1	91CMd
		0.1; $NaClO_4$	25	3.03 (0.04)	R1	96GEb
		0.1; KNO_3	25	2.97 (0.04)	R1	96GEb
		1.0; $NaClO_4$	25	3.16 (0.01)	R1	87BGc
		1.0; $NaClO_4$	35	3.19 (0.01)	R1	87BGc
	H_3L + H	1.0; $NaClO_4$	25	1.96 (0.01)	P	87BGc
		1.0; $NaClO_4$	35	2.02 (0.01)	P	87BGc
Mg^{2+} [b]	M + L	0.1; $NaNO_3$	25	9.69 (0.03)	P	87BGc
		0.1; $NaNO_3$	35	9.66 (0.03)	P	87BGc
	ML + H	0.1; $NaNO_3$	25	4.6 (0.2)	P	87BGc
Ca^{2+} [b]	M + L	0.1; $NaNO_3$	25	8.92 (0.01)	P	87BGc
		0.1; $NaNO_3$	35	8.74 (0.01)	P	87BGc
	ML + H	0.1; $NaNO_3$	25	5.06 (0.03)	P	87BGc
		0.1; $NaNO_3$	35	5.17 (0.05)	P	87BGc
Sr^{2+} [b]	M + L	0.1; $NaNO_3$	25	6.83 (0.01)	P	87BGc
		0.1; $NaNO_3$	35	6.76 (0.01)	P	87BGc
	ML + H	0.1; $NaNO_3$	25	6.30 (0.01)	P	87BGc
		0.1; $NaNO_3$	35	6.00 (0.01)	P	87BGc
Ba^{2+} [b]	M + L	0.1; $NaNO_3$	25	5.10 (0.01)	P	87BGc
		0.1; $NaNO_3$	35	5.06 (0.01)	P	87BGc
Ga^{3+}	M + L	0.1; KCl	25	31.0 (0.2)	P	91CMd
	ML + OH	0.1; KCl	25	4.1 (0.1)	P	91CMd
In^{3+}	M + L	0.1; KCl	25	26.2 (0.2)	P	91CMd
	ML + OH	0.1; KCl	25	7.2 (0.1)	P	91CMd
Fe^{3+}	M + L	0.1; KCl	25	28.3 (0.1)	P	91CMd

(*continues on next page*)

Table 5 (*Continued*).

Cation	Equilibrium	I/mol L^{-1}	t/°C	lg K	Category	References
Cu^{2+} [d]	M + L	1.0; $NaClO_4$	25	21.63 (0.03)	P	87BGc
		1.0; $NaClO_4$	35	21.30 (0.03)	P	87BGc
	ML + H	1.0; $NaClO_4$	25	2.74 (0.06)	P	87BGc
		1.0; $NaClO_4$	35	2.74 (0.06)	P	87BGc

[a] Enthalpy and entropy changes for protonation reactions have been derived from the temperature dependence of the protonation constants in the range 15–55 °C [87BGc]: for H + L, Δ_rH_c = –46 (7) kJ mol^{-1} and Δ_rS_c = 68 (22) J K^{-1} mol^{-1} (0.1 mol L^{-1} $NaNO_3$); Δ_rH_c = –46(4) kJ mol^{-1} and Δ_rS_c = 52 (12) J K^{-1} mol^{-1} (1 mol L^{-1} $NaClO_4$); for HL + H, Δ_rH_c = –8.8 (0.8) kJ mol^{-1} and Δ_rS_c = 80 (3) J K^{-1} mol^{-1} (0.1 mol L^{-1} $NaNO_3$); Δ_rH_c = –16.3 (0.4) kJ mol^{-1} and Δ_rS_c = 61 (2) J K^{-1} mol^{-1} (1 mol L^{-1} $NaClO_4$); for H_2L + H, Δ_rH_c = 5.9 (0.8) kJ mol^{-1} and Δ_rS_c = 80 (3) J K^{-1} mol^{-1} (0.1 mol L^{-1} $NaNO_3$); Δ_rH_c = 5.4 (0.4) kJ mol^{-1} and Δ_rS_c = 78 (1) J K^{-1} mol^{-1} (1 mol L^{-1} $NaClO_4$); for H_3L + H, Δ_rH_c = 5 (3) kJ mol^{-1} and Δ_rS_c = 54 (9) J K^{-1} mol^{-1} (1 mol L^{-1} $NaClO_4$).

[b] Enthalpy and entropy changes for complexation have been derived from the temperature dependence of the ML stability constants in the range 15–55 °C, or 25–55 °C, [87BGc]. For Mg^{2+} (where no value was determined at 15 °C because of the long equilibration time): Δ_rH_c = 2 (4) kJ mol^{-1} and Δ_rS_c = 188 (13) J K^{-1} mol^{-1}; for Ca^{2+}, Δ_rH_c = –25 (1) kJ mol^{-1} and Δ_rS_c = 88 (4) J K^{-1} mol^{-1}; for Sr^{2+}, Δ_rH_c = –9 (1) kJ mol^{-1} and Δ_rS_c = 100 (4) J K^{-1} mol^{-1}, for Ba^{2+}, Δ_rH_c = –5.8 (0.5) kJ mol^{-1} and Δ_rS_c = 78 (2) J K^{-1} mol^{-1}.

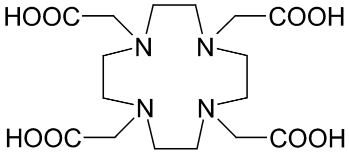
[c] For Mn^{2+}, an average value of lg K_{ML} = 14.6 (0.3) (I = 0.1 mol L^{-1} Me_4NCl, 25 °C, relaxation rate measurements, Conc. [90CBc], and I = 0.1 mol L^{-1} KNO_3, 25 °C, polarography, Conc. [75HTa]) is considered to be reliable.

[d] Enthalpy and entropy of complexation have been derived from the temperature dependence of the stability constants in the range 15–55 °C [87BGc]: for Cu + L, Δ_rH_c = –56 (3) kJ mol^{-1} and Δ_rS_c = 226 (8) J K^{-1} mol^{-1}, for CuL + H, Δ_rH_c = 0.4 (3) kJ mol^{-1} and Δ_rS_c = –54 (8) J K^{-1} mol^{-1}.

[e] Two publications [97WHb, 87CN] report stability constants for lanthanides. [97WHb] gives lg K(M + L) = 13.9 (0.1) for Eu^{3+} from a luminescence-based method, whereas [87CN] gives values for the full series of cations, but only as a graphical representation of the values obtained from competitive spectrophotometric measurements using arsenazo(III) as auxiliary ligand.

[f] The quality of data presented by [87BGc] is generally high. If there had been at least one lg K_{ML} value from an independent group to give R-level with [87BGc], then all of the values in Table 5 represented by [87BGc] could have been nominated as **R1**. No such result is available.

10. 2,2',2'', 2'''-(1,4,7,10-TETRAAZACYCLODODECANE -1,4,7,10-TETRAYL) TETRAACETIC ACID, DOTA, H_4dota



Cations studied: **H^+** [a,b]: 00BCa, 98BFa, 96CHc, 95KKa, 95PMa, 94KCa, 94TBb, 93KCa, 92CDd, 91DSa, 91CMa, 84DFa, 82DSa, 81DMa, 81SFa, 79DE, 76SFb

Li^+ [c]: 82DSa
Na^+: 00BCa, 82DSa, 81DMa, 79DE
K^+: 91CMa, 82DSa
Be^{2+} [d]: 82DSa
Mg^{2+}: 96CHc, 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Ca^{2+}: 00BCa, 96CHc, 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Sr^{2+}: 96CHc, 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Ba^{2+}: 96CHc, 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Al^{3+}: 95KKa
Ga^{3+}: 91CMb
In^{3+}: 91CMb
Mn^{2+}: 01BCa, 92CDd, 81SFa
Fe^{2+} [e]: 92CDd
Fe^{3+}: 91CMb
Co^{2+} [e]: 92CDd, 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Ni^{2+} [e]: 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Cu^{2+} [e]: 00BCa, 92CDd, 91CMa, 84DFa, 82DSa, 81SFa, 76SFb

Zn^{2+}: 00BCa, 92CDd, 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Cd^{2+}: 92CDd, 90CCa, 76SFb
Hg^{2+}: 94KOb
$Pb^{2+\,f}$: 95PMa, 92CDd, 90CCa, 81SFa, 76SFb
$Y^{3+\,g}$: 94KCa, 91BCc, 89CJ
$La^{3+\,h}$: 97WH, 94TBb, 91CMb, 87CN
$Ce^{3+\,h}$: 98BFa, 97WHa, 94TBb, 87CN
Pr^{3+}: 97WHb, 94TBb, 87CN
Nd^{3+}: 97WHb, 94TBb, 87CN
Sm^{3+}: 97WHb, 94TBb, 87CN
Eu^{3+}: 97WHb, 95WH, 94TBb, 87CN, 86LDb
$Gd^{3+\,h}$: 00BCa, 97WHb, 96PW, 96BCd, 95WH, 94TBb, 94KCa, 93KCa, 92AAb, 92WJa, 91CA, 91CMb, 87CN, 79DE
Tb^{3+}: 97WHb, 94TBb, 87CN, 86LDb
Dy^{3+}: 97WHb, 94TBb, 87CN
Ho^{3+}: 97WHb, 94TBb, 87CN
Er^{3+}: 97WHb, 94TBb, 87CN
Tm^{3+}: 97WHb, 94TBb, 87CN
$Yb^{3+\,h}$: 98BFa, 97WHb, 94TBb, 87CN
Lu^{3+}: 97WHb, 94TBb, 87CN, 86LDb

Experimental conditions of papers selected for critical evaluation:

I = 0.1 mol L^{-1} $(CH_3)_4NNO_3$, 25 °C, Conc., gl: 92CDd, 91BCc, 82DSa
I = 0.1 mol L^{-1} $(CH_3)_4NCl$, 25 °C, Conc., gl: 01BCa, 00BCa, 98BFa, 96BCd, 96CHc, 95PMa, 94KCa
I = 0.1 mol L^{-1} $(CH_3)_4NCl$, 25 °C, Conc., sp: 94KCa
I = 0.1 mol L^{-1} KNO_3, 25 °C, Conc., gl: 82DSa
I = 0.1 mol L^{-1} KCl, 25 °C, Conc., gl: 94KCa

Table 6 Recommended and provisional data for DOTA, 25 °C.

Cation	Equilibrium	I/mol L^{-1}	lg K	Category	References
$H^{+\,a,b}$	L + H	0.1; $(CH_3)_4NNO_3$/Cl	11.9 (0.2)	P	00BCa, 94KCa, 95PMa, 92CDd, 82DSa
	HL + H	0.1; $(CH_3)_4NNO_3$/Cl	9.72 (0.03)	R	00BCa, 94KCa, 95PMa, 92CDd, 82DSa
	H_2L + H	0.1; $(CH_3)_4NNO_3$/Cl	4.60 (0.05)	R	00BCa, 96CHc, 94KCa, 95PMa, 92CDd, 82DSa
		0.1; KNO_3/Cl	4.5 (0.1)	P	94KCa, 82DSa
	H_3L + H	0.1; $(CH_3)_4NNO_3$/Cl	4.13 (0.03)	R	00BCa, 98BFa, 94KCa, 95PMa, 92CDd, 82DSa
		0.1; KNO_3/Cl	4.3 (0.1)	P	94KCa, 82DSa
	H_4L + H	0.1; $(CH_3)_4NCl$	2.36 (0.05)	R	00BCa, 98BFa, 95PMa
Na^+	M + L	0.1; $(CH_3)_4NNO_3$/Cl	4.2 (0.2)	P	00BCa, 82DSa
K^+	M + L	0.1; $(CH_3)_4NNO_3$	1.6 (0.1)	R1	82DSa
Mg^{2+}	M + L	0.1; $(CH_3)_4NNO_3$/Cl	11.85 (0.09)	R	96CHc, 82DSa
Ca^{2+}	M + L	0.1; $(CH_3)_4NNO_3$/Cl	17.2 (0.1)	R	00BCa, 82DSa
	ML + H	0.1; $(CH_3)_4NNO_3$/Cl	3.7 (0.1)	P	00BCa, 82DSa
Sr^{2+}	M + L	0.1; $(CH_3)_4NNO_3$/Cl	15.0 (0.2)	P	96CHc, 82DSa
Ba^{2+}	M + L	0.1; $(CH_3)_4NNO_3$	12. 9 (0.1)	R1	82DSa
$Mn^{2+\,e}$	M + L	0.1; $(CH_3)_4NNO_3$/Cl	20.0 (0.2)	P	01BCa, 92CDd
	ML + H	0.1; $(CH_3)_4NNO_3$/Cl	4.21 (0.06)	R	01BCa, 92CDd
$Co^{2+\,e}$	M + L	0.1; $(CH_3)_4NNO_3$	20.2 (0.1)	R1	92CDd, 82DSa
	ML + H	0.1; $(CH_3)_4NNO_3$	4.04 (0.05)	R1	92CDd, 82DSa
	MHL + H	0.1; $(CH_3)_4NNO_3$	3.5 (0.1)	P	92CDd, 82DSa

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Table 6 (*Continued*).

Cation	Equilibrium	I/mol L^{-1}	lg K	Category	References
$Ni^{2+\,e}$	M + L	0.1; $(CH_3)_4NNO_3$	20.0 (0.1)	R1	82DSa
$Cu^{2+\,e}$	M + L	0.1; $(CH_3)_4NNO_3$/Cl	22.3 (0.1)	R	00BCa, 92CDd, 82DSa
	ML + H	0.1; $(CH_3)_4NNO_3$/Cl	4.30 (0.09)	R	00BCa, 82DSa
	MHL + H	0.1; $(CH_3)_4NNO_3$/Cl	3.55 (0.09)	R	00BCa, 92CDd, 82DSa
$Zn^{2+\,e}$	M + L	0.1; $(CH_3)_4NNO_3$/Cl	20.8 (0.2)	P	00BCa, 82DSa
	ML + H	0.1; $(CH_3)_4NNO_3$/Cl	4.24 (0.08)	R	00BCa, 92CDd, 82DSa
	MHL + H	0.1; $(CH_3)_4NNO_3$/Cl	3.51 (0.04)	R	00BCa, 92CDd, 82DSa
Cd^{2+}	M + L	0.1; $(CH_3)_4NNO_3$	21.3 (0.1)	R1	92CDd
	ML + H	0.1; $(CH_3)_4NNO_3$	4.39 (0.04)	R1	92CDd
	MHL + H	0.1; $(CH_3)_4NNO_3$	3.03 (0.05)	R1	92CDd

[a] The first two protonation constant values selected for DOTA were determined in $(CH_3)_4NCl$ or $(CH_3)_4NNO_3$ as supporting electrolyte. Indeed, this ligand forms stable complexes with Na^+ and also with K^+, and media containing these cations lead to lower K(H + L) and K(HL + H) values.

Another problem is the high value for the first protonation constant of this ligand, which is difficult to determine by the usual potentiometric methods. NMR titration is the preferred technique in such cases but the determined values have not the required precision, because it is difficult: (i) to control the ionic strength; (ii) to prevent reaction with CO_2 at high pH values through the contact with the atmosphere, and (iii) to convert the constant determined in D_2O to that in H_2O; see [91DSa]. No other spectroscopic technique is reported.

The protonation constants were also determined in five different mixtures of DMSO/H_2O (from volume fraction 10 to 50 %), at 25 °C and in 0.1 mol L^{-1} in $(CH_3)_4NNO_3$. The values extrapolated for 100 % of H_2O were identical to those obtained by the same authors by potentiometry in 0.1 mol L^{-1} $(CH_3)_4NNO_3$ [92CDd].

Reported values range from lg K(L + H) = 9.37 (determined in 0.1 mol L^{-1} NaCl [94KCa]) to 12.6 (in 0.1 mol L^{-1} $(CH_3)_4NCl$ [98BFa]). Thus, only a provisional assignment is possible.

[b] The values of stability constants determined for metal complexes of DOTA are highly dependent on the values used for K(L + H) and K(HL + H). The spread of values found in the literature for the stability constants of metal complexes of DOTA is mainly because of the large range of K(L + H) values used by different authors. Those using Na^+ as supporting electrolyte always obtained the lower values. The same occurs in K^+ medium, but as the stability constant of the K^+-DOTA complex is much smaller, other experimental errors are usually more significant in determining the final value.

[c] The single communication for Li^+, lg K_{LiL} = 4.3 (25 °C, 0.1 mol L^{-1} $(CH_3)_4NNO_3$ [82DSa]) is considered reliable, but needs further independent verification.

[d] DOTA demonstrates a high affinity toward Be^{2+} [82DSa], although its selectivity within Ca^{2+}, Mg^{2+}, Be^{2+} is not as high as that for TETA. The value lg K_{BeL} = 13.6 (25 °C, 0.1 mol L^{-1} $(CH_3)_4NNO_3$) is considered reliable, but for critical evaluation an independent measurement is required.

[e] The macrocyclic framework is folded because of the small size of the cavity. The metal center is coordinated by the four nitrogen atoms and only by two carboxylate groups of the ligand. As the size of the metal ions is not a critical parameter in such structures, the ligand is quite unselective.

[f] The stability constant for Pb^{2+} is very high and is difficult to determine by direct potentiometry. Among the values found in the literature, two can be mentioned: one from direct potentiometry, lg K_{PbL} = 22.69 (0.03) [90CCa, 92CDd], and the other from spectrophotometry using competition with EDTA (lg K_{PbL} = 24.3 (0.2) [95PMa]). The first value can not be accurate because it is too high to be determined by a direct potentiometric measurement, while the second is presented with a very high standard deviation. Without other values, it is impossible to recommend one.

[g] For Y^{3+}, an average value of 24.6 (0.3) for lg K_{YL} is considered reliable (I = 0.1 mol L^{-1} $(CH_3)_4NNO_3$ or $(CH_3)_4NCl$, 25 °C [94KCa, 91BCc, 89CJ]). The first two publications are based on potentiometric methods and the last one on competition with Arsenazo III followed by spectrophotometric methods.

[h] Several research groups have determined stability constants for the lanthanide-DOTA complexes, but the spread of values is significant. However, for some of the lanthanides, an indicative value is possible:

- lg K_{ML} = 25.0 (0.3) for Ce^{3+} (I = 0.1 mol L^{-1} KCl, 25 °C, luminescence, competition with EDTA [97WHb] and I = 0.1 mol L^{-1} Me_4NCl, 25 ºC, gl, out-of-cell [98BFa]);
- lg K_{ML} = 25.0 (0.5) for Gd^{3+} (I = 0.1 mol L^{-1}, 25 °C, gl [91CA], I = 0.1 mol L^{-1} Me_4NNO_3, 25 °C, fluorescence [93KCa], I = 0.1 mol L^{-1} Me_4NCl, 25 °C, competition with Arsenazo III, sp [94KCa], I = 0.1 mol L^{-1} Me_4NCl, 25 °C, competition with EDTA, gl [96BCd, 00BCa]);

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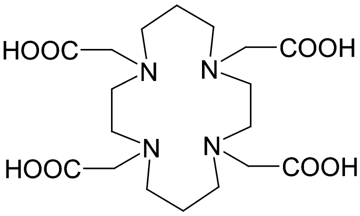
Table 6 (*Continued*).

- lg K_{ML} = 26.1 (0.3) for Yb^{3+} (I = 0.1 mol L^{-1} KCl, 25 °C, luminescence, competition with EDTA [97WHb], I = 0.1 mol L^{-1} Me_4NCl, 25 °C, gl, out-of-cell [98BFa]).

[i] Enthalpy and entropy changes for the protonation reactions of DOTA have been given in two publications [84DFa] and [00BCa], however, there are significant discrepancies between the reported values.
Enthalpy and entropy changes for the complexation reactions of DOTA with Co^{2+}, Ni^{2+}, Cu^{2+}, Zn^{2+}, and earth-alkaline metals (Ca^{2+} – Ba^{2+}) [84DFa], with Hg^{2+} [94KOb], and with Gd^{3+} [96BCd] have only been determined by one group.

11. 2,2',2'', 2'''-(1,4,8,11-TETRAAZACYCLOTETRADECANE-1,4,8,11-TETRAYL) TETRAACETIC ACID, TETA, H_4teta



Cations studied: **H^+** [a]: 92CDd, 91KKa, 91CMa, 84DFa, 82DSa, 81DMa, 81SFa, 76SFb

Na^+: 82DSa, 81DMa
Be^{2+} [b]: 82DSa
Mg^{2+}: 91CMa, 82DSa, 81SFa, 76SFb
Ca^{2+}: 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Sr^{2+}: 91KKa, 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Ba^{2+}: 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Al^{3+}: 95KKa
Ga^{3+} [d]: 91CMb
In^{3+} [d]: 91CMb
Pb^{2+}: 92CDd, 91KKa, 81SFa, 76SFb
Mn^{2+}: 92CDd, 81SFa
Fe^{2+}: 92CDd, 81SFa
Fe^{3+} [e]: 91CMb
Co^{2+}: 92CDd, 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Ni^{2+}: 92CDd, 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Cu^{2+}: 92CDd, 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Zn^{2+} [c]: 92CDd, 91CMa, 84DFa, 82DSa, 81SFa, 76SFb
Cd^{2+}: 92CDd, 81SFa, 76SFb
Hg^{2+}: 94KOb, 91KKa
Ag^+: 90KMc
Y^{3+}: 91KKa, 91BCc, 89CJ
La^{3+} [f]: 91KKa, 91CMb
Ce^{3+} [f]: 91KKa
Nd^{3+} [f]: 91KKa, 86LDb
Sm^{3+} [f]: 91KKa, 86LDb
Eu^{3+} [f]: 97WHb, 91KKa, 86LDb
Gd^{3+} [f]: 91KKa, 91CMb, 86LDb
Tb^{3+} [f]: 91KKa
Dy^{3+} [f]: 86LDb
Ho^{3+} [f]: 91KKa
Er^{3+} [f]: 86LDb
Tm^{3+} [f]: 91KKa
Yb^{3+} [f]: 86LDb
Lu^{3+} [f]: 91KKa

Experimental conditions of papers selected for critical evaluation:

I = 0.1 mol L^{-1} KNO_3, 25 °C, Conc., gl: 92CDd, 82DSa
I = 0.1 mol L^{-1} KCl, 25 °C, Conc., gl: 91CMa, 91CMb
I = 0.1 mol L^{-1} $(CH_3)_4NCl$, 25 °C, Conc., gl: 91CMa, 91CMb
I = 1.0 mol L^{-1} KCl, 25 °C, Conc., luminescence: 97WHb
I = 0.2 mol L^{-1} $NaNO_3$, 25 °C, Conc., gl: 91KKa; pol: 94KOb

Table 7 Recommended and provisional data for TETA, 25 °C.

Cation	Equilibrium	I/mol L^{-1}	lg K	Category	References
H^{+} [a]	L + H	0.1; $K/(CH_3)_4NCl$	10.7 (0.2)	P	92CDd, 91CMa, 82DSa
	HL + H	0.1; $K/(CH_3)_4NCl$	10.13 (0.03)	R	92CDd, 91CMa, 82DSa
	H_2L + H	0.1; $K/(CH_3)_4NCl$	4.10 (0.03)	R	92CDd, 91CMa, 82DSa
	H_3L + H	0.1; $K/(CH_3)_4NCl$	3.27 (0.08)	P	92CDd, 91CMa, 82DSa
Ca^{2+}	M + L	0.1; KNO_3/Cl	8.4 (0.1)	R	91CMa, 82DSa
	ML + H	0.1; KNO_3/Cl	7.2 (0.2)	P	91CMa, 82DSa
Sr^{2+}	M + L	0.1; KNO_3/Cl	5.82 (0.09)	R	91CMa, 82DSa
Ba^{2+}	M + L	0.1; KNO_3/Cl	4.1 (0.2)	P	91CMa, 82DSa
Mn^{2+}	M + L	0.1; KNO_3	11.3 (0.1)	R1	92CDd
Fe^{2+}	M + L	0.1; KNO_3	13.1 (0.1)	R1	92CDd
Co^{2+}	M + L	0.1; KNO_3/Cl	16.6 (0.1)	R	91CMa, 82DSa
	ML + H	0.1; KNO_3/Cl	4.2 (0.2)	P	92CDd, 91CMa, 82DSa
	MHL + H	0.1; KNO_3/Cl	2.84 (0.07)	R1	92CDd, 82DSa
Ni^{2+}	M + L	0.1; KNO_3/Cl	19.91 (0.07)	R	92CDd, 91CMa, 82DSa
	ML + H	0.1; KNO_3/Cl	4.2 (0.1)	R	92CDd, 91CMa, 82DSa
	MHL + H	0.1; KNO_3/Cl	3.2 (0.1)	R	92CDd, 91CMa, 82DSa
Cu^{2+}	M + L	0.1; KNO_3/Cl	21.7 (0.1)	P	91CMa, 82DSa
	ML + H	0.1; KNO_3/Cl	3.79 (0.09)	R	92CDd, 91CMa, 82DSa
	MHL + H	0.1; KNO_3/Cl	2.7 (0.2)	P	91CMa, 82DSa
Zn^{2+} [c]	ML + H	0.1; KNO_3/Cl	4.21 (0.08)	R	92CDd, 91CMa, 82DSa
Cd^{2+}	M + L	0.1; KNO_3	18.0 (0.1)	R1	92CDd
	ML + H	0.1; KNO_3	4.04 (0.01)	R1	92CDd
	MHL + H	0.1; KNO_3	2.93 (0.03)	R1	92CDd
Hg^{2+}	M + L	0.2 $NaNO_3$	25.7 (0.2)	P	94KOb, 91KKa
Pb^{2+}	M + L	0.1; KNO_3	14.3 (0.1)	R1	92CDd
	ML + H	0.1; KNO_3	4.75 (0.02)	R1	92CDd
	MHL + H	0.1; KNO_3	4.25 (0.03)	R1	92CDd
La^{3+} [f]	M + L	0.1; KCl	11.6 (0.1)	R1	91CMb
	ML + H	0.1; KCl	6.05 (0.01)	R1	91CMb
Eu^{3+} [f]	M + L	0.1; KCl	14.0 (0.1)	R1	97WHb
Gd^{3+} [f]	M + L	0.1; KCl	13.8 (0.1)	R1	91CMb
	ML + H	0.1; KCl	4.52 (0.05)	R1	91CMb

[a] Values of protonation constants (lg K) given at other temperatures and ionic strengths are accepted as provisional: (i) H + L 10.11 (0.06), HL + H 9.50 (0.02), H_2L + H 4.02 (0.02), H_3L + H 3.29 (0.04), H_4L + L 1.90 (0.15) ([81DMa], I = 1 mol L^{-1} NaCl, 80 °C, ^1H NMR in D_2O); (ii) H + L 10.84 (0.03), HL + H 9.50 (0.03), H_2L + H 4.20 (0.03), H_3L + H 3.07 (0.03) ([91KKa], I = 0.20 mol L^{-1} $NaNO_3$, 35 °C, potentiometry).

[b] TETA reveals a high affinity toward Be^{2+} [82DSa]. The data published for 25 °C and 0.1 mol L^{-1} KNO_3 (lg K_{ML} = 13.4; lg K(BeL + H) = 5.12) are considered reliable, but for critical evaluation an independent measurement is required. The Mg^{2+} complex presents a very low stability constant, but it is impossible to recommend a value owing to the large difference between the values reported [91CMa, 82DSa].

(*continues on next page*)

Table 7 (*Continued*).

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[c] The literature values for the Zn^{2+} complex (lg K_{ML}) vary widely as the kinetics of the reaction are slow. In [91CMa], the titration has been performed by a batch method and the reported value is taken as indicative only: 17.6 (0.3): I = 0.1 mol L^{-1} KCl, 25 °C, Conc., gl.

[d] Only one laboratory [91CMb] reports values for Ga^{3+} (lg K_{ML} = 19.74) and In^{3+} (lg K_{ML} = 21.89); both were derived by potentiometry. Despite the small standard deviation for Ga^{3+} (0.01; batch method) and the rather high value for In^{3+} these values are considered indicative.

[e] However, the value presented by the same authors for the Fe^{3+} complex (26.53 ± 0.01) [91CMb], obtained by competition with NTA is, in comparison with Ga^{3+} and In^{3+} complexes, too high to be accepted.

[f] Values determined at 80 °C and reported in [86LDb] for several lanthanides (I = 1 mol L^{-1} NaCl, 80 °C, Conc., gl) are accepted as indicative. For Nd^{3+}: 14.51 (M + L), 4.56 (ML + H); for Sm^{3+}: 14.97 (M + L), 3.90 (ML + H); for Eu^{3+}: 15.46 (M + L), 3.77 (ML + H); Gd^{3+}: 15.75 (M + L), 3.75 (ML + H); for Dy^{3+}: 16.04 (M + L), 3.10 (ML + H); for Er^{3+}: 16.49 (M + L), 3.50 (ML + H) and for Yb^{3+}: 16.55 (M + L), 2.44 (ML + H).

[g] Enthalpy and entropy changes for the protonation reactions of TETA and for its complexation reactions with Co^{2+}, Ni^{2+}, Cu^{2+}, Zn^{2+}, and earth-alkaline metals (Ca^{2+}–Ba^{2+}) [84DFa], with Hg^{2+} [94KOb], and with La^{3+}, Tb^{3+}, and Lu^{3+} [91KKa] were also determined, but each by a single group.

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