SYNTHESIS AND EXTRACTION PROPERTIES OF 1,2-BIS(AMIDOXIME) DERIVATIVES

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Four 1,2-bis(amidoxime)s bearing benzoate groups were synthesized. Their complexing properties were studied by the liquid–liquid extraction of selected alkali (Na^+ and K^+) and transition metals (Hg^{2+}, Cu^{2+}, Pb^{2+} and Ni^{2+}). It has been observed that all ligands show a high affinity to Cu^{2+} and Hg^{2+} ions, whereas almost no affinity to alkali metals.

Keywords: Extractions; Oximes; Amidoximes; Dioximes; Aminobenzoates; Alkali cations; Transition metal cations; Chelates.

1,2-Dioximes have received considerable attention as model compounds for mimicking biofunctions such as reduction of vitamin B_{12} (refs[1,2]). Some oxime metal chelates are biologically active[3] and are reported to possess semiconductive properties[4,5]. The complexation studies of 1,2-oximes were initiated by investigating the reaction of dimethylglyoxime with Ni(II) by Tschugaeff[6] in 1907. Their complexes have been the source, through the decades, of a never-ending series of interesting reports.

The process of solvent extraction is one of the most versatile procedures used for the removal, separation and concentration of metallic species. Its applications expand into the recycling of resources in the field of metallurgy and wastewater treatment as demands increase for the development of new approaches to resolve various problems. For this purpose, many oxime derivatives have been synthesized and their extraction properties investigated by solvent extraction.

Paping *et al.* studied the extraction of copper(II) and nickel(II) from aqueous solution by using camphorquinone[7] and nopinonequinone[8] dioximes, cyclic trioximes, 2,1,3-benzooxadiazole oximes[9] and long-chain aliphatic

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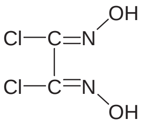
dioximes[10]. They found that the use of camphorquinone and nopinoquinone dioximes instead of aliphatic dioximes makes the extraction more convenient due to higher extraction rates, but the selectivity to nickel over copper disappears. They also showed that the use of trioximes for the extraction of copper(II) and nickel(II) is not an improvement compared with the dioximes. In the search for a selective nickel extraction, Paping and co-workers elaborated the extraction of copper and nickel ions from ammonia and acid solutions with symmetric dialkyl dioximes. Extraction from an ammonia solution showed great differences between copper and nickel.

Srinivasa Rao *et al.*[11] studied the separation and recovery of copper from solution which were 0.1 M in both copper and sodium sulfate using the new hydroxyoxime reagent under the trade name MOC 45 dissolved in kerosene. An increase in equilibrium pH and extractant concentration increases the percentage extraction of metal ions. Calligaro *et al.*[12] studied the extraction of copper, nickel, cobalt, zinc and iron from solutions using LIX-64N (a mixture of LIX-63 and LIX-65N) in kerosene.

The order of extractability as a function of pH was Cu(II) < Fe(III) < Ni(II) < Zn(II) < Co(II). Sastre and Alguacil[13] showed that LIX-622 extracts both copper and molybdenum from aqueous sulfate media. The extraction of both metals is pH-dependent; however, molybdenum is extracted preferably to copper as the equilibrium pH shifts to lower values.



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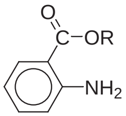

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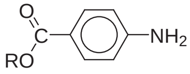
2b, R = CH_2CH_3



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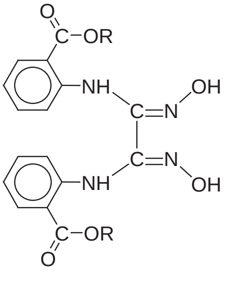
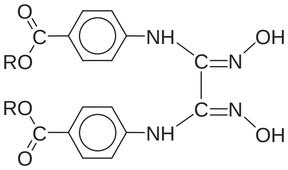


4, R = CH_3
5, R = CH_2CH_3



6, R = CH_3
7, R = CH_2CH_3

In this paper we report the synthesis of four new compounds bearing methyl or ethyl 2- and 4-aminobenzoate moieties and possible utilization of these compounds in metal ion extractions from aqueous solutions into organic solvents.

EXPERIMENTAL

Ethyl 2-aminobenzoate (Fluka), methyl 2-aminobenzoate, methyl 4-aminobenzoate, ethyl 4-aminobenzoate, hydroxylamine hydrochloride, chloral hydrate, benzene, absolute ethanol were purchased from (Merck) and used without further purification. All aqueous solutions were prepared with deionized water, which had been passed through a Millipore Milli-Q Plus water purification system.

Melting points were determined on a Büchi SMP-20 apparatus. ^{1}H NMR spectra were recorded on a Bruker 250 MHz spectrometer in $CDCl_3$. Chemical shifts are given in ppm (δ-scale), coupling constants (*J*) in Hz. IR spectra (ν in cm^{-1}) were recorded on a Perkin–Elmer 1605 FTIR spectrometer in KBr pellets. UV-VIS spectra were obtained on a Shimadzu 160 recording spectrophotometer.

Compound **1** was synthesized as described previously[14,15]. The other ligands (**4**, **5**, **6** and **7**) are synthesized as follows.

Dimethyl 2,2′-{[bis(hydroxyimino)ethylene]diimino}dibenzoate (**4**) and
Diethyl 2,2′-{[bis(hydroxyimino)ethylene]diimino}dibenzoate (**5**)

A solution of methyl (**2a**) (3.02 g, 0.02 mol) or ethyl 2-aminobenzoate (**2b**) (3.3 g, 0.02 mol) in absolute ethanol (20 ml) was added to a mixture of (*E*,*E*)-dichloroglyoxime (**1**) (1.57 g, 0.01 mol) and $NaHCO_3$ (3.00 g) in absolute ethanol (20 ml). The mixture was stirred efficiently at room temperature for 7 h. The mixture was filtered, the solid was washed with absolute ethanol, and the filtrate and washings were combined. The solution was then poured into 150 ml of cold distilled water with stirring. The precipitated solid was filtered off, washed thoroughly with distilled water, and dried under vacuum. Recrystallization of the crude product from benzene–ethanol furnished compounds **4** and **5**.

Compound **4**: yield 2.20 g (56%), m.p. 192–193 °C. IR (KBr): 3 408 (N–H), 3 251 (O–H), 2 975–2 906 (C–H), 1 728 (C=O), 1 618 (C=N), 974 (N–O). ^{1}H NMR ($CDCl_3$): 3.37 s, 6 H (OCH_3); 6.83–7.57 m, 8 H (Ar-H); 9.80 s, 2 H (NH); 11.11 s, 2 H (OH). For $C_{18}H_{18}N_4O_6$ (386) calculated: 55.96% C, 4.66% H, 14.51% N; found: 55.59% C, 4.79% H, 14.98% N.

Compound **5**: yield 2.07 g (50%), m.p. 187 °C. IR (KBr): 3 420 (N–H), 3 250 (O–H), 2 982–2 826 (C–H), 1 710 (C=O), 1 619 (C=N), 968 (N–O). ^{1}H NMR ($CDCl_3$): 1.25 t, 6 H, *J* = 7 (CH_3); 3.73 q, 4 H, *J* = 7 (O-CH_2-C); 6.79–7.52 m, 8 H (Ar-H); 7.60 s, 2 H (NH); 9.92 s, 2 H (OH). For $C_{20}H_{22}N_4O_6$ (414) calculated: 57.91% C, 5.31% H, 13.53% N; found: 58.22% C, 5.54% H, 13.75% N.

Dimethyl 4,4′-{[bis(hydroxyimino)ethylene]diimino}dibenzoate (**6**) and
Diethyl 4,4′-{[bis(hydroxyimino)ethylene]diimino}dibenzoate (**7**)

A solution of methyl (**3a**) (3.02 g, 0.02 mol) or ethyl 4-aminobenzoate (**3b**) (3.3 g, 0.02 mol) in absolute ethanol (40 ml) was added dropwise in the course of 2 h to solution of (*E*,*E*)-dichloroglyoxime (**1**) (1.57 g, 0.01 mol) in absolute ethanol (20 ml). The mixture was stirred efficiently at room temperature. The mixture was stirred efficiently for an additional 3 h, and left overnight. The mixture was cooled to 0 °C and a solution of $NaHCO_3$ (1.68 g, 0.02 mol) in a small amount of distilled water were added dropwise with stirring. The solution was then poured into 200 ml of cold water. The precipitated solid was filtered off, washed thoroughly with distilled water, and dried under vacuum. Recrystallization of the crude product from ethanol–water furnished compounds 6 or 7.

Compound **6**: yield 2.74 g (71%), m.p. 176 °C. IR (KBr): 3 422 (N–H), 3 229 (O–H), 2 960–2 975 (C–H), 1 718 (C=O), 1 614 (C=N), 966 (N–O). ^{1}H NMR ($CDCl_3$); 3.75 s, 6 H (OCH_3); 6.77–7.78 dd, 8 H (Ar-H); 8.06 s, 2 H (NH); 10.72 s, 2 H (OH). For $C_{18}H_{18}N_4O_6$ (386) calculated: 55.96% C, 4.66% H, 14.51% N; found: 56.30% C, 4.54% H, 14.38% N.

Compound **7**: yield 3.10 g (75%), m.p. 162–163 °C. IR (KBr): 3 413 (N–H), 3 230 (O–H), 2 927–2 885 (C–H), 1 718 (C=O), 1 625 (C=N), 966 (N–O). ^{1}H NMR ($CDCl_3$): 1.29 t, 6 H, J = 7.1 (CH_3); 4.22 q, 4 H, J = 7.1 (O-CH_2-C); 6.78–7.62 dd, 8 H (Ar-H); 8.08 s, 2 H (NH); 10.68 s, 2 H (OH). For $C_{20}H_{22}N_4O_6$ (414) calculated: 57.91% C, 5.31% H, 13.53% N; found: 57.45% C, 5.42% H, 13.82% N.

Solvent Extraction

Picrate extraction experiments were performed following Pedersen's procedure[16]. Ten ml of a $2.5 \cdot 10^{-5}$ M aqueous picrate solution and 10 ml of a $1 \cdot 10^{-3}$ M solution of ligand in CH_2Cl_2 were vigorously agitated for 2 min in a stoppered glass tube using a mechanical shaker, then magnetically stirred in a thermostated water-bath at 25 °C for 1 h, and finally left standing for additional 30 min. The concentration of the picrate ion remaining in the aqueous phase was then determined spectrophotometrically as previously described[17]. Blank experiments showed that no picrate extraction occurred in the absence of ligand.

The alkali picrates were prepared as described elsewhere[17,18] by successive addition of a $2.0 \cdot 10^{-2}$ M aqueous picric acid solution to an 0.14 M aqueous solution of alkali metal hydroxide until neutralization, which was checked with a glass electrode. They were then rapidly washed with ethanol and ether and dried *in vacuo* for 24 h. Transition metal picrates were prepared by successive addition of a $1 \cdot 10^{-2}$ M metal nitrate solution to a $2.5 \cdot 10^{-5}$ M aqueous picric acid solution and shaken at 25 °C for 1 h.

Log–Log Plot Analysis

To characterize the extraction ability, the dependence of the distribution coefficient D of the cation between the two phases upon the ligand concentration was examined. If the general extraction equilibrium is assumed to be given by Eq. (*1*)

$$M^{n+}_{(aq)} + n\,\mathrm{pic}^-_{(aq)} + x[L]_{(org)} \rightleftharpoons [M(L)^{n+}_x, (\mathrm{pic})^{n-}_n]_{(org)} , \qquad (1)$$

the overall extraction equilibrium constant is

$$K_{ex} = \frac{[M(L)^{n+}_x, (\mathrm{pic})^{n-}_n]_{(org)}}{[M^{n+}][\mathrm{pic}^-]^n\,[L]^x} \qquad (2)$$

and the distribution ratio D is defined by

$$D = \frac{[\mathrm{M(L)}_x^{n+}, (\mathrm{pic})_n^{n-}]_{(\mathrm{org})}}{[\mathrm{M}^{n+}]} \,. \qquad (3)$$

By introducing D into Eq. (*2*) and taking logarithm of both sides, Eq. (*4*) is obtained.

$$\log D = \log (K_{\mathrm{ex}} \, [\mathrm{pic}^-]^n) + x \log [\mathrm{L}] \qquad (4)$$

Under these assumptions, a plot of log D *versus* log [L] should be linear and its slope should be equal to the number of ligand molecules per metal cation in the extracted species.

RESULTS AND DISCUSSION

Four new 1,2-bis(amidoxime) compounds bearing methyl or ethyl 2- or 4-aminobenzoate moieties, dimethyl 2,2′-{[bis(hydroxyimino)ethylene]diimino}dibenzoate (**4**), diethyl 2,2′-{[bis(hydroxyimino)ethylene]diimino}dibenzoate (**5**), dimethyl 4,4′-{[bis(hydroxyimino)ethylene]diimino}dibenzoate (**6**) and diethyl 4,4′-{[bis(hydroxyimino)ethylene]diimino}dibenzoate (**7**) were prepared by one-step reaction of (*E*,*E*)-dichloroglyoxime (**1**) with methyl (**2a**) or ethyl 2-aminobenzoate (**2b**) and methyl (**3a**) or ethyl 4-aminobenzoate (**3b**) in absolute ethanol with excess of solid $NaHCO_3$ to neutralize HCl formed in the reaction. These compounds are soluble in common organic solvents such as methanol, chloroform, dichloromethane and hexane. All new compounds were characterized by a combination of ^{1}H NMR, IR and elemental analysis given in Experimental.

In the IR spectrum of the new compounds, an O–H stretching vibration is observed at 3 251 cm^{-1} for **4**, 3 250 cm^{-1} for **5**, 3 229 cm^{-1} for **6** and 3 230 cm^{-1} for **7** as a broad absorption. The other characteristic bands of bis(amidoxime)s C=N and N–O are at 1 618 and 974 cm^{-1} for **4**, 1 619 and 968 cm^{-1} for **5**, 1 614 and 966 cm^{-1} for **6**, 1 625 and 966 for **7**. The C=O vibrations of aminobenzoates are shown at 1 728 cm^{-1} for **4**, 1 710 cm^{-1} for **5**, 1 718 cm^{-1} for **6** and 1 718 cm^{-1} for **7**. These values are in accord with the previously reported bis(amidoxime) derivatives[19–24]. Furthermore, CH stretching vibrations are at 2 982–2 875 cm^{-1}.

In the ^{1}H NMR spectra of compounds, the OH and NH resonances appear as two singlets at δ 11.11 and 9.80 for **4**, 9.92 and 7.60 for **5**, 10.72 and 8.06 for **6**, 10.68 and 8.08 for **7**. A single chemical shift for OH protons indicates that, due to symmetry, the oxime groups are in the *anti* form[20,22,23].

As expected for the *para*-disubstituted benzene, the aromatic protons of compounds **6** and **7** give doublet-doublet peaks, which resemble an AB pattern of two distorted doublets, whereas the aromatic protons of compounds **4** and **5**, *ortho*-disubstituted, give multiplet signals. The ^{1}H NMR spectra of these compounds exhibit singlet for OCH_3 protons; δ 3.37 for **4**, 3.75 for **6**, quartets for OCH_2-C protons; 3.73 (J = 7) for **5**, 4.22 (J = 7.1) for **7**, triplets for C-CH_3 protons; 1.25 (J = 7) for **5**, 1.29 (J = 7.1) for **7**. These data are in agreement with previously reported for similar molecules[22–24].

1,2-Bis(amidoxime)s have been widely employed in the formation of transition metal complexes and in the study of inclusion phenomena due to their relatively easy preparation, remarkable stability and high versatility[20–24,27]. As yet, reports on solvent extraction with complexes of amidoxime compounds are scarce. Therefore, this work focused on elaboration of strategic requirements for the two-phase extraction measurements. For this purpose, solvent extraction experiments were performed to estimate the effectiveness of **4**–**7** in transferring alkali metals Na^+ and K^+ and transition metals Ni^{2+}, Cu^{2+}, Pb^{2+} and Hg^{2+} from the aqueous into organic phase. The results are summarized in Table I. These data were obtained using dichloromethane solutions of ligands (**4**–**7**) to extract metal picrates from aqueous solution according to Pedersen's procedure[16]. The equilibrium concentration of picrate in aqueous phase was then determined spectrophotometrically.

From the extraction data shown in Table I, it was deduced that the alkali metal cations were not significantly extracted by ligands **4**–**7**. However, all the compounds extracted transition metals, in particular Cu^{2+} and Hg^{2+}.

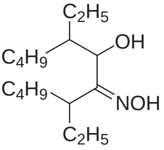
TABLE I
Extraction of metal picrates with the synthesized compounds[a]

Compound	Picrate salt extracted, %					
	Na^+	K^+	Ni^{2+}	Cu^{2+}	Hg^{2+}	Pb^{2+}
4	2.0	<1	18.0	96.1	94.7	13.0
5	3.0	2.0	26.0	96.8	93.6	35.0
6	2.2	<1	16.0	97.0	95.2	16.1
7	3.1	2.2	27.1	97.2	94.3	32.3

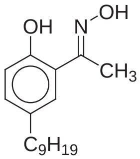
[a] Aqueous phase; [metal nitrate] = $1 \cdot 10^{-2}$ M; [picric acid] = $2.5 \cdot 10^{-5}$ M; organic phase; dichloromethane [ligand] = $1 \cdot 10^{-3}$ M; at 25 °C for 1 h.

The effectiveness in transferring transition metals rather than the other metal cations by the synthesized compounds indicates that a cation–π electron interaction is operative since the metal is bounded by the opposite N, N or N, O sites of these compounds. Moreover, in the case of **4** and **5**, the C=O group may participate in the complexation due to steric arrangement. Memon and co-workers[26] and Seangprasertkij and co-workers[28] who studied Schiff base derivatives of calix[4]arene obtained similar results. As compared in this work, the extraction data of **4**–**7** show a high increase in the extraction ability to Cu^{2+} and Hg^{2+}. The presence in the ligands of soft donor nitrogen groups, which show high affinity to transition metals, causes the increase in the extraction ability of these ligands[7–13,25,26,28].

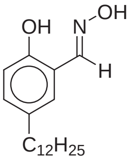
Despite the great affinity of nickel to amidoximes, surprisingly, after a contact time of 60 min, the extraction percentage of nickel is very low,



LIX-63



MOC 45



LIX-622

$$H_3C-\overset{\overset{HON}{\|}}{C}-\overset{\overset{NOH}{\|}}{C}-CH_3$$

8

$$C_7H_{15}-\overset{\overset{HON}{\|}}{C}-\overset{\overset{NOH}{\|}}{C}-C_7H_{15}$$

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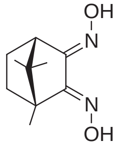

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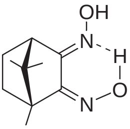

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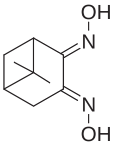

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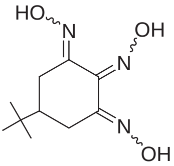

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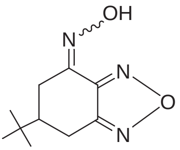

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but that of copper is very high. Dioxime materials usually form stable *N*,*N*- or *N*,*O*-chelated complexes[30] containing a conjugated –N=C–C=N– or –N=C–C=N-O– system, respectively. The substitution pattern of the bis(amidoxime) moiety affects the structure, stability and solubility of the complexes[7–12,29].

The high extraction percentage of mercury(II) is another surprising phenomenon in this work. The ionic radius of Hg^{2+}, which is 1.1 Å, cannot explain alone this phenomenon because Pb^{2+}, which has a similar ionic radius to Hg^{2+}, could not be extracted as much as Hg^{2+} by all ligands. In other words, the extraction behaviors of these compounds to Hg^{2+} and Pb^{2+} ions are very different. The type of the donor atoms, polarizability effects, steric hindrance and cavity size are probably important factors here.

Comparing the copper(II) extraction efficiencies of these new compounds with that of some commercial copper(II) extractants after a contact time of 1 h, it is seen that their efficiencies are higher than that of the industrial extractant LIX-63 and LIX-622 and just slightly lower than that of MOC-45 as verified by the transport and solvent extraction experiments carried out with these compounds. The copper extraction percentages of some industrial dioxime extractants, which are dimethylglyoxime (**8**), diheptylglyoxime (**9**), *E,E*-camphorquinone dioxime (**10**), *E,Z*-camphorquinone dioxime (**11**), *E,E*-nopinonequinone dioxime (**12**), 5-*tert*-butylcyclohexane-1,2,3-trione trioxime (**13**), 6-*tert*-butyltetrahydro-2,1,3-benzoxadioazol-4-one (**14**, derived from **13**), and the synthesized ligands are given in Table II.

TABLE II
The copper extraction percentages of some oxime compounds

Industrial extractans		Dioximes		Dioximes	
Name	% Cu extracted	Name	% Cu extracted	Name	% Cu extracted
MOC 45	>98	**8**	<75	**4**	96.1
LIX-622	96	**9**	<80	**5**	96.8
LIX-63	<55	**10**	<60	**6**	97
		11	<70	**7**	97.2
		12	<50		
		13	99.4		
		14	poor		

Figure 1 shows the extraction of Hg^{2+} and Cu^{2+} into dichloromethane at different concentrations of **4** and **5**. For the extraction of Hg^{2+} and Cu^{2+} with these ligands, the results are not linear. The observed metal–ligand ratios of Hg^{2+} and Cu^{2+} with **4** and **5** are 1 : 2.

For the systems the following logarithmic extraction constants corresponding to Eq. (*1*) were determined.

$\log K_{ex} = 12.38 \pm 3$ (for Hg^{2+} with **4**)
$\log K_{ex} = 12.05 \pm 4$ (for Hg^{2+} with **5**)
$\log K_{ex} = 12.67 \pm 3$ (for Cu^{2+} with **4**)

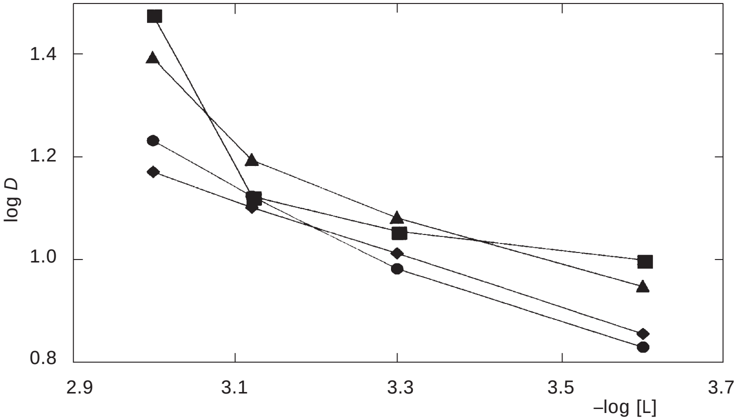

FIG. 1
log *D versus* log [L] for the extraction of Hg picrate with **4** (●) and **5** (◆) and Cu picrate with **4** (▲) and **5** (■) from an aqueous phase into dichloromethane at 25 °C

CONCLUSION

In the present study, four new aminobenzoate derivatives of symmetric bis(amidoxime)s have been synthesized. Liquid–liquid extractions of some alkali and transition metal ions with these compounds have been examined. All the compounds, in particular **4** and **6**, are excellent extractants for copper(II) and mercury(II) and can be used for copper and mercury recovery.

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