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Synthesis and Properties of *N*-Hydroxy-*N*-naphthylbenzamides

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Abstract

N-Hydroxy-*N*-naphthylbenzamide derivatives (**3, 5**) were prepared from nitronaphthalenes by reduction with raney Ni-hydrazine and benzoylation. Compounds (**3, 5**) were found to display unique absorption spectral changes in the presence of Cu^{2+} and Fe^{3+} and exhibited high Cu^{2+} selectivity. Compounds (**3, 5**) were effective for Cu^{2+} transport through a liquid membrane.

1. Introduction

Hydroxamic acids, a group of weak organic acids, have wide applications as antifungal agent, food additives, inhibitors for copper corrosion in metallurgy and in nuclear fuel processing. The isolation of *N*-hydroxyoxamic acid from the reaction products of diethyloxalate and hydroxylamine attracted much attention to the chemistry of hydroxamic acids.[1,2] The complexation of hydroxamic acids with metal ions constitutes the basis of many analytical determinations. A beautiful purple color of the Fe^{3+} and Cu^{2+} complexes enabled sensitive qualitative and quantitative determinations of carboxylic acids and their derivatives. They form stable transition-metal complexes and are used as analytical reagents.[3-7] Considerable attention has been devoted to the effective separation and recovery of heavy metal ions. In this respect, Bacon has reported transport of heavy metal ions such as Hg^{2+} and Pb^{2+} through a liquid membrane.[8,9] Liquid membrane methods are useful for assessing the partitioning of metal into and out of organic phases and are of considerable importance in medicine, water purificaton, and metallurgy. A liquid membrane, (which consists of an organic solvent placed at the bottom of a U-tube), has been widely used in order to study ion transport from one water compartment to the other, a process requiring, of course, passage through the chloroform barrier.[10,11] We have also been reported Hg^{2+} transport with troponoid dithiocrown ethers through a liquid membrane.[12-15] A few studies regarding metal ion transport with hydroxamic acid derivatives[5,6,16] such as *N*-hydroxy-*N*-

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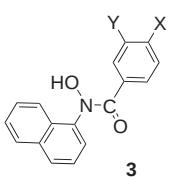
phenylbenzamide (BPA), *N*-hydroxy-*N*-phenylcinnamamide and *N*-hydroxy-*N*-naphthylbenzamide through liquid membranes have been reported. In this paper, we will report the synthesis and properties of *N*-hydroxy-*N*-naphthylbenzamides that can use as an analytical reagent and transporting agent of Cu^{2+} through a liquid membrane.



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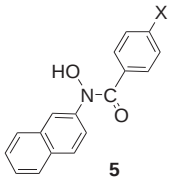
a (X=OCH_3, Y=H)
b (X=CH_3, Y=H)
c (X=H, Y=H)
d (X=F, Y=H)
e (X=Cl, Y=H)
f (X=H, Y=Cl)
g (X=CF_3, Y=H)
h (X=CN, Y=H)



5

Fig. 1. Chemical structures of **3** and **5**.

2. Experimental

Elemental analyses were performed by Perkin Elmer PE2400 series II CHNS/O analyzer. The melting points were obtained with a Yanagimoto Micro Melting Point Apparatus and were uncorrected. The NMR spectra were measured on a JEOL JNM-500 Model spectrometer in $CDCl_3$; the chemical shifts are expressed by an δ unit using tetramethylsilane as an internal standard. The IR spectra were recorded on a Hitachi Model 270-30 infrared spectrometer with KBr disks for crystalline compounds. The UV spectra were measured using a Shimadzu Model UV-2200 spectrophotometer.

2.1. Synthesis of *N*-Hydroxy-*N*-naphthalenyl-benzamides

Nitronaphthalene (0.02 mol, 5.18 g) was dissolved in ethanol/1,2-dichloroethane (1 : 1 *v*/*v*, 50 cm^3) and was cooled to 0 ºC. Raney nickel (type W-4; 1.0 g) was added by stirring into the solution. Then, 80 % hydrazine hydrate solution (2.0 cm^3, 0.064 mol) was added drop by drop, taking care to maintain the reaction temperature below 10 ºC. The complete (or nearly complete) nitronaphthalene conversion was confirmed by TLC analysis on silica gel using benzene as an eluent. The catalyst was removed by filtration. The solvent was evaporated and the residue was recrystallized from benzene to yield *N*-naphthylhydroxylamines. The residue of the *N*-arylhydroxylamine (which was obtained by the evaporation of solvents in the above procedure) was dissolved in benzene (40 cm^3) and was washed with ice-cold water (20 cm^3). This mixture was added with stirring into a previously cooled solution (0 ºC) of 5% sodium hydrogencarbonate in water (60 cm^3). The corresponding benzoyl chloride (0.03 mol) in benzene (30 cm^3) was added drop by drop to the well-stirred mixture in about 0.5 h, taking care to maintain the temparature of the reaction below 5 ºC. The mixture was stirred for 2h at 0-5 ºC. The benzene layer was separated and the aqueous layer was extracted with benzene (30 cm^3). The combined benzene extract was washed with water (40 cm^3) and then with 10% sodium hy-

droxide solution (150 cm^3). The aqueous alkaline solution was separated and acidified with a 10% hydrochloric acid solution when the *N*-Hydroxy-*N*-naphthylbenzamides were separated from the solution. The obtained crystals were purified by column chromatography over silica gel (70-230 mesh, Merck) using hexane and benzene as the eluent. Recrystallization from ethyl acetate or ethanol gave analytically pure samples (**3, 5**) with the following physical properties. The physical properties of *N*-hydroxy-*N*-1-naphthylbenzamide (**3c**) and *N*-hydroxy-*N*-2-naphthylbenzamide (**5c**) have been reported in our previous paper.[5]

N-Hydroxy-4-methoxy-*N*-1-naphthalenylbenzamide (**3a**)[17] : Colorless crystals. ^{1}H NMR ($CDCl_3$) δ= 3.70 (3H, s), 6.62 (2H, dt, J=9.8, 2.5 Hz), 7.24 (1H, dd, J=7.3, 1.2 Hz), 7.33-7.36 (3H, m), 7.58 (1H, dd, J=8.2, 7.0 Hz), 7.64 (1H, dd, J=8.2, 7.0 Hz), 7.87 (1H, d, J=8.2 Hz), 8.22 (1H, d, J=8.5 Hz), and 9.32 (1H, brs). ^{13}C NMR ($CDCl_3$) δ=55.2, 113.4, 123.3, 123.5, 125.3, 127.0, 127.3, 127.8, 128.3, 130.2, 130.5, 134.5, 136.5, 161.8, 167.0. IR (KBr) ν 612, 666, 753, 774, 915, 975, 1029, 1110, 1155, 1176, 1257, 1305, 1404, 1461, 1509, 1575, 1629, 2896, 3052, 3480 cm^{-1}. Found : C, 73.47 ; H, 5.28 ; N, 4.48%. Calcd for $C_{18}H_{15}NO_3$: C, 73.71 ; H, 5.15 ; N, 4.78%.

N-Hydroxy-4-methyl-*N*-1-naphthalenylbenzamide (**3b**)[17] : Colorless crystals, mp 149-151ºC. ^{1}H NMR ($CDCl_3$) δ=2.22 (3H, s), 6.92 (2H, d, J=8.0 Hz), 7.24 (1H, d, J=7.3 Hz), 7.27 (2H, d, J=8.0 Hz), 7.33 (1H, dd, J=8.2, 7.3 Hz), 7.58 (1H, dd, J=8.2, 7.0 Hz), 7.65 (1H, dd, J=8.2, 7.0 Hz), 7.87 (1H, d, J= 8.2 Hz), 7.90 (1H, d, J=8.0 Hz), and 9.32 (1H, brs). ^{13}C NMR ($CDCl_3$) δ=21.4, 123.3, 125.3, 127.0, 127.4, 127.8, 128.3, 128.5, 128.8, 130.3, 134.5, 136.1, 141.7, 167.1. IR (KBr) ν 543, 609, 666, 738, 777, 807, 831, 912, 972, 1416, 1506, 1572, 1632, 2878, 3094, 3448 cm^{-1}. Found : C, 77.97 ; H, 5.60 ; N, 4.81%. Calcd for $C_{18}H_{15}NO_2$: C, 77.96 ; H, 5.45 ; N, 5.05%.

4-Fluoro-*N*-hydroxy-*N*-1-naphthalenylbenzamides (**3d**) : Colorless crystals, mp 134.0-135.5ºC. ^{1}H NMR ($CDCl_3$) δ=6.81 (2H, t, J=8.7 Hz), 7.24 (1H, d, J=7.4 Hz), 7.35 (1H, t, J=8.2 Hz), 7.39 (2H, t, J =8.7 Hz), 7.59 (1H, dd, J=8.2, 6.8 Hz), 7.65 (1H, dd, J=8.2, 6.8 Hz), 7.89 (1H, d, J=8.2 Hz), 7.91 (1 H, d, J=8.2 Hz), 8.19 (1H, d, J=8.2 Hz), and 9.26 (1H, bs). ^{13}C NMR ($CDCl_3$) δ=115.2, 115.4, 123.1, 125.3, 127.1, 127.5, 128.0, 128.5, 130.4, 130.6. 130.8, 130.9, 134.5, 163.2, 165.2. IR (KBr) ν 609, 771, 849, 915, 972, 1155, 1227, 1401, 1416, 1509, 1605, 1632, 2878, 3052, 3448 cm^{-1}. Found : C, 72.97 ; H, 4.66 ; N, 4.91%. Calcd for $C_{17}H_{12}NO_2F$: C, 72.59 ; H, 4.30 ; N, 4.98%.

4-Chloro-*N*-hydroxy-*N*-1-naphthalenylbenzamide (**3e**) : Colorless crystals, mp 142.5-144.5ºC (*lit.* 148 ºC)[18,19]. ^{1}H NMR ($CDCl_3$) δ=7.10 (2H, d, J=8.7 Hz), 7.23 (1H, d, J=7.0 Hz), 7.30 (2H, d, J=8.7 Hz),

7.34 (1H, dd, J=8.2, 7.0 Hz), 7.59 (1H, dd, J=8.2, 7.0 Hz), 7.65 (1H, dd, J=8.2, 7.0 Hz), 7.89 (1H, d, J=8.2 Hz), 7.91 (1H, d, J=8.2 Hz), 8.19 (1H, d, J=8.2 Hz), and 9.31 (1H, bs). ^{13}C NMR ($CDCl_3$) δ= 123.1, 125.3, 127.1, 127.5, 128.0, 128.4, 128.5, 129.8, 130.0, 130.4, 130.7, 134.5, 135.6, 137.4, 165.9. IR (KBr) ν 558, 663, 744, 774, 804, 840, 912, 972, 1017, 1092, 1416, 1491, 1569,1629, 2878, 3112, 3586 cm^{-1}. Found : C, 68.86 ; H, 4.25 ; N, 4.84%. Calcd for $C_{17}H_{12}NO_2Cl$: C, 68.58 ; H, 4.06 ; N, 4.70%.

3-Chloro-*N*-hydroxy-*N*-1-naphthalenylbenzamide (**3f**) : Colorless crystals, mp 147.0-149.0ºC. ^{1}H NMR ($CDCl_3$) δ=6.98 (1H, dd, J=8.2, 7.5 Hz), 7.08 (1H, d, J=7.0 Hz), 7.23 (1H, d, J=8.2 Hz), 7.26 (1H, d, J=8.2 Hz), 7.35 (1H, dd, J=8.2, 7.5 Hz), 7.51 (1H, s), 7.59 (1H, dd, J=8.2, 7.0 Hz), 7.65 (1H, dd, J=8.2, 7.0 Hz), 7.89 (1H, d, J=8.2 Hz), 7.91 (1H, d, J=8.8 Hz), 8.18 (1H, d, J=8.8 Hz), and 9.45 (1H, bs). ^{13}C NMR ($CDCl_3$) δ=123.0, 125.2, 126.3, 127.0, 127.5, 128.0, 128.5, 128.7, 129.3, 130.0, 130.8, 131.2, 133.4, 134.3, 134.5, 135.3, 165.5. IR (KBr) ν 723, 777, 792, 810, 879, 927, 975, 1161, 1404, 1572, 1593, 1635, 2902, 3154, 3448 cm-1. Found : C, 68.71 ; H, 4.30 ; N, 4.75%. Calcd for $C_{17}H_{12}NO_2Cl$: C, 68.58 ; H, 4.06 ; N, 4.70%.

N-hydroxy-*N*-1-naphthalenyl-4-trifluoromethylbenzamide (**3g**) : Colorless crystals, mp 159.0-160.0ºC. ^{1}H NMR ($CDCl_3$) δ=7.24 (1H, d, J=7.0 Hz), 7.35 (1H, dd, J=8.2, 7.0 Hz), 7.39 (2H, d, J=8.2 Hz), 7.45 (2H, d, J=8.2 Hz), 7.60 (1H, dd, J=8.2, 7.0 Hz), 7.68 (1H, dd, J=8.2, 7.0 Hz), 7.90 (1H, d, J=8.5 Hz), 7.92 (1H, d, J=8.5 Hz), 8.20 (1H, d, J=8.2 Hz), and 9.27 (1H, bs). ^{13}C NMR ($CDCl_3$) δ=122.3, 122.9, 124.5, 125.1 (q, J=4Hz), 125.3, 127.2, 127.6, 128.1, 128.5, 128.8, 130.4, 130.9, 132.7 (q, J=33 Hz), 134.5, 135.1, 165.4. IR (KBr) ν 561, 693, 774, 804, 855, 876, 975, 1017, 1068, 1116, 1170, 1335, 1425, 1638, 2932, 3196, 3448 cm^{-1}. Found : C, 65.58 ; H, 3.90 ; N, 4.09%. Calcd for $C_{18}H_{12}NO_2F_3$: C, 65.26 ; H, 3.65 ; N, 4.23%.

4-Cyano-*N*-hydroxy-*N*-naphthalen-1-yl-benzamide (**3h**) : Colorless crystals, mp 150.0-153.0 ºC. ^{1}H NMR ($CDCl_3$) δ=7.23 (1H, d, J=7.3 Hz), 7.34 (1H, dd, J=8.2, 7.3 Hz), 7.41 (2H, d, J=8.2 Hz), 7.45 (2H, d, J=8.2 Hz), 7.61 (1H, dd, J=8.2, 7.3 Hz), 7.67 (1H, dd, J=8.2, 7.3 Hz), 7.90 (1H, d, J=8.2 Hz), 7.92 (1H, d, J=7.3 Hz), 8.17 (1H, d, J=8.2 Hz), and 9.28 (1H, bs). ^{13}C NMR ($CDCl_3$) δ=114.7, 117.7, 122.8, 125.2, 127.3, 127.6, 128.2, 128.6, 128.9, 130.3, 131.1, 131.9, 134.5, 134.8, 135.9, 164.8. IR (KBr) ν 750, 777, 807, 846, 912, 972, 1410, 1506, 1566, 1626, 2230, 2860, 3100, 3424 cm^{-1}. Found : C, 75.20 ; H, 4.67 ; N, 9.70%. Calcd for $C_{18}H_{12}N_2O_2$: C, 74.99 ; H, 4.20 ; N, 9.72%.

N-Hydroxy-4-methoxy-*N*-2-naphthalenyl-benzamide (**5a**) : Colorless crystals, mp 148.0-149.0ºC. ^{1}H NMR ($CDCl_3$) δ=3.75 (3H, s) , 6.72 (2H, d, J=9.1 Hz) , 7.30 (1H, d, J=8.9 Hz) , 7.44 (2H, d, J=9.1 Hz) , 7.47-7.52 (2H, m), 7.72 (1H, d, J=2.4 Hz), 7.74 (1H, dd, J=6.8, 2.4 Hz), 7.77 (1H, d, J=8.9 Hz), 7.81 (1H, dd, J=6.8, 2.4 Hz), and 9.21 (1H, bs). ^{13}C NMR ($CDCl_3$) δ=55.3, 113.6, 123.8, 123.9, 124.6, 126.9, 127.7, 128.2, 129.1, 131.0, 132.4, 133.2, 137.3, 161.8, 165.2. IR (KBr) ν 594, 660, 741, 759, 810, 834, 1026, 1053, 1161, 1179, 1257, 1302, 1407, 1428, 1512, 1569, 1590, 2914, 3112 cm^{-1}. Found : C, 73.89 ; H, 5.42 ; N, 4.71%. Calcd for $C_{18}H_{15}NO_3$: C, 73.71 ; H, 5.15 ; N, 4.78%.

2.2. Determination of Equilibrium Constant

An aqueous solution (3 cm^3) containing metal salts ([$CuCl_2$] or [$FeCl_3$] / mol dm^{-3}=0−0.1) was shaken with a chloroform solution (3 cm^3) of **3** or **5** ($5x10^{-5}$ mol dm^{-3}) for 5 min. The chloroform layer was measured spectrophotometrically (Cu^{2+} : λ=330 nm, Fe^{3+} : 430 nm) and the equilibrium constants were estimated by using the Benesi-Hildebrand approximation equation.[20]

2.3. Transport of Cu^{2+}

Transport experiments were performed using a liquid membrane system which consists of source phase (10 cm^3, 5 x 10^{-3} mol dm^{-3} **3** or **5**), and a receiving phase (10 cm^3, 2.0 moldm^{-3} HCl). A single apparatus and a constant stirring at 25ºC were used.[12,15] As described,[21] to measure Cu^{2+}, 0.5 cm^3 was also taken from aq. I and aq. II and was diluted with water to 5 cm^3. To the diluted solution (0.5 cm^3) aqueous citric acid solution (2.00 g, in 10 cm^3) was added to acidify the solution, and then aqueous EDTA solution (500 mg of EDTA hydrate in 10 cm^3) was added. The mixture was adjusted to pH= 9.0 by adding NH_3 solution. After 20 min the mixture was transferred into a separate funnel and was diluted with water to 50 cm^3. A sodium diethyldithiocarbamate (DDTC) solution (1 x 10^{-2} mol dm^{-3}, 5 cm^3) was added and was shaken with $CHCl_3$ (10 cm^3). The organic layer was dried onto a filter paper and was measured spectrophotometrically (λ=440 nm).

3. Result and Discussition

N-Hydroxy-*N*-naphthylbenzamides (**3, 5**) were prepared from nitronaphthalenes in two steps, as shown in **Fig. 2**.[22] Reduction of **1** with raney-nickel and hydrazine furnished the hydroxylamines (**2, 4**), which were converted to the corresponding *N*-hydroxy-*N*-naphthylbenzamides by benzoylation. The structure and purity of **3** and **5** were ascertained by NMR spectroscopy and elemental analysis.

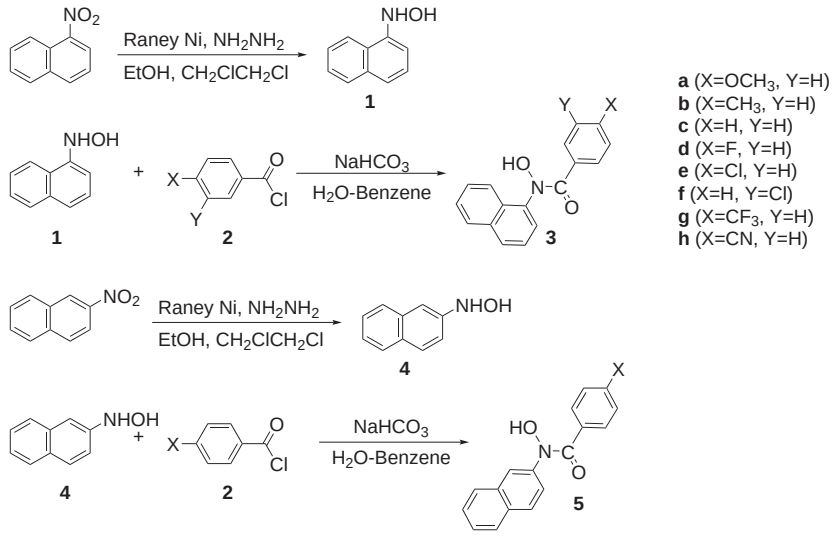


Fig. 2. Synthesis of **3** and **5**.

Extraction of various metal ions (1.0×10^{-2} mol dm^{-3}) into the chloroform solutions containing **3** and **5** (5.0×10^{-5} mol dm^{-3}) was checked by UV spectroscopy ; lithium, sodium, potassium, magnesium, calcium, barium, cobalt, nickel, and zinc ions revealed no indication of UV spectral change, but Cu^{2+} and Fe^{3+} showed a spectral change. In **Fig. 3** are shown the absorption spectra of **3a** and **5c** obtained in the presence of varying concentrations of Cu^{2+} or Fe^{3+}. The complexation of **3** and **5** with Cu^{2+} and Fe^{3+} (**Fig. 3**) showed an enhancement of the absorption, while the Fe^{3+} complex gave a new band at 430 nm. The composition of the complexes was determined as 1 (metal) : 2 (ligand) for the Cu^{2+}- *N*-hydroxy-*N*-naphthylbenzamides (**3, 5**) system and 1 : 3 for the Fe^{3+}- *N*-hydroxy-*N*-naphthylbenzamides (**3, 5**) system by the molar ratio method. The extraction equilibrium constants were determined by the Benesi-Hildebrand method. The equilibrium constants of Cu^{2+} complex were larger than those of Fe^{3+} complex. The deeasing orders of equilibrium constants for Cu^{2+} and Fe^{3+} were **5a** > **3g** > **5c** > **3e** > **3f** > **3d** > **3a** > **3c** > **3b** and **5a** > **3a** > **3d** > **5c** > **3b** > **3g** > **3c** > **3f** > **3e**. Hydroxylamines **3** and **5** captured Cu^{2+} ion under neutral conditions and liberated it upon acidification with hydrochloric acid as detected by UV spectroscopy. This means that **3** and **5** can serve as a transporting agent of Cu^{2+} through the liquid membrane.

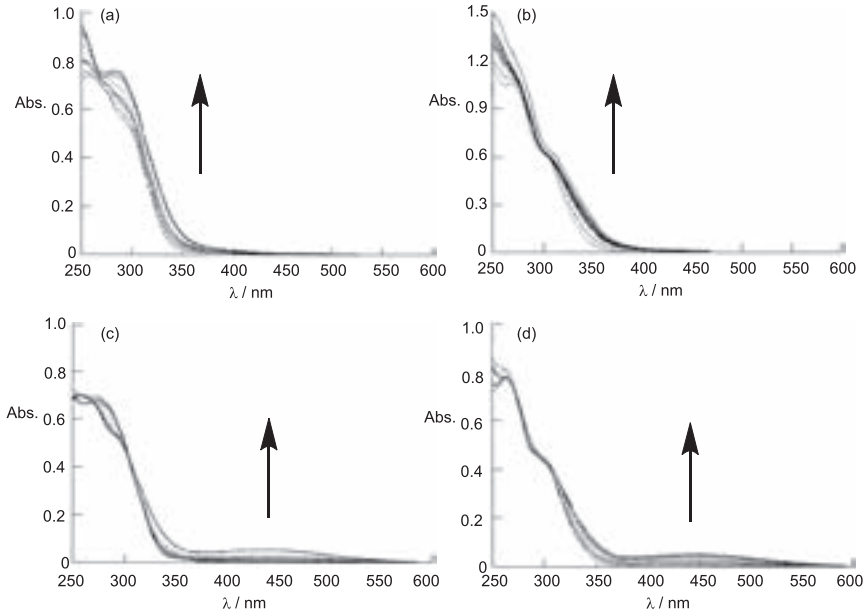


Fig. 3. UV-vis spectral changes of (a) **3a** with Cu^{2+}, (b) **5c** with Cu^{2+}, (c) **3a** with Fe^{3+}, and (d) **5c** with Fe^{3+} in $CHCl_3$.

Table 1. Extraction equilibrium constants of Cu^{2+} and Fe^{3+} complexes of **3** and **5**

	Cu^{2+}	Fe^{3+}
3a	25700	300
3b	21400	170
3c	25400	70
3d	34700	230
3e	47900	40
3f	35500	50
3g	62500	90
5a	73300	1900
5c	50400	200

Transport experiments were performed using a liquid membrane system (see experimental section). The Cu^{2+} concentrations in the aqueous compartments were monitored as a function of time by means of the colorimetric method. The transport data are an average of at least three runs whose experimental error is less than 5%. No movement of Cu^{2+} through the chloroform was observed unless a carrier was used. When an aqueous solution of $CuCl_2$ in the source phase was brought into contact with a chloroform solution of **3** or **5**, stirring with a magnetic bar at 25ºC, the concentration of Cu^{2+} in the

source phase decreased. **Fig. 4** shows the result of transport experiment of Cu^{2+} with **3c**. Cu^{2+} transport with **3** and **5** was promoted by the counterflow of protons from the receiving to the source phase, although proton concentration was not quantitatively investigated. **Fig. 5** shows that the reaction takes place at both interfaces.

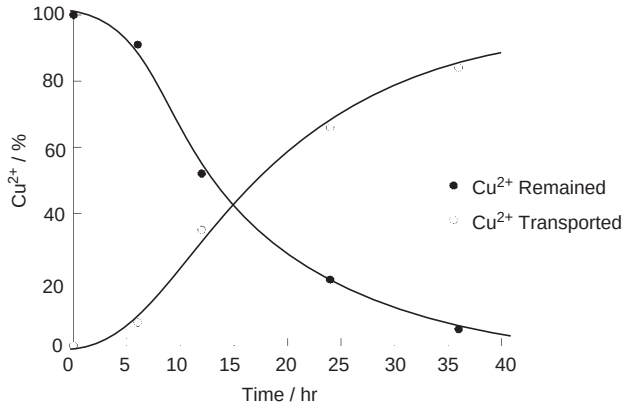


Fig. 4. Percent of Cu^{2+} in the source phase ($CuCl_2$, 5 x 10^{-3} mol dm^{-3}, 10 cm^3) and the receiving phase (2 mol dm^{-3} HCl, 10 cm^3) as a function of time (hr) using the chloroform phase (10cm^3) for carrier **3c** (5 x 10^{-3} mol dm^{-3}) (Reproduced from ref. 5.).

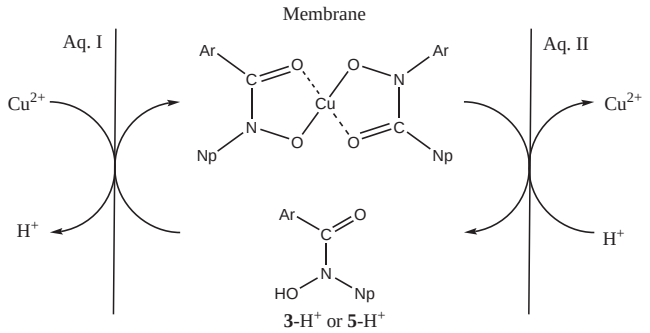


Fig. 5. Schematic presentation of the reactions taking place at the boundary of the source phase and the chloroform and at the boundary of the chloroform and the receiving phase (Modified from ref. 5.).

In conclusion, hydroxamic acid derivatives with naphthyl substituents were found to display unique absorption spectral changes in the presence of Cu^{2+} and Fe^{3+} and are used as analytical reagents for Cu^{2+}. *N*-Hydroxy-*N*-naphthylbenzamides were effective for Cu^{2+} transport through a liquid membrane and indicate that slight structural changes affect the extraction and transport rate of Cu^{2+} to a great extent. This makes it possible to design an effective carrier for Cu^{2+} separation.

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