Liquid−Liquid Equilibria of *p*-Aminophenol between Water and Trialkylamine, Trialkylphosphine Oxide, and Di(2-ethylhexyl)phosphoric Acid in Heptane

Wei Qin,* Deliang Li,† and Youyuan Dai

State Key Laboratory of Chemical Engineering, Tsinghua University, Beijing 100084, People's Republic of China

Extraction of *p*-aminophenol by trialkylamine, trialkylphosphine oxide, di(2-ethylhexyl)phosphoric acid, and the mixed solvent of trialkylamine + trialkylphosphine oxide in heptane was studied under initial pH values ranging from 3 to 10. Trialkylamine and trialkylphosphine oxide extractants extracted the neutral form of *p*-aminophenol. Di(2-ethylhexyl)phosphoric acid extracted both neutral and positive forms of *p*-aminophenol. When the initial pH value was between pK_{a1} and pK_{a2}, a maximum distribution coefficient occurred. This pH dependence can be explained by the variation of the molar fraction of neutral *p*-aminophenol with pH value. The mixed solvent consisting of trialkylamine + trialkylphosphine oxide had a higher distribution coefficient than the sum of those of trialkylamine and trialkylphosphine oxide; that is, there was a synergistic effect when extracting *p*-aminophenol with the mixed solvent.

Introduction

Amphoteric substances, such as aminosulfonic acid, aminophenol, and amino acid, are commonly used in human foods, animal feed additives, and the dye and pharmaceutical industries, as intermediates for production of dyes and pharmaceutics. They are the key components in industrial effluent from printing and dyeing mills, most of which is not biodegradable, for example, aminosulfonic acid and aminophenol; thus, it is necessary to recover the organic species from the effluent to reduce pollution.

Reactive extraction has received increasing attention for the recovery of polar organic species from dilute aqueous solutions. The strong interaction between organic species and the extractant allows for the formation of complexes and thus provides for a high equilibrium distribution coefficient. Reactive extractant can be classified in two major types: (a) phosphorus-bonded, oxygen-containing extractants and (b) high molecular weight aliphatic amines. Generally, a polar diluent with functional groups that enable greater solvation of the complex increases the extracting ability of relatively low polar amines by providing additional solvating power that allows the higher levels of polar-amine complexes to stay in the extractant phase,[1] while a larger polar phosphorus-bonded, oxygen-bearing extractant is also a polar diluent and allows the complexes to dissolve in themselves.[2] Therefore, the optimal composition for the extractant−diluent mixture is a tertiary amine + stronger polar diluent and phosphorus-bonded, oxygen-bearing extractants + nonpolar diluent.

Neutral phosphorus-bonded, oxygen-containing extractants, such as tributyl phosphate and trialkylphosphine oxide, and long-chain, aliphatic amines, such as trioctylamine, are more effective for the extraction of Lewis acid substances, and they have been used in the separation of acetic acid,[3−7] lactic acid,[8−11] propionic acid,[9,12] butyric acid,[9,13] succinic acid,[14] phenols,[15−17] and so forth. Additionally, the acidic, phosphorus-bonded, oxygen-containing extractants, such as di(2-ethylhexyl)phosphoric acid, can be used in the extraction of Lewis base substances, such as organic amines, effectively.[18]

It was found that most studies on the organic species separated from aqueous solutions through reactive extraction are with only one Lewis functional group, Lewis acid (−OH, −COOH, $-SO_3H$), or Lewis base ($-NH_2$), for example, carboxylic acids, phenols, and organic amine solutions. *p*-Aminophenol is a typical chemical with amphoteric functional groups, commonly used as the substrate for the production of pharmaceutical products such as paracetamol, practotol, and vitamin B_1. Additionally, there is increasing interest in using *p*-aminophenol in antioxidants, dyestuff, and photosensitive material. Since *p*-aminophenol has both a Lewis functional group (−OH) and a Lewis base functional group ($-NH_2$), it can be extracted from an aqueous phase into an organic phase with a Lewis base extractant (amines and neutral phosphorus-bonded, oxygen-bearing extractants) and a Lewis acidic extractant (acidic phosphorus-bonded, oxygen-bearing extractants).

In this work, *p*-aminophenol was selected as the extracted species. Trialkylphosphine oxide, trialkylamine, and di(2-ethylhexyl)phosphoric acid were studied for their abilities to extract *p*-aminophenol. Also, heptane was investigated for its ability to affect the extracting power of the extractants over a wide pH range of aqueous solution.

Experimental Section

Chemicals. *p*-Aminophenol, from Fluka, was an analytical reagent with purity > 97 mass %. Trialkylphosphine oxide was kindly supplied by CYTEC Canada Incorporation free of charge. Trialkylamine with purity > 97 mass % was from Jiangsu Feixiang Chemical Co. Ltd. Di(2-ethylhexyl)-phosphoric acid and the diluent, heptane, from Beijing Chemical Reagent Plant were chemical reagents with purity > 97 mass %. All of the extractants were washed with distilled water to remove water-soluble impurities.

* To whom correspondence should be addressed. E-mail: qinwei@tsinghua.edu.cn.
† Present address: School of Chemistry and Chemical Engineering, Henan University, Kaifeng 475001, China.

10.1021/je034124d CCC: $25.00
Published on Web 10/08/2003

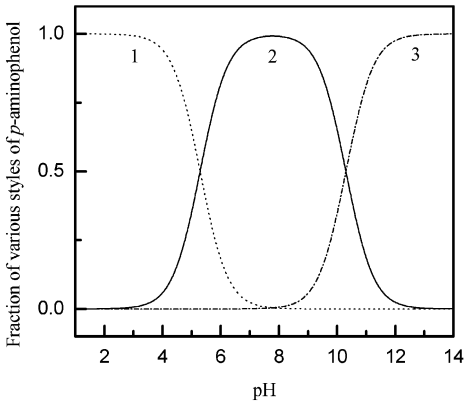

Figure 1. Dependence of the fraction of *p*-aminophenol styles on pH: 1, positive ion *p*-aminophenol; 2, neutral *p*-aminophenol; 3, negative ion *p*-aminophenol.

Extraction Experiments. For the solution preparations, approximately 0.2 g of *p*-aminophenol was dissolved in a 1 L H_2SO_4 solution with the concentration of 0.03 mol·L^{-1}; the effective molecular weight of aqueous *p*-aminophenol solution was 200 mg·L^{-1}.

All extraction experiments were conducted with 100 mL flasks at 25 °C. Unless otherwise noted, 20 mL of the mixture solvents and 20 mL of the organic compound solution were added to each flask. The flask containing the mixture was shaken for about 4 h in a shaker bath with a vibrating rate of 200 rpm and then was left to equilibrate for 1−2 h, followed by separating the two phases. The upper layer (organic phase) was removed, and an aqueous-phase sample was then taken from the bottom layer (aqueous phase) for pH and solute concentration analyses.

Sample Analysis. The aqueous samples were analyzed for *p*-aminophenol concentration (C_{aq}) with a HP-8452 UV spectrometer at 250 nm, when the concentrations of the samples were diluted to 1−10 mg·L^{-1} and the pH was adjusted to >6. The solute concentrations in the organic phase were calculated by material balance. However, the solute concentration in the organic phase (C_{org}) was also determined by first stripping the organic phase with a small amount of NaOH solution (0.02 mol·L^{-1}). The alkaline solution containing the organic salt was then analyzed by using a HP-8452 UV spectrometer. The results from these two methods agreed well within the deviation of 2%. The concentration of each form of the solute (the cation ion, anion ion, and neutral *p*-aminophenol) was calculated from the measured total solute concentration, the pH, and the dissociation constant, K_{a1} and K_{a2}, of the solute. The pH value of the aqueous phase was measured with a pH meter (Hanna pH 201 model) with a deviation of ±0.02.

Results and Discussion

p-Aminophenol has one Lewis acid group, −OH, and one Lewis base group, $-NH_2$. Two dissociation equilibria exist in aqueous solutions as follows:

$$NH_3^+ArOH \overset{K_{a1}}{\rightleftharpoons} NH_3^+ArO^- + H^+ \quad (1)$$

$$NH_2ArOH \overset{K_{a2}}{\rightleftharpoons} NH_2ArO^- + H^+ \quad (2)$$

where pK_{a1} and pK_{a2} are 5.29 and 10.3, respectively.[19]

As shown in Figure 1, the cation or positive ion of *p*-aminophenol exists at low pH, while the anion or negative ion appears at high pH, and neutral *p*-aminophenol dominates at an intermediate pH. Therefore, the extraction equilibrium behaviors of species with amphoteric functional groups are more complicated than that of the normal, and it is essential to understand the effects of pH on the extraction.

Table 1. Equilibrium Data of *p*-Aminophenol between Water and Trialkylphosphine Oxide in Heptane

init conc of extractant/ mol·L^{-1}	init/equil pH in aq phase	total equil conc of *p*-aminophenol/mg·L^{-1} in aq phase	in org phase	distribution coefficient
0.2571	3.37/3.67	151.3	48.7	0.322
0.2571	4.96/4.68	143.9	56.1	0.390
0.2571	5.98/5.71	113.6	86.4	0.761
0.2571	7.23/6.60	83.6	116.4	1.392
0.2571	8.18/6.73	81.1	118.7	1.464
0.2571	8.72/7.07	91.1	108.9	1.195
0.2571	9.37/7.48	114.6	85.4	0.745
0.2571	10.39/8.35	169.1	30.9	0.183
0.5142	3.74/3.72	149.6	50.4	0.337
0.5142	5.71/5.14	104.3	95.7	0.918
0.5142	6.68/6.04	70.2	129.8	1.849
0.5142	7.45/6.20	63.3	136.7	2.160
0.5142	8.01/6.34	58.3	141.7	2.431
0.5142	8.58/6.56	63.8	136.2	2.135
0.5142	9.04/6.77	85.2	114.8	1.347
0.5142	9.70/7.01	118.4	81.6	0.689
0.5142	10.25/7.48	142.1	57.9	0.407
0.7713	3.42/3.57	144.7	55.3	0.382
0.7713	4.60/4.34	136.0	64.0	0.471
0.7713	5.78/5.34	75.9	124.1	1.635
0.7713	6.78/6.27	43.2	156.8	3.630
0.7713	7.14/6.37	40.3	159.7	3.963
0.7713	7.60/6.46	36.8	163.2	4.435
0.7713	7.84/6.55	35.8	164.2	4.587
0.7713	8.22/6.68	39.2	160.9	4.105
0.7713	8.91/6.92	48.6	151.4	3.115
0.7713	9.69/7.14	59.8	140.2	2.344
0.7713	10.95/8.00	109.7	90.3	0.823

Table 2. Equilibrium Data of *p*-Aminophenol between Water and Trialkylamine in Heptane

init conc of extractant/ mol·L^{-1}	init/equil pH in aq phase	total equil conc of *p*-aminophenol/mg·L^{-1} in aq phase	in org phase	distribution coefficient
0.2285	3.34/4.64	186.9	13.1	0.070
0.2285	4.51/5.31	175.9	24.1	0.137
0.2285	5.59/5.91	169.1	30.9	0.183
0.2285	6.75/6.85	149.1	50.9	0.341
0.2285	7.06/7.02	140.1	59.9	0.428
0.2285	7.27/7.11	137.4	62.6	0.456
0.2285	7.82/7.21	151.0	48.9	0.324
0.2285	8.80/7.66	165.3	34.7	0.210
0.4570	3.24/5.05	180.9	19.1	0.106
0.4570	5.16/5.86	169.9	30.1	0.177
0.4570	6.14/6.63	166.2	33.8	0.203
0.4570	6.67/6.88	162.8	37.2	0.228
0.4570	7.05/6.97	159.9	40.1	0.251
0.4570	7.23/7.08	129.6	70.4	0.543
0.4570	7.54/7.20	123.2	76.8	0.623
0.4570	7.71/7.27	124.1	75.9	0.612
0.4570	9.40/7.85	152.2	47.8	0.314
0.6855	3.55/5.78	177.7	22.3	0.125
0.6855	4.85/5.95	170.3	29.7	0.174
0.6855	5.95/6.46	158.1	41.9	0.265
0.6855	6.71/7.01	139.8	60.2	0.431
0.6855	6.99/7.07	133.4	66.6	0.499
0.6855	7.33/7.24	110.2	89.8	0.815
0.6855	7.82/7.30	109.6	90.4	0.825
0.6855	8.23/7.40	122.1	77.9	0.638
0.6855	9.06/7.69	143.1	56.9	0.398
0.6855	10.11/8.17	157.7	42.3	0.268

Effect of pH. Tertiary amines and neutral organophosphorus extractants extract most Lewis acid functional groups from the aqueous phase by forming a complex with

Table 3. Equilibrium Data of *p*-Aminophenol between Water and Di(2-ethylhexyl)phosphoric Acid in Heptane

init conc of extractant/ mol·L^{-1}	init/equil pH in aq phase	total equil conc of *p*-aminophenol/mg·L^{-1}		distribution coefficient
		in aq phase	in org phase	
0.2991	3.58/2.72	156.4	43.6	0.279
0.2991	5.52/2.85	139.6	60.4	0.433
0.2991	6.70/2.94	130.2	69.8	0.536
0.2991	7.24/2.95	128.7	71.3	0.554
0.2991	7.44/2.96	125.3	74.7	0.596
0.2991	7.80/2.97	124.0	76.0	0.613
0.2991	8.40/3.03	127.6	72.4	0.567
0.2991	9.40/3.26	163.4	36.6	0.224
0.5982	4.38/2.68	108.6	91.4	0.842
0.5982	5.27/2.69	107.7	92.3	0.857
0.5982	5.92/2.73	98.1	101.9	1.039
0.5982	7.00/2.75	90.0	110.0	1.222
0.5982	7.35/2.76	89.1	110.9	1.245
0.5982	7.78/2.78	85.6	114.4	1.336
0.5982	8.03/2.79	89.7	110.3	1.230
0.5982	8.53/2.81	95.1	104.9	1.103
0.5982	9.65/2.83	136.3	63.7	0.467
0.8973	4.19/2.62	91.9	108.1	1.176
0.8973	5.71/2.65	82.3	117.7	1.430
0.8973	6.87/2.67	73.4	126.6	1.670
0.8973	7.22/2.68	68.1	131.9	1.937
0.8973	7.53/2.69	66.6	133.4	2.003
0.8973	7.80/2.70	59.2	140.8	2.378
0.8973	8.25/2.71	63.2	136.8	2.165
0.8973	9.19/2.73	75.4	124.6	1.653
0.8973	10.05/2.76	118.6	81.4	0.686

the neutral molecule, and acidic organophosphorus extractants extract most Lewis base functional groups by forming a complex with the neutral molecule or by ion exchange. Because the concentrations of the neutral molecule and positive ion of *p*-aminophenol are a function of pH, the extraction of *p*-aminophenol will greatly depend on the pH of the aqueous phase. As indicated in Tables 1−4 and Figures 2−4, the extraction of *p*-aminophenol with trialkylphosphine oxide, trialkylamine, and di(2-ethylhexyl)phosphoric acid is greatly affected by the pH. The distribution coefficient (*D*) value increased with an increase in the pH value and then decreased. There is a maximum *D* when the initial pH value is between pK_{a1} and pK_{a2}; that is, that the optimal initial pH is around 8.

Comparing the initial pH to the equilibrium pH, the equilibrium solution pH is different from the initial one. The equilibrium solution pH decreases sharply and was around 2−4 with di(2-ethylhexyl)phosphoric acid as the extractant, tended to drop 2 pH units with trialkylphosphine oxide, and increased slightly with trialkylamine as

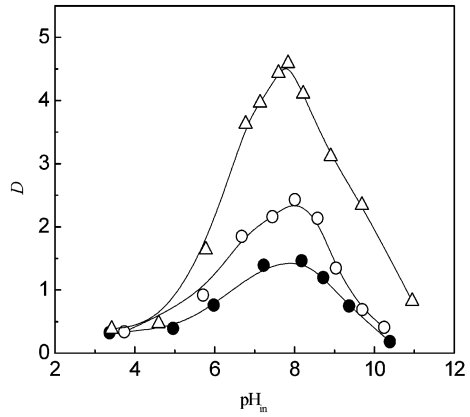

Figure 2. Extraction behavior of *p*-aminophenol with trialkylphosphine oxide: ●, 0.2571 mol·L^{-1}; ○, 0.5142 mol·L^{-1}; △, 0.7713 mol·L^{-1}.

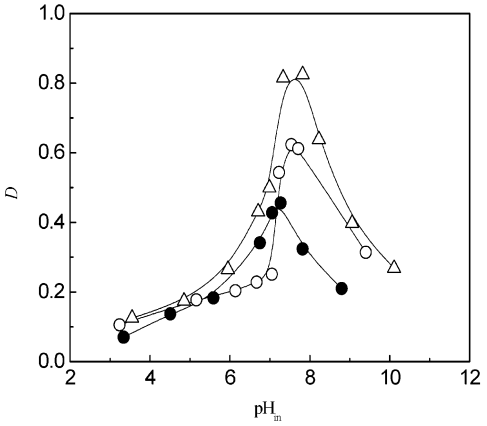

Figure 3. Extraction behavior of *p*-aminophenol with trialkylamine: ●, 0.2285 mol·L^{-1}; ○, 0.4570 mol·L^{-1}; △, 0.6855 mol·L^{-1}.

the extractant. Obviously, the variation extent of the equilibrium solution pH with extractant has the same order as the acidity of the extractants. Generally, the difference between the equilibrium solution pH and the initial one results from the removal of solute and a little dissolution of the extractant used. For the solute of *p*-aminophenol as the neutral species, the variation of solution pH mainly depends on the dissolution of the extractant used.

Effect of Extractant. It was similar to the results of phenol and aniline that, comparing Figure 1 to Figures 2−4, the positive ion of *p*-aminophenol was extractable with di(2-ethylhexyl)phosphoric acid but not with trialkylphos-

Table 4. Equilibrium Data of *p*-Aminophenol between Water and Trialkylamine + Trialkylphosphine Oxide in Heptane

init conc of extractant/mol·L^{-1}			total equil conc of *p*-aminophenol/mg·L^{-1}		
trialkylamine	trialkylphosphine	init/equil pH in aq phase	in aq phase	in org phase	distribution coefficient
0.6855	0.2571	3.56/6.46	40.1	159.9	3.987
0.6855	0.2571	5.74/7.25	38.6	161.4	4.181
0.6855	0.2571	6.62/7.28	38.4	161.6	4.208
0.6855	0.2571	7.12/7.41	36.5	163.5	4.479
0.6855	0.2571	7.63/7.43	36.3	163.7	4.510
0.6855	0.2571	8.10/7.47	47.1	152.9	3.246
0.6855	0.2571	8.51/7.51	48.0	152.0	3.167
0.6855	0.2571	9.29/8.03	60.6	139.4	2.300
0.2285	0.7713	4.64/5.45	47.5	152.5	3.211
0.2285	0.7713	5.97/6.67	26.4	173.6	6.576
0.2285	0.7713	6.59/6.88	24.1	175.9	7.299
0.2285	0.7713	7.22/7.16	23.3	176.7	7.584
0.2285	0.7713	7.46/7.20	22.6	177.4	7.850
0.2285	0.7713	8.11/7.37	22.9	177.1	7.734
0.2285	0.7713	8.50/7.47	28.1	171.9	6.117
0.2285	0.7713	9.38/7.54	58.9	141.1	2.396

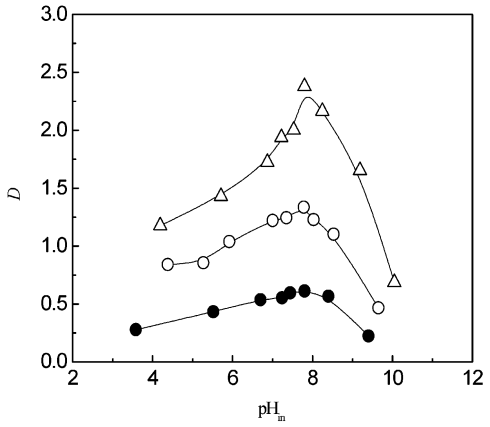

Figure 4. Extraction behavior of *p*-aminophenol with di(2-ethylhexyl)phosphoric acid: ●, 0.2991 mol·L^{-1}; ○, 0.5982 mol·L^{-1}; △, 0.8973 mol·L^{-1}.

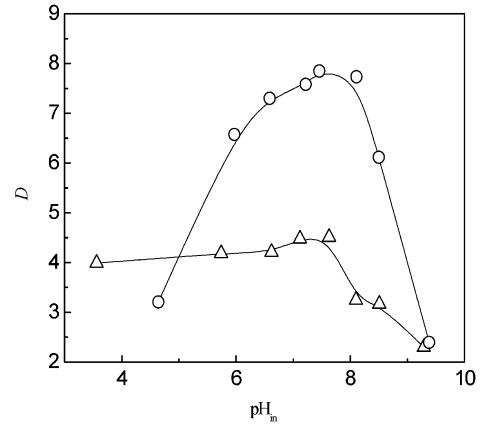

Figure 5. Extraction behavior of *p*-aminophenol with trialkylphosphine oxide + trialkylamine: △, 0.6855 mol·L^{-1} trialkylamine + 0.2571 mol·L^{-1} trialkylphosphine oxide; ○, 0.2285 mol·L^{-1} trialkylamine + 0.7713 mol·L^{-1} trialkylphosphine oxide.

phine oxide and trialkylamine, while trialkylphosphine oxide and trialkylamine are capable only of extracting neutral *p*-aminophenol, and the reactive extractions of *p*-aminophenol with various extractants were as follows:

$$\overline{(HP)_2} + NH_2C_6H_4OH \rightleftharpoons \overline{(HP)_2 \cdot NH_2C_6H_4OH} \quad (3)$$

$$\overline{(HP)_2} + NH_3{}^+C_6H_4OH \rightleftharpoons \overline{P^- \cdot NH_3{}^+C_6H_4OH} + \overline{HP} + H^+ \quad (4)$$

$$\overline{R_3P{=}O} + NH_2C_6H_4OH \rightleftharpoons \overline{NH_2C_6H_4OH \cdot O{=}PR_3} \quad (5)$$

$$\overline{R_3N} + NH_2C_6H_4OH \rightleftharpoons \overline{NH_2C_6H_4OH \cdot NR_3} \quad (6)$$

where $(HP)_2$, $R_3P{=}O$, and R_3N represent di(2-ethylhexyl)-phosphoric acid, trialkylphosphine oxide, and trialkylamine, respectively. The species in the organic phase are marked with an overbar.

As shown in Figures 2−5, the distribution coefficients *D* depended on the extractant type and its concentration (S_0). The values of maximum *D* are proportional to the concentration of the extractant, and they follow the order trialkylphosphine oxide + trialkylamine > trialkylphosphine oxide > di(2-ethylhexyl)phosphoric acid > trialkylamine. For extraction of *p*-aminophenol, trialkylphosphine oxide in the cosolvent trialkylamine + trialkylphosphine oxide not only is capable of extracting *p*-aminophenol directly but also provides a more favorable polar medium for the complex of trialkylamine and *p*-aminophenol than the nonpolar diluent, heptane. The cosolvent of trialkylamine + trialkylphosphine oxide had a higher distribution coefficient than the sum of those of trialkylamine and trialkylphosphine oxide; therefore, the extraction of *p*-aminophenol with the cosolvent showed the synergistic effect.

Compared with conventional mixture solvents such as extractant−carbon oxygen compounds, the cosolvent of trialkylphosphine oxide + trialkylamine can be considered as a selective solvent more attractive for extraction of *p*-aminophenol from the industrial effluent. The regeneration of the cosolvent according to a back-extraction by means of a basic aqueous solution, commonly used for recovery of carboxylic acids from amine base, can be used.

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Received for review June 25, 2003. Accepted September 1, 2003. This research was supported by the National Natural Science Foundation of China (Grant 29836130). D.L. received a financially supported study leave from Henan University, Kaifeng, China.

JE034124D