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Liquid−Liquid Equilibria for Aqueous Solutions of Lithium Sulfate or Lithium Formate and Triethylamine or Diisopropylamine

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Measurements of liquid−liquid equilibria were performed in order to study the suitability of triethylamine and diisopropylamine for the extractive crystallization of lithium sulfate or lithium formate from its aqueous solutions. In the temperature range studied, two liquid phases exist in all ternary-salt-saturated systems. Both amines selectively extract water from saturated lithium sulfate or lithium formate solutions at low temperatures, causing salt crystals to precipitate. Ranges of possible crystallization temperatures are given for each system. The variation with temperature of the two liquid phases compositions allows the amine recovery from the mother liquor. In all cases, a regeneration temperature of 20 °C is chosen.

Introduction

Recently, several papers have been published on the extractive crystallization of inorganic salts whose solubility in water is almost insensitive to temperature. Hanson and Lynn (1989) patented a process for production of sodium chloride from aqueous solution by contacting with an amine. Weingaertner et al. (1991) studied the crystallization of both sodium carbonate from water with 1-butanol and 1-propanol, and sodium chloride with diisopropylamine. Ting et al. (1992) recovered NaCl and Na_2SO_4 from water with *N*,*N*-diethylmethylamine, while Brenner et al. (1992) crystallized Na_2SO_4 with 1-propanol, 2-propanol, and 2-methylpropan-2-ol. Lynn et al. (1996) discussed the extractive crystallization of sodium carbonate, sulfate, and sulfite by using acetone, 2-propanol, and *tert*-butyl alcohol as precipitants. Zijlema et al. (1997) screened dimethylisopropylamine for the precipitation of NaCl.

In an extractive crystallization process, an organic component capable of extracting some of the water molecules surrounding the ions is added to a concentrated aqueous salt solution, decreasing the salt solubility and bringing about its precipitation. The added third component acts as a nonsolvent, extracting water from the aqueous solution, and leads to both a crystallized solid and two liquid phases. The salt-lean phase contains most of the precipitant, which may be recovered in subsequent steps, among other methods, by changing temperature, by countercurrent liquid−liquid extraction, or by fractional countercurrent extraction (Berry et al., 1997). A key to the process is the selection of a suitable precipitation agent. The nonsolvent should be capable both to extract water at the crystallization stage and to release water at the regeneration conditions. Ullmann et al. (1995) suggest the use of organic compounds that have a critical point of miscibility with the feed liquid to be extracted. In any case, knowing the phase diagram of the salt + water + nonsolvent ternary system is the first step necessary for a precipitant selection and a possible implementation of such a process. Measurements of the liquid−liquid−solid equilibrium for the ternary system saturated with salt are necessary to represent conditions that would be found in the crystallizer. Measurements of the liquid−liquid equilibrium for the ternary system subsaturated with salt are useful for calculation of the nonsolvent regeneration stages.

Lithium sulfate has technological interest as an ionic conductor material for laser technology (Heed et al., 1991; Lukaszewicz and Dorosz, 1992). Lithium sulfate has inverted solubility and a nearly zero temperature coefficient of solubility in aqueous solutions. Lithium formate monohydrate exhibits interesting piezoelectric properties (Aleksandrovski et al., 1974) and is a potentially useful nonlinear material for optical device applications (Baldini et al., 1975). Aqueous lithium formate solutions have normal solubility, showing little variation with temperature. In previous papers (Carton et al., 1994a; Carton et al., 1994b; Carton et al., 1996), we studied the purification of these salts by drowning-out with alcohols. This work considers the possibility of precipitation of both salts from aqueous solutions by extractive crystallization. Triethylamine (TEA) and diisopropylamine (DIPA) were considered as possible nonsolvents. Hence, measurements were performed to determine the liquid−liquid equilibria for the ternary systems TEA + H_2O + Li_2SO_4 (or HCOOLi) and DIPA + H_2O +Li_2SO_4 (or HCOOLi), as a function of temperature.

Experimental Section

Panreac PRS grade lithium sulfate monohydrate (99+%) and lithium formate monohydrate, prepared following the procedure described by Cartón et al. (1996), were used as solid reagents. Fluka p.a. triethylamine (99.5+% v/v) and diisopropylamine (99.5+% v/v) were used as nonsolvents. All reagents were used without further purification. Distilled water passed through a Millipore ultrapure cartridge kit was used in preparing solutions.

Duplicate experiments were performed for each ternary system saturated with salt. A solid phase and two saturated liquid phases were always present in these experiments in the temperature range studied. The saturation was achieved by stirring for >48 h, in closed glass cylinders, known masses of nonsolvent and water together with an excess of salt. The accuracy of the compositions was $\pm 10^{-5}$ kg. Approximately 250 cm^3 of liquid mixture was placed into each cylinder, yielding roughly equal

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Table 1. Equilibrium Data for the System Triethylamine (1) + Water (2) + Lithium Sulfate (3), Saturated with Lithium Sulfate

	aqueous phase composition (mass %)			organic phase composition (mass %)		
t (°C)	$100w_1$	$100w_2$	$100w_3$	$100w'_1$	$100w'_2$	$100w'_3$
−5.0	0.7	72.1	27.18	90.36	9.63	0.01
1.3	0.6	73.1	26.30	92.92	7.08	0.01
10.0	0.4	73.7	25.86	96.01	3.99	0.00
20.0	0.2	74.2	25.60	97.47	2.52	0.00
30.0	0.1	74.5	25.36	97.55	2.45	0.00

volumes of the top and the bottom phases. All flasks, fitted with a magnetic stirrer, were placed in a bath controlled to ±0.05 K, the temperature being measured with a digital thermometer (Yokogawa 7563) with a precision of ±0.01 K. A refrigerator was introduced into the bath in the low-temperature experiments. At the end of each run, the agitator was switched off, and the equilibrium mixture was allowed to stand for >1 h at constant temperature to enable any dispersed solid to settle down. Filtered samples of clear solution (between 80 and 90 g) from both liquid phases were taken following the procedure given by Cheluget et al. (1994). Except for the absence of a solid phase in the flasks, a similar experimental procedure was followed for a second set of experiments made for the ternary systems subsaturated with salt. Hence, two liquid phases are always present, but no solid-phase exists in these experiments.

The salt concentration in both liquid phases was determined, in duplicate, by evaporation to dryness of a known mass of solution (between 30 and 40 g). All masses were measured on an analytical balance with a precision of $\pm 10^{-7}$ kg. A constant mass of anhydrous salt was considered only when successive measurements of a sample differed by $<10^{-6}$ kg. Each reported experimental concentration of salt represents the mean of four measurements. On the basis of repeated determinations, the reproducibility of the gravimetric method was $\pm 10^{-4}$ kg of lithium sulfate (or lithium formate)/kg of solution. A Mettler DL-35 model automatic Karl Fischer titrator measured the concentration of water in each phase. Titrating 0.6 g of organic solution and 0.3 g of aqueous solution analyzed water in samples. Hydranal Solvent 34800 and Hydranal Tritrant 34801 (Riedel-deHaën) were used as solvent and tritrant, respectively. Between 35 and 40 mL of solvent reagent was tritrated with Hydranal 34801 in order to eliminate any water in the medium. Then, the known mass of solution was added to the cell and then titrated. After ulterior homogenization, a new aliquot was added to the cell, and then the procedure was repeated again once more. By changing the cell content, triplicates were run for each concentration so the average value of 18 determinations was reported. Measurements of the repeatability and uncertainty of the final data were performed with gravimetrically prepared standard mixtures, giving an error of ±0.25%. Having determined the lithium sulfate and water concentrations, the amine content is given by the difference.

Results

TEA + Water + Li_2SO_4. Table 1 shows the liquid−liquid−solid equilibrium data for the system TEA + water + lithium sulfate saturated with salt, as a function of temperature. The concentration of water in the organic phase increases as temperature decreases. Lithium sulfate remains virtually insoluble in this phase in the temperature range studied. Both of these characteristics indicate that water can be selectively extracted from a saturated lithium sulfate solution by contacting the solution with TEA at a low temperature. In addition, a low weight percent TEA in the aqueous phase is found. However, the water concentration in the organic phase is low, so the expected crystal yield is low. On the other hand, the concentration of lithium sulfate in the aqueous phase rises as temperature decreases, following an opposite trend to those of water extraction to the organic phase. At temperatures below 1.3 °C the increase of the water concentration in the organic phase does not compensate the increase of the salt concentration found in the aqueous phase, so the crystal yield decreases.

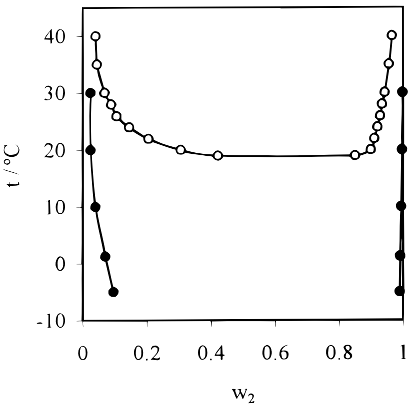

Figure 1. Liquid−liquid equilibrium data for the TEA (1) + water (2) (○) and TEA (1) + water (2) + saturated lithium sulfate (3) (●) systems with w as mass fraction on a salt-free basis.

The measured liquid−liquid equilibrium data for the TEA + water + lithium sulfate system (salt-free basis) are also displayed in Figure 1 together with the liquid−liquid equilibrium data for the TEA + water system (De Santis, 1976). Aqueous solutions of TEA separate into two homogeneous solutions above a certain lower critical solution temperature (LCST), estimated as 18.5 °C. Apparently, the ternary system also exhibits a LCST below −5 °C. The LCST of the amine−water system decreases as a result of the presence of lithium sulfate. The decreasing of the miscibility of the amine−water mixtures by the dissolved salt can be attributed to the fact that it prefers to be surrounded by water molecules and not by amine molecules (Weingaertner et al., 1991).

Data from Table 1 are also useful to gain insight into the recovery of TEA from the organic phase formed in the crystallizer. Increasing temperature would be necessary for nonsolvent regeneration. As the organic phase composition at 30 °C is very close to those of 20 °C, regeneration temperatures over 20 °C do not seem to be justified. Hence, a temperature of 20 °C was selected for regeneration. At such temperature an appreciable decrease of water solubility is produced in the organic phase, no salt is found in such a phase, and the weight percent TEA in the aqueous phase is small. All of these characteristics are favorable for nonsolvent recovery.

To design the amine regeneration step, subsaturated equilibrium data should be necessary. The liquid−liquid equilibrium results for the subsaturated ternary system at 20 °C are given in Table 2. A ternary phase diagram is also displayed in Figure 2. The concentration of lithium sulfate in the organic phase is close to zero. Low percentages of TEA in the aqueous phase lead to significant reduction in the concentration of lithium sulfate. Data from Table 2 show how the water + TEA binary equilibrium is modified due to the presence of lithium sulfate in the

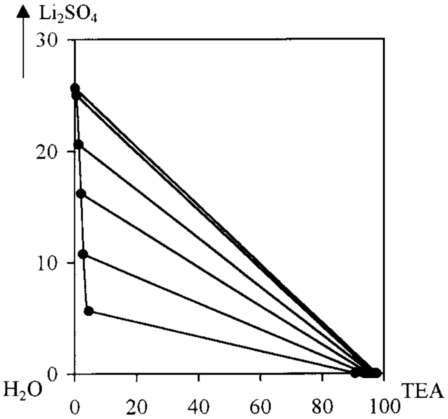

Figure 2. Phase diagram of the TEA (1) + water (2) + lithium sulfate (3) system at 20 °C.

Table 2. Liquid−Liquid Equilibrium of the Triethylamine (1) + Water (2) + Lithium Sulfate (3) System at 20 °C

aqueous phase composition (mass %)			organic phase composition (mass %)		
$100w_1$	$100w_2$	$100w_3$	$100w'_1$	$100w'_2$	$100w'_3$
4.6	89.8	5.67	90.77	9.17	0.06
2.8	86.4	10.78	93.44	6.52	0.05
2.2	81.5	16.22	95.14	4.84	0.02
1.3	78.1	20.62	96.14	3.84	0.02
0.6	74.5	24.96	96.58	3.41	0.01
0.2	74.2	25.60	97.47	2.52	0.00

Table 3. Equilibrium Data for the System Diisopropylamine (1) + Water (2) + Lithium Sulfate (3), Saturated with Lithium Sulfate

	aqueous phase composition (mass %)			organic phase composition (mass %)		
t (°C)	$100w_1$	$100w_2$	$100w_3$	$100w'_1$	$100w'_2$	$100w'_3$
−15.0	11.1	62.1	26.84	67.65	32.28	0.07
−5.0	1.8	71.6	26.57	78.70	21.25	0.05
1.3	0.7	73.1	26.24	81.24	18.72	0.01
10.0	0.5	73.8	25.74	84.91	15.08	0.00
20.0	0.0	74.5	25.48	88.41	11.59	0.00
30.0	0.0	74.8	25.22	91.31	8.69	0.00

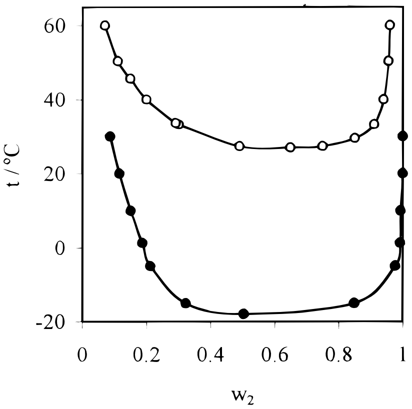

Figure 3. Liquid−liquid equilibrium data for the DIPA (1) + water (2) (○) and DIPA (1) + water (2) + saturated lithium sulfate (3) (●) systems with *w* as mass fraction on a salt-free basis.

Table 4. Liquid−Liquid Equilibrium of the Diisopropylamine (1) + Water (2) + Lithium Sulfate (3) System at 20 °C

aqueous phase composition (mass %)			organic phase composition (mass %)		
$100w_1$	$100w_2$	$100w_3$	$100w'_1$	$100w'_2$	$100w'_3$
2.2	89.1	8.73	71.46	28.49	0.06
1.1	84.2	14.64	79.44	20.54	0.02
0.7	80.1	19.22	84.03	15.95	0.01
0.2	76.5	23.28	87.19	12.80	0.01
0.1	75.4	24.53	87.86	12.13	0.01
0.0	74.5	25.48	88.40	11.60	0.01

aqueous phase. In fact, a 10 mass % TEA solubility in the aqueous phase was found from liquid−liquid equilibrium measurements of the water + TEA system at 20 °C (De Santis, 1976). Compared with the case of the binary system, an appreciable decrease in the TEA solubility in the aqueous phase was observed for the ternary system due to a salting-out effect induced by lithium sulfate.

DIPA + Water + Li_2SO_4. Table 3 shows the liquid−liquid equilibrium data for the system DIPA + water + lithium sulfate saturated with salt, as a function of temperature. As happened with TEA, DIPA is capable of extracting water to the organic phase. The most significant difference between them is that the concentration of water in the organic phase increases much more with DIPA than that found with TEA as temperature decreases. This increase of water concentration clearly compensates the small increase of salt concentration found in the aqueous phase when decreasing temperature. The crystallization is favored at moderate temperatures, in the range 1 to −5 °C, because there is a large extraction of water to the organic phase and the DIPA concentration in the aqueous phase is low.

Figure 3 shows the liquid−liquid equilibrium data for both the binary DIPA + water system (Davison, 1960) and the ternary DIPA + water + lithium sulfate system (salt-free basis). The ternary system exhibits a lower critical solution temperature measured at −18 °C and the liquid-phase composition 26.97 mass % lithium sulfate, 36.81 mass % water, and 36.22 mass % DIPA. The presence of lithium sulfate decreases the amine + water LCST and widens the two-phase envelope.

Above 20 °C the DIPA solubility in the aqueous phase is practically zero. Hence, increasing the temperature above 20 °C can allow the regeneration of DIPA from the organic phase formed in the crystallizer. Table 4 shows the liquid−liquid equilibrium data for the ternary system subsaturated with lithium sulfate at 20 °C. The organic phase compositions (essentially binary DIPA/H_2O mixtures) vary appreciably in this system. The solubility of lithium sulfate in the aqueous phase is strongly dependent on the DIPA to water concentration ratio, even though the DIPA concentration is always low in such a phase.

TEA + Water + HCOOLi. The liquid−liquid equilibrium data for the system TEA + water + lithium formate saturated with salt are given in Table 5, as a function of temperature. A relatively low mass percent TEA in the aqueous phase is observed in the whole temperature range studied. The crystallization is favorable at low temperatures, because of both increasing water extraction to the organic phase and falling formate solubility in the aqueous phase produced as temperature decreases. However, the crystallization temperature should not stand below 1.3 °C. In fact, at −5 °C lithium formate begins to appear in the organic phase and 3.6 mass % TEA appears in the aqueous phase.

Figure 4 reveals that, apparently, the TEA + water + lithium formate system exhibits a LCST below −5 °C, the dissolved salt decreasing the miscibility of the binary TEA + water mixtures. A similar effect was found in other normal solubility systems such as dimethylisopropylamine + water + sodium chloride (Weingaertner et al., 1991;

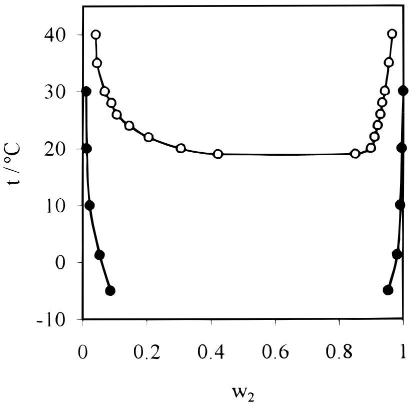

Figure 4. Liquid−liquid equilibrium data for the TEA (1) + water (2) (○) and TEA (1) + water (2) + saturated lithium formate (3) (●) systems with *w* as mass fraction on a salt-free basis.

Table 5. Equilibrium Data for the System Triethylamine (1) + Water (2) + Lithium Formate (3), Saturated with Lithium Formate

	aqueous phase composition (mass %)			organic phase composition (mass %)		
t (°C)	$100w_1$	$100w_2$	$100w_3$	$100w'_1$	$100w'_2$	$100w'_3$
−5.0	3.6	74.5	21.91	91.15	8.64	0.21
1.3	1.5	74.4	24.10	94.69	5.30	0.01
10.0	0.7	73.1	26.20	97.76	2.23	0.01
20.0	0.3	71.8	27.86	98.75	1.24	0.01
30.0	0.0	70.5	29.48	98.95	1.05	0.00

Table 6. Liquid−Liquid Equilibrium of the Triethylamine (1) + Water (2) + Lithium Formate (3) System at 20 °C

aqueous phase composition (mass %)			organic phase composition (mass %)		
$100w_1$	$100w_2$	$100w_3$	$100w'_1$	$100w'_2$	$100w'_3$
3.4	90.8	5.84	93.19	6.78	0.03
2.6	86.8	10.63	95.19	4.78	0.03
1.1	83.2	15.77	96.69	3.30	0.02
1.5	78.1	20.40	97.36	2.63	0.01
1.0	74.4	24.52	97.94	2.05	0.01
0.3	71.8	27.86	98.75	1.24	0.01

Zijlema et al., 1997) and *N*,*N*-diethylmethylamine + water + sodium chloride (or sodium sulfate) (Ting et al., 1992).

From analysis of the compositions given in Table 5 at higher temperatures, a temperature above 20 °C is convenient for TEA regeneration. Table 6 shows the liquid−liquid equilibrium data for the ternary system subsaturated with lithium sulfate at 20 °C. Lithium formate is almost insoluble in the organic phase, observing a slight change in the composition of this phase. The change in the composition found in the aqueous phase is very similar to that of the TEA + H_2O + Li_2SO_4 system. Both salts bring about a similar effect on the equilibrium of the TEA + H_2O binary system.

***DIPA* + *Water* + *HCOOLi*.** Table 7 shows the liquid−liquid equilibrium data for the system DIPA + water + lithium formate saturated with salt, as a function of temperature. The organic phase solvates much more water than that found in the TEA + water + lithium formate system. A crystallization temperature about 1 °C may be selected. A temperature in the range 20 to 30 °C is acceptable for DIPA regeneration.

Figure 5 shows the phase diagram of temperature versus mass percent DIPA concentration (salt-free basis). The liquid−liquid separation with the lower consolute temperature appears for −13 °C and 7.01 mass % lithium formate, 46.14 mass % water, and 46.85 mass % DIPA composition. An enlargement of the DIPA + water miscibility gap was observed with addition of lithium formate. A similar effect was found in the DIPA + water + NaCl system (Weingaertner et al., 1991; Zijlema et al., 1997).

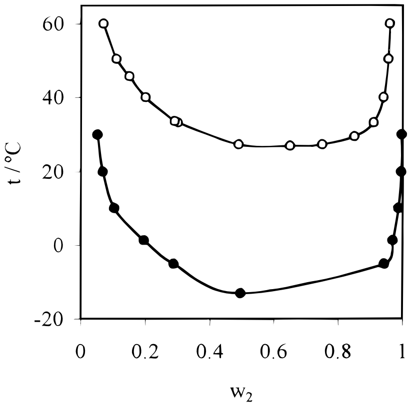

Figure 5. Liquid−liquid equilibrium data for the DIPA(1) + water(2) (○) and DIPA (1) + water (2) + saturated lithium formate (3) (●) systems with *w* as mass fraction on salt-free basis.

Table 7. Equilibrium Data for the System Diisopropylamine (1) + Water (2) + Lithium Formate (3), Saturated with Lithium Formate

	aqueous phase composition (mass %)			organic phase composition (mass %)		
t (°C)	$100w_1$	$100w_2$	$100w_3$	$100w'_1$	$100w'_2$	$100w'_3$
−5.0	4.4	73.9	21.70	68.60	27.68	3.72
1.3	3.3	73.7	23.03	79.19	19.24	1.57
10.0	0.9	73.3	25.78	89.54	10.39	0.07
20.0	0.3	71.9	27.76	93.12	6.86	0.02
30.0	0.2	70.6	29.18	94.81	5.17	0.02

Table 8. Liquid−Liquid Equilibrium of the Diisopropylamine (1) + Water (2) + Lithium Formate (3) System at 20 °C

aqueous phase composition (mass %)			organic phase composition (mass %)		
$100w_1$	$100w_2$	$100w_3$	$100w'_1$	$100w'_2$	$100w'_3$
5.3	86.4	8.25	75.20	24.36	0.44
3.3	82.5	14.21	85.07	14.75	0.18
2.0	78.8	19.22	89.33	10.58	0.09
1.1	75.5	23.41	91.84	8.11	0.06
0.6	74.0	25.34	92.67	7.28	0.04
0.3	71.9	27.76	93.12	6.86	0.02

Table 8 shows the liquid−liquid equilibrium data for the ternary system subsaturated with lithium formate at 20 °C. The concentration of DIPA varies appreciably in the organic phase, much more than that found with TEA. The variation of the lithium formate solubility in the aqueous phase is not as strong as that found with lithium sulfate.

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