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Solubility phenomena studies concerning brines in China*

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Abstract: Solubility equilibria are relevant to many aspects of chemistry and chemical engineering. Studies on solubility phenomena are of importance in understanding chemistry and in the development of chemical technology. Solubility equilibria are especially useful for geochemists investigating the formation and evolution of natural salt deposits. China possesses an enormous variety of such resources, including salt lakes, underground brines, oil-field brines, and natural soda and borate deposits, etc. With the ongoing economic development of China, the exploitation and comprehensive utilization of these kinds of mineral resources are making great progress. Although much research on aqueous salt systems has been published, solubility equilibria studies have long been an active research field in China. This paper presents a review of solubility research on salt/water systems, encompassing more than 200 papers from Chinese researchers.

Keywords: chemical thermodynamics; phase diagrams; solubility.

INTRODUCTION

Because of the great importance of solubility phenomena both in theoretical and engineering practice, studies on them have been an active research field in China. Research in China has covered a wide variety of areas: aqueous salt, molten salt, alloy, and mixed solvent systems. Moreover, the *CALPHAD* technique and different thermodynamic models have been applied in these studies. The purpose of the present paper is to provide an extensive review of solubility research in China concerning salt/water systems. Aqueous salt systems existing in nature, and those produced in chemical plants, are the major focus of this review.

China possesses abundant mineral salt resources with more than 1000 salt lakes distributed across the country [1]. Salt lakes on the Qinghai-Xizang (Tibet) Plateau are famous for their high Li and B concentrations. In recent years, underground brines with high contents of rare elements have been discovered in many provinces in China. These brines often have unusual chemical compositions, e.g., the high B,K,Br,Rb content in Sichuan, and the high Li,Sr,B,Cs content in Qinghai [2]. Salt-lake brines, underground brines, sea-salt bitterns, and oil-field brines are all important natural mineral resources. Solubility data and related equilibrium processes for these systems are essential for developing appropriate process technologies to extract valuable elements and to investigate the geochemical origin of these brines. The exploitation and comprehensive utilization of these salt resources has promoted solu-

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bility studies on salt/water systems in China. Table 1 lists the chemical compositions of various salt lakes around the world.

Table 1 Chemical composition of various salt lake brines around the world (in mass percent).

Salt lake	Country	Li	Na	K	Mg	Ca	Cl	SO_4	B	Density	Mg/Li	Ref.
Atacama[a]	Chile	0.150	7.60	1.85	0.96	0.031	16.04	1.65	0.064	1.223	6.4	[3]
Cauchari	Arg.	0.051	9.33	0.42	0.145	0.033	14.86	1.57	0.112	1.215	2.84	[3]
Hombre Muerto	Arg.	0.062	9.79	0.617	0.085	0.053	15.80	0.853	0.035	1.205	1.37	[3]
Rincon	Arg.	0.033	9.46	0.656	0.303	0.059	16.06	1.015	0.040	1.220	9.29	[3]
Uyuni[a]	Bolivia	0.035	8.75	0.72	0.65	0.046	15.69	0.85	0.02	1.211	18.6	[3]
Great Salt Lake	USA	0.004	7.0	0.4	0.8	0.03	14.0	1.5	0.071	1.218	200	[4]
Bonneville[a]	USA	0.007	9.4	0.6	0.4	0.12	16.0	0.5	0.0004	1.145	57.1	[4]
Searles Lake[a]	USA	0.0083	15.20	2.30	0.034	–	12.39	4.56	0.350	1.30	4.1	[4]
Silver Peak	USA	0.023	6.20	0.53	0.03	0.02	10.06	0.71	0.008	1.14	1.43	[3]
Dead Sea	Isr., Jor.	0.002	3.0	0.6	4.0	0.3	16.0	0.05	0.0033	1.232	2000	[4]
Karabogaz	Turkm.	0.0009	5.11	0.46	3.26	0.02	13.11	6.11	0.023	1.2[b]	3266	[4]
Bieletan[a]	China	0.01	1.882	1.87	5.172	0.035	19.34	0.54	0.0124	1.240	517	[5]
Chaerhan	China	0.0013	5.903	1.0	2.372	0.084	16.67	0.53	0.006	1.2088	1825	[5]
Dachaidan	China	0.02	10.6	0.4	1.3	0.04	18.7	2.25	0.040	1.174	65	[4]
Zabuye	China	0.097	10.81	2.64	0.001	0.007	12.16	5.24	0.286	1.297	0.01	[5]
Zacangchaca[a]	China	0.13	6.769	1.20	1.073	0.002	13.24	2.29	0.042	1.190	8.3	[5]
Xitaijinaier[a]	China	0.021	8.256	0.69	1.284	0.016	14.97	2.88	0.0310	1.2233	61	[5]
Yiliping[a]	China	0.022	6.694	0.91	2.0	0.031	16.17	1.14	0.0184	1.2152	91	[5]
Ocean water		0.000017	1.08	0.039	0.13	0.04	1.94	0.27	0.0004	1.0237647		

[a] Interstitial brine.
[b] Estimated.

Studies of solubility phenomena and related equilibrium processes on salt+water systems in China have been intertwined with the development of chemical processes to exploit these resources and possess distinct characteristics of resources. The salts involved in these studies are primarily halides of the alkali and alkaline-earth metals and the oxyacid salts of B, C, N, and S, i.e., the so-called salt-forming elements. Correctively, these studies have provided considerable new inorganic chemistry knowledge and have made solubility studies an active field in China. These studies have addressed both fundamental and practical questions, and a large number of papers have been published.

Salt lakes are naturally existing mixtures of salts and water and are divided into four types according to their chemical composition: carbonate, chloride, Mg sulfate, and Na sulfate. China possesses all four types. The basis for classification is given in Table 2 [1].

Table 2 Classification of chemical types of salt-lake brine.

Brine type	Characteristic coefficients			
	Kn_1	Kn_2	Kn_3	Kn_4
Carbonate style	>1	>1	>>1	>>1
Sodium sulfate substyle	≤1	≥1	>1	>/<1
Magnesium sulfate substyle	1	≤1	>>1	>/<1
Chloride style	<<1	<<1	≤1	<1

$$Kn_1 = \frac{n_{CO_3^{2-}} + n_{HCO_3^-}}{n_{Ca^{2+}} + n_{Mg^{2+}}},\ Kn_2 = \frac{n_{CO_3^{2-}} + n_{HCO_3^-} + n_{SO_4^{2-}}}{n_{Ca^{2+}} + n_{Mg^{2+}}},\ Kn_3 = \frac{n_{SO_4^{2-}}}{n_{Ca^{2+}}},$$

$$Kn_4 = \frac{n_{CO_3^{2-}} + n_{HCO_3^-}}{n_{Ca^{2+}}}\ (n, \text{equivalent of ions})$$

STUDIES OF AQUEOUS CARBONATE SYSTEMS WITH Li AND BORATE SALTS

Introducing Li^+ and borate ions into carbonate systems creates great differences in solubility phenomena and hence phase diagrams. Chinese chemists have studied solid–liquid phase equilibria for the aqueous quaternary and quinary systems listed in Table 3 (ternary systems are omitted).

Table 3 Aqueous quaternary and quinary systems of the carbonate type with Li^+ or borate.

No.	System	T/°C	Author[b]	Ref.
1	$Li,Na,K/Cl,CO_3–H_2O$	25	Zeng, Y.	6
2	$Li,Na,K/B_4O_7,CO_3–H_2O$	15	Sang, Sh. H.	7
3	$Li,Na/Cl,B_4O_7,CO_3–H_2O$	25	Yin, H. A.	8
4	$Li,K/Cl,B_4O_7,CO_3–H_2O$	25	Wang, Zh. J.	9
5	$Li,Na/SO_4,CO_3–,B_4O_7–H_2O$[a]	15	Sang, Sh. H.	10
6	$Li,K/SO_4,CO_3–,B_4O_7–H_2O$[a]	15	Sang, Sh. H.	11
7	$Na,K/Cl,CO_3,B_4O_7–H_2O$[a]	25	Zeng, Y.	12
8	$Na,K/SO_4,CO_3,B_4O_7–H_2O$[a]	0	Zeng, Y.	13
9	$Li/Cl,SO_4,B_4O_7,CO_3–H_2O$	25	Zeng, Y.	14
10	$Na,K/SO_4,CO_3–H_2O$	0	Zeng, Y.	15
11	$Na,K/CO_3,B_4O_7–H_2O$[a]	25	Zeng, Y.	16
12	$Na,K/CO_3,B_4O_7–H_2O$	15	Sang, Sh. H.	17
13	$Na,K/Cl,HCO_3–H_2O$	25	Wei, X. H.	18
14	$Na,K/SO_4,HCO_3–H_2O$	25	Wei, X. H.	18
15	$Li,Na/CO_3,B_4O_7–H_2O$	25	Zeng, Y.	19
16	$Li,Na/CO_3,B_4O_7–H_2O$	15	Sang, Sh. H.	20
17	$Li,Na/CO_3,B_4O_7–H_2O$[a]	0	Sang, Sh. H.	21
18	$Li,K/Cl,CO_3–H_2O$	25	Deng, T. L.	22
19	$Li,K/CO_3,B_4O_7–H_2O$	25	Zeng, Y.	23
20	$Li,K/CO_3,B_4O_7–H_2O$	15	Yin, H. A.	24
21	$Li,Na,K/CO_3–H_2O$	25	Zeng, Y.	25
22	$Na/Cl,CO_3,B_4O_7–H_2O$[a]	0	Wang, R. L.	26
23	$Na/SO_4,CO_3,B_4O_7–H_2O$	25	Peng, J.	27
24	$Na/SO_4,CO_3,B_4O_7–H_2O$	0	Zhen, Zhi. Y.	28
25	$Li/Cl,CO_3,B_4O_7–H_2O$	25	Yang, H. M.	29

[a] Metastable equilibrium solubility.
[b] First author only.

In the first nine quinary systems, no further double salts were found than those in their corresponding quaternary subsystems. For the system $Li/Cl,SO_4,B_4O_7,CO_3–H_2O$ (at 25 °C), complicated reciprocal interactions cause the crystallization areas of the stable solid phase to change dramatically, despite the system being a rather simple common-ion type. The solubilities of $Li_2B_4O_7·3H_2O$ and Li_2CO_3 in water in the temperature range between 0 and 100 °C (Fig. 1) show that $Li_2B_4O_7·3H_2O$ always has higher solubility than Li_2CO_3 in the temperature range. But in the system $Li/Cl,SO_4,B_4O_7,CO_3–H_2O$ (25 °C), this trend is reversed completely (Fig. 2).

The main cations of carbonate salt lakes are the alkali metals because Mg and Ca carbonates are sparingly soluble in water. Carbonates of Li, Na, and K are soluble to different levels: Li_2CO_3, 1.28; Na_2CO_3, 23.5; and K_2CO_3, 52.7 (at 25 °C) in mass percent. Borates of Li, Na, and K are soluble in similar quantities. All the same, the space phase diagrams of any quinary system involving $Li,Na,K/Cl,SO_4,B_4O_7$, $CO_3–H_2O$ are very complicated. As an example, a $Na,K/SO_4,B_4O_7,CO_3–H_2O$ phase diagram at 273.15 K is illustrated in Fig. 3. All of the above studies provide useful information

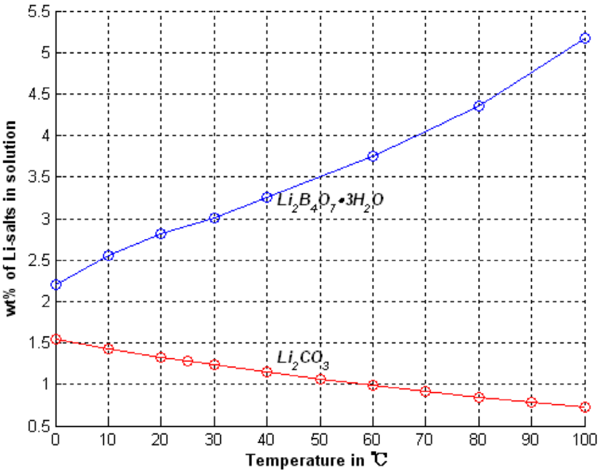


Fig. 1 Solubility of Li_2CO_3 and $Li_2B_4O_7 \cdot 3H_2O$ (in g/100g solution) in water from 0 to 100 °C.

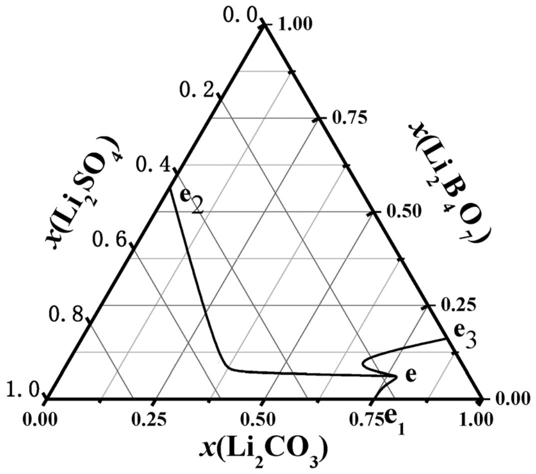


Fig. 2 Phase diagram of the system $Li^+/Cl^-, SO_4^{2-}, CO_3^{2-}, B_4O_7^{2-}–H_2O$ at 298.15 K. Projected on the bottom from the LiCl apex. x: Janecke index, $x_i = n_i / \sum_i n_i$ [14]; eutectic points: e_1-$Li_2CO_3 + Li_2SO_4 \cdot H_2O$, e_2-$Li_2SO_4 \cdot H_2O + Li_2B_4O_7 \cdot 3H_2O$, e_3-$Li_2CO_3 + Li_2B_4O_7 \cdot 3H_2O$, and e-$Li_2SO_4 \cdot H_2O + Li_2B_4O_7 \cdot 3H_2O + Li_2CO_3$.

for recovering Li_2CO_3 from the carbonate salt lakes in Xizang (e.g., Zabuye Salt Lake, whose brine has the second highest concentration of Li (1.0~1.2 g/L) in the world).

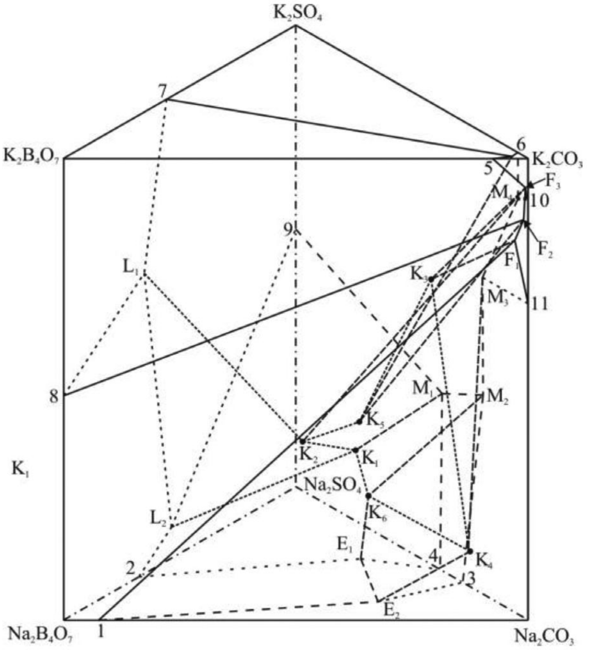


Fig. 3 Phase diagram of the quinary system $Na^+,K^+/SO_4^{2-},CO_3^{2-},B_4O_7^{2-}–H_2O$ at 273.15 K [13].

STUDIES OF AQUEOUS SULFATE AND CHLORIDE SYSTEMS WITH Li AND BORATE SALTS

Chinese chemists have also investigated the solubility equilibria for many aqueous sulfate and chloride systems. For example, solubility equilibria and phase diagrams have been determined for the following quinary systems: $Li,K,Mg/Cl,SO_4–H_2O$ (25 °C), $Li,Na,K,Mg/SO_4–H_2O$ (25 °C), $Li,Mg/Cl,SO_4,B_6O_{10}–H_2O$ (25 °C), $Li,Na,K/Cl,B_4O_7–H_2O$ (25 °C), $Li,Na,K/SO_4,B_4O_7–H_2O$ (0 °C), and $Na,K,Mg/SO_4,B_4O_7–H_2O$ (15 °C) (Table 4). All of the chosen systems share characteristics with salt lakes resources in China, especially those focused on brines with high Li and borate contents. Li sulfate easily forms double or even triple salts, with other alkali metal sulfates, e.g., $Li_2SO_4{\cdot}3Na_2SO_4{\cdot}12H_2O$ (Db1), $Li_2SO_4{\cdot}Na_2SO_4$ (Db2), $Li_2SO_4{\cdot}K_2SO_4$ (Db4), $2Li_2SO_4{\cdot}Na_2SO_4{\cdot}K_2SO_4$ (Db3), $Rb_2SO_4{\cdot}3Li_2SO_4{\cdot}2H_2O$, and $Li_2SO_4{\cdot}Cs_2SO_4$. These studies have widened Li chemistry knowledge. Figure 4 is a stereogram of the phase diagram of the system $Li,Na,K,Mg/SO_4–H_2O$ at 25 °C, and Fig. 5 shows the space volumes of each existing salt. Figures 4 and 5 illustrate that $Li_2SO_4{\cdot}H_2O$ and its double salts Db1, Db2, Db3, and Db4 occupy most parts of the tetrahedron.

Table 4 Aqueous quaternary and quinary systems of sulfate and chloride types.

No.	System	T/°C	Author[a]	Ref.
1	$Li,K,Mg/Cl,SO_4–H_2O$	25	Sun, B.	30
2	$Li,Na,K/Cl,B_4O_7–H_2O$	25	Deng, T. L.	31
3	$Li,Na,K/SO_4,B_4O_7–H_2O$	0	Zeng, Y.	32
4	$Li,Na,K,Mg/SO_4–H_2O$	25	Li, B.	33
5	$Li,Mg/Cl,SO_4,B_6O_{10}–H_2O$	25	Sun, B.	34
6	$Na,K,Mg/SO_4,B_4O_7–H_2O$	15	Sang, Sh. H.	35
7	$Li,Na/Cl,SO_4–H_2O$	25	Hu, K. Y.	36
8	$Li,K/Cl,SO_4–H_2O$	50,75	Ren, K. W.	37
9	$Li,Mg/SO_4,B_4O_7–H_2O$	25	Song, P. Sh.	38
10	$Li,Mg/Cl,SO_4–H_2O$	25	Ren, K. W.	39
11	$Li,Mg/SO_4$,borate–H_2O	25	Sun, B.	40
12	$Na,Mg/SO_4,B_4O_7–H_2O$	15	Sang, Sh. H.	41
13	$Na,K/Cl,B_4O_7–H_2O$	25	Yan, Sh. W.	42
14	$Na,K/Cl,B_4O_7–H_2O$	15	Li, H.	43
15	$Li,Mg/SO_4,B_4O_7–H_2O$	15	Sang, Sh. H.	44
16	$K,Mg/Cl,B_4O_7–H_2O$	15	Sang, Sh. H.	45
17	$K,Mg/SO_4,B_4O_7–H_2O$	15	Sang, Sh. H.	46
18	$Li,K,Mg/SO_4–H_2O$	25	Fang, Ch. H.	47
19	$Li/Cl,SO_4,B_4O_7–H_2O$	25	Song, P. Sh.	48
20	$Mg/Cl,SO_4,B_4O_7–H_2O$	25	Du, X. H.	49
21	$Li,K,Mg/Cl–H_2O$	25	Zhang, F. X.	51
22	$Li,Na,K/B_4O_7–H_2O$	10	Sang, Sh. H.	52

[a]First author only.

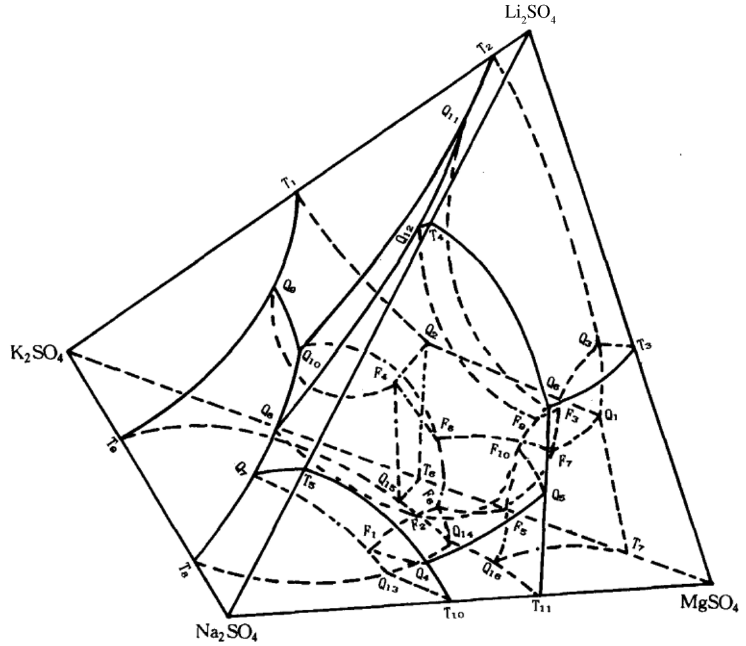
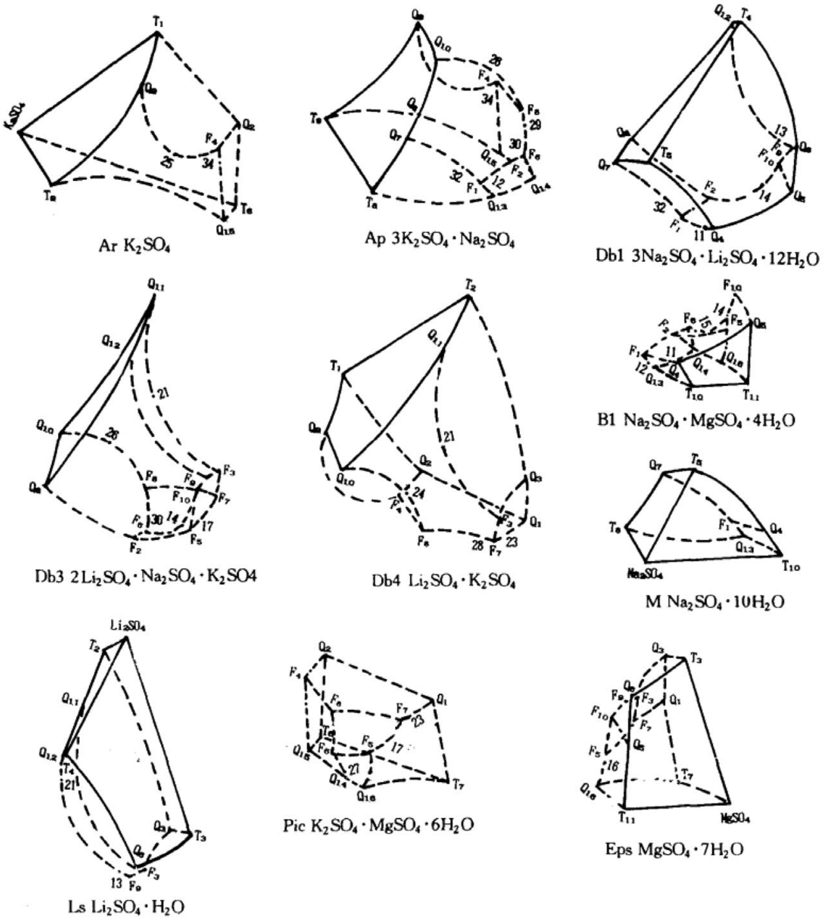


Fig. 4 Phase diagram of the system $Li,Na,K,Mg/SO_4–H_2O$ at 25 °C [33].



Ar K_2SO_4

Ap $3K_2SO_4 \cdot Na_2SO_4$

Db1 $3Na_2SO_4 \cdot Li_2SO_4 \cdot 12H_2O$

Db3 $2Li_2SO_4 \cdot Na_2SO_4 \cdot K_2SO4$

Db4 $Li_2SO_4 \cdot K_2SO_4$

B1 $Na_2SO_4 \cdot MgSO_4 \cdot 4H_2O$

M $Na_2SO_4 \cdot 10H_2O$

Ls $Li_2SO_4 \cdot H_2O$

Pic $K_2SO_4 \cdot MgSO_4 \cdot 6H_2O$

Eps $MgSO_4 \cdot 7H_2O$

Fig. 5 Exploded polyhedron of Fig. 4.

In the system Li,Na,K,Mg/Cl,SO_4–H_2O (25 °C) more equilibria should be considered with solid phases containing Li: $LiCl{\cdot}H_2O$, $Li_2SO_4{\cdot}H_2O$, and their double salts $LiCl{\cdot}MgCl_2{\cdot}6H_2O$ (Li-carnallite), Db1, Db2, Db3, and Db4, besides the simple and double salts in the classical sea-water system Na,K,Mg/Cl,SO_4–H_2O. Therefore, sulfate-type brines with high concentrations of Li have very complicated phase relationships. The two solids $Li_2SO_4{\cdot}H_2O$ and $Li_2SO_4{\cdot}K_2SO_4$ (Db4) were found in mixed salts precipitated during the solar pond evaporation of brines in the salt lakes, Laguocuo in China and Atacama in Chile [50]. Studies on solubility equilibria of this kind of Li brine have special importance to solar pond processes. In some studies, the physico-chemical properties of the equilibrium solutions were determined. The study on the phase diagram of the quaternary system LiCl–KCl–$MgCl_2$–H_2O at 25 °C is an example [51], and contours of physico-chemical properties were plotted in a phase diagram. In another study, an empirical equation for calculation of solution refractive indices was proposed [53], and the calculated and determined values were in good agreement. Later researchers used this empirical equation to accurately calculate the refractive indices of many solutions [54–61].

STUDIES OF METASTABLE EQUILIBRIUM AND NONEQUILIBRIUM SOLUBILITY

As is generally known, solubility phenomena in the metastable equilibrium state may be more important for many practical processes. For example, far more sea salts can be harvested from solar ponds of sea water due to the existence of a metastable equilibrium in which the precipitation of bloedite ($Na_2SO_4 \cdot MgSO_4 \cdot 4H_2O$) is delayed.

Metastable solubility here refers to solubility phenomena exhibited during isothermal evaporation of a salt water system. It should be repeatable equilibruim solubility rather than occasional phenomenon. This concept was first introduced by Russian chemists, who referred to it as a “solar phase diagram”.

Chinese chemists studied metastable equilibrium solubility extensively. Table 5 lists the quaternary and quinary systems where metastable solubility has been studied. All ternary systems are omitted for the sake of simplification. For the classical sea-water quinary system Na,K,Mg/Cl,SO_4–H_2O, the metastable equilibrium solubility and the whole phase diagrams at 15, 25, and 35 °C were completely determined by Jin et al. [62,63,65]. They found that the phase assemblage and space volume of some solid phases were significantly different from the stable equilibrium solubilities. The crystallization volume (Figs. 6 and 7) of Schonite ($K_2SO_4 \cdot MgSO_4 \cdot 6H_2O$) at 25 °C is the biggest from 15 to 35 °C. Quinary systems Na,K/Cl,SO_4,CO_3–H_2O (25 °C), Li,Na,K/B_4O_7,CO_3–H_2O (15 °C), Li,Na/SO_4,B_4O_7,CO_3–H_2O (15 °C), Li,K/SO_4,B_4O_7,CO_3–H_2O (15 °C) and other quaternary systems were also studied, all of which are relevant to industrial applications.

Table 5 Aqueous quaternary and quinary systems of metastable solubilities.

No.	System	T/°C	Author[a]	Ref.
1	Na,K,Mg/Cl,SO_4–H_2O	25	Jin, Z. M.	62
2	Na,K,Mg/Cl,SO_4–H_2O	35	Jin, Z. M.	63
3	Na,K,Mg/Cl,SO_4–H_2O	15	Su, Y. G.	64
4	Na,K,Mg/Cl,SO_4–H_2O	15	Jin, Z. M.	65
5	Li,Na,K/SO_4,B_4O_7–H_2O	15	Zeng, Y.	66
6	Li,Na,K/CO_3,B_4O_7–H_2O	15	Sang, Sh. H.	67
7	Li,Na/SO_4,CO_3,B_4O_7–H_2O	15	Sang, Sh. H.	10
8	Li,K/SO_4,CO_3,B_4O_7–H_2O	15	Sang, Sh. H.	11
9	Na,K/Cl,SO_4,CO_3–H_2O	25	Fang, Ch. H.	68
10	Li,Na/Cl,SO_4–H_2O	0	Wang, Sh. Q.	69
11	Li,Na/Cl,CO_3–H_2O	25	Deng, T. L.	70
12	Li,Na/SO_4,B_4O_7–H_2O	15	Sang, Sh. H.	71
13	Li,Na/CO_3,B_4O_7–H_2O	0	Sang, Sh. H.	21
14	Li,K/Cl,CO_3–H_2O	25	Yan, Sh. Y.	72
15	Li,K/Cl,B_4O_7–H_2O	25	Yan, Sh. Y.	73
16	Li,K/SO_4,CO_3–H_2O	15	Sang, Sh. H.	74
17	Li,K/SO_4,B_4O_7–H_2O	15	Sang, Sh. H.	75
18	Li,K/CO_3,B_4O_7–H_2O	0	Zeng, Y.	76
19	Li,K/CO_3,B_4O_7–H_2O	25	Yan, Sh. Y.	77
20	Li,Mg/Cl,SO_4–H_2O	25	Guo, Zh. Zh.	78
21	Li,Mg/Cl,SO_4–H_2O	35	Gao, J.	61
22	Li,Mg/Cl,SO_4–H_2O	50	Meng, L. Zh.	79
23	Na,K/Cl,NO_3–H_2O	25	Lin, N.	80
24	Na,K/Cl,NO_3–H_2O	25	Feng, T.	81
25	Na,K/Cl,CO_3–H_2O	0	Zeng, Y.	82
26	Na,K/Cl,B_4O_7–H_2O	0	Zeng, Y.	83

(*continues on next page*)

Table 5 (*Continued*).

No.	System	T/°C	Author[a]	Ref.
27	$Na,K/Cl,B_4O_7–H_2O$	35	Deng, T. L.	58
28	$Na,K/CO_3,B_4O_7–H_2O$	0	Zeng, Y.	84
29	$Na,Mg/Cl,SO_4–H_2O$	0	Deng, T. L.	85
30	$Na,Mg/Cl,SO_4–H_2O$	50	Guo, Y. F.	86
31	$Li/Cl,SO_4,B_4O_7–H_2O$	0	Yang, H. X.	87
32	$Li/SO_4,CO_3,B_4O_7–H_2O$	15	Sang, Sh. H.	88
33	$Li/SO_4,CO_3,B_4O_7–H_2O$	0	Qu, Sh. D.	89
34	$Na/Cl,SO_4,NO_3–H_2O$	25,50	Lin, W. Y.	90
35	$Na/Cl,CO_3,B_4O_7–H_2O$	0	Wang, R. L.	26
36	$K/Cl,SO_4,B_4O_7–H_2O$	35	Deng, T. L.	91
37	$K/Cl,CO_3,B_4O_7–H_2O$	25	Yan, Sh. Y.	92
38	$K/SO_4,CO_3,B_4O_7–H_2O$	0	Sang, Sh. H.	93
39	$Li,Na,Mg/B_4O_7–H_2O$	15	Peng, J.	94
40	$Li,Na,Mg/SO_4–H_2O$	–10	Li, Zh. Y.	95
41	$Li,Na,Ca/Cl–H_2O$	15	Deng, T. L.	96
42	$Li,K,Mg/SO_4–H_2O$	15	Deng, T. L.	97
43	$Na,K,Mg/B_4O_7–H_2O$	15	Wei, F.	98
44	$Na,K,Ca/Cl–H_2O$	15	Deng, T. L.	99

[a] First author only.

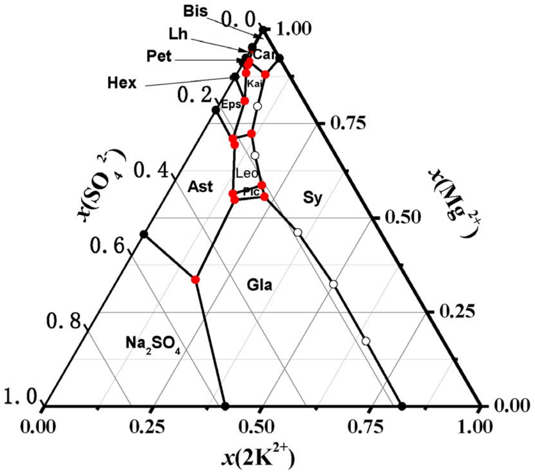


Fig. 6 Van t'Hoff's stable phase diagram of the $Na,K,Mg/Cl,SO_4–H_2O$ quinary system at 25 °C [62]. Ast: $Na_2SO_4·MgSO_4·4H_2O$; Bis: $MgCl_2·6H_2O$; Car: $KCl·MgCl_2·6H_2O$; Eps: $MgSO_4·7H_2O$; Gla: $Na_2SO_4·3K_2SO_4$; Hex: $MgSO_4·6H_2O$; Kai: $KCl·MgSO_4·3H_2O$; Leo: $K_2SO_4·MgSO_4·4H_2O$; Lh: $MgSO_4·4H_2O$; Pet: $MgSO_4·5H_2O$; Pic: $K_2SO_4·MgSO_4·6H_2O$; Sy: KCl.

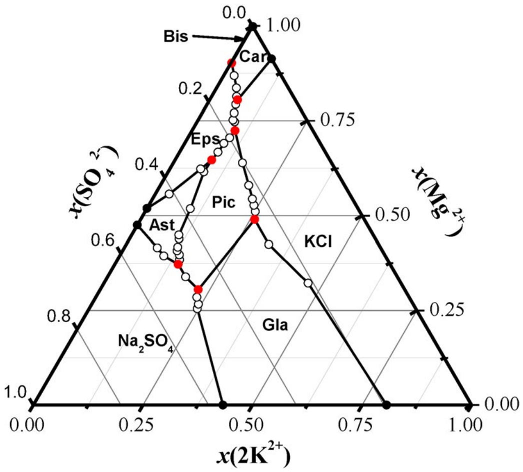


Fig. 7 Metastable stable phase diagram of the Na,K,Mg/Cl,SO_4–H_2O quinary system at 25 °C [62]. (For the meanings of the abbreviation nouns, see Fig. 6.)

Gao Sh. et al. studied Mg-borate solubilities in high-$MgCl_2$ brines in nonequilibrium states. Phase diagrams for the MgO–nB_2O_3–$wMgCl_2$–H_2O systems ($n = 0, 1, 2, 3, 1/3$; $w = 0.08, 0.18, 0.28$) at 0 and 20 °C were constructed [1,100–104].

Zhou et al. [105–109] studied nonequilibrium-state solubilities during the boiling evaporation process of Na,Mg/Cl,SO_4–H_2O and its subsystems. Their work showed interesting results, and they proposed the concepts of primary, extended, and overlay salt-forming regions to characterize differences of solubility phenomena between the corresponding stable and metastable equilibria.

STUDIES OF SYSTEMS CONCERNING PROCESSING TECHNOLOGY OF OTHER NATURAL MINERAL RESOURCES

In order to utilize certain mineral resources in China, e.g., ludwigite (syngenetic magnetite-ascharite), natural nitrate-containing Mg, natural soda and underground brines, solubility studies were carried out for the systems: Na,K,Mg/Cl,SO_4,NO_3–H_2O (25 °C), Na,Mg/Cl,SO_4,NO_3–H_2O (25 °C), Na,K/Cl,NO_3–H_2O (25, 50 °C), Na,K,Mg/Cl,NO_3–H_2O (25 °C), K,Mg/Cl,NO_3–H_2O (25 °C), K,Mg/SO_4,NO_3–H_2O (25 °C), and Na,K,Mg/NO_3–H_2O (25 °C) (Table 6). Figure 8 is the space phase diagram of the system Na,Mg/Cl,SO_4,NO_3–H_2O at 25 °C, the structure of which is very complicated. Borate-containing systems Mg/SO_4,Cl–H_2BO_3–H_2O (25, 100 °C), Mg/SO_4,NO_3–H_2BO_3–H_2O (25, 100 °C), Na/Cl,SO_4,–H_2BO_3–H_2O (25, 100 °C), Na,K/Cl,SO_4–H_3BO_3–H_2O (25 °C), and Na,Mg/Cl–H_2BO_3–H_2O (25, 100 °C), etc. were studied. Some good work has been completed for the quinary systems Na,K,NH_4/Cl,SO_4–H_2O (100 °C), Na/Cl,SO_4,CO_3,HCO_3–H_2O (150 °C), Na/Cl,SO_4,OH,CO_3–H_2O (150 °C), and their quarternary systems Na/Cl,SO_4,OH–H_2O (150 °C), Na/Cl,OH,CO_3–H_2O (150 °C), Na/Cl,SO_4,OH,–H_2O (150 °C), and Na/Cl,SO_4,CO_3–H_2O (150 °C). Some research on aqueous Sr salt systems [135–137] has been reported for certain Sr-rich underground brines.

Table 6 Aqueous quaternary and quinary systems of borate, nitrate. and others.

No.	System	*T*/°C	Author[a]	Ref.
1	$Na,Mg/Cl,SO_4,NO_3–H_2O$	25	Huang, X. L.	110
2	$Na,K,Mg/Cl,SO_4,NO_3–H_2O$	25	Huang, X. L.	111
3	$Na,K,Mg/Cl,SO_4,NO_3–H_2O$	25	Huang, X. L.	112
4	$Na,K,Mg/Cl,SO_4,NO_3–H_2O$	25	Huang, X. L.	113
5	$Na,Mg/Cl,SO_4,NO_3–H_2O$	25	Huang, X. L.	114
6	$Na,K/Cl,NO_3–H_2O$	25	Liu, N.	80
7	$Na,K/Cl,NO_3–H_2O$	25	Feng, T.	81
8	$Na,K/Cl,NO_3–H_2O$	25,50	Lin, W. Y.	90
9	$K,Mg/SO_4,NO_3–H_2O$ $Na,K,Mg/NO_3^-–H_2O$	25	Yang, Zh.	115
10	$K,Mg/Cl,NO_3–H_2O$	25	Li, S. W.	116
11	$Na,K,Mg/Cl,NO_3–H_2O$	25	Huang, X. L.	117
12	$Mg/SO_4,Cl–H_2BO_3–H_2O$	25,100	Cao, J. L.	118
13	$Mg/SO_4,NO_3–H_2BO_3–H_2O$	25,100	Cao, J. L.	119
14	$Na/Cl,SO_4–H_2BO_3–H_2O$	25,100	Cao, J. L.	120
15	$Na,K/Cl,SO_4–H_3BO_3–H_2O$	25	Cao, J. L.	121
16	$Na,Mg/Cl–H_2BO_3–H_2O$	25,100	Cao, J. L.	122
17	$H_3BO_3–MgSO_4–MgCl_2–H_2O$	15	Chen, H. Ch.	123
18	$H_3BO_3–MgSO_4–MgCl_2–H_2O$	0	Gao, Z. L.	124
19	$K/CO_3,SO_4,B_4O_7–H_2O$	30	Yin, J. Zh.	125
20	$Na/Cl,SO_4,CO_3–H_2O$	150	Li, X. S.	126
21	$Na/Cl,SO_4,OH,CO_3–H_2O$	150	Liu, J. R.	127
22	$Na/Cl,SO_4,OH–H_2O$	150	Zhang, Ch. D.	128
23	$Na/Cl,OH,CO_3–H_2O$	150	Zhang, Ch. D.	128
24	$Na/Cl,SO_4,OH–H_2O$	150	Zhang, Ch. D.	128
25	$Na/Cl,SO_4,CO_3–H_2O$	150	Zhang, Ch. D.	128
26	$Na,NH_4/Cl,SO_4,HCO_3–H_2O$	35	Zhang, M. J.	129
27	$Na,K,NH_4/Cl,SO_4–H_2O$	100	Zhang, M. J.	130
28	$Na/Cl,CO_3,B_4O_7–H_2O$	0	Zhang, F. X.	131
29	$Na/CO_3,HCO_3,B_4O_7,BO_2–H_2O$	25,35,45	Li, J.	132
30	$Na/CO_3,HCO_3,B_4O_7,BO_2–H_2O$	0,15	Li, J.	133
31	$Na/CO_3,HCO_3,B_4O_7,BO_2–H_2O$	0–45	Zhang, F. X.	134

[a] First author only.

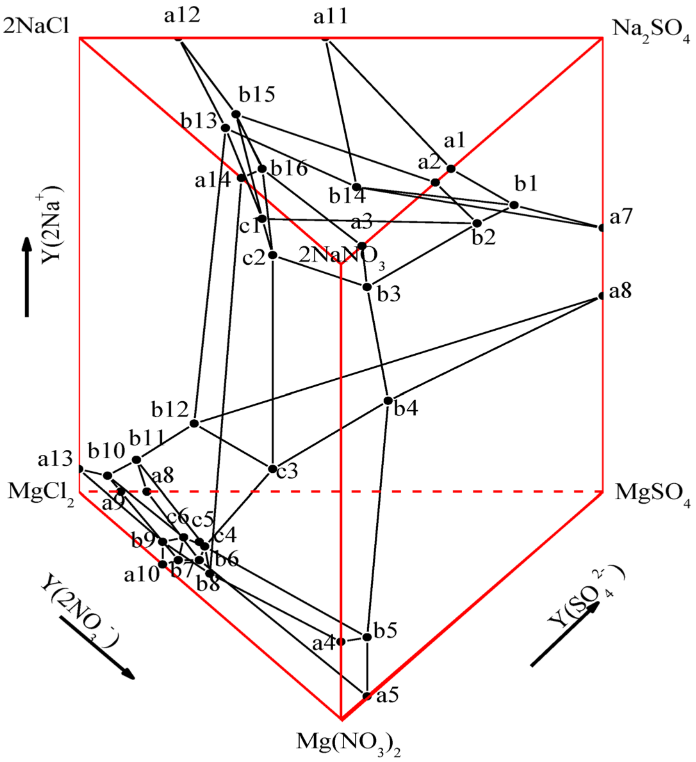


Fig. 8 Phase diagram of Na,Mg/Cl,SO_4,NO_3–H_2O at 298.15 K [110].

STUDIES OF SALT-WATER SYSTEMS WITH ORGANIC COMPONENTS

Tables 7 and 8 summarize research on salt-water systems also containing one or more organic alcohols. Most systems containing Rb and/or Cs salts in Table 7 were studied in the search for more effective methods of separation from their solutions and purification of the salts. Since the alcohol content is low in most cases, only a homogeneous solution phase exists.

Table 7 Rb- and Cs-containing aqueous systems with organic components.

System	*T*/°C	Ref.
Rb_2CO_3–C_2H_5OH–H_2O	0–60	138–141
Rb_2SO_4–CH_3OH/C_2H_5OH–H_2O	30,50	142–144
RbCl/$RbNO_3$/Rb_2SO_4/Rb_2CO_3–C_2H_5OH–H_2O	25	145–147,159
RCl–R'Cl–HOAc(R,R' = K^+, Rb^+, Cs, R ≠ R')	25,35,45	148
RbCl/RbBr/CsCl/CsBr/CH_3OH/C_2H_5OH–H_2O	25	149
CsCl–C_2H_5OH–H_2O	20–60	150–151
Cs_2CO_3–C_2H_5OH–H_2O	10,30,50	152–153
$CsNO_3$/Cs_2SO_4–ROH–H_2O	−10–50	154–159
Cs_2SO_4–polyethylene glycol–H_2O	25,35,45	158,160
Cs_2CO_3–C_3H_7OH–H_2O	25,35,45	161
CsCl–Cs_2SO_4/Cs_2CO_3–*n*-C_3H_7OH/C_2H_5OH–H_2O	20,25	162–163
NaCl–RbCl–CH_3OH/C_2H_5OH/1–C_3H_7OH/2–C_3H_7OH–H_2O	15,25,35	164
KCl–CsCl–CH_3OH/C_2H_5OH/1–C_3H_7OH/2–C_3H_7OH–H_2O	15,25,35	165

Table 8 Other aqueous salt systems with mixed solvents.

No.	System	*T*/°C	Author[a]	Ref.
1	Li_2SO_4–C_2H_5OH–H_2O	–20~50	Xia, Sh. P.	166
2	K/Cl,NO_3–CH_3OH–H_2O	25	Liu,Y. F.	167
3	K,NH_4/NO_3–CH_3OH–H_2O	25	Liu,Y. F.	167
4	K,NH_4/NO_3–CH_3OH–H_2O	10	Yuan, J. Sh.	168
5	Mg,NH_4/Cl,–CH_3OH–H_2O	0–79	Hu, C. H.	169
6	Li,Mg/SO_4–C_2H_5OH–H_2O	–5–50	Zhang, J.	170
7	Na,Mg/SO4–C_2H_5OH–H_2O	25	Zhang, F. X.	171
8	Na,K/Cl,SO_4,C_2H_5OH–H_2O	25	Guo, Zh. Zh.	172
9	K/Cl,SO_4–$CO(NH_2)_2$–H_2O	25	Zhang, F. X.	173
10	Mg,/Cl,SO_4–$CO(NH_2)_2$–H_2O	25	Zhang, F. X.	174
11	K,NH_4/SO_4–$CO(NH_2)_2$–H_2O	25	Zhang, Y. J.	175
12	K,Mg/Cl,SO_4–$CO(NH_2)_2$–H_2O	25	Zhang, F. X.	174
13	K,Ca/Cl,SO_4–$CO(NH_2)_2$–H_2O	25	Sun, B.	176
14	NaCl–$CO(NH_2)_2$–H_2O_2–H_2O	15	Zhang, C.	177
15	NH_4Cl–$CO(NH_2)_2$–H_2O_2–H_2O	15	Guo, K. N.	178
16	Na/Cl,CO_3–H_2O_2–H_2O	25	Cao, J. L.	179
17	K,NH_4/SO_4–NH_3–H_2O	25	Cao, J. L.	180
18	K,NH_4/Cl,SO_4–NH_3–H_2O	40	Shu, L.	181
19	K,Ca/Cl,SO_4,36 %NH_3–H_2O	10	Huang, X. L.	182
20	H,Li/Cl–H_2O	25	Li, G.	183
21	H,Mg/Cl–H_2O	–10	Wang, J. Sh.	184
22	H,Li,Mg/Cl–H_2O	0	Hu, K. Y.	185
23	H,Li,Mg/Cl–H_2O	–10	Wang, J. Sh.	186
24	M,M'/Cl–HOAc (M,M' = K, Rb, Cs, M ≠ M')	25	Ran, X. Q.	187
25	Li,M/Cl(M = K, Rb, Cs)–HOAc	25,40	Ran, X. Q.	188
26	CsCl–$PrCl_3$–13 %HCl–H_2O	25	Li, Y. H.	189
27	CsCl–$PrCl_3$–42 %HOAc–H_2O	30	Li, Y. H.	89

[a]First author only.

Other investigated systems containing organic components such as carbamide, carboxylic acids, or mannitol, and mixed solvents of water with ammonia, hydrogen peroxide, hydrochloric acid, sulfuric acid, or nitric acid are listed in Table 8.

THERMODYNAMIC MODELS AND SOLUBILITY EQUILIBRIUM PREDICTIONS

Solubility prediction using thermodynamic models of electrolyte solution is a useful tool for studying aqueous salt systems, particularly when employing Pitzer's ion-interaction model. A Pitzer model for predicting solubilities in aqueous salt-water systems was first used in China in 1983 [190]. As mentioned above, salt lake brines on the Qinghai-Xizang (Tibet) Plateau belong to the system Li^+, Na^+, K^+, Mg^{2+}/Cl^-, SO_4^{2-}, and borate-H_2O. Our work aimed to establish Pitzer model parameters of the system, which could be used for the entire range of concentrations up to saturation.

Because the thermodynamic properties of Li-containing aqueous solutions were not available in the literature, Song et al. determined experimentally osmotic or activity coefficients of Li-salts solutions [191]. Solubility predictions were performed for different types of aqueous systems listed in Table 9. Our work on the model for the system Li,Na,K,Mg/Cl,SO_4–H_2O included predicting solubilities for the system itself and all its subsystems, the thermodynamic properties of Li-bearing brines, and their applications in chemical process technology. References [195–197] briefly summarize the results obtained while Fig. 9 shows the predicted space phase diagram for the quinary system Li,K,Mg/Cl,SO_4–H_2O at 25 °C.

Table 9 Prediction of solubilities for aqueous salt systems.

No.	System or key words	T/°C	Author[a]	Ref.
1	Na,K/Cl–H_2O, H,Na/Cl–H_2O et al.	25	Song, P. Sh.	190
2	MCl–HCl–H_2O (M = Li,Na,K,Rb,Cs)	25	Li, Y. H.	192
3	Calculation of isohydrores	25	Song, P. Sh.	193
4	Na,K/Cl,SO_4–CO_3–H_2O	25	Fang, Ch. H.	194
5	Li,K,Mg/Cl,SO_4–H_2O	25	Song, P. Sh.	195
6	Li,Na,K,Mg/Cl,SO_4–H_2O	25	Song, P. Sh.	196
7	Li,K/Cl,SO_4–H_2O	25	Song, P. Sh.	197
8	H,Na/Cl–H_2O	25	Li, Y. H.	198
9	H,K/Cl–H_2O	25	Li, Y. H.	199
10	HCl–LiCl–$MgCl_2$–H_2O	0,20,25	Li, Y. H.	200–202
11	HCl–CsCl–H_2O	25	Li, Y. H.	203
12	HCl–RbCl–H_2O	25	Li, Y. H.	204
13	HCl–$MgCl_2$–H_2O	–5	Li, Y. H.	205
14	Na,K,Mg/NO_3,Cl,SO_4–H_2O	25	Huang, X. L.	206
15	Na,K/NO_3,Cl–H_2O	25	Song, P. Sh.	207
16	K,Mg/NO_3,SO_4–H_2O	25	Zhang, J.	208
17	K,Mg/NO_3,Cl–H_2O	25	Song, P. Sh.	209
18	K/NO_3,Cl,SO_4–H_2O; Mg/NO_3,Cl,SO_4–H_2O	25	Song, P. Sh.	210
19	K,Na/Cl,SO_4,NO_3–H_2O and its subsystems	25	Huang, X. L.	211

[a] First author only.

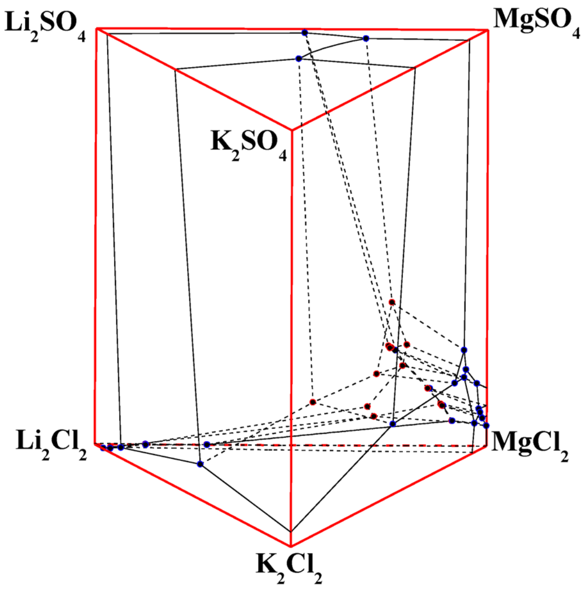


Fig. 9 Predicted phase diagram for Li,K,Mg/Cl,SO_4–H_2O at 298.15 K [this work].

Additionally, Zeng et al. [214–219] found that the Brunauer–Emmett–Teller (BET) model [212,213] is especially suitable for representing the solubility equilibria of highly soluble salts, such as chlorides and nitrates of Li, Mg, Ca, etc. For the systems consisting of both moderately soluble and highly soluble salts, such as sulfates and chlorides of alkali metals and rare earth metals, a Pitzer–Simonson–Clegg (PSC) model would be more suitable [220–224]. For representing the solubility of complicated systems related to salt-lake brines over a wide temperature range, more thermodynamic data, such as water and salt activities and solubility isotherms, at temperatures other than 25 °C are needed for model parameterization.

SUMMARY AND CONCLUSIONS

Solubility phenomena are relevant to chemistry, chemical engineering, and geochemistry. For a long time, Chinese researchers have studied the solubilities of salt-water systems. This comprehensive review has demonstrated that Chinese chemists have contributed much to the understanding of solubility phenomena in natural multicomponent aqueous salt systems. Most of these studies explored Li and B salt systems at 0 and 25 °C. The presence of the highly soluble salt LiCl and borate salts that readily form complicated species make the relevant phase diagrams for salt lakes considerably more complicated than those sea-water systems. While phase diagrams of multicomponent brine systems have been widely investigated at low temperatures, their characterization at temperatures higher than 25 °C would be useful to chemical engineers in generating new ideas for the extraction of valuable resources from salt lakes and will therefore benifit from further investigation.

Valuable elements are mostly extracted from salt lakes using evaporation processes employing the natural energy of the sun. To develop optimal evaporation routes and improve the separation efficiency, simulations of evaporation process based on phase diagram calculations can save a huge amount of experimental work. Appropriate thermodynamic models should be chosen to perform the simulation work. Among all of the thermodynamic models developed, the PSC model should be the prospective one. For the parameterization of the model, solubility data should be critically evaluated and selected. Component activities, i.e., water activity and mean activity coefficient of salts in multicomponent systems of brines, should be measured accurately further, especially at temperatures higher than 25 °C.

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