

Solubilities of BaCl₂ in NaCl and/or KCl Aqueous Solutions at Temperatures from 50 to 140°C

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Solubilities of BaCl₂ (exactly BaCl₂ · 2H₂O) in the quaternary system BaCl₂-NaCl-KCl-H₂O have been determined by mean of the electric conductivity method between 50 and 140°C under vapor-saturated conditions with relative errors less than 6 %. At constant temperatures, depending largely on NaCl concentration, and less on KCl one, the solubilities of BaCl₂ · 2H₂O in NaCl, KCl and NaCl+KCl solutions having salt/(salt+water) weight ratios of 0, 0.05, 0.10 and 0.15 decrease with increasing salt concentrations. On the other hand, when the concentration of a solvent is constant, the solubilities increase with increasing temperature. The result in the quaternary system indicates that the solubilities of BaCl₂ in the system can be linearly estimated from the data of systems BaCl₂-NaCl-H₂O and BaCl₂-KCl-H₂O.

Introduction

Solubility data give us one of the most important keys for discussion on mineral precipitation from hydrothermal solutions. For this reason, many authors have tried to determine solubilities of various kinds of salts, and reported many available data. Unfortunately, most of them are restricted, however, within binary systems including H₂O, and only a few are obtained in ternary systems. On contrast, a mineral usually precipitates in a multiple component system. For instance, barite (BaSO₄), which is one of the important gangue minerals characterizing the kuroko-type ore deposit and also the submarine hydrothermal deposit occurring in back-arc basins, is believed to be formed by mixing of ascending fluid and sea water, and hence requires a phase relation in the system Ba²⁺-Na⁺-SO₄²⁻-Cl⁻-H₂O for the discussion on the formation.

Shoji et al.⁷⁾ have developed a new technique by which solubilities of salts in hydrothermal solution are easily determined. To check the applicability of the technique to multicomponent systems, solubilities of BaCl₂ in NaCl, KCl and NaCl+KCl aqueous solutions have been determined.

Based on the results of the X-ray powder diffraction, the species containing barium

is $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$. Exactly speaking, accordingly, the obtained data are the solubilities of $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$, but are described here as the solubilities of BaCl_2 because of simplifying the statement.

Experimental and Determination Procedure

Solubilities of barium chloride have been determined between 50 and 140°C in vapor-saturated conditions. The solvents are pure water, and NaCl, KCl and NaCl+KCl aqueous solutions; where salt/(salt+water) or salts/(salts+water) weight ratios are 0, 0.05, 0.10 and 0.15, and NaCl/(NaCl+KCl) weight ratios are 0, 0.5 and 1. Reagent grade BaCl_2 , NaCl and KCl were used after drying at temperatures above 110°C over 24 hours.

In this study has been used the electric conductivity method, where condition of a sample solution in a TEFLON reaction vessel is monitored by the electric conductivity at elevated temperatures⁷⁾. For instance, the solubility of BaCl_2 in pure water is determined by the following procedure. Let weight fraction of component BaCl_2 in solution (simply BaCl_2 concentration) and weight fraction of component BaCl_2 in bulk composition (simply bulk BaCl_2 composition) be respectively $W_{dis}/(W_{dis} + W_{wat})$ and $(W_{dis} + W_{slid})/(W_{dis} + W_{slid} + W_{wat})$, where W_{dis} , W_{slid} and W_{wat} are weights of dissolved BaCl_2 , solid BaCl_2 and water. As shown in Figure 1 (selected data are listed in Table A1), primary data are given as a temperature-conductivity relation for a constant bulk BaCl_2 composition. The conductivity increases with increasing temperature as a function of both temperature and concentration, where BaCl_2 concentration is less than bulk BaCl_2 composition (i.e. saturation). On the other hand, where BaCl_2 concentration equals to the bulk BaCl_2 composition (i.e. undersaturation), the conductivity increases with increasing temperature as a function of only temperature. This implies that a temperature-conductivity curve at a constant bulk composition bends at the saturation point as shown in Figure 1. Figure 2 shows the conductivity-bulk composition relations at constant temperatures, which are obtained from the above data. When temperature is constant, the conductivity of a solution is a function of BaCl_2 concentration. Consequently, in Figure 2 the conductivity at a temperature increases with increasing BaCl_2 bulk compositions until the BaCl_2 solubility (i.e. where the bulk composition is less than the solubility), and becomes constant at a range where the bulk composition exceeds the solubility. The solubility at each temperature is given, therefore, by the value at which a composition-conductivity curve bends and becomes flat.

Errors in the measurement are produced probably in 1) every experimental step, and 2) finding a bend of each bulk composition-conductivity curve. Among them, the second source should be taken into more account. The error increases as the bulk composition-conductivity curve becomes gentle (Fig. 2). The amount of relative error is estimated to be at most 6 % for all diagrams drawn in this work.

Results and Discussion

At first solubilities of BaCl_2 in the system $\text{BaCl}_2\text{-H}_2\text{O}$ were determined under

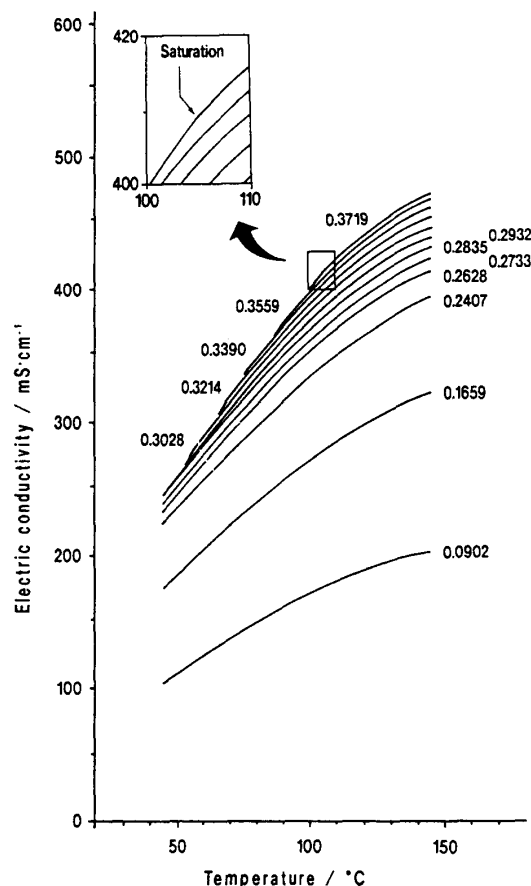


Fig. 1 Temperature-conductivity relations of BaCl₂ solutions having various bulk compositions, BaCl₂ / (BaCl₂ + H₂O). The disappearance of the last crystal of BaCl₂ occurs at the bend of each curve.

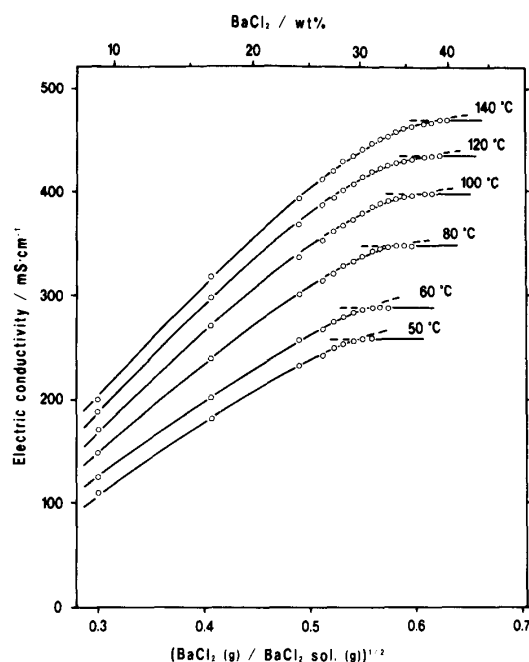


Fig. 2 Bulk composition-conductivity relations at constant temperatures in the system BaCl₂-H₂O. The solubility at each temperature is given at the bending point of each curve. The horizontal axis is represented by the square root of the bulk composition, because the conductivity of a dilute solution changes linearly with the square root of the concentration.

vapor-saturated conditions by the above stated procedure. The present results are compared with the previous data in Figure 3, and fairly agree with them in the whole range from 50 to 140°C.

At a constant temperature in the ternary system BaCl₂-NaCl-H₂O (Table A2), the conductivity of a solution having a constant NaCl/(NaCl+H₂O) ratio also increases until the BaCl₂ saturation point, and becomes constant after the saturation. Consequently, the same determination procedure as used in the binary system is available in the ternary system. Figure 4 shows solubilities of BaCl₂ in solutions having NaCl/(NaCl+H₂O) weight ratios of 0, 0.05, 0.10 and 0.15. Figure 4 also shows the results reported by Precht and Wittjen⁵⁾, whose NaCl/(NaCl+H₂O) ratios were 0 and 0.09. Except the data at the temperatures ranging from 60 to 80°C, and with the NaCl/(NaCl+H₂O) ratio of 0.09, the present data scarcely agree with their result both in the values and the slopes, and are generally lower than them. The reason of this difference is not clear at present. It should be mentioned, however, that Precht and Wittjen⁵⁾ used the quench method in which the solution was separated from a mixture of water and

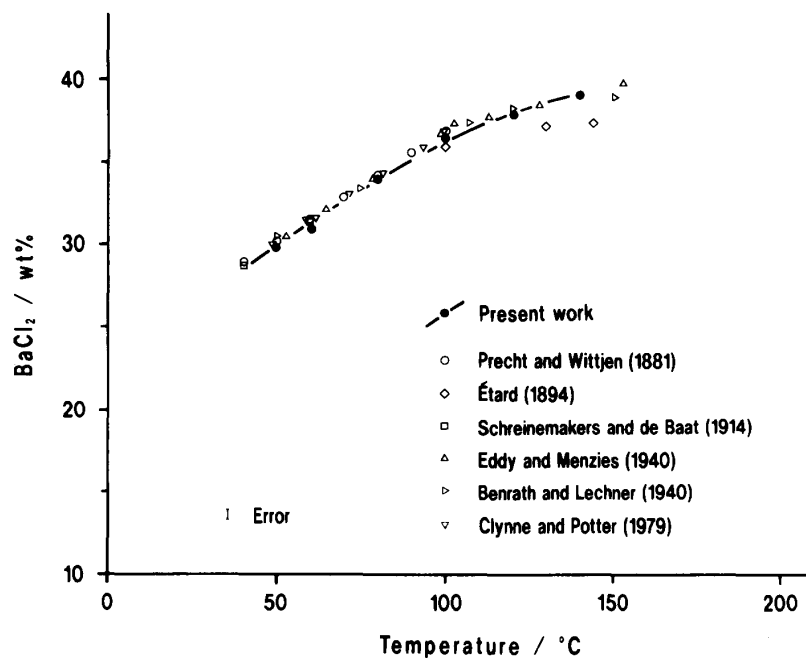


Fig. 3 Solubilities of BaCl_2 in vapor-saturated water.

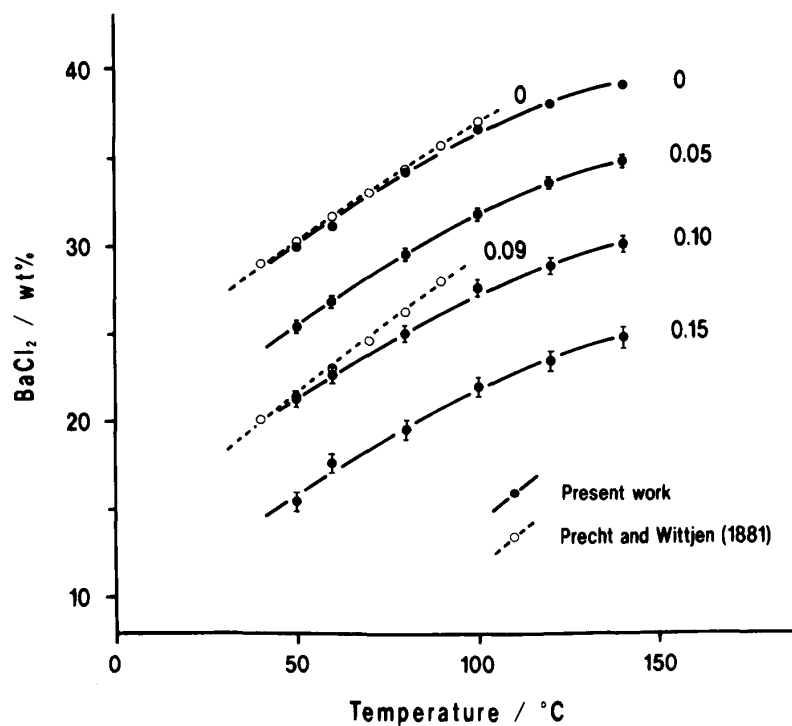


Fig. 4 Solubilities of BaCl_2 in the system $\text{BaCl}_2\text{-NaCl-H}_2\text{O}$ having $\text{NaCl}/(\text{NaCl}+\text{H}_2\text{O})$ weight ratios of 0, 0.05, 0.10 and 0.15, and those reported by Precht and Wittjen⁵⁾ for the $\text{NaCl}/(\text{NaCl}+\text{H}_2\text{O})$ weight ratio of 0 and 0.09.

salt crystals by using a paper filter, and that if small crystals passed through the filter, the obtained solubility would be higher than the true value.

Figure 5 shows solubilities of BaCl₂ at constant KCl/(KCl+H₂O) ratios ranging from 0 to 0.15 (Table A3). Schreinemakers and de Baat⁶⁾ measured solubilities of BaCl₂ in KCl solutions at 40 and 60°C. However, the data cannot be compared with the present result, because all their KCl/(KCl+H₂O) weight ratios are higher than 0.25.

The same procedure has been applied to the quaternary system BaCl₂-NaCl-KCl-H₂O. Figure 6 shows solubilities of BaCl₂ in the NaCl+KCl solutions as functions of temperature and (NaCl+KCl)/(NaCl+KCl+H₂O) ratios ranging from 0 to 0.15, where the NaCl/(NaCl+KCl) weight ratio in each solvent is 0.5. When the (NaCl+KCl)/(NaCl+KCl+H₂O) ratio of the solvent is 0.15, the conductivity does not change remarkably with an increase of BaCl₂ concentration (Table A4). Therefore, determining a saturation point at each temperature is relatively difficult, and hence the accuracy of the solubility data is low. In this case, the relative error is 6 % as stated previously.

Figure 7 is a stereopair showing a liquidus surface BaCl₂·2H₂O in the system BaCl₂-NaCl-KCl-H₂O at 50°C and 140°C. Figure 8 shows the solubility of BaCl₂ in NaCl, KCl and NaCl+KCl solutions having the KCl/(NaCl+KCl) weight ratios of 0, 0.5 and 1 at constant temperatures. These diagrams show that solubilities of BaCl₂ at a constant temperature decrease with increasing concentrations of the other salts, and that the solubilities depend largely on the NaCl concentration, and less on the KCl concentra-

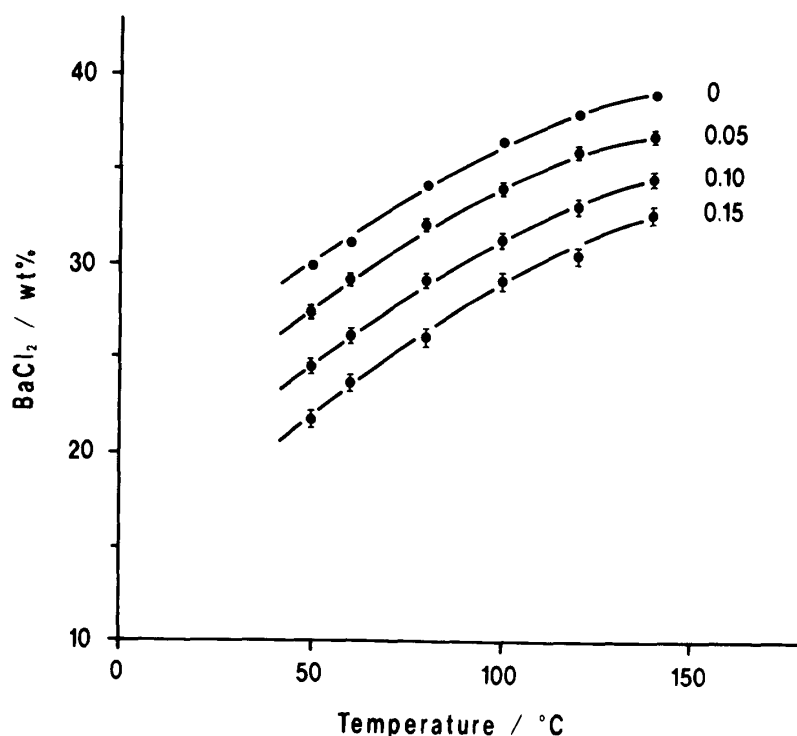


Fig. 5 Solubilities of BaCl₂ in the system BaCl₂-KCl-H₂O having KCl/(KCl+H₂O) weight ratios of 0, 0.05, 0.10 and 0.15.

tion.

According to Figure 8, the BaCl_2 solubility linearly increases with increasing $\text{KCl}/(\text{NaCl}+\text{KCl})$ ratio, when temperature and $(\text{NaCl}+\text{KCl})$ concentration are constant. This fact indicates that solubilities of some salts in a quaternary system can be estimated from data of corresponding ternary systems. It is clarified after the measurement, however, whether a solubility can be linearly estimated or not. It is

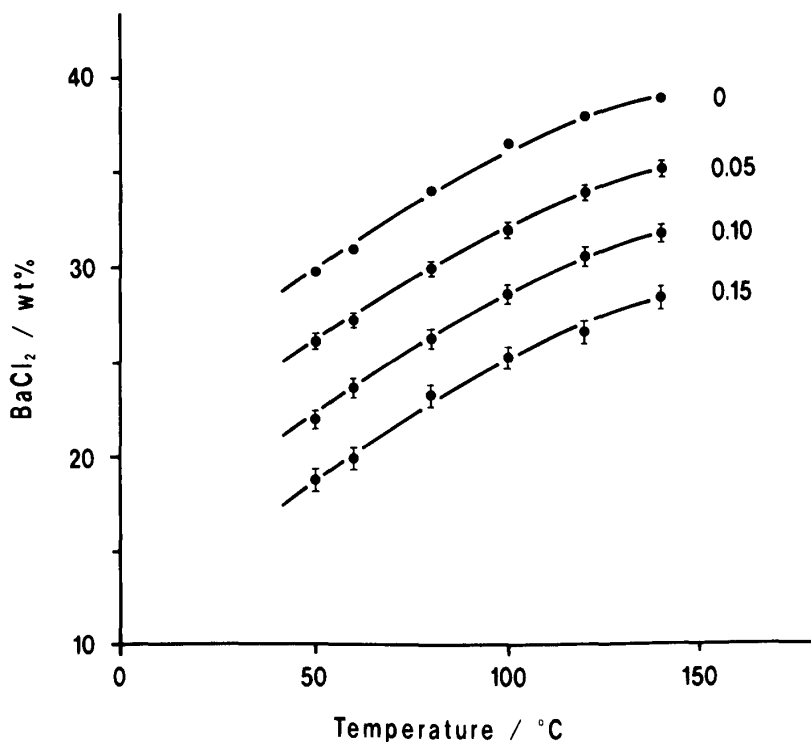


Fig. 6 Solubilities of BaCl_2 in the system $\text{BaCl}_2\text{-NaCl-KCl-H}_2\text{O}$ having $(\text{NaCl}+\text{KCl})/(\text{NaCl}+\text{KCl}+\text{H}_2\text{O})$ weight ratios of 0, 0.05, 0.10 and 0.15, and a $\text{KCl}/(\text{NaCl}+\text{KCl})$ weight ratio of 0.5.

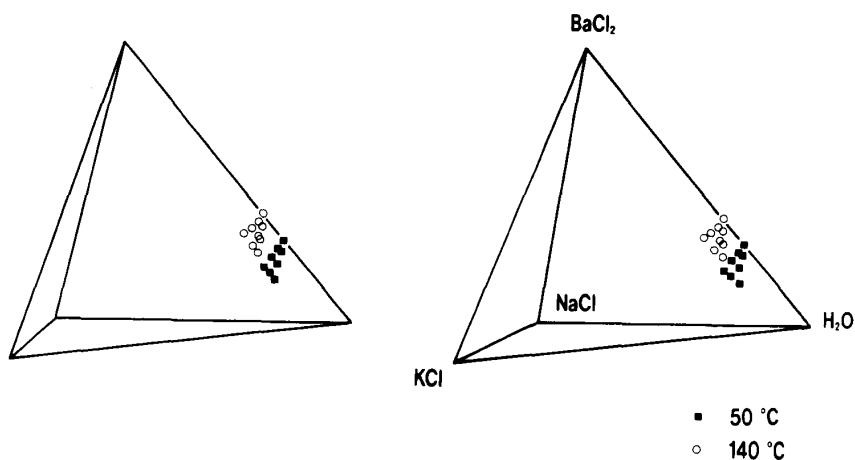


Fig. 7 A stereopair of a tetrahedral diagram showing the $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$ liquidus surface in the system $\text{BaCl}_2\text{-NaCl-KCl-H}_2\text{O}$.

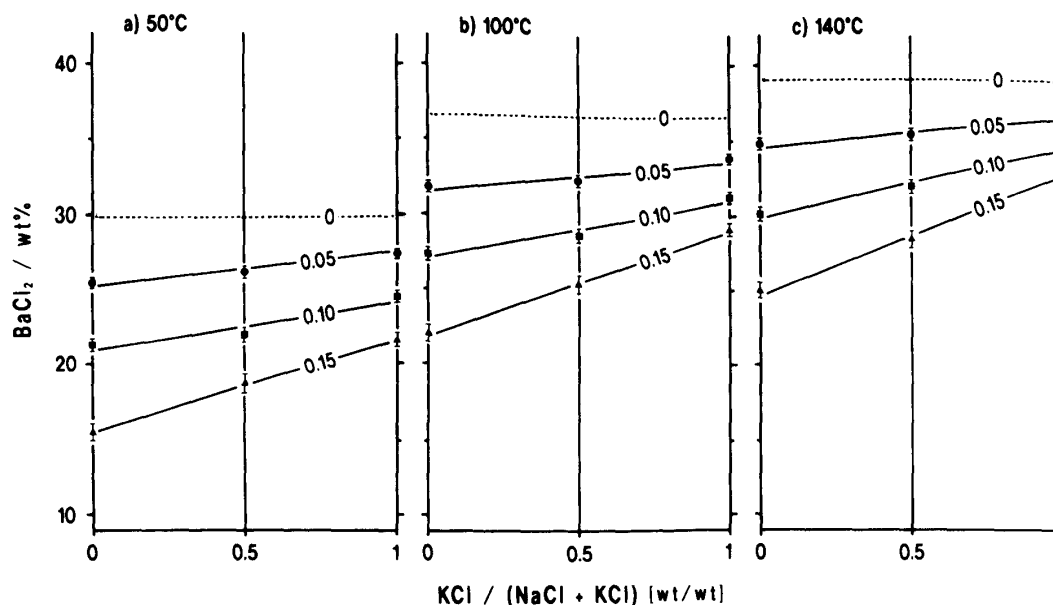


Fig. 8 Solubilities of BaCl₂ as a function of KCl/(NaCl+KCl) weight ratios : a) 50°C, b) 100°C and c) 140°C. The parameter is NaCl, KCl and NaCl+KCl weight fractions in solvents.

concluded, therefore, that the measurement in a quaternary system is always necessary in order to check this point.

As stated above, the solubility measurement in a quaternary system is necessary, even though an accurate solubility can be estimated from data in ternary systems. The present work shows that the electric conductivity method is available for determining solubilities in a quaternary system. However, the present work dealt with a quaternary system containing only one anion species. We should check the availability to systems involving two anion species at the next step.

Conclusions

Solubilities of BaCl₂ (exactly BaCl₂ · 2H₂O) in NaCl, KCl and NaCl+KCl solutions have been determined at temperatures from 50 to 140°C under vapor-saturated conditions with relative errors less than 6%. When temperature is constant, the solubilities decrease with an increase of NaCl, KCl and NaCl+KCl concentrations of the solvent, and depend largely on the NaCl concentration and less on the KCl one. On the other hand, the solubilities in each system increase with increasing temperature, when the concentration of the solvent is constant. The present result suggests that solubilities of some salts in a quaternary system can be linearly estimated from data of corresponding ternary systems with a some amount of errors. However, the solubility measurement in a quaternary system is always necessary in order to check whether the solubilities can be estimated on the basis of data in the simpler systems or not.

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Appendix

Table A 1 Bulk compositions (wt. %), temperatures (T in °C), pressures (p in kPa) and conductivities (κ in mS/cm) in the system BaCl₂-H₂O.

T	p	κ	p	κ	p	κ	p	κ	p	κ	p	κ	p	κ	p	κ		
<i>BaCl₂</i>	9.02		16.56		24.07		26.28		27.33		28.35		29.32		30.28		31.23	
50	16	110	15	183	10	233	16	243	7	250	10	254	2	257	8	258	3	258
60	26	125	22	203	17	257	20	268	11	274	15	279	7	283	12	286	8	288
70	38	138	30	223	24	280	29	293	19	298	21	302	12	307	20	311	15	314
80	48	149	40	241	35	302	40	315	30	321	33	329	22	333	31	338	25	343
90	64	158	52	257	48	322	54	336	41	343	47	348	38	353	43	357	39	361
100	84	172	74	272	78	337	72	354	60	362	66	367	57	373	60	380	57	385
110	110	180	98	285	97	357	100	352	87	379	92	385	83	391	89	395	83	402
120	145	189	135	299	130	368	135	387	120	394	127	401	118	407	123	415	117	420
130	194	194	172	309	170	385	175	402	160	409	167	416	156	422	160	427	160	434
140	248	201	220	319	225	393	223	412	208	420	212	430	200	435	201	442	205	447
<i>BaCl₂</i>	32.14		33.47		33.90		34.76		35.59		37.19		37.96		38.71		39.80	
50	7		8		14		7		15		17		11		16		10	
60	12	288	12	287	19		12		20		20		17		20		14	
70	20	316	19	318	26	318	20		27		30		22		29		22	
80	30	345	28	347	37	348	29	348	38	347	40		33		39		33	
90	43	364	42	366	49	368	41	371	50	372	52	372	45		50		43	
100	60	388	58	392	67	393	60	395	68	396	69	398	62	398	67		59	
110	87	403	86	405	92	409	83	411	93	413	93	416	87	416	89	416	82	
120	123	423	125	426	118	429	113	430	127	432	138	434	117	436	113	436	110	436
130	160	436	151	438	152	442	144	446	165	448	162	450	150	452	142	453	144	452
140	209	451	192	453	192	459	190	461	212	463	208	466	192	467	189	469	184	469

Table A 3 Bulk compositions (wt. %), temperatures (T in °C), pressures (p in kPa) and conductivities (κ in mS/cm) in the system $BaCl_2$ -KCl- H_2O .

T	p	κ	p	κ	p	κ	p	κ	p	κ	p	κ	p	κ	p	κ		
KCl/(KCl+H ₂ O)=0.05 [wt/wt]																		
<i>BaCl₂</i>	8.17	15.12	21.08	23.24	25.28	26.74	27.68	28.14	29.04									
50	19	182	19	231	12	263	17	272	12	280	13	283	10	285	19	284	17	
60	25	203	25	257	18	291	22	300	18	308	20	312	16	315	23	316	22	318
70	32	218	32	280	26	315	30	322	25	334	28	338	23	340	30	341	30	343
80	45	240	44	302	38	340	42	352	37	360	39	364	34	366	42	370	40	372
90	60	254	59	318	52	356	57	368	50	378	52	383	48	387	56	388	55	392
100	82	274	80	341	70	384	73	397	69	408	70	412	67	413	73	416	72	418
110	106	287	101	357	91	400	97	411	90	421	90	428	87	432	97	433	93	436
120	137	302	130	374	117	421	122	429	115	444	117	448	114	451	123	453	120	455
130	170	306	160	383	143	430	150	443	141	454	140	461	141	465	155	466	147	471
140	230	317	215	395	210	445	205	457	205	471	200	474	200	482	202	485	202	488
<i>BaCl₂</i>	29.93	30.76	31.64	32.86	34.81	36.96												
50	17	14	11	15	13	12												
60	22	318	20	18	21	20	18											
70	30	344	27	345	26	344	28	26	26									
80	41	375	38	375	38	376	40	376	40	38								
90	58	394	51	397	50	398	52	399	50	399	48							
100	75	419	68	421	69	425	70	427	70	427	68							
110	97	439	89	441	88	443	94	444	90	445	85	442						
120	122	455	113	456	110	458	114	461	112	463	106	463						
130	150	474	139	476	138	479	142	481	140	484	135	482						
140	198	493	195	493	198	496	187	500	190	502	180	504						
KCl/(KCl+H ₂ O)=0.10 [wt/wt]																		
<i>BaCl₂</i>	17.01	21.46	23.51	24.50	25.46	26.39	27.30	28.19	29.48									
50	15	313	18	325	15	330	12	332	19	332	11	332	22	19	16			
60	20	344	24	363	18	366	19	369	22	370	19	370	28	370	21	21		
70	29	374	31	391	26	397	27	400	32	402	27	403	32	406	29	406	29	
80	40	407	42	423	37	430	38	431	43	433	37	435	45	437	40	439	40	439
90	55	434	58	452	50	458	51	460	53	463	50	464	60	466	55	468	52	469
100	75	463	77	480	69	490	71	493	77	496	66	497	78	499	74	500	70	500
110	96	482	99	500	90	508	91	510	97	513	89	515	100	518	93	519	90	522
120	123	498	125	522	119	530	120	534	110	535	112	538	128	541	120	546	115	547
130	155	516	152	539	149	547	150	551	129	554	140	556	155	560	152	562	140	566
140	210	534	200	559	200	567	199	571	205	574	200	577	199	579	195	583	185	587
<i>BaCl₂</i>	30.73	31.94	32.71	33.47	34.22	34.96	35.67											
50	20	28	18	20	20	20	17	16										
60	26	32	20	23	24	20	23	23										
70	30	38	29	30	30	28	37	37										
80	42	437	50	39	40	41	38	47										
90	56	471	63	471	50	52	54	40	60									
100	73	501	78	501	63	499	68	69	63	72								
110	92	524	98	525	78	526	83	526	87	83	88							
120	115	548	127	550	98	551	105	552	102	552	98	107						
130	140	568	155	571	133	572	136	573	130	573	130	573	136					
140	200	588	185	590	185	591	190	591	185	592	180	592	185	591				
KCl/(KCl+H ₂ O)=0.15 [wt/wt]																		
<i>BaCl₂</i>	19.04	19.70	21.70	22.72	23.71	24.20	26.08	26.98	28.32									
50	10	350	20	352	18	358	16	358	11	18	18	20	20	20	6			
60	16	382	27	385	22	390	22	393	18	393	24	388	25	26	10			
70	24	408	34	412	30	420	30	424	25	426	32	429	34	429	32	15		
80	34	439	45	442	42	453	40	458	35	461	42	464	44	467	44	468	25	468
90	48	466	60	470	56	479	55	484	50	486	55	488	58	495	58	496	38	497
100	68	494	78	497	74	508	72	514	68	517	74	518	72	525	75	526	52	527
110	90	516	102	520	95	530	94	535	88	538	94	541	94	547	96	549	72	552
120	113	542	124	547	122	556	120	562	113	565	115	565	116	574	115	575	92	577
130	144	561	155	564	152	574	148	578	144	581	140	584	142	591	140	594	116	598
140	195	576	198	580	199	589	198	585	186	603	190	605	182	616	182	619	180	622
<i>BaCl₂</i>	29.17	30.00	30.41	30.81	31.21	31.99	33.64											
50	16	8	20	12	20	14	10											
60	22	12	28	18	26	20	15											
70	30	20	36	26	32	28	24											
80	40	30	44	38	44	38	34											
90	52	497	42	56	48	60	52	46										
100	70	528	58	528	74	62	74	66										
110	90	554	76	554	96	554	80	92	82	78								
120	112	578	95	579	108	580	96	580	112	579	102	98						
130	134	599	122	601	134	602	130	602	138	604	130	604	124	603				
140	180	624	172	625	185	626	178	627	185	628	172	629	168	629				

Table A 4 Bulk compositions (wt. %), temperatures (T in °C), pressures (p in kPa) and conductivities (κ in mS/cm) in the system BaCl₂-NaCl-KCl-H₂O.

T	p	κ	p	κ	p	κ	p	κ	p	κ	p	κ	p	κ	p	κ
(NaCl+KCl)/(NaCl+KCl+H ₂ O)=0.05 [wt/wt]																
BaCl ₂	8.25		15.24		21.25		22.35		23.42		24.46		25.47		26.45	28.35
50	22	167	12	219	17	255	10	257	18	261	8	266	19	270	12	270
60	27	184	17	240	20	283	16	285	25	290	12	297	23	300	19	302
70	37	201	25	266	30	304	22	310	32	314	20	318	30	322	27	325
80	48	219	34	285	40	330	35	337	42	341	31	347	41	351	38	353
90	63	235	49	307	55	346	48	355	59	361	39	366	57	371	51	374
100	83	252	67	324	62	372	67	380	77	386	44	387	77	397	70	397
110	110	265	93	342	87	388	89	397	100	404	60	409	100	415	95	419
120	137	277	119	358	128	410	116	418	130	425	105	430	130	433	123	442
130	180	288	156	372	160	420	148	430	170	438	130	444	165	450	158	455
140	225	295	208	383	205	432	194	441	208	454	185	458	205	466	200	470
BaCl ₂	29.26		30.15		31.02		32.69		35.05		36.60					
50	13		20		16		18		21		20					
60	30		26		20		26		27		33					
70	38	328	35		28		34		32		40					
80	48	358	44	358	38		40		45		52					
90	60	384	59	385	51	385	56		56		67					
100	80	409	76	410	68	410	70	410	70	407	85					
110	102	431	100	434	91	436	98	437	91	438	107	435				
120	130	451	128	455	120	458	128	461	118	465	137	463				
130	165	468	160	471	154	474	162	478	145	482	169	482				
140	203	481	200	483	197	485	198	491	190	497	203	495				
(NaCl+KCl)/(NaCl+KCl+H ₂ O)=0.10 [wt/wt]																
BaCl ₂	8.69		16.00		18.60		21.05		22.22		22.79		23.36		25.53	27.58
50	21	254	23	279	19	286	20	288	15	290	19	290	8	20	20	15
60	28	282	30	308	23	317	26	320	22	321	25	322	13	322	26	320
70	37	308	38	337	33	344	35	346	29	350	34	350	22	352	33	352
80	48	332	46	361	42	371	45	374	40	377	43	377	30	378	40	380
90	63	355	63	386	58	393	60	398	54	400	60	401	45	403	58	405
100	84	378	82	406	77	416	80	422	72	424	78	425	63	426	77	428
110	108	395	108	426	102	434	105	440	97	443	100	445	85	445	96	450
120	143	416	141	444	133	454	134	457	128	461	130	462	112	464	129	471
130	182	427	180	458	170	467	170	473	163	476	169	478	149	479	164	484
140	235	439	220	470	215	481	217	486	210	492	213	493	196	495	205	500
BaCl ₂	30.46		32.25		33.96											
50	17		20		10											
60	23		24		15											
70	30		32		23											
80	39	376	41		34											
90	55	405	52	402	47											
100	73	431	69	422	64											
110	94	454	86	454	88											
120	125	476	113	477	114	472										
130	160	490	145	491	145	490										
140	202	505	195	508	190	506										
(NaCl+KCl)/(NaCl+KCl+H ₂ O)=0.15 [wt/wt]																
BaCl ₂	11.65		14.15		15.94		17.42		18.53		19.61		20.66		21.68	22.69
50	12	331	22	338	18	342	15	344	16	345	20	346	10	346	19	15
60	19	370	28	376	20	380	20	381	20	382	28	383	15	384	23	384
70	28	398	35	404	28	410	30	434	29	416	37	418	23	419	29	420
80	39	433	47	437	40	448	41	449	40	452	49	452	34	454	41	455
90	54	463	60	472	55	476	57	480	54	481	63	483	48	485	56	487
100	74	498	80	505	72	513	75	514	72	517	80	518	66	520	73	522
110	96	520	102	529	95	536	98	540	92	541	104	544	86	546	93	548
120	123	546	130	558	119	563	127	565	118	568	127	569	110	573	117	574
130	150	564	160	575	152	582	155	585	145	588	155	591	135	593	140	595
140	200	581	200	588	195	603	195	605	195	606	193	608	180	612	185	614
BaCl ₂	23.66		24.62		25.72		26.61		27.49		28.35		29.20			
50	22		6		18		18		15		10		10			
60	30		10		24		24		20		15		15			
70	35		13		31		32		28		23		22			
80	43	456	25	456	42		39		36		28		33			
90	57	489	38	489	55	488	40		40		30		45			
100	73	523	54	524	73	525	67	525	63		60		60			
110	92	551	73	552	94	554	82	554	78	552	77		80			
120	112	578	95	560	120	561	115	562	103	562	98		104			
130	134	599	118	601	150	602	139	603	133	604	133	603	139			
140	180	618	175	621	182	621	178	621	180	623	172	624	175	624		