

# Calculation of Salt Activities in Molten Salt Hydrates Applying the Modified BET Equation, I: Binary Systems\*

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**Summary.** On the basis of the modified BET model according to Stokes and Robinson, an equation for the calculation of salt activities in molten salt hydrates has been derived. The equation is used to describe successfully the liquidus curves of the hydrates of  $\text{MgCl}_2$ ,  $\text{Mg}(\text{NO}_3)_2$  and  $\text{CaCl}_2$  in  $T$ - $x$  diagrams. A promising feature of the model is the small number of adjustable parameters and its extrapolative power.

**Keywords.** Molten hydrates; Phase diagrams; Solid-liquid equilibria; Thermodynamic activities.

## **Berechnung von Salz-Aktivitäten in geschmolzenen Salzhydraten mittels der modifizierten BET-Gleichung, 1. Mitt.: Binäre Systeme**

**Zusammenfassung.** Auf der Grundlage des modifizierten BET-Modells nach Stokes und Robinson wurde eine Gleichung zur Berechnung der Salzaktivitäten in geschmolzenen Salzhydraten abgeleitet. Mit der Gleichung wurden die Liquiduslinien der Hydrate von  $\text{MgCl}_2$ ,  $\text{Mg}(\text{NO}_3)_2$  und  $\text{CaCl}_2$  im  $T$ - $x$ -Diagramm beschrieben. Besondere Charakteristika des Modells sind die kleine Anzahl adjustierbarer Parameter und die Möglichkeiten zu Extrapolationen.

## **Introduction**

The phase diagrams of a large number of salt–water systems are characterized by a series of congruently or incongruently melting hydrates  $M_nX_m \cdot R \text{H}_2\text{O}$  with varying values of  $R$ . As long as  $R$  does not exceed the coordination number  $z$  of the cation ( $z = 4 \dots 9$ ), the water of hydration is coordinated around the cations [1]. With  $R < 6$ , for hydrates of salts like  $\text{MgCl}_2$ ,  $\text{Mg}(\text{NO}_3)_2$  or  $\text{CaCl}_2$  also in molten state the water molecules are quite firmly bound within the first coordination sphere of the cations. Arguments for this structural situation came from spectroscopic [2–4] and physico-chemical investigations [5, 6]. Also the very low water activities of these hydrous melts support this view. For example, molten  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$  at  $120^\circ\text{C}$  (that

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\* Dedicated to o. Univ.-Prof. Dipl.-Ing. Dr. mont. Heinz Gamsjäger on the occasion of his 60th birthday

is near the metastable congruent melting point) has a water activity of  $a_w = 0.127$ . Even at a temperature of 250 °C  $a_w$  of this melt is not higher than 0.284 [7]. According to Raoult's law on an ionized basis a value of 0.67 would be expected.

Independent from the existence of crystalline hydrates, the term "molten salt hydrate" is useful to depict a range on the concentration scale salt–water, where essentially all water is coordinated to ions (mostly cations) and the contact interactions ion–water are predominant [8]. Passing the concentration scale from the range of solutions to molten hydrates, characteristic changes in the thermodynamic properties occur, especially in respect to the partial molar properties [8–10]. The sharpness of this "transition behaviour" and its location on the concentration scale is correlated with the coordination number and the hydrophilic character of the cations [8]. Systems of the type  $MNO_3-H_2O$  with  $M = Na, K, Rb, Cs, NH_4,$  and  $Ag$  exhibit no pronounced transition behaviour. In these cases activities can be described covering the complete concentration scale by means of simple non-electrolyte mixing models extended by a Debye-Hückel term [11, 12]. However, this approach fails in the case of stronger hydrating salts like  $MgCl_2$  or  $CaCl_2$ . The widely used ion-interaction model of Pitzer as a typical solution model is also not suited to describe activity data of molten hydrates [13].

On the other hand, already 1948 Stokes and Robinson [14] proposed a modified form of the BET equation [15] to describe the water activities of very concentrated electrolyte solutions. The equation contains only two adjustable parameters. Later this equation was frequently used to describe the solvent activity of hydrous [5, 16–19] as well as of non-aqueous melts [20]. All results confirm that as long as the solvent activity remains below approx. 0.5, the model is able to reproduce the solvent activities of all binary systems with reasonable accuracy, independent from the individual hydrating properties of the electrolytes.

There is considerable need for modelling and predicting solid-liquid equilibria in systems of melting salt hydrates, for instance for heat storage applications [21]. Calculations of salt activities in the liquid phase by means of a suited model equation represent a presumption for this purpose. In spite of the fact that the BET equation is the most successful model used in describing water activities of molten hydrates, it was never used to calculate salt activities. The reason has to be seen in some inherent difficulties in deriving salt activities from this model by means of the Gibbs–Duhem relation.

It is the aim of this paper to derive an equation for the calculation of salt activities from water activities of molten hydrates on the basis of the BET model. The usefulness of the equation obtained will be demonstrated by calculating liquidus curves of salt hydrates in binary salt–water systems.

## Results and Discussion

### *Modified BET Equation*

Usually the BET equation is formulated in its linearized form, where either  $m$ , the molality of electrolyte (Eq. 1a), or  $x_w$ , the mole fraction of water (Eq. 1b), is chosen

as the concentration variable,

$$\frac{ma_w}{55.51(1-a_w)} = \frac{1}{cr} + \frac{(c-1)a_w}{cr}, \quad (1a)$$

$$\frac{(1-x_w)a_w}{x_w(1-a_w)} = \frac{1}{cr} + \frac{(c-1)a_w}{cr}, \quad (1b)$$

$r$  and  $c$  represent the adjustable parameters.

Having in mind the physical background of an adsorption layer,  $r$  is interpreted as a hydration number expressing the maximum number of water molecules, which can adhere at the surface of a molecular unit of the salt. The parameter  $c$  is related to  $\Delta E$ , the difference between the energy of adsorption at the salt surface  $E_d$  and the condensation energy of pure water  $E_L$ ,

$$c = \exp(-\Delta E/RT) = \exp\{-(E_d - E_L)/RT\}. \quad (2)$$

The magnitude of  $r$  and  $\Delta E$  correlates well with the general hydration tendencies of electrolytes.

As mentioned above, the typical validity range of Eq. (1) is limited to mixtures of water activities below 0.5, although some systems are known, where the activity data follow Eq. (1) up to values of  $a_w = 0.7$  [22]. In contrast to the limitation toward more dilute solutions, there exists no definite evidence for systematic deviations from the BET equation on the molten salt side of the concentration scale. Measurements down to water activities of 0.03 fulfilled Eq. (1). Only Richardson and Kurtz [23] noticed deviations at water activities below 0.02 determined in highly supersaturated LiI solutions. The authors used a bithermal isopiestic method adapted for sample drops of microscopic sizes (10  $\mu\text{m}$  in diameter). The drops carried a negative electrical charge and were weighed in the vapour of supercooled pure water by means of an electrodynamic balance. Thermal equilibrium was assumed between the drop and its surrounding held at a temperature of 23  $^\circ\text{C}$ . But, at water activities below 0.02 the absolute vapour pressure is lower than 50 Pa. At those low pressures it is difficult to reach thermal equilibrium. Considering the critical experimental conditions in that range of water activity the results are not convincing to state a validity limit of the BET equation toward the molten salt side of the concentration scale.

### *Salt Activities from BET Equation*

The water activity is the experimentally available activity quantity for most of the concentrated solutions and molten hydrates. In order to obtain salt activities the Gibbs–Duhem equation has to be applied, which for a binary salt–water system at constant pressure and temperature is given by

$$x_w d \ln a_w + x_s d \ln a_s = 0. \quad (3)$$

The subscript  $s$  denotes the salt component. Eq. (3) has to be integrated to calculate the salt activity at a given mole fraction of water

$$\int_{\ln a_s(x_w=0)}^{\ln a_s(x_w)} d \ln a_s = - \int_{\ln a_w(x_w=0)}^{\ln a_w} \frac{x_w}{x_s} d \ln a_w. \quad (4)$$

Since the BET equation cannot be used in dilute solutions, the alternative reference state, the pure (supercooled) liquid salt ( $X_w = 0$ ) has to be chosen. Unfortunately, this choice of reference state yields an infinite lower limit of the integral on the right-hand side of Eq. (4). Also several transformations of the integral do not improve the situation. In addition to this infinity problem, also the substitution of  $d \ln a_w$  by an analytic expression from the BET equation is complicated because of the quadratic form of the function

$$0 = \frac{1}{1-c} + a_w \frac{(c-2) - (cr)/R}{1-c} + (a_w)^2. \quad (5)$$

Both solutions of Eq. (5) can be relevant depending on the actual values of  $a_w$  and the parameter  $c$ .

Obviously, these difficulties prevented the use of the BET model to calculate salt activities. Abraham [19, 24] tried to avoid these complications by deriving an equation for the salt activity on the basis of a 2-dimensional lattice statistics with a physical background similar to the BET model. The result was an equation (Eq. 6) very similar in structure to the BET equation. However, as will be shown later, Eq. (6) is not identical with the equation, which follows from application of the Gibbs–Duhem relation,

$$\frac{\lambda(1-x_s)}{x_s(1-\lambda)} = \frac{r}{c} + \frac{r(c-1)}{c} \lambda, \quad (6)$$

$$\lambda = a_s^{(1/r)}.$$

Now, it will be shown that by appropriate substitutions and transformations in Eq. (3) an integral is obtained, which yields a simple analytic expression. For this aim Eq. (3) is rearranged to Eq. (7) and the latter one is multiplied by a factor  $(1 - a_w)$ , which gives Eq. (8),

$$\frac{x_w}{(1-x_w)} \cdot \frac{da_w}{a_w} + d \ln a_s = 0, \quad (7)$$

$$\frac{x_w(1-a_w)}{(1-x_w)a_w} da_w + (1-a_w) d \ln a_s = 0. \quad (8)$$

From Eq. (8) it can easily be recognized that the factor of  $da_w$  is equal the reciprocal of the left-hand side of the BET equation (Eq. 1b). After substitution of Eq. (1b) into Eq. (8) the expression in Eq. (9) results,

$$\frac{cr}{1+(c-1)a_w} da_w + (1-a_w) d \ln a_s = 0. \quad (9)$$

Some further elementary rearrangements give rise to a differential equation, which can be integrated (Eq. 10) without the difficulties discussed above,

$$\int_{\ln a_s(x_w=0)}^{\ln a_s(x_w)} d \ln a_s = -\frac{cr}{(1-c)} \int_{a_w=0}^{a_w} \frac{1}{(a_w)^2 + a_w \frac{(c-2)}{(1-c)} + \frac{1}{(1-c)}} da_w. \quad (10)$$

The integral on the right-hand side of Eq. (10) belongs to the basic form of Eq. (11), the solution of which can be found in mathematical handbooks [25],

$$\int \frac{dx}{X} = \frac{1}{\sqrt{-\Delta}} \ln \frac{2fx + b - \sqrt{-\Delta}}{2fx + b + \sqrt{-\Delta}}, \quad \Delta < 0, \quad (11)$$

$$X = fx^2 + bx + d, \quad \Delta = 4fd - b^2. \quad (12)$$

The condition  $\Delta < 0$  is fulfilled as can be seen from inserting the corresponding coefficients into the definition of  $\Delta$ , which gives the inequality

$$\Delta = -\frac{c^2}{(1-c)^2} < 0. \quad (13)$$

Finally, performing the integration in the denoted limits yields the desired relation for the salt activity

$$\ln a_s = r \ln \frac{a_w - 1}{a_w(1-c) - 1}. \quad (14)$$

In order to get an explicit formulation as a function of  $x_w$ , or  $R$ , respectively, the solution of Eq. (5) has to be inserted into Eq. (14), from which Eq. (15) is obtained,

$$\lambda = a_s^{(1/r)} = \frac{1}{1-c} \left\{ 1 + \frac{2}{(z+1) \pm \sqrt{(1-z)^2 + \frac{4z}{c}}} \right\} \quad z = (r/R). \quad (15)$$

Let us return once more to Eq. (6) derived by Abraham. For a comparison all  $\lambda$ -terms are collected on the left-hand side, which yields the simple expression

$$K = \frac{x_w}{(1-x_w)} \cdot \frac{c}{(1-\lambda)} - \frac{r}{\lambda} = r(c-1). \quad (16)$$

The exact expression for  $K$  follows from Eq. (14) and is more complex,

$$K = \{c-1 + (1/a_w)\} \left\{ \frac{x_w}{(1-x_w)} + \frac{r}{1-(1/a_w)} \right\}. \quad (17)$$

Only at the limit  $a_w \rightarrow 0$ , both equations become identical. This means, the assumptions introduced into the statistical treatment by Abraham are more restrictive than those of the BET model.

### *Modelling of Liquidus Curves of Salt Hydrates*

The main draw-back for a quantitative description of solid-liquid equilibria in systems of molten hydrates is the lack of a suited model equation for the component activities in the liquid phase. The application of the BET model in form of its complementary Eqs. (5) and (14) can initiate progress in this respect. An attractive feature of the BET equation is the small number of adjustable parameters.

In the following the application of the BET model for this purpose will be demonstrated at the systems  $\text{MgCl}_2\text{-H}_2\text{O}$ ,  $\text{Mg}(\text{NO}_3)_2\text{-H}_2\text{O}$  and  $\text{CaCl}_2\text{-H}_2\text{O}$ , selected as typical examples of systems forming fusible hydrates.

In Figs. 1-3 the calculated liquidus curves and the corresponding experimental

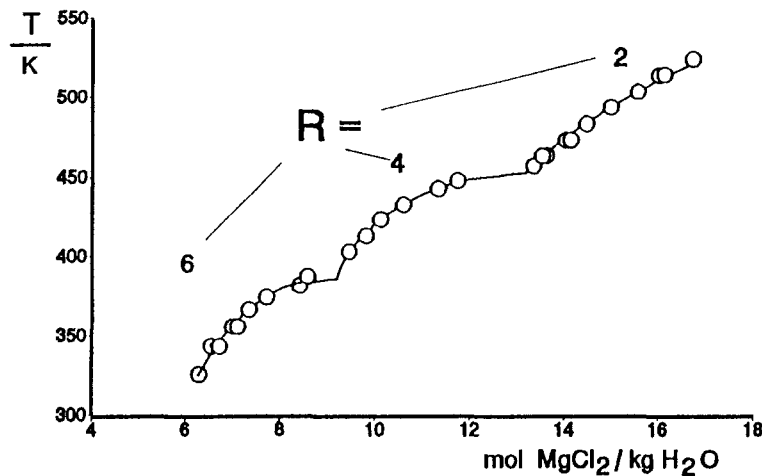


Fig. 1. Liquidus curves of  $\text{MgCl}_2 \cdot R \text{H}_2\text{O}$  with  $R = 6, 4, 2$ ; lines: calculated; points: experimental data [31, 32]

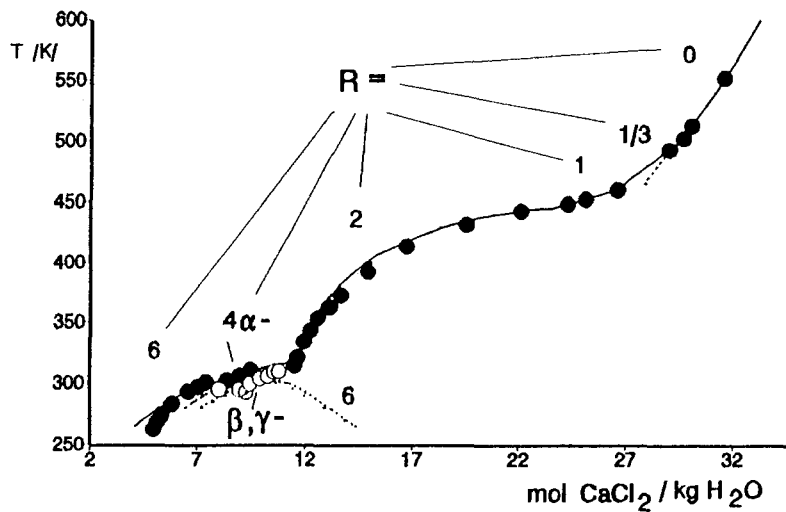


Fig. 2. Liquidus curves of  $\text{CaCl}_2 \cdot R \text{H}_2\text{O}$  with  $R = 6, 4, 2, 1, 1/3, 0$ .  $\beta, \gamma$  denote metastable tetrahydrates; lines: calculated (— stable, ---- metastable equilibria); points: experimental data [33] (● stable, ○ metastable equilibria)

data are compared. Calculations are based on

$$\ln K_s = \ln a_{MX_n}^{(l)} + R \ln a_w^{(l)}, \quad (18)$$

where  $K_s$  represents the solubility constant of a hydrate  $\text{MX}_n \cdot R \text{H}_2\text{O}$  at a given temperature and the superscript (l) denotes the liquid phase. As a consequence of the BET model, ionic dissociation is formally not considered in Eq. (18) and the reference state of  $a_{MX_n}^{(l)}$  is the anhydrous pure molten salt  $\text{MX}_n$ . The calculation of activities according to Eqs. (5) and (14) requires the BET parameters within the temperature range of the considered sections of the phase diagrams. For the systems

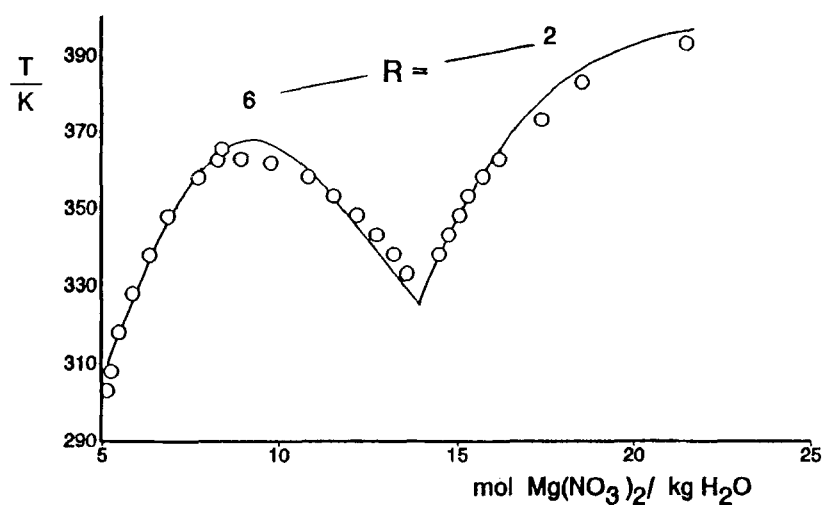


Fig. 3. Liquidus curves of  $\text{Mg}(\text{NO}_3)_2 \cdot R \text{H}_2\text{O}$ ; lines: calculated; points: experimental data [29, 30]

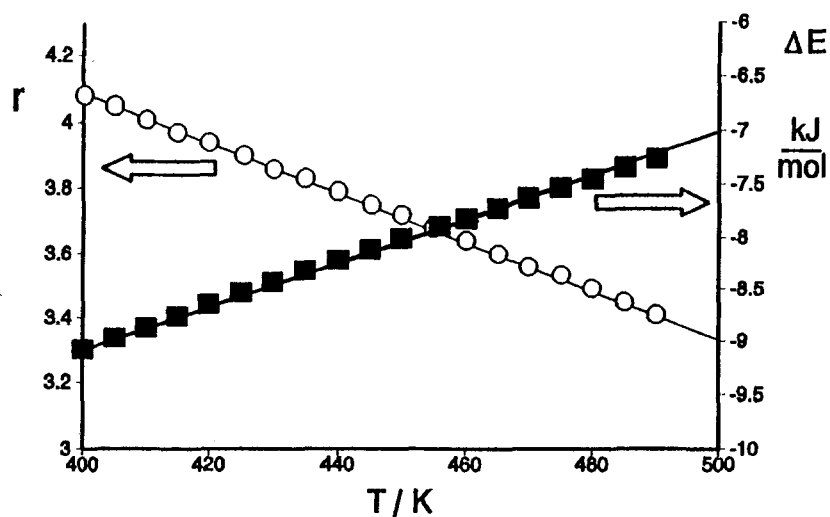


Fig. 4. Temperature dependence of the BET parameters  $r$  and  $\Delta E$  of the system  $\text{CaCl}_2\text{-H}_2\text{O}$

Table 1. Coefficients  $a_0, a_1$  of the empirical function  $(r, \Delta E) = a_0 + a_1 T$  describing the temperature dependence of the BET parameters  $r$  and  $\Delta E$

System	$r$		$\Delta E$ (kJ/mol)		Ref.
	$a_0$	$a_1 \cdot 10^3$	$a_0$	$a_1 \cdot 10^2$	
$\text{MgCl}_2\text{-H}_2\text{O}$	8.225	-6.137	-23.665	2.457	[26]
$\text{CaCl}_2\text{-H}_2\text{O}$	7.067	-7.453	-17.175	2.033	[27]
$\text{Mg}(\text{NO}_3)_2\text{-H}_2\text{O}$	5.579	-	-	-3.1348	[28-30]

**Table 2.** Solubility constants  $K_s$  of solid phases according to Eq. (18)

Solid Phase	$T_1$ [K]	$\ln K_s$	$T_2$ [K]	$\ln K_s$
$\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$	326.2	-37.884	374.6	-29.216
$\text{MgCl}_2 \cdot 4\text{H}_2\text{O}$	403.2	-21.353	443.0	-17.641
$\text{MgCl}_2 \cdot 2\text{H}_2\text{O}$	463.0	-11.318	493.0	-10.021
$\text{CaCl}_2 \cdot 6\text{H}_2\text{O}$	293.1	-26.223	302.1	-24.417
$\alpha\text{-CaCl}_2 \cdot 2\text{H}_2\text{O}$	303.1	-21.603	319.1	-18.595
$\beta\text{-CaCl}_2 \cdot 4\text{H}_2\text{O}$	303.1	-20.886	*	
$\gamma\text{-CaCl}_2 \cdot 4\text{H}_2\text{O}$	303.1	-20.810	*	
$\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$	319.1	-14.561	443.1	-7.335
$\text{CaCl}_2 \cdot \text{H}_2\text{O}$	449.1	-5.018	460.1	-4.714
$\text{CaCl}_2 \cdot 1/3\text{H}_2\text{O}$	460.1	-3.284	503.1	-2.808
$\text{CaCl}_2$	503.1	-2.174	553.1	-1.908

\* The same linear dependence of  $\ln K_s$  on  $(1/T)$  as for  $\alpha\text{-CaCl}_2 \cdot 4\text{H}_2\text{O}$  was assumed

$\text{MgCl}_2\text{-H}_2\text{O}$  [26] and  $\text{CaCl}_2\text{-H}_2\text{O}$  [27] BET parameters have been derived from recent vapour pressure measurements over a broad interval of temperature. A plot of  $r$  and  $\Delta E$  shows a linear dependence on temperature as is demonstrated in Fig. 4 for the system  $\text{CaCl}_2\text{-H}_2\text{O}$ . In Table 1 the coefficients of the corresponding empirical equations are listed. In the case of  $\text{Mg}(\text{NO}_3)_2\text{-H}_2\text{O}$  the parameters were estimated from data of larger uncertainty and covering a smaller interval of temperature (30–85 °C). In particular, data of Ewing et al. [28], who measured the vapour pressure above solutions (melts) saturated in respect to the solid phase, were combined with solubility data from elsewhere [29, 30]. Therefore, the temperature dependence of  $r$  was neglected and the temperature coefficient of  $\Delta E$  in Table 1 results from a constant value of  $c = 43.40$  (see Eq. (2)).

Solubility constants of the various solid phases are given in Table 2. They were calculated using Eq. (18) at selected points of the experimental  $T$ - $x$  diagrams. To account for the temperature dependence,  $\ln K_s$  of the systems  $\text{MgCl}_2\text{-H}_2\text{O}$  and  $\text{CaCl}_2\text{-H}_2\text{O}$  were calculated at two different temperatures  $T_1$  and  $T_2$ . Interpolation and extrapolation were carried out using the linear dependence of  $\ln K_s$  on the reciprocal absolute temperature.

Only for  $\text{Mg}(\text{NO}_3)_2\text{-H}_2\text{O}$  an empirical temperature function (Eq. 19a, b) was estimated by least square fitting of  $\ln K_s$  calculated at all available experimental liquidus points,

$$\ln K_s(\text{Mg}(\text{NO}_3)_2 \cdot 2\text{H}_2\text{O}) = -6.402 - 1999.8/T, \quad (19a)$$

$$\ln K_s(\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}) = -13.739 - 3570.9/T. \quad (19b)$$

As can be seen from Figs. 1–3, the liquids curves calculated from the data in Tables 1 and 2 coincide quite well with the experimental data. Some deviation is observed at the liquidus curve of  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  (Fig. 3). Certainly, the approximation of temperature independent BET parameters is too rough, but for essential improvements reliable vapour pressure measurements within the range of molten hydrates are required.



For the system  $\text{MgCl}_2\text{-H}_2\text{O}$  the agreement is within the uncertainty of the solubility data [31, 32].

A larger number of hydrates exists in the system  $\text{CaCl}_2\text{-H}_2\text{O}$ . Special attention should be paid to the crystallization branch of the dihydrate, which covers a molality range from about 11 to 25 mol  $\text{CaCl}_2/\text{kg H}_2\text{O}$ . Although the solubility constants were fixed at the limits of this concentration interval (see Table.2), the calculated line nicely fits the pronounced curvature of the liquidus curve. This gives strong support for the validity of the salt activities calculated from Eq. (14). Also the BET parameters of the system  $\text{CaCl}_2\text{-H}_2\text{O}$  were derived from vapour pressure data in a molality range of 7–22 mol/kg  $\text{H}_2\text{O}$  and temperatures between 400 K and 490 K. Therefore, those parts of the diagram in Fig. 2 with  $m > 22$  mol  $\text{CaCl}_2/\text{kg H}_2\text{O}$  and  $T > 520$  K or  $T < 400$  K, respectively, are based on an extrapolated activity model of the liquid phase. The extrapolative power of the BET model, especially toward higher concentrations, provides a solid base for calculations of metastable solid–liquid equilibria, as it was done for the tetra- and hexahydrates of calcium chloride (Fig. 2).

In the low temperature region of the diagram in Fig. 2 a divergence between calculated and experimental data becomes evident. The differences grow with decreasing molality of  $\text{CaCl}_2$ . Here the water activities exceed values of 0.5 and the BET model is no longer valid. The latter fact emphasizes that the agreement observed in the other parts of the diagram is not simply the result of suited chosen solubility constants, but due to successful described activity data, both for water and the salt components.

## Conclusions

The BET model provides a convenient basis to describe the thermodynamic activities of both components in binary salt–water systems within the region of molten hydrates. A generalization to solvents other than water, as alcohols, formamides or dimethylsulfoxide, seems possible.

An extension of this approach to ternary or multi-component systems requires mixing rules for the BET parameters. In nitrate mixtures simple additivity relations were found to be valid [16, 19]. However, introducing of such dependencies, the Gibbs–Duhem integration becomes more complex. In addition, salt activities of anhydrous molten salt mixtures in a supercooled state have to be used as a reference. These aspects will be treated in the second part of this paper.

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