Direct Production of Crystalline Boric Acid through Heterogeneous Reaction of Solid Borax with Propionic Acid: Operation and Simulation

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Abstract−The production of boric acid through reaction of borax crystals with propionic acid was investigated in batch mode. It was found that the product boric acid precipitates on the solid borax reactant. An increase in the coefficient of variation of feed crystals resulted in an increase in the time of completion of the reaction. A sharp interface model with variable bulk fluid concentration of liquid reactant was developed for simulation of the process. The analytical solution of the series of rate equations involving liquid film, solid boric acid layer and chemical reaction resistances was obtained, and a new method for simultaneous determination of kinetic parameters was established. The chemical reaction and diffusion through the solid layer of boric acid were the rate controlling mechanisms. A numerical algorithm was also developed for predicting the fractional conversion of multisize borax crystals using the mono size kinetic parameters determined according to the proposed analytical method.

Key words: Boric Acid, Reaction, Crystallization, Simulation, Sharp Interface

INTRODUCTION

In the industrial two stage process of boric acid production from sodium borates, borax ($Na_2B_4O_7 \cdot 10\ H_2O$) is reacted with sulphuric acid and the product boric acid is then recovered from the solution phase by cooling crystallization [McKetta, 1999]. In this process, the mean size of the product crystals is about $100\ \mu m$ and larger sizes cannot be attained without seeding operation. The larger boric acid crystals have lower caking tendency in the warehouse (which reduces the product loss during conservation and shipment) and many more applications in chemical and process industries [Gerhartz, 2001]. Since a large amount of propionic acid is being produced as a by-product in some petrochemical sites, the establishment of a plant for boric acid production near to the main site is economically justified [ZareNezhad, 2003]. Also, depending on the requirements, different ranges of boric acid crystals can be produced: e.g., mean sizes up to 462 and $780\ \mu m$ by using propionic acid in one stage without seeding operation.

In this work, a one stage process for production of boric acid crystals through reaction of solid borax with propionic acid solution is proposed. The reaction can be presented by the equation:

$$Na_2B_4O_7 \cdot 10\ H_2O_{(S)} + 2\ C_2H_5COOH_{(L)}$$
$$\rightarrow 2\ C_2H_5COONa_{(L)} + 4\ H_3BO_{3(S)} + 5\ H_2O \quad (1)$$
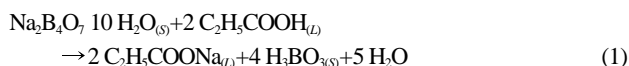
The suggested process can be used as an alternative way of production of crystalline boric acid. Effects of process variables, namely, stirrer speed, temperature, propionic acid concentration, mean size and size distribution of borax crystals on the reaction kinetics, conversion profiles and crystal size distribution of the product boric acid are discussed and a mathematical model for description of the process is presented.

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EXPERIMENTAL PROCEDURES

The crystallizer used was a 1.5 L jacketed baffled cylindrical vessel (11 cm i.d., height 16 cm) maintained at constant temperature. The contents were stirred with a six blade disk turbine impeller at a speed 15% higher than the minimum suspension speed [Baldi et al., 1978; ZareNezhad, 2003]. In a typical experiment 1 L propionic acid solution of a known concentration was charged into the vessel and the contents were heated to the desired temperature. A known amount of borax crystals, sieved in the size range of 462-$780\ \mu m$, was quickly added to the solution at the start of an experiment. Sampling was achieved by pipetting about 5 ml of the contents from the crystallizer at different time intervals and filtering it through a $0.4\ \mu m$ pore size filter paper. The pipette tip diameter was 7 mm, adequate for collecting the product crystals in the size range of 8-$1,200\ \mu m$.

Samples of the solid phase were analyzed for crystal size distribution using a multichannel Coulter Counter (Model Multisizer II) [Coulter Electronics Ltd., 1993]. The solid samples collected on the filter paper were quickly dispersed into the Coulter cell containing filtered Isoton II saturated with borax and boric acid. The samples of solids and filtrates were also weighed and analyzed to determine changes in the overall compositions of the solid and solution phases during the process. A weighed aliquot of the dried solid phase was dissolved in water and titrated against standard sulphuric acid solution for borax and against standard sodium hydroxide solution for boric acid [Jeffery et al., 1989]. Propionic acid and boric acid concentrations in the solution were determined by pH titration with standard sodium hydroxide solution and sodium propionate by sodium ion analysis using an atomic absorption spectrophotometer at a wavelength of 589 nm.

A series of experiments was carried out at different operating conditions covering the variable ranges shown in Table 1. The initial molal concentration of propionic acid (c_0) was varied from 10 to

Table 1. Ranges of run conditions for the reaction of borax crystals with propionic acid solution

| Run	c_0 (kmol/m³)	CV (dimensionless)	N (Hz)	L_B (μm)	T (K)
S1-S5	10-20	0.05	16	780	293
S6-S10	15	0.05-0.82	17	780	293
S11-S15	10	0.05	18-34	780	293
S16-S20	10	0.05	17	462-780	293
S21-S25	20	0.05	19	780	288-304

20 kmol/m³ water, stirrer speed (N) from 16 to 34 Hz, temperature (T) from 288 to 304 K, mean size of borax crystals (L_B) from 462 to 780 μm and coefficient of variations of borax crystals (CV) from 0.05 to 0.82. The loading of borax crystals, M_B was always kept at 1 kmol/m³.

MODELLING OF THE PROCESS

The experimental results confirmed that in the reaction of propionic acid solution with borax crystals, the product boric acid deposits mainly on the surface of borax crystals. This evidence suggests that application of the unreacted shrinking core model is appropriate. According to the sharp interface model [Levenspiel, 1998; Froment and Bischoff, 1990], when the product of a reaction between solid and liquid reactants precipitates as a solid layer around the solid reactant, the reactant in the solution diffuses through the liquid film surrounding the particle and then through the deposited solid product to reach the surface of the solid reactant where it reacts.

Fig. 1 shows the concentration gradients of propionic acid, the fluid reactant, from the bulk solution through the liquid film and then through the solid layer of boric acid to the surface of the borax core where it reacts to produce boric acid and sodium propionate. Boric acid crystals are assumed to form on the borax surface further thickening the product layer.

Three important stages - transportation of propionic acid from the bulk of the solution to the solid-liquid interface, diffusion of the reactant through the solid boric acid to the surface of unreacted core and chemical reaction at the surface of the solid reactant (borax) -
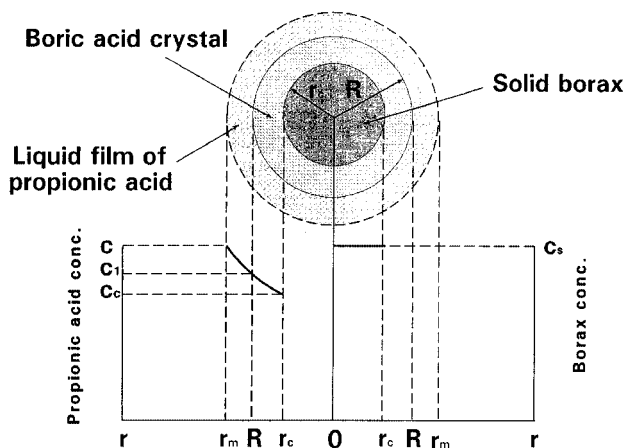
Fig. 1. Concentration profiles of the reactants according to the sharp interface model.

are considered in this study. For monosized spherical particles suspended in a constant volume reacting fluid, a general kinetic expression including three first-order steady-state processes can be obtained. Following Levenspiel [1998], the molar rate of transport of propionic acid in each of the steps can be written as follows:

(i) In the liquid film:

$$-\frac{dN_A}{dt} = k_1^*(c - c_1) \tag{2}$$

where N_A is the number of moles of propionic acid in the solution phase.

(ii) In the boric acid layer, integration of the 1st order diffusion rate equation for points of radius $r_c < r < R$ and concentration $c_c < c < c_1$ yields:

$$-\frac{dN_A}{dt}\left(\frac{1}{\xi} - 1\right) = k_a^*(c_1 - c_c) \tag{3}$$

(iii) At the surface of unreacted borax core according to a 1st order chemical reaction:

$$-\frac{dN_A}{dt} = \xi^2 k_s^* c_c \tag{4}$$

where $k_1^* = A_e k_1$, $k_a^* = A_e k_a = A_e(D_e/R)$, $k_s^* = A_e k_s$, $A_e = 4\pi R^2$ and $\xi = r_c/R$. k_1, k_a and k_s are the bulk mass transfer, product layer diffusion and reaction rate coefficients.

The consumption of propionic acid can be related to that of solid borax through the following relationship:

$$-\frac{dN_A}{dt} = -2\rho_B R A_e \xi^2 \frac{d\xi}{dt} \tag{5}$$

Levenspiel [1998] assumes a constant fluid concentration. Since in this work, propionic acid concentration varies as the reaction proceeds, we here introduce a variable concentration as follows:

$$c = c_0\left(1 - \frac{2X}{\kappa}\right) \tag{6}$$

where c_0 is the initial propionic acid concentration and X is the solid borax conversion defined as:

$$X = 1 - (r_c/R)^3 = 1 - \xi^3 \tag{7}$$

and $\kappa = (c_0/M_B)$, the molar ratio of charged reactants.

Combination of Eqs. (6) and (7) gives:

$$c = 2\frac{c_0}{\kappa}(\alpha^3 + \xi^3) \tag{8}$$

where $\alpha = (\kappa/2 - 1)^{1/3}$. The multiplier 2 which appears in Eqs. (5)-(8) and also in the definition of α is the number of moles of propionic acid consumed per one mole of solid borax.

Combining Eqs. (2)-(5) and (8) gives:

$$\frac{d\xi}{dt} = -\frac{c_0(\alpha^3 + \xi^3)}{\kappa\rho_B R\left[\frac{1}{k_s} + \frac{R}{D_e}(\xi - \xi^2) + \frac{\xi^2}{k_1}\right]} \tag{9}$$

Eq. (9) can be integrated by using the boundary condition $\xi = 1$ at t=0:

$$\frac{1}{k_s}\int_1^\xi \frac{d\xi}{\alpha^3+\xi^3} + \frac{R}{D_e}\int_1^\xi \frac{(\xi-\xi^2)d\xi}{\alpha^3+\xi^3} + \frac{1}{k_1}\int_1^\xi \frac{\xi^2 d\xi}{\alpha^3+\xi^3} = -\frac{c_0}{\kappa\rho_B R}\int_0^t dt \qquad (10)$$

The analytic solution of Eq. (10) can be derived as:

$$\frac{t}{Q} = \eta_1 \ln\left[\frac{(\alpha+\xi)^3}{\alpha^3+\xi^3}\frac{1+\alpha^3}{(1+\alpha)^3}\right]$$
$$+ \eta_2 \tan^{-1}\left[\frac{\sqrt{3}\alpha(\xi-1)}{2\alpha^2-\alpha(\xi+1)+2\xi}\right] + \eta_3 \ln\frac{\alpha^3+\xi^3}{1+\alpha} \qquad (11)$$

where

$$Q = \frac{R\kappa\rho_B}{c_0} \qquad (12)$$

and the three coefficients, η_1, η_2 and η_3 are derived as:

$$\eta_1 = \frac{1}{6\alpha}\left[-\frac{1}{k_s\alpha} + \frac{1}{D_e/R}\right] \qquad (13)$$

$$\eta_2 = \frac{1}{\sqrt{3}\alpha}\left[-\frac{1}{k_s\alpha} - \frac{1}{D_e/R}\right] \qquad (14)$$

$$\eta_3 = \frac{1}{3}\left[-\frac{1}{k_1} + \frac{1}{D_e/R}\right] \qquad (15)$$

Having k_s, k_1 and D_e, Eqs. (7) and (11) can be used for prediction of solid conversion with time. The method of determining these rate constants is discussed in the next section.

1. Simultaneous Determination of the Rate Constants

In order to calculate the rate constants, Eq. (10) can be transformed into the following form:

$$\frac{t}{Q} = \frac{1}{k_s}F_1(\xi) + \frac{1}{k_a}F_2(\xi) + \frac{1}{k_1}F_3(\xi) \qquad (16)$$

where

$$F_1(\xi) = -\frac{1}{3\alpha^2}G(\xi) + \frac{1}{6\alpha^2}H(\xi) - \frac{1}{\sqrt{3}\alpha^2}K(\xi) \qquad (17)$$

$$F_2(\xi) = \frac{1}{3\alpha}G(\xi) - \frac{1}{6\alpha}H(\xi) - \frac{1}{\sqrt{3}\alpha}K(\xi) + \frac{1}{3}M(\xi) \qquad (18)$$

$$F_3(\xi) = -\frac{1}{3}M(\xi) \qquad (19)$$

and the functions $G(\xi)$, $H(\xi)$, $K(\xi)$ and $M(\xi)$ are defined as:

$$G(\xi) = \ln\frac{\alpha+\xi}{\alpha+1} \qquad (20)$$

$$H(\xi) = \ln\frac{\alpha^2-\alpha\xi+\xi^2}{\alpha^2-\alpha+1} \qquad (21)$$

$$K(\xi) = \tan^{-1}\frac{\sqrt{3}\alpha(\xi-1)}{2\alpha^2-\alpha(\xi+1)+2\xi} \qquad (22)$$

$$M(\xi) = \ln\frac{\alpha^3+\xi^3}{\alpha^3+1} \qquad (23)$$

Substituting the values of k_1 obtained by the correlation of Asai et al. [1986], the values of k_s and k_a for each set of experiments can be determined by minimization of the square of errors defined as:

$$\varepsilon_r = \sum_{i=1}^n\left[\psi_s F_1(\xi_i) + \psi_a F_2(\xi_i) + \frac{1}{k_1}F_3(\xi_i) - \frac{t_i}{Q}\right]^2 \qquad (24)$$

where ψ_s and ψ_a are equal to $1/k_s$ and $1/k_a$ respectively and determined by simultaneous solution of two linear equations obtained at $\partial\varepsilon_r/\partial\psi_s = 0$ and $\partial\varepsilon_r/\partial\psi_a = 0$.

Values of the k_s and k_a were determined by linear regression analysis of 160 data points from 25 experimental runs. These parameters were then correlated by Arrhenius type equations [Eqs. (25) and (26)]. Also the values of k_1 can be expressed in the more convenient form of Eq. (27) adapted from Asai et al., 1986 for turbulent mixing condition. The standard errors in parameter estimates were determined at 95% confidence limits and the following correlations were obtained:

$$k_s = (8.7\pm0.63)\times10^3 \exp(-(49.14\pm2.62)/RT) \qquad (25)$$

$$k_a = (7.97\pm0.92)\times10^{-7} \text{ L}^{-1} \exp(-(19.21\pm1.86)/RT) \qquad (26)$$

$$k_1 = (4.62\pm0.71)\times10^{-4} \text{ L}^{-0.22} \text{N}^{0.58} \exp(-(12.2\pm1.54)/RT) \qquad (27)$$

The influence of feed particle size distribution on the fractional conversion of a multiparticle system [Pais and Portugal, 1994] can be studied by using the monosize particle kinetic parameters. In applying Eq. (11) to a feed containing a distribution of particle sizes, the weight fraction, w_i, in N size intervals can be used for predicting the overall conversion of the solid reactant as:

$$X = \sum_{i=1}^N w_i(L_i^*)X_i(L_i^*, c, t) \qquad (28)$$

where L_i^* represents the average initial diameter of the particle in the interval L_i to L_{i+1} and in weight fraction w_i defined as $(L_i+L_{i+1})/2$, and X_i refers to the fraction reacted in that weight fraction. Hence, the overall conversion of solid reactant with feed size distribution $w_i(L_i^*)$ can be predicted as long as we know the conversion for the individual particles of original size L_i^* for any time t. The latter quantity, $X_i(L_i^*, c, t)$ depends in general on the concentration of propionic acid, at time t.

In order to calculate the overall conversion of an assembly of feed crystals, Eqs. (2)-(5) and (7) were combined in the following form:

$$\frac{dX_i}{dt} = \frac{\delta c(1-X_i)^{2/3}}{\frac{1}{k_s}+\frac{1}{k_a}[(1-X_i)^{1/3}-(1-X_i)^{2/3}]+\frac{1}{k_1}(1-X_i)^{2/3}} \qquad (29)$$

where $\delta = 6/(2\rho_B L_i^*)$ and c is the concentration of propionic acid in the solution phase at time t.

In general, the overall conversion, X, must be evaluated according to the following steps:

(1) Determination of k_s, k_a and k_1 at each size fraction, L_i^* by Eqs. ((25)-(27))

(2) Numerical integration of Eq. (29) for each size for a small time interval

(3) Overall conversion, X computed from Eq. (28)

(4) Reduction in concentration of propionic acid calculated from Eq. (6)

(5) Repeating the steps 2, 3 and 4 until the completion of the process

In order to study the effect of feed size distribution at a specific mean size, the coefficient of variation (CV) of feed crystals (around the mean) can be used as a parameter. CV in its discrete approximation form can be written as:

$$CV = \frac{\sqrt{\sum_{i=1}^{N}(L_i^* - L_B)^2 w_i}}{L_B} \tag{30}$$

where L_B is the weight mean size defined as:

$$L_B = \sum_{i=1}^{n} L_i^* w_i \tag{31}$$

The fourth order Runge-Kutta method with time step of 0.5 s was used for numerical integration of Eq. (29) in step 2, and the number of size intervals, N, used for the prediction was 20.

RESULTS AND DISCUSSION

Crystal size distributions (CSDs) of three solid phase sample obtained at different time intervals in a typical run are shown in Fig. 2. This figure shows that the differential volume percent of fine particles (<350 μm) in 60 s sample is low. The fine crystals could have been created as a consequence of attrition of larger particles. The
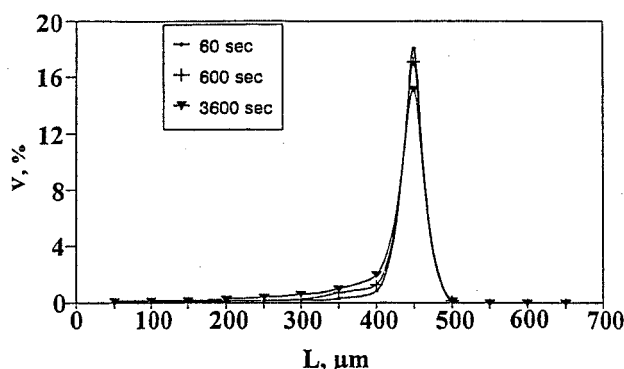

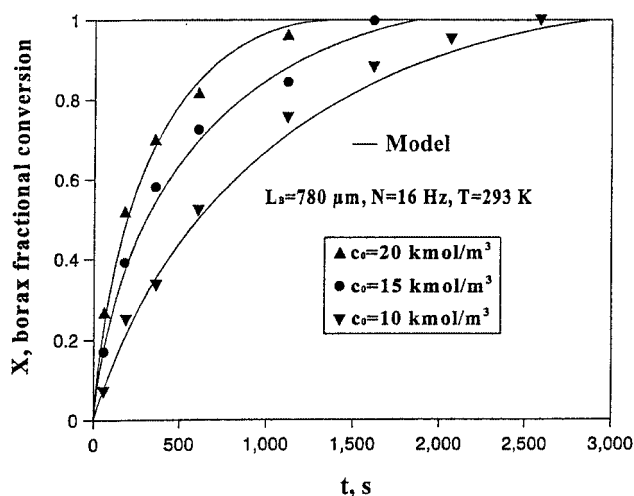Fig. 2. Crystal size distributions at different time intervals.

other CSDs obtained at 600 and 3,600 s look almost similar to the one at 60 s. All volume percentages of crystals at different times are very close to each other and concentrated around the borax seed size. Since the initial crystals of borax have retained more or less their original size, it can be concluded that the product boric acid deposits mainly as an outer solid crust around the unreacted core of solid borax.

The data obtained from the experiments were plotted as conversion fraction versus time, described as X=(the amount of reacted borax in the solid reactant)/(the amount of original borax in the solid reactant) in Figs. 3-6. The conversion profiles of different runs all at N of 16 Hz and different initial propionic acid concentrations are shown in Fig. 3. For a conversion X=0.9 the reaction time is about two times longer for $c_0=10$ kmol/m^3 compared to an experiment with a higher propionic acid concentration, $c_0=20$ kmol/m^3. At a given borax loading, the batch run time was always affected by the available propionic acid concentration in the solution phase.

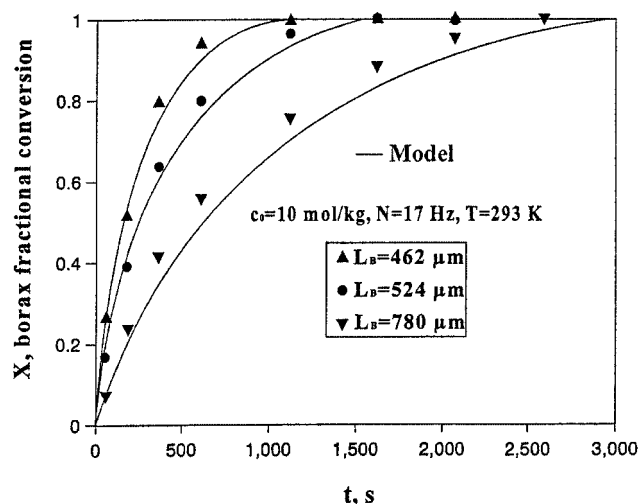Fig. 4 shows the influence of borax crystal size on the solid reac-

Fig. 4. Comparison of the simulated and measured solid borax conversion at different sizes of solid reactant.

Fig. 3. Comparison of the simulated and observed solid borax conversion at different initial propionic acid concentrations.
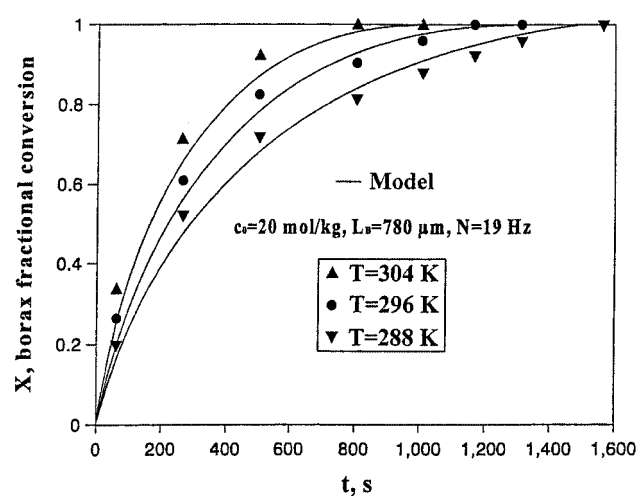

Fig. 5. Comparison of the simulated and measured solid borax conversion at different temperatures.

tant fractional conversion. The stirrer speed in these three runs was 17 Hz but the reactant borax crystals were of different size ranges. The figure shows that increase in the reactant seed size slightly affects the initial rate of conversion, but after 300 seconds, there was a sharp decrease in the rate of fractional conversion and a substantial increase in the time of completion of reaction from 900 to 2,400 s as the borax crystal size increased from 462 to 780 μm. For 780 μm reactant borax crystals, the time of reaction completion decreased from 2,500 to 2,400 s when the stirrer speed increased from 17 to 34 Hz. Thus the experimental results indicated that the dissolution of borax in propionic acid was affected primarily by the size of borax crystals with the stirrer speed having a smaller effect.

The effect of reaction temperature on the solid reactant conversion profiles was examined at different temperatures from 288 to 304 K, as shown in Fig. 5. According to this figure, an increase in temperature from 288 to 304 K resulted in a sensible increase in the initial rate of borax conversion from 5×10^{-3} to 8.7×10^{-3} s^{-1} and a decrease of time of completion of reaction from 1,580 to 1,000 s. These together with the results concerning the effects of stirrer speed and borax seed size, show that the dissolution of borax crystals in propionic acid solution is mainly controlled by the chemical reaction and/or diffusion through the solid product layer.

The influence of the spread of the feed crystal size distribution about a fixed mean size on the solid borax conversion is shown in Fig. 6. The effect is not as dramatic as for changes in the mean, but is still significant, indicating that the average particle size of the feed alone is not sufficient to completely characterize the dissolution process. It is interesting to note that the curves in this figure cross over one another; those for small CV values have low initial rates and relatively high rates of conversion for later times, and those for high CV values have high initial rates and low conversion rates at later stages of the dissolution process. This occurs because increasing values for the coefficient of variation result in an increasing abundance of both small and large diameters (relative to the mean) in the population of feed crystals. The small particles react at a high rate giving rise to the initial shape of the curves, whereas the large particles react at a slow rate giving rise to the shape of the curves at
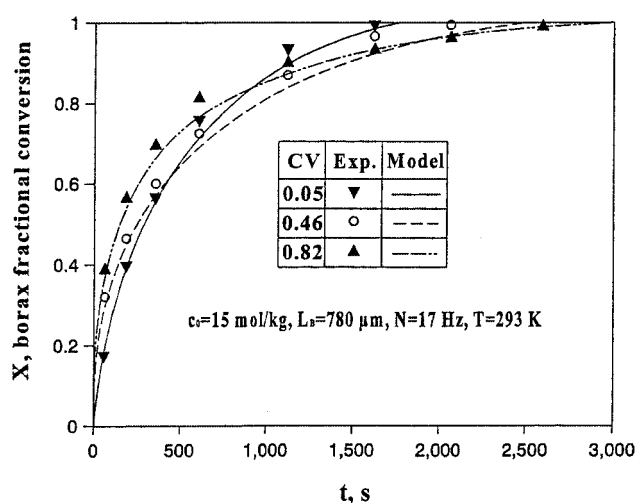

Fig. 6. Comparison of the measured and predicted solid borax conversion at different coefficient of variations of feed crystals.
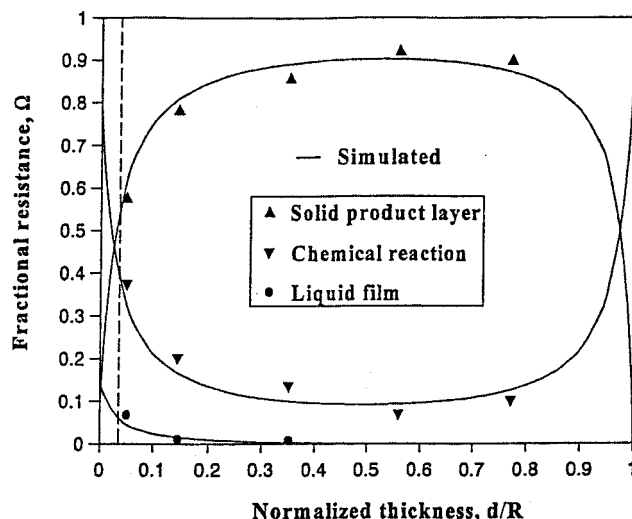
Fig. 7. Comparison of the simulated and measured variations of fractional resistances during the reaction for run S1.

longer batch run times.

The contribution of the three stages on the overall rate of reaction between borax crystals and propionic acid solution may be determined by comparing the chemical reaction, solid product layer and liquid film resistances. According to Eq. (9) these can be defined as follows:

$$\Omega_m = \frac{R_m}{\sum_{m=1}^{s} R_m} \qquad (32)$$

where $R_l = 1/k_l^*$, $R_a = (1 - \xi)/(\xi k_a^*)$ and $R_s = 1/(\xi^2 k_s^*)$ are the normalized resistances of liquid film, solid product layer and chemical reaction, respectively.

Fig. 7 shows a plot of the fractional resistances, Ω_m, against normalized thickness of the solid product, $d/R(=1-\xi)$, according to Eq. (32) for run S1. This figure indicates that at the initial stages of the reaction, the dissolution process is mainly controlled by the chemical reaction. The liquid film fractional resistance declines quickly, such that at the boric acid thickness of 0.02 (X=0.06) about 10% of the total resistance is due to diffusion through the liquid film and it approaches zero when the normalized thickness of the solid product layer exceeds 0.2. As the reaction proceeds the product layer resistance dominates and when the normalized thickness of the product layer reaches around 0.04 (dotted line in Fig. 7: thickness=15.6 μm and X=0.11), the resistance offered by diffusion through the solid product layer exceeds the sum of the other two resistances. At the normalized solid product thickness of 0.6 (X=0.94), about 90% of total resistance is contributed by solid boric acid layer. This is in good agreement with results obtained from run S1. According to the model at the final stage of the process, the dissolution is again chemical reaction controlled at the normalized solid product thickness of 0.96 (X=0.99).

Predicted borax conversion at different operating conditions is compared with typical experimental data in Figs. 3-6. Eqs. (7), (8), (11-15) and (25-27) are used to predict the conversion of monosized borax crystals with time. The fractional conversion of multisize borax crystals is also predicted by combining these equations
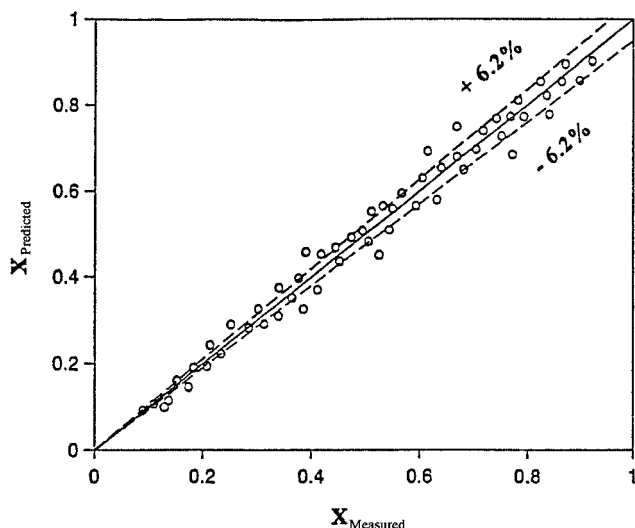
Fig. 8. Comparison of the measured and predicted conversion of solid borax.

with Eqs. (28) and (29).

The predicted overall conversions of the solid borax are also compared with experimental data for all runs in Fig. 8 at a relative deviation of ±6.2%. Figs. 3-8 indicate that the process is well described by the proposed model. Overall, the predicted results are more accurate at lower borax crystal sizes, lower temperatures, higher propionic acid concentrations and lower coefficient of variations of feed crystals.

CONCLUSIONS

A sharp interface model with variable bulk fluid concentration of liquid reactant was developed for simulation of the reaction of borax crystals with propionic acid solution. A novel method for simultaneous determination of all kinetic parameters based on the analytical solution of the rate equations was presented. The chemical reaction and diffusion through the boric acid solid layer were the main rate controlling mechanisms. The effects of process variables, namely, propionic acid concentration, borax mean crystal size, temperature and the coefficient of variation of feed crystals on the fractional conversion of borax particles, were simulated by the proposed model. The suggested one stage process shows the advantage of using an organic acid for direct production of crystalline boric acid.

NOMENCLATURE

A_e : external surface of a borax crystal [m^2]
c_0, c : initial and bulk concentrations of propionic acid respectively [kmol (m)$^{-3}$]
c_1, c_c : conc. at the solid-liquid interface and at the surface of core [kmol (m)$^{-3}$]
CV : coefficient of variation of borax crystals [-]
D_e : diffusivity of propionic acid in the porous solid layer of boric acid [m^2 s^{-1}]
d : thickness of the solid boric acid layer (R−r_c) [m]

i : experiment index [-]
k_l : bulk mass transfer coefficient [ms^{-1}]
k_a : diffusion rate of acid in solid layer of product [ms^{-1}]
k_s : chemical reaction rate coefficient [ms^{-1}]
k_l^*, k_a^*, k_s^* : normalized forms of k_l, k_a and k_s respectively [-]
L_B : mean size of feed borax crystals [m]
L_i^* : average diameter of a cut of the feed borax crystals [m]
M_B : loading of borax crystals [kmol (m)$^{-3}$]
N : stirrer speed [Hz]
N_A : moles of propionic acid in the solution phase [mol]
n : number of experiments [-]
Q : parameter defined in Eq. (12) [s]
R : gas constant (=8.314) [J mol^{-1} K^{-1}]
R_i : dimensionless resistance [-]
R_l : dimensionless liquid film resistance [-]
R_a : dimensionless resistance of solid boric acid layer [-]
R_s : dimensionless resistance of chemical reaction [-]
r : radius in spherical coordinate [m]
r_c : radius of unreacted core [m]
T : temperature [K]
t : time [s]
w_i : weight fraction of solid borax with mean size of L_i^* [-]
X_i : fractional conversion of reacted crystals of the mean cut size L_i^* [-]
X : fractional conversion of borax crystals [-]

Greek Letters

ρ : solid borax density [kmol (m)$^{-3}$]
κ : molal ratio of the reactants [-]
α : parameter in Eq. (8) (=κ/2-1) [-]
η_{1-3} : three parameters defined in Eqs. (13-15) [m^{-1}s]
ε_r : sum of square errors [-]
ξ : dimensionless core radius (=r_c/R) [-]
ψ_a, ψ_s: parameters defined in Eq. (24) (=1/k_a, 1/k_s respectively) [m^{-1}s]
Ω_m : fractional resistance [-]
δ : parameter defined in Eq. (29) (=6/(2$\rho_B L_i^*$) [kmol^{-1} m^2]

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