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# Kinetics of reaction of benzyl chloride with sodium acetate using tetrahexylammonium chloride as a phase-transfer catalyst

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**Abstract**

Benzyl acetate was synthesized by the reaction of benzyl chloride with sodium acetate in a two-phase system of an aqueous solution and toluene using tetrahexylammonium chloride (QCl) as a phase-transfer catalyst at 85 °C. The distribution coefficient and chemical equilibrium constants of QCl and tetrahexylammonium acetate were used to analyze the kinetics of the esterification. Two models such as the simple ion exchange across the interface (interface model) and the anion exchange in the aqueous phase (bulk model) were used to obtain the reaction rate constant.

The overall reaction can be described by a first-order reaction with respect to the molar concentration of benzyl chloride and the active intermediate (tetrahexylammonium acetate) produced by the reaction between QCl and acetate ion in the aqueous phase, respectively.

*Keywords:* Phase-transfer catalyst; Esterification; Tetrahexylammonium chloride; Benzyl chloride; Sodium acetate

## 1. Introduction

The reaction between two mutually insoluble phases can be promoted by a phase-transfer catalyst (PTC) under mild operating conditions to give a high yield of products [1]. The catalyst first reacts with the reactant in the aqueous phase to form an active intermediate that there transfers into the organic phase to react with the other organic reactant. Usually, the reactivity of reaction by phase-transfer catalysis is controlled by the reaction rate in the organic phase and two-phase-transfer steps between the organic and aqueous phase, as well as the partition equilibrium of the catalysis between the two phases. The mass transfer problems as well as the kinetics are very important in the field of phase-transfer catalysis. In general, it is reasonable to assume that the resistance of mass transfer can be neglected for a slow organic phase reaction by phase-transfer catalysis. Because one or more equilibria are coupled with one or more relatively slow reactions in phase-transfer catalysis, the kinetics of these systems may become extremely complicated, although in practice most systems studied have been found to be relatively simple. Most of the work published to data has dealt with the kinetics of anion transfer displacement reactions [2] using one of the reaction mechanisms with the anion exchange across the aqueous–organic interface and with the anion exchange in the aqueous phase.

Benzyl esters are synthesized by the reaction of benzyl chloride with sodium carboxylate catalyzed by tertiary amines such as triethylamine [3–5] and by

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**Nomenclature**

| | |
|---|---|
| $C_0$ | initial concentration of $Cl^-$ in toluene/water system ($kmol/m^3$) |
| $k_0$ | reaction rate constant defined by Eq. (5) ($m^3/(kmol\,s)$) |
| $K$ | equilibrium constant defined by Eq. (7) |
| $K_1$ | equilibrium constant defined by Eq. (1) ($kmol/m^3$) |
| $K_2$ | equilibrium constant defined by Eq. (2) ($m^3/kmol$) |
| $N_0$ | initial concentration of $CH_3COONa$ ($kmol/m^3$) |
| $Q_0$ | initial concentration of QCl ($kmol/m^3$) |
| $Q^+$ | tetrahexylammonium cation $(CH_3(CH_2)_5)_4\ N^+$ |
| $R_0$ | initial concentration of RCl ($kmol/m^3$) |
| $t$ | reaction time (s) |
| $x$ | fraction of feed $\overline{RCl}$ converted to $\overline{CH_3COOR}$ |
| *Greeks* | |
| $\alpha_1$ | distribution coefficient of THAC defined by Eq. (3) |
| $\alpha_2$ | distribution coefficient of $CH_3COOQ$ defined by Eq. (4) |
| *Superscript* | |
| — | organic phase |
| *Subscript* | |
| 0 | initial |

quaternary ammonium [6–8]. In the anion transfer displacement reactions such as esterification of benzyl chloride by sodium acetate referred by these papers, the reaction was described by a first-order reaction with respect to the molar concentration of benzyl chloride and an active intermediate formed by the reaction of the quaternary onium cation with the reactant in the aqueous phase, respectively. They determined the pseudo-first-order reaction rate constant under the condition of constant concentration of the active intermediate. When the concentration of the active intermediate are not maintained constant, then the reaction rate must take these variations into account.

The concentration of the active intermediate can be directly measured from the reaction of PTC with reactant in the aqueous phase [9], or estimated from the equilibrium experiment [10] between PTC and reactant in the aqueous phase. In order to estimate the concentration of active intermediate, it is necessary to know the physico-chemical properties such as the chemical equilibrium constants between PTC and the active intermediate, and the distribution coefficients of these components between the organic and aqueous phase.

In view of the fact that the circumstances of reaction condition in a heterogeneous reaction such as reaction between benzyl chloride dissolved in toluene and sodium acetate dissolved in water are different from those a homogeneous reaction, it is considered worthwhile to investigate the role of the active intermediate in the kinetics.

There is scanty information in the effect of the active intermediate, of which concentration is calculated from the equilibrium data, on the kinetics of liquid–liquid phase-transfer esterification of benzyl chloride with sodium acetate [11–13].

In the present study, the role of the active intermediate is investigated from the esterification of benzyl chloride with sodium acetate to produce benzyl acetate via liquid–liquid phase-transfer catalysis using tetrahexylammonium chloride in a well-stirred batch reactor. The intrinsic reaction rate constant is obtained from the experimental conversion of benzyl chloride to ester and the concentration of the active intermediate, which are calculated using the equilibrium constants of PTC and the active intermediate, and the distribution coefficients of these components between the organic and aqueous phase from the equilibrium of PTC and sodium acetate in the aqueous phase. Two models were used to analyze the kinetic of reaction of benzyl chloride with sodium acetate in the liquid–liquid heterogeneous phase, i.e., (i) simple ion exchange across interface, (ii) anion exchange in the aqueous phase.

## 2. Experimental

All chemicals used in this study were of reagent grade and used without further purification. tetrahexylammonium chloride (THAC or QCl, $[CH_3(CH_2)_5]_4$-

NCl) was received from Aldrich Chemical Company, Inc., USA.

### 2.1. Two-phase reaction

A reactor was used for experimental studies of esterification reaction. The reactor was a stirred vessel with a flat liquid–liquid interface. The vessel was composed of a Pyrex cylinder, two-bladed glass disk turbine stirrers 0.04 m in diameter and a reflux condenser. The vessel was 0.07 m in diameter and 0.12 m in length. Four vertical baffles, 0.004 m wide, were oriented $\pi/2$ rad and fitted from 0.005 to 0.07 m above the bottom. Two stirrers were used to agitate the organic and the aqueous phase, respectively, positioned at the middle of each phase, and set at the constant speed of agitation. The vessel was kept in a constant temperature bath with a temperature control of ±0.55.

An aqueous solution of $1 \times 10^{-4}\,\mathrm{m}^3$ having dissolved sodium acetate was placed in the vessel, and a toluene solution of $1 \times 10^{-4}\,\mathrm{m}^3$ having dissolved benzyl chloride and THAC was then added into the vessel slowly so as not to disturb the aqueous solution. The reaction mixture was heated to a desired temperature and then stirred at a given speed of agitation. Samples were withdrawn from the toluene solution at a regular interval and analyzed by gas chromatography to measure the concentrations of benzyl chloride and benzyl acetate. The stoichiometric analysis for esterification was confirmed by the comparison of the measured concentrations of benzyl chloride with those of ester. The measured conversions of benzyl chloride to ester as a function of reaction time were used to get the rate of reaction at the reaction time. During the experiment in this study, the ranges of concentration of sodium acetate, benzyl chloride and THAC were 0.3–1.0, 0.3 and 0.01–0.2 kmol/m$^3$, respectively, those of speed of agitation, 0–1200 rev/min, and the reaction temperature, 85 °C.

### 2.2. Equilibria of PTC and sodium acetate

Measurements of the dissociation and distribution equilibria of THAC and tetrahexylammonium acetate ($CH_3COOQ$) were carried out for systems of toluene/water, aqueous electrolyte solutions, or aqueous sodium acetate. Using a shaker, $2.5 \times 10^{-5}\,\mathrm{m}^3$ of toluene solution of THAC was shaken vigorously for about 2 h with the same volume of aqueous solution of sodium acetate. The distribution constant and the equilibrium constant of THAC and $CH_3COOQ$ were measured at 358 K along the procedure similar to those reported elsewhere [14].

### 2.3. Measurement of concentrations

The concentration of $Q^+$ ion in the aqueous phase was determined by the orange II-chloroform method with an ultraviolet spectrophotometer (HP 8452A Model, 482 nm). The concentration of $Cl^-$ in the aqueous phase was measured by an ion chromatograph (Water, 600E Model). The concentration of $CH_3COO^-$ ion in the aqueous phase was measured by the titration method with 0.1 kmol/m$^3$ HCl. The concentration of benzyl chloride and benzyl acetate in the organic phase were measured by a gas chromatograph (Young-in, 680D Model, Korea). The GC column, 0.002 m in diameter and 1.8 m in length, was packed with 10% SE30 as liquid phase adsorbed on 80–100 mesh Chromosorb WHP as a solid support. The column temperature was programmed with an initial temperature at 150 °C for 2 min, increased at 20 °C/min to 270 °C, and maintained at 270 °C for 10 min. Helium was used as the carrier gas with a flow rate of 25 ml/min. An FID detector was used at the temperature of 290 °C. Under this analysis condition, the retention time of benzyl chloride and benzyl acetate were 4.8, 6.1 min, respectively.

## 3. Results and discussion

### 3.1. Kinetic scheme

Fig. 1 presents the reactions characterizing the esterification of benzyl chloride by sodium acetate with a phase-transfer catalyst in toluene/water two-phase system. It is assumed that dissociation of QCl and $CH_3COONa$, formation of $CH_3COOQ$ in the aqueous phase and formation of ester in the organic phase occur along the reactions such as reactions of (i)–(iii) in Fig. 1, where Q refers to tetrahexylammonium cation. The equilibrium constants, $K_1$ and $K_2$ in reactions (i) and (ii) in Fig. 1, and distribution constants, $\alpha_1$and $\alpha_2$ of QCl and $CH_3COONa$ are defined as follows,

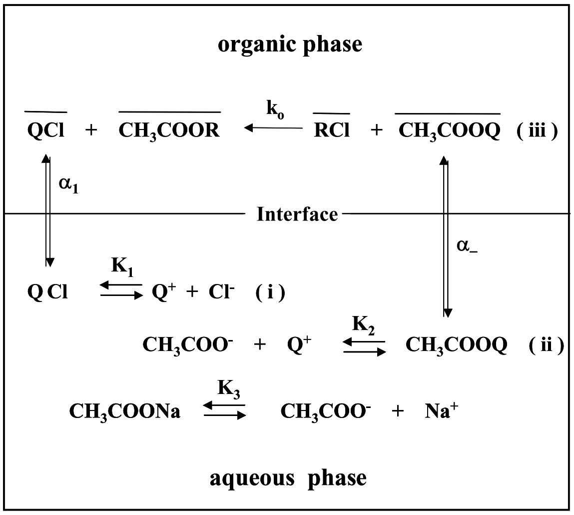

Fig. 1. Esterification path with phase-transfer catalyst in an aqueous–organic two phase system (Q: $[CH_3(CH_2)_5]_4N$, R: $C_6H_5CH_2$).

respectively:

$$K_1 = \frac{[Q^+][Cl^-]}{[QCl]} \tag{1}$$

$$K_2 = \frac{[CH_3COOQ]}{[CH_3COO^-][Q^+]} \tag{2}$$

$$\alpha_1 = \frac{[QCl]}{[\overline{QCl}]} \tag{3}$$

$$\alpha_2 = \frac{[CH_3COOQ]}{[\overline{CH_3COOQ}]} \tag{4}$$

where the brackets and overbar refer to the concentration and organic phase, respectively.

For an experiment where a highly oleophilic catalyst is used such that essentially all of it stays in the organic phase through out the reaction, the rate of esterification of $\overline{RCl}$ will be given by

$$-\frac{d[\overline{RCl}]}{dt} = k_0[\overline{RCl}][\overline{CH_3COOQ}] \tag{5}$$

Two reaction mechanisms may be possible according to the formation path of component, $\overline{CH_3COOQ}$ as follows.

### 3.1.1. A simple ion exchange across the interface (interface model)

If a simple ion exchange between $\overline{QCl}$ and $CH_3COO^-$ occurs across the interface, the overall reaction between $\overline{QCl}$ and $CH_3COO^-$ is

$$\overline{QCl} + CH_3COO^- \overset{K}{\rightleftarrows} \overline{CH_3COOQ} + Cl^- \tag{6}$$

and the overall reaction equilibrium constant is defined as follows:

$$K = \frac{[\overline{CH_3COOQ}][Cl^-]}{[\overline{QCl}][CH_3COO^-]} \tag{7}$$

By combining Eq. (7) with (1)–(4), the overall equilibrium constant defined in Eq. (7) is expressed as follows:

$$K = \frac{K_1 K_2 \alpha_1}{\alpha_2} \tag{8}$$

The mass balance of component Q in the organic phase is

$$Q_0 = [\overline{CH_3COOQ}] + [\overline{QCl}] \tag{9}$$

By combining Eq. (7) with (9), the concentration of $\overline{CH_3COOQ}$ is given by

$$[\overline{CH_3COOQ}] = \frac{KQ_0[CH_3COO^-]}{K[CH_3COO^-] + [Cl^-]} \tag{10}$$

Substitution of Eq. (10) into Eq. (5) gives

$$\frac{dx}{dt} = \frac{k_0 K Q_0 R_0 (1-x)^2}{KN_0(1-x) + C_0 + xR_0} \tag{11}$$

which may be integrated to

$$A\left(\frac{x}{1-x}\right) + B\ln(1-x) = Ct \tag{12}$$

where $A = C_0 + R_0$, $B = R_0 - KN_0$ and $C = k_0 K Q_0 R_0$.

$R_0$, $N_0$, and $Q_0$ are the initial concentration of RCl, $CH_3COONa$ and THAC, respectively, and $C_0$ is the initial concentration of $Cl^-$ dissociated from THAC in the toluene/water system.

The initial concentration of $Cl^-$ in the THAC/$H_2O$ system, $C_0$, is obtained as follows.

The mass balance of component, $Cl^-$ is

$$Q_0 = [\overline{QCl}] + [QCl] + [Cl^-] \tag{13}$$

Because the concentration of $Q^+$ and $Cl^-$ are same each other in this system, Eq. (13) is arranged to the following equation using Eqs. (1) and (3):

$$C_0 = \frac{-1 + \sqrt{1 + 4Q_0(1+\alpha_1)/\alpha_1 K_1}}{2(1+\alpha_1)/\alpha_1 K_1} \tag{14}$$

### 3.1.2. Anion exchange between $Q^+$ and $CH_3COO^-$ in the aqueous phase (bulk model)

If the component Q transfers back and forth across the interface with anion exchange in the aqueous phase as shown in Fig. 1, the mass balance of component Q in the organic and aqueous phase is

$$\begin{aligned} Q_0 &= [\overline{CH_3COOQ}] + [CH_3COOQ] + [\overline{QCl}] \\ &\quad + [QCl] + [Q^+] \end{aligned} \tag{15}$$

where $Q_0$ is the feed concentration of QCl.

By combining Eq. (15) with Eqs. (1)–(4), the concentration of $\overline{CH_3COOQ}$ is given by

$$[\overline{CH_3COOQ}] = \frac{K_1 K_2 Q_0 [CH_3COO^-]}{\alpha_2 K_1 + ((1+\alpha_1)/\alpha_1)\alpha_2 [Cl^-] + (1+\alpha_2) K_1 K_2 [CH_3COO^-]} \tag{16}$$

The concentrations of components, $CH_3COO^-$ and $Cl^-$ are presented as follows:

$$[CH_3COO^-] = N_0(1-x) \tag{17}$$

$$[Cl^-] = C_0 + xR_0 \tag{18}$$

where $x$ is the fraction of original $\overline{RCl}$ converted to $\overline{CH_3COOR}$ in reaction (iii) in Fig. 1, $N_0$, $R_0$, and $C_0$ are the feed concentrations of $CH_3COONa$, RCl and $Cl^-$, respectively.

Substitution of these values of $[CH_3COO^-]$ and $[Cl^-]$ into Eq. (5) through Eq. (16) gives

$$\frac{dx}{dt} = \frac{k_0 K_1 K_2 Q_0 R_0 (1-x)^2}{\alpha_2 K_1 + ((1+\alpha_1)/\alpha_1)\alpha_2 (C_0 + xR_0) + (1+\alpha_2) K_1 K_2 N_0 (1-x)} \tag{19}$$

which may be integrated to

$$A\left(\frac{x}{1-x}\right) + B \ln(1-x) = Ct \tag{20}$$

where

$$A = \alpha_2 K_1 + \left(\frac{1+\alpha_1}{\alpha_1}\right)\alpha_2 (C_0 + R_0),$$

$$B = \left(\frac{1+\alpha_1}{\alpha_1}\right)\alpha_2 R_0 - (1+\alpha_2) K_1 K_2 N_0,$$

$$C = k_0 K_1 K_2 Q_0 R_0$$

with the initial conditions that $x = 0$ when $t = 0$.

The intrinsic reaction rate constant, $k_0$, may be obtained from Eq. (12) or (20) by using the measured concentration of $\overline{RCl}$ in the organic phase against the reaction time.

## 3.2. Equilibria of $QCl/CH_3COONa$ system

The distribution constants, $\alpha_1$ and $\alpha_2$, and the equilibrium constants, $K_1$ and $K_2$ in Fig. 1 must be given in order to get the values of the left-hand side of Eq. (12) or (20), and these constants are obtained along the procedure similar to those reported elsewhere [14] in the equilibrium of QCl–toluene/$CH_3COONa$–$H_2O$ heterogeneous system. The evaluated distribution constants of THAC and the active intermediate, $CH_3COOQ$ formed between tetrahexylammonium cation and $CH_3COO^-$ were correlated as a function of the ionic strength in the aqueous phase as follows [14]:

$$\begin{aligned} \log\left(\frac{\alpha_1}{0.0709}\right) &= -(0.0566[NaCl] \\ &\quad + 0.0564[CH_3COONa]) \end{aligned} \tag{21}$$

$$\log\left(\frac{\alpha_2}{0.1105}\right) = -0.1501[CH_3COONa] \tag{22}$$

Also, the equilibrium constants in the reactions of (i) and (ii) of Fig. 1, $K_1$ and $K_2$ are 0.0064 kmol/m$^3$ and 472.42 m$^3$/kmol, respectively [14].

## 3.3. Determination of reaction rate constant ($k_0$)

Using the measured concentration of $\overline{RCl}$ against the reaction time at $Q_0 = 0.05$ kmol/m$^3$, $R_0 = 0.3$ kmol/m$^3$, and $N_0 = 0.3$ kmol/m$^3$, the values of the left-hand side of Eqs. (12) and (20) are plotted against the reaction time in Fig. 2, respectively. The values of $\alpha_1$ and $\alpha_2$ used in calculation of the values the left-hand side of equation are 0.0046 and 0.1041,

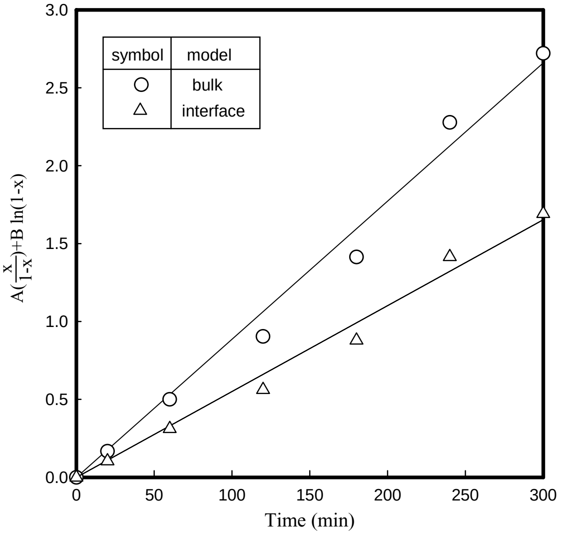

Fig. 2. Plot of left-hand side of Eqs.(12) and (20) vs. reaction time ($Q_0 = 0.05\,\mathrm{kmol/m^3}$, $R_0 = 0.3\,\mathrm{kmol/m^3}$, $N_0 = 0.3\,\mathrm{kmol/m^3}$).

respectively, which are obtained from Eqs. (21) and (22). As shown in Fig. 2, the plots satisfies the straight line very well with the correlation coefficient of 0.991. The reaction rate constants are obtained from the slopes of the straight lines in Fig. 2 by a least-square method, and the reaction rate constants are $1.58 \times 10^{-3}$ and $1.74 \times 10^{-3}\,\mathrm{m^3/(kmol\,s)}$ according to the reaction mechanisms of the interface model and bulk model, respectively. These reaction rate constants are almost same each other, and then it is very hard to distinguish between the interface model and the bulk model.

The concentration of $\overline{\mathrm{CH_3COOQ}}$ in Eq. (16) in the bulk model through Eq. (7) is rearranged to the following equation:

$$[\overline{\mathrm{CH_3COOQ}}] = \frac{KQ_0[\mathrm{CH_3COO^-}]/\alpha_1 K_1}{1 + ((1+\alpha_2)K/\alpha_1 K_1)[\mathrm{CH_3COO^-}] + ((1+\alpha_1)/\alpha_1 K_1)[\mathrm{Cl^-}]} \tag{23}$$

If the values of $\alpha_1$, $\alpha_2$ and $K_1$ are less than 1, the values of $(1+\alpha_2)K/\alpha_1 K_1$ and $(1+\alpha_1)/\alpha_1 K_1$ in the denominator of Eq. (23) become to be less than 1, and Eq. (23) comes to be Eq. (10), which is based on the interfacial model. Because the values of $\alpha_1$, $\alpha_2$ and $K_1$ in the reaction condition of Fig. 2 are 0.0046, 0.1041, and $0.0064\,\mathrm{kmol/m^3}$, respectively, the concentration of $\overline{\mathrm{CH_3COOQ}}$ in the two model becomes to be same. In the case of a highly oleophilic catalyst, $\alpha_1$ and $\alpha_2$ are less than 1, and then the esterification of benzyl chloride and sodium acetate by a highly oleophilic phase-transfer catalyst such as THAC occurs at the interface between the organic and aqueous phase.

### 3.4. *Effect of agitation speed on the reaction rate constant*

It is necessary to indicate the experimental conditions for the diffusion effect of the components, QCl and $\mathrm{CH_3COOQ}$, to be neglected in the case that the chemical reaction occurs in the heterogeneous phase like liquid–liquid system [15]. The concentration of $\overline{\mathrm{RCl}}$ was measured against the reaction time in the range of agitation speed of 0–1200 rev/min under the reaction condition such as $Q_0 = 0.01\,\mathrm{kmol/m^3}$, $R_0 = 0.3\,\mathrm{kmol/m^3}$, and $N_0 = 0.3\,\mathrm{kmol/m^3}$.

The obtained reaction rate constants obtained by using Eq. (12) are plotted against to agitation speed in Fig. 3. As shown in Fig. 3, no increase of $k_0$ is observed when the agitation rate exceeds 600 rev/min. Therefore, the agitation rate was set at 600 rev/min for studying the reaction kinetics from which the resistance of mass transfer between two phases is kept at a constant value.

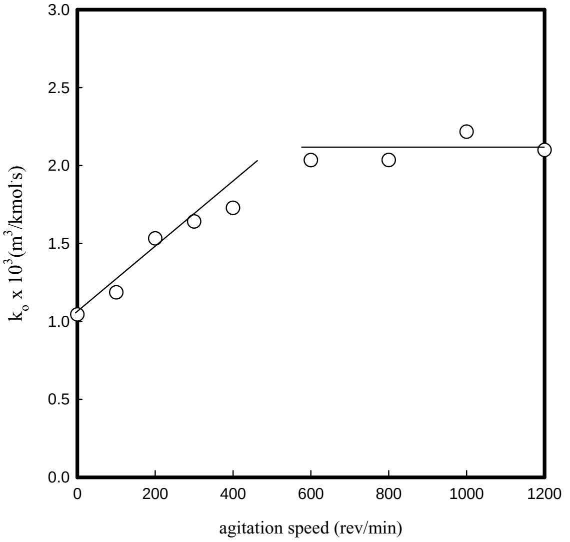

Fig. 3. Effect of agitation speed on reaction rate constant ($Q_0 = 0.01\,\mathrm{kmol/m^3}$, $R_0 = 0.3\,\mathrm{kmol/m^3}$, $N_0 = 0.3\,\mathrm{kmol/m^3}$).

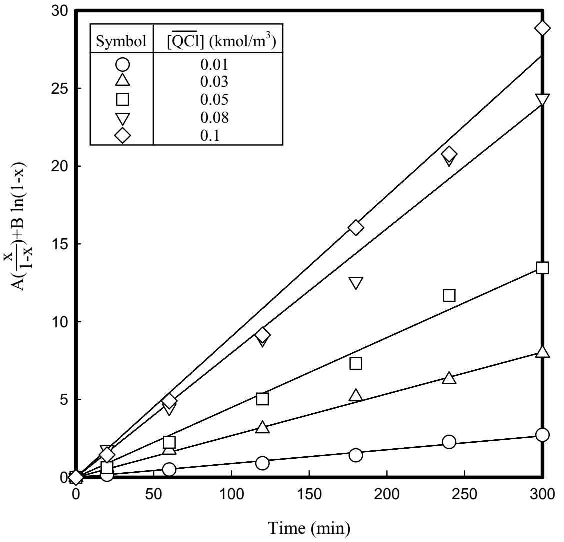

Fig. 4. Plot of left-hand side of Eq. (12) vs. time with the concentration of QCl as parameter ($R_0 = 0.3\,\mathrm{kmol/m^3}$, $N_0 = 0.3\,\mathrm{kmol/m^3}$).

### 3.5. Effect of concentration of QCl

The following operating conditions are specified for studying the effect of the phase-transfer catalyst, on the reaction rate constant: $0.3\,\mathrm{kmol/m^3}$ ($R_0$), and $0.3\,\mathrm{kmol/m^3}$ ($N_0$). The values of the left-hand side of Eq. (12) are plotted against the reaction time for various concentrations of the phase-transfer catalyst in Fig. 4. As shown in Fig. 4, the plots are linear very well, and the slopes increases as the concentration of the phase-transfer catalyst increases. The reaction rate constants are obtained from the slope of these plots by a least-square method, and shown in Fig. 5. As shown in Fig. 5, the reaction rate constant is kept at a constant value.

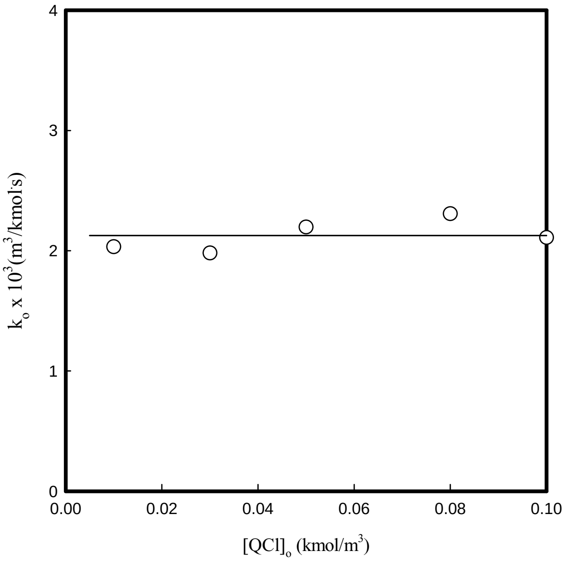

Fig. 5. Effect of QCl concentration on reaction rate constant ($R_0 = 0.3\,\mathrm{kmol/m^3}$, $N_0 = 0.3\,\mathrm{kmol/m^3}$).

## 4. Conclusions

Reaction kinetics on the synthesis of benzyl acetate from benzyl chloride and sodium acetate by liquid–liquid phase-transfer catalysis have been studied at 358 K. The observed conversion of benzyl chloride to benzyl acetate was used to analyze the esterification mechanism with a cyclic phase-transfer initiation step ion the heterogeneous liquid–liquid system. The distribution constants and equilibrium constants of QCl and $CH_3COOQ$ obtained from the equilibrium experiments were used to calculate the concentration of the active intermediate, $\overline{CH_3COOQ}$, in the organic phase.

The rate of esterification was formed to be first-order in both the concentrations of benzyl chloride and the activate intermediate. The heterogeneous liquid–iquid system with the active intermediate was used to obtain the reaction rate constant.

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