# Kinetics of Liquid Phase Synthesis of Ethyl *tert*-Butyl Ether from *tert*-Butyl Alcohol and Ethanol Catalyzed by β-Zeolite Supported on Monolith

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ABSTRACT: This paper compared the performance of  $\beta$ -zeolite and Amberlyst-15 catalysts on a liquid phase synthesis of ethyl *tert*-butyl ether (ETBE) from ethanol (EtOH) and *tert*-butyl alcohol (TBA)  $\beta$ -Zeolite was synthesized and deposited on monolith support. Its structure was confirmed by an XRD measurement and its composition was analyzed by an XRF measurement. It was found that even though the catalytic activity of  $\beta$ -zeolite was lower than that of Amberlyst-15, the selectivity of ETBE was much higher than that of Amberlyst-15, resulting in almost the same level of ETBE yield. The dehydration of TBA to isobutene (IB) was the major side reaction. The kinetic study of the reaction catalyzed by  $\beta$ -zeolite supported on monolith was carried out by using a semibatch reactor. The effect of external mass transfer was investigated by varying stirring speeds. The activity-based rate expressions were developed taking into account of water inhibition. Three temperature levels of 323, 333, and 343 K were performed in the study to obtain the parameters in the Arrhenius's equation and the Van't Hoff's equation. © 2002 Wiley Periodicals, Inc. Int J Chem Kinet 34: 292–299, 2002

# INTRODUCTION

Although MTBE, commercially produced from the reaction of isobutene (IB) and methanol, currently

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predominates in the world industries, a recent investigation revealed that it has tendency to pollute underground water because of its high solubility in water. As a result, there is a pending legislation in a number of states in the US [1]. A review on the market expansion of oxygenate fuels and the catalytic aspect of their synthesis was given by Ancilloti and Fattoro [2].

Similar to MTBE, ETBE showed a considerable reduction in CO, a small reduction in unburned

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hydrocarbon, and a nonsignificant effect on  $NO_x$  [3,4]. It may be a good alternative compared to lighter alcohols because of its lower blending Reid vapor pressure (bRvp), low vaporization latent heat, very low solubility in water and so on which account for its full fungibility in the petroleum refining and distribution system [5], and compared to other tert ethers such as TAME and TAEE because of their high cost. ETBE is also attractive from the viewpoint of environment, as it is derived from EtOH which can be obtained from renewable resources such as biomass [6,7]. It was expected that by 2005, 5% of fuel used for transportation in France should be produced from renewable sources [8]. ETBE outranks MTBE as an octane enhancer and is more attractive than MTBE for low bRvp blends as required (less than 55 kPa) in some hot places during summer or in tropical countries because ETBE has low bRvp (28 kPa) than MTBE (55-69 kPa) [5]. In addition, because the water solubility of MTBE (43 kg/m<sup>3</sup>) is about 4 times higher than that of ETBE  $(12 \text{ kg/m}^3)$ , the use of ETBE reduces the risk of water contamination.

Generally, ETBE can be produced by an exothermic reversible reaction between EtOH and IB. However, the supply of IB is mainly limited from refinery catalytic cracking and steam cracking fractions. Hence, alternative routes for the synthesis of ETBE were currently explored [9]. TBA, a major byproduct of propylene oxide production from IB and propylene in the ARCO process, can be employed instead of IB as a reactant [10]. TBA was first investigated for the ETBE synthesis about 60 years ago [11]. There are two methods to produce ETBE from TBA, namely the indirect and the direct methods. In the indirect method, TBA is first dehydrated to IB in a reactor and then the produced IB is reacted with EtOH to produce ETBE in a second reactor. In the direct method which is of our interest, ETBE is produced directly from TBA and EtOH in only one reactor. It is favorable because it shortens the process itself [12]. However, selection of proper catalysts with high activity and selectivity is a major concern for the success of this process.

A number of researches are investigating the ETBE synthesis by the direct method using various catalysts such as Amberlyst-15, heteropoly acid (HPA), KHSO<sub>4</sub>, NaHSO<sub>4</sub>, and H<sub>2</sub>SO<sub>4</sub>. Yin and coworkers [12] compared the performance of Amberlyst-15 and HPA by performing the reactions in a semibatch reactor. The conversion and selectivity of Amberlyst-15 (at 338 K and 8 h reaction time) were 62% and 43%, respectively. Heteropoly acid was found to yield superior selectivity (79%); however, it was less attractive because it was significantly inhibited by the presence of water. Matouq and coworkers [13] employed KHSO<sub>4</sub>, NaHSO<sub>4</sub>, H<sub>2</sub>SO<sub>4</sub>, and Amberlyst-15 as catalysts for the

direct synthesis of ETBE from TBA and EtOH in reactive distillation columns. NaHSO<sub>4</sub> failed to synthesize ETBE. The homogeneous catalyst KHSO<sub>4</sub> was found to be superior to H<sub>2</sub>SO<sub>4</sub> and Amberlyst-15 which produced IB as a main product; however, a subsequent catalyst separation unit was required. Recent research compared three cation-exchange resins of S-54, D-72, and Amberlyst-15 [14]. It was observed (at T = 338 K) that S-54 showed improvements (compared to those of Amberlyst-15) of activity and selectivity of 6 and 5%, respectively, while D-72 showed improvements of 10 and 1%, respectively.

The purpose of this investigation was to compare the performance of  $\beta$ -zeolite catalyst with the commercial Amberlyst-15, which is usually used for *tert*ethers synthesis, for the production of ETBE from TBA and EtOH by the direct method. The zeolitic catalyst was chosen as it showed promising properties, high thermal stability and no acid fume emission against conventional resin-based catalysts [15]. In this study, the kinetic parameters based on an activity model of  $\beta$ -zeolite catalyst were determined. The obtained parameters were used for modeling the ETBE production in a reactive distillation with or without a combined pervaporation unit (as proposed in our earlier work [10]) and a pervaporative membrane reactor.

# **EXPERIMENTAL**

# **Catalyst Preparation**

Synthesis of  $\beta$ -zeolite powder was carried out in an autoclave at 408 K and 300 kPa. Colloid of SiO<sub>2</sub> and NaAlO<sub>2</sub>, which were the main reagents employed in the zeolite synthesis, were used to obtain a Si/Al ratio of 42. A compound of tetraethylammonium hydroxide was used as a template for crystals. Sodium and potassium ions, contained in NaCl, NaOH, and KCl were used as seeds for the crystals, as well as to balance the ionic charges in the crystals. All these components when mixed together formed a gel. To avoid quick solidification of the gel, HCl was added to keep pH at low level. The gel was stirred thoroughly at room temperature before transferring it to the autoclave. Then, the mixture was stirred and heated for 40 h. In order to remove the template from the catalyst precursor, the catalyst was calcined at 813 K for 3.5 h. After this, the catalyst in the Na<sup>+</sup> form was ion exchanged twice with the solution of 2 M NH<sub>4</sub>NO<sub>3</sub> at 353 K for 1 h. Then, the ion-exchanged crystals were dried in an oven at 383 K for at least 3 h. The resulting crystals were in the  $NH_4^+$ form. Finally, calcination of the catalyst at 773 K for 2 h was necessary to dissociate the ammonium ion,  $NH_4^+$ into NH<sub>3</sub> and protonated form of hydrogen, H<sup>+</sup>. NH<sub>3</sub>

escaped in the atmosphere while H<sup>+</sup> stayed onto the catalyst to balance the ionic charge. The catalyst was characterized by an X-ray diffraction (XRD-SIEMENS D5000) and its composition was measured using an X-ray fluorescence spectrometer (XRF-model Fision).

# Preparation of Supported β-Zeolite

Supported  $\beta$ -zeolite was prepared by coating the obtained powder catalysts on a cordierite monolith support (400 cell/in<sup>2</sup>.) which was cut into small cubes of  $0.5 \times 0.5 \times 0.5$  cm<sup>3</sup>. The supports were weighted and soaked in 2.5 wt% acetic acid solution for 2 min. After this they were washed with distilled water several times to remove residual acid solution, and then dried in an oven at 383 K until the weight became constant. β-zeolite powder was added into 2.5 wt% acetic acid solution to give 30-50% w/v washcoat. The obtained slurry was stirred for 10 min. Then, the monolith supports were dipped into the prepared washcoat for 15 min, followed by drying at 383 K overnight in the oven. The supports were repeatedly dipped in the washcoat 2-3 times and calcined at 773 K for 3.5 h in air atmosphere. The amount of catalyst was calculated as the increased weight of the monolith. The uniformity of the catalyst distribution was examined by measuring distribution of Al using a scanning electron microscopeenergy dispersive X-ray spectrometer (SEM-EDX). It was shown that the catalyst was well dispersed on the monolith surface.

# **Kinetic Study**

The powder  $\beta$ -zeolite was used in a preliminary study to compare the catalyst performance between  $\beta$ -zeolite and Amberlyst-15 (obtained from Fluka). The latter is a sulfonated styrene divinyl benzene copolymer with a macroreticular structure. The mesh size was in the range of 20–50 and the ion exchange capacity was 4.96 mol-H<sup>+</sup>/kg-dry resin. A certain amount of catalyst was weighted and pretreated by leaving it in an oven at 363 K overnight to remove moisture from the catalyst. A desired amount of EtOH and TBA was mixed and placed into a slurry reactor consisting of 250 cm<sup>3</sup> three-necked flask fitted with a condenser in the central opening. The mixture was stirred and heated up to 338 K by circulating hot water through the jacket. The reaction was started by adding the catalyst into the reaction mixture. Liquid samples of 1 cm<sup>3</sup> were taken to measure concentrations of H<sub>2</sub>O, EtOH, TBA, IB, and ETBE at different reaction times. They were analyzed by a TCD gas chromatography with a column packed with Gaskuropack 54.

In the kinetic study,  $\beta$ -zeolite supported on monolith was packed in a specially designed basket-type reactor

as shown in Fig. 1a. A frame of four catalyst baskets was equipped with a rotating shaft which was driven by a motor via an inverter controller to determine the accurate start-up time in a semibatch reactor [16]. The cylindrical baskets were made of stainless steel tubes with a wall made of stainless steel mesh. The frame was held above the liquid level by upper hooks as shown in Fig. 1b. When the temperature was maintained at a desired value, the reaction was started by inverting the direction of agitation so that the frame of baskets dropped into the liquid mixture. The lower hooks were securely connected with slots on the disk turbine and the frame was rotated without slip. Three temperature levels, T = 323, 333, and 343 K, were performed under atmospheric pressure. It is noted that the liquid samples taken from the reactor were small compared to the total liquid volume.

# **RESULTS AND DISCUSSION**

#### **Catalyst Characterization**

Synthesized  $\beta$ -zeolite was analyzed by an XRD measurement to identify crystal structure. Figure 2 showed an XRD pattern which agreed well with those reported in Ref. [17]. This indicates that the synthesized catalysts had the same structure as  $\beta$ -zeolite. The particle size of the obtained  $\beta$ -zeolite powder was approximately 0.5  $\mu$ m. The Si, Al, and Na contents of  $\beta$ -zeolite measured by XRF spectrometer (XRF-model Fision) were 96.97, 2.30, and 0.21 wt%, respectively, thus yielding the Si/Al ratio of 42.

# Comparison Between Catalyst Performances

Two types of catalysts, i.e. commercial Amberlyst-15 and powder  $\beta$ -zeolite were tested in a slurry reactor to compare the performance on the synthesis of ETBE from EtOH and TBA. The experiments were carried out under the following conditions: catalyst weight = 4 g, T = 338 K, stirring speed = 660 rpm, and the initial amount of ethanol and TBA = 0.5 and 0.5 mol, respectively. The reactions taking place in the reactor can be summarized as follows:

$$TBA + EtOH \Leftrightarrow ETBE + H_2O \tag{1}$$

$$TBA \Leftrightarrow IB + H_2O$$
 (2)

$$IB + EtOH \Leftrightarrow ETBE$$
 (3)

Figure 3 shows the number of moles of TBA, EtOH, ETBE, and  $H_2O$  in liquid mixtures at different reaction time of both catalysts. Filled and empty symbols



Figure 1 Schematic diagrams of (a) experimental setup and (b) catalyst basket.

represent results of Amberlyst-15 and  $\beta$ -zeolite, respectively. It should be noted that the side product, IB was mainly present in the gas phase. Considering the disappearance of TBA, it was found that  $\beta$ -zeolite was less active than Amberlyst-15. However, when considering the formation of ETBE, it is obvious that the ETBE yields of both catalysts were almost the same. By defining selectivity as the ratio of the produced amount of ETBE to the produced amount of water, it was found that the selectivities of  $\beta$ -zeolite and

Amberlyst-15 were 70% and 34% respectively. It can be concluded that  $\beta$ -zeolite was much more attractive than Amberlyst-15 and, consequently, the following studies would consider only  $\beta$ -zeolite catalyst.

# **Kinetic Study**

*Effect of External Mass Transfer.* The kinetic study was investigated in the basket-type reactor using  $\beta$ -zeolite supported on monolith. The effect of external



**Figure 2** XRD pattern of  $\beta$ -zeolite (Si/Al = 41).

mass transfer of catalyst was studied by varying stirring speeds. Figure 4 shows the relationship between the conversion of TBA (at 7 h) and the stirring speed. It was found that the conversion increased with increasing speed and, finally, it leveled off at speed of 1210 rpm. It can be concluded that the effect of external mass transfer can be neglected when the speed is higher than 1210 rpm. In the subsequent studies, a speed of 1210 rpm was used to ensure negligible external mass transfer resistance.

**Development of Mathematical Models.** Mathematical models were developed by following our previous study [6]. The reverse reaction in Eq. (2) and the reaction in Eq. (3) were neglected since the operating pressure in this study was at atmospheric pressure and,



**Figure 3** Comparison between different catalysts. Filled symbol: Amberlyst-15; empty symbols:  $\beta$ -zeolite (catalyst weight = 4.0 g,  $m_{\text{TBA},0} = 0.5 \text{ mol}$ ,  $m_{\text{EtOH},0} = 0.5 \text{ mol}$ , T = 338 K, and reactor type = slurry reactor).

consequently, only small amount of IB can be dissolved in the liquid. It was also confirmed by our experimental results that the concentration of IB was very small in liquid mixture. As a result, the rate laws of reactions (1) and (2) can be expressed in terms of activities as

$$r_1 = k_1 \frac{\left(a_{\text{TBA}} a_{\text{EtOH}} - \frac{a_{\text{ETBE}} a_{\text{H}_2\text{O}}}{K_1}\right)}{1 + K_{\text{W}} a_{\text{H}_2\text{O}}} \tag{4}$$

$$r_2 = k_2 \frac{a_{\rm TBA}}{1 + K_{\rm W} a_{\rm H_2O}} \tag{5}$$

where  $k_j$  is the reaction rate constant of reaction j (j = 1, 2),  $a_i$  is an activity of species i, and  $K_W$  is the water inhibition parameter.



**Figure 4** The effect of speed on the conversion (catalyst =  $\beta$ -zeolite, catalyst weight = 15 g,  $m_{\text{TBA},0} = 2 \text{ mol}$ ,  $m_{\text{EtOH},0} = 2 \text{ mol}$ , T = 333 K, time = 7 h, and reactor type = basket reactor).

The expression of  $K_1$  is given as follow [18].

$$K_1 = \exp(1140.0 - 14580/T + 232.9 \ln T + 1.087 T) - 1.114 \times 10^{-3} T^2 + 5.538 \times 10^{-7} T^3)$$
(6)

It should be noted that the resistance of pore diffusion plays an important role in the reaction rate because of the very fine pore size of  $\beta$ -zeolite. However, it was assumed that this effect was included into the kinetic parameters obtained from the data fitting.

By performing the material balance for a semibatch reactor, the following expressions are obtained.

$$-\frac{dm_{\text{TBA}}}{dt} = \frac{dm_{\text{H}_2\text{O}}}{dt} = W(r_1 + r_2)$$
(7)

$$-\frac{\mathrm{d}m_{\mathrm{EtOH}}}{\mathrm{d}t} = \frac{\mathrm{d}m_{\mathrm{ETBE}}}{\mathrm{d}t} = Wr_1 \tag{8}$$

where  $m_i$  and W represent the number of mole of species *i* and the catalyst weight, respectively. It is noted that the number of moles in the liquid phase at any time is constant because IB can only slightly dissolved in the liquid phase. In addition, every 1 mol of TBA consumption produces 1 mol of water, and every 1 mol of EtOH consumption produces 1 mol of ETBE. The activity can be calculated from the following relation.

$$a_i = \gamma_i x_i \tag{9}$$

where  $x_i$  is mole fraction of species *i* in the liquid mixture and  $\gamma_i$  is the activity coefficient. The activity coefficients can be calculated using the UNIFAC method [19].

*Kinetic Parameter Determination.* A set of experiments was carried out at three temperature levels of 323, 333, and 343 K to investigate the kinetic parameters. Figure 5 shows typical results of mole changes with time at T = 343 K. The initial moles of each species are given in the figure legend. It should be noted that the experimental results showed good agreement with the simulation results. In addition, the experimental results showed that the production of ETBE became higher with the increase of temperature as expected in the Arrhenius's equation.

A curve fitting method was employed to find the kinetic parameters  $k_1$ ,  $k_2$ , and  $K_w$  at each temperature. Initial guess values of the parameters  $k_1$  and  $k_2$  were obtained by using an initial rate method [20]. The lines in the figure represent the simulation results. It was found that within the ranges of the study, the model fit the experimental results well with the sum square



**Figure 5** Mole changes with time (catalyst =  $\beta$ -zeolite, catalyst weight = 15.0 g,  $m_{\text{TBA},0} = 1.91$  mol,  $m_{\text{EtOH},0} = 1.98$  mol,  $m_{\text{ETBE},0} = 0.01$  mol,  $m_{\text{H}_2\text{O},0} = 0.10$  mol, T = 343 K, and reactor type = basket reactor).

of the residual (at T = 343, 333 and 323 K) of 0.061, 0.099 and 0.002, respectively.

Figure 6 shows the Arrhenius's plot of the reaction rate constants and the van't Hoff's plot of the adsorption parameters using the results at three temperatures. The following equations were determined from the plots.

$$k_1 = \exp(3.55 - 2286/T) \tag{10}$$

$$k_2 = \exp(36.57 - 13653/T) \tag{11}$$

$$k_{\rm W} = \exp(-16.16 + 6636/T) \tag{12}$$

Table I compares the values of the activation energy of the reactions (1) and (2) and the heat of adsorption of water with those values of different catalysts. It was observed that the heat of adsorption of water of  $\beta$ -zeolite catalyst was quite close to the values of other catalysts except that of HPA which showed significant water-inhibition effect. The activation energy of the dehydration reaction of  $\beta$ -zeolite was within the range



Figure 6 Arrhenius and van't Hoff plots.

Catalysts	Activation Energy of Reaction (1) (kJ/mol)	Activation Energy of Reaction (2) (kJ/mol)	Heat of Adsorption of Water (kJ/mol)	Ref.
β-zeolite	19	114	55	This work
Amberlyst-15	57	85	63	[12]
HPA	95	108	20	[12]
S-54	48	74	67	[14]
D-72	43	30	63	[14]
Amberlyst-15	_	142	55	[21]

Table I Comparison of Activation Energy and Heat of Adsorption of Water Among Different Catalysts

reported in the other catalysts. This indicates that the production of the undesired product, IB was greatly enhanced with increasing temperature. However, the activation energy of the etherification reaction (1) of  $\beta$ -zeolite was much lower than those of the other catalysts. This may indicate the pore-diffusion limitation arising from the transport of large molecule ETBE from the pore of  $\beta$ -zeolite.

The obtained rate expressions will be used in our subsequent studies on a reactive distillation column with and without a pervaporation unit and a pervaporative membrane reactor; however, it should be noted that it is desirable to operate the reactor at low operating temperature. This is because the activation energy of the dehydration reaction (2) is higher than the main reaction (1), and consequently the selectivity to ETBE decreases with increasing temperature.

# CONCLUSION

It was found that  $\beta$ -zeolite showed superior performance over the commercial Amberlyst-15 for the direct production of ETBE from TBA and EtOH. Even though the activity was moderate, the selectivity was much higher. The kinetic study of the reaction catalyzed by  $\beta$ -zeolite supported on monolith was carried out by using a semibatch reactor. The effect of external mass transfer was negligible at the stirring speed higher than 1210 rpm. The activity-based model taking into account of the effect of water inhibition was developed. Three temperature levels of 323, 333, and 343 K were used in the study to obtain the parameters in the Arrhenius's equation of the reaction rate constants and the van't Hoff's equation of the water inhibition coefficient.

# NOMENCLATURE

- $a_i$  Activity of species i
- ETBE Ethyl *tert*-butyl ether, 2-ethoxy 2-methyl propane (IUPAC)

#### EtOH Ethanol

H<sub>2</sub>O Water

- $k_1$ Reaction rate constant of reaction (1) in the<br/>activity-based model (mol kg^{-1} s^{-1}) $k_2$ Reaction rate constant of reaction (2) in the<br/>activity-based model (mol kg^{-1} s^{-1})
- $K_1$  Equilibrium constant of reaction (1) in the activity-based model
- *K*<sub>W</sub> Water inhibition parameter in the activitybased model
- $m_i$  Number of mole of species  $i \pmod{i}$
- $r_j$  Reaction rate of reaction  $j \pmod{\text{kg}^{-1} \text{s}^{-1}}$
- t Reaction time (s)
- *T* Temperature
- TBA *tert*-Butyl alcohol, 2-methyl 2-propanol (IUPAC)
- W Catalyst weight (kg)
- $x_i$  Mole fraction of species *i* in liquid mixture
- $\gamma_i$  Activity coefficient of species *i*
- ()<sub>0</sub> Initial value a t = 0

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