

ADSORPTION OF MIXED ORGANIC SOLVENT BY Y TYPE ZEOLITE

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ABSTRACT

Fixed-bed adsorption experiments of laboratory-scale were carried out to remove organic solvent vapors by Y type high silica zeolite. Experimental results for binary component system, which showed azeotropic mixture systems, could be correlated satisfactorily.

Keywords

azeotropic adsorption, breakthrough curves, Stop & Go method

1. INTRODUCTION

Discharge of organic substances into air has been strongly prohibited since some decades ago, to preserve comfortable natural environment. Though hydrocarbons, alcohols and chlorinated hydrocarbons had been used as the degreasing agent in industries, these solvents may also affect to our environment, and it is necessary to remove them from air as much as possible. This study was performed aiming at presenting useful data for the design of adsorption processes, especially the removal of the solvent vapors as above from air by adsorption. The solvents used were trichloroethylene (TCE) and ethanol (EtOH). Moreover, the simulation in the same EtOH-TCE system as an experiment was performed using the STOP & GO method which calculates substance income and outgo for every cell.

2. EXPERIMENTAL

2.1 Adsorbent

Fixed-bed adsorption experiments of laboratory-scale were carried out to remove organic solvent vapors by Y type high silica zeolite. Experimental results for binary component system, which showed azeotropic mixture systems, could be correlated satisfactorily.

2.2 Experimental Procedure

Solvents used were ethanol (EtOH) and trichloroethylene (TCE). Adsorbents were packed in a glass column of 0.10m length and 0.0156m i.d. Experimental conditions were as follows: linear flow rate; 0.2 m/s, influent concentration; in the range of 0.004~0.2 mol/m³, and temperature; 298 K. Experimental apparatus are shown in Figure 1.

2.3 Repeated experiment Procedure

After the adsorption run, desorption was performed by circulating only dry air in a column, and the outflow gas concentration from the column exit was measured by gas chromatograph. These adsorption and desorption run were continuously repeated 2~3 times for an EtOH-TCE system and the total concentration of 2500~3500 ppm.

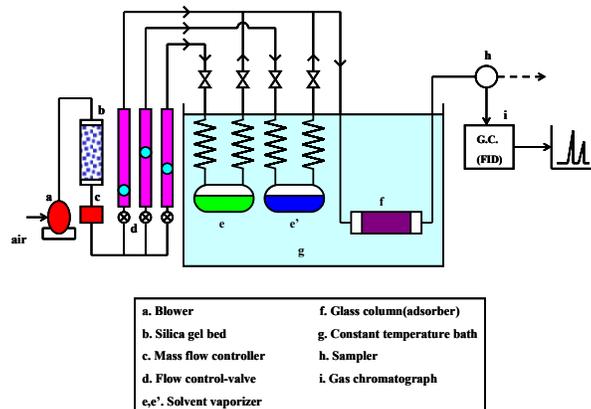


Figure 1: Schematic diagram of experimental apparatus for adsorption

3. RESULT AND DISCUSSION

3.1 Breakthrough Curves

Figure 2 show several types of breakthrough curves obtained for EtOH-TCE -Y-type zeolite system. In Figure 2, the range of the total concentration of EtOH and TCE are 2500~3500ppm. For this system reversal of the order of breakthrough (turn over) occurred once at concentrations of 0.222 mole fractions of EtOH. When the mole fractions were 0.222, the mixture of two components behaved as if it was a single component system.

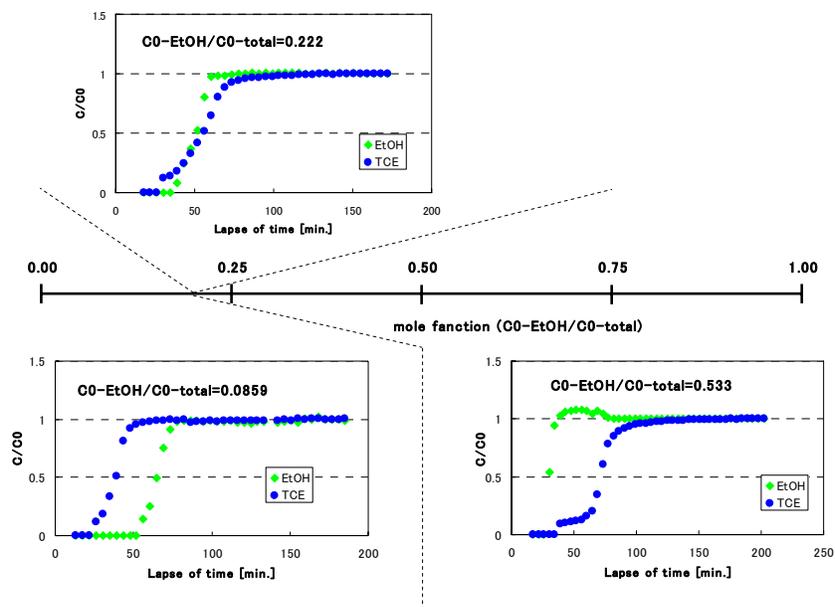


Figure 2: Several types of breakthrough curves observed for EtOH-TCE-Y-type system (2500~3500ppm)

3.2 Adsorption Equilibria

Binary adsorption equilibria for the system of EtOH-TCE of several total concentration are shown in Figure 3 in form of X-Y diagram (X and Y are the mole fractions of gas phase concentration and the amount adsorbed at equilibrium, respectively). Those equilibria have one azeotropic point, which attribute to the reversal of breakthrough order.

For vapor-liquid equilibrium, when the saturation vapor pressures of each component were almost the same, appearance of one azeotropic points were reported. It is thought, therefore, that the phenomenon occurred in adsorption equilibrium for these systems can be ascribed to the fact that the saturation vapor pressure and the boiling points of each component were almost the same.

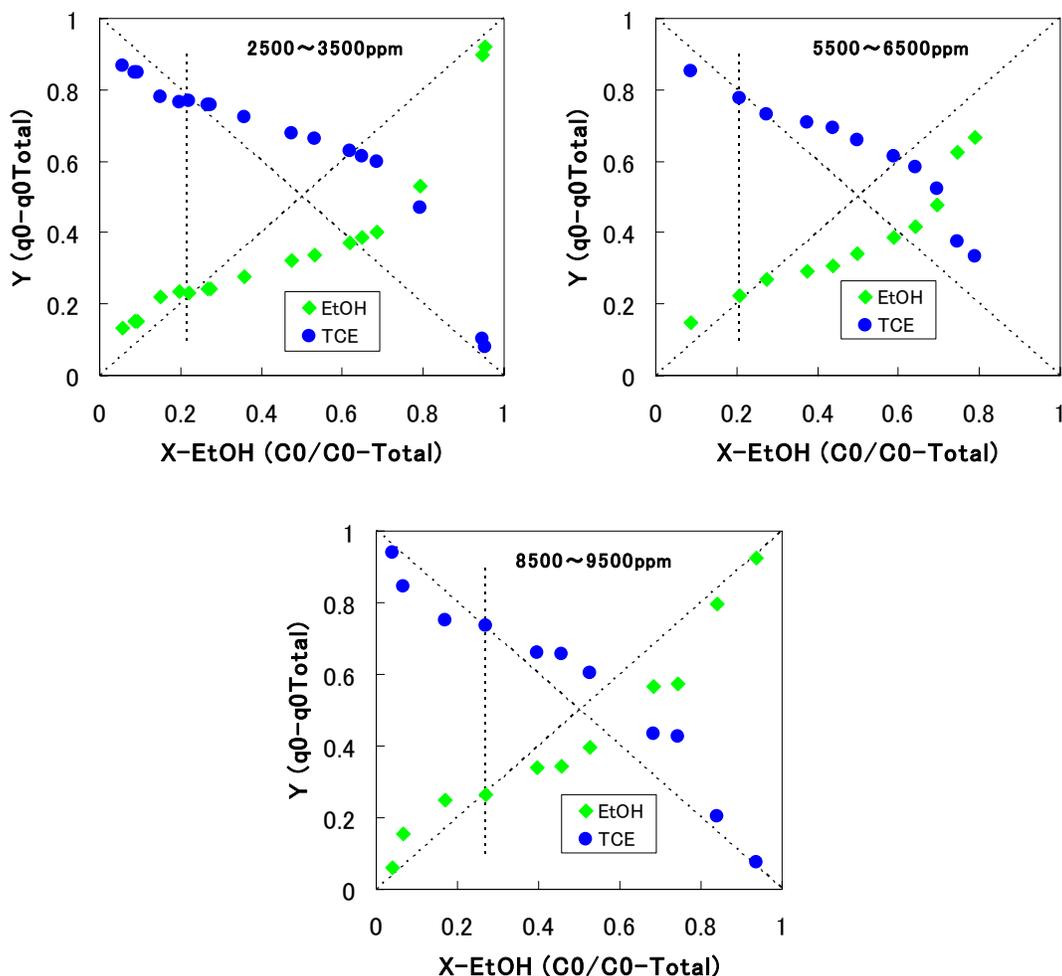


Figure 3: Comparison of adsorption equilibrium for experimental concentration 2500~3500, 5500~6500, 8500~9500ppm on EtOH-TCE-Y-type zeolite system

3.3 Repeated Experiment

After the first adsorption run, desorption and next adsorption were repeated twice to check the reproducibility. As for the obtained breakthrough curve, the results of EtOH and TCE overlapped mostly in the first time, the second time, and the third time (Figure 4). Therefore, it can be said that the solvent did not remain in the adsorbent after desorption. It turned out that degradation by using Y type high silica

zeolite repeatedly was seldom seen. In consequence, this adsorption can be reproduced and this azeotropic adsorption is reversible phenomenon.

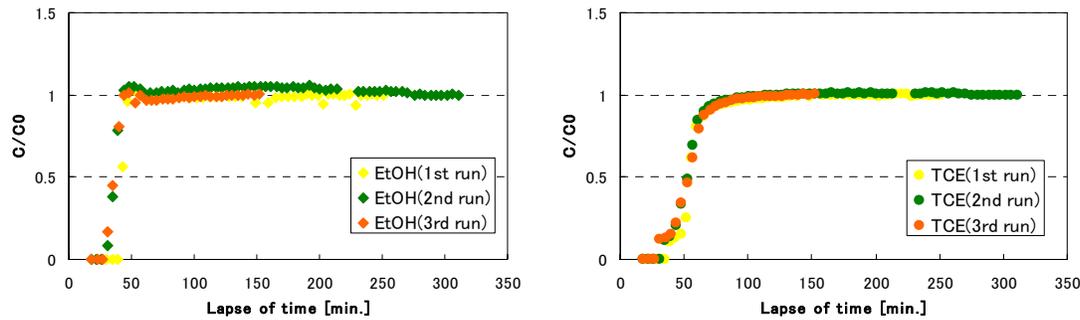


Figure 4: breakthrough curve obtained in the repetition experiment

3.4 Stop & Go Method

(1) Stop & Go Method: Figure 5 shows a concept of Stop & Go method. The Stop & Go method used in this study can be explained briefly as follows. Each column is divided to N complete mixed cells in axial direction. While the flow between cells is stopped for a certain short time (dt), the adsorbed or desorbed amount of the adsorbate within this time interval is calculated (Stop calculation). After this Stop calculation, the flows between cells are calculated by using inflow, remained gas amount and column pressure for dt time (Go calculation). These Stop and Go calculations within dt are repeated alternatively to get the pulse response.

(2) Simulation Result: Since Langmuir equation was not able to be used for binary systems, in which adsorption equilibrium data, one azeotropic points were observed, the following calculation was performed. Then, the equation of the azeotropic theory in a liquid-vapor azeotropic equilibrium was applied instead of Langmuir equation. The equation of azeotropic theory in a liquid-vapor azeotropic equilibrium applied for this simulation is shown.

$$y_i \gamma_i P_i^* = x_i P \quad (1)$$

where P_i^* is the vapor pressure and γ_i the activity coefficient of component i.

The activity coefficient was first calculated from experiment value. The relation between a gaseous phase molar fraction and the activity coefficient are shown in Fig.6. γ vs. X equation was obtained by curve fitting of this figure. This equation was substituted into Eq. (1) to get two correlations between a gaseous phase molar fraction and an adsorption layer molar fraction as shown in Figure 7. Stop & Go [1] method for simulation of breakthrough curve with these correlations (Figure 6, 7 and 8) was applied to get Figure 9. Substitution of the order of breakthrough was seen once in the simulation as well as the experiment. Also breakthrough curves were almost simulated in all cases.

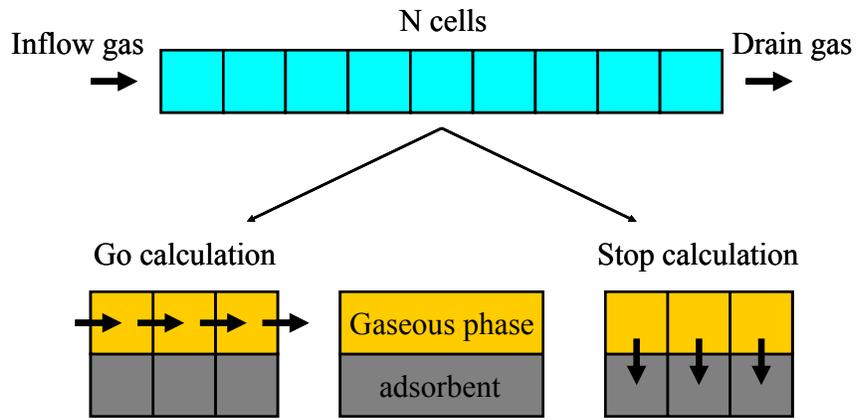


Figure 5: A concept of Stop & Go method

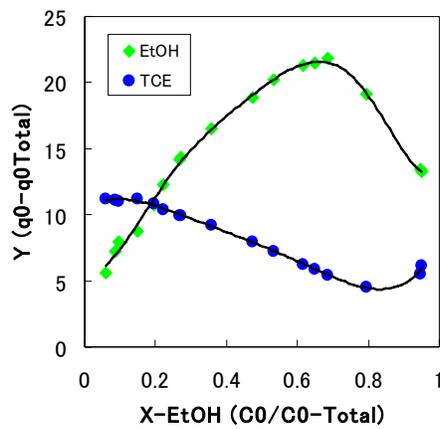


Figure 6: The relation between a gaseous phase Molar fraction and the activity coefficient

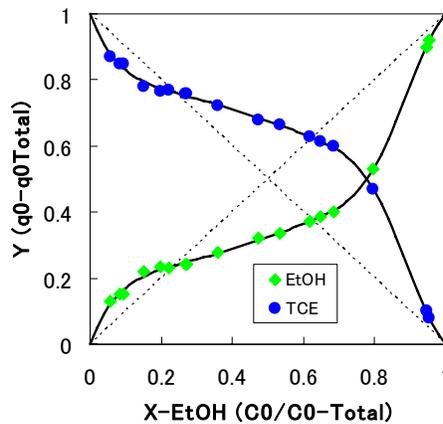


Figure 7: Comparison of the actual measurement of adsorption equilibrium data and estimated value

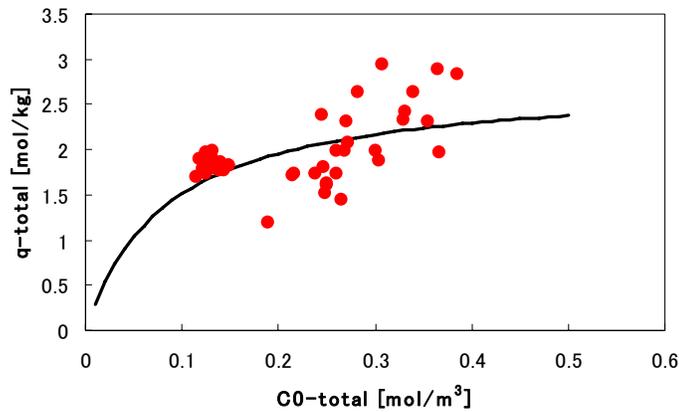


Figure 8: adsorption isotherm as the total of EtOH and TCE

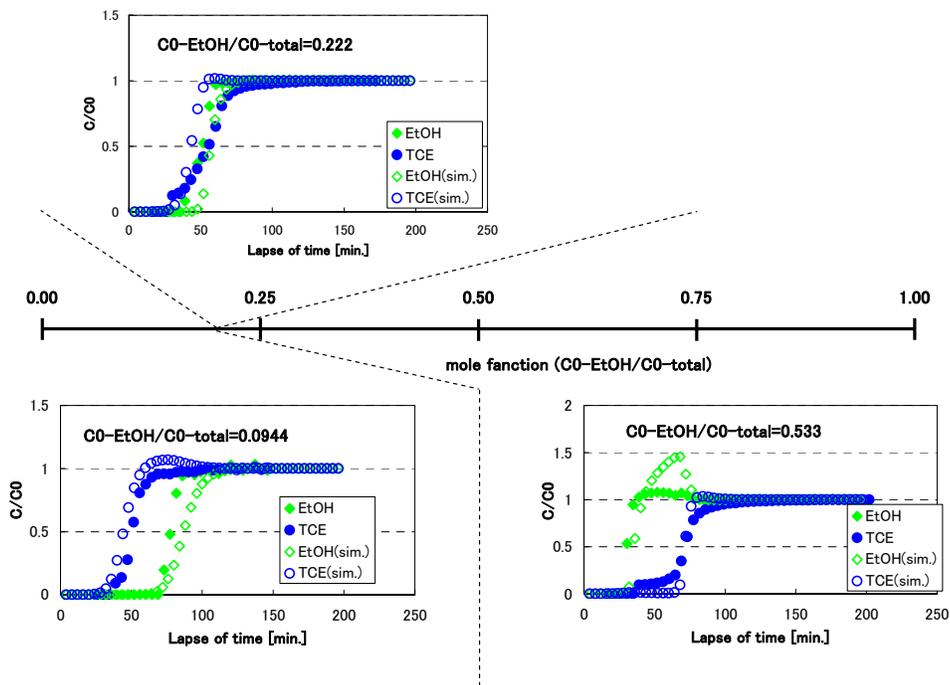


Figure 9: Comparison of the breakthrough curves for experiments and simulations

4. CONCLUSION

Adsorption of EtOH-TCE vapors onto HSZ was studied. One azeotropic points appeared in the adsorption equilibrium for this EtOH-TCE -Y-type system. For this binary systems, adsorption equilibrium data could be expressed by proposed equation, similar to liquid-vapor azeotropic equilibrium equation. Moreover, It can be said from the repetition experiment of adsorption and desorption that adsorption is an reversible physical phenomenon. Breakthrough curve could be simulated using the Stop & Go method in the whole range for azeotropic mixture systems.

REFERENCE

1. K Chihara et al., Proceeding of the 7th International Zeolite Conf, 563-570 (1986)