

**MATHEMATICAL MODELING AND EXPERIMENT
OF BREAKTHROUGH CURVE FOR DEACTIVATED ADSORBENTS
PACKED IN A MULTI-LAYER GAS ADSORBER:
CASE OF HIGH DEACTIVATION**



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ABSTRACT

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A multi-layer adsorber consisting of commercial activated alumina and two sizes of 4A molecular sieve is one system that is used to remove water from natural gas. The removal of vapour water from natural gas passing through the adsorber can be described by using the modeling of breakthrough time based on mass balance. To predict the breakthrough time, which changes along the time of service, the adsorbents were acceleratingly aged, especially the molecular sieve, which was studied only at the low degrees of deactivation in previous works. Therefore, in this work, the two sizes of molecular sieve were boiled at 600°C, and the number of batch cycles was varied to increase the aging. The SEM results showed that the average crystal size decreased when the number of batches increased. Also, the parameters in the mass balance equation and the water adsorption isotherms of adsorbents changing with the percentage of deactivation were determined to be used in the breakthrough time model in accordance with deactivation. It was found that the curve of the adsorption isotherm becomes more flat with higher degrees of deactivation, and Aranovich-Donohue for Toth was employed to explain the adsorption isotherm at various degrees of deactivation. Then, the breakthrough time from the experiments and theorem was compared which it was found that the modified mathematical model gave good agreement with the experiments. Additionally, the breakthrough time prediction for any degree of deactivation can be

accomplished by using the predicted adsorption isotherm that was written as a function of degree of deactivation.

บทคัดย่อ

สังขรรณ พลแสน : แบบจำลองทางคณิตศาสตร์และการทดลองสำหรับการเบรคทูรของน้ำในหอดูดซับที่บรรจุด้วยชั้นของตัวดูดซับหลายชนิด: กรณีที่ตัวดูดซับมีระดับการเสื่อมสภาพสูง (Mathematical Modeling and Experiment of Breakthrough Curve for Deactivated Adsorbents Packed in a Multi-Layer Gas Adsorber: Case of High Deactivation) อ. ที่ปรึกษา : ผศ. ดร. ศิริรัตน์ จิตการคำ, ผศ. ดร. กิติพัฒน์ สีมานนท์, 90 หน้า

หอดูดซับที่บรรจุด้วยตัวดูดซับหลายชนิดที่ประกอบด้วยตัวดูดซับทางการค้า อะลูมินา และโมเลกุลาสีฟชนิด 4A สองขนาด (1/16 และ 1/8 นิ้ว) นั้น เป็นระบบหนึ่งที่ใช้ในการดูดซับน้ำออกจากก๊าซธรรมชาติ ปริมาณน้ำที่ถูกกำจัดน้ำออกจากก๊าซธรรมชาติด้วยหอดูดซับนี้ สามารถหาได้จากแบบจำลองเวลาของการเบรคทูรโดยใช้หลักการสมดุลมวล ที่ใช้ในการที่จะทำนายเวลาของการเบรคทูรที่ซึ่งเปลี่ยนแปลงไปตามเวลาของการใช้งาน ตัวดูดซับได้ถูกเร่งให้มีการเสื่อมสภาพ โดยเฉพาะโมเลกุลาสีฟ เนื่องจากได้ถูกศึกษาไปแล้วในระดับที่มีการเสื่อมสภาพต่ำ ดังนั้นในงานนี้โมเลกุลาสีฟทั้งสองขนาดได้ถูกดัมที่อุณหภูมิ 600 องศาเซลเซียส โดยที่เปลี่ยนแปลงจำนวนครั้งของการดัมเพื่อเพิ่มระดับการเสื่อมสภาพ หลังจากตัวดูดซับได้ถูกเตรียมให้มีระดับการเสื่อมสภาพที่ต่างกันไปแล้ว ค่าขนาดเฉลี่ยของผลึกได้ถูกวิเคราะห์โดยใช้เครื่องสแกนนิ่งอิเล็กตรอนไมโครสโคป (SEM) ซึ่งพบว่าขนาดผลึกลดลงเมื่อระดับการเสื่อมสภาพเพิ่มขึ้น นอกจากนี้ได้ทำการทดลองหาค่าตัวแปรต่างๆ ในสมการสมดุลมวลและไอโซเทอร์มของการดูดซับที่เปลี่ยนแปลงตามการเสื่อมสภาพเพื่อนำไปใช้ในแบบจำลองเวลาของการเบรคทูรในกรณีตัวดูดซับเสื่อมสภาพ จากการศึกษาไอโซเทอร์มของการดูดซับพบว่า กราฟของไอโซเทอร์มแบนราบมากขึ้นเมื่อระดับการเสื่อมสภาพเพิ่มสูงขึ้น และแบบจำลองของ Aranovich-Donohue ที่ปรับปรุงใช้กับสมการของ Toth สามารถอธิบายไอโซเทอร์มของการดูดซับได้ครอบคลุมทั้งระดับการเสื่อมสภาพที่ต่ำและสูง หลังจากนั้น เมื่อนำเอาไอโซเทอร์มที่ได้ไปใช้ในแบบจำลองเพื่อทำนายเวลาของการเบรคทูร พบว่าเวลาของการเบรคทูรที่ได้นั้น สอดคล้องกับค่าที่ได้จากการทดลองในห้องปฏิบัติการ นอกจากนี้ไอโซเทอร์มของการดูดซับยังสามารถทำนายได้โดยเขียนเป็นฟังก์ชันกับระดับการเสื่อมสภาพ ทำให้สามารถทำนายเวลาของการเบรคทูรที่ค่าการเสื่อมสภาพใดๆ ได้

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| C Porosity as a function of the ration of particle diameter to bed diameter (Leva, 1947). | 79 |

LIST OF SYMBOLS

SYMBOL

| | |
|-----------|---|
| $a(t)$ | activity of catalyst |
| t | time, (sec) |
| r_d | rate of catalyst decay |
| F_A | molar fluxes of a species A |
| W_A | molar flux of a species A |
| v | interstitial velocity of fluid, (cm/s) |
| \bar{q} | adsorbate concentration averaged over crystal and pellet |
| z | distance measured from column inlet, (cm) |
| c_A | adsorbate concentration in fluid phase, (mol/l) |
| q^* | equilibrium adsorbed phase concentration |
| D_e | effective diffusivity, (cm ² /s) |
| D_L | axial dispersion coefficient, (cm ² /s) |
| D | diameter of particle in the bed, (cm) |
| G | superficial mass velocity, (kg/m ² s) |
| A | cross section area of bed, (cm ²) |
| k | overall mass transfer coefficient, (1/sec) |
| K | equilibrium constant |
| k_f | external fluid film mass transfer coefficient, (cm/s) |
| R_p | radius of adsorbent pellet, (cm) |
| r_p | pore radius, (cm) |
| D_k | Knudsen diffusivity, (cm ² /s) |
| D_m | molecular diffusivity or bulk diffusivity, (cm ² /s) |
| M | molecular weight of diffusing species |
| T | temperature, (K) |
| M_A | average molecular weight of bulk species |
| M_B | molecular weight of adsorbate species |
| M_B | molecular weight of adsorbate species |
| P | total pressure, (atm) |

**GREEK
LETTERS**

| | |
|-----------------|---|
| ε | bed void fraction |
| ε_p | porosity of particle |
| μ | viscosity of mixing gas, (g/cms) |
| ρ | density of mixing gas, (g/cm ³) |
| ρ_b | bulk density, (g/cm ³) |
| τ | tortuosity factor |
| σ_{AB} | collision diameter from Lennard-Jones potential |
| Ω_{AB} | collision integral |