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Retraction

Retracted: The Adsorption of VOCs by Honeycomb Ceramics Loaded with Molecular Sieves

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This article has been retracted by Hindawi following an investigation undertaken by the publisher [1]. This investigation has uncovered evidence of one or more of the following indicators of systematic manipulation of the publication process:

1. Discrepancies in scope
2. Discrepancies in the description of the research reported
3. Discrepancies between the availability of data and the research described
4. Inappropriate citations
5. Incoherent, meaningless and/or irrelevant content included in the article
6. Peer-review manipulation

The presence of these indicators undermines our confidence in the integrity of the article's content and we cannot, therefore, vouch for its reliability. Please note that this notice is intended solely to alert readers that the content of this article is unreliable. We have not investigated whether authors were aware of or involved in the systematic manipulation of the publication process.

Wiley and Hindawi regrets that the usual quality checks did not identify these issues before publication and have since put additional measures in place to safeguard research integrity.

We wish to credit our own Research Integrity and Research Publishing teams and anonymous and named external researchers and research integrity experts for contributing to this investigation.

The corresponding author, as the representative of all authors, has been given the opportunity to register their agreement or disagreement to this retraction. We have kept a record of any response received.

References

[1] P. Zhang, "The Adsorption of VOCs by Honeycomb Ceramics Loaded with Molecular Sieves," *Journal of Chemistry*, vol. 2022, Article ID 7207403, 7 pages, 2022.

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Research Article

The Adsorption of VOCs by Honeycomb Ceramics Loaded with Molecular Sieves

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In order to improve the adsorption performance of molecular sieve honeycomb ceramics for VOCs, the author proposes a research method for the adsorption performance of VOCs based on the honeycomb ceramics loaded with molecular sieves. The method prepared cordierite honeycomb ceramic-supported HY, Na Y, USY, ZSM-5-300, and ZSM-5-360 monolithic adsorption materials by the coating method. Using toluene, ethyl acetate, isopropanol, and acetone as adsorbates, the adsorption properties of single-component and mixed-component organic compounds on the supported molecular sieve honeycomb ceramics were investigated. The result shows ZSM-5-300 has the largest adsorption capacity for the four adsorbates, which are toluene 68.0 mg/g, ethyl acetate 118.7 mg/g, isopropanol 70.9 mg/g, and acetone 72.5 mg/g. The test results show that ZSM-5-300 molecular sieve has high saturated adsorption capacity and is an ideal VOCs adsorption material.

1. Introduction

The development of modern industry, while bringing us good quality and cheap goods, also brings environmental problems. In recent years, the frequent occurrence of haze weather has seriously affected people's life and health; volatile organic compounds are one of the main reasons for the formation of haze. Volatile organic compounds (VOCs) refer to the saturated vapor pressure greater than 70 Pa at room temperature, an organic compound with a boiling point below 260°C under normal pressure. According to chemical structure, it can be divided into alkanes, alkenes, alkynes, aldehydes, acids, esters, alcohols, benzene series, halogenated hydrocarbons, ketones, acids, and others, a total of 12 kinds of substances. It has the characteristics of many types, complex components and long release period; it is a kind of chemical substance with "three hazards" (carcinogenic, teratogenic, and mutagenic) to the human body; it participates in photochemical reactions and causes the greenhouse effect; at the same time, it is also the main material for the formation of atmospheric particulate matter, which is harmful to the atmospheric environment and human body [1]. Volatile organic compounds (VOCs) are toxic and polluting organic compounds; it comes from various fields of industrial production, motor vehicle emissions, and human activities, and the emissions are huge. The catalytic combustion method is clean and efficient, with low reaction temperature and low energy consumption, and is an effective means to control the emission of VOCs [2].

According to amendments to the Clean Air Act passed by the United States in 1990, of the nearly 190 air pollutants that are strictly controlled, about 70% are volatile organic compounds. In May 2010, the General Office of the State Council forwarded the "Notice of Guiding Opinions on Promoting Joint Prevention and Control of Air Pollution to Improve Regional Air Quality" (Guobanfa (2010) No. 33) issued by the Ministry of Environmental Protection and other departments; for the first time, the pollution prevention and control of VOCs was proposed at the national level. The guidance states "engaged in production operations that emit VOCs such as spray painting, petrochemicals, shoe industry, and electronic products, pollution control should be carried out in accordance with specific relevant technical specifications." The treatment of VOCs has become one of the major problems to be solved urgently in contemporary environmental pollution [3].

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2. Literature Review

Due to the harm of VOCs to the environment and human body, all countries in the world have formulated relevant laws and regulations on the emission of VOCs and minimize or even eliminate the emission of VOCs [4]. Although it is possible to transform the production process and process, improve the production equipment, so that it can achieve zero emissions in the production process. However, limited by the level of production technology and cost pressure, most ways to control the emission of VOCs can only be in the exhaust gas treatment stage. There are two general methods for controlling VOCs emission content: recycling technology and destruction technology [5]. The so-called recycling technology is to recover and recycle VOCs, which are generally used for VOCs with high concentration or relatively economical value. Destruction technology is to degrade and destroy VOCs to generate CO_2 and H_2O and other harmless substances, which are suitable for medium and low concentrations of VOCs [6]. Common technologies for recovering VOCs mainly include adsorption technology, absorption technology, condensation technology, and membrane separation technology. Common technologies for destroying VOCs mainly include combustion technology, photocatalytic degradation technology, biodegradation technology, and plasma technology. These technologies for removing VOCs have their own advantages and disadvantages; in actual operation, one or a combination of several is selected according to the specific conditions of the air volume, concentration, type, and economic value of VOCs in the exhaust gas [7].

Adsorption is the process of converting a mixture into two or more products with different components using an adsorbent. Adsorbents are often porous solid materials. The adsorption method is widely used in the treatment of organic waste gas; due to its low adsorption capacity, the adsorption method is mainly used for the purification of organic waste gas with low concentration and large air volume [8]. Organic waste gases that have been successfully treated by adsorption are the painting process (toluene, xylene, and benzene), organic solvent volatilization, acetone waste gas, ethyl acetate, paint production waste gas styrene, and xylene. The key to adsorption technology is the adsorbent. Commonly used adsorbents for adsorbing organic waste gas are activated carbon particles, activated carbon fibers, molecular sieves, and adsorption resins. The advantages of adsorption technology are high removal rate, the removal rate can reach more than 95%, no secondary pollution, high purification efficiency, convenient operation, and automatic control [9].

Molecular sieve is a natural zeolite or synthetic aluminosilicate, which has the functions of sieving molecules, adsorption, ion exchange, and catalysis; it is mainly composed of silicon, oxygen, aluminum, and other metal cations, and it has high thermal stability and chemical stability [10, 11]. Since the 1970s, a large number of artificial zeolite molecular sieves have been synthesized; because of their unique channel zeolite molecular sieves, they have become indispensable catalytic materials and adsorption materials in petrochemical and fine chemicals and are used in more and more fields. In the 1940s, zeolite molecular sieves were artificially synthesized for the first time; up to now, more than 200 kinds of zeolite molecular sieves with framework structures have been found. Sodium silicate and sodium aluminate are commonly used as raw materials to synthesize molecular sieves. However, due to the problems of high energy consumption, high environmental pollution, and low efficiency in the synthesis process, it is gradually replaced by the synthesis method using kaolin and diatomite as raw materials.

Yda et al. synthesized ZSM-5 molecular sieve with kaolin as raw material by the nonorganic template method; the samples were characterized by XRD, SEM, FTIR, N_2 adsorption and desorption, and temperature-programmed elution, and its catalytic cracking performance was studied, and the results showed that the molecular sieve has high hydrothermal stability and strong acidity, which is beneficial to the formation of propylene and butene in oil [12]. Enesca A. et al. developed three molecular sieve adsorption runners for the separation and concentration of different VOCs and pointed out that Y-type molecular sieves were used for amines, esters, ethers, and macromolecular VOCs; Y-type and ZSM composite molecular sieves are used for aromatic hydrocarbons, ethers, alcohols, and aldehydes; ZSM molecular sieves are used for ketones and chlorinated alkanes (alkenes) [13].

The authors prepared cordierite honeycomb ceramic-supported HY, Na Y, USY, ZSM-5-300, and ZSM-5-360 monolithic adsorption materials by the coating method. Using toluene, ethyl acetate, isopropanol, and acetone as adsorbates, the adsorption properties of single-component and mixed-component organic compounds on the supported molecular sieve honeycomb ceramics were investigated, which has an important value for industrial application.

3. Research Methods

3.1. VOCs Control Technology. The pollution prevention and control of VOCs mainly manages the source and process of pollution sources by formulating standards, improving systems, improving energy structure, changing raw materials, and improving technological processes; however, most of the exhaust gas emitted in the production process adopts terminal treatment, which is mainly divided into two technologies: destruction and recycling [14, 15]. Destruction technology mainly converts toxic and harmful organic pollutants into nontoxic, harmless, or low-toxic substances through chemical reactions and discharges them into the atmosphere, including the catalytic combustion method, photocatalytic method, biodegradation method, and low temperature plasma method, the recovery technology mainly separates and enriches organic pollutants through physical methods such as temperature, pressure, and filtration and realizes the resource utilization of VOCs, including the adsorption method and condensation method [16, 17]. There are many types of VOCs with different properties; according to the types, concentrations, and emission methods of VOCs, VOCs treatment technologies

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have different applications in actual production, and even a combination of several treatment technologies is used. Different treatment technologies have different characteristics and application ranges; in actual projects, the most suitable technology is selected according to the type of VOCs, gas flow, and concentration; condensation and membrane separation technologies are mostly used to treat VOCs gases with a concentration greater than 10000 mg/m^3; catalytic combustion and adsorption are mostly used to treat VOCs gases with a concentration of 2000–10000 mg/m^3, and biodegradation and plasma technologies are mostly used to treat concentrations less than 2000 mg/m^3 VOCs gas [18]. Figure 1 shows common VOCs treatment technologies:

3.2. Preparation of Loaded Honeycomb Ceramics. Commercial powder HY molecular sieve (Si/Al = 5.1, specific surface area 548 m^2/g), NaY (Si/Al = 3.0, specific surface area 581 m^2/g), US (Si/Al = 7.8, specific surface area 545 m^2/g), ZSM-5-300 (Si/Al = 300, specific surface area 358 m^2/g), and ZSM-5-360 (Si/AI = 360, specific surface area 346 m^2/g) were taken.

Weigh an appropriate amount of the above 5 kinds of molecular sieves with an electronic balance and put them in a 200 mL beaker, add silica sol binder, dilute to a certain concentration with deionized water, stir for 30 min, slowly immerse the prepared honeycomb ceramics in the above solution, soak for 15 min, dry at 100°C for 1 h, roast in a muffle furnace at 400°C for 2 h, and cool to room temperature for later use.

3.3. Characterization of Adsorbents. The specific surface area of molecular sieves was measured by N_2 adsorption on the JW-BK200C specific surface and pore size analyzer; before the measurement, the samples were degassed at 200°C for 6 hours and calculated by the BET multipoint method.

3.4. Adsorption Experiments. The entire adsorption system of the experiment consists of a VOCs generator, a gas flow control system, a temperature control system, and an adsorption device. The size of the honeycomb ceramics used in the experiment is 100 × 100 × 50 mm, and the pore density is 200 holes/im^2; the abovementioned honeycomb ceramics are processed into cylinders with a diameter of 2.0 cm, and 4 pieces are one unit, which are placed in a quartz glass tube with a diameter of 2.2 cm. The VOCs adsorption experiment uses air as the carrier gas; after being dried and purified by the air purifier, it is divided into 3 paths through a four-way pipe and enters the flowmeter of the 3 gas paths, respectively; one of them passes through a mass flow meter and an organic gas generator (placed in an ice-water bath) bring out a certain concentration of VOCs gas; 1 pass through the mass flow meter into the water vapor generator (placed in a 40°C waterbath) and bring out a certain concentration of water vapor. The first route is the dilution gas, and the last 3 routes are merged into 1 route gas. Adjust the flow of the 3-way gas through the flow meter to prepare the inlet gas with a certain VOCs concentration, relative humidity, and air velocity. When the VOCs content and relative humidity at the inlet concentration no longer change, it is connected to the adsorption device for experiments. The inlet concentration of VOCs before the reaction and the outlet concentration after the reaction were detected by a gas chromatograph (GC1120). The total gas flow rate is 0.32 m^3/h, the single-component organic matter experiment, and the adsorbate concentration is 500 mg/m. In the multicomponent organic matter experiment, the concentration of each adsorbate is 125 mg/m^3, the outlet VOCs concentration reaches 10% of the inlet concentration as the breakthrough point, and when it reaches 90%, it is saturated adsorption, and the VOCs adsorption amount is calculated through the integration of the breakthrough curve; the formula is

$$q = \frac{F \times C_0}{60 \times W}\left[t_s - \int_0^{t_s} \frac{C_i}{C_0} \mathrm{d}t\right]. \tag{1}$$

In the formula, q is the adsorption amount of organic gas, mg/g; F is the total gas flow rate, m^3/h; C_0 is the initial concentration of inlet VOCs, mg/m^3; t_s is the adsorption time, min; C_i is the concentration of VOCs at the min exhaust outlet, mg/m^3; W is the adsorbent loading, g.

4. Results Analysis

4.1. Phase Confirmation of Molecular Sieves. Figure 2 shows that HY, NaY, USY, ZSM-5-300, and ZSM-5-360 molecular sieves are mixed with silica sol (mass ratio is 9 : 1), respectively; XRD pattern after calcination is at 400°C. It is completely consistent with the corresponding Y-type molecular sieve XRD spectrum (PDF45-0112) and ZSM-5 molecular sieve XRD spectrum (PDF44-0003), and no SiO_2 diffraction peak is observed; it shows that SiO_2 is in a highly dispersed state, only for bonding. Molecular sieves are calcined at 400°C and their structures remain stable [19].

4.2. Adsorption Performance of Molecular Sieves for Single-Component VOCs. Using toluene and ethyl acetate as adsorbates, the adsorption properties of five molecular sieves were determined, respectively. Figure 3 shows that the five molecular sieves have obvious "shoulder peaks" in the toluene penetration curves, the reason may be that the polarities of toluene and water are quite different, and the Y-type molecular sieves with low silica-alumina ratio are in the presence of a large amount of water vapor, great competition relationship, and the more polar water will replace part of the toluene adsorbed on the molecular sieve. There is no "shoulder peak" in the toluene breakthrough curves of ZSM-5-300 and ZSM-5-360, indicating that the high-silicon ZSM-5 molecular sieve has the adsorption breakthrough curves of benzene and ethyl acetate. As shown in Figures 3 and 4, the saturated adsorption time sequence of five molecular sieves for toluene and ethyl acetate is USY > ZSM-5-300 > HY > ZSM-5-360 > NaY, and NaY has the worst adsorption performance. HY, NaY, and USY have strong hydrophobicity, and water vapor has no effect on the adsorption of toluene. Ethyl acetate also has no obvious "shoulder peak" on the five adsorption materials, which may

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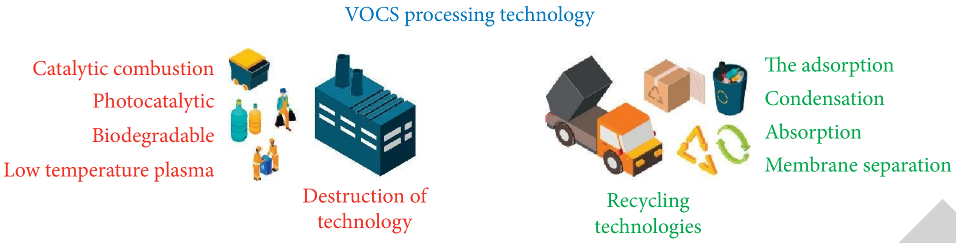

Figure 1: Common VOCs treatment technology.

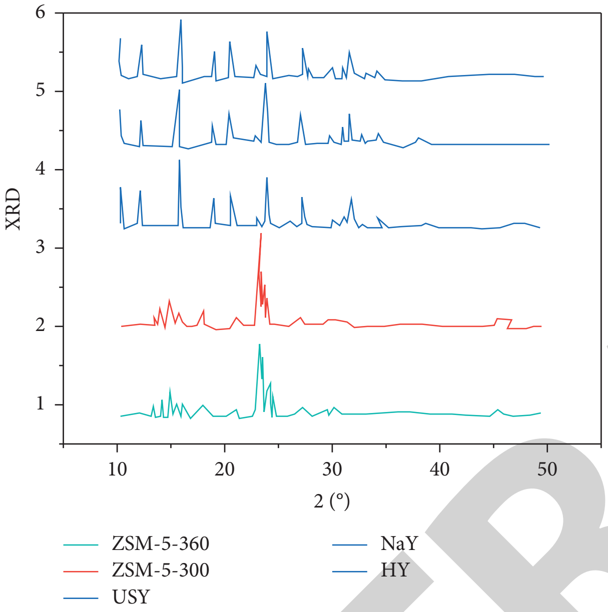

Figure 2: XRD patterns of the mixture of HY, Na Y, USY, ZSM-5-300, and ZSM-5-360 molecular sieves and silica sol.

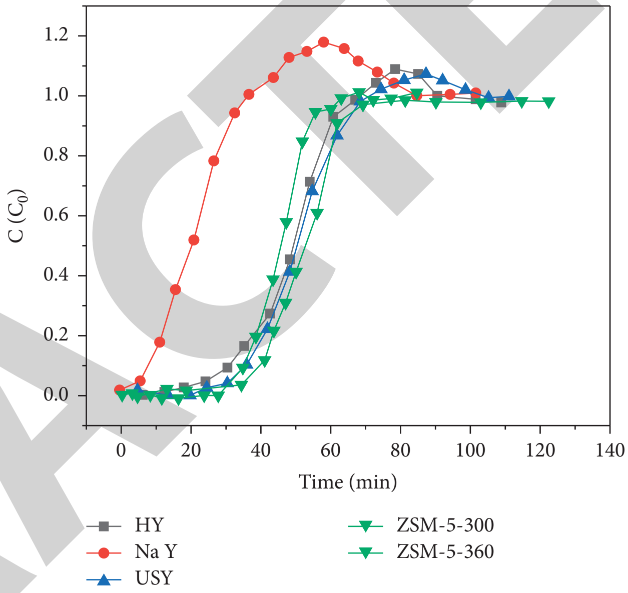

Figure 3: Adsorption breakthrough curve of molecular sieve for toluene. Note: Airspeed 5000 h^{-1}. The adsorption conditions are normal pressure, (35 ± 2) °C, molecular sieve loading 2.0 g, toluene and ethyl acetate concentration 500 mg/m^3, and water vapor RH = 60%.

be due to the similar polarity of ethyl acetate and water, and the substitution effect is not obvious [20].

Table 1 provides the saturated adsorption capacity of 5 kinds of adsorption materials for toluene, ethyl acetate, isopropanol, and acetone [21]. It can be seen that ZSM-5-300 has the largest adsorption capacity for the four adsorbates, which is closer to USY. The adsorption capacity of HY and ZSM-5-300 is in the middle. The saturated adsorption capacity of NaY is the smallest because there is a large competitive adsorption relationship between toluene and water vapor in NaY with a low Si/Al ratio; in the presence of a large amount of water vapor, the adsorption capacity of VOCs decreased significantly compared with the dry gas condition [22].

4.3. Adsorption Performance of Multicomponent VOCs. In the actual industrial VOCs adsorption, the components are often complex, so it is necessary to investigate the adsorption performance of molecular sieves for mixed VOCs [8]. Figure 5 shows the adsorption breakthrough curve of ZSM-5-300 molecular sieve to the mixed components of toluene, ethyl acetate, isopropanol, and acetone. As shown in Figure 5, the adsorption process of the four mixed components is more complicated than that of a single component; the adsorption process of the mixed components is roughly divided into three stages: in the initial stage (before 50 min), the four mixed components are equally adsorbed on the molecular sieve, each occupying the "active site" of the molecular sieve. As the adsorption process continued, the outlet concentrations of acetone, toluene, isopropanol, and ethyl acetate gradually increased, the outlet concentration of ethyl acetate rose relatively slowly, and acetone, toluene, and isopropanol all showed a downward trend after reaching the highest point successively and then gradually tended to balance; there is an obvious "hump" on the breakthrough curve, that is, the outlet concentration of acetone is greater than the inlet concentration within a certain breakthrough time, which is in line with the typical two-component adsorption process curve. In the saturation equilibrium stage, the concentration of the mixed components no longer increases or decreases and tends to be stable [23]. During the adsorption process, the boiling point of acetone is 56°C, which is relatively low, so the adsorption strength of acetone on the surface of molecular sieve is weak

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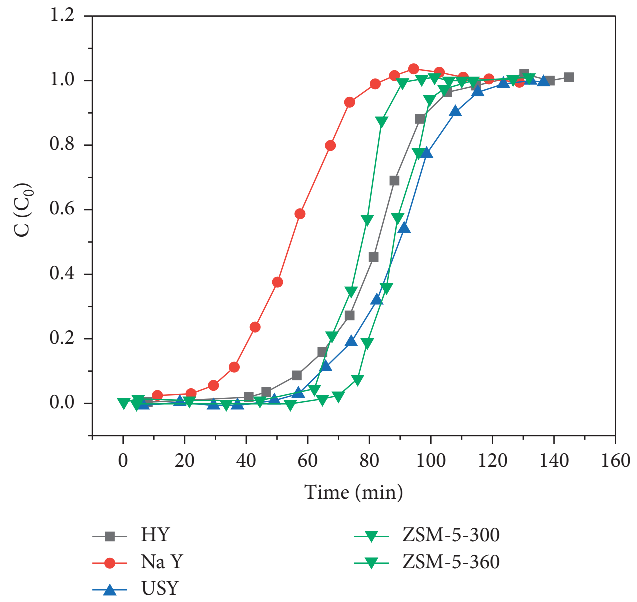

Figure 4: Adsorption breakthrough curve of molecular sieve for ethyl acetate. Note: Airspeed 5000 h^{-1}. The adsorption conditions are normal pressure, (35 ± 2) °C, molecular sieve loading 2.0 g, toluene and ethyl acetate concentration 500 mg/m^3, and water vapor RH=60%.

Table 1: The saturated adsorption capacity of VOCs adsorbed by molecular sieves (mg/g).

Molecular sieve	Toluene	Ethyl acetate	Isopropyl alcohol	Acetone
HY	62.7	108.0	63.4	67.3
NaY	24.0	73.1	37.2	63.4
USY	65.3	117.3	68.6	71.2
ZSM-5-300	68.0	118.7	70.9	72.5
ZSM-5-360	58.6	101.9	59.6	64.7

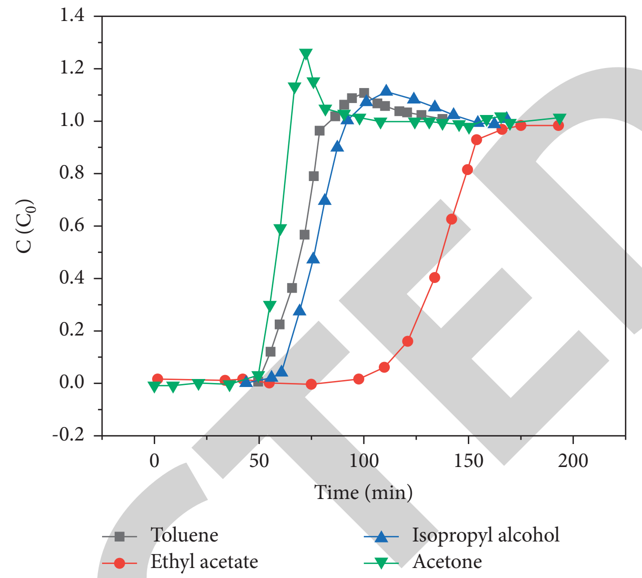

Figure 5: Adsorption breakthrough curve of ZSM-5-300 molecular sieve for mixed components of toluene, ethyl acetate, isopropanol, and acetone. Note: Airspeed 5000 h^{-1}. The adsorption conditions were normal pressure, (35 ± 2) °C, molecular sieve loading 2.0 g, four VOCs concentrations were 125 mg/m^3, and water vapor RH = 60%.

Table 2: Adsorption parameters of mixed VOCs on ZSM-5-300 molecular sieve.

VOCs	Saturated adsorption capacity/(mg·g^{-1})	Breakthrough adsorption/(mg·g^{-1})
Toluene	22.3	17.1
Ethyl acetate	45.0	35.8
Isopropyl alcohol	25.0	19.8
Acetone	19.0	16.5

at a certain temperature, and it is easily replaced by other adsorbates under the drive of airflow; there is also an obvious "hump" on the breakthrough curves of toluene and isopropanol because the polarity of ethyl acetate is stronger than that of toluene and isopropanol; acetone, toluene, and isopropanol adsorbed in the molecular sieve channels are replaced by ethyl acetate, resulting in an obvious "hump." The pore size of ZSM-5-300 is 0.51–0.56 nm, and the molecular diameters of toluene, ethyl acetate, isopropanol, and acetone are 0.60, 0.52, 0.47, and 0.50 nm, respectively, under the combined action of the surface polarity of molecular sieve and the superposition of pore walls, with the completion of the "hump" of acetone, toluene, and isopropanol, the outlet concentration of ethyl acetate slowly increased; finally, adsorption saturation was reached, and it can be seen from the breakthrough curve that there was a competitive adsorption between the four components. Table 2 provides the adsorption parameters of mixed VOCs on ZSM-5-300 molecular sieves [24]. Comparing Table 1 and Table 2, it can be seen that the saturated adsorption amount of mixed VOCs is significantly reduced compared with that of single component, and the decline range is 67.2% of toluene, isopropyl alcohol 64.7%, acetone 73.8%, and ethyl acetate 62.1%, indicating that when the molecular sieve surface area "effective adsorption site" remains unchanged, the adsorption capacity of each adsorbate will be significantly reduced when the molecular sieve adsorbs mixed VOCs.

5. Conclusion

The author studied the adsorption performance of VOCs on the honeycomb ceramics loaded with molecular sieve and screened out the optimal adsorption material, which achieved the expected research purpose; the development of a new type of adsorption material with a good coating effect, excellent adsorption performance, and good regeneration is of great value for industrial application. The results of the study are as follows: the order of saturated adsorption capacity of single-component organics toluene, ethyl acetate, isopropanol, and acetone on the supported molecular sieve honeycomb ceramics is ZSM-5-300 > USY > HY > ZSM-5-360 > NaY, ZSM-5-300 It is a more suitable adsorption material for adsorbing complex

VOCs. Compared with the single-component adsorption of ZSM-5-300 molecular sieve for the adsorption of mixed-component VOCs, the saturated adsorption of mixed VOCs is significantly reduced; this indicated that the adsorption capacity would change significantly when the mixed VOVs were adsorbed, which was attributed to the coadsorption and competitive adsorption between the four VOCs. The research on the adsorption performance of VOCs on the supported molecular sieve honeycomb ceramics can be further considered in the following aspects: the loading, dispersibility, and adsorption strength of ZSM-5-300 molecular sieve on honeycomb ceramics have an important influence on improving its adsorption of VOCs; it is necessary to further study the molecular sieve coating process, such as stirring speed and stirring time, pH, type of binding material, molecular sieve ratio, and other conditions during the experiment, in order to prepare honeycomb ceramic adsorption materials with strong stability and good adsorption effect. Further research was conducted to synthesize porous ZSM-5 molecular sieve adsorption materials by the template method, nanograin method, and postprocessing method or composite ZSM-5 molecular sieve and mesoporous molecular sieve to form mesoporous-microporous composite materials, adsorption and desorption performance of industrial waste gas.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The author declares that there are no conflicts of interest.

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