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Catalytic pyrolysis of plastic vapors by ZSM-5/MCM-41 core-shell catalyst

Aim

Investigate the catalytic pyrolysis using micro-pyrolyzer. Modification of molecular sieve catalysts to prepare a micro-mesoporous composite catalyst, investigating the catalytic mechanism to enhance the selectivity of olefins and monocyclic aromatics, studying the deactivation mechanism to extend catalyst lifetime, and exploring reaction kinetics.

Justification

The extensive use of plastics has led to widespread white pollution. The recycling of plastics is not only benefit for mitigating environmental pollution but also for reducing excessive dependence on resources. Catalytic pyrolysis of plastics, which can convert plastics into lightweight olefins or monocyclic aromatics, is a highly promising technology. Efficient and durable catalysts constitute the core of this process. The Brønsted acid sites (BAS) of zeolite catalysts are considered as active sites for catalytic cracking of large hydrocarbon molecules. The BAS active sites are located on the Si-O-Al bridging structures, where the negative charge of the $[\text{AlO}_4]^-$ tetrahedra is compensated by H^+ to form BAS. For ZSM-5 catalysts, a lower silicon-to-aluminum ratio corresponds to a higher abundance of acidic sites. However, ZSM-5 is a microporous catalyst with pore diameters ranging from 5.2 to 5.9 angstroms. Due to the larger molecular chains of plastic vapors, the acidic sites located within the microporous structure are significantly affected by steric hindrance and diffusion resistance. On the other hand, coking is the primary factor leading to deactivation of zeolite catalysts[1]. Recently, research has indicated that the relationship between BAS surface density and catalyst lifetime exhibits a non-linear correlation [2]. In general, the introduction of mesoporous structures in ZSM-5 catalysts is beneficial for enhancing the catalytic activity and extending the catalyst lifespan.

This study will employ a commercial molecular sieve catalyst, subjected to mesopore expansion through the erosion of silica-alumina framework in a NaOH solution. Simultaneously, assisted by the mesoporous template CTAB, a regular mesoporous structure can be re-crystallized under hydrothermal conditions. The acidity sites of the catalyst will be characterized through NH_3 -TPD, and the specific surface area will be determined using a nitrogen adsorption instrument. The density of acidic sites can be calculated. Catalytic pyrolysis experiments will be conducted on a micro-pyrolyzer coupled with a two-dimensional gas chromatography instrument. The effects of residence time and temperature on product distribution will be studied. Based on the experimental data, the relationship between catalyst structure and pyrolysis performance will be studied to elucidate the catalytic pyrolysis mechanism. A lump model will be employed to investigate the catalytic pyrolysis kinetics and deactivation kinetics, and predictive modeling will be applied to assess catalytic pyrolysis performance.

Program

1. Literature review on hierarchical catalyst for plastic pyrolysis.
2. ZSM-5/MCM-41 preparation and characterization.
3. ZSM-5/MCM-41 pyrolysis performance and the structure-activity relationship.
4. Catalytic pyrolysis kinetics based on lumping method.

References

- [1] D. Trueba, et al., *Chemical Engineering Journal*, 451, (2023) 138709.
- [2] L. Dai, et al., *Applied Catalysis B: Environmental*, 318, (2022) 121835.