Cyclic plasma-catalytic system applied for VOC removal with repetitive deactivation and regeneration of catalysts

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Abstract: This study investigates a cyclic system of catalyst deactivation and regeneration used for toluene removal by plasma catalysis. The system was used with TiO_2 and Pt/γ-Al_2O_3 catalysts and exhibited higher toluene removal efficiency with plasma-regenerated catalysts than with those non-regenerated. The results demonstrate the potential of nonthermal plasma as an effective alternative for VOC removal and catalyst regeneration.

1. Introduction

Volatile organic compounds (VOCs) are significant contributors to the air pollution. One of the methods for their removal is catalysis. However, catalysts often lose their catalytic activity and/or selectivity over time, for example, due to the adsorption of undesired by-products on their surface, which limits their lifespan. This requires regular replacement or regeneration of the deactivated catalysts. Although regeneration (i.e., restoring of catalytic activity/selectivity) is preferred over replacement, conventional (thermal) methods of regeneration are often energy-demanding. Therefore, there is a need to develop alternative regeneration methods. An innovative method for removing VOCs from a gas [1] and regenerating catalysts that have been deactivated by undesired by-products, such as those generated in the VOC removal process, involves the use of nonthermal plasma [2].

2. Methods

This research was focused on testing the cyclic plasma-catalytic system that removes a model VOC pollutant (toluene) from the air using a packed-bed dielectric barrier discharge reactor in the first step. Then, the system was applied for oxygen plasma regeneration of previously deactivated catalyst in the second step. The cycle of toluene removal associated with catalyst deactivation followed by its regeneration was repeated three times using various catalysts (TiO_2, Pt/γ-Al_2O_3). To evaluate regeneration efficiency, three cycles without regeneration were also performed. Toluene removal efficiencies (TREs) of regenerated and non-regenerated catalysts were compared.

Regeneration efficiency was assessed by monitoring gaseous products by FTIR spectroscopy. TGA, SEM, and GC-MS were used for the identification of solid by-products. Additionally, plasma regeneration was compared to other regeneration methods (ozone, thermal and sequential plasma regeneration).

3. Results and Discussion

The Fig. 1 shows the TRE obtained with both plasma-regenerated and non-regenerated Pt/γ-Al_2O_3 catalyst in each of the three cycles. It is evident that the TRE decreases as the number of cycles increases for both catalysts. This reduction is due to gradual deactivation of the catalyst caused by the deposition of solid carbon-containing by-products. The GC-MS analysis revealed the presence of long-chain alkanes and toluene-based aromatic hydrocarbons with various added functional groups. However, plasma regeneration of the catalyst produced gaseous oxidation products (CO_2, CO, HCOOH, H_2O) and partially restored the catalyst's activity, resulting in higher TRE across all cycles compared to the TRE obtained with non-regenerated catalyst.

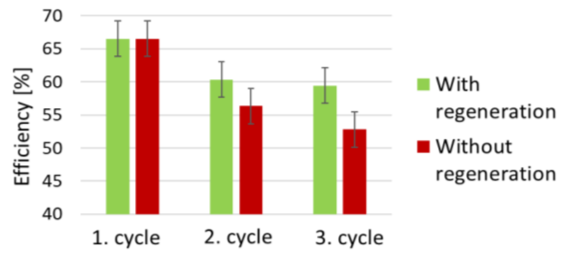

Fig. 1. Toluene removal efficiency obtained with Pt/γ-Al_2O_3 for cycles with/without plasma regeneration.

4. Conclusion

Investigation of cyclic plasma-catalytic system applied for VOC removal demonstrated that plasma-regenerated catalysts maintained higher TRE in each cycle, compared to those non-regenerated. This indicates that plasma regeneration partially restored the catalyst's activity, offering a cost-effective regeneration alternative.

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