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Nonthermal plasma regeneration and repetitive use of deactivated catalysts

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Nonthermal plasma regeneration of deactivated catalysts was investigated at ambient and elevated temperatures and compared with ozone regeneration. A level of catalyst regeneration strictly depended on the catalytic material and gas temperature. Regenerated catalysts were reused in several cycles and the obtained results were compared with those of non-regenerated ones.

1 Introduction

A catalyst deactivation, i.e., the loss of catalytic activity and/or selectivity over time, is generally one of the major and inevitable concerns in conventional as well as plasma catalysis. After a decrease of catalyst activity, the catalyst can be regenerated (restored, reactivated), used for another application, recycled, or discarded [1]. From the economic and environmental point of view, the first option is the most appropriate.

It is well known that nonthermal plasma can create highly reactive environment even at ambient conditions. In this sense, a potential of nonthermal plasma generated by atmospheric pressure dielectric barrier discharge (DBD) for regeneration of deactivated catalysts was investigated.

2 Experiment and analysis

The experimental procedure consisted of two steps. Firstly, plasma catalytic removal of a model volatile organic compound (toluene) or polycyclic aromatic hydrocarbon (naphthalene) was employed using packed-bed DBD reactors in synthetic air as a carrier gas [2, 3]. The reactors had a cylindrical coaxial geometry and were packed with various catalysts (TiO_2, Pt-, Pd-γAl_2O_3, $BaTiO_3$). Decomposition of the model compounds resulted in formation of various gaseous and solid compounds (products) often found as carbon containing deposits on surface of the catalysts. These solid deposits were responsible for deactivation of catalysts and decrease of their activity over time.

Secondly, the deactivated catalysts were regenerated using the packed-bed DBD reactors of the same geometry in oxygen as a carrier gas at ambient (25°C) or elevated (100°C) temperatures for 2 hours. Besides, in order to evaluate a role of ozone in the regeneration process, the catalysts were also regenerated by ozone from an ozonator.

Regeneration led to formation of gaseous oxidation products (CO_2, CO and HCOOH) and temporal evolution of their concentrations was monitored by infrared absorption spectroscopy FTIR. Surface of the catalysts was analysed by scanning electron microscopy SEM and optical microscopy. In addition, the thermogravimetric analysis TGA of the catalysts was also performed.

3 Results

The results showed that the concentrations of the oxidation products rapidly increased, reached maxima and then gradually decreased with time. Moreover, efficiency of the catalyst regeneration depends predominantly on the used catalytic material, i.e., its dielectric constant. It determines not only an interaction between catalyst and plasma, but also an exhibiting discharge mode (surface or localised filamentary mode). Surface analysis of the catalysts proved oxidation and partial removal of solid carbon deposits. Efficiency of catalyst regeneration was higher with nonthermal plasma compared to ozone regeneration alone and depended on gas temperature.

Finally, some of the regenerated catalysts were reused and tested for the removal of toluene by plasma catalysis in three repetitive cycles. The results showed that regenerated catalysts exhibited higher efficiency of toluene removal than non-regenerated ones. This effect was the most evident for Pt/γAl_2O_3, and then for TiO_2.

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