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Effect of pellet catalyst properties on gas cleaning process

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The work deals with gas cleaning process, i.e. removal of model hydrocarbons (toluene and naphthalene) by a dielectric barrier discharge (DBD) reactor filled with various pellet catalysts [1,3]. We studied the effect of pellet material properties on removal efficiency. The plasma reactors were powered by AC high voltage and operated at various specific input energies (SIE) and at the constant gas flow rate (0.5 L/min). After the gas cleaning process, selected pellet catalysts were regenerated by various methods to restore their activity and subsequently re-used again for gas cleaning [3]. Gaseous and solid carbon-containing products (coke deposits) of toluene and naphthalene decomposition were analyzed by means of FTIR, TGA, GC-MS, SEM, EDX analytical methods.

In case of napthalene, the effect of the pellet materials of various catalytic activity (glass, Al2O3, Pt/Al2O3, TiO2, BaTiO3, ZrO2), shape and size (spherical, cylindrical, 3-8 mm), dielectric constant (5-4000) and specific surface area (35-250 m2/g) in various carrier gases (ambient air, oxygen or nitrogen) on removal efficiency and CO2 selectivity were investigated. The experiments were carried out at relatively low temperature (<150°C) and with relatively high initial concentration of naphthalene (5,000 ppm). In case of toluene, its removal efficiency in air, formation of solid products (coke deposits), their accumulation on pellet catalysts followed by their gradual deactivation was investigated. The deactivated pellet catalysts were then regenerated either by plasma, ozonation or heat. The experiments were carried out with four different pellet materials (Al2O3, Pt/Al2O3, Pd/Al2O3, TiO2) at room temperature and with the initial concentration of toluene (2,770 ppm).

The results showed that naphthalene removal efficiency (NRE) increased indeed with the increase of the amplitude and frequency of the applied voltage. The carrier gas had significant effect on the NRE, as different reactive species (N2(A), N, O, O3, OH) and products were formed. In nitrogen, the NRE was the poorest (<25%) with mono-/hetero- N-containing aromatic compounds being main products. In ambient air, the NRE was significantly higher (up to 88%) and accompanied with the formation of CO/CO2 and O-containing hydrocarbons or hydrocarbons with fewer carbon atoms. In oxygen, complete naphthalene removal was achieved. With respect to the pellet materials, the highest NRE was obtained with TiO2 catalyst (88%, 320 J/L), and slightly lower with Pt/Al2O3 (78%), but much higher CO2 selectivity suggesting its best oxidation abilities compared to other materials. The NRE for other materials followed a sequence: ZrO2 (72%)> Al2O3 (66%)> glass (64%)> BaTiO3 (51%). The NRE increased with increasing material surface area, while shape of material was found to be more sensitive and important: higher NRE was found for cylindrical and smaller pellets. Surface analysis of materials revealed differences in distribution of solid carbon products on the surface of various materials, with spherical materials covered almost uniformly, while cylindrical in characteristic circular patterns probably as a result of different discharge modes depending on the used material. Besides the main gaseous products (CO2, CO, H2O and HCOOH), several other complex gaseous and solid by-products were identified (phthalic anhydride, maleic anhydride, benzoquinone naphthoquinone, etc). Toluene removal by plasma combined with pellet catalyst as well as subsequent pellet catalyst regeneration were tested for various materials. Removal efficiency, selectivity and carbon balance of the process were evaluated. The RE of toluene followed a sequence: Pd/Al2O3 (76%, 760 J/L)> Pt/Al2O3 (74%)> Al2O3 (65%)> TiO2 (56%). These results were affected not only by catalytic properties of materials, but also dielectric constant and specific surface area (Al2O3). Pd/Al2O3 exhibited the highest toluene RE and the lowest energy demand [kWh/kg]. All catalysts showed the highest selectivity for CO2 (13–29%) and smaller for CO, HCOOH. For Al2O3 production of other than above species was the highest, resulting in the worst carbon balance.

The used pellets materials were regenerated for 2 hours by three different methods - by DBD plasma, O3 (8,000 ppm), and (at 100°C). The efficiency of regeneration was evaluated based on the production of gaseous products (CO2, CO and HCOOH) in time. Plasma regeneration was the best although still only partial and spatially non-uniform. Regenerated catalysts were re-used for toluene removal and the results showed that regenerated catalysts exhibit higher toluene RE after each cycle than those non-regenerated.

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References

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