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Effect of Time on Stream on the Performances of Plasma Assisted Fe-Doped Cryptomelanes in Trichloroethylene (TCE) Oxidation

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Abstract : Environmental issues, especially air pollution, have become a huge concern of environmental legislation as a consequence of growing awareness in our global world. In this regard, control of volatile organic compounds (VOCs) emission has become an important issue due to their potential toxicity, carcinogenicity, and mutagenicity. The research of innovative technologies for VOC abatement is stimulated to accommodate the new stringent standards in terms of VOC emission. One emerging strategy is the coupling of 2 existing complementary technologies, namely here non-thermal plasma (NTP) and heterogeneous catalysis, to get a more efficient process for VOC removal in air. The objective of this current work is to investigate the abatement of trichloroethylene (TCE-highly toxic chlorinated VOC) from moist air (RH=15%) as a function of time by combined use of multi-pin-to-plate negative DC corona/glow discharge with Fe-doped cryptomelanes catalyst downstream i.e. post plasma-catalysis (PPC) process. For catalyst alone case, experiments reveal that, initially, Fe doped cryptomelane (regardless the mode of Fe incorporation by co-precipitation (Fe-K-OMS-2)/ impregnation (Fe/K-OMS-2)) exhibits excellent activity to decompose TCE compared to cryptomelane (K-OMS-2) itself. A maximum obtained value of TCE abatement after 6 min is as follows: Fe-KOMS-2 (73.3%) > Fe/KOMS-2 (48.5) > KOMS-2 (22.6%). However, with prolonged operation time, whatever the catalyst under concern, the abatement of TCE decreases. After 111 min time of exposure, the catalysts can be ranked as follows: Fe/KOMS-2 (11%) < K-OMS-2 (12.3%) < Fe-KOMS-2 (14.5%). Clearly, this phenomenon indicates catalyst deactivation either by chlorination or by blocking the active sites. Remarkably, in PPC configuration (energy density = 60 J/L, catalyst temperature = 150°C), experiments reveal an enhanced performance towards TCE removal regardless the type of catalyst. After 6 min time on stream, the TCE removal efficiency amount as follows: K-OMS-2 (60%) < Fe/K-OMS-2 (79%) < Fe-K-OMS-2 (99.3%). The enhanced performances over Fe-K-OMS-2 catalyst are attributed to its high surface oxygen mobility and structural defects leading to high O_3 decomposition efficiency to give active species able to oxidize the plasma processed hazardous\by-products and the possibly remaining VOC into CO_2. Moreover, both undoped and doped catalysts remain strongly capable to abate TCE with time on stream. The TCE removal efficiencies of the PPC processes with Fe/KOMS-2 and KOMS-2 catalysts are not affected by time on stream indicating an excellent catalyst stability. When using the Fe-K-OMS-2 as catalyst, TCE abatement slightly reduces with time on stream. However, it is noteworthy to stress that still a constant abatement of 83% is observed during at least 30 minutes. These results prove that the combination of NTP with catalysts not only increases the catalytic activity but also allows to avoid, to some extent, the poisoning of catalytic sites resulting in an enhanced catalyst stability. In order to better understand the different surface processes occurring in the course of the total TCE oxidation in PPC experiments, a detailed X-ray Photoelectron Spectroscopy (XPS) and Time of Flight-Secondary Ion Mass Spectrometry (ToF-SIMS) study on the fresh and used catalysts is in progress.

Keywords : Fe doped cryptomelane, non-thermal plasma, plasma-catalysis, stability, trichloroethylene
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