Enhancement of catalytic activity and stability during PPC for total oxidation of TCE in humid air over Fe-doped cryptomelane.

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Cryptomelane catalyst K-OMS-2 of ideal formula $K(Mn_7^{IV}Mn^{III})O_{16}$ is modified with Fe doping and has been previously used as catalyst in post-plasma catalysis (PPC) in the course of total trichloroethylene (TCE) oxidation in moist air (RH=15%). It was shown that adding iron to cryptomelane allows a better functioning of catalyst, consequently, combining with plasma significantly enhanced the catalytic performances (at 150°C). The issue we want to address herein is the effect of time on stream on the performance of Fe-doped cryptomelane in plasma assisted TCE oxidation. It is found that, next to enhanced catalyst activity, non-thermal plasma (NTP) remarkably improves the stability of the catalyst. A combined 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.

1. Introduction

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 NTP and heterogeneous catalysis, to get a more efficient process for VOC removal in air.

In this study it is shown that Fe-doped (by co-precipitation: Fe-KOMS-2 and by a successive Fe(OH)x (x = 2; 3) precipitation: Fe/KOMS-2) cryptomelanes with the assistance of NTP are efficient candidates for the abatement of TCE (highly toxic chlorinated VOC) in terms of activity, selectivity and stability.

2. Results and Discussion

Our results shows that, initially Fe doped cryptomelane (solid red and green) (regardless the mode of Fe incorporation) exhibits excellent activity to decompose TCE compared to cryptomelane itself (fig. 1). A maximum obtained value of TCE abatement after 6 min is as follows: Fe-KOMS-2 (75.5%) > Fe/KOMS-2 (48.5) > KOMS-2 (22.6%). However, with prolonged operation time, the abatement of TCE decreases. Clearly, this phenomenon indicates catalyst deactivation either by chlorination or by blocking the active sites. Nonetheless, both undoped and doped catalysts (dashed lines) used in a PPC process remain strongly capable to abate TCE. 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 by time on stream, however, it is noteworthy to stress that still a constant abatement of 83% is observed during at least 30 minutes.

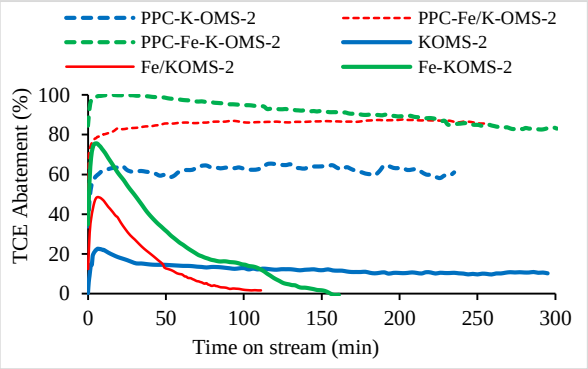

Figure 1: The effect of time on stream on the performance of the catalyst alone (150°C) and PPC (150°C and energy density 60 J/L) towards TCE removal for 3 catalysts.

3. Conclusion

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 XPS and ToF-SIMS study on the fresh and used catalysts is in progress.