

## DEGRADATION OF CHLORINE CONTAINING ORGANIC POLLUTANTS USING CATALYTIC PEROXONE PROCESS

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### 1. INTRODUCTION

Halogenated - and more specifically chlorinated - volatile organic compounds (CVOCs) are widely used for various purposes commercially; however, there exists a well-known demand to decompose the waste of these chemicals to protect the environment. As dry cleaning and electronic industry solvents, trichloro- (TRCE) and tetrachloroethene (TCE), as well as dichloroethane (DCE) (starting material for PVC production) are those organochlorinated compounds that are released into the atmosphere in the largest quantities. The diminution of the X-VOC emission is of high importance and the advanced oxidation (AO) processes are promising method for elimination of these compounds [1]. The „peroxone” process is an effective AO method applying ozone and hydrogen peroxide for degradation of organochlorine water or air pollutants [2]. Our starting point is to enhance the efficiency of the homogeneous peroxone process using modified mesoporous materials as catalyst.

We report here our investigations concerning the application of ozone + H<sub>2</sub>O<sub>2</sub> in water for oxidation of airborne DCE, TRCE and TCE in the presence of transition metal (Ti or Fe) containing MCM41 or SBA15 type mesoporous catalysts.

### 2. EXPERIMENTAL

Four catalyst samples (Ti-MCM41, Fe-MCM41, Ti-SBA15 and Fe-SBA15) were synthesized in our laboratory followed by the suggestion of the literature [3,4]. The structure of the catalysts was studied by X-ray diffraction, IR spectroscopy and transmission electron microscopy techniques; their thermal stability was followed by thermal analytical method. The specific surface area and pore size distribution of samples were determined from the data of nitrogen adsorption/desorption isotherms.

For determination of optimal parameters of the reaction system containing three phases (gaseous with the VOC, water + H<sub>2</sub>O<sub>2</sub> and the

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solid catalyst) more experimental arrangements were tested, and finally a recirculating bubble column system was chosen as the most efficient solution. The reactions were followed by gaschromatographic analysis (organic components) UV-spectroscopy (ozone) and iodometric method ( $\text{H}_2\text{O}_2$ ).

### 3. RESULTS AND DISCUSSIONS

The characteristics of catalyst samples have corresponded to the data of literature, the synthesized materials have had mesoporous structure and they have contained transition metals in the planned quantity (1% Ti or Fe). A run for testing the peroxone method for the oxidation of DCE feeding by oxygen stream into a buffer solution with pH 6.2 can be seen in Fig 1.

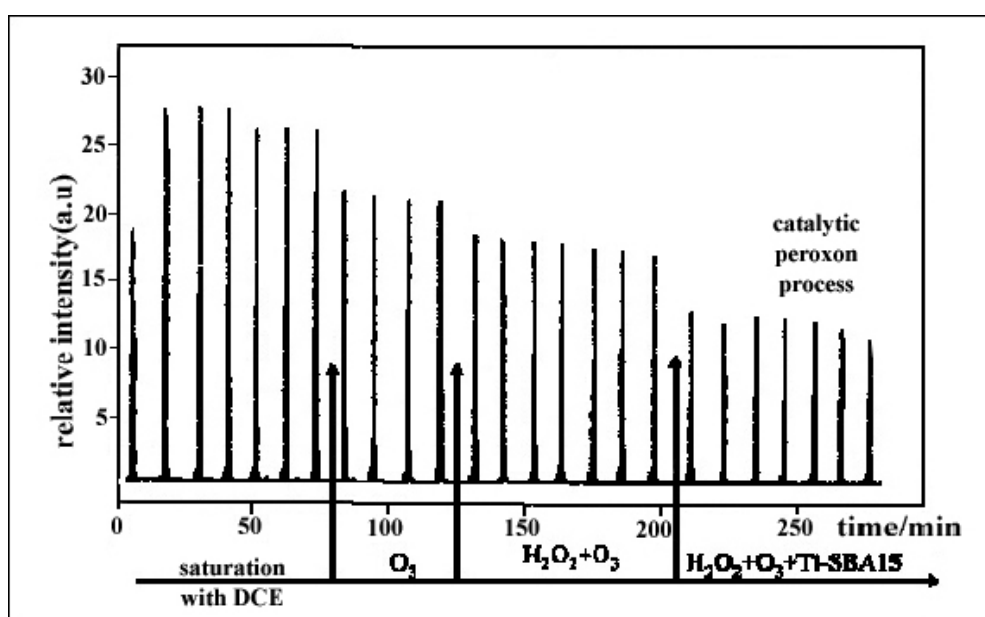


Fig. 1: Reaction of 1,2-dichloroethane in the presence of ozone,  $\text{H}_2\text{O}_2$  and Ti-SBA15 catalyst

The conversion of DCE reached about 22% even in the presence of ozone formed in the oxygen stream, and a further increase (~ 30%) could be observed at the addition of hydrogen peroxide. In the presence of Ti-SBA15 catalyst the level of the conversion heightened for 51-55%, which unequivocally proves the catalytic effect of the material used.

Similar results have been observed using the other catalysts, generally an increase of 15-25% has detected in comparison to the homogeneous peroxon process. The data are summarized on Figures 2 and 3, and the highest conversion can be established for Ti-MCM41 both in the presence of oxygen or air as carrier gas.

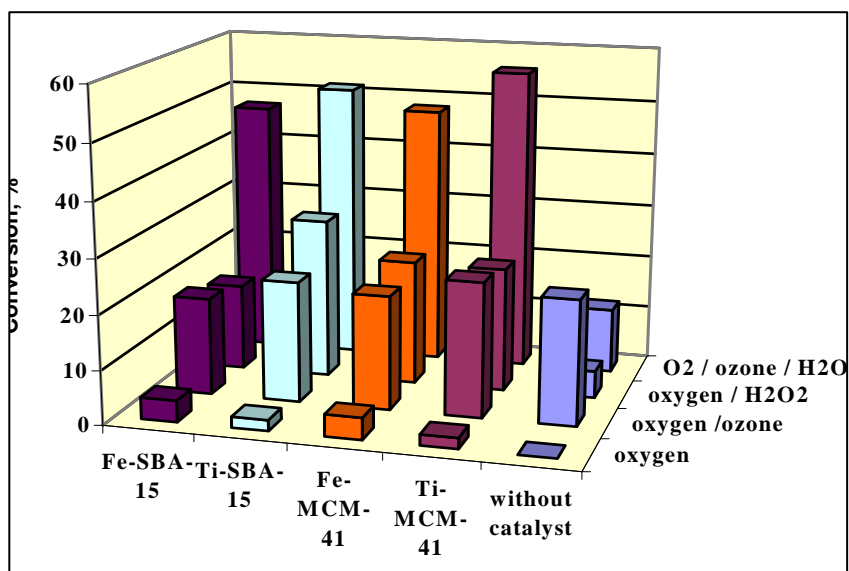


Fig. 2: Oxidation of 1,2-dichloroethane by peroxon process at 25 °C, in the presence of oxygen as gas phase; pH: 6.2 – 6.8

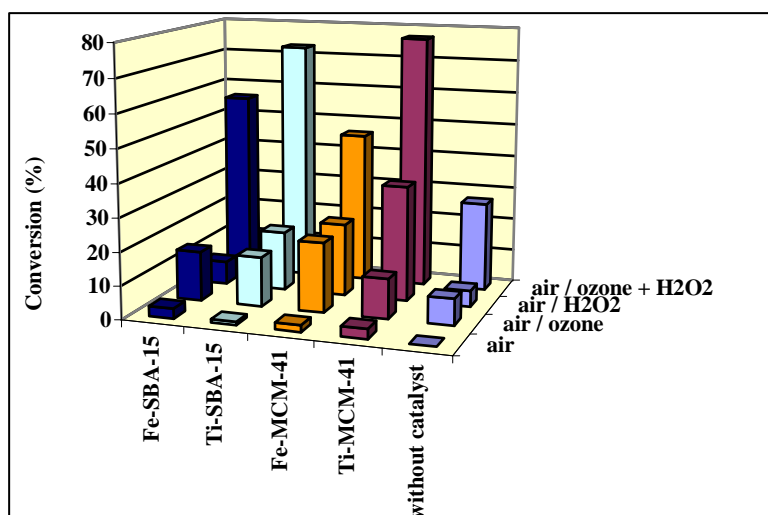


Fig. 3: Oxidation of 1,2-dichloroethane by peroxon process at 25 °C, in the presence of air as gas phase; pH: 6.2 – 6.8

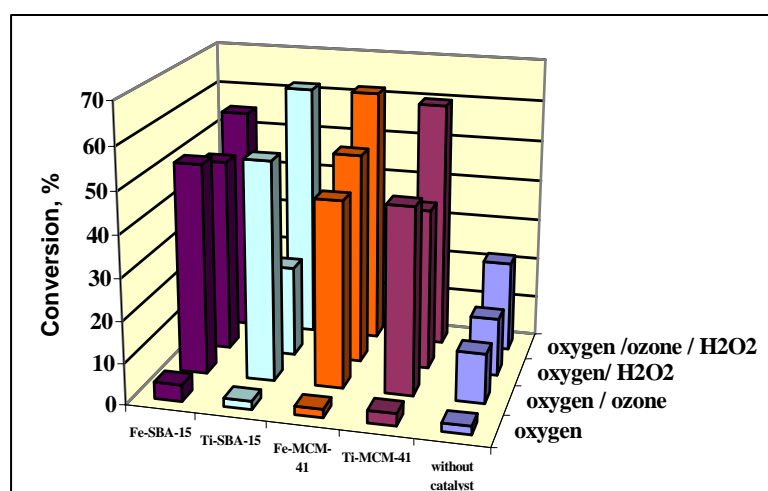


Fig. 4: Oxidation of trichloroethene by peroxon process at 25 °C, in the presence of oxygen as gas phase; pH: 6.2 – 6.8

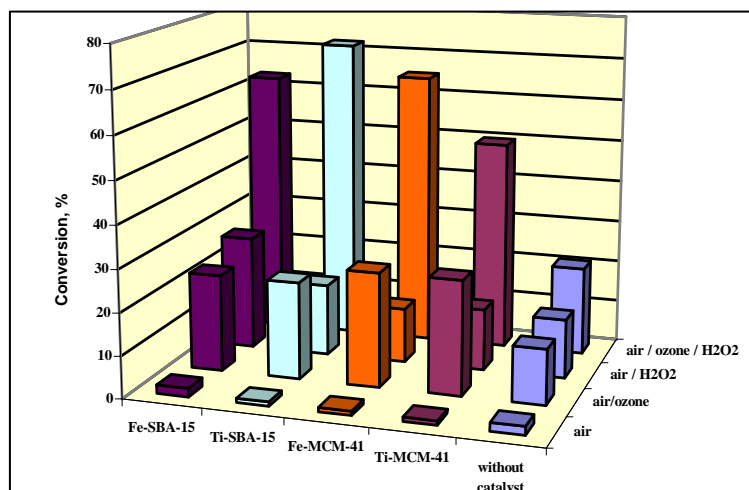


Fig. 5: Oxidation of trichloroethene by peroxon process at 25 °C, in the presence of air as gas phase; pH: 6.2 – 6.8

On Fig. 4 and 5 the data obtained in the reaction of trichloroethene can be seen. The conversion of TRCE in the presence of oxygen is higher in comparison to the presence of air, and the catalytic effects are well established.

#### 4. CONCLUSIONS

There are increasing demands (society) for developing of total and environmental benign degradation of X-VOC.

The peroxone process - application of  $O_3$  and  $H_2O_2$  - is an effective method, however, its efficiency could be increased by the addition of proper heterogeneous catalyst.

In the presence of modified (Ti or Fe) mesoporous silicates (MCM-41 and SBA15) the conversion of DCE and TRCE could be increased, the effect of Ti is higher.

The reaction system is too complicated, the details, namely description of the transport processes and assignation of the mechanism of surface radical reactions involved ozone and hydrogen peroxide is in progress.

#### 5. REFERENCES

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