

Nanocarbon materials grown on carbon felts as catalysts for the oxidation of organic compounds

João Restivo¹, José J.M. Órfão¹, M. Fernando R. Pereira¹, Estelle Vanhaecke^{2*}, Magnus Rønning², De Chen²,

¹Laboratório de Catálise e Materiais (LCM), Laboratório Associado LSRE/LCM, Departamento de Engenharia Química, Faculdade de Engenharia, Universidade do Porto, Portugal

²Chemical Engineering Department, NTNU, Sem Sælands vei 4, 7193 Trondheim, Norway
estelle.vanhaecke@chemeng.ntnu.no

Introduction

Catalytic ozonation is an innovative technology for the elimination of organic pollutants in water and wastewater. Different supported and unsupported catalysts have been tested in the ozonation of several organic compounds. Among the studied materials, activated carbon has been found to be a promising alternative for this reaction [1]. Carbon nanofibers are potential catalysts for ozonation due to the extended mesoporosity, which may have a very positive impact by decreasing the mass transfer resistance usually present in the microporous activated carbon.

Experimental

The graphite felt (CF) pieces are cylindrical, 5 mm thick and 25 mm of diameter. The catalyst precursor is a nickel solution corresponding to 2% loading of nickel. Impregnation by dip coating is done on all pieces until incipient wetness. The samples are then dried overnight at room temperature. First, the samples are reduced for 1 hr at 650°C in H₂/Ar and then C₂H₆ is introduced instead of Ar for 16 hrs. During the cooling down process, the reactor is flushed with Ar. The as synthesized samples are used directly (ACF/CF) or after an oxidation treatment with HNO₃ (oxidized ACF/CF) in ozonation tests.

Ozonation experiments were conducted on a reactor specially designed for the use of structured catalysts. This reactor consists of two columns which allow the circulation of the liquid phase between both. A larger column is used for contact with the gas phase (50 g O₃/m³) and a smaller column is used for contact with the catalyst. Oxalic acid at 1mM was used as a model compound due to its refractory nature to direct ozonation and its presence among the final products of the oxidation of several organic pollutants. Liquid flow between the two columns was kept at 100 ml/min and gas flow was kept at 100 cm³/min.

Results/Discussion

The results obtained in the previous reactor show that the adsorption and the ozonation by itself were not effective to remove the oxalic acid. The best performance was observed for the untreated sample. The presence of oxygen-containing surface groups (present in high amount

on the sample oxidized ACF/CF) has a detrimental effect. Therefore, the basic surface groups (mainly the electron-rich oxygen-free Lewis basic sites on the carbon basal planes) are the active sites for this reaction.

But since the catalysts had performed particularly well when they were cut to smaller pieces and tested in a conventional semi-batch tank reactor, the question whether the liquid phase had actually come into contact with the catalyst or if a bypass of the liquid through the outer side of the catalyst existed was raised. Further experiments were made with the catalyst placed at the bottom of the smaller column, thus forcing the liquid to go through the inside of the carbon felt. This placing is pictured in Fig. 1.

As suspected and shown in fig.1, the performance of the catalyst was much improved by this new placing. *Top* and *Bottom* refer to whether the catalyst was placed at the top or at the bottom of the column in similar fashion as to what is seen in Fig 3.

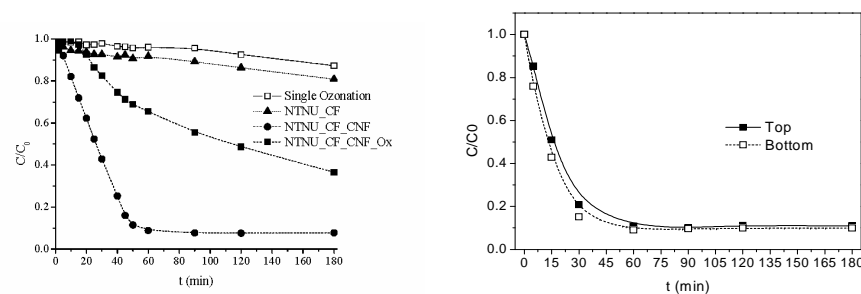


Figure 1. Dimensionless removal of oxalic acid in catalytic ozonation experiments using the powder catalysts (left) and the supported nanocarbon materials by forcing the liquid phase through the carbon felt catalysts placed either at the bottom or at the top of the column.

This new results agree with the results obtained in the semi-batch reactor previously mentioned as it is shown on fig 1. The carbon felts tested have shown very promising results for the catalytic ozonation of organic compounds.

References.

1. P.C.C. Faria, J.J.M. Órfão, M.F.R. Pereira, *Appl. Catal. B-Environ.* 79 (2008) 237.

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