PREPARATION OF CuO, Co$_3$O$_4$, AND Cr$_2$O$_3$ CATALYSTS SUPPORTED BY 13X ZEOLITE

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Supported catalysts utilized in oxidative abatement of volatile organic compounds (VOCs) are usually prepared by wet impregnation method, which is both cost-effective and eco-friendly [1]. Active sites are formed through the process of adsorption of thermally destructible salts on the selected support and their subsequent calcination. 13X zeolite is very durable material for catalyst preparation, because of its thermal stability, good mechanical strength and micro pore structure, which defines its adsorptive and chemical properties [2]. The best catalytic activity is achieved when the content of active component reaches 10 % so in order to optimize composition of catalysts it is necessary to evaluate adsorptive nature of support, characterize active component and evaluate its distribution after calcination.

Adsorptive properties of 13X support were determined in the series of solutions containing Cu, Co and Cr nitrates of various concentrations. Aforementioned elements and their mixtures have very wide application as active components in the preparation of catalysts. XRD analysis of dried and calcined samples showed that CuO, Co$_3$O$_4$ and Cr$_2$O$_3$ were formed as active components on the surface of zeolite. AAS analysis of solutions and calcined samples showed, that adsorptive capacity of 13X zeolite is in the range of 85.21 – 88.35 mg/g for all elements. This leads to the content of the oxides being in the range of 9.96 – 11.18 %. Adsorption isotherms were compiled and several models were used to describe peculiarities of the process – Langmuir, Freundlich, Dubinin-Radushkevich, Temkin and Elovich. All models are derived based on various premises, so each fit of the model by linear regression analysis can confirm or disprove those assumptions. Adsorption isotherms of Cu and Cr were best described by Langmuir model, which indicated that monolayer of adsorbates were formed. This was confirmed by optical microscopy, because active components were found only in the outer, up to 0.4 mm, layer of support. Egg shell type distribution of active components resulted in decrease of mechanical strength as well – up to 15 N/m^2 when using higher concentration impregnation solutions. Also high concentration of CuO in the outer layer of catalyst resulted in some pores being blocked together with NO$_x$ gases failing to escape and part of nitrates remaining undecomposed. In order to maximize active surface area of catalyst and optimize distribution of the oxides, adsorption of Cu and Cr should be performed in several shorter cycles with calcination steps in between. Co adsorption was less affected by diffusion limitations and with increasing concentration of the solution it evenly covered all surface of zeolite. Adsorption modeling resulted in Freundlich isotherm being the best fit for Co, which indicates the formation of several layers of adsorbate and adsorption proceeding in several steps. Cross section of catalyst granule was evenly covered with no visible imperfections. Therefore, preparation of Co$_3$O$_4$ catalyst on the 13X support can be performed in single adsorption-calcination cycle.

References
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2. A. Abdelrasoul, H. Zhang. C. Cheng, H. Doan. Micropor. Mesopor. Mat. **242** (2017) 294.