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ACCEPTED MANUSCRIPT

Cobalt – carbon nanocomposite catalysts of gas-phase hydrodechlorination of chlorobenzene

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The catalytic efficiency of three different cobalt-carbon composites has been compared in the gas-phase hydrodechlorination (HDC) of chlorobenzene (CB) in a flow type fixed bed reactor. The Co@C composite was synthesized by evaporation of overheated liquid drop of Co in the flow of an argon-butene mixture; the Co/C composite - by pyrolysis of sawdust impregnated with $Co(NO_3)_2$ water solution, and the Co/CNT composite – by the impregnation of carbon nanotubes with Co(NO₃)₂ water solution. Different oxidation states of cobalt were observed in the studied composites according to XPS, TPR, and in situ vibrating sample magnetometry results: predominantly Co^0 in Co@C, CoO in Co/C, and Co_3O_4 in Co/CNT. TPR showed the possibility of the reduction of Co_3O_4 to CoO and even to Co^0 under conditions of HDC and established the temperatures of these transitions. TEM revealed that Co⁰ nanoparticles in the Co@C composite are encapsulated by the thin carbon shell, CoO nanoparticles in Co/C are immersed in the carbon matrix, and Co₃O₄ nanoparticles in Co/CNT are located both on the surface and inside the channels of CNTs. All the composites demonstrated activity in HDC of CB. The efficiency of not only Co⁰ but CoO in the CB HDC was shown. The CB conversion at 150-250 °C for Co/C and Co/CNT composites, containing predominantly cobalt oxides, was higher than that for Co@C one, containing Co⁰ nanoparticles coated with the thin carbon shell. The Co/C composite with the lowest Co content (1.3 wt. %) was more effective in HDC than Co/CNT (14.6 wt. % of Co) and Co@C (79.1 wt. % of Co) ones. High CB conversion for the Co@C composite at 350 °C resulted from the catalytic action of the graphene shell of Co particles, activated by subsurface Co^0 .

Keywords: cobalt, carbon, nanocomposite, hydrodechlorination, chlorobenzene

1. Introduction

Organochlorine compounds, because of their negative ecological and biological effects, are considered as a class of the most dangerous organic pollutants. Catalytic reduction by hydrogen, or hydrodechlorination (HDC), is a promising and effective method for utilization of Download English Version:

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