Size effect of carbon nanotubes supported iron nanoparticles on their catalytic properties in hydrogenation processes

I. B. Bychko,¹, Ye. Yu. Kalishyn¹, P. E. Strizhak¹

¹ L.V. Pisarzhevsky Institute of Physical Chemistry, Prospect Nauki 31, Kyiv-03028, Ukraine E-mail: <u>igorbychko@ukr.net</u>

Catalytic activity of carbon nanotubes (CNTs) and Fe nanoparticles supported on CNT have been studied in hydrogenation of crotonealdehyde and ethylene. Catalysts have been prepared by deposition colloidal solution of Fe_xO_y nanoparticles dispersed in hexane on CNTs and dried in air. Preparation colloidal solution in hexane of Fe_xO_y nanoparticles was performed by a modification of procedure described earlier which is based on iron (II) oleate thermal decomposition [1]. CNTs were synthesized by the catalytic decomposition of ethylene according to procedure described elsewhere [2].

The average diameter of prepared iron nanoparticles was 4.3-8.0 nm with standard deviation σ =0,6-0,8 nm. TEM, electron diffraction and Mössbauer spectroscopy data allows one to conclude that the initially formed iron nanoparticles are partially oxidized due to contact with atmospheric oxygen and water.

Iron content in catalyst was determined by oxygen titration method. The size of supported nanoparticles is the same as in colloid solution and, there are no changes of nanoparticles morphology or chemical composition during their deposition. It was shown, that Fe_xO_y/CNT fully reduces to Fe/CNT at 350 °C without nanoparticles agglomeration.

It was found, that pure CNT is active in hydrogenation of crotonealdehyde and ethylene. Measured activity of CNT is much lower, than Fe/CNT. Products of hydrogenation of crotonaldehyde over CNT and Fe/CNT are croton alcohol, butanal and butanol. For CNT, selectivity for the hydrogenation of the carbonyl group is much higher than for C=C bond, while C=C bond is better hydrogenates on Fe/CNT. It was found, that selectivity depends on the size of the iron nanoparticles, the highest selectivity to croton alcohol and butanol, shows 6.8 nm nanoparticles.

Hydrogenation of ethylene on CNT and Fe/CNT shows difference on catalytic properties of CNT and iron nanoparticles. So, only ethane is formed on the CNT, while on Fe/CNT, above 200 $^{\circ}$ C, is also formed methane. Also, it was shown, that reducing the size of the iron nanoparticles leads to a decrease in their catalytic activity in ethylene hydrogenation.

1. Don Lee DL, Kim YH, Zhang X-L, Kang YS Curr Appl Phys 6,786

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2. A.I.Tripol'skii, N.V. Lemesh, V.A. Khavrus' and P.E. *Theor Exp Chem* 4,240 (2008)