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LETTERS TO THE EDITOR

Catalytic Activity in Hydrogenation Processes of Catalysts Containing Nanodimensional Palladium on Carbon Support. Concentration Effect of Palladium and Precursor

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We showed earlier that catalysts obtained by depositing nanodimensional palladium on various nanocarbon materials: fullerenes, nanotubes, fullerene soot, and also activated coconut carbon [1, 2] possessed substantially higher catalytic activity in alkene hydrogenation in comparison with industrial catalysts, where palladium was deposited on activated carbon. At the same time, we found that the nature of nanocarbon material has no essential effect upon catalytic activity. The obtained catalysts posses a pronounced affinity towards hydrogen absorption. The dependence of the absorbed hydrogen amount on the palladium content in the catalyst is of extremal character. The absorption maximum is achieved at comparatively low palladium content in the catalyst (0.1 wt %) and equals to 30–35 mol of H_2 per 1 mol of Pd.

This work was devoted to studying influence of various concentrations of precursor $[Pd(H_2O_4)](ClO_4)_2$ (here and further is designated as Pd^{+2}_{aq}) in the initial solu-tion and palladium nanoclusters formed as a result of its reduction in the synthesized catalyst upon its specific catalytic activity in hydrogenation processes. In our case the number of H_2 mole reacted with substrate per 1 mol of Pd per second was measured.

The influence of this factor was studied by the example of liquid-phase hydrogenation of vegetable oils with hydrogen in the presence of the examined palladium catalyst. It was shown [1, 2] that these catalysts are competitive by their catalytic activity with those obtained on the base of nickel and used presently in fat-and-oil industry to produce hydrogenated fats.

Results obtained are presented in the table. By these data, the specific activity essentially increased as the concentration of Pd^{+2}_{aq} and palladium content in catalyst decreased. Catalysts have the heightened specific

Influence of Pd^{+2}_{aq} concentration and palladium content on the carrier upon specific catalytic activity

Pd content on the carrier, wt%	Precursor concentration, mol $l^{-1}\times10^5$							
	0.94	1.87	2.83	3.74	4.67	9.30	18.7	46.7
	Specific catalytic activity, H_2 mol/1 mol Pd per s							
0.1	13.10	15.8	16.1	17.3	14.3	13.0	5.0	3.0
0.2	13.60	15.7	14.2	15.0	15.2	14.0	12.0	4.2
0.3	14.00	13.0	13.0	12.8	15.0	14.4	12.1	5.5
0.4	13.10	8.5	8.80	11.3	11.0	12.0	11.8	6.2
0.5	10.00	5.1	5.20	9.5	9.5	8.8	8.0	6.2
1.0	4.00	3	3.00	2.35	6.7	6.0	4.5	4.2
2.0	2.50	1	1.10	0.94	3.3	2.5	3.1	1.8
5.0	1.00	0.85	1.00	0.9	–	–	–	1.0

activity, when precursor concentration in the initial solution is within $(1–5)\times10^{-5}$ mol l^{-1} and palladium concentration on the carrier is 0.1–0.5 wt %. Evidently this change in the catalytic activity corresponds to the size of palladium nanoclusters optimal for hydrogenation processes. Note that this dependence is parallel to the hydrogen absorption dependence on the palladium concentration in the obtained catalysts.

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The results obtained were protected by a patent of Russian Federation [3]. Procedure for catalyst preparation and the conditions of experiments were given in the patent.

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