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ABSTRACTS

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- Physical methods, including in situ and operando techniques, in catalysis
- Kinetics and mechanisms of catalyzed processes
- Advanced catalyst systems addressing current challenges: energy, materials, sustainability

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The Effect of Pd/Al₂O₃ Modification with Si,W-Heteropolyacid on the Mechanism of 1,3,5-Trichlorobenzene Multi-Phase Hydrodechlorination

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Hydrodechlorination (HDC) provides the convenient way to transform chlorinated organics which are well-known ecotoxicants into hydrocarbons. The most active are Pd catalysts, so the development of improved Pd-containing systems is of practical and theoretical importance. In this work 2 wt.% of Pd was supported by wet impregnation on two types of alumina (E, produced by Engelhard, and C, produced by calcination of boemite at 600°C), both pristine and modified with 20 wt.% of H₈[Si(W₂O₇)₆]×6H₂O (HPS), and tested in multi-phase HDC [1] of 1,3,5-trichlorobenzene (TCB, 3.5×10⁻⁴ M, isooctane, 0.1 g of catalyst, Aliquat-336 as interphase transfer agent, 20% KOH water solution, 5 ml/min of H₂, 50°C). TEM HR demonstrated uniform distribution of HPS and Pd on the surface of alumina, Pd particle size is less than 20 nm and increases in the raw Pd/HPS/Al₂O₃ (C) < Pd/HPS/Al₂O₃ (E) < Pd/Al₂O₃ (C) < Pd/Al₂O₃ (E). According to TPR data, significant part of Pd easily reduces even at RT and form PdH_x in H₂, but the other part is more difficult to reduce, and it is lower in HPS-modified catalysts. This fraction grows after catalytic tests.

Table 1. Catalytic properties

Catalyst	TOF, h ⁻¹	t ₅₀ , min
Pd/Al ₂ O ₃ (E)	11	180
Pd/Al ₂ O ₃ (C)	30	250
Pd/ГПС-Al ₂ O ₃ (E)	18.2	62
Pd/ГПС-Al ₂ O ₃ (C)	34.6	105

Using IR DO spectroscopy after CO

adsorption, it was found that relatively large Pd⁰ particles are the major form of palladium in non-modified catalysts. In modified catalysts the fraction of Pd⁰ is much lower, and single Pd⁺ and Pd²⁺ ions are also found. The features of Lewis

acidity of the catalysts surface and the presence of HPS and/or the products of its thermal decomposition define the possibility of TCB adsorption and activation not only on Pd, but also on a support. Then adsorbed moieties can be reduced with hydrogen spilling over the surface from Pd and W centers. Also the new types of active centers can be formed on the border between Pd and HPS particles. All listed phenomena provide an improvement of catalysts activity (Table 1, t₅₀ is time of 50% TCB conversion) and stability in aggressive reaction medium after modification of a support with HPS. The catalysts on Al₂O₃ (E) have better performance due to the feature of structure, as it was found by TEM and XRD.

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