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Author: Cecília A. Badari Ferenc Lónyi Eszter Drotár
Alexander Kaszonyi József Valyon



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A study of the hydrodenitrogenation of propylamine over supported nickel phosphide catalysts using amorphous and nanostructured silica supports

Cecília A. Badari¹, Ferenc Lónyi^{1,*}, Eszter Drotár¹, Alexander Kaszonyi², József Valyon¹

¹ *Institute of Materials and Environmental Chemistry, Research Centre for Natural Sciences, Hungarian Academy of Sciences, H-1117 Budapest, Magyar tudósok körútja 2., Hungary*

² *Department of Organic Technology, Slovak University of Technology, Radlinského 9, Bratislava, SK-81237, Slovak Republic*

Research Highlights

- Propylamine (PA) hydroconversion was applied as model reaction of bone oil HDN.
- The Ni₂P/SiO₂ catalyst was active in the C-N but inactive in C-C hydrogenolysis.
- Operando DRIFTS study confirmed that DPA is intermediate of the PA HDN reaction.
- The protonated N-bases over the Ni₂P/SiO₂ catalyst are spectator species in the reaction.
- Defective Ni₂P crystals of fresh catalysts take up ordered structure during reaction.

Abstract

Pyrolysis of animal by-products provides pyro-oil that contains about 10 wt% nitrogen mainly in aliphatic compounds and virtually no sulfur. The nitrogen should be removed from the oil preferably by hydrodenitrogenation (HDN) to obtain liquid fuel and ammonia. In order to understand the catalytic HDN processes of such bio-oils the HDN of propylamine (PA) was studied as model reaction over silica-supported Ni₂P and Ni catalysts. Catalysts were prepared by H₂-reduction of catalyst precursor NiO/phosphated silica gel and NiO/phosphated SBA-15 silica material and NiO/silica gel. The catalyst surface was characterized by Diffuse Reflectance Infrared Fourier Transform Spectroscopic (DRIFTS) examination of adsorbed CO. It was shown that the Ni₂P particles had a highly defected structure in the initial reduced state of the catalyst but were rapidly converted to ordered crystals in contact with PA under reaction conditions. The HDN reaction of PA was studied using a flow-through tube reactor and also by *in situ* DRIFTS experiments in the 16-60 bar pressure and 200-400 °C temperature range. Below about 300 °C the Ni₂P catalysts were active in the direct

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