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研究论文

Application of Hydrodechlorination in Environmental Pollution Control: Comparison of the Performance of Supported and Unsupported Pd and Ni Catalysts

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- 摘要
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摘要 Catalytic hydrodechlorination (HDC) is an innovative means of transforming chlorinated waste streams into a recyclable product. In this study, the gas phase HDC of chlorobenzene (CB) has been studied over bulk Pd and Ni and ((8 ± 1) wt%) Pd and Ni supported on activated carbon (AC), graphite, graphitic nanofibers (GNF), Al_2O_3, and SiO_2. Catalyst activation was examined by temperature-programmed reduction (TPR) analysis and the activated catalysts characterized in terms of BET area, transmission electron microscopy, scanning electron microscopy, H_2 chemisorption/temperature-programmed desorption, and X-ray diffraction measurements. Metal surface area (1–19 m^2/g), TPR, and H_2 uptake/release exhibited a dependence on both metal and support. The Pd system delivered specific HDC rates that were up to three orders of magnitude greater than that recorded for the Ni catalysts, a result that we link to the higher H_2 diffusivity in Pd. HDC was 100% selective over Ni while Pd also produced cyclohexane (selectivity < 4%) as a result of a combined HDC/hydrogenation. Bulk Pd outperformed carbon supported Pd but was less active than Pd on the oxide supports. In contrast, unsupported Ni presented no measurable activity when compared with supported Ni. The specific HDC rate was found to increase with decreasing metal surface area where spillover hydrogen served to enhance HDC performance.

关键词： hydrodechlorination chlorobenzene bulk palladium and nickel catalysts activated carbon graphite carbon nanofiber alumina silica spillover hydrogen

Abstract： Catalytic hydrodechlorination (HDC) is an innovative means of transforming chlorinated waste streams into a recyclable product. In this study, the gas phase HDC of chlorobenzene (CB) has been studied over bulk Pd and Ni and ((8 ± 1) wt%) Pd and Ni supported on activated carbon (AC), graphite, graphitic nanofibers (GNF), Al_2O_3, and SiO_2. Catalyst activation was examined by temperature-programmed reduction (TPR) analysis and the activated catalysts characterized in terms of BET area, transmission electron microscopy, scanning electron microscopy, H_2 chemisorption/temperature-programmed desorption, and X-ray diffraction measurements. Metal surface area (1–19 m^2/g), TPR, and H_2 uptake/release exhibited a dependence on both metal and support. The Pd system delivered specific HDC rates that were up to three orders of magnitude greater than that recorded for the Ni catalysts, a result that we link to the higher H_2 diffusivity in Pd. HDC was 100% selective over Ni while Pd also produced cyclohexane (selectivity < 4%) as a result of a combined HDC/hydrogenation. Bulk Pd outperformed carbon supported Pd but was less active than Pd on the oxide supports. In contrast, unsupported Ni presented no measurable activity when compared with supported Ni. The specific HDC rate was found to increase with decreasing metal surface area where spillover hydrogen served to enhance HDC performance.

Keywords： hydrodechlorination, chlorobenzene, bulk palladium and nickel catalysts, activated carbon, graphite, carbon nanofiber, alumina, silica, spillover hydrogen

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