Catalysis Letters (2024) 154:3457–3471
https://doi.org/10.1007/s10562-023-04568-9

Highly Effective Zirconia/γ-Alumina for Continuous Vapor Phase *N*-methylation of Aniline

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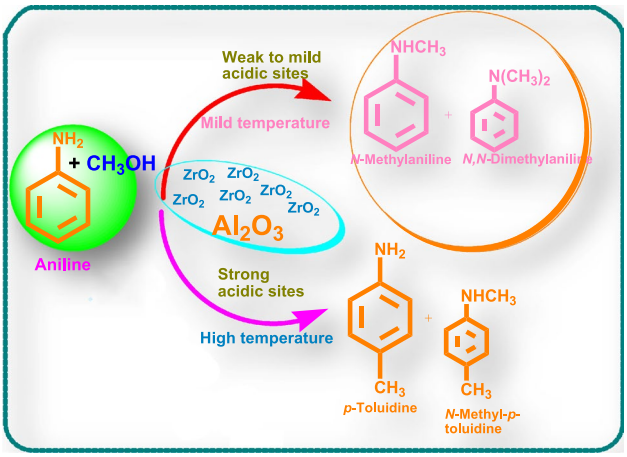
Received: 9 November 2023 / Accepted: 22 December 2023 / Published online: 30 January 2024

Abstract

High-surface-area $ZrO_2/\gamma\text{-}Al_2O_3$ catalysts were used for the selective *N*-methylation of aniline with methanol at mild temperature and atmospheric pressure conditions. The activity and stability of these catalysts were maintained throughout the process without significant changes in conversion and selectivity of the target products. These $\gamma\text{-}Al_2O_3$-supported ZrO_2 catalysts were synthesized by a facile wet impregnation method, and their zirconia loadings varied from 10 to 50%. In order to enhance the catalytic activity, changes were implemented on the zirconia loadings under varying reaction parameters. *N*-methyl aniline can be produced preferentially at 270 °C with a catalyst composed of 20 wt % $ZrO_2/\gamma\text{-}Al_2O_3$. This catalyst was able to acquire a higher aniline conversion during methylation because of its enough surface area and the presence of weak and moderate Lewis acidic sites in its $\gamma\text{-}Al_2O_3$ and ZrO_2 components. The catalytic activity and product distribution were also assessed by adjusting a variety of reaction parameters, like temperature, ratios, and catalyst loadings, respectively. The structural and textural properties of synthesized catalysts are thoroughly characterized using a number of different analytical techniques. Thus, the current protocol can be considered as a simpler, reproducible, and environmentally benign approach for *N*-methylation of amines.

Graphical Abstract

Zirconia/γ-alumina for *N*-methylation of aniline in vapor-phase continuous mode



Keywords Aniline · Methanol · *N*-methyl aniline · *N*, *N*-dimethyl aniline · Methylation

Extended author information available on the last page of the article