Improving Nickel Catalyst Dispersion on Phenol Hydrodeoxygenation by Amine-Assisted Impregnation

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Significance and Relevance

This work explores nitrogen-chelating agents to enhance nickel dispersion on support surfaces further. Obtaining a 70% dispersion of Nickel as active phase in SiO_2 changing the product distribution over phenol hydrodeoxygenation, this study may be applicable in all transition metal to improve their dispersion with an affordable treatment.

Preferred and 2nd choice for the topic: Green chemistry and biomass transformation, renewable resources conversion(preferred), Sustainable and clean energy production and transport (2nd choice)
Preferred presentation: Poster

Introduction and Motivations

Catalytic reactions occur on the metal surface, making dispersion and surface properties key for catalyst activity and selectivity. Common methods for preparing Ni-supported catalysts include incipient wetness impregnation, ion exchange, and precipitation-deposition. Nickel nitrate is often preferred for its solubility and low decomposition temperature, though it can lead to metal agglomeration on high-surface-area supports [1].

To improve metal dispersion, sacrificial transition metal complexes have been used in impregnation methods [5], with chelating agents like citric acid [2], ethylenediamine [3], and EDTA [4]. Recent studies, such as Gao *et al.* [5], show that Ni-amine complexes yield high dispersion and small Ni particles.

Materials and Methods

Ni/SiO_2 catalysts with amines of different N numbers were prepared via incipient wetness impregnation with a 10 wt% nickel loading. $Ni(NO_3)_2{\cdot}6H_2O$ was dissolved in water with various chelating agents—NH_3, ethylenediamine (en), diethylenetriamine (DETA), and pentaethylenehexamine (PEHA)—at amine/Ni molar ratios of 6, 3, 2, and 1, respectively, to form $Ni(NH_3)_6$, $Ni(en)_3$, $Ni(DETA)_2$, and Ni(PEHA) complexes. After aging at room temperature and drying at 90°C, the precursors were calcined at 550°C for 2 h and labeled Ni/SiO_2, $Ni(NH_3)/SiO_2$, $Ni(en)/SiO_2$, $Ni(DETA)/SiO_2$, and $Ni(PEHA)/SiO_2$. Catalytic stability was evaluated in the vapor phase at 250°C using a fixed-bed quartz U-tube reactor. All catalysts were reduced in situ with H_2 (60 mL/min, 550°C) before introducing phenol into the reactor via an H_2 flow through a saturator at 100°C. Products were analyzed using a Nexis 2030 gas chromatograph.

Results and Discussion

Catalysts active phase dispersion, particle diameter and surface area are displayed in Table 1. It is observed a marked improvement in the dispersion of the nickel particles with the used of Ni-Amine clomplexes as a sacrificial agent. It is worth noting that the most dispersed active phase was found to be the Ni(PEHA) complex with a 70% dispersion and a 1.44 nm particle size obtained by CO-chemisorption and correlated with the particle size distribution of TEM. Besides, figure 1 is observed the difference of the selectivity distribution between Ni/SiO_2 and $Ni(PEHA)/SiO_2$ which could be related to the dispersion of the active phase.

Table 1: catalysts surface area, particle diameter obtained by TEM and CO-Chemisorption and dispersion

Catalysts	S_{BET} (m^2/g)	dp_{TEM}(nm)	dp_{CO}(nm)	Ni (%) Dispersion
SiO_2	347	-	-	-
Ni/SiO_2	285	9.89	14.79	7
$Ni(NH_3)_6/SiO_2$	383	1.97	1.56	64
$Ni(en)_3/SiO_2$	368	2.95	1.72	59
$Ni(PEHA)/SiO_2$	292	1.60	1.44	70

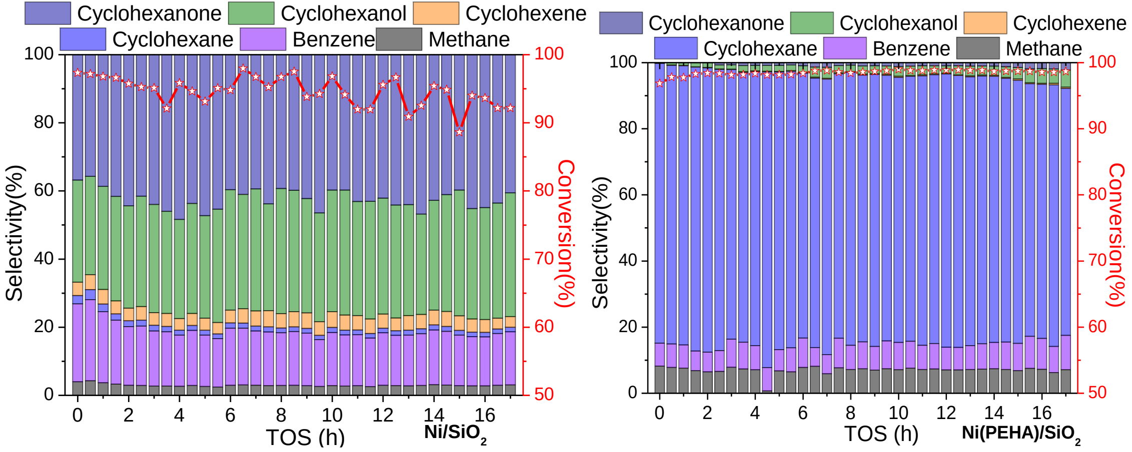

Figure 1 conversion and selectivity fixed-bed hydrodeoxygenation of phenol

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Acknowledgements

This research was funded by the following sources: Fondequip No. EQM160070, The Millennium Science Initiative Program-NCN2021_090, FONDECYT regular (Grant No. 1220763), FONDEF ID23I10149, ANID ACT240050; C.C.D. was funded by Agencia Nacional de Investigacion y Desarrollo (ANID), Chile through doctoral grant 2021-(grant No. 21211077); and C.A.L. was funded by ANID, Chile, through FONDECYT Postdoctoral (Project3220395).