

Catalytic hydrogenation of biomass derived levulinic acid using zero valent non-precious metal Fe catalysts based on N-triphos ligand

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The global concern on declining crude oil resources and efforts to reduce the anthropogenic emission of CO₂ has led to an intensified search for renewable and environmentally benign alternative sources of carbon for the production of transportation fuels and chemical. Biomass remains the most suitable and sustainable alternative that meets both the demand for clean energy and the production of liquid transportation fuels and chemicals.¹ Levulinic acid (LA) is an important biomass derived platform molecule that can be transformed to more valuable chemicals and fuels, with catalysis playing a key role in its transformations. Ru-triphosphine complexes have recently proven to be excellent homogeneous catalysts for effecting the hydrogenation of LA to gamma-valerolactone (GVL), 1, 4-pentanediol (1, 4-PDO) and 2-methyltetrahydrofuran (2-MTHF), however, there are few reports on non-precious based metal catalyst for this transformation.^{2,3}

Herein we explore the catalytic hydrogenation of LA to GVL, 1, 4-PDO and 2-MTHF using more sustainable non-precious metal Fe complexes as catalysts (fig.1). The Fe metal precursors Fe₃CO₁₂ and FeCO₅ were chosen because of their previously reported catalytic activity and commercial availability. The novel bimetallic [Fe(CO)₃(K²-NP3^{Ph})(μ-PPH₂)Fe(CO)₄] and monometallic Fe(CO)₃(K²-NP3^{Ph}) Fe⁰ complexes were synthesized from

the reaction of N, N, N- tris(diphenylphosphinomethyl) amine (N-triphosPh) ligand with Fe₃CO₁₂ and Fe(C₈H₈)(CO)₃ precursors respectively. Catalysis of LA was performed in a high pressure autoclave under the conditions of 150o C and 50 bar H₂. Near quantitative conversion of LA was observed in most cases, yields were determined by GC.



Fig. 1 Hydrogenation pathway of levulinic acid (LA)

[1] M. J. Climent, A. Corma, S. Iborra. *Green Chem.* 2014, 16 (2), 516. [2] F. M. A. Geilen, B. Engendahl, A. Harwardt, W. Marquardt, J. Klankermayer, W. Leitner. *Angew.Chemie - Int. Ed.* 2010, 49 (32), 5510 –5514. [3] H. Zhong, Q. Li, J. Liu, G. Yao, J. Wang, X. Zeng, Z. Huo, F. Jin. *ACS Sustainable Chem. Eng.* 2017, 5, 6517– 6523.

Biography

Uwaila Omoruyi obtained her BSc (2006) and MSc (2013) in industrial chemistry from the University of Benin, Nigeria. She then proceeded to United Kingdom in 2014 to pursue a PhD in the Department of Chemistry, Imperial College London under the supervision of Dr. Philip Miller. She is currently in the final year of her PhD and her research is focused on the catalytic hydrogenation of biogenic acids using novel phosphine complexes. Uwaila is a recipient of a scholarship award from the Nigerian Petroleum Technology Development Fund (PTDF)

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