

Insight into the activity, selectivity, and stability of heterogeneous catalysts in the continuous flow hydrogenation of levulinic acid and its esters

József M. Tukacs, Áron Sylvester and László T. Mika
Department of Chemical and Environmental Process Engineering, Budapest University of Technology and Economics, Budapest, Hungary

Abstract
The global efforts to reduce the carbon dioxide emission and replacement of fossil-based resources demand new and innovative strategies for the green production of fuels, platform molecules and value-added chemicals. The selective conversion of non-edible carbohydrates, *e.g.* cellulose, chitin, chitosan into platform molecules plays a key role in sustainable development.[1] The acid catalyzed dehydration of non-edible carbohydrates results in equimolar formation of levulinic acid (LA) and formic acid.[2] Subsequently, the LA and levulinic acid esters (methyl- and ethyl-levulinate) can be converted to γ-valerolactone GVL, which can be considered as a sustainable liquid,[3] either by homogeneous,[4] or heterogeneous catalyst systems. Recently, the catalytic transfer hydrogenation of LA to GVL in a continuous alternative system was reported.[5] The H-Cube® tubular reactor is one of the most promising techniques for the high-throughput heterogeneous catalytic hydrogenation under continuous-flow conditions. Here we report the application of the H-Cube® and H-Cube Pro™ tubular reactors for the hydrogenation of LA and LA esters using environmentally bening solvents such as water and alcohols in the presence of $(C_6H_4\text{-}m\text{-}SO_3Na)_2PBu$ phosphine modified catalyst. We have demonstrated, that the activity of the Ru-based catalyst system can be increased by the application of phosphine ligands[6, 7] as well as the LA and LA esters can be selectively and quantitatively converted to GVL in the presence of 5% Ru/C, 5% Pt/C, 10% Pd/C and Raney Ni catalyst and bidentate $Ph_2P(CH_2)_nPPh_2$ (n = 1 – 3) ligands. Details of the reaction conditions, the effect of chelating ring on the activity and the catalyst recycling will be presented.

Biography
József M. Tukacs finished his studies in Organic and Organometallic Chemistry at Budapest University of Technology and Economics, Budapest, Hungary. He works as an assistant professor at the University, in the research team of catalytic processes, led by László T. Mika. It is also apparent from his list of publications. He has published more than 10 papers in reputed journals and he has co-author of a book chapter. He received his papers more than 160 independent references.

Presenting author details
Full name: József M. TUKACS
Session name/ number: Green Solvent: Replacing Organic Solvents/ Track 4
Category: Oral
Email id: tukacsjozsi@mail.bme.hu
Contact number: +36 30 463 2574

[1] Mika, L. T.; Cséfalvay, E.; Németh, Á. *Chem. Rev.* **2018**, *118*, 505.
[2] Szabolcs, Á.; Molnár, M.; Dibó, G.; Mika, L. T. *Green Chem.*, **2013,** *15*, 439.
[3] Horváth, I. T.; Mehdi, H.; Fábos, V.; Boda, L.; Mika, L. T. *Green Chem.*, **2008,** *10*, 238.
[4] Tukacs, J. M.; Király, D.; Strádi, A.; Novodárszki, Gy.; Eke, Zs.; Dibó, G.; Kégl, T.; Mika, L. T. *Green Chem.*, **2012,** *14*, 2057.
[5] Kopetzki, D.; Antonietti, M. *Green Chem.*, **2010,** *12*, 656.
[6] Tukacs, J. M.; Jones, R. V.; Darvas, F.; Dibó, G.; Lezsák, G.; Mika, L. T. *RSC Adv.*, **2013,** *3*, 16283.
[7] Tukacs, J. M.; Novák, M.; Dibó, G.; Mika, L. T. *Catal. Sci. Technol.*, **2014,** *4*, 2908.