ISGC 2022

N°11 / OC
TOPIC(s) : Waste and side streams valorization / Homogenous, heterogenous and biocatalysis

Renewable methanol synthesis from CO2 with indium-based catalysts formed by pulsed laser ablation in liquids

AUTHORS

Patrick SCHUEHLE / UNIVERSITY ERLANGEN-NUREMBERG, EGERLANDSTRASSE 3, ERLANGEN
Kinran LAU / UNIVERSITY DUISBURG-ESSEN, BUNDESSTRASSE 27, DUISBURG
Sven REICHENBERGER / UNIVERSITY DUISBURG-ESSEN, BUNDESSTRASSE 27, DUISBURG
Shunxing LIANG / UNIVERSITY DUISBURG-ESSEN, BUNDESSTRASSE 27, DUISBURG
Jakob ALBERT / INSTITUTE FÜR TECHNICAL AND MACROMOLECULAR CHEMISTRY, BUNDESSTRASSE 45, HAMBURG

PURPOSE OF THE ABSTRACT

With intensifying global warming and depleting fossil fuels, it is desirable to convert anthropogenic CO2 into valuable chemicals. In this respect, methanol is a promising feedstock for producing commodity chemicals such as olefins and formaldehyde, since its demand has been increasing continuously over the past decade.[1] Moreover, the energy generated from renewable sources has to be stored in certain chemical energy carriers before usage.[2] With a high volumetric energy density (4.33 kWh L-1), methanol is an ideal candidate for chemical energy storage, which can be produced from CO2 and hydrogen derived from water electrolysis.[3]
Recently, supported In2O3-catalysts have shown excellent long time stability and higher methanol selectivity compared to traditional Cu/ZnO-based systems.[4] However, the activity of indium-based catalysts is very susceptible to the preparation method which is typically wet impregnation or co-precipitation.[5] An alternative and innovative catalyst preparation method, namely "pulsed laser ablation in liquids" (PLAL) was successfully applied for several common active metals (Pt, Pd, Au, Cu) in the past.[6] Thereby, the method offered high process control for the design of catalyst parameters like particle size and defined elemental composition.
Here, we explore a laser-based synthesis route to prepare In2O3-catalysts of varying indium size and load. To the best of our knowledge, In2O3 catalysts prepared by "pulsed laser ablation in liquids" (PLAL) were applied in thermally activated catalysis for the first time. Several batches, each consisting of catalysts with different indium loadings (2-25 wt%) on ZrO2, were prepared using individual laser synthesis parameters.
By choosing the wavelength of the (infrared or green) laser light, we were able to adjust the size distribution of the In2O3-nanoparticles, while the surrounding liquid of the synthesis process influenced their chemical composition. After deposition of In2O3 on several commercially available oxidic supports (e.g. TiO2, CeO2, ZrO2) the properties of the catalysts were analyzed using XRD, XPS, H2 TPR and HR-TEM. Depending on the parameters of the PLAL process, the catalysts showed a distinct difference in crystallinity and morphology.
Subsequently, the catalysts were applied for methanol synthesis from CO2 in a slurry reactor concept that has shown interesting process features in our previous publication.[7] The choice of the support had a clear effect on the catalytic performance. Although crystallinity and morphology of the indium species were depending on the settings of the laser process, catalytic activity stayed comparably robust. Post-reaction analysis of these catalysts revealed a stepwise convergence of their composition and properties by oxidation and recrystallization processes, explaining the similar activity in CO2-hydrogenation. Part of these scientific results were published in 2021 in ACS Applied Energy Materials.[8]

FIGURES

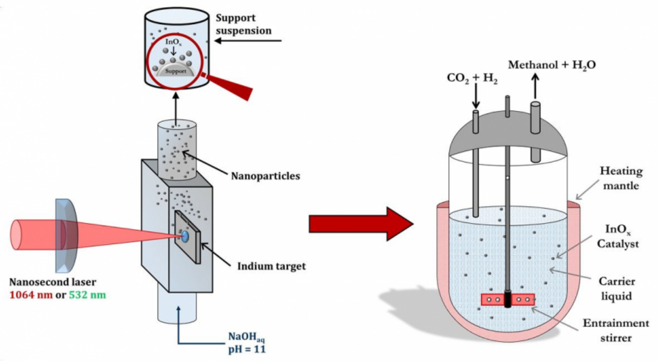

FIGURE 1

Synthesis of InOx-catalysts by PLAL and their usage for slurry phase CO2-hydrogenation.

Schematic representation of the PLAL setup and procedure to synthesize supported InOx nanoparticles.

These particles are applied as catalyts in slurry methanol synthesis afterwards.

FIGURE 2

KEYWORDS

Pulsed laser ablation in liquids | CO2-hydrogenation | indium catalysts | slurry methanol synthesis

BIBLIOGRAPHY