Synthesis of Functional Materials for Organic Transformation of Carbohydrates to Furan Based Compounds

By

Chatterjee, AMRITA

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Examination Committee:
Prof. Volkan Kursun, Chairman
Prof. Xijun HU, Supervisor
Prof. Minhua SHAO, Prof. Frank Leung-Yuk LAM, CBE
Prof. Irene Man Chi LO, CIVL
Prof. Duong DO, University of Queensland

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Abstract

Synthesis of the platform chemical, furfural, which is one of the precursors for biofuels production, can be done by the biomass dehydration process. Absence of a suitable heterogeneous catalyst for highly selective production of furfural has been a long-standing issue in the industrial world. Metal-Organic-Frameworks (MOFs) are one of the promising catalysts due to their ultra-high porosity and high flexibility to provide anchorage through the organic linkers. Tuning the organic moieties impart functionality as well as alter the pore size, which is crucial for product yield and selectivity. However, MOFs always suffer from low hydrothermal stability, which occurs due to the hydrolysis of 3-D structure in boiling water or acidic medium of pH~1-2. Further they lack Brønsted acidity, which is pivotal in catalytic syntheses of fine chemicals in bio-refineries.

In this work, for the first time we have explored the properties of the Lewis acidic MOF, MIL-101 (Cr), as the heterogeneous catalyst for xylose-to-furfural organic transformation. It has been found that the pristine MIL-101 (Cr), yields 49 % furfural and collapses after 4 cycles of catalytic runs. While organosilane-coated MOF, synthesized at room temperature has enhanced hydrothermal stability and can withstand twice the number of catalytic runs than pristine MOF. Simultaneous improvement of Brønsted acidity with hydrothermal stability of MOFs have been achieved by compositing it with materials like activated coal-fly-ash waste and mesoporous tin phosphate. These dual-acidic core shell hybrids have successfully achieved 71 % and 87 % of furfural yield respectively at 150°C, which surpasses the present performances of solid acid catalysts at such lower temperature. Relevant post-reaction characterizations indicate that the crystallinity, porosity and acidity of the developed MOF-based catalysts are maintained even after 10 cycles of catalytic runs. This study reveals new possibilities of tailoring MOFs not only as catalysts but also for other proton-conducting applications which require hydrothermal stability.

- ALL ARE WELCOME -