Metal-organic frameworks for catalytic conversion of CO_2

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Growing CO_2 emissions into the atmosphere is one of the key problems to solve in the 21st century. Methods that can help reduce CO_2 emissions include its chemical conversion into valuable chemicals and fuels. The motivation for the research described in this doctoral dissertation is the need to develop new efficient catalysts for methanol and cyclic carbonates synthesis. Metal-organic frameworks (MOFs) are inviting candidates for this role due to their properties, i.e. crystalline structure, unique textural properties, and the availability of exposed active sites. Moreover, MOFs can be extensively modified to improve their catalytic properties.

The research aimed to determine the impact of modifications on HKUST-1, MIL-100(Fe), and PCN-222 physicochemical properties and activity in two CO_2 conversion reactions, i.e. (i) synthesis of cyclic carbonates in the cycloaddition reaction CO_2 to epoxides and (ii) methanol synthesis via hydrogenation of CO_2. Modifications included MOF/GO composites and bimetallic structures synthesis as well as the post-synthetic modification of MOFs via a metal phase and organic compound introduction.

The doctoral thesis begins with an extensive literature introduction, which addresses the issue of excessive CO_2 emissions and carbon capture and utilization methods. The application potential of metal-organic frameworks in the CO_2 conversion to cyclic carbonates and methanol was also discussed. The following chapters present the aim and scope of the research as well as a description of the performed experiments. The results of my studies presented in Chapter 4 are divided into four parts regarding:

1. Research on the performance of HKUST-1/GO and HKUST-1/rGO composites in cyclic carbonates synthesis (HKUST-1/rGO composites were obtained via a new method involving cooper-modified reduced graphene oxide).
2. Research on the performance of cerium-modified HKUST-1/rGO composites in a styrene carbonate synthesis.
3. Research on the activity of bimetallic metal-organic frameworks MIL-100(Fe/Zn) in cyclic carbonates synthesis (including synthesis without an external cocatalyst).
4. Research on the performance of PCN-222 modified with cerium and/or copper in methanol synthesis.

The dissertation concludes with a summary and the most important inferences arising from the presented research.

The results showed in this doctoral dissertation deepen the knowledge about the physicochemical properties of modified metal-organic frameworks and open new perspectives in the development of CO_2 conversion catalysts.