Invitation à la soutenance publique de thèse

Pour l’obtention du grade de Docteur en Sciences agronomiques et ingénierie biologique

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Catalytic hydrogenation of carbon dioxide under mild reaction conditions

This thesis focuses on the study of the hydrogenation of CO₂ at mild conditions (Temperature < 200°C, Pressure < 7 bar) through heterogeneous catalysis. The fundamental aspects of the reactions are assessed by studying the physicochemical properties of the catalysts, the kinetic parameters of the reactions and looking at the reaction intermediates by in-situ infrared spectroscopy.

The first part of the work is devoted to the study of the effect of support and the size of Rh particles on the transformation of CO₂ to methane. Rh/Al₂O₃ and Rh/TiO₂ catalysts with different Rh particle size are studied. Selectivity to methane is 100%. Results put in evidence the effect of Rh nanoparticle size on the activity. Results are explained by studying the type and stability of surface intermediates at steady state and in transient experiments. The effect of the addition of another metallic phase (Pd/Al₂O₃) to Rh/Al₂O₃ catalyst is also studied. It is shown that Pd/Al₂O₃ (itself not active at low temperatures) can be activated when contacted with Rh/Al₂O₃ leading to a synergistic effect. A mechanistic model is proposed to explain these results.

The second part of the work concerns the hydrogenation of CO₂ to methanol. Cu/ZnO catalysts are prepared by wet impregnation and citrate methods. A cooperation effect between Cu and ZnO is evidenced using mechanical mixtures. The effect of copper cluster size and support on activity and selectivity is studied. Selectivity to methanol is near 100% at temperatures lower than 160°C. Results underline the key effect of the size of Cu particles on selectivity.

It is expected that the results presented in this thesis could serve as guidelines for preparing active and selective catalysts in industrial CO₂ hydrogenation processes operating at mild reaction conditions in order to improve the sustainability of catalytic processes and reduce energy consumption.

Membres du jury :
Professeur P. RUIZ BARRIENTOS (UCL) (Promoteur)
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Professeur G. HEYEN (ULg)
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Vendredi 30 août 2013 à 15h00
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