THE EFFECT OF THE REACTION CONDITIONS ON THE DEACTIVATION OF A HIGHLY ACTIVE Ru/TiO_2 CATALYST FOR CO_2 HYDROGENATION.

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ABSTRACT

The hydrogenation of carbon dioxide (CO_2) is a reaction of strategic importance both in terms of reducing the levels of a key Greenhouse Gas from the atmosphere but also in the production of fuels and high added value chemicals. For the case of the CO_2 methanation (Sabatier reaction) and the parallel Reverse Water Gas Shift (RWGS) reaction, a wide range of catalysts have been examined including noble (Ru, Rh, Pd) and non-noble (Ni, Cu, Fe) metals, on a variety of oxide supports ranging from reducible (CeO_2, TiO_2) to irreducible (Al_2O_3), zeolites and many more.[1-3] Reducible supports tend to enhance the adsorption and activity of CO_2, due to the role of oxygen vacancies in proposed redox cycles, and in particular TiO_2 shows a high catalytic interest due to its' multiple structures of anatase and rutile.[1,4] This difference in crystalline structure can affect the interactions of the metal oxide support with the catalytically active metal nanoparticles dispersed on the surface and therefore modify the reactivity of the catalyst.[5]

Ru/TiO_2 catalysts have been reported to display remarkable performance both in terms of activity and selectivity towards methane (CH_4).[5-8] In this project, a conventional Ru/TiO_2(P25) catalyst prepared by wet impregnation was used to study the methanation reaction at a temperature range of 150-450 ^oC and atmospheric pressure. Conventional light-off experiments confirm the catalyst's initial high activity and selectivity towards the Sabatier reaction, reaching a maximum yield of ca. 86% (375 ^oC, P_{CO_2}:P_{H_2}=1:7), and on stream experiments at 400 ^oC show a deactivation of ca. 5% after 80 hours on stream. The deactivation pattern of the catalyst, however, changes when the catalyst is tested under light-off conditions. Here, we explore the structural and electronic changes during the light-off experiments as a deactivation mechanism of the catalyst.

KEYWORDS: CO_2 methanation, Ru/TiO_2, catalyst deactivation, Sabatier

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