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The Power of in situ Characterization Techniques in Heterogeneous Catalysis: A Case Study of Deacon Reaction

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Abstract : Introduction: The conventional approach of characterizing solid catalysts under static conditions, i.e., before and after reaction, does not provide sufficient knowledge on the physicochemical processes occurring under dynamic conditions at the molecular level. Hence, the necessity of improving new in situ characterizing techniques with the potential of being used under real catalytic reaction conditions is highly desirable. In situ Prompt Gamma Activation Analysis (PGAA) is a rapidly developing chemical analytical technique that enables us experimentally to assess the coverage of surface species under catalytic turnover and correlate these with the reactivity. The catalytic HCl oxidation (Deacon reaction) over bulk ceria will serve as our example. Furthermore, the in situ Transmission Electron Microscopy is a powerful technique that can contribute to the study of atmosphere and temperature induced morphological or compositional changes of a catalyst at atomic resolution. The application of such techniques (PGAA and TEM) will pave the way to a greater and deeper understanding of the dynamic nature of active catalysts. Experimental/Methodology: In situ Prompt Gamma Activation Analysis (PGAA) experiments were carried out to determine the Cl uptake and the degree of surface chlorination under reaction conditions by varying p(O2), p(HCl), p(Cl2), and the reaction temperature. The abundance and dynamic evolution of OH groups on working catalyst under various steady-state conditions were studied by means of in situ FTIR with a specially designed homemade transmission cell. For real in situ TEM we use a commercial in situ holder with a home built gas feeding system and gas analytics. Conclusions: Two complimentary in situ techniques, namely in situ PGAA and in situ FTIR were utilities to investigate the surface coverage of the two most abundant species (Cl and OH). The OH density and Cl uptake were followed under multiple steady-state conditions as a function of p(O2), p(HCl), p(Cl2), and temperature. These experiments have shown that, the OH density positively correlates with the reactivity whereas Cl negatively. The p(HCl) experiments give rise to increased activity accompanied by Cl-coverage increase (opposite trend to p(O2) and T). Cl2 strongly inhibits the reaction, but no measurable increase of the Cl uptake was found. After considering all previous observations we conclude that only a minority of the available adsorption sites contribute to the reactivity. In addition, the mechanism of the catalysed reaction was proposed. The chlorine-oxygen competition for the available active sites renders re-oxidation as the rate-determining step of the catalysed reaction. Further investigations using in situ TEM are planned and will be conducted in the near future. Such experiments allow us to monitor active catalysts at the atomic scale under the most realistic conditions of temperature and pressure. The talk will shed a light on the potential and limitations of in situ PGAA and in situ TEM in the study of catalyst dynamics.

Keywords : CeO2, deacon process, in situ PGAA, in situ TEM, in situ FTIR
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