Séminaire :

« Understanding selectivity in partial oxidation of olefins »

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Salle RAPHAEL 1 – site d’Ivry-sur-Seine

A periodic density functional study of the epoxidation mechanism of ethylene and propylene on Cu(111) and Ag(111) surfaces will be presented. This is a competitive reaction leading to epoxide and aldehyde from a common oxametallacycle intermediate. For the ethylene epoxidation reaction it is found that while Ag presents a larger catalytic activity, it also presents a lower selectivity towards the epoxide. The enhanced selectivity of Cu is attributed to the late nature of the transition state forming epoxide from the common oxametallacycle. A similar mechanism holds for propylene epoxidation although here it competes with proton abstraction from adsorbed oxygen leading to an adsorbed allyl species which cannot evolve to the oxametallacycle intermediate. Precisely, H abstraction is found to be preferred on Ag(111) and this is the reason for the poor performance of Ag in forming propylene oxide. On the other hand, it is shown that Cu does not exhibit this trend and hence could be the basis for an alternative catalyst for propylene epoxidation.