Invitation à la soutenance publique de thèse

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

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Bio-ingénieur en chimie et bio-industries à finalité spécialisée

Behavior of Keggin heteropolyacid catalysts in the gas phase methanol-to-dimethylether reaction

Keggin heteropolyacids (HPAs) are metal-oxygen clusters being nowadays considered as highly promising catalysts for the gas phase conversion of methanol to dimethylether (DME), namely the so-called “fuel for the 21st Century”. Indeed, thanks to their unusually strong Brönsted acidity approaching the super-acid region, Keggin HPAs allow reaching high methanol conversions at significantly lower temperatures than required by other, more conventional, acid catalysts. Lower temperatures imply a better selectivity to DME, a less rapid catalyst deactivation by coking, and a more energy-efficient process. However, trying to optimize the catalytic performance of the HPAs, which is what many studies currently aim at, requires a full understanding of their working mechanism that is to date not provided. Indeed, the literature still debates on whether the methanol reaction takes place at the surface or within the bulk of Keggin crystals. To clarify this was precisely the aim of the present thesis. The strategy was to use operando spectroscopy allowing to monitor the HPAs at work during the reaction. First, our results demonstrate that Keggin HPAs can actually operate through both the surface-type and the bulk-type mechanisms, depending on how they were pre-treated. Second, they show that activating the bulk-type mechanism renders pure Keggin HPAs so active that they largely win the competition against their hitherto preferred TiO$_2$-supported concurrents. In other words, our results overthrow the hitherto admitted views that pure HPAs are necessarily less active in the methanol-to-DME reaction than supported ones and that TiO$_2$ is an excellent support. Third, we show that challenging the performance of pure bulk-activated Keggin HPAs is actually possible with supports, but only provided that they do not lower the acid strength of the HPAs (a known phenomenon on TiO$_2$). As a successful example of supports meeting the latter requirement, we reveal here for the first time hexagonal boron nitride (BN).

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Mardi 12 septembre 2017 à 15h00
Salle de Séminaire ISV (B.059)
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