

In Situ Vibrational Spectroscopy of Reactants and Probe Molecules on Oxide Catalysts



MAX-PLANCK-GESELLSCHAFT

Friederike C. Jentoft



Vibrational spectroscopy is not a priori a surface-sensitive method but it becomes surface-sensitive when either functional groups present on the surface are analyzed or when observing an adsorbed probe or reactant. In this respect, it is a unique tool that often allows identifying changes of the catalyst itself upon adsorbing a molecule and the nature of the adsorbed species. Moreover, it can be applied in situ, i.e. while a catalytic reaction is in progress, and changes of surface functionalities and adsorbates can be monitored and correlated to catalytic performance. The accessible temperature and pressure ranges are wide, from liquid helium temperature to several hundred degrees Celsius and from the sub-Pa range to MPa. The possibilities of IR spectroscopy in investigating powder catalysts will be illustrated using two examples:

1) *Reactive probe molecule spectroscopy on silica-supported silver catalysts*

Silica-supported silver catalysts are suitable for the hydrogenation of α,β -unsaturated aldehydes to the respective alcohols [1]. In order to elucidate the mechanism of H_2 activation the interaction of H_2 with pure silica and an 8 at% Ag-containing catalyst were studied. Interaction of H_2 (or D_2) with the hydroxyl groups of the silica support was observed at 77 K. At 100 mbar D_2 and room temperature or above, exchange of OH- to OD-groups occurred. The exchange reaction requires cleavage of the hydrogen molecule and can thus be taken as a model reaction for hydrogen activation. The kinetics of the exchange reaction was studied spectroscopically.

2) *In situ DRIFTS investigations of n-butane isomerization catalyzed by sulfated zirconia*

With the diffuse reflectance method, spectra can be obtained of powder beds, which are more suitable for catalytic reactions than wafers, and possible changes to the sample through pressing—as have been found for sulfated zirconia [2]—can be avoided. Spectra taken of sulfated zirconia catalysts during isomerization at temperatures of 323 to 378 K and 1 kPa *n*-butane partial pressure are highly complex since they exhibit intense gas phase absorptions, alterations to the sulfate bands, and bands that possibly indicate the formation of water. Series of spectra were analyzed for simultaneous changes of various features, and correlations between spectral developments and the catalytic performance will be discussed.

[1] P. Claus, H. Hofmeister, J. Phys. Chem. B 103 (1999) 2766-2775; P. Claus, P.A. Crozier, P. Druska, Fresenius J. Anal. Chem. 361 (1998) 677-679.

[2] B.S. Klose, R.E. Jentoft, A. Hahn, T. Ressler, J. Kröhnert, S. Wrabetz, X. Yang, F.C. Jentoft, J. Catal. 217 (2003) 487-490.