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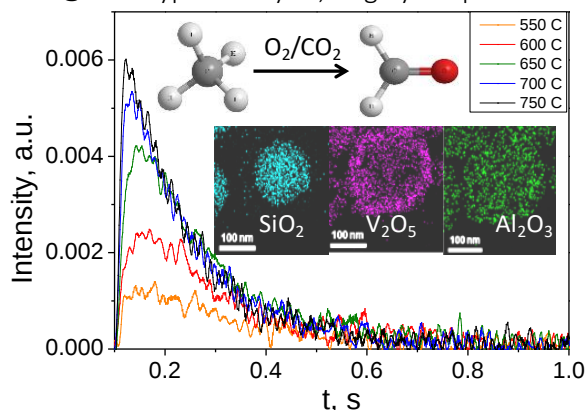
Selective oxidation of bio-methane to formaldehyde: core-shell catalyst approach

Aim

The selective oxidation of methane to formaldehyde, which avoids the production of carbon dioxide by full oxidation and is therefore important for the versatile utilization of methane, is still viewed as challenging. Here, we utilize catalyst synthesis followed by atomic layer deposition (ALD) and nanocoating to prepare an efficient and thermally stable catalyst based on $\text{SiO}_2@V_2O_5@Al_2O_3$ core@shell nanostructures.

Justification

Methane, the main component of biogas, is mostly used for heating and electricity generation. Recent progress in bio-methane technology presents a further stimulus for converting abundant methane to more valuable chemical feedstocks and thus reducing dependence on petroleum resources. Nevertheless, the four strong C-H bonds of methane (bond energy = 413 kJ/mol) present a serious obstacle to its chemical conversion. At elevated temperatures, methane can be catalytically converted to syngas, which can be used as a feedstock for the catalytic production of added-value hydrocarbons or alcohols. Although a number of indirect processes for the oxidative conversion of methane to formaldehyde (HCHO), methanol (CH_3OH), and ethylene (C_2H_4) have been developed and applied industrially, direct conversion of methane by partial oxidation is still challenging because of the above mentioned high C-H bond energy and the need to avoid carbon dioxide release as a greenhouse gas. V_2O_5/SiO_2 was identified as a good catalyst for methane partial oxidation, achieving methane to HCHO conversion and selectivity of 10 and 65%, respectively, at 600°C. The thermal stability of the catalytically active surface species can be increased by protection/encapsulation with an oxide. In well-designed core@shell-type catalysts, highly disperse active species in the core can be protected against



deactivation caused by sintering or coking during high-temperature reactions.

Atomic layer deposition (ALD) or nanocoating are considered attractive growth techniques for homogeneous encapsulation of active species, e.g. by Al_2O_3 , allowing the surface of core species to be uniformly coated with layers of controlled thickness at an atomic scale. Here, we will design highly disperse V_2O_5 nanocatalysts supported on SiO_2 spheres with Al_2O_3 overcoating for the direct oxidation of methane to HCHO.

Program

Transmission electron microscopy (TEM), scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDS), in situ X-ray diffraction (XRD), Raman spectroscopy, H_2 temperature programmed reduction (H_2 TPR), and diffuse reflectance UV-vis spectroscopy analyses will be used to determine the mechanism of core vanadium species preservation at high temperature and the type of core@shell structure capable of achieving maximal methane-to-HCHO conversion without undergoing deactivation. New catalysts will be compared with conventional mesoporous silica supported V_2O_5 , prepared by impregnation. The effect of the presence of CO_2 as well as using CO_2 as soft oxidant will be investigated.