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Flame synthesized vanadia catalysts for oxidative dehydrogenation of propane

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Flame spray pyrolysis (FSP) is a novel one-step method for continuous production of nanoparticles with well defined crystalline structure and high thermal stability[1]. FSP has been utilized for synthesis of supported noble metal catalysts as well as mixed metal oxides [2,3].

In this study a series of five alumina supported vanadia catalysts with 2, 3, 5, 7.5 and 10 wt.% nominal vanadium content have been synthesized by FSP. Aluminum(III)- and vanadium(III)acetylacetonate salts were dissolved in toluene and sprayed as fine droplets and ignited by a premixed methane-oxygen supporting flame. The high combustion enthalpies of the solutions resulted in high flame temperatures and atomically dispersed product vapors, leading to particle formation from the gas phase. The powder products, which were the calcined oxide catalysts, were collected on glass fiber filters.

The catalysts were characterized by BET, XRD, Raman and UV-vis spectroscopy. The specific surface areas were between 140 to 170 m²/g, resulting in apparent vanadia surface densities from 1.4 to 8.4 VO_x/nm². Raman (see Fig. 1) and UV-vis spectroscopy showed that supported vanadia monomers were the surface species at low loadings (2 and 3 wt. %) while vanadia oligomers were the main species at high loadings (7.5 and 10 wt.%) and a mixture of vanadia monomers and oligomers were formed at intermediate loadings (5 wt.%). Only traces of crystalline V₂O₅ were detected at 10 wt. %.

The vanadia catalysts were tested for oxidative dehydrogenation of propane (ODP). The catalysts were diluted with SiC for better mass and energy transport and loaded in fixed bed reactors. Depending on the vanadia loading between 150 and 75 mg of catalyst was used. The gas composition was C₃H₈/O₂/N₂ = 5/25/70, the total flow was between 50 and 260 Nml/min and temperature between 360 and 500 °C.

The major drawback of ODP is limited propene selectivity since both propane and propene are easily combusted to CO and CO₂. The propene selectivity depended highly on the type of vanadia species present on the catalyst (see Fig. 2). The 2 and 3 wt.% V catalysts, containing mainly vanadia monomers, showed superior selectivity compared to the 5 to 10 wt.% V catalysts containing vanadia oligomers. The propane conversion and propene yields could be interpreted in terms of a kinetic model using the ODP reaction, parallel propane combustion and sequential propene combustion [4]. This showed that particularly the direct combustion of propane was a limiting factor for the propene selectivity at high vanadia loadings.

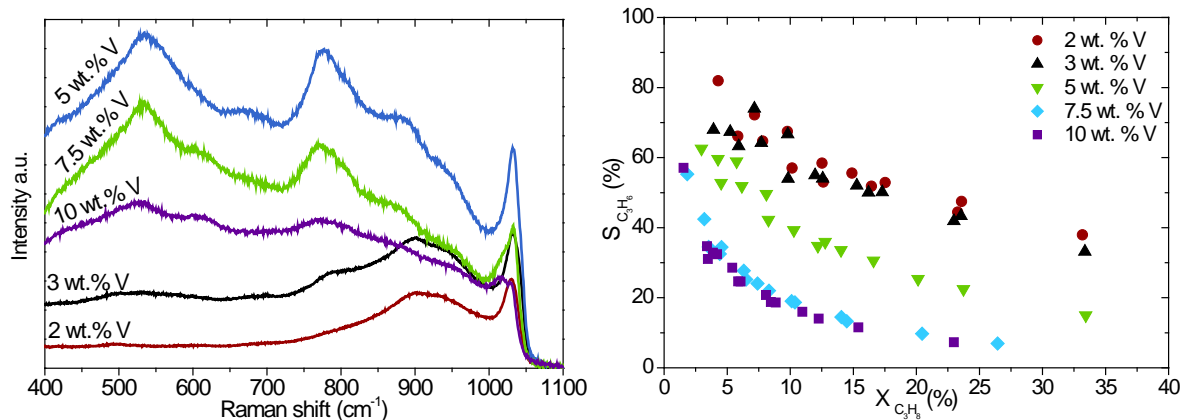


Figure 1. Raman spectra of the vanadia catalysts.

Figure 2. Propene selectivity as function of conversion.

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