Different Types of Diffusion of Carbon Dioxide in Samaria/Alumina Aerogel Catalyst by High Field Diffusion NMR

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Introduction

Mass transport of reactant and product molecules can play an important role in the overall performance of catalysts and is a multi-length scale process: the rate of diffusion inside individual catalyst particles can be quite different from that at macroscopic length scales comparable with the size of a particle bed. In this work pulsed field gradient (PFG) NMR was used to investigate the self-diffusion of carbon dioxide in alumina stabilized samaria aerogel catalyst, a promising porous catalyst for gas-phase reactions featuring high porosity and high surface area. For diffusion studies, the catalyst was prepared in two sample packing types, macroscopic monoliths (macroscopic cylindrical particles) and powder beds with particle sizes around 200 µm that are considered for catalytic applications.

Experimental

Samples of alumina stabilized samaria aerogels were formed in the laboratory of Prof. Bäumer following the epoxide addition sol-gel method. Diffusion data were obtained by ^{13}C PFG NMR using a wide-bore 17.6 T Bruker BioSpin spectrometer located at AMRIS, University of Florida. Magnetic field gradients with the amplitude of up to 30 T/m were generated using diff60 diffusion probe and Great60 gradient amplifier (Bruker BioSpin).

Results and Discussion

Fig. 1 shows the pore size distributions in the studied alumina stabilized samaria aerogel catalyst. Fig.2 presents the results of PFG NMR measurements of CO_2 diffusion in the catalyst samples as a function of the CO_2 loading pressure. The results show that at small CO_2 loading pressures there is a significant difference between the long-range diffusivity and the diffusivity inside individual catalyst particles (intraparticle diffusivity). This difference becomes smaller with the increasing loading pressure.

Conclusions

PFG NMR was applied to investigate different types of gas diffusion in the samaria/alumina aerogel catalyst.

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References

[1] Mueller, R. *et al*., J. Chem. Phys.**139**, 154703 (2013).

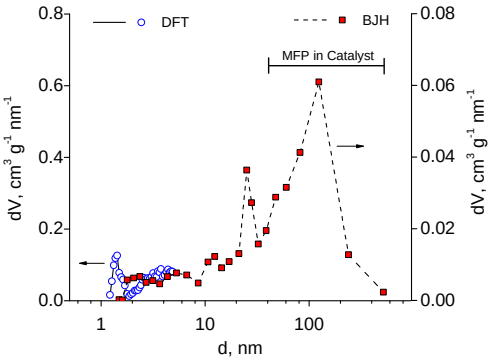

Fig.1 Pore size distributions for the studied alumina stabilized samaria aerogel. The distributions were obtained from N_2 adsorption isotherms measured at 77 K by applying density functional theory (open circles, left-axis) and Barrett-Joyner-Halenda (filled squares, right-axis) methods.

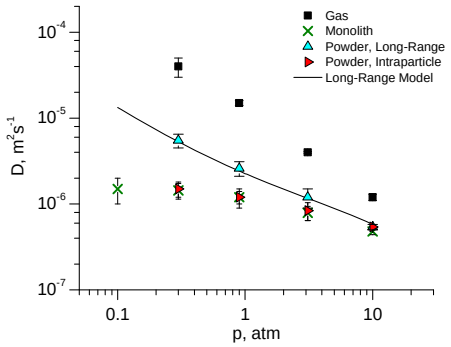

Fig.2 CO_2 self-diffusivities in the alumina stabilized samaria aerogel catalyst at 297 K for different diffusion types. The diffusivities are shown as a function of the CO_2 loading pressure in the samples. Also shown for comparison are the gas-phase self-diffusivities of CO_2.