8th European Congress of Chemical Engineering, 25-29 September 2011, Berlin (Germany)

Magnesium Oxide as Model Catalyst for the OCM Reaction

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Introduction

The influence of the morphology of the primary MgO particle as well as the type and the concentration of surface defects (like steps, corners and kinks) on the reactivity of the oxidative coupling of methane (OCM) reaction were studied in this work.

Experimental

MgO with different primary particle morphology has been prepared according to 5 different preparation methods: precipitation of $Mg(NO)_3$ with NaOH (P-MgO) and Na_2CO_3 (B-MgO), sol-gel synthesis (SG-MgO), hydrothermal post treatment of MgO in a microwave autoclave (HT-MgO) and smoke MgO (S-MgO). These materials were compared with ultra pure MgO (99.998% m. b., Alfa Aesar, C-MgO). The precursors were calcinated at 850 ℃ for 3 hours, h eating rate 5 ℃/min, in a flow of 20 % O_2 /80 % Ar. The catalytic activity of the catalysts in the OCM reaction has been tested at 750 ℃ and a contact time of 0.05 g.s/ml in a plug flow reactor set up equipped with GC analytics applying a feed composition ($CH_4/O_2/N_2$) of 4/1/4.

Results and discussion

The TEM pictures of the catalysts prepared by different synthesis methods show different morphology from perfect cubic for S-MgO to highly defected structures for HT-MgO or SG-MgO. The size of the particles is varying from ca. 100 nm to ca. 10 nm. Analysis of the used catalysts with TEM show that the structure is modified during OCM reaction probably due to sintering of the primary particle to larger rounded one. The first catalytic results show different conversion for the 6 samples, but also different C_2 selectivity for the same methane conversion in steady state. Further analyses like photoluminescence, UV-Vis and IR spectroscopy of adsorbed CO of the fresh and used catalysts are in progress in order to understand the catalytic reactivity of the OCM reaction with MgO and especially to understand the role played by the low coordinated ions present on the catalyst surface and the morphology of the primary particle.