

Synthesis of transition metal molybdates and tungstates and their reactivity in alkane oxidation

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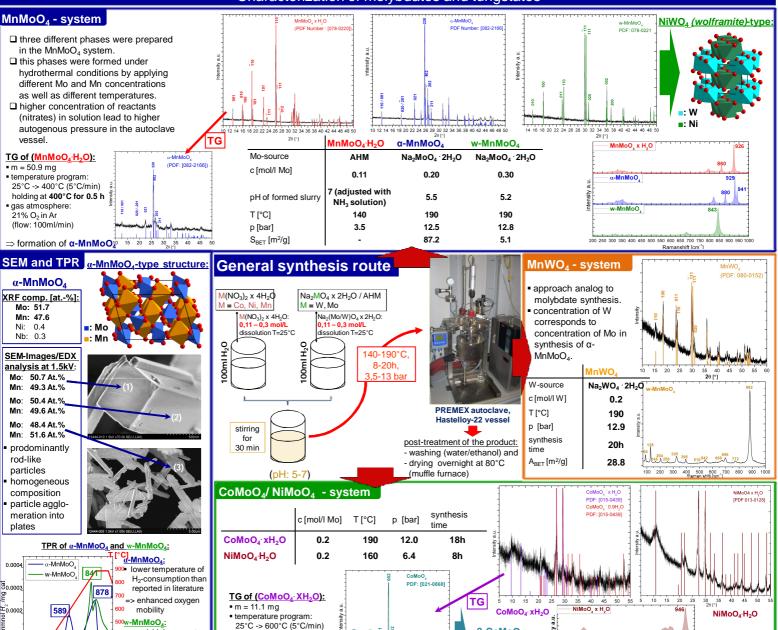
Introduction

Ni-Co molybdates have been reported to show high selectivities for propene in the oxidative dehydrogenation of propane [1, 2]. Furthermore transition metal molybdates of the type (Mn,Co,Ni)MoO₄ were also investigated in the selective oxidation of propane to acrylic acid and acrolein [3] and in the ODH of isobutane.[4] With the addition of Mn, sodium tungstate, which is structurally related to the corresponding molybdate, also showed considerable activity in the oxidative coupling of methane [5]. In this work, we have synthesized a series of phase-pure MMoO₄ (M = Co, Ni, Mn) catalysts and a corresponding MnWO₄ catalyst via a simple hydrothermal route [6]. Systematic investigations of the synthesized phases were performed by means of selective oxidation of propane, ethane and the oxidative coupling of methane. First results of this work are presented here.

Conclusion

- a simple hydrothermal synthesis route to transition metal molybdates and tungstates is presented here.
- ❖ high thermal stabilities, relatively high specific surface areas, and partially increased oxygen mobility (concluded from TPR measurements) of anhydrous synthesized samples suggest the applicability of hydrothermally synthesized molybdates and tungstates as catalysts in selective oxidation reactions of alkanes and alkenes.

Characterization of molybdates and tungstates



25 30 35 40 20 [°]

20

Propane oxidation

much higher onset

temperature of H₂-

> possible catalyst

for OCM reactions

consumption

pretreatment:

air for 1h

6000

total flow

700°C in synthetic

catalytic testing:

W/F [ml/gh]:

• feed: 10/5/85

30 mln/min

C₃H₈/O₂/He

α-MnMoO₄ S(C,H,) S(CH_.) 0,8 0,8 <u>≩</u> €0,6 **≨**0,6 ^೫0,4 克0,4 0.2 0,0 0,0 550 570 590 610 T [°C] 550 570 590

holding at 600°C for 0.5 h

gas atmosphere:

(flow: 100ml/min)

References

500 600 700 800 Raman shift [cm⁻¹]

- [1] Y. -S. Yoon, N. Fujikawa, W. Ueda, Y. Moro-oka, K. -W. Lee; Catalysis Today 24 (1995), 327-333,
- [2] D. L. Stern, R. K. Grasselli; Journal of Catalysis 167 (1997), 550-559.
- [3] Fujikawa, N., K. Wakui, et al. (2001); Catalysis Today 71(1-2): 83-88.

β-CoMoO₄:

α-MnMoO₄-

structure

- [4] Agafonov, Y., N. Nekrasov, et al. (2009); Kinetics and Catalysis 50(4): 577-582. [5] Z. Gholipour, A. Malekzadeh, R. Hatami, Y. Mortazavi, A. Khodadadi; Journal of
- Natural Gas Chemistry 19 (2010), 35-42 [6] W. Xiao, J. S. Chen, C. M. Li, R. Xu, X. W. Lou; Chem. Matter. 22 (2010) 746-