Accepted Manuscript

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PII: S0920-5861(17)30539-4

DOI: http://dx.doi.org/doi:10.1016/j.cattod.2017.08.019

Reference: CATTOD 10962

To appear in: Catalysis Today

Received date: 31-5-2017 Revised date: 4-8-2017 Accepted date: 7-8-2017

Please cite this article as: Samira Parisha, Patrick Littlewood, Aleks Arinchtein, Vinzenz Fleischer, Reinhard Schomäcker, Chemical looping as a reactor concept for the oxidative coupling of methane over the MnxOy-Na2WO4/SiO2 catalyst, benefits and limitation, Catalysis Todayhttp://dx.doi.org/10.1016/j.cattod.2017.08.019

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ACCEPTED MANUSCRIPT

Chemical looping as a reactor concept for the oxidative coupling of methane over the Mn_xO_y-Na₂WO₄/SiO₂ catalyst, benefits and limitation

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Highlights

- The secondary oxidation of C₂ products is successfully suppressed in CLR
- The high temperature needed for OCM on Mn-Na₂WO₄/SiO₂ leads to H₂ by TDH
- H₂ formed through the TDH is responsible for the yield limitation

Abstract

The chemical looping concept (CLC) has been implemented to suppress the formation of COx in an oxidative coupling of methane (OCM) reactor under a wide range of conditions. In comparison to the normal co-feeding strategy, this technique resulted in an enhanced C2 selectivity at the same methane conversions. Nevertheless, the obtained yield never exceeded 25 % which is still lower than the minimum value needed for industrializing OCM. The CLC was applied in mechanistic studies to investigate the consecutive reaction of the main products of OCM, i.e. ethane and ethene. The performance of the reaction of C2 components at 750°C in the chemical looping reactor was compared with that obtained in the co-feeding experiments. The effect of the surface adsorbed oxygen species on the reaction of both ethane and ethene was investigated. The results of these experiments reveal that some of the mechanistic assumptions about the OCM reaction are not compatible with the nature of Mn_xO_y-Na₂WO₄/SiO₂, one of the most stable and best performing catalysts known for this reaction. The yield limitation is shown to be inherent to the catalyst. However, this limitation should be solvable through the modified process concept aiming at the production of ethane in a separated first step.

Keywords: Oxidative coupling of methane; Mn_xO_y-Na₂WO₄/SiO₂; Chemical looping.

Introduction

Ethene (C_2H_4) is the building block for producing a vast range of chemicals from plastics to antifreeze and solvents. It is among the most produced organic compounds of the petrochemical industry. The market demand of ethene was more than 150 million tons in 2016 and its global growth rate is presumed to be around 3.5 % over the next 5 years [1]. This chemical is usually produced in steam-cracking units from a range of petroleum-based feedstocks, such as naphtha, and therefore its production capacity and

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