



Experimental study of the deactivation of Ni/AlMCM-41 catalyst in the direct conversion of ethene to propene

L. Alvarado Perea^{a,*}, T. Wolff^b, C. Hamel^{c,d}, A. Seidel-Morgenstern^{b,d}

^a Unidad Académica de Ciencias Químicas y Posgrado en Ciencias de la Ingeniería, Universidad Autónoma de Zacatecas, Carretera Zacatecas-Guadalajara km 6, Ejido La Escondida, 98160 Zacatecas, Mexico

^b Max Planck Institute for Dynamics of Complex Technical Systems, Sandtorstr. 1, 39106 Magdeburg, Germany

^c Anhalt University of Applied Sciences, Bernburgerstr. 55, 06366 Köthen, Germany

^d Otto Von Guericke University, Institute of Process Engineering, 39106 Magdeburg, Germany



ARTICLE INFO

Article history:

Received 14 September 2016

Received in revised form

24 December 2016

Accepted 27 December 2016

Available online 31 December 2016

Keywords:

ETP-reaction

Propene

Deactivation

Ni/AlMCM-41 catalyst

Filamentous carbon

Regeneration

ABSTRACT

In this work, an experimental study of the deactivation and regarding a possible regeneration procedure in the ETP-reaction using Ni/AlMCM-41 (Si/Al = 60) was carried out. ETP experiments at different temperatures and long time *on*-stream were carried out. Characterization of the catalyst after experiments was performed using powder-XRD, N₂-physisorption, NH₃-TPD, TPO and TEM. At 250 and 350 °C the catalyst did not suffer significant deactivation after 107 h *on*-stream. Nevertheless, the yield of propene was low. A severe deactivation of the catalyst was observed at 450 °C where propene was the main reaction product. Regeneration of the catalyst after 30 h *on*-stream at temperatures lower than 350 °C could be performed using a mixture of 5 vol.% oxygen in N₂. The regeneration of the catalyst was not possible after 30 h *on*-stream at 450 °C. Filamentous carbon and Ni reduction are considered responsible of the deactivation of the catalyst at 450 °C. The analysis of the obtained results suggests that a conjunct polymerization process takes place and is the precursor of the reaction products and the carbon species. However, a detailed study of the mechanism of the ETP-reaction should be done for a better understanding of the deactivation of Ni/AlMCM-41.

© 2017 Elsevier B.V. All rights reserved.

1. Introduction

Propene is one of the main block-buildings in the chemical industry and traditional sources are not sufficient to provide its present worldwide increasing demand [1,2]. For this reason, and the shifting to lighter feedstocks in steam cracker units from the low-cost natural shale gas, have motivated the development of the so called *on*-purpose technologies [1,3]. Inside of these technologies, it is possible to find the dehydrogenation of propane [3,4], the catalytic cracking of butenes to propene [5,6], the metathesis of ethene and 2-butene [5,7] and the direct conversion of ethene to propene [8–18]. Recent results have shown that the direct conversion of ethene to propene (ETP-reaction) represents one of the most attractive alternatives to face the increasing demand of propene [9,11,12,14,17–20]. In this reaction, propene it is believed that is produced based on the dimerization of ethene to 2-butene and its metathesis with another molecule of ethene to produce propene

[9]. However, the catalyst used underwent severe deactivation where the highest production of propene was observed [14].

In this sense, some of the most active catalysts in this reaction are based on Ni ion loaded on MCM-41 and MCM-48 and Ni on zeolites of the type H-ZSM-5 and SAPO-34 [9,12,14,17,18,21]. More recently, it has been shown that Ni doped MCM-41 and Ni doped FSM-16, Y modified ceria, and Sc on In₂O₃, are attractive candidates for the direct conversion of *bio*-ethanol to propene [22–24]. In this last process, ethanol is dehydrated to ethene which is transformed in a second step to propene. These catalysts open an attractive alternative to produce propene from renewable raw materials.

On the other hand, deactivation of catalysts represents an important issue in many processes of great economical interest [25]. This deactivation is influenced by different factors during a chemical reaction, which include poisoning of the catalyst, formation of deposits, thermal degradation, mechanical damage and corrosion or leaching by the reaction mixture [25]. Therefore, catalyst deactivation is highly relevant in the operation of catalytic processes and represents an issue with many scientific and technological challenges [25–41]. Several options to decrease the rate of deactivation or to restore the catalytic activity of the catalyst can

* Corresponding author.

E-mail address: leoap@uaz.edu.mx (L.A. Perea).