Experimental Investigation of the Reaction Network of Ethene to Propene over Ni/AlMCM-41 Catalysts

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Dedicated to Prof. Dr. Wilhelm Schwieger on the occasion of his 65th birthday

The reaction network of the ethene-to-propene (ETP) reaction over Ni/AlMCM-41 catalysts was studied. This reaction consists of the dimerization of ethene, the isomerization of 1-butene, and the metathesis of ethene and 2-butene to propene. This work focused on the latter. ETP experiments followed by in situ diffuse reflectance infrared Fourier transform spectroscopy were carried out. Metathesis activities were evaluated by the isomerization of butenes, the metathesis of ethene and 2-butene, and the retro-metathesis of propene. Metathesis activities of Ni/AlMCM-41 catalysts were not observed. Thus, a conjunct polymerization of ethene to propene is proposed.

Keywords: Ethene, Metathesis, Ni/AlMCM-41 catalysts, Propene, Reaction network

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1 Introduction

Nowadays, the petrochemical industry is facing a major squeeze regarding propene availability as a result of both the strong global demand of propene derivatives and the shifting to lighter feedstocks in steam cracker units from low cost natural shale gas [1 – 3]. Propene is used as raw material for the production of a broad spectrum of chemical products, e.g., polypropene, propene oxide, acrylonitrile, cumene, and acrylic acid [3]. Therefore, many efforts have been directed to develop new alternatives for propene production, the so-called on-purpose technologies [3 – 14]. Amongst these technologies, the direct conversion of ethene to propene (ETP) represents one of the most attractive alternatives [8 – 14]. In this reaction, Ni doped on MCM-41 materials has shown high catalytic activity [8, 12]. However, these materials underwent an important deactivation at the reaction conditions where the highest production of propene was observed [12, 15].

Ni-based catalyst systems constitute one of the most important catalysts for olefin dimerization [16]. The specific activity of Ni is high, and it is one of the less expensive transition elements. Because of these advantages, Ni-catalyzed dimerization and oligomerization have been extensively studied [16 – 26]. In the case of heterogeneous catalysts, Hartmann et al. showed that the dimerization of ethene and the isomerization of 1-butene can proceed over Ni/AlMCM-41 [18]. Hulea and Fajula performed the oligomerization of ethene over Ni/AlMCM-41 and found that these catalysts show high activity for this reaction [25]. The presence of uniform pore-size distribution is favorable for the oligomerization process, in which only the formation of hydrocarbons with even carbon numbers occured. In both cases, the presence of both metal and acid sites is required for the dimerization and oligomerization of ethene.

On the other hand, olefin metathesis is one of the very few fundamentally novel organic reactions discovered in the last 50 years. This reaction was discovered by Banks and Bailey in 1964 [27]. Since then, there is a great interest in industrial applications of olefin metathesis for the synthesis of various chemicals. For the metathesis of ethene and

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