



A novel CeO₂ supported on carbon nanotubes coated with SiO₂ catalyst for catalytic cracking of naphtha

Kamyar Keyvanloo^{a,*}, Ali Mohamadalizadeh^b, Jafar Towfighi^a

^a Chemical Engineering Department, Tarbiat Modares University, Tehran, 14115-143, Iran

^b Gas Research Division, Research Institute of Petroleum Industry, Tehran, 14665-1998, Iran

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ABSTRACT

Carbon nanotubes (CNTs) supported ceria catalysts coated with silica were used in catalytic cracking of naphtha to increase the yield of ethylene and propylene. Coating of CNTs was carried out with a pH control chemical deposition method using Si precursor. TGA results on coated CNTs showed 80 °C improvement in the thermal stability compared to uncoated CNT. XRD, SEM, and TEM analysis indicated that the structure of the CNT remained intact after modification. The silicon partly coated the CNT walls and the CeO₂ entered the CNT channel or was deposited on the wall. Measurement of the total acidity and number of Brønsted acid sites with NH₃-TPD and FTIR measurements exhibited that the total acid concentration and Brønsted acid sites of modified CNTs increased. TPR results showed that the presence of Si species slightly improved the reduction of CeO₂. As a result, catalyst Si–Ce/CNT showed a better catalytic cracking performance than catalysts Ce/CNT, Si/CNT or CNT.

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1. Introduction

Light olefins, such as ethylene and propylene, are very important building blocks of the petrochemical industry. They are mostly produced by steam cracking of naphtha at high temperatures. There is an increasing demand for light olefins which highlights development of alternative processes that would require less demanding reactor conditions and higher yield of olefins. Among these processes, steam catalytic cracking showed promising. Catalytic cracking of hydrocarbons over zeolites appears attractive since zeolites facilitate transformation of long paraffins to short olefins via their acidic sites [1–6].

Acidic zeolites such as HZSM-5 are potential catalysts for cracking of hydrocarbons. However, these solid catalysts suffer from relatively slow heat transfer, which consequently limits the mass transfer [7]. The other disadvantage of zeolite-based catalysts is that they tend to be deactivated due to coke formation and dealumination in high temperature and humidity [3,8,9].

Carboneous materials such as activated carbon have also been used to improve catalytic cracking due to their large surface area and high pore volume, low cost, resistance to high temperature and availability [10–14]. The influence of carbon active sites on the pyrolysis of propylene has been studied [10,11]. The results showed

that the carbon active surface greatly enhanced the rate of cracking of hydrocarbons to elemental carbon which was deposited directly on the surface and coke has been an important product for most of the samples. However, carbon supported catalysts have been less prone to coke deposition than Al₂O₃ supports [11,12]. Researchers have shown that an activated carbon supported Ni–Mo catalyst can be able to maintain high activities over longer periods of operation for the hydrogenation of saturated hydrocarbons such as ethylene [13]. Carbon nanotubes are another carboneous material which has been widely used as a catalyst support [15–17]. Carbon nanotubes exhibit mechanical strength as well as electrical and thermal properties which render them useful for several applications [18–21]. The high aspect length to diameter, high external surface area and the mesoporous structure of them significantly reduces mass transfer limitations and allows high catalytic performance compared to some traditional supports like activated carbon supports which have large number of micropores with high diffusion resistance [22]. Furthermore, the tubular morphology of the carbon nanotubes accelerates the occurrence of a confinement effect and increases the reactivity of the gaseous and liquid reactants [23]. Thermal resistance of carbon nanotubes has been increased by coating with more resistant materials which make them capable for reactions requiring higher temperatures [24–26].

In this study, for the first time, we tune carbon nanotubes properties in order to be used in thermal catalytic cracking of naphtha for production of light olefins of a high operating temperature processes. With this aim, CeO₂ supported on a multiwall carbon

* Corresponding author.

E-mail address: kamyar.keyvanloo@yahoo.com (K. Keyvanloo).