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Enhanced catalytic activity of the Rh/ γ -Al₂O₃ pellet catalyst for N₂O decomposition using high Rh dispersion induced by citric acid

Min-Jae Kim^{a,b}, Young-Ji Kim^c, Seung-Jae Lee^b, In-Soo Ryu^b,
Hye Jeong Kim^b, Yeonjin Kim^b, Chang Hyun Ko^{c,*}, Sang Goo Jeon^{b,*}

^a Department of Chemical and Biological Engineering, Korea University, Anam-ro 145, Seongbuk-gu, Seoul, Republic of Korea

^b Korea Institute of Energy Research, 152 Gajeong-ro, Yuseong-gu, Daejeon 34129, Republic of Korea

^c School of Chemical Engineering, Chonnam National University, 77, Yongbong-ro, Buk-gu, Gwangju 61186, Republic of Korea

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ABSTRACT

Two types of egg-shell-type Rh-based spherical γ -alumina pellets were prepared by simple impregnation and incipient wetness methods with citric acid as an additive. In the case of the simple impregnation method, most of the Rh precursor molecules were selectively located in the outermost region of the spherical γ -alumina pellet because of strong interactions between the Rh precursor and alumina. Thus, it formed a very thin Rh shell (Rh/Al₂O₃-Thin) around the pellet. On the other hand, the Rh precursor molecules were widely dispersed in the outer part of the spherical γ -alumina pellet when citric acid was used as an additive. This method led to the formation of a thick Rh shell (Rh/Al₂O₃-Thick). The two samples were characterized via N₂ adsorption isotherms and CO chemisorption, and used as catalysts for the decomposition of N₂O under different reaction conditions and at varying gas hourly space velocities. The N₂O conversion achieved using the Rh/Al₂O₃-Thick catalyst was higher than that attained with the Rh/Al₂O₃-Thin catalyst. This implied that Rh dispersion is an important factor governing the performance of Rh-based spherical γ -alumina pellet catalysts in N₂O decomposition.

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1. Introduction

It is very important to reduce the amount of nitrous oxide (N₂O) present in the atmosphere because it contributes to the destruction of stratospheric ozone and the greenhouse effect (Kapteijn et al., 1996; Konsolakis, 2015). Several researchers have investigated methods for controlling N₂O emissions including thermal decomposition, selective catalytic reduction (SCR), and catalytic decomposition of N₂O (Centi et al., 2000; Kapteijn et al., 1996; Shimizu et al., 2000). Among these methods, catalytic decomposition of N₂O (2N₂O → 2N₂ + O₂) has received significant attention from both an academic and industrial point of view (Kapteijn et al., 1996; Konsolakis, 2015). This is because

catalytic decomposition of N₂O offers a simple and efficient way of removing N₂O without using any reductant such as ammonia, hydrocarbon, and carbon monoxide (Centi et al., 2000; Kapteijn et al., 1996; Konsolakis, 2015; Shimizu et al., 2000). However, none of the catalysts proposed in literature exhibit a good performance and stability under space velocity and feed composition conditions which are realistic for industrial applications. Therefore, several different types of catalysts such as noble metals (Bueno-López et al., 2006; Centi et al., 2000; Yuzaki et al., 1997), ion-exchanged zeolites (Li and Armor, 1992; Melián-Cabrera et al., 2005), and metal oxides (Abu-Zied and Soliman, 2009; Karásková et al., 2010; Pasha et al., 2008) have been investigated for efficient N₂O decomposition. Among these various types, rhodium

* Corresponding authors.

E-mail addresses: chko@jnu.ac.kr (C.H. Ko), sgjeon@kier.re.kr (S.G. Jeon).

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