Highly efficient removal of ethylene at 0 °C over Au-Pt/zeolite

([1]Tokyo Metropolitan University, [2]Yantai University)
○Mingyue Lin[*1], Haifeng Wang[*1], Tetsuya Shishido[*1], Hiroki Miura[*1], Masatake Haruta[*1], Toru Murayama[*1,2]

1. Introduction

Ethylene (C_2H_4) released from the plants could fasten the mature and deterioration of fresh fruits and vegetables even at low temperatures since it is a natural gaseous plant hormone. To prolong the storage time of the fresh plants during the transportation, removing trace amounts of C_2H_4 is important, and selective catalytic oxidation of C_2H_4 to CO_2 is an ideal method.[1] However, almost all of the reported catalysts would be deactivated due to the water adsorption, usually within 1 h. Therefore, it is necessary to develop a more effective material with a high C_2H_4 removal efficiency and long-term stability at low temperatures (0~5 °C).

Here, we report an Au-Pt/zeolite (ZHM20) with a highly active C_2H_4 removal efficiency (81%) and long-term stability (40 h) for C_2H_4 elimination at 0 °C.

2. Experimental

Mordenite 20 (ZHM20, SiO_2/Al_2O_3=18.3) provided by the Catalysis Society of Japan was used as the support. The loading amount of nanoparticles was prepared as 1wt%. Sol immobilization method[2] was applied to prepare ZHM20, Pt/ZHM20, Au/ZHM20, and Au_xPt_y/ZHM20 (x and y represent the molar ratio of Au and Pt, respectively) and they were calcined at 500 °C for 2 h before being used.

A fixed-bed flow reactor was used to conduct the C_2H_4 removal test. The sample (0.2 g) was firstly pretreated at 150 °C for 2 h under N_2 with a flow rate of 50 mL min^{-1} before being evaluated under the reactant gas that contained 50 ppm C_2H_4, 20% O_2, and N_2 balance with a total flow rate of 10 mL min^{-1}. The inlet and outlet concentrations of C_2H_4 and CO_2 were analyzed by an online 490 Micro GC system (Agilent) for calculating the removal efficiency of C_2H_4 and the yield of CO_2.

3. Results and Discussion

As shown in Figure 1, the initial C_2H_4 removal efficiency over ZHM20 was 100% at 0 °C and it reached the maximum adsorption capacity after flowing 11 h of 50 ppm C_2H_4. The removal efficiency curves of Pt/ZHM20 and Au/ZHM20 were U-shaped. Initially, the removal efficiency was 100% and it decreased to 19% and 15% after 10 h-on-stream over Pt/ZHM20 and Au/ZHM20, respectively, which was the same with that observed from ZHM20, suggesting that this step might be owing to the adsorption of C_2H_4 on ZHM20. Then, the C_2H_4 removal efficiency increased until reaching the steady-state of 56% on Pt/ZHM20 and 45% on Au/ZHM20. $Au_{54}Pt_{46}$/ZHM20 also showed a U-shape removal efficiency curve, but the turning point time of 3.5 h was much shorter than the other two. Moreover, the C_2H_4 removal efficiency of $Au_{54}Pt_{46}$/ZHM20 at the steady-state was 81% and it lasted for as long as 40 h. According to the electronic state analysis, $Au_{54}Pt_{46}$/ZHM20 possessed electron-deficient Pt species and electron-rich Au species, which might be helpful for the faster adsorption and transformation of C_2H_4 on $Au_{54}Pt_{46}$ NPs than that on Au NPs and Pt NPs.

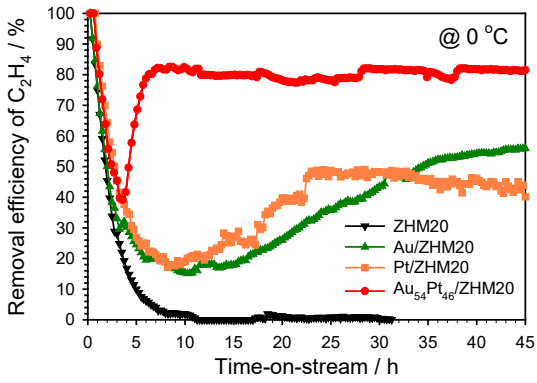

Figure 1. The removal efficiency of C_2H_4 with time-on-stream over ZHM20, Au/ZHM20, Pt/ZHM20, and $Au_{54}Pt_{46}$/ZHM20 at 0 °C.

1) C. Jiang, K. Hara, A. Fukuoka, Angew. Chem. Int. Ed., 52(24), 6265 (2013)
2) H. Miura, Y. Tanaka, K. Nakahara, Y. Hachiya, K. Endo, T. Shishido, *Angew. Chem. Int. Ed.*, 130(21), 6244 (2018)

