Catalytic combustion of acrylonitrile over 3d-transition metals (Cu, Co, Fe) or Pt/SBA-15, Cu/SBA-16 and Cu/KIT-6 mesoporous catalysts

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Introduction

Nitrile gases such as acrylonitrile (C_2H_3CN), acetonitrile (CH_3CN), and hydrocyanic acid (HCN) are hazardous properties, which are commonly classified as volatile organic compounds (VOCs). If the nitrile waste gases are not strongly demanded to prevent emission into the atmosphere, they can lead to seriously environmental problems and affect human beings. The efficiency of removal of them by either incineration or catalytic combustion becomes essential. The relatively lower operating temperature for catalytic combustion associated with a less NO_x formation makes this technology especially suitable for gaseous nitrile elimination. SBA-15 supported transition or noble metals catalysts were applied to the catalytic combustion of acetonitrile (CH_3CN) and four relevant kinds of mechanism were proposed[1]. Cu/SBA-15 exhibited a nearly complete CH_3CN conversion associated with a N_2 selectivity of around 80 % T > 350 ^oC. Although hydrocyanic acid (HCN), acetonitrile (CH_3CN) and acrylonitrile (C_2H_3CN) can be assigned to nitrile material, they have different structures and chemical properties. Hence, to detailedly investigate the system of nitrile gases catalytic combustion, expand research should be conducted to other nitrile gases over metals/SBA-15. The different types of ordered mesoporous materials, such as MCM-41, SBA-15, SBA-16, and KIT-6, have different space structures, surface areas, pore volumes and pore size distributions. Scare data related to the character of mesoporous support on the copper particles dispersion, reducibility and catalytic behaviour of C_2H_3CN have been reported. Whether the mechanisms for the C_2H_3CN over the metal/SBA-15 conform to the four kinds of reaction mechanisms for CH_3CN catalytic combustion we have proposed needs to be further proved.

Materials and Methods

A series of SBA-15 with different metals (Cu, Co, Fe, and Pt) and the copper loading different mesoporous zeolites (SBA-15, SBA-16, and KIT-6) were used for the catalytic combustion of C_2H_3CN. Meanwhile, the activity and selectivity for the three kinds of nitrile gases (C_2H_3CN, CH_3CN, HCN) over Cu/SBA-15 were contrasted under the similar condition. The catalysts were prepared and characterized by XRD, N_2 adsorption, TEM, H_2-TPR, XPS. Moreover, an attempt to verify the related combustion mechanism has been done based on the diffuse reflectance infrared Fourier transform spectra (DRIFTS) studies.

Results and Discussion

As presented in **Figure 1**, C_2H_3CN conversions achieving over the investigated catalysts follow a trend of Pt/ = Co/ ≈ Cu/ > Fe/SBA-15 at 400 ^oC, however, the yield of mainly desired product N_2 follow the trend of Cu/ > Fe/ > Pt/ > Co/SBA-15 at 400 ^oC, being correlated well with the redox abilities, metallic state and the chemical nature of the loaded metal species. The conversion of C_2H_3CN and the yield of N_2 were sequentially followed by the Cu/SBA-15 > Cu/SBA-16 > Cu/KIT-6 at above 350 ^oC, due to the straight cylindrical pores with 2-D arrangement of SBA-15 are beneficial not only to homogeneous distribution of the loaded copper along the pore surface, but also to the formation of highly dispersed Cu^{2+} ions.

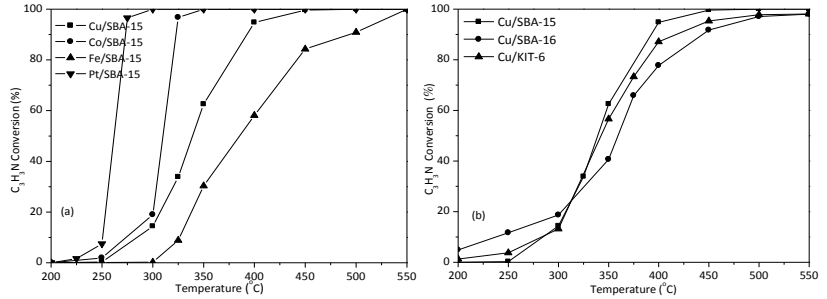

Figure1. Catalytic performance as a function of temperature during C_3H_3N combustion: (a) (Cu, Co, Fe, Pt)/SBA-15; (b) Cu/ (SBA-15, SBA-15, KIT-6)

As presented in **Figure 2**, the -NCO (2198 cm^{-1}) being the most intermediate over Cu/SBA-15 can be directly oxidized to N_2 and CO_2 under the oxygen-rich condition. However, the CN band (2237 cm^{-1}) of C_2H_3CN is able to be hydrolyzed into acylamino over Fe/SBA-15, leading to an enhancement in the related bands (1571, 1659 cm^{-1}).

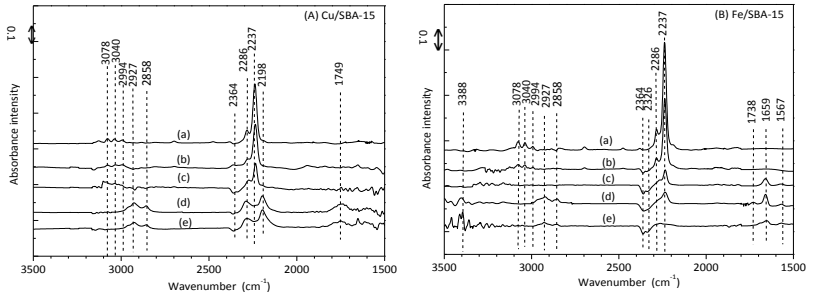

Figure 2. DRIFTS of adsorbates produced from the flow of C_2H_3CN (0.3 vol %) + O_2 (8 vol%) + He (91.7 vol%) for 25 min: (a) Cu/SBA-15; (b) Fe/SBA-15

Significance

Acrylonitrile was efficiently removed over Cu/SBA-15 with high conversion and high yield of N_2. Meanwhile, the C_2H_3CN combustion mechanisms separately complied with the "N_2 formation" mechanism over Cu/SBA-15 and the "NH_3 formation" mechanism over Fe/SBA-15.

References

1. Runduo, Z.; Dongjun, S.; Ning, L.; Biaohua, C. Applied Catalysis B: Environmental, 2014, 146, 79-93.