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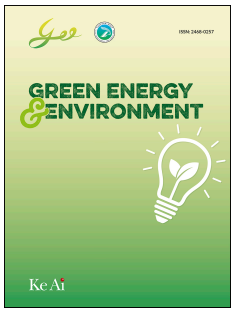

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Highly Efficient Cobalt-doped Carbon Nitride Polymers for Solvent-Free Selective Oxidation of Cyclohexane

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Abstract: Selective oxidation of saturated hydrocarbons with molecular oxygen has been of great interest in catalysis, and the development of highly efficient catalysts for this process is a crucial challenge. A new kind of heterogeneous catalyst, cobalt-doped carbon nitride polymer (g-C_3N_4), was harnessed for the selective oxidation of cyclohexane. X-ray diffraction, Fourier transform infrared spectra and high resolution transmission electron microscope revealed that Co species were highly dispersed in g-C_3N_4 matrix and the characteristic structure of polymeric g-C_3N_4 can be retained after Co-doping, although Co-doping caused the incomplete polymerization to some extent. Ultraviolet–visible, Raman and X-ray photoelectron spectroscopy further proved the successful Co doping in g-C_3N_4 matrix as the form of Co(II)–N bonds. For the selective oxidation of cyclohexane, Co-doping can markedly promote the catalytic performance of g-C_3N_4 catalyst due to the synergistic effect of Co species and g-C_3N_4 hybrid. Furthermore, the content of Co largely affected the activity of Co-doped g-C_3N_4 catalysts, among which the catalyst with 9.0 wt% Co content exhibited the highest yield (9.0%) of cyclohexanone and cyclohexanol, as well as a high stability. Meanwhile, the reaction mechanism over Co-doped g-C_3N_4 catalysts was elaborated.

Keywords: Selective oxidation of cyclohexane; Oxygen oxidant; Carbon nitride; Co-doping

1. Introduction

C–H activation is always a spotlight for the development of chemical industry owing to the ubiquity of C–H bonds in organic molecules [1-3]. However, saturated hydrocarbons consist of only strong and localized single bonds, C–C and C–H bonds, so that they generally have no empty orbitals of low energy or filled orbitals of high