Effect of mesoporous MCo_2O_4 (M = Cu, Zn and Ni) spinel catalysts on catalytic combustion of methane

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Natural gas vehicles (NGVs) have been receiving considerable attention as effective ways to reduce emission of NOx, HCs and CO compared to diesel- and gasoline- powered vehicles. However, unburned methane emitted from NGVs has 21 times higher global warming potential than carbon dioxide so that it is regarded as a potent greenhouse gas. Thus, reducing the emission of unburned methane is necessary for expediting the use of NGVs [1].

Metal cobalt spinel oxides (MCo_2O_4; M = Cu, Ni, Zn, Mn, etc.) are obtained by the substitution of transition metal for cobalt in cobalt spinel oxide, leading to special magnetic and catalytic properties for the various industrial fields [2]. However, the previous study claimed that single metal oxide phase is formed from the thermal decomposition of MCo_2O_4 spinel phase above 400 °C; therefore, it requires the enhanced thermal stability [3]. Mesoporous metal oxides have been widely studied for various catalytic applications recently because they have special textural properties like well-defined mesopore and high surface area. In this study, we aimed at investigating the relationship between mesoporous and bulk MCo_2O_4 spinel catalysts and finding the optimized catalyst having superior catalytic performance for catalytic combustion of methane.

Mesoporous MCo_2O_4 spinel catalysts were prepared by nano-replication method using mesoporous silica KIT-6 template and the bulk ones were prepared by typical co-precipitation method. Both catalysts were applied to methane combustion reaction. N_2 adsorption-desorption, ICP-AES, TEM, XRD, XPS and EXAFS were utilized to investigate textural and structural property of all the catalysts.

Fig. 1 indicates light-off curves of methane combustion on all the catalysts. Provided that T_{90} set as the criterion of catalytic activity for methane combustion, descending order of activity is as follows: m-$CuCo_2O_4$ > m-$ZnCo_2O_4$ > m-$NiCo_2O_4$ > b-$NiCo_2O_4$ > b-$CuCo_2O_4$ > b-$ZnCo_2O_4$. Consequently, the activity of the meso catalysts are strikingly higher than that of the bulk ones.

Based on characterization, the combined N_2 adsorption-desorption, TEM and XRD results can clarify the excellent textural property and thermal stability of the meso samples such as high BET surface area, amount of MCo_2O_4 phase [4]. Therefore, such superior properties prove that the meso spinel catalysts reveal higher catalytic activity for methane combustion than bulk counterparts.

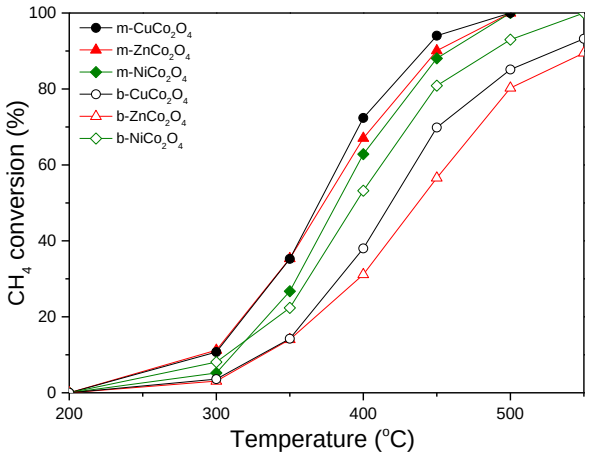

Fig. 1 Light-off curves of methane combustion on all the catalysts.

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