Applied Catalysis A: General 445–446 (2012) 121–127

Contents lists available at SciVerse ScienceDirect

Applied Catalysis A: General

journal homepage: www.elsevier.com/locate/apcata

Catalytic growth of high quality single-walled carbon nanotubes over a Fe/MgO catalyst derived from a precursor containing Feitknecht compound

Yi Jin, Gaowei Wang, Yongdan Li*

Tianjin Key Laboratory of Catalysis Science and Technology and State Key Laboratory for Chemical Engineering, School of Chemical Engineering, Tianjin University, Tianjin 300072, China

ARTICLE INFO

Article history:
Received 29 March 2012
Received in revised form 23 July 2012
Accepted 11 August 2012
Available online 20 August 2012

Keywords:
Methane catalytic decomposition
Single-walled carbon nanotubes
FC compound
High quality carbon nanotubes
Metal–support interaction

ABSTRACT

A catalyst is derived from a Feitknecht-compound (FC) precursor prepared via a co-precipitation method and is proved to manifest a strong metal–support interaction which facilitates the growth of high quality single-walled carbon nanotubes (SWCNTs) with a narrow diameter distribution. The effect of the rinsing condition of the precipitate on the growth of the SWCNTs is also examined. The pore structure of the catalysts rinsed by ethanol is modified and the carbon yield is enhanced while the diameter distribution of the SWCNTs is unchanged. The correlation between the metal–support interaction and the SWCNT growth is discussed.

1. Introduction

Single-walled carbon nanotubes (SWCNTs) have arose a great research interest due to their unique mechanical, electrical and thermal conductive properties [1,2]. The traditional SWCNT production methods include carbon-arc discharge [3], laser ablation [4] and chemical vapor deposition (CVD) [5–7]. CVD can be carried out continuously at a relatively low temperature and has been thought to be the most promising process for large-scale production [8]. However, mass production of the high quality SWCNTs is still a challenge which needs a rational catalyst design and reactor optimization.

Supported metal catalysts with Fe, Co and Ni as the active components have been widely used for the growth of the carbon nanotubes (CNTs) and carbon nanofibers [7,9–12]. Metal–support interaction (MSI) in the supported metal catalysts plays an important role in CNT growth. Generally, a strong MSI decreases the mobility of the metal particles on the support surface and enhances the metal dispersion [13]. The size of the metal nanoparticle has a direct effect on the number of walls and the diameter distribution of the CNTs. When the size of the metal nanoparticle is smaller than 5 nm, the main products are single and double-walled CNTs (DWCNTs), while larger metal nanoparticles lead to multi-walled CNTs (MWCNTs) [14]. Mattevi et al. [15] compared Fe catalysts supported on Al_2O_3 and SiO_2 and noted that a stronger MSI between Fe and Al_2O_3 restricts the Fe nanoparticle mobility and results in a much smaller Fe particle size. Ago et al. [16] found that after a thermal treatment in Ar, the iron nanoparticles become smaller and are appropriate for the SWCNT growth. They believed that Fe atoms diffuse into the MgO lattice during the heat treatment which results in a strong MSI. Ning et al. [17] found that a uniform $MgFe_2O_4$/MgO solid solution formed in the Fe/MgO catalyst after 900–1000 °C calcination also manifests a strong MSI. However, this high temperature treatment leads to a low surface area and also a low SWCNT yield. An ideal catalyst for SWCNT production should have a large specific surface area with a strong MSI.

Feitknecht compound (FC) has been recognized as a good precursor for many catalysts since 1970s [18–20]. The catalyst prepared via FC structured precursor enables a strong MSI in the reduced catalyst, and facilitates the formation of the mixed oxide with a high uniformity and large total metal surface area. Ni–Cu–Al catalysts derived from FC structure have been applied in the carbon nanofiber growth via methane catalytic decomposition reaction [21–26]. And also, Fe–Mg–Al, Co–Mg–Al and Ni–Mg–Al catalysts derived from FC structures were also employed as the catalysts for the SWCNT growth in a fluidized bed reactor [27]. However, the removal of Al_2O_3 support material from produced SWCNTs during the purification process is a laborious work.

Magnesium oxide supported iron catalyst (Fe/MgO) has been used as the catalyst for SWCNT production [13,16,17,28]. Fe/MgO catalyst is environmentally benign and attractive in the economic aspects. And also, Fe and MgO can be easily removed by acid,

* Corresponding author. Tel.: +86 22 27405613.
E-mail address: ydli@tju.edu.cn (Y. Li).

0926-860X/$ – see front matter
http://dx.doi.org/10.1016/j.apcata.2012.08.010