Chinese Chemical Letters Vol. 15, No. 2, pp 231 – 233, 2004
http://www.imm.ac.cn/journal/ccl.html

Preparation of Carbon Nanotubes with High Yield and Narrow Diameter Distribution from C_2H_2 over $LaCu_{0.2}Ni_{0.8}O_x$

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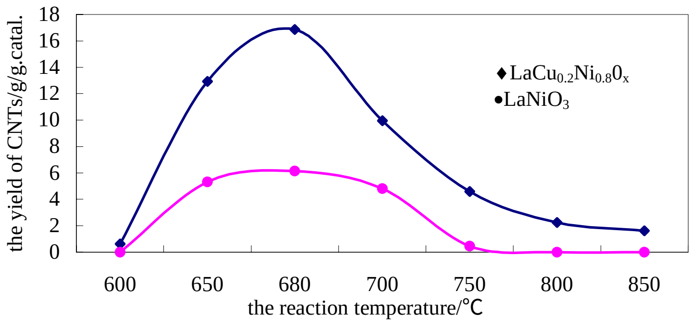
Abstract: Carbon nanotubes (CNTs) were prepared by decomposition of C_2H_2 over newly developed $LaCu_{0.2}Ni_{0.8}O_x$ in the temperature range from 600 to 850℃. The effect of the reaction temperature on the yield of CNTs was investigated in detail. At 680℃,the yield of CNTs reaches 17 g/g.catal. or so. The morphology of CNTs was examined by TEM. The diameter of CNTs rangs from 9 nm to 14 nm.

Key words: CNTs, C_2H_2, $LaCu_{0.2}Ni_{0.8}O_x$.

Research on the synthesis of CNTs has been carried out widely throughout the world, since the discovery of CNTs by Iijima[1] in 1991. So far, various methods such as arc-discharge, laser-vaporiation, chemical vapor deposition (CVD) method have been proposed for the synthesis of CNTs. From the viewpoint of application, high yield, high purity, diameter controllable and large-scale production are very important, so CVD method is becoming more and more popular in the last several years. To a large degree, the yield and tube diameter of CNTs are dependent on the nature and particle size of

Figure 1 The relationship between the yield of CNTs and the reaction temperature



C_2H_2 flow rate=10 mL/min, H_2 flow rate=30 mL/min, N_2 flow rate=130 mL/min, reaction time= 1 h.

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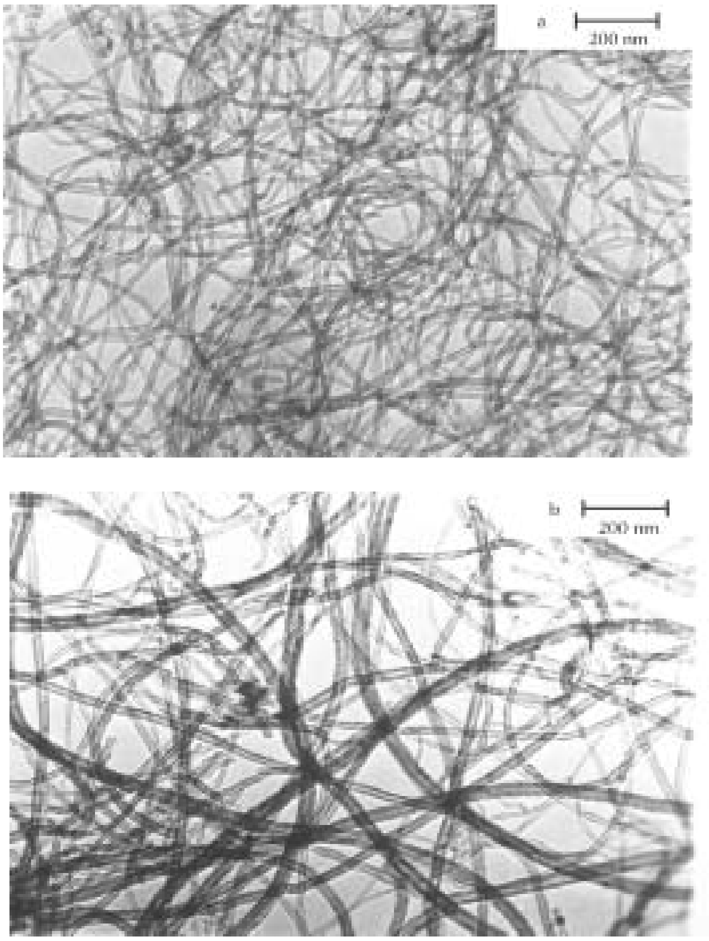
Figure 2 TEM images of CNTs prepared at 680℃ by (a) $LaCu_{0.2}Ni_{0.8}O_x$ (b) $LaNiO_3$



C_2H_2 flow rate=10 mL/min, H_2 flow rate=30 mL/min, N_2 flow rate=130 mL/min.

catalyst in CVD method[2]. In the present work, we describe a newly developed catalyst, $LaCu_{0.2}Ni_{0.8}O_x$ which is favorable for mass production of CNTs with high yield and even diameter by decomposition of C_2H_2.

$LaCu_{0.2}Ni_{0.8}O_x$ was prepared by the citric acid complexing method reported by B.C. Liu *et al.*[3]. The reaction was carried out in a quartz tube (35 mm in diameter,1400 mm in length). Reaction temperature was adjusted by ±1°C. 10 mg of the catalyst ($LaCu_{0.2}Ni_{0.8}O_x$) was loaded into a quartz boat lying in the central part of the quartz tubes, followed by heating in a flow of N_2. When the temperature rose to the desired temperature, a mixed gas of C_2H_2 (10 mL/min), H_2 (30 mL/min) and nitrogen (130 mL/min) as reaction mixture was introduced instead of N_2 for 1 h. After reaction, when the whole reaction system was cooled down to room temperature in the flow of N_2 automatically, fresh made CNTs were collected, then purified by diluted nitric acid and dried in the heating oven. The obtained CNTs were characterized by TEM.

The relationship between the reaction temperature and the yield of CNTs was investigated in detail. As shown in **Figure 1**, the yield of CNTs with catalyst $LaCu_{0.2}Ni_{0.8}O_x$ increases rapidly with rise of reaction temperature in the range 600 ～ 680℃. Above 680℃ , the yield of CNTs decreases with rise of reaction temperature. From **Figure 1**, it is clear that the yield of CNTs with catalyst $LaCu_{0.2}Ni_{0.8}O_x$ is much higher than that of CNTs with catalyst $LaNiO_3$ in the same reaction condition. The bimetallic catalyst may be more active in this reaction than monometallic one[4]. Above results show that $LaCu_{0.2}Ni_{0.8}O_x$ is a favorable catalyst for synthesis of CNTs.

The morphology of CNTs with catalysts $LaCu_{0.2}Ni_{0.8}O_x$ and $LaNiO_3$ at 680℃ was illustrated in **Figure 2**. According to **Figure 2 (a)**, CNTs prepared with catalyst $LaCu_{0.2}Ni_{0.8}O_x$ are all multi-walled and the diameter of CNTs rangs from 9 nm to 14 nm, while the diameter of CNTs prepared with catalyst $LaNiO_3$ is larger, ranging from 10 nm to 40 nm. $LaCu_{0.2}Ni_{0.8}O_x$ is favorable for the formation of CNTs with more uniform diameter. The reason of above results was proposed that the particle size of $LaCu_{0.2}Ni_{0.8}O_x$ is smaller than that of $LaNiO_3$.

In conclusion, CNTs with higher yield and more uniform diameter are obtained by decomposition of acetylene over $LaCu_{0.2}Ni_{0.8}O_x$ instead of $LaNiO_3$. The yield of CNTs prepared by $LaCu_{0.2}Ni_{0.8}O_x$ at 680°C is high up to 17 g/g.catal. and the diameter distribution of the CNTs is narrow, ranging from 9 nm to 14 nm.

Acknowledgments

This work was supported by the Sciential Innovation Project of Chinese Academy of Sciences.

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Received 8 January, 2003