Journal of Alloys and Compounds 521 (2012) 126–133

Contents lists available at SciVerse ScienceDirect

Journal of Alloys and Compounds

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

Field emission characteristics of carbon nanotube films fabricated on a metal mesh by filtration

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ARTICLE INFO

Article history:
Received 30 September 2011
Received in revised form 5 January 2012
Accepted 15 January 2012
Available online 28 January 2012

Keywords:
Field emission
Carbon nanotubes
Vacuum filtration
Metal mesh
Emission life span

ABSTRACT

A novel-structured field emitter was fabricated by vacuum-filtrating in sequence an aqueous suspension of thin, highly crystalline multiwalled carbon nanotubes (MWCNTs) and an aqueous suspension of thick, defective MWCNTs through a metal mesh placed on a polymer membrane. We could form a mechanically strong CNT film by weaving carbon nanotubes (CNTs) through the metal mesh residing inside the film. On drying at 70 °C, the polymer membrane was spontaneously separated from the CNT-mesh hybrid structure due to a large difference between their thermal expansion coefficients, producing vertically aligned bushes of thin CNTs on the CNT film. On top of the bushes, sharp CNT tips were well developed and worked excellently as field emitters. This structure was designed in such a way that the thin CNTs worked as field emitters while the thick CNTs supported the emitter CNTs. Our CNT emitters showed a high emission current density of 220 mA/cm^2 at 4.3 V/μm, and its emission life span, measured at 40 mA/cm^2 in a DC bias mode, was ~171 h. We expect that the CNT emitters with a novel structure are promising for the applications to field emission sources requiring small area but high current, for example, X-ray generators, microwave amplifiers, etc.

1. Introduction

Carbon nanotubes (CNTs) have drawn much attention as one of the superior materials for field emitters due to their high aspect ratios and nanometer scale in radii of curvature at tip and excellent electrical, mechanical, thermal, and chemical properties [1–3]. CNT emitters have been extensively explored not only for large area applications such as backlight units (BLUs) of liquid crystal displays (LCD) [4,5] and field emission lamps (FELs) [6,7] but also for small area but high current applications such as X-ray tubes [8–11] and microwave amplifiers [12,13]. For the latter applications, in particular, emission of high current from the small area CNT emitters may cause failure to them in a short period of time mainly due to Joule heating [14], field evaporation [15], or electrostatic interaction [16]. In order to solve such failure problems owing to high current field emission, CNT emitters should satisfy the requirements such as low electrical resistance, high crystallinity, optimum aspect ratio and distribution density, height uniformity, low outgassing rate, strong adhesion on substrates, etc. CNT emitters have been usually fabricated by vertical growth on substrates using chemical vapor deposition (CVD)[17], screen printing [18], spray coating [19], vacuum filtration [20], electrophoresis [21], and so on. Over the other methods, the vacuum filtration is of advantage since, in this method, we can choose CNTs having excellent electrical properties and crystallinity to prepare the CNT suspension and use a filtration membrane with a controlled pore size and morphology to optimize the aspect ratios, distribution density, and height uniformity of CNT emitters. This method, based on the solution processes, is simple and low cost. Furthermore, the filtration method is characteristic of low outgassing in vacuum during field emission because surfactants and solvent involved in the CNT suspension can be completely removed on annealing the filtrated CNT film.

Qian et al. [22] have reported that the CNT emitters prepared by vacuum filtration showed better field emission properties than those by screen printing for large area LCD-BLU applications. Although vacuum filtration is more appropriate for fabricating small area CNT emitters, there have been few studies on the application of vacuum filtration to fabrication of small area but high current CNT emitters. Thus, we investigate the fabrication of the CNT emitters by using vacuum filtration and their field emission characteristics for small area, high current applications. In this work, CNTs were filtrated through a metal mesh placed on a polymer membrane. A mechanically strong CNT film was designed to be formed by weaving CNTs through the metal mesh residing inside the film. Two types of CNT suspensions were filtrated in sequence:

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doi:10.1016/j.jallcom.2012.01.074

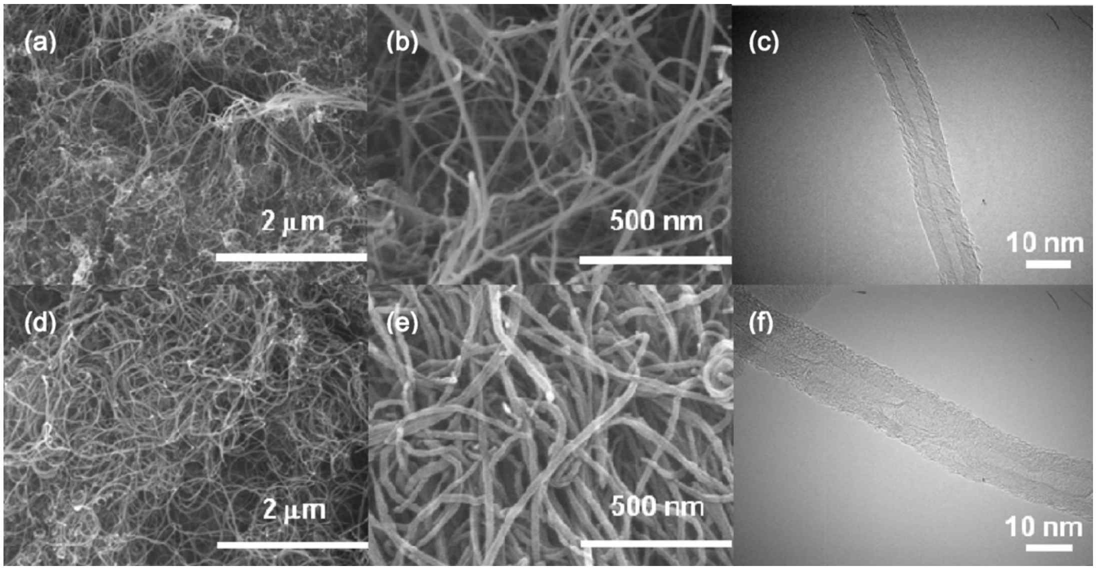

Fig. 1. SEM and TEM images of (a–c) thin and (d–f) thick CNTs used for emission and supporting layers, respectively. (c) and (f) are TEM images, and others are SEM images.

multi-walled CNTs (MWCNTs) with small diameters, short length, and high crystallinity and MWCNTs with large diameters, long length and low crystallinity. The former and latter CNTs were called as thin and thick CNTs, respectively. The thin CNTs formed an emission layer on the surface of the CNT film while the thick CNTs were strongly entangled with the metal mesh, forming a supporting layer of the emission layer. When the CNT film was separated by itself from the membrane during drying, the vertical CNT bushes were automatically developed with sharp tips on the film surface without further surface activation. In this report, we were able to fabricate small area but high current field emitters by incorporating the metal mesh, combining the two different types of CNTs, and optimizing the pore size of membrane.

2. Experimental

This study used thin and thick MWCNTs (Hanwha Nanotech Inc., Korea) as shown in Fig. 1. The CNTs were characterized by using Raman spectroscopy, thermogravimetric analysis (TGA), scanning electron microscopy (SEM), and transmission electron microscopy (TEM). CNTs featured the G peak at $\sim$1582 cm^{-1} and the D peak at $\sim$1350 cm^{-1}, which were attributed to stretching C-C bond and structural defects or impurities such as amorphous carbon, respectively [23]. In most cases, the crystallinity of CNTs is evaluated by the intensity ratio of G peak to D peak. In other way, the crystallinity of CNTs may be assessed by their oxidation

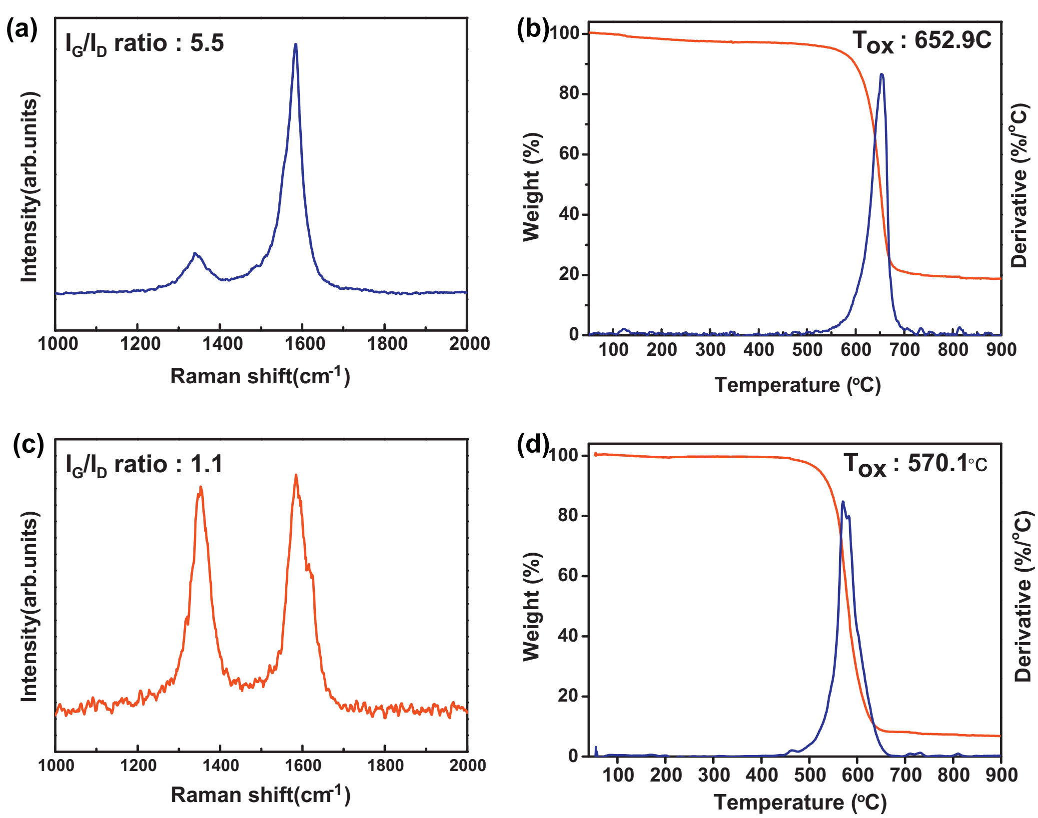

Fig. 2. Raman spectra and thermogravimetric analysis (TGA) and derivative thermogravimetric (DTG) curves (a, b) thin and (c, d) thick MWCNTs.