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Preparation of a miniature carbon nanotube paste emitter for very high resolution X-ray imaging

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ABSTRACT

A miniature carbon nanotube (CNT) paste emitter dot with a diameter of ~50 μm was fabricated onto the cross-sectional surface of Kovar (nickel-cobalt ferrous alloy) rod without using a sophisticated photolithography. A highly adhesive CNT paste was prepared with silicon carbide and nickel nanoparticles, and then the small CNT emitter was fabricated by a novel and facile technique, called point contact method. The prepared emitter generates very high current density of ~11.2 A/cm^2 when measured using a diode configuration. We further observed for the first time the brightness of electron beam generated from the CNT paste emitter that is a more practical point electron source than the individual CNT emitter. The triode structure fabricated with a miniature CNT paste emitter and a gate aperture (20 μm in diameter) generated a collimated electron beam by which very high resolution (>16 lp/mm) X-ray imaging was achieved.

1. Introduction

A carbon nanotube (CNT) has shown the excellent field emission properties including high current density and long-term emission stability due to its large aspect ratio, small radius of curvature, and chemical stability [1–3]. Thus, CNT has been conceived to be an ideal field emitter for being used in various field emission devices, such as backlight unit [4], field emission display [5], and X-ray tube [6–8]. Among the aforementioned applications, the X-ray tube using CNT emitters has recently attracted much attention because the CNT-based filed emitter has a number of advantages over conventional thermionic tungsten filament that has long been used for the generation of X-ray. The strong points of CNT emitter include low operating temperature, little power consumption, and narrow energy dispersion of electron beams [9,10]. Furthermore, the triode structure with a gate electrode can be easily manufactured using CNT emitters, which makes possible the operation of an X-ray tube with digital addressing.

CNT emitters have been in general fabricated by either the direct growth of CNT using chemical vapor deposition (CVD) [11,12] or the screen printing of CNT paste [13,14]. The CVD is useful for the fabrication of CNT emitters on a desired position. However, apart from its complicated high-cost process, the CVD suffers from the limited growth temperature, causing the poor crystallinity of CNT. On the other hand, the screen printing of CNT paste is not only cost-effective but also capable of using high quality CNTs synthesized independently. The preparation of a CNT paste emitter on a desired location can also be achieved using a patterned screen. Nevertheless, CNT paste emitter is also difficult to be used in a miniature X-ray tube since the preparation of a very small CNT emitter using the screen printing is a challenge.

Another importance for the fabrication of a very small emitter arises from the efficient operation of triode structure. Since the gate electrode is extracting the electrons from the emitter, the CNT emitter with a large dimension causes a high leakage current to the gate. Meanwhile, the gate electrode with a very small aperture is preferred to generate a collimated electron beam that is useful for high resolution X-ray imaging. Therefore, the diameter of CNT

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emitter should be small enough to minimize a leakage current and to generate a high brightness collimated electron beam. Based on these motivations, we made use of a novel technique, called point contact method, to fabricate a CNT paste dot with a diameter of ~50 μm, followed by the generation of a collimated electron beam through the small gate aperture. Note that such a small emitter could be prepared without using photolithography. We measured the brightness of electron beam emitted from the CNT paste emitter. The collimated electron beam generated was then used to accomplish very high resolution X-ray imaging.

2. Experimental

CNT paste was prepared by an optimized ball-milling process using few-walled carbon nanotubes purchased from Xinnano Inc. First, 0.36 g of silicon carbide (SiC) and 0.11 g of nickel (Ni) nanoparticles were mixed with 4 g of terpineol by rotating and spinning the closed bucket. SiC and Ni nanofillers were found to endow CNT paste with strong adhesion to the substrate when the paste emitter was thermal-treated in a vacuum. Second, 0.15 g of carbon nanotubes was added and then, mixed by rotating and spinning with 5 mm zirconia balls. Finally, we added 0.2 g of an organic binder (ethyl cellulose), followed by the mixing again with zirconia balls.

Kovar (nickel-cobalt ferrous alloy) wire with a diameter of 1 mm was purchased, and then cut into rod shape having the length of 15 mm. The as-cut rod was found to have very rough surface. Such a rugged surface leads to non-uniform heights of CNT emitting tips, which causes short-term emission stability [15]. Besides, the sharp edges of cut surface may give birth to arcing when a high voltage is applied for electron emission. For these reasons, we polished the Kovar rod using a barreling process in order to improve the smoothness of the cut surface. As a consequence, the polished surface, which is smooth enough to give CNT paste emitters the uniform tip-heights, was obtained. It was expected that the barrel-polished cut Kovar surface would make it possible to form a stable field emitter on it.

The field emitter was fabricated on a cross-sectional surface of Kovar rod by a point contact method. The detailed procedure is described in the next section. The field emission properties of a CNT paste emitter were evaluated using both diode and triode structures in a vacuum chamber of 1×10^{-7} Torr. We also simulated the trajectory of a collimated electron beam using a commercial simulator, OPERA 3D. A transmission type X-ray generating system was constructed in a vacuum chamber. The tungsten film deposited on a beryllium plate by DC magnetron sputtering was used as an anode target. The X-ray generated at the anode target transmits through a beryllium window positioned behind the target. The line pair phantom is placed in front of the beryllium window. The high resolution X-ray image of a phantom was obtained using an image intensifier which is placed 280 mm away from the window.

3. Results and discussion

Fig. 1a and b are the optical microscope images showing a bare tungsten probe and the probe with surrounding CNT paste, respectively. The radius of curvature at very end point of the bare tip is 5 μm. Immersing the probe into the CNT paste resulted in the preparation of the probe embedded in the paste (Fig. 2b) through the equilibrium between gravitational force and adhesive strength of CNT paste to the probe surface. Fig. 1c shows the schematic diagram of point contact method developed for the fabrication of a miniature paste emitter in this study. The tungsten probe with surrounding CNT paste was installed in a position controller which can move the probe in *x*, *y*, and *z* axis directions. The probe is connected to a micro-ohmmeter that is plugging into the bottom surface of Kovar rod. After the probe is positioned at right above the center of Kovar surface by *x*–*y* direction control, the position controller moves the probe downward very slowly. While the tungsten probe is being moved downward, the micro-ohmmeter displays the resistance as extremely soon as the probe touches the Kovar surface. The position controller moves the probe upward at the time of sensing a resistance, resulting in the formation of a miniature CNT paste dot on the Kovar surface. Note that this is quite a simple technique that can be easily scaled-up for practical applications. Fig. 1d shows the optical microscope images of CNT paste dots prepared on a polished Kovar surface using the point contact method. After the contact of CNT paste to the surface, the Kovar rod was dried at 90 °C for 10 min to evaporate a solvent. Then, the rod with a dried paste on it was heat-treated at 300 °C for 3 h in air atmosphere to burn off organic ingredients, followed by physical surface treatment using an adhesive roller for the exposure and vertical alignment of nanotubes. These vertically aligned carbon nanotubes have been used as an emitter in field emission devices [4,7,8,13–16]. However, the thermal treatment temperature of 300 °C is not high enough to get rid of organic ingredients completely. To make things worse, we cannot increase the temperature further in air atmosphere since the nanotubes would be burned off at high temperature. Hence, there is a possibility of outgassing from the paste during field emission. In order to overcome this problem, we carried out one more thermal treatment in a vacuum, in which the thermal-treated CNT paste emitter was heat-treated again at 810 °C for 10 min at a pressure of 1×10^{-7} torr. This treatment was found to eliminate the residual organic materials completely. Furthermore, the adhesion of paste emitters was greatly improved by the reaction between Kovar surface and nano-sized fillers. We recently reported on the detailed mechanism for improving the adhesion through the vacuum annealing [16]. The optical microscope images were taken after each step; drying, firing, physical surface treatment, and vacuum annealing/surface treatment. Fig. 1d shows that the size of CNT dot was not changed during a series of above mentioned procedures, confirming the sufficient adhesion of the prepared emitter. The side-view SEM image taken after vacuum annealing/surface treatment is shown in Fig. 1e. In order to observe a clear image, we took three images on the slightly different lateral positions and then combined those pictures into one micrograph. The diameter of CNT paste dot was found to be as small as ~50 μm. It has been reported that the CNT paste emitter with a diameter of smaller than 50 μm could be fabricated using a photolithography [4,14]. However, such small emitters are very difficult to be prepared using a conventional screen printing without the sophisticated technique like photolithography. Besides, the photolithography is not easy to be conducted on the rod-type cathode that is useful for the preparation of a point-type CNT emitter. It is therefore believed that the point contact method is one of the promising techniques for the preparation of a miniature CNT paste emitter. Although the CNT tips formed on the center position are a bit higher than those on edge parts, the vertically aligned carbon nanotubes with relatively uniform heights were prepared throughout the whole emitter area.

Field emission properties of the CNT paste emitter prepared on the cross-sectional surface of Kovar rod was evaluated using a diode configuration in a vacuum chamber. Fig. 2a is the photograph showing the diode configuration used for the field emission measurements. Bare Kovar rod without an emitter was placed 200 μm away from the cathode surface for being used as an anode. The emission current (*I*) versus applied voltage (*V*) curve was measured for three consecutive times with continuous DC mode. Fig. 2b represents the obtained *I*–*V* curves. The lower limit of measurement system (Keithley 248 High Voltage Supply) is 1 μA. The turn-on voltage was, therefore, defined as the voltage for the emission