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Fabrication and field emission characteristic of microcrystalline diamond/carbon nanotube double-layered pyramid arrays

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ABSTRACT

In this study, microcrystalline diamond (MCD)/carbon nanotube (CNT) double-layered pyramid arrays have been fabricated by microwave plasma chemical vapor deposition (MPCVD). First, the silicon template with pyramidal cavity arrays was manufactured using photolithography with anisotropic wet-etching. MCD film was then grown on the silicon template by MPCVD. After the sol–gel catalyst which contains ferric nitrate, tetrabutyl titanate and n-propanol was spun on the MCD film, CNT film was grown by MPCVD. In order to etch away the silicon template, hydrofluoric acid and nitric acid are used for 2 h. From the results, the turn-on electric field of MCD/CNT double-layered pyramid arrays is about 2.84 V/μm and lifetime over 100 h.

1. Introduction

Microcrystalline diamond (MCD) or carbon nanotube (CNT) as the cathode for the application of field emission (FE) has been investigated in recent years [1–6]. Diamond has excellent physical and chemical properties, including high thermal conductivity [7] and negative electron affinity [8], which make it suitable to become the FE emitter and operate stably and reliably. CNT is also suitable as FE emitter due to its small tip radius, low work function, large aspect ratio and good chemical stabilities [4,9]. CNT as FE emitters are widely used on display, x-ray tubes [10], lamp [11] and electron-gun sources [12].

In general, the turn-on electric field of the diamond emitter is higher than that of CNT [13–15]. Though CNT has the excellent electron conductivity and the low turn-on electric field as FE emitter, it can be easily broken down due to the heat accumulation. This is probably caused by the poor thermal contact at the interface between CNT and the substrate resulting in CNT local evaporation and destruction [16]. Some studies have focused on improving the FE stability of the CNT by enhancing the heat conducting at the interface. Varshney et al. [17] fabricated a monolithic CNT/diamond composite; they found that the seamless integration of CNT and diamond microcrystals helps to remove the heat from the CNT and avoid the heat accumulation. Zou et al. [18] proposed a diamond-coated multi-walled CNT“teepee”structure. Yang et al. [19] created a vertically aligned CNT/diamond double-layered structure.

In the current study, MCD/CNT double-layered pyramid arrays had been fabricated. CNT is designed as FE emitters, while MCD is the heat dispersion, which prevented CNTs from heat accumulation. Furthermore, MCD is fabricated into pyramid arrays to reduce FE turn-on field.

2. Experimental details

In order to compare FE characteristics with different structures and materials, four types of FE devices “MCD plain plate”, “MCD/CNT double-layered plain plate”, “MCD pyramid arrays” and “MCD/CNT double-layered pyramid arrays” were fabricated. The schematic diagram of these FE devices are shown in Fig. 1.

The fabrication processes of MCD/CNT double-layered pyramid arrays are illustrated in Fig. 2. A titanium layer was deposited on silicon substrate and patterned by a conventional photolithography process. The inverted pyramidal cavities were fabricated by anisotropic wet-etching which the KOH aqueous solution (30% KOH + 70% D.I. water, 80°C) was used to etch the Si substrates that contain titanium pattern. The obtained substrates were then soaked into the etching aqueous solution (ammonia, hydrogen peroxide and D.I. water (1:1:2)) to remove titanium pattern. The opening of the pyramidal cavities is a 12 μm by 12 μm square with the depth of 8.4 μm. The interval between cavities is 25 μm. Diamond nucleation were ultrasonically seeded on the templates in a solution with diamond powders (diameter of 4 ~ 12 nm), titanium powders and methanol for 30 min. After that, the templates were ultrasonically cleaned by acetone to remove the rest adhered particles. The MCD film was grown by an MPCVD process, using Ar/CH_4/H_2 (50/1/49 sccm) plasma at 10.7 kPa with 1.2 kW for 3 h. The thickness of MCD film is about 1.4 ~ 2 μm. The sol–gel catalyst, containing ferric nitrate, tetrabutyl titanate and n-propanol was coated on the MCD film by spin coating. MPCVD was used to grow CNT layer with CH_4/H_2/N_2 (30/120/60 sccm) plasma at 5.3 kPa and 800 W for 30 min. The thickness of CNT layer is about 12 μm. An aluminum plate was used to sustain the

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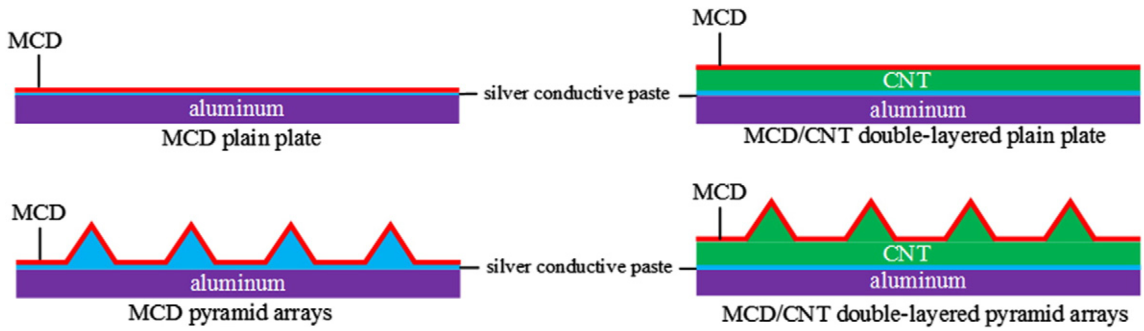

Fig. 1. The schematic diagram of four types of FE devices.

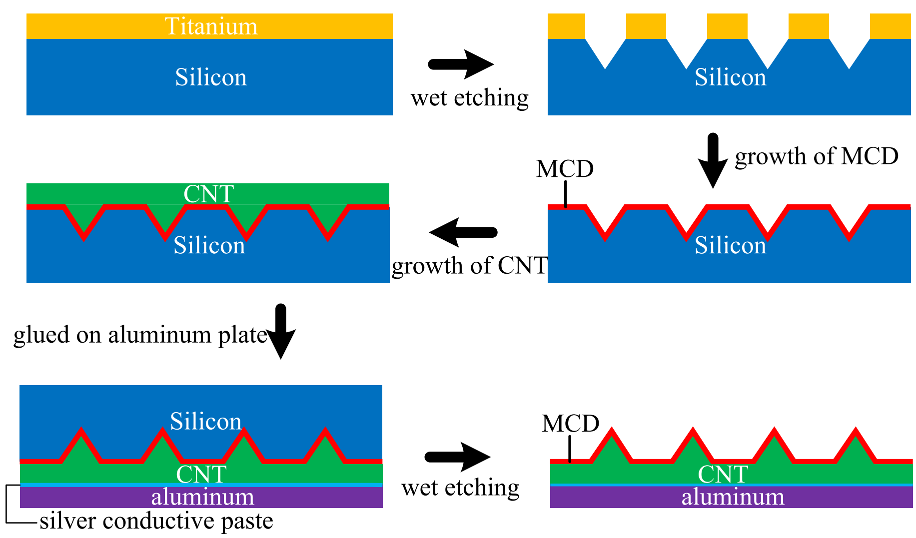

Fig. 2. The schematic diagram of MCD/CNT double-layered pyramid arrays fabrication procedures.

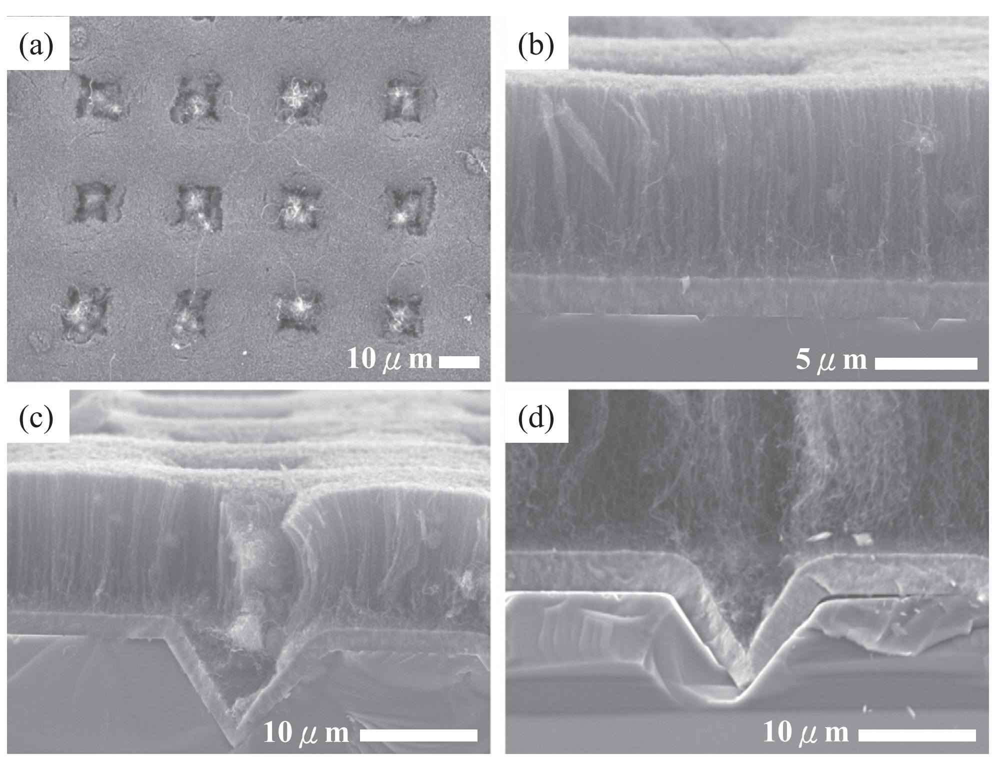

Fig. 3. SEM images of the MCD/CNT double-layered grow on the silicon template. (a) Top view of the CNT layer. (b, c and d) Cross section.