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Study of nanometric thin pyrolytic carbon films for explosive electron emission cathode in high-voltage planar diode

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ABSTRACT

We report on an experimental study of explosive electron emission properties of cathode made by nanometric thin pyrolytic carbon (PyC) films (2–150 nm) deposited on Cu substrate via methane-based chemical vapor deposition. High current density at level of 300 A/cm^2 in $5 \cdot 10^{-5}$ Pa vacuum has been observed together with very stable explosive emission from the planar cathode. The Raman spectroscopy investigation proves that the PyC films remain the same after seven shots. According to the optical image analysis of the cathode before and after one and seven shots, we conclude that the most unusual and interesting feature of using the PyC films/Cu cathode for explosive emission is that the PyC layer on the top of the copper target prevents its evaporation and oxidation, which leads to higher emission stability compared to conventional graphitic/Cu cathodes, and therefore results in longer working life.

1. Introduction

The unique capability of the explosive-emission cathode to emit high current density pulsed electron beams has made it a good candidate for applications such as electron source for high-current diodes. A number of works analyze the possible causes contributing to the generation of currents initiating explosive emission. As has been stated in earlier work, structure imperfections, artificial or natural, of the cathode surface, on which the electric field strength can be enhanced hundreds of times, are a necessary prerequisite for explosive emission to begin [1].

The operation of cathodes under explosive emission from the cathode surface is determined by the cathode plasma which can be considered as a metal surface with close to zero electronic work function. A nonstationary potential barrier is formed near the cathode surface: the value of the potential barrier is small as compared to the voltage applied across the cathode, the minimum of the potential is virtually the same as the applied voltage and the position of the potential minimum practically coincides with the cathode surface [2]. Hence we can suppose the initial velocity of the outcoming electrons to be equal to zero.

Given the cathode–anode gap, the amplitude of the voltage pulse, and the rise time, there is a possibility to improve the homogeneity of the emitting plasma using materials with small delay times, as well as materials and cathode configurations providing a high field amplification factor.

Explosive-emission cathodes fall into several groups: cathodes with one or several tips, multi-tip and metal-dielectric cathodes, and flat metal or graphite cathodes with rough surface. The latter cathodes are advantageous because of manufacturing simplicity, reproducibility and controllability. All mentioned types of cathodes are characterized by the presence of the elements enhancing the local electric field strength across the particular areas of the cathode surface and promoting the start of the explosive emission under smaller diode voltage, decreasing the delay time of the cathode plasma formation (emission startup), as well as improving the homogeneity of the electron beam.

Historically, cathodes with graphite emission area were among the earliest cathodes used in high-current diodes of planar geometry. Natural roughness of a graphite surface promotes the build-up of a large number of emission centers under comparatively moderate average electric field strength in a diode, maintaining quite a homogeneous structure of microroughness on the cathode surface immediately after mechanical treatment when manufacturing, as well as during the cathode operation. For this reason, just graphite cathodes are used in devices for which increased cathode life is required.

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Multi-tip cathodes consist of arrays of thin wires, foils, thin-walled tubes or carbon fibers that form the ordered structure of the emission centers (roughness) enhancing the electric field on the cathode. Development of such cathodes requires special technologies and design solutions that can provide high reliability (operability), performance, life, and necessary beam characteristics.

Carbon-fiber cathodes [3–5] are attractive for their unique capability to operate at very small values of the average electric field strength in a diode. Carbon fibers start to emit intensively when the average electric field strength in the diode is as small as tens of kilovolts per centimeter.

Studies of nanostructured carbon films interfaced with different materials [6] urge consideration of the emission properties of cathodes with carbon coating [7]. In the present communication we will compare cold explosive emission cathodes made of polished copper with those made of polished copper covered with a pyrolytic carbon (PyC) film in a planar high-current diode.

2. Experimental details

2.1. Pyrolytic carbon deposition on Cu substrate

We fabricated five identical cathodes (1 cm thick, 5 cm in diameter) made of M1 copper, four of them were covered with PyC films of different thicknesses. In the experiment, PyC films were grown directly on the cathode surface by chemical vapor deposition (CVD). The conventional hot wall CVD set-up consisted of computer controlled gas flow controllers for hydrogen and methane, vacuum pump and tubular furnace with a quartz reaction chamber. The sample was first heated up to 1000 °C in hydrogen atmosphere (~10 Pa). After that the chamber was vacuumed and CH_4:H_2 gas mixture was injected in the tube for PyC synthesis. The thickness of the PyC layer is typically controlled by the process time, methane concentration and pressure. In our experiment, we used different hydrogen:methane concentrations (15:60, 15:120, 15:180, 15:240) and pressures to control the thickness of the PyC layer on copper substrate. After 1 h, the chamber was vacuumed and the reaction chamber was cooled down overnight in H_2 atmosphere.

PyC film thickness was measured by a stylus profiler (Veeco Instruments, Dektak 150) with an accuracy of 1.5 nm. The thicknesses of PyC films are dependent on the methane concentration [8] and were measured to be 2 nm, 10 nm, 120 nm and 150 nm for 15:60, 15:120, 15:180, and 15:240 CH_4:H_2 respectively. Hereinafter we label PyC/Cu cathodes with the numbers 15:60, 15:120, 15:180 and 15:240, meaning the H_2:CH_4 concentration during the synthesis.

In our experiment, graphitization process was done in static atmosphere, i.e. there was no gas flow in the chamber during the PyC synthesis. This greatly reduces gas consumption and, more importantly, since PyC grows following a combination of heterogeneous and homogenous reactions the static atmosphere allows more time for the homogenous reactions to take place.

It is worth noting that copper substrates are conventionally used in graphene synthesis. This is because carbon solubility in copper is so low that the catalytic reactions between hydrocarbon precursor and copper are considered to take place only on the very surface of copper substrate. At low hydrocarbon concentrations, this results in deposition of a monolayer graphene. However, by modifying the process parameters one can grow a few layered graphene on copper via template graphene growth [9]. Moreover, if the concentration of methane is increased, the graphene layers are coated with thicker PyC layer resulting a composite film consisting of a few graphene layers coated with pyrolytic carbon (see results of structural characterization below).

2.2. Structural investigation

High resolution transmission electron microscopy (HRTEM) measurements were carried out by using a FEI Tecnai F20 electron microscope operating at 200 keV. For TEM measurements PyC carbon films were transferred onto a 3 mm copper grid (1000 mesh). Electron diffraction patterns were collected using the selected area electron diffraction (SAED) technique.

One can observe (see Fig. 1) that the graphene layers are coated with thicker PyC layer resulting in a composite of a few layered graphene and amorphous pyrolytic carbon film. SAED pattern (Fig. 1 inset) taken from the area of 200 nm corresponds to a mixture of amorphous carbon and graphite as revealed by the presence of sharp spots. The presence of [002] spots shows that some graphite planes are perpendicular to the surface of the film. The presence of graphite flakes [001] parallel to the surface is also clearly visible on the image.

Our synthesis process is similar to that of synthesis of graphene. But in order to synthesize graphene, i.e. atomically thin film composed of carbon atoms arranged in the honeycomb lattice, the graphitization process should take place at low concentration of carbon precursor (e.g. methane) in presence of a catalyst (i.e. copper). However, it is worth noting that graphitization process or deposition of carbon atoms on the substrate that occurs at temperature of about 1000 °C does not depend on the substrate. In Refs. [8,10], we demonstrated that on dielectric substrates, one can deposit PyC films whose thicknesses vary from 5 to 110 nm, depending on the methane concentration in the chamber. In the current paper, we employ a copper cathode as a substrate. The presence of copper results in formation of a few-layer graphene films on the copper surface (see Fig. 1). However, extended deposition time and high concentration of carbon precursor in the CVD chamber (H_2:CH_4 ratio up to 15:240) results in formation of a thick carbon layer on the top of the graphene. Specifically, the first layer of graphene is grown catalytically on a copper surface at a temperature of about 1000 °C, which is high enough for spontaneous methane decomposition [11] that takes place inside the chamber (i.e., not only on the copper surface). The decomposed hydrocarbon molecules start to form a new layer of carbon on the top of the first graphene layer. This process is referred to as template graphene growth and is usually considered as unwanted phenomena because crystallinity of these extra graphene layers is lower than that of the first ones, which were grown on the surface of the copper catalyst. Moreover the crystallinity of the

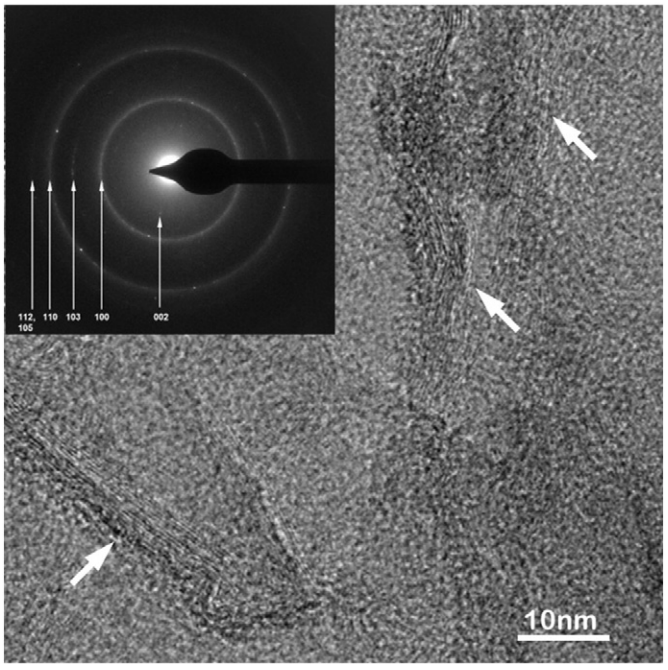

Fig. 1. HRTEM image of the PyC film. Indexed SAED pattern is shown in the inset. The graphite flakes are marked with white arrows.