

# Thermionic Electron Emission from Ultrananocrystalline Diamond Films

Pramila S<sup>1</sup>, Mohanty R<sup>1</sup> and Sankaran KJ<sup>2\*</sup>

<sup>1</sup>Department of Chemical Engineering, CV Raman College of Engineering, Bhubaneswar, India

<sup>2</sup>Advanced Materials Technology, CSIR-Institute of Minerals and Materials Technology, Bhubaneswar, India

## Abstract

In this work, we synthesized CH<sub>4</sub>/Ar-based ultrananocrystalline diamond (UNCD) films by using a microwave plasma enhanced chemical vapor deposition method. These films exhibit improved thermionic electron emission (TEE) properties with a work function value of 4.8 eV and a high current density of 0.47 mA/cm<sup>2</sup>. The high field enhancement factor and the amorphous carbon phases at the grain boundaries of UNCD films are the reliable features for the improved TEE characteristics of UNCD films. The results encourage the potential use of UNCD films for efficient thermionic electronic emission devices.

\*Correspondence to: Kamatchi Jothiramalingam Sankaran, CSIR-Institute of Minerals and Materials Technology, Bhubaneswar, India; E-mail: [kjsankaran@imtm.res.in](mailto:kjsankaran@imtm.res.in)

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## Introduction

Yielding clean and renewable energy is regularly connected with photovoltaic cells or solar cells. These cells generate valuable electricity by the accumulation of solar energy. On the other hand, easily obtained heat energy is being believed and estimated as an efficient way of energy conversion and to decrease the reliance on the usual sources of energy. An appropriate option is to convert thermal energy to electrical energy and the loss of thermal energy due to mechanical work is minimal. Hence, thermionic electron emission (TEE) is a potential renewable energy source that converts thermal energy into electric energy and possesses several applications in communication and space propulsion [1].

Basically, field electron emission (FEE) is the emission of electron from a material's surface due to an external applied field but in TEE the electrons are ejected from the material into vacuum space by an external applied thermal energy [2]. The thermionic electron emission is modelled by Richardson-Dushman (R-D) theory [2].

Diamond possesses high electron mobility, high strength, high breakdown voltage and stable chemical properties [3]. On account of wide band gap nature, diamond can endure at high temperature and hence it is a suitable cathode for TEE devices [4].

## Experimental

One square cm silicon substrates were used to synthesis UNCD films in CH<sub>4</sub> (1%) and Ar (99%) gas mixtures using an IPLAS microwave plasma enhanced chemical vapor deposition (MPECVD) reactor. The microwave power, pressure and growth time were retained at 1200 W, 120 Torr, and 60 min respectively. The silicon substrates were

pre-seeded using nanodiamond powder and titanium powder using ultrasonication process.

The as-synthesized UNCD films were characterized using various characterization techniques. A tunable parallel plate setup has been used to analyze the FEE characteristics of the UNCD films. The obtained UNCD film was used as a cathode, whereas Mo rod of diameter 3 mm used as an anode. The distance between cathode and anode was controlled using a micrometer. Using Keithley 2400 electrometer, the current (I)-voltage (V) characteristics were measured and Fowler-Nordheim (F-N) theory [5] was applied to model the FEE properties of these films. A vacuum system with 10<sup>-8</sup> Torr base pressure was used to perform the TEE measurements. A thermo coax cable was used to heat the sample and the temperature is measured by using a K type thermocouple. The emitter was UNCD films and the collector was a molybdenum disc. A 150 μm thick alumina spacer was used to separate the emitter and collector. TEE current was recorded as function of temperature ranging from 573 K to 873 K for an applied bias of 10 V.

Diamond films synthesized using chemical vapor deposition technique comprises defects and *sp*<sup>2</sup>-phases [6]. While decreasing the grain size, increases the *sp*<sup>2</sup>-content, ensuing in improved electronic conductivity. Among diamond films, ultrananocrystalline diamond (UNCD) film has *sp*<sup>3</sup>character grains and *sp*<sup>3</sup>character (amorphous carbon) grain boundaries [7]. Here, we examined the FEE and TEE characteristics of UNCD films and the morphology of UNCD films was studied under transmission electron microscopy to know the FEE and TEE mechanism of UNCD films.

## Results and Discussion

Scanning electron micrograph (JEOL JSM-6500F) in Figure 1(a)



shows random, spherical and equiaxed ultrasmall nanostructured grains in the UNCD films. The Raman spectrum (Renishaw, excitation wavelength= 632 nm) of UNCD films presented in Figure 1(b), which shows the  $\nu_1$  and  $\nu_2$  modes at around  $1160\text{ cm}^{-1}$  and  $1475\text{ cm}^{-1}$  consequent to trans-polyacetylene phases exist at the grain boundaries of UNCD films [8]. The D\*band at  $1340\text{ cm}^{-1}$  denotes the disordered  $sp^2$  carbon, whereas the G band observed at  $1540\text{ cm}^{-1}$ .

The FEE characteristics of UNCD films are presented in Figure 2(a) and the inset of Figure 2(a) illustrates the respective F-N plots.

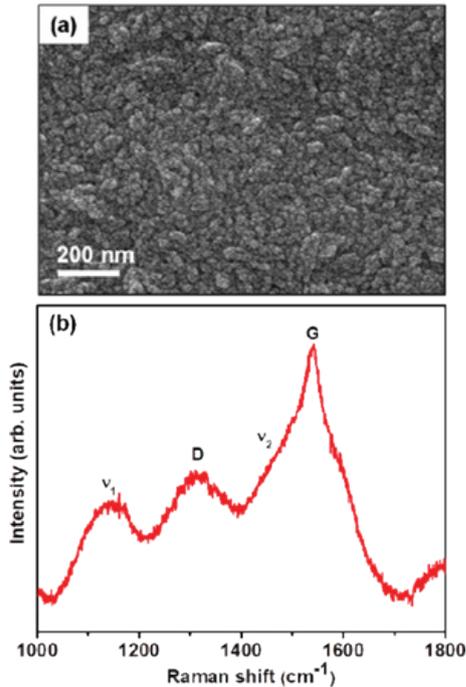


Figure 1: (a) SEM micrograph and (b) Raman spectrum of UNCD films.

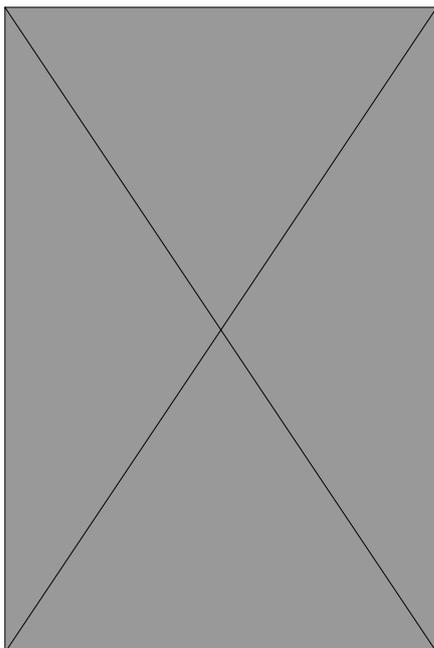


Figure 2: (a) FEE and (b) TEE characteristics of UNCD films. The inset of (a) displays the corresponding F-N plot of Figure 2(a). The inset of (b) shows the schematic representation of TEE measurement setup.

The UNCD films show a value of  $10.2\text{ V}/\mu\text{m}$  turn-on field with the FEE current density value of  $1.48\text{ mA}/\text{cm}^2$  (at an applied field of  $35.5\text{ V}/\mu\text{m}$ ) and the field enhancement factor value of 650. While high efficiency electron sources based on superior FEE materials (i.e. containing low effective work functions) show very useful applications, another potential application based on similar characteristics is a thermionic electron emitter.

Figure 2(b) displays the TEE characteristics of UNCD films and the schematic of a TEE setup is presented in the inset of Fig. 2(b). The TEE current density versus T for the UNCD films with an applied voltage of 10 V is displayed in Figure 2(b). A gradual escalation in TEE current density with the rise in temperature and finally achieved  $0.47\text{ mA}/\text{cm}^2$  at the temperature of 923 K for UNCD films. Moreover, the calculated work function ( $\phi$ ) and Richardson constant of UNCD films obtained from the R-D equation are 4.8 eV and around  $0.6\text{ A}/\text{cm}^2\text{ K}^2$ , respectively.

Now a question arises, how do UNCD films acquire enhanced FEE and TEE properties? To know the answer, the microstructure of the films was examined under transmission electron microscopy (TEM; JEOL 2100). Ultra-small (5-10 nm) spherical diamond grains are observed from the bright field (BF) TEM image of UNCD films (Figure 3(a)). The SAED (selected area electron diffraction) pattern displayed in Figure 3(b) shows diamond diffraction rings such as (111), (220) and (311), and a diffuse ring at the center represents amorphous carbon phases in the UNCD films.

From the above observations, the grain boundary phases are the genuine factors for the enhanced electron emission characteristics of

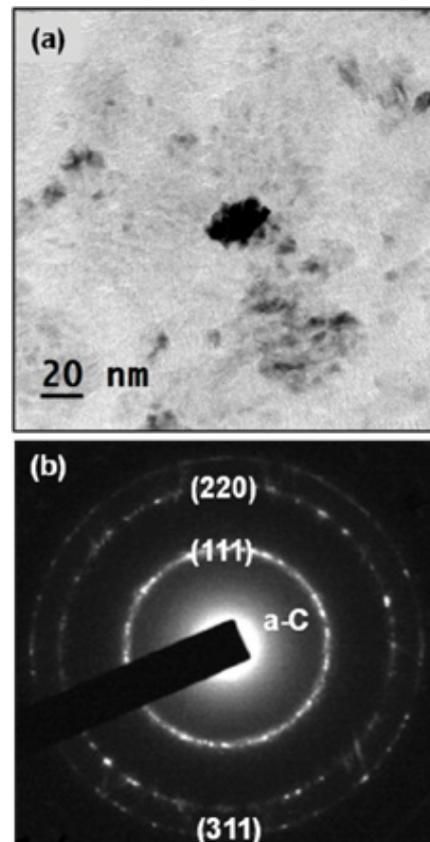


Figure 3: (a) Bright field TEM and (b) SAED pattern of UNCD films.



diamond films [6]. We propose here the same conduction mechanism for UNCD films. The amorphous carbon assists the electrons to move easily from the grain boundaries to the emitting surface and hence emits to vacuum. Consequently, ultranano-sized diamond grains and amorphous carbon are the reliable features for the superb FEE/TEE properties of UNCD films.

## Conclusion

UNCD films were successfully synthesized using  $\text{CH}_4/\text{Ar}$  plasma. Ultrananocrystalline diamond grains and amorphous carbon phases exist in UNCD films. The UNCD films show enhanced FEE and TEE characteristics. The amorphous carbon phases and high field enhancement factor are the dependable factors for the enhanced FEE and TEE properties of UNCD films. Therefore, UNCD-based thermionic emitters are considered as an efficient cathode for thermionic electron sources.

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