Stable and high current density electron emission using coniferous carbon nano-structured emitter

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The electron emission properties of a coniferous carbon nano-structure (CCNS) based field emission electron gun have been measured. The CCNS was grown on a stainless steel substrate using chemical vapor deposition (CVD) and biased to about 50 kV. Stable, high-current-density electron emission (102 mA/cm²) was measured continuously for more than 1300 h. This result compares favorably to printed and directly grown carbon nanotube (CNT) based emitters. A wide range of potential applications are foreseen for CCNS based electron emitters, such as high intensity X-ray sources.

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1. Introduction

The carbon nanotube (CNT) was discovered in 1991 [1]. Many measurements of CNT as a field emission electron source were reported from 1995 [2–4] to recent years towards applications in field emission displays, X-ray sources, and so on. For such applications both long lifetime and high emission current are required. CNT can generate high emission current densities although it has the disadvantage that the lifetime of the emitter is short. Emission stability measurements for printed CNT emitters [5–13] and CNT emitters directly grown on substrates [14–17] have been reported. For printed CNT emitters these experiments were typically performed at an emission current density of about 1 mA/cm² or less. The longest such measurement for a printed CNT emitter was 45,000 h at an emission current density of about 1.27 mA/cm² operated at 60 Hz and a 10% duty ratio [5]. Directly grown CNT emitters can generate higher emission current densities than printed CNT emitters. The longest reported stability measurement of a directly grown CNT emitter was 200 h at an emission current density of 150 mA/cm² and continuous DC emission [14].

Recently, a new type of carbon based electron emitter, the so-called coniferous type carbon nano structure (CCNS), was discovered by A. Hiraki, M. Haba and H.-X. Wang [18]. This carbon nano structure can generate an electron beam of more than 100 mA/cm² [19]. At AIST we are developing X-ray tubes for X-ray imaging and X-ray fluorescence spectrometry using CCNS emitters [20]. In this study, we measured the long term electron emission stability for CCNS at an emission current density of 102 mA/cm².

2. Experiment

The experimental setup for the emission stability measurement is shown in Fig. 1. A vacuum chamber in insulating oil was kept at a pressure below 1 × 10⁻⁷ Torr by an ion pump and non-evaporable getter (SAES Getters St 172). In the vacuum chamber, the electrodes consist of the cathode, the anode, and the extraction electrode. The anode and the cathode are insulated from the extraction electrode by 35-mm-long ceramics, and a DC voltage up to ±65 kV can be applied. The cathode consists of a CCNS coated substrate for the electron emission source, and a cap for focusing the electron beam [21]. The gap between the cathode and the extraction electrode can be adjusted between 3.0 and 10.0 mm. In this measurement, the gap was 3.0 mm. The electric field around the cathode and extraction electrode was calculated by the TriComp simulation code (Field Precision). The simulation was used to design the electron source such that the electron beam at the anode has a beam size of between 0.5 and 1.0 mm. The cap electrode ensures a uniform electric field between the emitter and extraction electrode. For a cathode voltage of 60 kV, the average electric field on the surface within 1 mm in diameter of emitter substrate was calculated to be 7.86 MV/m. Although the electric field increases towards the axial center, the difference in value at 1.0 mm in diameter is uniform to within 1%. This value was used to calculate the electric field from the applied cathode voltage. Electrons emitted from the cathode pass through a 4-mm-diameter hole in the extraction electrode and collide with the anode. In the present measurement the anode voltage was 20 kV and does not affect the electric field around the emission surface. In order to prevent heating of the electrodes, the vacuum chamber was cooled by circulating insulating oil through a chiller to maintain a temperature of 15 °C. Before this measurement,
we observed no electric breakdown of the equipment at an applied voltage of about ±65 kV to the anode and the cathode without CCNS. In this condition, the leak current was less than 0.02 mA.

The CCNS used in the present experiment for the electron source, as shown in Fig. 2, was grown on a SUS304 substrate using the DC plasma chemical vapor deposition (DCPCVD) method [18]. The substrates were treated in H2 plasma at a pressure of 30 Torr for 5 min. CCNS were then grown on the substrate in a mixed gas of CH4 and H2 (CH4/H2: 8%) at a pressure of 75 Torr for 90 min with a substrate temperature of around 1100 °C. The substrate has a concave structure with a radius of curvature of 8.5 mm and a diameter of 6.0 mm. The 6.0-mm-diameter CCNS coated area on the substrate was shaved off to a diameter of 1.0 mm using a lathe. A photograph of the 1.0 mm diameter CCNS coated substrate is shown in Fig. 2(a). Electron microscopy images of the CCNS are shown in Fig. 2(b, c). The error in the diameter of the CCNS coated area was estimated to be ±0.05 mm from the SEM image. Individual CCNS grow randomly on the substrate, and the diameter of a single structure ranges from several μm to 10 μm, as shown in Fig. 2(b). The structure has a shape like a coniferous tree as shown in Fig. 2(c). The CCNS has the same tip shape as a CNT [18], and the structure becomes thicker towards the substrate side.

In this measurement, a 1.0-mm-diameter CCNS coated substrate was installed at the cathode. The emission current was increased gradually from 0 mA to 0.8 mA in 0.02 mA steps and at each step the CCNS was aged for about 1 h. Finally, the CCNS was aged for 10 min at an emission current of 1.6 mA (emission current density of 204 mA/cm²). Applying the electric field gradually in this aging process prevents sudden electrical breakdown which can damage the CCNS emitter and allows for subsequent stable emission.

The theory of field emission was proposed by R. H. Fowler and L. W. Nordheim [22] and typically the F–N equation is given by:

\[
\ln\left(\frac{I}{E^2}\right) = \ln\left(\frac{1.56 \times 10^6}{A \beta^2 / \phi}\right) - \left(\frac{6.83 \times 10^2 \times \phi^{1.5}}{\beta E}\right)
\]

where \(I\) is the emission current, \(E\) is the electric field, \(A\) is the effective emission area, \(\beta\) is the field enhancement factor, and \(\phi\) is the work function. A plot of \(1/E^2\) versus \(\ln(I/E^2)\) will produce a straight line, the so called F–N plot.

3. Results and discussion

The present I–V and F–N plots are shown in Fig. 3(a) and (b), respectively. The I–V characteristics were measured after the long term stability measurement (which was performed at a fixed emission current of 0.8 mA i.e., an emission current density of 102 mA/cm²), and shows the emission current up to 1.6 mA as a function of the applied voltage. In Fig. 3(a) the emission current density \((J)\) and electric field \((E)\) are also shown. The values for the F–N plot were converted from the measured I–V values according to Eq. (1) and this plot is shown in Fig. 3(b). The linearity of F–N plot shows that electrons are emitted by field electron emission from CCNS. In Fig. 3(b), the values of the slope
and intercept in the linear function of $I/E$ were determined in a least-squares fit analysis of the experimental F-N plot. The result from this analysis, after being converted back into the $I$–$V$ curve, is plotted as a solid line in Fig. 3(a). The values of the slope and intercept of the F–N plot are $-3.77$ and $-48.88$, respectively. Considering that CCNS has the same tip shape as CNT, the field enhancement factor of CCNS is calculated using the reported value for the work function for CNTs, i.e., 5.0 eV. The value of the field enhancement factor for the present CCNS is calculated as 1562, smaller than other CNT emitters for which values of between 2500 and 3980 were previously reported [8,9,14,15]. This lower value for CCNS may be explained by the loss of the most fragile CNT tips, which is expected to occur during the aging process. The remaining CCNS is stable but the field enhancement factor is reduced compared to CNT.

Fig. 4 shows the time dependence of the applied voltage at a constant emission current of 0.8 mA. In this long term stability measurement, a DC voltage was applied continually for 1308 h, except for a 2 day period at 915.5 h caused by a power outage. This long term emission stability measurement was performed immediately after the aging process. Initially the applied voltage decreased by about 5% in the first 200 h. This phenomenon is considered to be dependent on the pressure in the vacuum chamber. Although we did not measure the pressure below 1 x 10$^{-7}$ Torr directly, at the start of the stability measurement the vacuum quality is expected to be somewhat reduced due to outgassing during the aging process. The lower outgassing load during the measurement should cause the vacuum condition to improve resulting in improved emission performance. A similar effect can be seen after the 2 day stop at 915.5 h. After 200 h, the change in applied voltage was less than 3%. We have demonstrated high stability of CCNS at an emission current density of 102 mA/cm$^2$ for 1308 h.

A comparison of the present stability measurement for the CCNS emitter and similar measurements for various CNT emitters [5–17] is shown in Fig. 5. A graphene flower cloth emitter measured at an emission current density of about 0.4 mA/cm$^2$ by Iwai et al. [23] for an X-ray tube is also shown in Fig. 5. For printed CNT emitters measurements were typically performed at an emission current density of about 1 mA/cm$^2$ or less [5–11], although in recent years large improvements have been reported [12,13], with the most recent report by Kim et al. describing emission at 110 mA/cm$^2$ for around 100 h. Printed CNT emitters show very good stability at these low emission current densities, as measured by Furuta et al. [5] and Saito et al. [11]. Directly grown CNT emitters [14–17], including the present CCNS, typically report higher emission current densities of more than 1 mA/cm$^2$. However, for these emitters, long term stability has not been previously reported, with most measurements lasting only a few tens of hours. The longest measurement (200 h) at high emission current density (150 mA/cm$^2$) previously reported was measured at a low emission current density of 3 μA by Fujii et al. [14] using one CNT bundle with a diameter of 50 μm. On the other hand, the present CCNS emitter, with a similar current density, has a diameter of 1 mm and thus a correspondingly higher current (0.8 mA). The total current can be increased by increasing the size of the emitting area as the emission current is proportional to emission area. We have measured emission currents of more than 10 mA using larger area emitters [20]. Due to the combination of high emission

![Fig. 3. (a) $I$–$V$ characteristic and (b) Fowler–Nordheim (F–N) plot of CCNS after the long term emission stability measurement. The solid lines show the fit to the present data using Eq. (1).](image-url)

![Fig. 4. Long term stability measurement at the emission current of 0.8 mA (emission current density of 102 mA/cm$^2$) using a CCNSs of diameter 1.0 mm.](image-url)

![Fig. 5. Comparison of the previously reported stability measurements for printed CNT [5–13] (black ●), directly grown CNT [14–17] (blue ▲), and graphene [23] (green ▼), with the present result for CCNS emitter (red ○). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)](image-url)
current density and long term stability the present CCNS emitter is suitable for applications such as a high power electron source. We are currently developing compact, high power, X-ray sources for industrial applications using CCNS emitters.

4. Conclusion

In conclusion, we have measured the electron emission stability of a CCNS based field emission electron source. At a high current density of 102 mA/cm$^2$ the emission was stable over the 1308 h measurement time. Such long term stability at high emission current has not been reported for other CNT emitters. CCNS shows very favorable time. Such long term stability at high emission current has not been reported for other CNT emitters. CCNS shows very favorable characteristics and should prove useful for various applications involving high current density electron beams.

Prime novelty statement

We measured stable emission for 1308 h at a current density of 102 mA/cm$^2$ using a coniferous carbon nano-structure (CCNS) emitter as a field emission electron source. Long term stable emission at such a high current density has not been reported in a CNT based emitter previously.

References