# Emission site density depending on size and surface morphology of nanotube film emitters

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The influence of emitter surface morphology on emission site density (ESD) is investigated for carbon nanotube (CNT) films. We show that the ESD varies with cathode-anode distance for rough surface emitters while it is almost invariant for smooth ones. In addition, it has been revealed that the ESD decreases with the increase of the film size, resulting in low emission current density for large-size films. The present study suggests that the high ESD is more important to achieve high emission current density and long lifetime for film emitters than the high field enhancement factor.

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## I. Introduction

Since their discovery, carbon nanotubes (CNTs) have attracted intense interests in both research and industry, owing to their remarkable properties such as high aspect ratio, high electrical and thermal conductivity, strong mechanical strength, and high chemical inertness [1-3]. In particular, they are considered as a promising candidate for cold field emission (FE) sources [4-6]. Such sources exhibit extremely low power-consumption, which is ~0.14 W for an ionization vacuum gauge equipped with a CNT-film cathode, being a few percents of that for commercial ones [7]. Additionally, excellent emission current vs. applied voltage (*I*-*V*) characteristics and long lifetime have been achieved from CNT films [8-9]. The field enhancement effect of CNTs was extensively stressed in previous reports [10-12]. However, studies on emission uniformity and emission site density (ESD) are relatively rare although they are indeed more important especially for practical applications such as fabricating flat panel displays (FPDs) [12]. The influence of surface morphology on the *I*-*V* characteristics was investigated for CNT film emitters in our recent studies [13-15], and an empirical expression was obtained to describe their ESD. Based on this expression, we found that the ESD is influenced by the cathode-anode distance (gap) for rough surface films [14].

In this paper, we begin with a comparative study on the gap dependence of the ESD for film emitters with smooth and rough surfaces. No clear gap dependence of the ESD was found for smooth surface emitters, being different from rough surface emitters [14]. The film-size dependence of ESD is subsequently investigated, and it is found that an increase in the film size reduces the ESD, being responsible for low emission current densities for large-size films. The present study reveals that the high ESD is more important to achieve high emission current density and long lifetime for film emitters than the high field enhancement factor.

## II. Experiment

A commercial solution of CNTs made by an arc-discharge technique was selected in this experiment [13]. 70% of these CNTs are single-walled CNTs and the others are double-walled CNTs. Their diameters range from 1 to 3 nm. Several drops of the solution were put on a Ta-substrate covered with a thin Ti-film (2 μm in thickness), and a CNT film was formed by evaporating the solvent at ~80$^{o}$C. We then annealed the substrates to ~1000$^{o}$C in vacuum in order to form a TiC layer [13]. Figure 1(a) shows the surface morphology of a produced emitter, for which the surface roughness is < 10 μm. The length of CNTs protruding from the surface is ~2 μm as revealed by the inset of Fig. 1(a). Ten film emitters with sample size of ~1 mm$^2$ were prepared. In addition to them, five other emitters with relatively rough surface were prepared, as shown in Fig. 1(b), so as to clarify the surface-roughness dependence of the ESD. The detailed procedures for preparing rough surface emitters were given in the previous report [14].

An FE measurement setup with a phosphor screen was adopted in this experiment [14]. The cathode-anode gap was adjustable and varied between 0.1 and 0.8 mm. This setup was installed an ultra-high-vacuum chamber with a base pressure of $10^{-9}$ Pa. As for the FE measurements, emitters were firstly subjected to two-hour aging to stabilize the emission. Subsequently, they were measured at various gaps from 0.1 to 0.8 mm. After these measurements, we checked the FE performance at the gap of 0.1 mm to confirm the emission reproducibility.

## II. Results and discussion

Figure 2(a) shows the emission current density vs. applied field ($J$-$E_A$) curves for a typical smooth surface emitter at various gaps from 0.1 to 0.8 mm. An increase in the gap distance causes a clear shift of the $J$-$E_A$ curve to the low $E_A$ regime. The corresponding Fowler-Nordheim (FN) plots in Fig. 2(b) suggest that the field enhancement factor ($\beta_{FN}$) monotonically increases with the gap distance. The reason for this increase has been explained in our recent paper [13]. The good linearity of these FN plots is attributed to the sufficient aging of the emitter. Assuming 4.6 eV for the work function $\phi$ of CNTs [16], $\beta_{FN}$'s are derived from these FN plots. By using these $\beta_{FN}$ values, plots of $\{\ln(J/E_A^2) - 2\ln\beta_{FN}\}$ vs. $\frac{1}{E_A}$ at various gaps, hereafter being called modified FN plots [13-14], are produced in Fig. 2(c). All these modified FN plots possess a unique intercept with the vertical axis.

According to the FN theory, the intercept $B$ in the modified FN plot contains information of the total emission area of a film emitter. Our recent study has experimentally proved that the ESD ($n$) of the CNT film emitter is well described by

$$n = \frac{1}{A_{CNT}} e^{B - \ln C}$$
$$C = \frac{1.42\times10^{-6}}{\phi} \exp\left(\frac{9.89}{\sqrt{\phi}}\right) \qquad (1),$$

where $B$ is the intercept in the modified FN plots, $A_{CNT}$ the emission area of one site, and $C$ related to the work function $\phi$.

An appropriate $A_{CNT}$ value is necessary to determine the ESD value for film emitters. For emitters composed of capped MWNTs with diameter of >4 nm, our recent field emission microscope (FEM) study revealed that $A_{CNT}$ is almost constant with a value ~1.8 nm$^2$ [17], corresponding to the 6 pentagons and their neighbored area at a MWNT cap due to their local curvatures. However, this emission area cannot be applied to small-diameter (~2 nm) CNTs used in the present study. The previous study [18] revealed that small-diameter CNTs exhibits a uniform FEM pattern, suggesting that the electric field evenly distributes at the cap. We then consider that $A_{CNT}$ is roughly the tip area for such CNTs. By using a hemisphere approximation for the CNT cap, $A_{CNT}$ value is calculated to be 1.6 nm$^2$. Note that the lower limit of 1 nm for the CNT diameter range is used here because such CNTs have high field enhancement factors for a given CNT length and they should dominate the emission.

The solid circles in Fig. 2 (d) exhibits the ESD as a function of the gap distance for this smooth surface emitter, which is calculated from eq. (1). The gap dependence of the ESD is not apparent in the range from 0.1 to 0.8 mm. The emission patterns taken at various gaps are quite similar, and one is given in the inset of Fig. 2(d). It has been addressed that CNTs standing on the surface protrusion dominate the emission because of their strong local electric field [13]. Since the spacing (~ 20 μm) between these protrusions is quite large compared to the protrusion sizes (< 10 μm) (see Fig. 1(a)), the CNTs located at different protrusions give emission independently. Therefore, the ESD is almost invariable during the change in the gap distance.

In contrast, our recent study [14] revealed that ESD for rough surface emitters monotonically decreases with increasing the gap distance (*d*), showing high consistence with emission patterns taken at various gaps [14]. For an easy comparison, the ESD as a function of the gap distance is also given by the hollow stars in Fig. 2(d) for one film emitter with rough surface (See Fig. 1(b)). Note that $A_{CNT}$ = ~1.8 nm$^2$ is used for this film in the calculation of these ESD values since the corresponding CNT diameter is ~10 nm. The decrease in the ESD is ascribed to the increased screening effect among different emission sites during increasing the gap distance [14].

The variation in the ESD is actually expected to influence the lifetime of rough-surface film emitters. In order to explain this, the mean emission current per site in a film emitter $I_{mean}$ is introduced. For a given $J$ value for this rough surface emitter, $I_{mean}(d=0.8\text{ mm})$ is about 4 times of $I_{mean}(d=0.1\text{ mm})$ due to the decreased ESD. Therefore, the lifetime and emission stability should be reduced when this emitter works at large gaps due to the increased $I_{mean}$ value.

We further employed eq.(1) to investigate the influence of the emitter size ($S$) on the maximum emission current density of $J$. Some previous studies declared extremely high $J$ values up to > 100 A/cm$^2$ from $S$ <50x50 μm$^2$ [11,19]. However, such high $J$ values have never been achieved from film emitters with $S$ of several mm$^2$. In the present study, the maximum $J$ is about 1.5 A/cm$^2$ for $S$=1 mm$^2$. In order to investigate the reason, we tested five other rough surface emitters with $S$= ~0.1 mm$^2$, which produced a $J$ value up to 10 A/cm$^2$. Figure 3(a) shows the $J$-$E_A$ curves for two typical emitters with $S$=0.1 and 1 mm$^2$, respectively, which are obtained after sufficient aging of the emission. The inset of Fig. 3(a) shows the corresponding modified FN plots. Although there is a small difference (<5%) in their $\beta_{FN}$'s, a more obvious difference is attributed to their intercepts, and the derived ESD for $S$=0.1 mm$^2$ is about 5 times of that for $S$=1 mm$^2$. Based on eq. (1), the $I_{mean}$-$E_A$ characteristics for single emission sites, i.e., the $J$-$E_A$ curves divided by their corresponding ESD's, are derived for both emitters and shown in Fig. 3(b). These two curves are quite similar, and the corresponding maximum $I_{mean}$ are 0.9 and 0.8 μA, respectively, though the $J$-$E_A$ curves in Fig. 3(a) are very different. The high consistence between these two curves in Fig. 3(b) indicates that the FE properties of CNTs in both films are almost the same. It is reasonable since these two emitters were produced by the same procedures and the only difference is their film sizes.

The film-size dependence of the ESD is possibly due to the influence of the film edges, where the electric field is stronger than that inside the film. These edges may contribute a large proportion of the emission. Since the edge area does not increase proportionally with the film size, the ESD should decrease with the increase in the film size.

The field enhancement factor $\beta_{FN}$ of CNT films appears to be stressed overmuch in previous studies, and it was often considered as the key factor to achieve a high emission current density [10-12]. In fact, a higher $\beta_{FN}$ value suggests only a lower turn-on field to start emission while the upper limit ($I_{max}$) of the emission current induced from one CNT is indeed determined by its intrinsic properties. For CNTs with a given diameter, $I_{max}$ decreases clearly with the increase in the CNT length (or $\beta_{FN}$) due to damages caused by Joule heating [20], indicating that high $I_{max}$ values cannot be expected from large $\beta_{FN}$'s. In contrast, the ESD is directly responsible for the emission current density for CNT films; the higher the ESD, the higher the $J$ values. Therefore, effective methods to increase the ESD are the key issue to achieve high $J$ values for film emitters. However, it should be noted that the sample size strongly affect the ESD and it is not surprising to achieve high $J$ values up to 100 A/cm$^2$ from an sample size of ~50 x 50 μm$^2$ [11,19]. Hence, standardization of the film size is necessary to universally estimate the ESD and $J$ values for different CNT films. In addition, smaller

$I_{max}$ protects emission sites from damages induced by Joule heating. Therefore, high ESD values are expected to benefit the lifetime of film emitters for a given $J$ value.

## IV. Conclusion

We have carried out a comparative study concerning the dependence of the emission site density (ESD) on the cathode-anode distance for nanotube film emitters with different surface morphology. It was found that film emitters with smooth surface exhibits almost a constant ESD independent of the gap distance, resulting in a unique emission pattern. For rough-surface emitters, on the other hand, the ESD is reduced with the increase in the gap, which is expected to shorten their lifetime and to decrease the emission stability. In addition, it is experimentally proved that the ESD is remarkably reduced by increasing the film size, responsible for low current densities obtained for large-size emitters. This result reveals that the maximum emission current densities for CNT films are determined by their ESD's instead of their field enhancement factors. Finally, high ESD's are also expected to benefit the lifetime of emitters.

## V. Acknowledgement

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Figure captions:

Fig.1 (a) SEM images of a film emitter with a smooth surface (Surface protrusion height: <10 μm). The inset shows the CNT length of ~ 2 μm. (b) SEM images of a film emitter with a rough surface (Surface protrusion height: ~ 50 μm). The inset shows the CNT length of ~2 μm.

Fig. 2. a) Emission current density (*J*) vs. applied field ($E_A$) characteristics for a smooth-surface emitter at various cathode-anode gaps. b) The corresponding Fowler-Nordheim (FN) plots. c) The corresponding modified FN plots with a unique intercept. d) Solid-circle curve: The emission site density (ESD) as the function of the gap distance for this emitter. The emission pattern (See the inset) is almost invariant during the change in the gap. Hollow-star curve: The emission site density (ESD) as the function of the gap for a rough-surface emitter. Both the ESD and emission pattern varies with the gap (See Ref.[14]).

Fig. 3. $J$-$E_A$ characteristics for typical emitters with sample sizes (*S*) of 0.1 $mm^2$ and 1 $mm^2$. The inset shows the corresponding modified FN plots, suggesting that the main difference is caused by the intercept, i.e., the ESD. b) Mean emission current ($I_{mean}$) vs. $E_A$ for one site (or CNT). No large difference is found for these two film emitters.

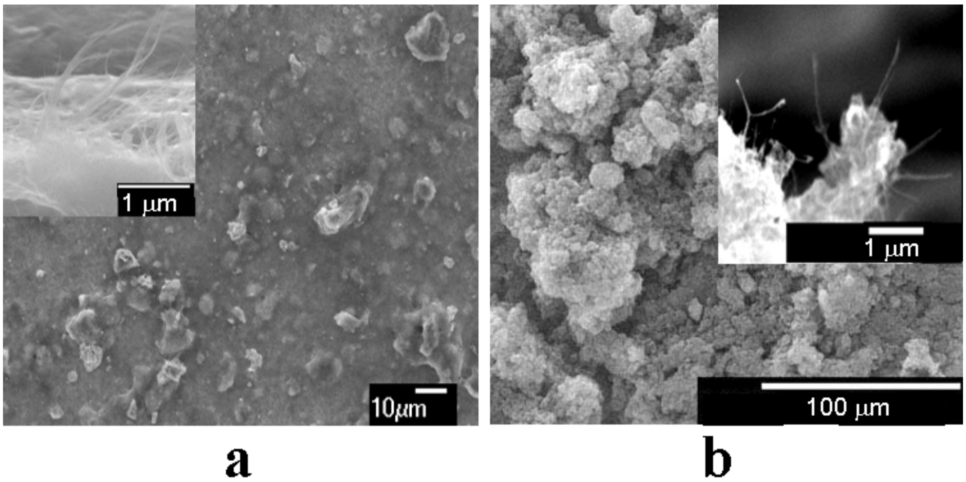
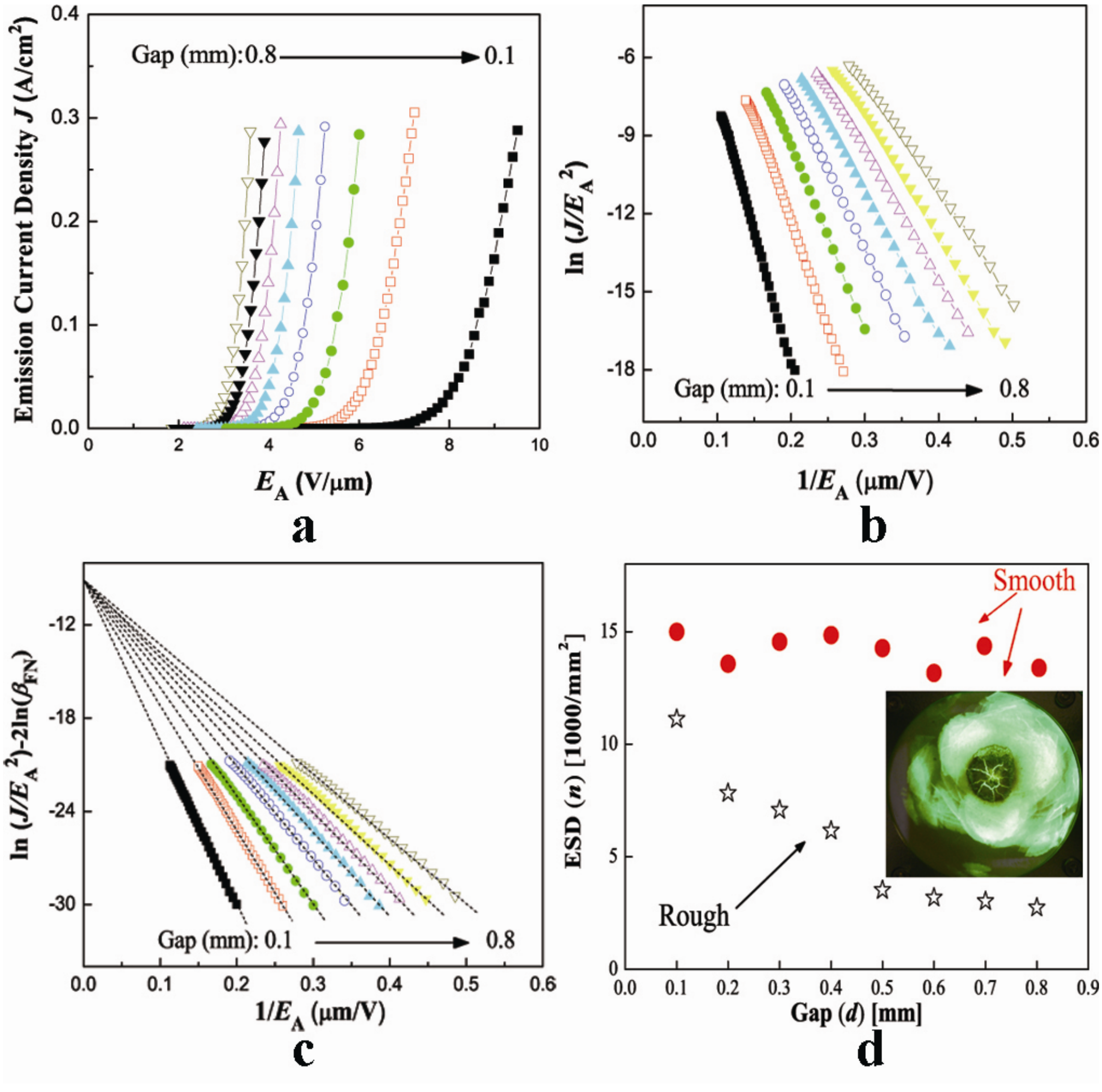



Figure 1



Gap (mm):0.8 → 0.1
Emission Current Density J (A/cm2)
EA (V/μm)
a
ln (J/EA2)
Gap (mm): 0.1 → 0.8
1/EA (μm/V)
b
ln (J/EA2)-2ln(βFN)
Gap (mm): 0.1 → 0.8
1/EA (μm/V)
c
Smooth
Rough
ESD (n) [1000/mm2]
Gap (d) [mm]
d


Figure 2

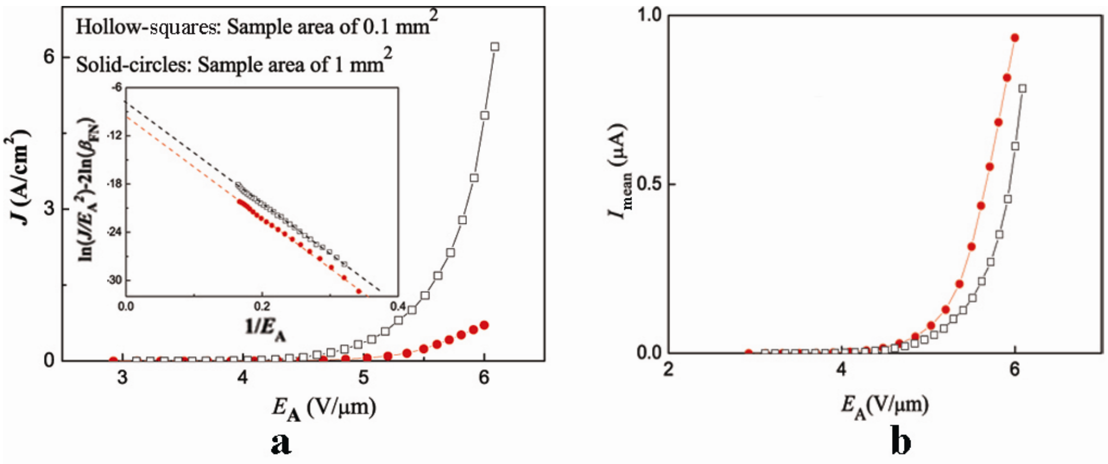

Hollow-squares: Sample area of 0.1 mm2
Solid-circles: Sample area of 1 mm2
J (A/cm2)
E_A (V/μm)
ln(J/E_A2)-2ln(β_FN)
1/E_A
a
I_mean (μA)
E_A(V/μm)
b

Figure 3