Review on development of carbon nanotube field emission cathode for space propulsion systems

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Abstract: In recent years, small-size, low-weight aerospace propulsion systems have developed rapidly for space exploration, on-orbit scientific instruments and extra space missions, while additional electron emission devices are commonly required in those propulsion systems. Carbon nanotube (CNT) based field emission cathodes exhibit extraordinary field emission properties and are regarded to be an ideal alternative of conventional thermionic or hollow cathodes. In this study, the authors give an overview of present status of researches on CNT-based electron emission cathodes for utilising as neutralisers or electron sources particularly in space electric propulsion systems, the theory and characteristics of CNT are also illustrated. Furthermore, challenges, problems and possible solutions before actual applications of CNT in a space mission are discussed accordingly.

1 Introduction

With the rapid development of space technology, the scope of mankind's space exploration is extensively expanding. Yet the increasing demand of deep space flight cannot be fulfilled by mere means of conventional chemical propulsion, which calls for new power sources including electric propulsion. Electric propulsion technology has significant advantages over traditional chemical propulsion in deep space flight due to its characteristics of high specific impulse, small size, long life and small thrust. Over the past decades, numerous types of electric thrusters have been developed and used in space missions worldwide [1].

Electric propulsion can be mainly divided into three classes: electro-thermal, electrostatic and electromagnetic. Among them, Hall type and gridded ion thrusters are the most advanced, mature technologies. Usually, these thrusters need electron sources, such as hollow cathodes to provide primary electron in the discharge chamber of an ion thruster, radio-frequency (RF)/microwave discharge neutralisers to supply free electron to neutralise ions in the plume. In addition, small spacecraft and satellites (e.g. Cuestas) have attracted increasing attentions. As stated recently by Levchenko et al. [2], small spacecraft or satellites are very efficient on performing remote earth sensing, precision agriculture, monitoring erosion and sea pollution, precision weather prediction and so on. In addition, small spacecraft or satellites could also efficiently be used for exploring our solar system and beyond. On the other hand, the increasing market of small spacecraft proposed even strict request on low-weight, low-power consumption electron sources.

Most of the previous electron sources are the thermionic or hollow cathodes offering a higher electron current [1, 3–5]. However, to generate free electrons, a heated device is needed, leading to power consumption and thermal load. In addition, working gas requirement also results in a heavy mass and complex support system, which brings further burden in space missions. Field emission cathodes (FECs) provide a promising alternative, due to the following benefits: (i) no propellant is consumed in FECs, and the whole system-specific impulse can be increased; (ii) reduction of the propulsion system power requirements. In particular, carbon nanotube (CNT) based FECs have become popular in last decades and have demonstrated numerous applications [6, 7] in X-ray imaging tools [8, 9], RF sources [10], compact mass spectrometry [11], flat-panel displays [12–14], electron-beam systems [15], vacuum tubes [6], lighting devices [16], amplifiers and electric propulsion devices [2]. Many efforts have been paid to summarise numerous potential applications of CNT-FECs and further details are presented in several previous reviews [6, 7, 17, 18]. Particularly, Levchenko et al. [2] presented a very detailed review on the progress of space electric propulsion systems on smart nanomaterial, they marked that nanotechnology is very useful for future advanced, adaptive spacecraft. In this paper, our aim is not to make our scope of discussions too broad, but to focus on the progress of CNT-FECs applications in particular on space electric propulsion systems, for use as a neutraliser or an electron source.

The structure of the paper is as follows. In Section 2, the theory of field emission and characteristics of CNT emission are illustrated. Then, we summarise the status of researches on CNT-FECs applications in space propulsion systems, especially, neutralisers or electron sources, in Section 3. In addition, potential challenges and problems before practical application in space missions are discussed, future development of CNT-FECs is also predicted. Finally, conclusions are drawn in Section 4.

2 Descriptions on FEC

2.1 Field emission theory

To release electrons that are trapped inside a solid material (e.g. a semiconductor or a metal), two methods can be used: one is adding energy to trapped electrons and the other is decreasing the height and width of the solid surface potential barrier. In past, four ways were frequently attempted, which are thermionic emission, photoelectron emission, secondary electron emission and field emission. In a thermionic emission, the solid material is heated to a high temperature (>1000°C), so the trapped electrons can have sufficient energies to overcome the potential barrier of the material. Photoelectron emission uses light or electromagnetic radiation to pass energy to trapped electrons, while differently, secondary electron emission hits the surface of solid material with energetic electrons or ions. Field emission (the electric field threshold is in the order of 10\textdegree V/m) takes advantages of sharp (can be nano-
Fig. 1 Effects of an external electric field on the energy barrier for electrons at a metal surface. The figure is reproduced from [17]

Fig. 2 Schematic of a Spindt-type array cathode

scale) electrodes to enhance the local electric field, comparing to other three methods, field emission process does not transfer energy directly to trapped electrons inside the material, but through deforming the potential barrier on the material's surface.

In the field emission case, when an external electric field is applied, the potential barrier varies with applied field. As shown in Fig. 1, the potential is represented as a simple plane with the electron requiring sufficient energy to escape from the material, this energy names the work function of the material usually represented in electron-volts (eV). The dash line shows the potential barrier without an external electric field, only trapped electrons with energy higher than work function ($\phi = 4.5$ eV usually) can escape from the metal surface through thermionic emission. In case a large external electric field (usually in an order of $10^9$ V/m) is applied, the width and height of potential barrier decreases to the order of electrons' wavelength, thus, cold trapped electrons near Fermi level ($E_F$) can escape into the vacuum by so-called 'quantum tunnelling' effect, hence, field emission occurs [19–21].

The fundamental physics of field emission is relatively well investigated and understood; the best representation is given by Fowler and Nordheim in 1928 [19]. The well-known Fowler-Nordheim equation is described as [19, 22]

$$J = \frac{e^2 E^2}{8\pi h \phi^2 \sqrt{2m}} \exp \left\{ -\frac{8\sqrt{2m} \phi^{1/2}}{3\hbar E} v(y) \right\},$$

$$y = \frac{e^2 E}{2 \pi \hbar \phi},$$

where $J$ is the current density, $\phi$ is the work function of material, $m$ is the electron mass, $E$ is the electric field, $h$ is the Plank's constant, $e$ is the electron charge and $\hbar$ is the permittivity of free space. The functions $t(y) = 1 + 0.1107 y^{1.55}$ and $v(y) = 1 - y^{1.50}$ can be approximated [22]. Assuming there is a triangular surface potential barrier, insert them and constants into (1), we obtain

$$J = A \beta E^2 \exp \left\{ -\frac{B \phi^{3/2}}{\beta E} \right\},$$

In which, $\beta$ is the field enhancement factor, $A = 1.56 \times 10^{-6}$ A V$^{-2}$ eV, $B = 6.83 \times 10^3$ eV$^{-3/2}$ V$^{-1}$

2.2 CNT-based field emission characteristics

Through aforementioned descriptions in Section 2.1, we note that a large electric field is required to trigger field emission process. In last decades, a usual way to obtain a high electric field was to use very sharp cathode with a tip diameter of a few hundred nanometres, with the help of micro- and nano-fabrication technology developments. Thus, a strong field enhancement can be obtained at a sharp tip electrode and hence the external trigger and extraction electric voltage can be reduced to a few hundred volts or even less. From 1960s, Spindt tip cathodes have become the most popular and successful cathodes for field emission, as shown in Fig. 2. Typically, molybdenum or tungsten were used to fabricate tip emitters because of their high melting point, and eventually with the development of micro-fabrication technology, silicon tip emitters dominated and Spindt array FECs became more and more used worldwide [23–25]. Up to now, the maximum emission current from a single tip can reach 1 mA and the maximum current density is higher than 2000 A/cm$^2$ for Spindt type FEC [26]. However, with the increasing applications of Spindt-type cathodes in vacuum electronic devices, intrinsic weak points of this type of emitters became highlight gradually and led to fails of electron emission, in particular, in complex plasma environments. These fails include, emission efficiency is highly dependent on sharpest tip and decreases with time, uniformity of electron emission becomes low, and frequent catastrophically arcing causes high temperature that can damage the whole device [27].

CNT was first discovered in 1991 by Iijima [28], single-walled CNT and multi-walled CNT were classified according to the number of graphene layer [29] (see Fig. 3). A single-walled CNT is a graphite sheet that is rolled into a cylinder of a few micrometres in length and a few nanometres in diameter. A multi-walled CNT consists of several single-walled CNTs nested inside each other. One carbon atom is bound to three other carbon atoms, thus, the activation energy for surface migration of the emitter atoms is very large and can undertake the intense external electric field that is needed for field emission [18]. Electron field emission under low trigger electric field and high current density from CNTs was reported in 1995, by three research groups [30–32], from then on, CNT-based electron field emission characteristic and applications on various fields have been investigated extensively in the world.

Fig. 4 shows a conceptual design of a CNT-based FEC, similar as a Spindt-type field cathode in Fig. 2. Electrons are extracted from tips of CNTs, the curvature of tips can be much smaller than conventional sharp metal tips. Comparing to other type of field emitters, CNT presents the following properties [31, 33]: (i) excellent conduction, (ii) extrem high temperature, (iii) Young modulus and maximal tensile strength (∼200 GPa); (iii) low sputter coefficient, which is very benefit as an electron source that multiple particles can bombard to; (iv) chemically inert, which is hard to react with, except under extreme conditions or high temperature; (v) nano-scale high aspect-ratio (can reach 10$^5$), which leads to strong field enhancement at tips to facilitate field emission. In general, chemical vapour deposition (CVD) and arc discharge method are frequently used for CNT synthesis [34, 35]. Especially, arc discharge method contributes to CNT with low defect, improving the field emission characteristics. Note that Fowler-Nordheim theory presented in Section 2.1 can partially describe the field emission process from CNTs field emitter at a certain current range, but fails to explain the emission threshold of CNTs is lower than conventional metal emitters, thus, a complete theory of CNT field emission is still under investigation.

3 Recent progresses of CNT-FEC applications on space propulsion and discussions

In 1995, the field emission characteristic of CNT was reported as stated in Section 2.2. From then, numerous applications of CNT-FEC have been attempted worldwide. Interest of using CFN-FECs into space propulsion systems started around 2000 [37, 38], in this section, we specifically summarise the development of two typical applications: neutraliser or electron source of potential electric propulsion systems.
3.1 CNT-FEC serves as a neutraliser of space propulsion systems

In 2001, Marrese-reading et al. [39] from JPL demonstrated charge neutralisation of a field emission electric propulsion (FEEP) thruster with three electron sources: a mixed metal thermionic cathode, a CNT-FEC and a Spindt-type molybdenum field emission array cathode. The experiments demonstrated that CNT-FEC can significantly decrease the power-to-thrust ratio by 50% under their test environment, which was very inspiring at that time. Configuration, microstructure and assembling of CNT cathode in JPL’s FEEP thruster system are shown in Fig. 5. The FEC was grown on a silicon base with a CVD process, which demonstrated 160 μA at applied voltage of 380 V on the gate electrode with 1.3 mA collected by the gate electrode. The efficiency of CNT-FEC is better than thermionic cathode at μA current level, but showed an unstable performance during test since it is more sensitive to the environmental conditions because of ion sputtering process generated near the tips.

In 2007, Takao et al. [40] have carried out experiment to check possibility of a CNT-FEC as a neutraliser for a small-scale microwave discharge ion thruster. They fabricated two types of CNT-FECs. As shown in Fig. 6, the first type is single-walled CNT made at Toyohashi university, the copper plate with a cylindrical hole filled with single-walled CNTs (diameter of 2 mm and depth of 0.5 mm); the second type is a graphite disk (diameter of 2 mm) that is dug into the copper plate. Experiments presented that a 0.56
mA electron current can be extracted with single-walled CNTs, at applied voltage of 380 V and electrode interval of 0.5 mm. Note that electrode interval strongly affects the extracted electron current during tests, indicating that single-walled CNT cathode is very applicable to the neutraliser for the small-scale microwave discharge ion engine, but the applied voltage can be reduced because of the energy loss. In addition, graphite disk cathode demands much larger applied voltage than single-walled CNTs, which is higher than 1500 V, as shown in Fig. 7. This is because graphite disk was not treated with arc discharge.

Velasquez-Garcia and Akinwande [41] from MIT reported a test on MEMS CNT-based neutraliser for micro-propulsion applications, in 2007. The CNT-FEC is fabricated with the help of plasma enhanced chemical vapour deposited technology, which allows them to set a small and uniform emitter-to-grid interval, thus to reduce the required voltage for electron emission. I–V characteristics of the emitter were obtained using a triode configuration, as shown in Fig. 8, a maximum gate current of 2.1 mA was measured at applied voltage of 375 V. Their experiments also confirmed that Fowler-Nordheim theory could basically describe the electron emission from the CNT substrate.

In 2014, Singh et al. [42] have tested a CNT field emission array in a Hall thruster plume environment, as shown in Fig. 9, most CNT field emission arrays were installed in the furthest radial locations from the Hall thruster (the centre) to capture the effects of plasma plume, with minimising the risk of damaging. The CNT field emission arrays were exposed in this environment for 40 min, field emission showed no influence by the plasma because of the physical structures, which indicated that a refined design of CNT field emitter can be an alternative for conventional thermionic cathode for Hall thrusters. Although present study shows that no fundamental incompatibilities exist of CNT field emitter in the plume, coupling between CNT field emitter and the Hall thruster itself should be further explored in the future.

Recently, in 2019, Yamamoto et al. [43] have further investigated the demonstration of the CNT-FEC as neutraliser for microwave ion thruster. The schematic and setup of CNT-FEC is shown in Fig. 10. The size of FEC is 88 × 88 mm² and the maximum emission current is 20 mA at potential difference of 500 V between gate grid and emitter. The CNT-FEC was mounted at 20 cm from the ion thruster, as shown in Fig. 10b. It confirmed that the emission current from FEC is well balanced with the ion beam current from the thruster chamber. The weak point is the electron emission cost is very expensive that reaches 360 W/A, much higher than a conventional hollow cathode (≤30 W/A). However, the total specific impulse of the system is 1.25 times higher than a conventional ion thruster system with their own microwave discharge neutralisers, in addition, remark that no propellant is required for a CNT-FEC, which is very competitive.

In China, although a great number of investigations on CNT-FEC have performed, only very few reports can be found...
concerning on space propulsion systems application, to the best of our knowledge. In 2017, University of Electronic Science and Technology [44] has reported their study on CNT emitters works as a neutraliser of electric propulsion system, while growth of CNT emitters, design and optimisation the neutraliser's electronic optics system were explored, in addition, field emission properties of CNT-based neutraliser were measured in lab, with an emission current of 473 mA and the grid transmission rate is about 60%. Furthermore, in 2018, Li et al. [45] reviewed the researches on preparations and applications of CNTs in Lanzhou Institute of Physics, the experimental method for fabricating CNT films, the field emission characteristics of different CNT-FECs and recent applications of CNTs on ion thrusters were summarised and analysed.

### 3.2 CNT-FEC serves as an electron source of EDT

From 2010, a group from Japan Aerospace Exploration Agency (JAXA) started their demonstration on CNT-FEC as electron sources of an electrodynamic tether (EDT), for the purpose of testing active removal of debris in the low earth orbit (LEO) [36, 46]. They reported that the reasons for choosing CNT-FEC are, low extraction voltage, high electron extraction efficiency and durability. First, a laboratory model of 0.5 mA-class CNT cathode was designed, fabricated and tested. Effect of emitter-gate separation distance, width of slit apertures and performance variability during different periods of operations were investigated in detail, as shown in Fig. 11. Under their selected configuration, the desired emission current of 0.5 mA can achieve with an applied voltage of 600 V and a drain current is less than 0.05 mA. Then, a further endurance test of a breadboard model of CNT-FEC was conducted, at a constant emission current density of 5 mA/cm². The pressure in the vacuum tank can be pumped to 5 × 10⁻³ Pa. The experiment showed that lowering the emission current density helps extending the FEC life in 50 h test, and no severe breakdown and short-circuit were observed during 1500 h endurance test. Remark that the FEC life might be even longer than 1500 h on real orbit because of the lower gas density environment.

In 2017, an on-orbit demonstration of CNT-FEC of EDT is conducted by JAXA, on H-II transfer vehicle 6 (HTV-6), in Japan, the mission is called Kounotori Integrated Tether Experiment (KITE). Note that HTV-6 was launched in December 2016 and KITE began in January 2017 after HTV-6 left the International Space Station (ISS). The operating orbit during the mission was around 370 km.

To supply electrical current through tether, eight CNT-FECs were designed from the beginning of 2013 and were completed in the spring of 2016. As shown in Fig. 12, since the tolerance of CNT-FEC to atomic oxygen (AO) is very low, the electron emitter surface of the flight model was set parallel to the orbital motion axis to avoid the direct impact of AO flux. JAXA's experiment showed that all eight CNT-FECs worked very well in the orbit; the total operation time is 50 h in the total exposure time of 130 h. The measured maximum 'emitter current' (electron current emitter from the CNT emitters) and 'emission current' (the electron current extracted to the outside) are 10 and 6 mA, respectively [47, 48].

In addition, I–V characteristics of a single FEC obtained at various stages were recorded, as shown in Fig. 13. It was found that almost no performance change occurred after 10 and 60 h exposure, indicating that CNT-FEC is at least able to work in AO irradiation and plasma condition of 10¹⁸ atoms/cm². Remark that current experiment was performed at 370 km, where the residual neutral gas density is much higher than normal orbit of satellites (around 600–1400 km). Further comparison showed after 125 h exposure with 5 h direct impact, the performance of CNT-FEC decreased a lot due to AO impact and erosion, which needs to be avoided in future low orbit missions [48].

#### 3.3 Discussions

CNT-FEC as a promising electron source has been well demonstrated to be an ideal candidate of neutraliser or electron source in space electric propulsion systems. Although plenty of theories, simulations and experiments have been attempted, many aspects still are not fully understood and further efforts are required before using CNT-FEC as a common electron source in space propulsion system. In this section, we propose some existing problems and possible suggestions, further potential development of CNT technology on space missions is also predicted.

(i) **Theoretical challenge:** In Section 2.1, we illustrated that well-known Flowler-Nordheim theory has been commonly used to
describe field emission process of CNT. In fact, many assumptions were used for Fowler-Nordheim theory, as (i) electrons obey Fermi-Driac distribution, (ii) surface of the metal is smooth enough without considering roughness, (iii) potential barrier is induced by the mirror charge force [44]. As we know, the size of CNT is in nano-scale, electrons are mainly emitted from its sharp tips, thus the smooth assumption of Fowler-Nordheim theory is not precise anymore for CNT emitters. A new theory has to be further investigated to express the field emission process of CNT better, although Fowler-Nordheim theory can be used as a verification tool of experimental tests at a certain current range.

(ii) Space charge limit: Due to the limit of techniques, a single CNT is very hard to fabricate, instead, a bunch of CNTs is fabricated meantime, which leads to so called space charge limit. Under the same environment, the length of two adjacent CNTs can be different, thus only few longest CNTs would emit electrons after an external strong electric field is applied, which affects the emission efficiency and uniformity.

This problem should be solved in the stage of fabrication. In past, many approaches have been used to fabricate CNTs, such as patterned growth of CNT bundles by CVD on the semiconductor or metallic substrates and patterned electrophoretic deposition of CNT-based inks. Or some recent approaches, such as, by screen printing [49], by assembling arrays from premade [50] and so on.

(iii) Extraction efficiency: Beyond emission efficiency, high extraction efficiency is also aimed to further reduce power consumption and suppress thermal load. As shown in Fig. 14, because of the electron current loss to gate, extraction efficiency of the conventional CNT-FEC is not high. Ohkawa et al. [51] proposed a geometrical treatment to decrease the electron loss to gate, by assembling multiple emitters locating under the emission holes, or by covering unnecessary CNTs area under the gate with proper masks and to make electron trajectories converge by distorting the field, as shown in Fig. 14.

(iv) Low tolerance to AO: Due to the vacuum limit, residual gas exists in chamber during ground test of CNT-FECs, furthermore, this is one of the critical issues when the CNT-FEC is used in LEO. We should avoid AO flux impinge the emitter surface directly. This could overcome by coating the surface of the CNT with AO resistant material, or avoiding direct exposure to AO.

Indeed, significant difficulties and constraints are expected during implementation of CNT-FECs to a space propulsion system, and not all the thruster systems are suitable for such a new technology. However, aiming at the potential benefits, it is a worthwhile effort because CNT technology has a large possibility to revolutionise the whole space propulsion system, as a cathode with long life, high current emission and no propellant requirement via advanced material and technology. Furthermore, the NASA's 2015 nanotechnology roadmap presented a so-called self-healing spacecraft concept, which includes self-healing field mission neutralisers, self-cleaning surfaces, self-healing repair mechanisms...
and so on [2]. CNT technology would benefit space propulsion system greatly while aforementioned challenges are solved eventually with the development of industrial and manufacturing level.

4 Conclusions

CNT-FECs clearly have many interesting properties, such as low voltage operation, long lifetime and good emission stability. Numerous applications of CNT-FECs have been attempted in various research fields in both academy and industry. In this review, we summarise recent tremendous progress of CNT-FECs as a promising candidate electron source for space propulsion mission, in particular, serving as a neutraliser of charges in various electric propulsion systems or an electron source for EDT systems. Moreover, we present the existing problems, challenges of using CNT-FECs in real space missions, and possible solutions are advised accordingly. Although, aforementioned several hurdles and challenges exist, a worthwhile effort on CNT-FECs is deserved since it can benefit the development of space propulsion much in the near future, and the current progress and technology development have already proved it is conceptually available.

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