Regenerative Field Emission Cathodes: Surface Morphology and Performance

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This paper reports on a field-emission cathode for use in electric propulsion that has the potential for very long lifetime due to its ability to be regenerated when the emitter tip(s) become damaged. The field-emitting tips were formed by applying an ion-extracting electric potential to a heated indium-coated tungsten needle, known as a liquid-metal ion source. The liquid-metal ion source was then cooled, freezing in a solid nanotip at the apex. When the modified emitter was then subjected to electron-extracting potentials, stable and long-lived electron emission was observed. The first goal of this investigation was to operate and quench a liquid-metal ion source at ion emission currents from 1 to 30 μA to acquire micrographs of the surface morphology as a function of the ion emission current at quench. Micrographs of the quenched emitter tips revealed Taylor-cone-shaped structures. Some of the quenched emitters exhibited multiple nanoprotuberances on the tapered surface of the microscale Taylor cone, which were capable of electron field emission. The second goal of this investigation was to compare regenerative field emitters with single-needle tungsten field emitters. Each type of emitter was used to obtain electron emission at a vacuum chamber pressure of 10⁻⁴ Torr, and then the emitters were exposed to increased pressure, up to 10⁻³ Torr, to observe how long they could sustain emission. In all cases, emission from the regenerative emitters lasted 10s of hours longer at increased pressure, and it was demonstrated that the tungsten emitters would eventually permanently fail, whereas the regenerative emitters could be repaired when they became damaged.

Nomenclature

\[ \begin{align*}
\alpha & = \text{Fowler–Nordheim empirical relation} \\
b' & = \text{Empirical relation between tip radius and gap spacing} \\
k & = \text{Fowler–Nordheim field-voltage proportionality factor} \\
m & = \text{slope} \\
r_t & = \text{emitter tip radius} \\
s_m & = \text{standard error} \\
t_e & = \text{elapsed time} \\
\alpha & = \text{Fowler–Nordheim image-correction factor} \\
\varphi & = \text{work function}
\end{align*} \]

I. Introduction

Historically, liquid-metal ion sources (LMISs) have found extensive use as ion sources of high brightness in focused ion beam materials processing applications [1] and, more recently, as electric propulsion (EP) thrusters via field-emission EP (FEEP) technology [2-4]. In an LMIS or FEEP thruster, an intense electric field is created near the surface of a low-melting-temperature liquid metal, such as indium, by a downstream electrode. A balance between the liquid surface tension and electrostatic forces causes a structure known as a Taylor cone to form in the liquid [5]. Because the Taylor cone has a very sharp tip, geometric enhancement of the local electric field is achieved at the cone tip. The electric field enhancement enables the electric force on the liquifed metal to overcome the surface tension force that holds the liquid together. When the electric field becomes great enough, a beam of charged particles can be directly extracted from the apex of the liquid Taylor cone. The charged particles emerge from a very narrow (femtometer diameter) liquid jet at the cone apex and are subsequently accelerated by the electric field either to produce thrust (FEEP) or for materials processing applications (LMIS). Other applications and areas of interest for the use of focused ion beams include lithography, semiconductor doping, sample preparation for transmission electron microscope imaging, circuit repair, scanning ion microscopy, and scanning ion mass spectroscopy [6].

For low-power EP applications (e.g., FEEP systems), an electron source is a necessary thruster component to maintain spacecraft neutrality since an operating thruster will cause a global charge imbalance on a spacecraft. Typical electron sources, or cathodes, that have been proposed for use with low-power thrusters include field-emission cathodes. Field-emission cathodes use nanoscale sharpened electrodes with locally enhanced electric fields to cause electrons to escape from the surface of the electrode into vacuum via a quantum tunneling effect known as Fowler–Nordheim (F–N) emission. The local electric field is inversely proportional to the electrode tip radius, so the sharper the emitter tip, the lower the electric potential needed to obtain electron field emission. Many types of electron field emitters have been used in the past, with the most popular being the Spindt-type array [7] and, more recently, carbon nanotube field-emission arrays [8,9]. A significant weakness with field-emission cathodes is the limited lifetime associated with the devices. The nanoscale or microscale features are fragile, and when the features become damaged, the electron source loses functionality. Researchers have found some ways to minimize damage to the emitters [10], and they have also found more spatter-resistant longer-life emitter materials [8,11-13]. However, all electron field emitters become damaged over time [14,15]; it is just a matter of how much time it will take. Some of the mechanisms that cause damage to field emitters include arcing between the emitter and the extraction electrode, localized heating causing emitter tip melting/destruction, and sputtering of the sharp emitter tips.

In 2007, Makela and King [16] proposed and demonstrated a technique for regenerating solid-metal field-emitter tips using a liquid-metal ion source [17,18]. The LMIS was constructed of an electrochemically etched tungsten emitter that was coated with a thin layer of the low-melting-temperature metal indium. Ion emission was established and simultaneous formation of a Taylor cone structure, by heating the emitter tip and applying an ion-extracting
field between the emitter tip and an extraction electrode [19]. By cooling the LMIS during ion emission, Makela and King [16] were able to solidify the metal Taylor cone into a permanent solid structure. This quenched Taylor cone could then be repurposed as an efficient electron emitter for space applications. Makela and King demonstrated the feasibility of creating electron field-emitting tips by quenching the ion-emitting LMIS at emission currents ranging from 1 to 30 μA. It was shown that sharp nanostructures could be regenerated as long as there was a sufficient supply of indium to obtain ion emission. It was also found that the electron current-voltage (I-V) characteristics of a field emitter could be altered by changing the ion emission current during the quench (quench current). The results of those experiments showed that as ion emission current during quenching was increased, the subsequent electron emission performance increased, inferring that the resulting solid emitter tip radii decreased. Applying the Fowler–Nordheim model to the electron I-V data yielded radii ranging from 230 to 80 nm at quench currents of 1 to 30 μA, respectively [17,18,20].

The goal of this paper is to further explore the formation process and the durability of frozen nanotips used for electron field emission when grown using Makela et al.’s technique [17]. The research reported here has two primary objectives:

1) Use electron microscopy to understand how the surface morphology of frozen nanostructures depends on the ion current that was emitted during the quench process.

2) Quantify the lifetime and performance degradation of frozen nanotip electron emitters when operated at ambient pressures above 10^{-8} torr.

II. Description of Apparatus

The emitter fixture that was used for the reported experiments was designed to be operated within the specimen chamber of a field-emission scanning electron microscope (FE-SEM) to perform in situ experiments within the FE-SEM. The indium-coated LMIS was constructed from a Y-shaped tungsten wire configuration, as shown in the electrical schematic in Fig. 1. Two legs of the tungsten wire were connected to a power supply to provide resistive heating to liquefy the indium. The remaining end of the tungsten wire was electrochemically etched in a NaOH solution and subsequently coated with indium using the process described by Makela et al. [17]. The fixture was also equipped with an electrical connection for the extraction electrode to provide the necessary potential to operate the LMIS.

For vacuum compatibility, the materials that were used to build the custom LMIS and support fixture included Teflon, stainless steel, and tungsten. Gap spacing between the emitter tip and the extraction electrode was ≤ 0.5 mm for all of the experiments. During the surface morphology testing, the LMIS was operated in the specimen chamber of the FE-SEM, and during the lifetime comparison testing, the LMIS was operated in an ultrahigh vacuum (UHV) system. The specimen chamber in the FE-SEM was evacuated using a series of three ion pumps, and pressure of 10^{-7} torr was maintained throughout testing. The UHV chamber is approximately 0.5 m in diameter by 0.5 m deep and has a base pressure of 10^{-9} torr, which is achieved by pumping with a single 280 liter/s turbomolecular pump that is backed by a 110 liter/min dry scroll pump. The UHV chamber is also equipped with a 300 liter/s combination ion-sublimation pump to reach UHV.

III. Experimental Procedures

To achieve ion emission, the resistive emitter heater, shown previously in Fig. 1, was supplied with 2.75 A, 1.3 V for the purpose of maintaining the indium-coated electrode above the melting temperature of indium, which is 156.6°C. Although the precise temperature of the emitter tip was unknown, the tip temperature was likely between 156.6 and 200°C since the heater power required for emission was determined by first applying a constant extraction potential of several kilovolts while slowly increasing the heater current until stable ion emission could be achieved. The method of first applying an extraction potential and then increasing heater current allowed for charged particle emission at the lowest possible temperature. After the heater I-V characteristics were determined, the heater supply could be returned to 2.75 A, 1.3 V for each subsequent test.

Once the heater power was set and emission was established, the extraction supply voltage was adjusted until the desired charged particle emission current was reached. For each set of experiments, the extraction electrode voltage was held constant after emission stabilized while a predetermined time was allowed to elapse tq. Leaving the extraction voltage constant, the heater power was turned off to quench the operating ion source by quickly reducing the heater current. When the heat was removed, ion emission ceased within 2 s. After the LMIS was allowed to cool for 30 s, the extraction supply was also turned off. Additional procedures are included in the following subsections, since separate experiments were performed for the surface morphology studies in the FE-SEM and the lifetime comparison studies in the UHV chamber.

IV. Experimental Results and Discussion

Results from the surface morphology research performed in the FE-SEM and the lifetime comparison research performed in the UHV chamber are reported in Secs. I and II, respectively. The surface morphology experiment was divided into two studies with different durations of ion emission time before quenching, which are described in more detail at the beginning of Sec. IV.A, and the lifetime comparison experiment was divided into three subsections for each type of field-emitter tested, which are described in more detail at the beginning of Sec. IV.B.

A. Surface Morphology

The FE-SEM surface morphology experiment was divided into two experiments, reported in Secs. IV.A.1 and IV.A.2. The experiments were divided to investigate the temporal evolution of the surface morphology during several short (~10 s) quenches in an effort to understand the length of time required to preserve a Taylor cone structure and to investigate the surface morphology from quenched emitters that had been operated for 2 min before quenching. The duration of ion emission time before quenching was chosen for two reasons:

1) Ten seconds were approximately the minimum time required to establish and stabilize ion emission before quenching.

2) Two minutes of ion emission time before quenching were used in a previous study [17] so comparisons could be made with this study.

Section IV.A.1 describes the evolution of a regenerative emitter tip during the subsequent 10 s quenches. Section IV.A.2 comprises micrographs of quenched emitters and Fowler–Nordheim analyses from the quenched emitters at ion emission quench currents ranging from 1 to 30 μA, where quench current refers to the ion emission current when the heater power is quickly reduced.
I. Incremental Growth Through Successive Quenching

To investigate the evolution of nanotips during the growth process, the LMIS was operated for 10 s intervals and was imaged at the end of each interval. The first experiment was performed at 10 μA of the ion emission current, quenching the source every 10 s to show the evolution of the emitter tip after seven consecutive quenches at a constant ion emission current, as shown in Fig. 2. For the 10 μA quenches, successive quenches created surface modification after the fourth 10 s quench \( (t_q = 40 \, \text{s}) \).

As shown in the micrograph, slight surface modification occurred after the first 10 s quench \( (t_q = 10 \, \text{s}) \) at 10 μA. After the second 10 s quench \( (t_q = 20 \, \text{s}) \), the surface roughness appears to get more defined and not much change can be seen between the second and third quench. After the fourth 10 s quench \( (t_q = 40 \, \text{s}) \), the emitter tip appears to have grown a Taylor cone structure. After the fifth 10 s quench \( (t_q = 50 \, \text{s}) \) at 10 μA, the Taylor cone becomes more pronounced. After the sixth and seventh quenches \( (t_q = 60 \, \text{s} \text{ and } t_q = 70 \, \text{s}) \), surface texture starts to become visible on the surface of the Taylor cone.

After the seven successive quenches, the emitter tip surface morphology was reset to eliminate any nanostructure or microstructure on the surface. To reset a smooth surface, the extraction voltage was increased to 7 kV. An emission current of 100 μA was sufficient to melt the sharp tips, and the extraction voltage was high enough to cause arcing to destroy any locally sharp points on the apex.

The same emitter was then reset and used to observe nanostructure formation at higher ion quenching currents. The experiment was performed by operating the regenerable source at an extraction voltage between 3.4 and 3.8 kV. The same emitter was then reset and used to observe nanostructure formation at higher ion quenching currents. The experiment was performed by operating the regenerable source at an extraction voltage between 3.4 and 3.8 kV. After the seven successive quenches, the emitter tip surface appeared to have grown a Taylor cone structure. After the fifth 10 s quench \( (t_q = 50 \, \text{s}) \) at 10 μA, the Taylor cone becomes more pronounced. After the sixth and seventh quenches \( (t_q = 60 \, \text{s} \text{ and } t_q = 70 \, \text{s}) \), surface texture starts to become visible on the surface of the Taylor cone.

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The micrographs show an interesting depression at the apex that resembles a microvolcano that formed after the fourth 10 s quench \( (t_q = 40 \, \text{s}) \). It is possible the surface of the Taylor cone formed a thin oxide layer, which could be responsible for holding the shape of the emitter between successive quenches. Indium tends to form indium oxide, In₂O₃, even when in a vacuum environment. Indium oxide melts at 1910°C, so it could be possible that during subsequent quenches, the emission current was sustained at the apex by liquid indium \( (T_{\text{melt}} = 156.6°C) \) beneath the oxide layer that was able to break through the layer, resulting in the microvolcano structure due to depletion of indium. The emitter tip most certainly had to be at a greater temperature than the melting temperature of indium in order to sustain ion emission, which was the intent of heating the source with about 3 W during testing. Off-the-shelf LMISs used for focused ion beams are similar to the emitters used for the testing reported here and typically use about the same amount of heater power. Whether...
composed of indium or In$_2$O$_3$, the microvolcano structure is not completely understood at this time.

After seven consecutive quenches, totaling 70 s of ion emission, the same regenerable source was again reset. The regenerable source was operated at 30 $\mu$A of ion emission current and then quenched after 10 s of operation. The emitter tip was imaged and operated at 30 $\mu$A of ion emission current for six additional quenches at 10 s intervals totaling 70 s at 20 $\mu$A of ion emission, as shown in Fig. 4.

The apex of the emitter began to form a cone after the first 10 s quench ($t_e = 10$ s). The cone became more defined after the second 10 s quench ($t_e = 20$ s), and some nanostructures began to form along the tip surface. It was not until after the third 10 s quench ($t_e = 30$ s) that the nanostructure really started to become defined, as shown in the micrographs. Also, the microvolcano structure appeared following the fourth 10 s quench ($t_e = 40$ s), which was at the same point as in the 20 $\mu$A series when the structure formed.

To determine if the micrographs showed the evolution of the emitter tip or if the emitter tip relaxed after each time it was heated (between pictures in a given series of images) and formed a new structure during each 10 s emission iteration, an experiment was performed to determine how much the tip would relax during a heating cycle with no ion emission. This experiment consisted of resetting an emitter tip, operating the emitter at 20 $\mu$A of ion emission current for 20 s to form a Taylor cone structure and then quenching the emitter to solidify the Taylor cone. The Taylor cone was imaged with the FE-SEM and then heated in the absence of an electric field (no ion current) to observe if the emitter tip relaxed, as shown in Fig. 5.

The same emitter was then operated at 20 $\mu$A of ion emission current (without resetting) by heating the emitter and increasing the extraction voltage to establish emission. Ion emission was adjusted to 20 $\mu$A for 20 s and then quenched. After quenching, the emitter was imaged and heated for 20 s in the absence of an electric field. Following the heating experiment, the emitter was imaged and heated again for an additional 20 s, as shown in Fig. 6.

Although it is impossible to say with certainty what mechanisms are the most influential in causing surface morphology changes between subsequent thermal cycles, the micrographs show that the emitter tip surface does not completely relax during each thermal cycle. That said, the images in Figs. 2–4 likely show the change in surface morphology as a result of the cumulative charged particle emission time coupled with multiple other factors that were relatively consistent between tests, including the quantity of indium at the apex of the emitter, the local electric field, the underlying emitter geometry, and the operating temperature.

2. Nanostructure Formation with Fowler–Nordheim Modeling

The purpose of this experiment was to use the FE-SEM to visually verify the trends that were implied via application of the Fowler–Nordheim modeling.
Nordheim model to data taken in an earlier study within an UHV chamber where an LMIS was operated for 2 min durations at a desired ion emission current before quenching [17]. In the earlier tests, the UHV chamber was equipped with an optical microscope that had 90X magnification, which was not a sufficiently high magnification to resolve the nanostructure and microstructure of quenched emitters. Thus, obtaining higher resolution micrographs motivated this series of experiments.

The regenerable emitters were quenched at multiple ion emission currents inside the FE-SEM. The FE-SEM was then used to image the nanostructure formation after quenching. Also, electron emission I-V sweeps were performed after each quench to estimate the emitter tip radii using the Fowler–Nordheim model. The emitter tip estimations were compared with the FE-SEM micrographs. The ion quenching currents that were chosen for the experiment ranged from 2 to 20 μA, operating each for 2 min exactly, as done by Makela et al. [17]. After each quenched emitter was imaged and an electron I-V sweep was acquired, the emitter tip was then reset using the same procedure described previously of exposing the emitter tip to destructive electron emission conditions to smooth out and remove surface features. The quenching experiments were performed in a randomized order but are presented from lowest to highest ion quenching current. The actual order of ion emission current before quenching was 20, 10, 5, 15, 20, 6, 16, 3, 10, and then 2 μA. After showing representative trends of individual results, a summary is provided with all of the compiled data, including data from previous work in the UHV chamber [17].

For each experiment, the regenerable emitter was first exposed to electron reset conditions. Sample data from the reported experiment, where the emitter was operated at 2 μA of ion emission current (at 3.3 kV) for 2 min and quenched, are shown in Fig. 7. The image is a postquench micrograph at 9000X magnification, which was the highest resolution that was possible to obtain during testing. As shown, a cone-type structure formed with nanoscale features on the surface. The volcano-type structure that was observed in the previous section was present in this test.

After quenching and imaging the emitter tip, an electron I-V sweep was performed on the solidified emitter. Using the electron I-V data and the Fowler–Nordheim model,

\[
\frac{I}{V^2} = a \left[ \exp \left( \frac{-b' \varphi^{3/2}}{V} \right) \right]
\]

an estimate of the emitter tip radius could be made. Gomer’s technique of using the Fowler–Nordheim model was applied to predict the emitter tip radii [21]. Using a plot of \(a(I/V^2) vs 1/V\), where \(I\) is the emission current and \(V\) is the extraction voltage, the slope of the Fowler–Nordheim plot was used to estimate the nanostructure size. An emitter radius estimate was made using the Fowler–Nordheim \(b'\) coefficient, which is shown in the following equations:

\[
m = b' \varphi^{3/2}
\]

where

\[
b' = 6.8 \times 10^3 \alpha kr_i
\]

Fig. 7 Micrograph showing a regenerable emitter tip after a 2 μA quench for 2 min at magnification of 9000X.

For all of the tests, the experimental I-V data were also analyzed to determine the experimental error. To verify the error in the slope, and thus the emitter radii error, the standard error of the linear regression from the Fowler–Nordheim plots was calculated using a data analysis toolkit in readily available software. The standard error \(s_m\) is an estimate of the standard deviation and bounds the slope \(m\) of a linear regression with a 95% confidence interval as [22]

\[
m - 2s_m \leq m \leq m + 2s_m
\]

The experimental error could then be compared against the Fowler–Nordheim criteria of 20% set by Gomer [21]. For each set of Fowler–Nordheim data that were collected, the experimental error in the emitter tip radii that were calculated were less than 20%; however, ±20% of the emitter tip radius was used when plotting data to maintain a conservative estimation.

For the sample set of data, the Fowler–Nordheim analysis was performed by increasing the extraction voltage to 2.9 kV at 50 V increments. A Fowler–Nordheim plot was created from the sweep data, as shown in Fig. 8, with an \(R^2\) of 0.86 for the linear curve fit. Applying the Fowler–Nordheim model to the data resulted in an emitter tip radius estimate of 7.0 ± 1.4 nm. Table 1 shows the complete set of test conditions that were explored and includes the ion current before quench, the extraction voltage at quench, the estimated emitter radii, and the \(R^2\) value from the linear regression analysis of the Fowler–Nordheim data.

A FE-SEM micrograph from a representative experiment is shown in Fig. 9. Many of the quenched emitter tips that were tested developed a texture on the surface, similar to what is shown in Fig. 9, while some of the emitters developed microdroplets (shown previously in Fig. 3 at \(t_i = 30\) and \(40\) s) that have been more commonly observed [1,19]. Also, some of the emitter tips formed the microvolcano structure shown previously in Fig. 4. The complete set of micrographs are available in the work of Makela [23].

As shown, the emitter tips are on a microscale with nanoscale features that formed on the surface of the emitters, which are referred to as nanostructures for the remainder of this document. Although the exact size of the nanostructures cannot be distinguished, electron I-V sweeps were acquired from each quenched emitter tip so that Fowler–Nordheim estimations could be made, as previously reported in Table 1 and plotted in Fig. 10. As displayed in the FE-SEM data, within the error bars, a discernible trend is not apparent between the emitter tip radius and the ion current, although the estimated emitter tip radii are on the same order as reported in the UHV data.

Unfortunately, comparing the FE-SEM data, the UHV data, and the FE-SEM micrographs shown previously, it was not possible to make accurate emitter tip radii estimations visually. The authors believe that the resolution of the micrographs was limited due to...
vibration in the fixture, since the LMIS was cantilevered from a Teflon fixture during testing and imaging.

Investigation of the surface structure of a quenched ion source using a FE-SEM resulted in the observation of a Taylor-cone-shaped structure, which was expected. However, some cones had multiple nanostructures that were solidified on the surface of the emitter tip. In fact, nanostructures were observed on most of the quenched emitters at quench currents covering the entire range tested, from 2 to 20 μA, with the exception of three experiments (i.e., 5, 10, and 15 μA quenches). Most of the micrographs revealed that the nanostructures were evenly spaced and approximately the same size all over the surface. Although electron emission was most likely occurring at the apex of the emitter (due to the apex being in closest proximity to the extraction electrode), any of the nanostructures were capable of emission if/when the sharpest structures at the apex became incapable of emission.

To summarize the major findings from the FE-SEM research, nanostructures were formed from quenching a LMIS at emission currents ranging from 2 to 30 μA. It was shown that a well-defined Taylor cone substructure formed after 20 to 30 s of ion emission, and a visual inspection revealed that the definition of nanostructures that were formed on the surface of the Taylor cone tended to reach a maximum after 30 to 40 s of ion emission. The exceptions were three of the quenches: for unknown reasons, a 5, 10, and 15 μA quench did not produce a Taylor cone substructure or multiple nanostructures; however, they still had some sharp nanostructures. Another interesting observation was that the Taylor cones did not completely melt by resistive heating in the absence of an electric field. While the temperature of the indium must have been sufficient to liquefy the indium, since ion emission could be established and the bulk shape of the emitter could be deformed, it is possible that an indium oxide coating on the surface of the emitter could have been responsible for maintaining the shape of the emitter when heated without an applied electric field. Indium oxide, In₂O₃, has a much higher melting temperature than indium, so the oxide layer could have formed a solid skin on the emitter surface. The Fowler–Nordheim model approximations reported in this paper had comparable tip radii estimations to those reported previously by Makela et al. [17]. Visually inspecting the nanostructures with the FE-SEM could not provide sufficient resolution to accurately determine the emitter tip radii; however, applying the Fowler–Nordheim model to the I-V data provided emitter tip radii estimations from 5 to 50 nm.

### B. Lifetime Comparison

This section is focused on the comparison between regenerable field emitters and pure tungsten field emitters. Tungsten field emitters were chosen due to the extensive characterization of single-needle tungsten emitters historically and since Spindt-type arrays have been made from refractory metals like molybdenum and tungsten [7,24]. Since Spindt-type emitters are made up of thin films of refractory metals, such as molybdenum or tungsten, the solid pure tungsten emitters should have had similar performance. To establish a baseline to compare the regenerable emitters with, the emission

<table>
<thead>
<tr>
<th>Ion current at quench, μA</th>
<th>Extraction voltage at quench, kV</th>
<th>Chronological order of data</th>
<th>Emitter tip radius estimate, nm</th>
<th>$R^2$ value from F–N plot</th>
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<tbody>
<tr>
<td>2</td>
<td>3.3</td>
<td>10</td>
<td>7.0 ± 1.4</td>
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<tr>
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<td>22.6 ± 4.5</td>
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<td>3.2</td>
<td>6</td>
<td>9.0 ± 1.8</td>
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<td>5</td>
<td>20.4 ± 4.1</td>
<td>0.77</td>
</tr>
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</table>

*Ion current and extraction voltage at quench are presented, as well as the chronological order that the data points were acquired. Also, emitter tip radius estimates from Fowler–Nordheim analyses [21] of regenerated emitter tips are reported at currents ranging from 2 to 20 μA.*
characteristics of tungsten emitters were investigated for long durations of time and at an elevated background pressure. For the reported experiments, two types of electrochemically etched tungsten emitters were used. The first was dc etched in a 2M NaOH solution to create a sharp and smooth emitter tip, following the procedure described by Makela et al. [17], as shown in Fig. 11 [25]. The second type of tungsten emitter that was tested was dc and then ac electrochemically etched in a 2M NaOH solution to create a roughened surface structure (like the emitters that were coated with indium), as shown in Fig. 12 [26]. The roughened structure was intended to create an emitter with surface grooves, exactly the same as the field emitter that was coated with indium for the regenerable experiments. A possible indirect benefit of adding surface grooves was that the number of field-emitting nanostructures on the surface may have also increased.

To characterize the emitters, a series of experiments were performed to observe the emission characteristics of each emitter for long durations of time in a UHV environment. Furthermore, each type of tungsten emitter and the regenerable emitters were tested for 2 to 24 h, and then the ion pump was turned off to let the vacuum chamber pressure slowly increase while observing the electron emission current. Once emission ceased, the extraction voltage was increased until emission was sustained. Each field emitter was exposed to elevated background pressure conditions multiple times to determine which emitter was the most robust.

To establish electron emission from the tungsten emitters, the extraction electrode was biased positively with respect to the emitter (which was at ground potential) and the extraction voltage was increased until emission was achieved. The emission current was recorded using a custom microammeter that was placed inline with the extraction supply. For lifetime studies, the extraction voltage was held constant and the emission current was observed while at pressure $10^{-6}$ torr. For the elevated background pressure experiments, emission was first established at a pressure less than $10^{-6}$ torr for 2 to 24 h, and then the ion pump was turned off to let the vacuum chamber pressure slowly increase while observing the electron emission current. Once emission ceased, the extraction voltage was decreased and the ion pump was turned back on to reduce the vacuum chamber pressure. When the pressure returned to less than $10^{-6}$ torr, the same procedure was used to establish emission and expose the operating electron source to elevated background pressure. The process was repeated several times until emission could no longer be achieved at reasonable (~4 kV) extraction voltages. Ultimately, exposing the emitter tips to increased pressure multiple times would destroy all of the sharp nanostructures. The regenerable emitters were then tested using the same procedure as the pure tungsten emitters.

1. **Smooth Bare Tungsten Emitters**

The first experiment was performed with a smooth bare tungsten field emitter, as shown previously in Fig. 11. The emitter was electrochemically dc etched using the procedure described by Makela et al. [17]. The smooth bare tungsten emitter was tested in the UHV chamber at a pressure of $10^{-9}$ torr. A Fowler–Nordheim sweep was taken before the lifetime experiment and yielded a tip radius estimate of 13.9 ± 1.2 nm. Electron emission was then achieved by increasing the extraction electrode to 1.4 kV to obtain an emission current of 3 μA. The extraction voltage was then held constant and emission current was observed for 625 h. The test was concluded after 625 h due to the necessity to perform additional experiments in the UHV facility. To conclude the experiment, at $t_e = 625$ h, the ion pump on the UHV chamber was turned off to slowly increase the chamber pressure. At a pressure of $10^{-7}$ torr, and after less than 2 min from the time the pump was turned off, the emission ceased, as shown in Fig. 13. A Fowler–Nordheim analysis was not performed after the 625 h test because electron emission could not be achieved at an extraction voltage of 4 kV. For the elevated background pressure experiments, if field emission required more than an extraction voltage of 4 kV to obtain electron emission, testing with the emitter was discontinued. The extraction voltage of 4 kV was arbitrarily chosen as an upper bound for all of the experiments reported. Since emission could not be established, the emitter tip must have been catastrophically damaged by the increase in background pressure. Exposure to elevated background pressure rendered the bare tungsten emitter useless as a field-emission electron source. A second smooth bare tungsten field emitter was tested under similar conditions and was permanently damaged after two exposures to elevated background pressure.

2. **Roughened Bare Tungsten Emitters**

Two roughened bare tungsten emitters were tested in elevated background pressure environments for comparison. The roughened emitters were electrochemically dc and ac etched to create surface grooves, as shown previously in Fig. 12. Etching the surface grooves on the emitter was intended to make the electrode as close as possible to the indium-coated regenerable electrodes. An electron I-V sweep was acquired on the new roughened bare tungsten emitter at $10^{-9}$ torr. Applying the Fowler–Nordheim model to the electron emission I-V sweep yielded a tip radius estimate of 4.6 ± 0.9 nm. After performing an electron I-V sweep, the extraction voltage was adjusted to establish electron emission.

The roughened bare tungsten emitter was then operated for about 20 h at a relatively constant emission current. Holding the extraction voltage constant, at $t_e = 21.2$ h, the vacuum chamber pressure was increased. The pressure was increased by turning off the ion pump. As the vacuum chamber pressure increased, the electron emission current was observed. Electron emission sustained for about 7 h at background pressures between $10^{-5}$ and $10^{-3}$ torr (the region where the vacuum gauges did not overlap), as shown in Fig. 14, which was about 7 h longer than emission sustained using smooth bare tungsten emitters. The region of unknown pressure was due to a lack of overlap in the cold cathode and thermocouple pressure gauges that were equipped on the vacuum facility. Analyzing data from a subsequent elevated pressure test using a gauge with a
wider vacuum measurement range, an approximate pressure of $5 \times 10^{-6}$ torr was estimated when emission ceased.

Once emission ceased, the pressure was decreased to $10^{-9}$ torr and another electron I-V sweep was acquired. The I-V sweep was applied to the Fowler–Nordheim model and yielded a tip radius estimate of $9.9 \pm 2.0$ nm, which was an increase in the emitter tip radius that was observed before exposing the operating field emitter to an increase in pressure.

The roughened bare tungsten emitter was then used to reestablish electron emission. After emission was stabilized at a pressure of about $10^{-9}$ torr, the pressure was increased while emission was observed. When emission ceased, the extraction voltage was reduced, the vacuum chamber was returned to high vacuum, and electron emission was reestablished. A Fowler–Nordheim I-V sweep was acquired, and then emission was stabilized; the vacuum chamber pressure was increased, and the emission current was observed as pressure was increased. This process was repeated several times until electron emission could no longer be achieved with less than 4 kV of extraction potential.

To summarize, the first roughened bare tungsten emitter that was tested survived seven exposures to elevated background pressure within the test conditions of using a maximum extraction voltage of 4 kV to establish electron emission. Also, the first roughened bare tungsten emitter sustained electron emission for 1.5 to 20 h after the increase in pressure began for each experiment. A second roughened tungsten field emitter was tested and survived four exposures to elevated pressure, and emission was sustained from 1.4 to 20 h while pressure was increased. The maximum pressure that was measured before emission ceased was approximately $9.9 \times 10^{-9}$ torr. Both roughened emitters demonstrated longer lasting emission and survived more exposures to elevated pressure than the smooth bare tungsten emitters, as outlined in Table 2.

3. Regenerable Field Emitters

To investigate the lifetime of regenerative emitters, a dual ion/electron source was placed in the UHV chamber and the vacuum chamber pressure was reduced to $10^{-9}$ torr. The emitter was then heated and operated at 20 $\mu$A of ion emission current for 1 min and then quenched. The polarity of the extraction electrode was then reversed to achieve electron emission, and the extraction electrode was increased to obtain electron emission at 4 $\mu$A. The voltage required for 4 $\mu$A of electron emission current was 4.1 kV; the extraction electrode was then voltage-limited at 4.1 kV for the remainder of the experiment. The reason for choosing 4 $\mu$A as a target was because operating a single-needle electron source at emission currents of 10s of microamperes for long durations of time can cause undesirable heating of the emitter apex, which can destroy the sharp emitter tip.

Within the first hour of the lifetime test, the emission current increased to approximately 11 $\mu$A for a few minutes and then quickly decreased down to about 3 $\mu$A, as shown in Fig. 15. At $t = 950$ h of operation, the experiment was voluntarily shut off for 5 h to observe if the emission current would return to the same magnitude when turned back on. The length of the shutdown period was arbitrarily chosen while modifications were made to data acquisition software. Shutting off the experiment was achieved by simply decreasing the extraction voltage from 4.1 to 0 kV.

The experiment was turned off for 5 h, and then the extraction electrode was increased back to the original setpoint of 4.1 kV. The emission current increased to almost 10 $\mu$A after the onset of emission but quickly decreased to about 5 $\mu$A at $t = 952$ h. Then, the electron emission current remained at 5 $\mu$A until approximately $t = 975$ h and then rapidly decreased to 2 $\mu$A. Once reaching 2 $\mu$A, the emission current slowly increased up near 3 $\mu$A until $t = 1025$ h. The experiment was then voluntarily shut down for a span of 4 h. During the 4 h period of downtime, an electron I-V sweep was taken to estimate the emitter tip radius. Applying the Fowler–Nordheim model to the I-V data yielded a tip radius estimate of $9.5 \pm 1.9$ nm.

After the I-V sweep was taken, the extraction voltage was increased back to the setpoint of 4.1 kV. As the extraction voltage was increased, the electron emission current increased to approximately 3 $\mu$A for 435 h of operation, bringing the total duration of the lifetime test thus far to 1460 h. At $t = 1460$ h, the experiment was voluntarily shut down again to obtain another electron emission I-V sweep. The I-V sweep was taken to determine if any change in tip radius was apparent from the first I-V sweep. The Fowler–Nordheim

![Fig. 14: Electron emission current from a roughened bare tungsten emitter held at a constant extraction voltage of 2.8 kV from $t_e = 3.1$ h to $t_e = 28.9$ h as background pressure was increased for the first time. The region of unknown pressure was due to a lack of overlap in the cold cathode and thermocouple pressure gauges that were equipped on the vacuum facility.](image1)

![Fig. 15: Electron emission lifetime experiment from a single-needle LMIS quenched at ion emission current of 20 $\mu$A, showing locations of voluntary shutdowns.](image2)

![Table 2: Emitter tip comparison table displaying how long the smooth and roughened bare tungsten emitter tips lasted at an elevated background pressure, how many exposures to increased pressure caused permanent failure, and the maximum vacuum chamber pressure that emission could sustain.](table1)

<table>
<thead>
<tr>
<th>Emitter type</th>
<th>No. of tested emitters</th>
<th>Emission duration at elevated pressure</th>
<th>No. of exposures before failure</th>
<th>Approx. maximum tank pressure reached</th>
</tr>
</thead>
<tbody>
<tr>
<td>Smooth bare tungsten</td>
<td>2</td>
<td>1–5 min</td>
<td>1–2</td>
<td>$10^{-7}$ torr</td>
</tr>
<tr>
<td>Roughened bare tungsten</td>
<td>2</td>
<td>1.4–20 h</td>
<td>4–7</td>
<td>$10^{-5}$ torr</td>
</tr>
</tbody>
</table>
The vacuum chamber pressure was estimated at 0.3 h to t = 20 h as background pressure was increased.

Following the I–V sweep, the extraction voltage was again increased to 4.1 kV. The emission current gradually increased back to near 3 µA as the extraction voltage was increased. The lifetime experiment was continued for 300 additional hours, bringing the total lifetime test to about t = 1750 h. The lifetime experiment was ended after 1750 h due to the necessity to use the UHV chamber to perform other experiments. To end the lifetime experiment, the extraction voltage was turned off, and then a final electron emission I–V sweep was taken to estimate the nanostructure tip radius. The Fowler–Nordheim plot was applied to the electron I–V data from the fourth sweep and yielded a tip radius estimate of 16.2 ± 3.2 nm.

In addition to the long-duration experiment, elevated background pressure tests were performed with the regenerable emitters to compare with the single-needle bare tungsten field emitters that were exposed to increased pressure. The LMIS was first operated as an ion source for 1 min at 20 µA while heating the LMIS with 2.25 A at 0.3 V. After 1 min, the heater was turned off and the ion emitter was quenched. The extraction voltage was then reversed to obtain electron emission, and an I–V sweep was performed at 10−8 torr. The Fowler–Nordheim model was applied, and the model resulted in a tip estimate of about 13.7 ± 2.8 nm.

After the I–V sweep, the extraction voltage was increased to 3.8 kV to obtain electron emission. During the first 20 min of operation at 10−8 torr, the emission current had increased to about 25 µA, so the extraction voltage was increased to 2.1 kV in an attempt to keep the emission current around 5 µA. After the emission current remained relatively constant for approximately 1 h, the ion pump on the UHV chamber was turned off to increase the background pressure. The background pressure was then recorded until the ion gauge turned off due to an overpressure. Unfortunately, vacuum chamber pressure could not be recorded for the duration of the test due to a lack in overlap of measuring range between the ion gauge and thermocouple gauge on the UHV system. The data that were recorded are shown in Fig. 16, along with a bound of the possible background pressure. The vacuum chamber pressure was estimated at 8 × 10−5 torr. As shown, electron emission from the quenched LMIS was sustained for over 15 h, which was 15 h longer than the smooth bare tungsten emitters.

Once emission ceased, the ion pump on the vacuum facility was turned back on and pressure was decreased to 10−8 torr. Then, an electron I–V sweep was performed to obtain an estimate of the nanostructure radius that was emitting electrons. Applying the Fowler–Nordheim model resulted in a tip radius estimate of 12.8 ± 2.6 nm. The extraction voltage was then increased to obtain an electron emission current. Once the emission current remained stable, the ion pump on the UHV system was turned off again to increase the background pressure. Once again, electron emission was sustained for 10s of hours longer than with a single bare tungsten field emitter. The process of operating the field emitter in a high vacuum environment while slowly increasing the pressure until emission ceased was repeated several times until emission could no longer be established.

Some of the emitter tips that were exposed to elevated vacuum chamber pressure demonstrated better electron emission performance after exposure. While no definitive argument is proposed, some of the possible reasons for that phenomenon are discussed. One possibility is that electron emission could have occurred from multiple emission sites simultaneously. During some of the experiments in the UHV chamber, when an emitter was used to obtain over 10 µA of electron emission current, multiple locations on the emitter tips could be observed through the optical microscope that were glowing blue. It is possible that the local electric field enhancement was very similar at numerous locations, so any of the locations that sustained damage due to elevated pressure conditions could have easily been replaced by other sharp locations. Another possibility is that ion sputtering at higher vacuum pressure could have sputtered away some of the atoms on the field-emitting nanostructures, leaving an even sharper emitter [27].

The results from the comparison between the smooth bare tungsten, the roughened bare tungsten, and the regenerable emitters are summarized in Table 3. The table includes the length of time that emission could be sustained at elevated pressure, the number of times the emitter was exposed to an increased pressure before permanent failure was observed, and the magnitude of the maximum pressure that was recorded while emission sustained.

<table>
<thead>
<tr>
<th>Emitter type</th>
<th>No. of tested emitters</th>
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<td>Roughened bare tungsten</td>
<td>2</td>
<td>1.4–20 h</td>
<td>4–7</td>
<td>10−5 torr</td>
</tr>
<tr>
<td>Regenerable emitter</td>
<td>2</td>
<td>6–45 h</td>
<td>Unlimited ^</td>
<td>10−7 to 10−8 torr</td>
</tr>
</tbody>
</table>

^Unlimited refers to the capability to regenerate the tip as long there is a sufficient metal supply available on the emitter tip.

![Graph showing electron emission current from a quenched LMIS held at a constant extraction voltage of 2.1 kV from t = 0.3 h to t = 20 h as background pressure was increased.](image)
quenched emitters proved to be more robust, sustaining electron emission at pressures greater than 10^{-5} torr. Lifetime experiments demonstrated that the regenerable field emitters could sustain electron emission current for long periods of time, and Fowler–Nordheim modeling of the emitter tip during voluntary shutdowns throughout the lifetime experiment yielded sharp emission images on the order of 100s of nanometers. The sharp nanostructures successfully demonstrated electron emission at an emission current of about 3 μA for 1750 h from a single needle that was quenched at an ion emission current of 20 μA. Emission current from the regenerable emitters stayed relatively constant for the long-duration tests, while the current degraded over time for both types of bare tungsten emitters.

V. Conclusions

As expected, quenching an operating LMIS yielded a well-defined Taylor cone. An interesting and unanticipated observation from the micrographs revealed multiple nanostructures covering the tapered section of the Taylor cone. As the quenching process was repeated, the nanostructures that were formed became more and more defined, and between subsequent quenches, the overall structure of the Taylor cones maintained their shape, even when subjected solely to heat in the absence of an electric field.

Another interesting feature was also present in many of the micrographs that were taken after quenching an operating LMIS multiple times. A hollow depression that looked like a microvolcano formed at the emitter tip apex after subsequent 10 s quenches and appeared to form after the fourth 10 s quench in most cases. It is not clearly understood at this time, but it is possible that a thin oxide layer is present on the surface of the emitter, causing the emitter tip to retain its shape during repeated quenching while the emission is sustained by depleting liquid indium from the apex.

Within the error bars of the Fowler–Nordheim analysis from quenched emitters, it was not possible to determine if the ion quenching current had a significant effect on the radius of the quenched emitter. However, the Fowler–Nordheim model revealed that the emitter radii from quenched LMISs are on the order of other state-of-the-art field emitters (e.g., carbon nanotubes and Spindt-type arrays).

To determine performance characteristics of regenerable and tungsten emitters, lifetime experiments were performed using three types of field emitters: smooth bare tungsten, roughened bare tungsten, and the regenerable emitters. Single-needle tungsten field emitters were used for comparison rather than Spindt-type emitters to avoid complicated microfabrication that is required to build Spindt devices. Since Spindt-type emitters are made up of thin films of refractory metals, such as molybdenum or tungsten, the pure tungsten emitters should have had similar (or better) performance.

The main purpose of the comparison experiments was to determine if regenerable field emitters had similar emission characteristics to bare tungsten emitters and to determine what, if any, advantages regenerable emitters had over bare tungsten emitters that were approximately the same physical size and geometry. The experiments with the electrochemically dc etched smooth bare tungsten emitters demonstrated the least reliable operation and were the most prone to permanent damage after the fewest exposures to elevated background pressure environments. The smooth bare tungsten emitters ceased to function after one to two cycles from 10^{-8} to 10^{-2} torr. In addition, emission would cease after only a few minutes at increased pressure.

The regenerable emitters demonstrated the longest lasting performance at an elevated pressure of the three emitters. The regenerable emitters survived up to 45 h at pressures up between 10^{-3} to 10^{-5} torr. While the regenerable emitters could only be exposed to elevated pressure two times before emission could no longer be achieved at up to 4 kV of extraction voltage, they could then be regenerated and subsequently operated for 100s of hours or longer. Therefore, the regenerable emitters could potentially be cycled to increased pressure an almost unlimited number of times. Not only did the regenerable emitters prove to be the most reliable when exposed to unfavorable vacuum conditions, the emission current remained more stable (with less decrease) than the tungsten emitters throughout the duration of the lifetime experiments. Also, the experiments reported using Fowler–Nordheim modeling of regenerable emitters have estimated that the regenerable emitters can be used to create the same size nanostructures as state-of-the-art field emitters: 1–100 s of nm.

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