Re-generable Field Emission Cathodes for Electric Propulsion

By:

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This dissertation, "Re-generable Field Emission Cathodes for Electric Propulsion," is hereby approved in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the field of Mechanical Engineering – Engineering Mechanics.

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If we did all the things we are capable of doing, we would literally astound ourselves.

-Thomas Edison

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Abstract

The thrusters that are used with electric space propulsion systems are equipped with an electron source for spacecraft neutralization and often for propellant ionization. The research presented in this dissertation was intended to explore a type of electron source that can be regenerated when its performance degrades – restoring the electron source to its original like-new condition. The electron source of interest is a field emission electron source, which relies on electric field enhancement from nano- and micro-scale sharp emitter tips to create a beam of electrons. The re-generable emitter tips are formed by taking advantage of Taylor cone formation from an operating liquid metal ion source. Tip formation was accomplished by solidifying, or quenching, the ion-emitting cone to preserve the sharp ion-emitting nano-structures so that they can then be used for electron emission, and subsequently re-generated when they become damaged.

To examine the feasibility of using re-generable field emitters with electric propulsion systems, a series of experiments were conducted to investigate the re-generable emitter tips. The experiments involved re-generating multiple emitter tips so that the electron performance of each re-generated tip could be evaluated and applied to the Fowler-Nordheim model to estimate the emitter tip radii. The re-generable emitter tips were also subjected to long duration electron emission tests under UHV conditions, as well as repeatedly being subjected to elevated pressures. The same duration and pressure elevation experiments were also performed with smooth and roughened pure tungsten field emitters for comparison purposes. Another experiment was conducted in the specimen chamber of a Field Emission Scanning Electron Microscope (FE-SEM) so that the nano-structures could be re-generated at ion emission current before quenching of 2 to 20 μ A and then investigated while inside the microscope. After quenching, Fowler-Nordheim modeling was used to estimate the emitter tip radii and then the FE-SEM optics were used so that the surface morphology of the quenched emitters could be investigated visually to compare with the model.

It was demonstrated that as the ion emission before quenching was increased, a decrease in emitter tip radius was observed. At ion emission currents greater than 10-15 μ A a minimal plateau was reached for the tip radii that were formed. The re-generable emitter tips also demonstrated more stable electron emission than pure tungsten field emitters during 100's of hours of testing. Where the emission current from the pure tungsten emitters decreased over time, the emission current from the re-generable sources remained more stable. The FE-SEM investigation revealed that multiple nano-structures were formed upon quenching an operating liquid metal ion source and it was apparent that 30 seconds of ion emission operation is approximately the minimum time required before quenching to form all of the nano-structures that are generated. The multiple nano-structures could then be used to operate much like an array of electron field emitters since each of the nano-structures had similar electric field enhancement.

The results of the experiments allowed for several conclusions to be made. The regenerable emitters proved to be more robust than pure tungsten emitters. Re-generable emitters continued to operate to higher vacuum pressure, on the order of 10^{-4} Torr, and for 10's of hours longer than the tungsten emitters. Also, repeated exposure to elevated pressure eventually caused catastrophic failure of the pure tungsten emitters whereas the re-generable emitters demonstrated the ability to re-generate sharp nano-structures after the emission deteriorated. The re-generated tips could then be used at lower extraction voltage than before re-generation. The FE-SEM investigation revealed that the Fowler-Nordheim estimation of emitter tip radii appeared to accurately predict the emitter tips to at least an order of magnitude. Higher resolution micrographs would be necessary but within the resolution of the acquired micrographs the model approximations appear realistic. The FE-SEM experiments also revealed multiple nano-structures that were never observed before. The structures created multiple locations of electric field enhancement which is beneficial for a field emitter. Combining all of information gathered from this research about emitter tip lifetime, tip robustness in elevated pressure, re-generability after performance degradation, and the fact that multiple nano-structures are created on the emitter tip, re-generable field emitters appear to have a great amount of potential to be further developed for electric propulsion systems.

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Nomenclature

а	Fowler-Nordheim term
Α	Total emitting area (m ²)
b'	Fowler-Nordheim term
d	Tip-to-extraction electrode spacing (m)
Ε	Electric field at emitter tip apex (V/m)
F _E	Electric field force (N)
F _{ST}	Surface tension force (N)
F _T	Thrust (N)
g	Acceleration due to gravity (m/s^2)
I	Current (A)
I _{sp}	Specific impulse (s)
k	Empirical relation between tip radius and gap spacing
1	Groove length (m)
т	slope
'n	Mass flow rate (mg/s)
<i>m</i> _a	Anode mass flow rate (mg/s)
$\dot{m_c}$	Cathode mass flow rate (mg/s)
ΔP	Capillary pressure (Pa)
p_{int}	Internal pressure, due to surface tension (Pa)
p_{ext}	External pressure, due to electric field (Pa)
r_{g}	Longitudinal groove radius (m)
r_h	Extraction electrode hole radius (m)
r_t	Emitter tip radius (m)
S _m	standard error for slope
t _e	Elapsed Time (s)
Т	Temperature (K)
<i>ν</i> ̈́	Volumetric flow rate (m ³ /s)
V _{max}	Extraction voltage required for 0.5 μ A of electron emission current (V)
V_o	Extraction electrode potential (V)
α	Nordheim image-correction factor
γ	Surface tension (N/m)
\mathcal{E}_{o}	Permittivity of free space constant ($8.854 imes10^{-12}$ F/m)
η	Viscosity (kg/m·s)
θ	Contact angle (degrees)
μ	Fowler-Nordheim term
ϕ	Work function (eV)

Chapter 1

1 Introduction

1.1 Field Emission Cathode Overview

Electric space propulsion systems have a wide range of power levels, from a couple of Watts to 10's of kilowatts.¹⁻³ The electron sources that are employed for propellant ionization and spacecraft neutralization for most electric propulsion systems are thermionic and require on the order of 10-100 Watts of heater power and 1 to 10's of mg/s of propellant to operate.^{4, 5} Since thermionic electron sources require power and propellant, both of which can't be directly converted to thrust, they contribute to a decrease in the efficiency and specific impulse of electric propulsion systems. For thrusters in the sub 500 Watt power range, a thermionic cathode can decrease the efficiency and specific impulse of the system by over 20% and 15%, respectively, with this penalty decreased to 2% and 10% as power levels are increased over 5 kW. Ideally, lower power propulsion systems could really benefit from implementing a different electron source. Fortunately, another type of electron source is available which requires less power and propellant than thermionic sources, the field emission cathode.

Field emission cathodes are devices that take advantage of locally enhanced electric fields to emit a beam of "cold" electrons. Electric field enhancement is achieved by using nanoscale sharp electrodes. Applying a potential between the sharp emission electrode and an extraction electrode causes a beam of electrons to be extracted from the sharp emitter. The required potential to achieve electron emission is a few volts up to a few kV, depending on the sharpness of the emission electrode. A more detailed analysis of field emitters is presented in Chapter 2.

The only drawback to field emission cathodes is the limited lifetime associated with the devices. The nano- or micro-scale features are fragile and when the features become damaged, the electron source loses functionality. Researchers have found some ways to minimize damage to the emitters⁶ and they have found more robust, longer-life, emitters, such as carbon nanotubes.⁷⁻¹⁰ However, all electron field emitters can and will become damaged over time^{11, 12} – it's just a matter of how much time it will take.

The research reported in this dissertation presents a method for re-generating damaged field emitter tips *in situ*. Having the ability to re-generate damaged emitter tips has the potential to greatly increase the efficiency and lifetime of field emission electron sources.

1.2 Contribution of this Research

The goal of this research was to examine single-needle re-generable electron sources to determine if the re-generable electron source is a feasible technology to be used with electric propulsion systems. Since the emitter tip sharpness is the most important factor influencing local electric field enhancement, for re-generable emitters to ever be implemented in space propulsion devices it was important to demonstrate that re-generable emitter tips could be created just as small as current state-of-the-art emitters. Emitter tip robustness was also examined to ensure that re-generated emitters could operate in elevated vacuum conditions and for long durations.

The experiments that are reported in this dissertation were designed (1) to examine the electron emission current-voltage (I-V) characteristics of a re-generable electron source, (2) to investigate the lifetime of the re-generable electron source, (3) to determine the robustness of the re-generable electron source against elevated neutral pressure conditions, (4) to compare the exposure time and maximum-pressure-before-failure of re-generated emitters with pure tungsten field emitters, and (5) to visually investigate the surface morphology and emitter tip geometry after re-generating the electron sources.

Experiments 1-4 were performed in an ultra high vacuum (UHV) chamber in MTU's Ion Space Propulsion (Isp) Lab and Experiment 5 was performed inside of a Field Emission Scanning Electron Microscope (FE-SEM) at MTU's Applied Chemical and Morphological Analysis Laboratory. For Experiment 1 emitter tips were re-generated numerous times and after the tips were re-generated, the emitter was operated as an electron source so that the extraction voltage could be increased while recording the emission current. The current-voltage data were then applied to the Fowler-Nordheim model to estimate the emitter tip radii. Experiment 2 involved observing the emission current of a re-generated emitter tip at a constant extraction voltage for about 1,750 hours. Experiment 3 was performed to investigate the robustness of the re-generated emitter tips by exposing the operating emitters to elevated vacuum conditions. After multiple exposures to elevated pressure, an emitter was re-generated to enhance the local electric field of the nano-structure and then to compare the electron emission performance before and after tip re-generation. For Experiment 4 bare tungsten emitter tips were exposed to elevated pressure conditions to compare with the re-generable emitters. Experiment 5 involved performing tip re-generation experiments inside of the FE-SEM to investigate the surface morphology of the emitter tips. Custom components were built and implemented in FE-SEM to provide an electrical interface to perform the experiments. Using the microscope it was possible to look at the nano- and micro-structure of the emitter tips after re-generating the tips and then to compare the results with Fowler-Nordheim tip radius estimations. The data were also compared with the results from Experiment 1 that were performed in the UHV facility, a facility that didn't have the capability to visually inspect the emitter tips.

As stated, this research was intended to examine the feasibility of employing regenerable electron sources with electric propulsion systems so there are a few things that are out of the scope of this work but are probable future steps. One future step is to build arrays of tightly packed emitter tips. Since each emitter tip can only provide on the order of 10 μ A, a large number of emitters would be necessary to supply enough electron current for most thrusters. Another future experiment should involve operating the re-generable electron sources near a plasma source. While Marrese presented findings from operating other field emission devices near a Hall-effect thruster environment, it would be necessary to examine regenerable emitters in a plasma environment since they are made from different materials than what Marrese tested.¹³

1.3 Document Roadmap

Following this introduction, Chapter 2 will provide a background and a review of prior work with field emission electron sources and liquid-metal-ion-sources (LMIS). Chapter 3 describes the experimental apparatus and methods used for each set of experiments in this dissertation. The following three chapters, Chapters 4 - 6, include results from each set of experiments that were performed. In particular, Chapter 4 includes applying the Fowler-

Nordheim model to experimental data to estimate emitter tip size. In Chapter 5, re-generable emitter tips are operated for long durations of time and at elevated pressure and then they are compared with bare tungsten field emitters that were exposed to the same conditions. In Chapter 6, emitter tips were operated in the Field Emission Scanning Electron Microscope to observe the nano-structure of re-generated emitter tips. Chapter 7 includes the conclusions for the research in this dissertation, as well as some potential applications for re-generable electron sources. The Appendices at the end of this document include results from a statistical analysis performed on the experimental data that is reported in Chapter 4, observations of anomalous behavior from one of the emitter tips tested in the FE-SEM, an explanation of *in situ* imaging of an operating re-generable source using the FE-SEM, and the results from an experiment that was performed as an undergrad research experiment on NASA's microgravity airplane.

References for Chapter 1

- [1] Marcuccio, S., A. Genovese and M. Andrenucci, "FEEP Thruster: Development Status and Prospects", *Second European Spacecraft Propulsion Conference*, 1997.
- [2] deGrys, K., N. Meckel, G. Callis, D. Greisen, A. Hoskins, D. King and F. Wilson, "Development and Testing of a 4500 Watt Flight Type Hall Thruster and Cathode", 27th International Electric Propulsion Conference, IEPC-01-011, Pasadena, CA, 2001.
- [3] Randolph, T. M. and J. E. Polk, "An Overview of the Nuclear Electric Xenon Ion System (NEXIS) Activity", 40th AIAA/ASME/SAE/ASEE Joint Propulsion Conference and Exhibit, AIAA-2004-3450, Fort Lauderdale, FL, 2004.
- [4] Goebel, D. M. and R. M. Watkins, "LaB6 Hollow Cathodes for Ion and Hall Thrusters", 41st AIAA/ASME/SAE/ASEE Joint Propulsion Conference & Exhibit, AIAA 2005-4239, Tucson, Arizona, 2005.
- [5] Carpenter, C. and M. Patterson, "High-Current Hollow Cathode Development", 27th International Electric Propulsion Conference, IEPC-01-274, Pasadena, California, 2001.
- [6] Marrese, C. M., "A Review of Field Emission Cathode Technologies for Electric Propulsion Systems and Instruments", *IEEE*, 85-97, 2000.
- [7] Gamero-Castano, M., V. Hruby, P. Falkos, D. Carnahan, B. Ondrusek and D. Lorents, "Electron Field Emission from Carbon Nanotubes, and Its Relevance in Space Applications", 36th Joint Propulsion Conference, AIAA-2000-3263, Huntsville, Alabama, 2000.
- [8] Gasdaska, C. J., P. Falkos, V. Hruby, M. Robin, N. Demmons, R. McCormick, D. Spence and J. Young, "Testing of Carbon Nanotube Field Emission Cathodes", 40th AIAA/ASME/SAE/ASEE Joint Propulsion Conference and Exhibit, AIAA 2004-3427, Fort Laurderdale, Florida, 2004.
- [9] Jonge, N. d., M. Allioux, M. Doytcheva, M. Kaiser, K. B. K. Teo, R. G. Lacerda and W. I. Milne, "Characterization of the field emission properties of individual thin carbon nanotubes", *Applied Physics Letters*, 85, 9, 1607-1609, 2004.
- [10] Saito, Y. and S. Uemura, "Field emission from carbon nanotubes and its application to electron sources", *Carbon*, 38, 169-182, 2000.
- [11] Aplin, K. L., C. M. Collingwood and B. J. Kent, "Reliability tests of gated silicon field emitters for use in space", *Journal of Physics D: Applied Physics*, 37, 2009-2017, 2004.
- [12] Bonard, J.-M., C. Klinke, K. A. Dean and B. F. Coll, "Degradation and failure of carbon nanotube field emitters", *Physical Review B*, 67, 11, 2003.
- [13] Marrese, C. M., "Compatibility of Field Emission Cathode and Electric Propulsion Technologies", *Doctoral Dissertation*, University of Michigan, 1999.

Chapter 2

2 Background and Review of Prior Research

The purpose of this chapter is to present a review of the concepts that are necessary to understand the research reported in this dissertation and to show what research has been done in the past to provide the foundation for this study. To begin, the phenomenon of electron field emission is discussed. Following is a discussion of liquid metal ion sources (LMIS) and how a LMIS can be used as a re-generable electron source. Then, there is an explanation of how regenerable cathode can be applied to space propulsion.

2.1 Electron Field Emission

Field emission typically refers to emission from metals into a vacuum environment and is defined as "the emission of electrons from the surface of a condensed phase into another phase, usually a vacuum, under the action of high (0.3-0.6 V/angstrom) electrostatic fields",¹ where 0.3-0.6 V/angstrom is equivalent to 3x10⁹ to 6x10⁹ V/m. Most commonly, field emission cathodes employ sharp nano-scale electrodes to establish electron emission by taking advantage of the locally enhanced electric field created by the nano-scale electrodes. Electric field enhancement is used to stimulate electrons to escape from the surface of the electrode into vacuum via a quantum tunneling effect known as Fowler-Nordheim emission. If the electric field at the sharp electrode is great enough ($\sim 10^9$ V/m) to deform the potential barrier, electrons can escape through the barrier at the metal surface, ¹ as illustrated in Figure 1.² The potential energy diagram in Figure 1 shows a conductive surface in the absence and in the presence of an external electric field. In order for an electron to escape from the surface the electron requires more energy than ϕ , the material's work function, which can be achieved thermionically or via photoemission. Since the emitted electrons require the addition of energy to escape they are considered hot. Field emission is achieved by deforming the potential barrier so that lower-energy, cold electrons can escape, as shown by the potential diagram in Figure 1 that was adapted from Charbonnier.² The height and width of the barrier are deformed so that cold electrons can escape and accelerate down the potential hill into vacuum.



Figure 1. Potential energy diagram adapted from Charbonnier² for electrons at a conductive surface showing the barrier for thermionic emission (without an electric field applied) and the deformed barrier for field emission due to an applied electric field. Also shown are two other types of electron emission that involve heat and electric field – thermal field emission and Schottky emission.

As mentioned, electric field enhancement is achieved by using sharp nano-scale electrodes as field emitters. For comparison, the electric field between two flat plates is governed by,

$$E = V/d$$

Equation 2.1

which means the voltage, V, must be increased or the gap spacing between the electrodes, d, must be very small for field emission of electrons to occur in order to meet the 10^9 V/m field emission criteria. For the flat plate example, achieving field emission simply isn't feasible due to the magnitude of voltage required to obtain the necessary electric field. Gomer explained it best when he said, "It is clearly not feasible or desirable to obtain such fields by brute force".¹ Therefore, implementing sharp needle-like electrodes to take advantage of their locally enhanced electric fields is desirable since the electric field follows,

$$E = \frac{2V}{r_t \ln \left(d/r_t \right)}$$
 Equation 2.2

where V is the potential difference between the emitter tip and extraction electrode, r_t is the emitter tip radius, and d is the gap spacing between the emitter tip and extraction electrode, as shown in Figure 2 with a typical electrical schematic. Equation 2.2 is only valid when r_t is less than d and when the emitter tip is a parabolic shape or the equipotentials formed by the emitter tip are parabolic in shape.



Figure 2. Single-needle field emission cathode schematic.

When Equation 2.2 is plotted, as shown in Figure 3, it becomes obvious that increasing the potential difference between the emitter and the extraction electrode increases the electric field but, more importantly, decreasing the emitter tip radius increases the electric field. While the gap spacing, *d*, is an important factor influencing the onset voltage of a field emitter (which is discussed in detail later in this section),³ the largest effect on the electric field is from the emitter tip radius. Using emitter tips with smaller radii allows for minimization of the extraction voltage that is necessary.



Figure 3. Plot showing the electric field enhancement from a parabolic field emitter.

The first theories to attempt to describe field emission were published back in the early 1920's by Schottky.⁴ In 1928, Fowler and Nordheim reported a detailed theoretical analysis with experimental electron field emission data,⁴ which etched their names in history since electron field emission is now synonymous with Fowler-Nordheim emission. In addition to being used as neutralizers for space propulsion devices, field emission electron sources are used in flat panel displays, focused electron beams for electron microscopy, and neutralization for spacecraft

mass spectrometry.⁵⁻⁸ Single needle field emission cathodes and Spindt-type arrays of field emission sites are the most commonly researched. In 1961 Gomer published a book devoted to explain the physics of electron field emission from single needle sources.¹ Single needle field emission cathodes are typically used in electron microscopy and can be readily purchased from a number of vendors. Commonly, refractory metals have been used as single-needle field emitters and include tungsten and molybdenum.⁹⁻¹¹ For space applications, the most common field emission cathode researched has been the Spindt-type array that was developed in the 1960's. Spindt-type arrays require micro-fabrication techniques and are typically made of a silicon sub-structure that is coated with a thin layer of molybdenum, tungsten, boron nitride, or diamond.^{5, 12} More recently interest has shifted towards using multi-wall carbon nanotube field emission arrays.^{13, 14}

Spindt-type arrays have many advantages as field emission electron sources. Micromanufacturing techniques allow for large arrays of micro-scale emitters to be produced relatively easily and the packing density of the arrays is high for small emitter tips, up to 10⁷ emitters/cm^{2.6} As mentioned, Spindt-type emitters can be made from a wide range of materials, including refractory metals like molybdenum and tungsten.^{12, 15} Also, microfabrication techniques can be used to implement a series of gated electrodes that can 'tune' the acceleration and to focus the beam of electrons from the tips, as well as to prevent ions from sputtering away the sharp emitter tips,⁵ leaving blunt tips. Although the gated electrodes help minimize sputtering, tip degradation is unavoidable.

Emitter tip degradation is an inherent problem with field emission cathodes and is commonly due to ion sputtering of the tip, excessive heating, and arcing between the emitter tip and the extraction electrode. Problems arise when the sharp emitter tips become blunt from one of the three mechanisms that were just mentioned. When the tip becomes blunt the local electric field enhancement diminishes, which causes failure of the emitter as a field emission electron source. The effect of diminishing tip radius is easy to see from looking back to the electric field equation shown previously in Equation 2.2. As r_t is increased, the field enhancement decreases. A micrograph of a tungsten emitter tip that has been operated at an excessive emission current (> 100 μ A) for multiple minutes, and therefore has experienced excessive heating, is shown in Figure 4. Implementing complicated circuitry can minimize arcing,⁸ however, eliminating tip degradation is impossible.



10μm SEI 1700X Damaged Tungsten Emitter Tip

Figure 4. Micrograph of a damaged tungsten field emitter that has been operated at > 100 μ A of emission current for multiple minutes.

In 1999 Marrese published extensive studies showing the feasibility of using Spindt-type arrays in the vicinity of Hall-effect thrusters.¹⁵ Results from the research were promising in xenon environments but measures had to be taken in order for the field emission arrays (FEA) to survive in the neutral gas environment. In order for molybdenum or silicon FEAs to survive in pressures of 2×10^{-5} Torr or greater, the extraction voltages for the molybdenum and silicon FEAs had to be less than 19 V and 30 V, respectively, unless a series of extraction and acceleration electrodes were implemented. Also, to prevent the tips from catastrophically arcing, currentlimiting circuitry had to be implemented into the system. Ultimately, Marrese determined that field emission arrays are feasible for electric propulsion neutralizers but careful design considerations must be made and the physical location of the FEAs near a plasma source is critical. In addition, depending on the environment larger arrays may be necessary because of limitations of electron emission current due to space-charge. Other studies on FEAs revealed the possibility that most of the emission current from an operating Spindt-type array comes from only a fraction of the "sharpest tips" and as the sharpest tips fail emission must continue from the blunter tips, causing degradation to the arrays to happen faster and faster as the sharp tips fail.¹⁶

Since about 2000 there has also been a lot of interest in using multi-wall carbon nanotubes (CNTs) as neutralizers for spacecraft.¹⁷ Carbon nanotubes are ideal field emitters due to their mechanical properties, ease and cost of manufacturing, and, most importantly, their high aspect-ratio.¹⁸ The diameter of CNTs is so small that extraction potentials are minimal to establish electron emission. Busek Co. Inc. performed some initial studies with CNTs to determine if they were feasible to replace traditional hollow cathodes for low power Hall and ion thrusters.¹⁷ Busek used a proprietary technique to grow a tangled web of CNTs to form a fibrous mat. Busek's cathodes have been used to demonstrate lifetime experiments in which the multi-wall CNT field emission cathodes were operated for over 13,000 hours at electron currents of 100 μ A and for approximately 6,000 hours at electron currents of about 1 mA in a vacuum environment of 10⁻⁸ to 10⁻⁹ Torr.^{14, 19} However, just as with other field emission cathodes the extraction voltage necessary to maintain a constant emission current increased

over time which implies that some of the emitter tips became damaged or destroyed over time. For the 13,000 hour test at 100 μ A of emission current the extraction voltage was gradually increased from about 400 V at the beginning to approximately 1000 V when the experiment was concluded. Additional experiments were reported where an operating CNT was exposed to high-purity oxygen. During those experiments the emission current decreased from about 130 μ A at 10⁻⁶ Torr to about 60 μ A at high 10⁻⁴ Torr over a 15 minute period. When the chamber pressure was restored to 10⁻⁶ Torr the emission current returned to the original setpoint.

However, CNTs can, and do, become damaged by a number of methods during operation as field emission electron sources. Much like the Spindt-type arrays, single-needle emitters, and any other field emitter, damage to CNTs has been observed via field evaporation, ion bombardment, oxidation, thermal failure due to excessive emission current, and arcing.^{18, 20} It has also been noted that the stress due to the electric field on CNTs can cause individual nanotubes to be removed from the underlying substrate.^{14, 21}

For all of the reasons listed previously, optimum operation of a field emission electron source requires maintaining high local electric field enhancement. Since the local electric field is inversely proportional to the electrode tip radius, the sharper the emitter tip, the lower the electric potential needed to obtain electron field emission.⁵ A typical bare tungsten singleneedle field emitter has a tip radius on the order of 10's of nm to a few microns and can produce up to $100 \,\mu A$ of electron emission current.¹ As mentioned, electron field emission is commonly referred to as Fowler-Nordheim emission, after the researchers who proposed one of the first adequate theories for how the field emission process works. Emission characteristics are governed by the Fowler-Nordheim equation,

$$\frac{I}{V^2} = a \exp\left(\frac{-b' \phi^{3/2}}{V_o}\right)$$
 Equation 2.3

where a and b' are,

$$a = A \left[6.2 \times 10^{6} \left(\mu/\phi \right)^{1/2} \left(\mu + \phi \right)^{-1} \left(\alpha k r_{t} \right)^{-2} \right]$$
 Equation 2.4
b' = 6.8×10⁷ \alpha k r_{t}. Equation 2.5

In this series of equations I is the emission current measured in Amperes, V_o is the extraction voltage measured in volts, ϕ is the work function in eV, A is the total emitting area, α is the Nordheim image-correction factor, and kr_t is the field voltage proportionality factor.¹ When the extraction voltage is increased high enough to obtain electron emission current, the voltage where current is first established is called the onset voltage, V_o . As the extraction voltage is increased beyond the onset voltage there is an exponential relationship between the emission current and the extraction voltage, as shown in Equation 2.3 and in Figure 5. Another characteristic of the I-V curve is apparent when the extraction voltage can be decreased below the onset voltage while continuing to emit current. This is an important concept that is discussed further in Chapter 4.



Figure 5. Diagram showing the current-voltage relationship of Fowler-Nordheim emission from an electron field emitter.

For the experiments reported in this dissertation, the work function that was used for tungsten was 4.5 eV²² and the work function of indium was 3.8 eV.²³ When the Fowler-Nordheim model is applied to electron I-V data, the graph of $\ln(I/V_o^2)$ versus I/V_o is linear, as shown in Figure 6, and has an intercept of $\ln(a)$ and a slope of $b'\phi^{3/2}$.



Figure 6. A sample Fowler-Nordheim plot of electron field emission.

Equating the slope with Equation 2.5 and inputting values for α and k, the tip radius, r_t , can be approximated.¹ In practice, α is an image-correction factor that varies between 0.9 and 1.0 and k is used to account for the field distribution at the emitter tip due to the tip's geometry and is given by,

$$k = \frac{1}{2} \ln(d / r_t)$$
 Equation 2.6

for parabolic emitters, where r_t is the emitter tip radius and d is the gap spacing between the emitter tip and the extraction electrode. The value typically used for k is 5, which allows for an emitter tip approximation to within 20% when coupled with estimating α at 1.0. A more detailed description of error analysis is included with experimental data in Chapter 4. To estimate emitter tip radii for all of the data reported in this dissertation, the Fowler-Nordheim model was applied to experimental data.

2.2 Liquid Metal Ion Sources

To operate a liquid metal ion source an electric field is created near the surface of a low melting-temperature liquefied metal, such as indium, by a downstream extraction electrode. At the surface of the liquid-metal two main forces are responsible for ion emission, the electrostatic attractive force, F_{Er} , on the liquid metal due to the extraction electrode and the surface tension force, F_{ST} , on the liquefied surface of the metal, as shown in Figure 7.²⁴ The electrostatic force is independent of field polarity and causes charge separation at the surface of the liquid metal which 'pulls' the liquid metal towards the extraction electrode. The surface tension force is caused by cohesive intermolecular forces between the like-molecules in the liquid metal reservoir.



Figure 7. Diagram showing the interacting electrostatic and surface tension forces at a liquefied metal's surface, which are the forces responsible for ion emission.

As early as 1600 Gilbert had reported that a liquefied conductor deformed when exposed to an electric field.²⁵ Gilbert noticed when a spherical drop of water was placed on a dry surface and then a charged material was brought close to it, the sphere was attracted toward the charged material. After Gilbert's observations, over a hundred years went by until scientists started investigating the electrified liquids in more detail. Around 1745 Bose reported that "water threads" formed at the nozzle of an electrified capillary. And in 1882 Lord Rayleigh first calculated the electric stress on conducting fluids. In 1917, Zeleny observed jets forming from fluids exposed to an electric field and in 1931 Macky reported water droplet deformation when the drops were exposed to an electric field.^{26, 27} It wasn't until over 300 years after Gilbert first observed fluid interaction with electric fields that Taylor proposed the first adequate theory for their observations. In the mid 1960's, Taylor showed that when a conductive liquid is exposed to an electric field, the liquid is attracted towards the field and forms a cone with a half angle of 49.3°.²⁸ Taylor's investigations and contributions to understanding the physics surrounding the interaction between liquids and electric fields led to the 98.6° cone formed by the delicate balance between the electric field force and the surface tension force of a liquid to be named the Taylor cone. Some refer to the cone as a Gilbert-Taylor cone to credit Gilbert's original observations. The cone will be referred to as a Taylor cone for the remainder of this document.

When an ion extracting field is used it is possible to establish a stable beam of ions from the liquefied metal. Research reported the in the late 1950's and early 1960's realized the potential that liquid metal ion sources had for space propulsion.²⁹ It wasn't until the 1980's that research really started producing significant results with liquid metal ion sources and in the 1990's two main types of field emission electric propulsion devices emerged in response to the need for a high precision and low-thrust (1 μ N to 1 mN) propulsion system for science missions.^{30, 31} Cesium-Field Emission Electric Propulsion (FEEP) thrusters using a slit-type geometry were developed by Centrospazio and ALTA in Italy and indium-FEEP thrusters using single-needles were being developed by Austrian Research Center's Seibersdorf Research.^{30, 31} Also in the 1990's liquid metal ion sources found extensive use as ion sources of high brightness in focused ion beam materials processing applications.³²

As discussed in the previous section, electron field emitters can be easily damaged. For that reason, researchers made attempts at extracting electrons from liquefied metals, since a

liquid metal is essentially impossible to damage and Taylor cone formation is independent of field polarity. The earliest documentation of using a Taylor cone as a liquid-metal *electron* source (LMES) was the work of Swanson and Schwind in 1978.³³ Because the formation of a Taylor cone is independent of field polarity, Swanson and Schwind applied electron-extracting fields to a liquid metal in an effort to obtain electron emission from the liquid cone. Their experiments showed that stable electron emission was impossible to achieve from a liquid metal and they observed pulses of emission current during their experiments operation. They referred to the emission as "explosive" since small portions of the liquid metal were expelled as the emission pulsed. The phenomena responsible for pulsed electron emission was supported by Gomer the following year.³⁴ Two possible explanations for the inability to obtain stable DC electron emission and absence of a space-charge, both of which are thought to have stabilizing effects on an operating liquid metal ion source.^{35, 36}

One main condition must be met in order to induce ion emission from liquid metal, the liquid metal must be deformed by the electric field. This is accomplished when the pressure outside of the liquid (p_{ext}) due to the applied electric field exceeds the pressure of the surface tension (p_{int}) within the liquid indium at the tip of the tungsten electrode,

$$p_{ext} \ge p_{int}$$
. Equation 2.7

The surface tension pressure is fixed once the tungsten electrode has been etched to a desirable geometry and coated with a thin film of metal. The surface tension pressure on the liquid indium can be calculated using the following relationship,

$$p_{int} = \frac{2\gamma}{r_t}$$
 Equation 2.8

where γ is the surface tension of the liquid metal, for indium 592 dyne/cm (0.592N/m) at 185°C, and r_t is the emitter tip radius.^{37, 38}

For ion emission to occur the applied external electrostatic pressure must be greater than p_{int} The external pressure is,

$$p_{ext} = \frac{1}{2} \varepsilon_o E_o^2$$
 Equation 2.9

where E_o is the electric field at the tip apex and ε_o is the permittivity of free space, $8.854 \times 10^{-12} F/m$.³⁹ By applying a potential of V_o to the extraction electrode, the electric field approximation in Equation 2.2 for a parabolic emitter tip shape is valid;

$$E_o = \frac{2V_o}{r_t \ln(d/r_t)}$$

Equation 2.10

where, r_{t} is the tip radius and d is the tip-to-extraction electrode spacing.

When the electric field force (p_{ext}) on the liquid metal exactly balances with the surface tension force (p_{int}) the Taylor cone is formed, as shown in Figure 8.^{25, 40} It was found that the Taylor cone has a half-angle of 49.3°. It should be noted that the equilibrium condition of $p_{ext} = p_{int}$ is extremely delicate and nearly impossible to stably maintain. Therefore, as p_{ext} is increased and the electrostatic attraction becomes greater on the liquid metal, a jet-like protrusion of liquefied metal forms at the apex of the Taylor cone.



Figure 8. Taylor cone and jet-like protrusion schematic on a liquid metal ion source

Because the Taylor cone has a sharp tip, geometric enhancement of the local electric field at the cone apex is sufficient to extract metal ions directly from the liquid. In most metals the ions emerge from a very narrow (few nanometer diameter) liquid jet at the cone apex and are subsequently accelerated by the electric field to either produce thrust^{30, 31} (FEEP) or for materials processing applications (LMIS).³²

As the liquid metal is depleted from the apex during ion emission, the indium must be replenished in order to continue ion source operation. The mechanism used to supply the indium is by creating a series of micro-grooves⁴¹ on the underlying tungsten electrode. The micro-grooves, shown in Figure 9, act as capillaries to feed indium to the apex.



SEI AC etched tip 1000x

Figure 9. Micrograph showing the longitudinal grooves that act as capillaries to aid indium flow to the emitter apex during ion source operation.

The grooves are used to create a pressure difference to transport indium to the needle apex using capillary forces and can be estimated by the following;

$$\Delta P \approx \frac{\gamma \cos \theta}{r}$$

Equation 2.11

where ΔP is the capillary pressure, γ is the surface tension, θ is the contact angle, and r_g is the groove radius, as shown in Figure 10. For an indium coating on a tungsten electrode the surface tension and contact angle, γ , θ , are approximately 0.592 N/m and 9°, respectively.⁴²



Figure 10. Capillary action diagram of a cross-section of a tube placed in a reservoir of liquid, indicating the physical attributes of Equation 2.10. In this case the tube would represent tungsten and the reservoir of liquid would be indium.
The pressure difference causes a volumetric flow rate of indium to the emitter apex as follows;

$$\dot{V} \simeq \frac{\pi \Delta P r_g^4}{8\eta l}$$
 Equation 2.12

where η is the viscosity and l is the length over which the pressure drop, ΔP , occurs (corresponding to the longitudinal groove length).^{41, 43} Swanson and Li have used gallium to show that 10's of μ A of emission current can be obtained from each micro-capillary that has a 1 μ m groove radius and 2 mm groove length,⁴³ while Bell and Swanson have shown that up to 1 A of emission current is feasible from each groove of radius 100 μ m and length of 300 μ m.⁴¹ The variability in the groove radius and length allows for flexibility in emitter tip design. For indium, η is approximately $16.5 \times 10^{-3} poise$ ($1.65 \times 10^{-3} kg/m \cdot s$),⁴⁴ which compares with gallium at $20 \times 10^{-3} poise$.⁴³

2.3 Dual Electron/Ion Sources

In 1989, Rao et al found that it was possible to obtain DC electron emission using gallium and indium if a source was first operated as a liquid metal ion source (LMIS) and then the heat was quickly reduced, solidifying the liquid metal so that the Taylor cone was "frozen in" (quenched). Once the sharp Taylor cone was quenched, electron emission could be induced from a solid, rather than liquid, metal tip.³⁶ In 1997 Chen and Wang proposed using a dual electron/ion source for microlithography and micro-imaging.²³ Up until then, integrating a focused ion beam (FIB) for microlithography and a scanning electron microscope (SEM) for in situ imaging involved using two separate sources with complicated alignment.⁴⁵ Although using a single focused ion beam for lithography and then sample inspection, like an SEM, is possible the ion beam causes sputtering of the sample due to the large momentum of the ions, which is undesirable. In 1997 Chen and Wang demonstrated the feasibility of their dual source concept by operating a dual electron/ion source from an indium-coating on a tungsten needle.²³ In 1998 Chen and Wang demonstrated the first focused ion/electron beam (FIEB) system for lithography and imaging from a single emission source.⁴⁶ The following year, Saito et al observed very interesting behavior from an indium-coated tungsten emitter. They saw that the electron emission pattern from a quenched indium LMIS on a circular phosphor screen wasn't a single point-source but had a circular emission pattern,⁴⁷ implying emission from more than one quenched nano-structure. In 2001 Sheu and Wang reported results using an indium-gold alloy for a more stable dual electron/ion FIEB emission source.⁴⁸ Two years later, in 2003, Hsieh et al demonstrated the use of an indium-bismuth alloy as a dual electron/ion source that had comparable emission properties as the indium-gold alloy but could be operated at much lower temperature.49

2.4 Summary

How does all of the information in this chapter tie together? The most important reason is that each type of electron source that has been studied shares common characteristics and each source can be catastrophically damaged due to a number of factors. Some of the factors that were mentioned include thermal failures, arcing, and sputtering of the field-emitting nano-structures. The beauty of the dual electron/ion source is that the sharp nano-structures can be generated over and over, as long as there is a sufficient coating of liquid metal

to form the sharp emitters. Clearly, employing a neutralizer on a spacecraft that has the ability to re-generate itself when it becomes damaged would be extremely advantageous to increase the lifetime of a mission.

References for Chapter 2

[1] Gomer, R., <u>Field Emission and Field Ionization</u>. New York, NY: American Institute of Physics, 1961.

- [2] Charbonnier, F., "Developing and using the field emitter as a high intensity electron source", *Applied Surface Science*, 94/95, 26-43, 1996.
- [3] Liu, P., F. Arai, L. Dong, T. Fukuda, T. Noguchi and K. Tatenuma, "Field Emission of Individual Carbon Nanotubes and its Improvement by Decoration with Ruthenium Dioxide Super-Nanoparticles", *Journal of Robotics and Mechatronics*, 17, 4, 475-476, 2005.
- [4] Fowler, R. H. and L. Nordheim, "Electron Emission in Intense Electric Fields", Proceedings of the Royal Society of London. Series A, Containing Papers of a Mathematical and Physical Character, 119, 781, 173-181, 1928.
- [5] Marrese, C. M., "A Review of Field Emission Cathode Technologies for Electric Propulsion Systems and Instruments", *IEEE*, 85-97, 2000.
- [6] Spindt, C. A., C. E. Holland, A. Rosengreen and I. Brodie, "Field-Emitter Arrays for Vacuum Microelectronics", *IEEE Transactions on Electron Devices*, 38, 10, 2355-2363, 1991.
- [7] Goldstein, J., <u>Scanning Electron Microscopy and X-Ray Microanalysis</u>, <u>3rd Ed.</u> New York, NY: Springer, 2003.
- [8] Curtis, C. C. and K. C. Hsieh, "Spacecraft mass spectrometer ion source employing field emission cathodes", *Rev. Sci. Instrum.*, 57, 989, 1986.
- [9] Dyke, W. P., F. M. Charbonnier, R. W. Strayer, R. L. Floyd, J. P. Barbour and J. K. Trolan, "Electrical Stability and Life of the Heated Field Emission Cathode", *Journal of Applied Physics*, 31, 5, 790-805, 1960.
- [10] Martin, E. E., J. K. Trolan and W. P. Dyke, "Stable, High Density Field Emission Cold Cathode", *Journal of Applied Physics*, 31, 5, 782-789, 1960.
- [11] Song, Y., E. Garate and N. Rostoker, "High current density electron beam generation from field emission tip cathodes", *Journal of Applied Physics*, 76, 1, 609-611, 1993.
- [12] Park, Y. J., J. H. Choi, H. W. Lee, N. S. Lee, J. W. Kim, S. S. Hong, N. S. Park, J. E. Jung and J. M. Kim, "Fabrication of Spindt-type tungsten microtip field emitter arrays with optimized aluminum parting layers", J. Vac. Sci. Technol. B 18, 2, 2000.
- [13] Spindt, C. A., "A Thin-Film Field-Emission Cathode", *Journal of Applied Physics*, 39, 3504-3505, 1968.
- [14] Gasdaska, C. J., P. Falkos, V. Hruby, M. Robin, N. Demmons, R. McCormick, D. Spence and J. Young, "Testing of Carbon Nanotube Field Emission Cathodes", 40th AIAA/ASME/SAE/ASEE Joint Propulsion Conference and Exhibit, AIAA 2004-3427, Fort Laurderdale, Florida, 2004.
- [15] Marrese, C. M., "Compatibility of Field Emission Cathode and Electric Propulsion Technologies", *Doctoral Dissertation*, University of Michigan, 1999.
- [16] Aplin, K. L., C. M. Collingwood and B. J. Kent, "Reliability tests of gated silicon field emitters for use in space", *Journal of Physics D: Applied Physics*, 37, 2009-2017, 2004.
- [17] Gamero-Castano, M., V. Hruby, P. Falkos, D. Carnahan, B. Ondrusek and D. Lorents, "Electron Field Emission from Carbon Nanotubes, and Its Relevance in Space Applications", 36th Joint Propulsion Conference, AIAA-2000-3263, Huntsville, Alabama, 2000.
- [18] Bonard, J., H. Kind, T. Stockli and L. Nilsson, "Field emission from carbon nanotubes: the first five years", Solid-State Electronics, 45, 893-914, 2001.

- [19] Ziemer, J. K., M. Gamero-Castaño, V. Hruby, D. Spence, N. Demmons, R. McCormick and T. Roy, "Colloid Micro-Newton Thruster Development for the ST7-DRS and LISA Missions", 41st AIAA/ASME/SAE/ASEE Joint Propulsion Conference and Exhibit, AIAA 2005-4265, Tucson, Az, 2005.
- [20] Bonard, J.-M., C. Klinke, K. A. Dean and B. F. Coll, "Degradation and failure of carbon nanotube field emitters", *Physical Review B*, 67, 11, 2003.
- [21] Deline, C. A., T. M. Liu, D. P. Morris and B. E. Gilchrist, "Demonstration of Field Emission Cathode Operation in a Plasma Environment", 41st AIAA/ASME/SAE/ASEE Joint Propulsion Conference & Exhibit, AIAA-2005-3663, Tucson, AZ

2005.

- [22] Lee, Y.-H., C.-H. Choi, Y.-T. Jang, E.-K. Kim, B.-K. Ju, N.-K. Min and J.-H. Ahn, "Tungsten nanowires and their field electron emission properties", *Applied Physics Letters*, 81, 4, 745-747, 2002.
- [23] Chen, L. W. and Y. L. Wang, "Stable field-induced electron emission from a solidified liquid metal ion source", Appl. Phys. Lett., 72, 3, 389-391, 1997.
- [24] Suvorov, V. G. and E. A. Litvinov, "Dynamic Taylor cone formation on liquid metal surface: numerical modeling", *Journal of Applied Physics*, 33, 11, 1245-1251, 2000.
- [25] Gilbert, W., <u>De Magnete</u>. London: 1600.
- [26] Zeleny, J., "Instability of Electrified Liquid Surfaces", The Physical Review, X, 1, 1917.
- [27] Macky, W. A., "Some Investigations on the Deformation and Breaking of Water Drops in Strong Electic Fields", *Proc. Roy. Soc. Lond. Series A*, 133, 822, 565-587, 1931.
- [28] Taylor, S. G., "Disintegration of water drops in an electric field", *Proceedings Royal Society*. *Mathematical, physical, and engineering sciences*, 280, 1382, 383-397, 1964.
- [29] Mackenzie, R. A. D. and G. D. W. Smith, "Focused ion beam technology: a bibliography", Nanotechnology, 1, 163-201, 1990.
- [30] Tajmar, M., A. Genovese and W. Steiger, "Indium Field Emission Electric Propulsion Microthruster Experimental Characterization", *Journal of Propulsion and Power*, 20, 2, 211-218, 2004.
- [31] Marcuccio, S., A. Genovese and M. Andrenucci, "FEEP Thruster: Development Status and Prospects", *Second European Spacecraft Propulsion Conference*, 1997.
- [32] Driesel, W., C. Dietzsch and R. Muhler, "In situ observation of the tip shape of AuGe liquid alloy ion sources using a high voltage transmission electron microscope", *Journal of Vacuum Science and Technology B*, 14, 5, 3367-3380, 1996.
- [33] Swanson, L. W. and G. A. Schwind, "Electron emission from a liquid metal", *Journal of Applied Physics*, 49, 11, 5655-5662, 1978.
- [34] Gomer, R., "On the Mechanism of Liquid Metal Electron and Ion Sources", *Applied Physics*, 19, 4, 365-375, 1979.
- [35] Driesel, W., C. Dietzsch, H. Niedrig and B. Praprotnik, "HV TEM in situ investigations of the tip shape of a gallium liquid-metal ion/electron emitter", *Ultramicroscopy*, 57, 45-58, 1995.
- [36] Rao, K. A., A. E. Bell, G. A. Schwind and L. W. Swanson, "A combination electron/ion field emission source", *Journal of Vacuum Science and Technology B*, 7, 6, 1793-1797, 1989.
- [37] Prewett, P. D. and G. L. R. Mair, <u>Focused Ion Beams from Liquid Metal Ion Sources</u>. Somerset, England: Research Studies Press Ltd, 1991.

- [38] Shackelford, J. F. and W. Alexander, <u>CRC Materials Science and Engineering Handbook</u>. 3rd. Boca Raton: CRC Press, 2001.
- [39] Griffiths, D. J., Introduction to Electrodynamics. 2nd. New Jersey: Prentice Hall, 1989.
- [40] Taylor, S. G., "Electrically driven jets", Proc. Roy. Soc. Lond., A, 313, 453-475, 1969.
- [41] Bell, A. E. and L. W. Swanson, "The Influence of Substrate Geometry on the Emission Properties of a Liquid Metal Ion Source", *Applied Physics A*, 41, 335-346, 1986.
- [42] Wright, J., "Report on the wetting of tungsten thin film surfaces by liquid indium." *Technical Report*, Alameda Applied Sciences Corporation, San Leandro, CA, 2006.
- [43] Swanson, L. W. and J. Z. Li, "The influence of electrode geometry on liquid metal ion source performance", *Journal of Vac. Sci. Technol. B*, 6, 4, 1062-1065, 1988.
- [44] Culpin, M. F., "The Viscosity of Liquid Indium and Liquid Tin", *Physical Society, Section B*, 70, 11, 1069-1078, 1957.
- [45] Tamura, H., N. Saitou and K. Unemura, "Hybrid Charged Particle Apparatus", U.S. Patent 4,740,698. 1988.
- [46] Chen, L. W. and Y. L. Wang, "Operation of a single column focused ion/electron beam system based on a dual ion/electron source", *Applied Physics Letters*, 73, 15, 2212-2214, 1998.
- [47] Saito, Y., H. Nakane and H. Adachi, "Field electron emission from W covered with In", *Journal of Vac. Sci. Technol. B*, 18, 2, 1093-1096, 1999.
- [48] Sheu, B. L. and Y. L. Wang, "Emission properties of a dual ionÕelectron source based on Au– In alloy", Applied Physics Letters, 80, 8, 1480-1482, 2001.
- [49] Hsieh, W.-P., B. L. Sheu and Y. L. Wang, "Emission properties of a dual ion/electron point emitter based on In–Bi alloy", *Applied Physics Letters, American Institute of Physics*, 83, 11, 2277-2279, 2003.

Chapter 3

3 Experiments

The experiments presented in this dissertation were intended to lay the groundwork necessary to determine if re-generable field emission cathodes have potential to be implemented with electric space propulsion systems. To that end, a series of experiments have been designed and performed and the equipment and procedures that were used to perform the experiments will be described within this Chapter. The experiments that are reported in this dissertation were designed (1) to examine the electron emission current-voltage (I-V) characteristics of a re-generable electron source, (2) to investigate the lifetime of the regenerable electron source, (3) to determine the robustness of the re-generable electron source against elevated neutral pressure conditions, (4) to compare the exposure time and maximum-pressure-before-failure of re-generated emitters with pure tungsten field emitters, and (5) to visually investigate the surface morphology and emitter tip geometry after re-generating the electron sources.

Chapter 4 includes experiments used to study the effects that ion quenching current had on the electron I-V characteristics from the re-generated emitter tips. Fowler-Nordheim modeling of experimental data was used to estimate the tip radii using the electron emission I-V data. Chapter 5 includes the experiments that were designed to examine I-V characteristics of re-generable emitter tips that were operated for long durations in UHV conditions and to determine how long electron emission from the re-generable emitters lasted when operated in an environment of increasing neutral pressure. The data were compared with pure tungsten field emitters that were operated for long durations of time and exposed to elevated vacuum pressure. The experiments in Chapter 6 were performed in an effort to observe the regenerated emitter tip morphology using a Field Emission Scanning Electron Microscope (FE-SEM). Experiments were performed inside of the FE-SEM to directly determine the guenched emitter tip radii to compare with the estimates made using the Fowler-Nordheim model in the previous experiments. Past researchers have observed operating liquid metal ion sources¹⁻⁵ but no investigation had been performed to look at quenched LMISs. An additional experiment is included in Appendix D that was performed on NASA's microgravity C-9B aircraft⁶ where LMISs were operated and quenched in a microgravity environment as part of an undergraduate research experiment.

3.1 Equipment and Experimental Setup

The purpose of this Chapter is to describe the experimental setup that was used for all of the experiments that were performed. For each experiment there were slight modifications to the experimental apparatus but they were minimal and a schematic and picture of each are included to highlight the differences – but most importantly the similarities. Also, separate vacuum facilities were used for some of the experiments so each facility will be described.

3.1.1 Re-generable Field Emitters

The re-generable electron source that was used for the research reported in this document was fabricated in-house. The re-generable source, more commonly known as a liquid

metal ion source, was made by electrochemically etching a 0.25 mm diameter tungsten wire and then coating the tungsten wire with a layer of indium. The indium-coated tungsten electrode was then fixed to a Teflon block using stainless steel hardware, as shown in Figure 11.



Figure 11. Re-generable electron source.

To electrochemically etch the tungsten wire, the wire was suspended in the center of a 250-mL cylindrical beaker with a cylindrical stainless-steel mesh placed along the perimeter of the beaker, as shown in the cross section schematic in Figure 12. The beaker was then filled with a 2M solution of NaOH.



Figure 12. Electrochemical etching set-up.

By applying a DC bias between the partially submerged tungsten wire (anode) and the immersed stainless-steel electrode (cathode), the following reaction takes place at the air/electrolyte surface:⁷

Cathode
$$6H_2O + 6e^- \rightarrow 3H_2(g) + 6OH^-$$

Anode $W(s) + 8OH^- \rightarrow WO_4^{2-} + 4H_2O + 6e^-$
 $W(s) + 2OH^- + 2H_2O \rightarrow WO_4^{2-} + 3H_2(g)$

The reaction causes the oxidative dissolution of tungsten to soluble tungstate (WO_4^{2-}) anions at the tungsten anode and reduces the water to hydrogen gas bubbles and OH^- ions at the immersed stainless-steel cathode. ⁷ This type of DC electrochemical etching yields a sharpened tungsten electrode with a smooth surface finish and emitter tip radius of 10's of nm

to 10's of μ m depending on the current and voltage chosen for the DC power supply.⁷ Unfortunately, most liquid metal coatings on smoothly etched needles tend to pull away from the needle apex in an effort to minimize area and surface energy. Therefore, one additional electrochemical etch had to be performed to obtain a surface finish with longitudinal microgrooves on the electrode.⁸

The additional etch was performed using a function generator to produce a sinusoidal AC waveform.⁸ The AC conditions cause modification of the tungsten electrode's surface rather than at the air/electrolyte interface. For this etching procedure, the majority of the tungsten electrode must be submerged in the 2M NaOH solution. At a minimum, the area that requires indium coating must be submerged. By biasing the electrode using the AC conditions mentioned, the geometry of the micro-grooves can be altered by adjusting the amplitude and frequency of the sinusoidal input. The exposure time of the tungsten electrode to the AC conditions also can alter the surface finish. One example of an etched electrode is shown in Figure 13. The grooves act as capillaries that aid indium flow to the tip of the electrode.⁸



Figure 13. Electrochemically etched tungsten electrode showing longitudinal micro-grooves that act as capillaries. The capillaries supply liquid indium to the electrode tip.

The electrochemically etched tungsten emitters were coated with indium by heating the sharpened electrodes in a vacuum environment of 10^{-6} Torr in an effort to evaporate any oxide coating. The tungsten emitters were resistively heated at a current of 3.5 - 4.0 Amperes (at 2.0 - 2.5 V) so that the emitter tip would glow bright orange, characteristic of over 900°C on a metal temperature color chart. Then, the glowing tungsten emitter was dipped in a crucible of liquefied indium (heated to 650-700°C) multiple times. The reason for dipping multiple times was to break through any oxide layers on the surface of the indium reservoir and to evenly coat the tungsten emitter was turned off while the tungsten emitter tip was immersed in the indium reservoir and then the emitter tip was extracted immediately, leaving an indium coating on the tungsten emitter. A schematic of the dipping apparatus is shown in Figure 14.



Figure 14. Schematic showing the rotary-motion indium-coating apparatus, the tungsten emitter, and the indium crucible.

The vacuum environment was necessary to minimize oxidation of the indium during the coating process. The etched and coated tungsten emitter was then cooled, removed from the vacuum environment, and then inserted into the Teflon fixture with the planar stainless-steel extraction electrode placed approximately 0.5 mm away, as shown in the schematic in Figure 15. Each experiment that was mentioned previously and that will be described in great detail in the next three Chapters use variations of the experimental apparatus shown but each had the same general components and geometry.



Figure 15. Re-generable electron source experimental apparatus.

A bipolar high-voltage power supply was used for the extraction supply. The heater supply that was used to melt the metal coating could supply 5 A, 18 V, however, only 2-3 A at 1-2 V was necessary to liquefy the indium during testing. The emission current in each experiment was recorded using a custom micro-ammeter that was designed and built by Washeleski and Makela in the Isp Lab. The ammeter specifications will be described in more detail in Section 3.1.3. An electrical schematic of the experimental apparatus is shown in Figure 16.



Figure 16. Electrical schematic for the re-generable electron source.

For size reference, a picture of the re-generable source that was used in the Field Emission Scanning Electron Microscope is shown in Figure 17. The re-generable source is shown on the left-hand-side and an electrical interface that will be described in detail in Chapter 6 is on the right.



Figure 17. Re-generable field emitter apparatus and the custom electrical interface mounted inside the FE-SEM that will be discussed in Chapter 6.

Although a planar extraction electrode was used for the experiments reported in this dissertation, the extraction electrode for a flight-model re-generable source would have a hole in the extraction electrode to allow the ion or electron beam to escape. Therefore, a brief analysis was performed to investigate how the extraction voltage, emitter tip-to-extraction electrode gap-spacing, and extraction hole diameter influence the local electric field. Using Ansoft's Maxwell SV software, electric field simulations were performed to determine what factors had the greatest influence on the local electric field enhancement (excluding the obvious field enhancement from a sharp emitter tip). The simulations were performed with the geometry shown in Figure 18.





A flat metal plate, shown in dark gray, was held at ground potential and another flat electrode, also shown in dark gray, was positioned a distance, *d*, from the bottom metal electrode. An insulating sheet of Teflon, shown in light gray, was placed between the two electrodes and a hole of radius r_h was simulated in the Teflon and the top electrode. The top electrode was biased at a voltage, *V*, with respect to the bottom electrode for the simulations. For the first set of simulations the gap height was set constant at 0.5 mm (since 0.5 mm is commonly used in practice) and the extraction voltage was held constant at -10 kV. The hole-diameter, r_h , was varied from 25 μ m to 5 mm. The maximum electric field was recorded and plotted, as shown in Figure 19.



Hole Radius (mm)

Figure 19. Electric field simulation data with a fixed extraction voltage of -10 kV and fixed gap height of 0.5 mm while varying the extraction electrode hole radius from 25 μ m to 5 mm.

The electric field appears to vary drastically when the hole radius is increased from 25 μ m up to 5 mm, however, looking at the scale the electric field stays relatively constant at about 2 to 2.1 x 10⁷ V/m. The simulation indicates that the hole radius doesn't play a large role in the electric field that can be achieved. The second simulation was executed while holding the hole radius constant at 0.5 mm and the extraction voltage constant at -10 kV while increasing the gap height from 50 μ m to 1 mm. The electric field that was estimated using the simulation is shown in Figure 20.



Figure 20. Electric field simulation data with a fixed extraction voltage of -10 kV and fixed hole radius of 0.5 mm while varying gap height from 50 μ m to 1 mm.

Unlike the hole-diameter, the gap height plays a large role in the electric field that is generated. The simulation results show over an order of magnitude increase in electric field can be created by decreasing the distance between the two electrodes over the plotted range. With micro-manufacturing techniques, gap spacing has the potential to be reduced greatly which decreases the voltage required to achieve emission. The last simulation was executed while holding the extraction electrode hole radius constant at 0.5 mm and the gap spacing at 0.25 mm while increasing the voltage from 1 to 50 kV, as shown in Figure 21. The gap spacing was chosen as 0.25 mm because it is a small enough spacing to benefit from the electric field enhancement, but is large enough to avoid shorting the emitter tip to the extraction electrode.



Extraction Voltage (kV)

Figure 21. Electric field simulation data with fixed gap height of 0.25 mm and fixed extraction hole radius of 0.5 mm while varying extraction voltage from 1 to 50 kV.

In addition to the gap height playing an important role in the electric field, the applied voltage plays a large role in the electric field that can be generated. As expected from looking back the local electric field enhancement equation, Equation 2.2, for each order of magnitude increase in extraction potential there is an order of magnitude increase in the local electric field. However, applying potentials in excess of 5-10 kV can cause electrical isolation issues and arcing so the extraction voltage for testing was set a maximum of 4 kV.

Using the electric field simulations and excluding tip radius, it is apparent that gap height and the applied potential both are important factors in achieving the required electric field. The extraction hole radius had little, if any, influence on the electric field enhancement. Since all of the experiments in this dissertation were assembled and aligned by-hand in the Isp Lab, the gap spacing was held between 0.25 and 0.5 mm.

3.1.2 Pure Tungsten Field Emitters

In the experiments that involved pure tungsten emitter comparison with re-generable emitters (reported in Chapter 5) two types of tungsten field emitters were tested. The first type was electrochemically etched to have a smooth surface with a single sharp apex and the second was electrochemically etched to have a series of longitudinal surface grooves so that the surface was roughened. The smooth tungsten emitters were formed by electrochemically etching tungsten wire in a 2M NaOH solution using the same process that was reported for the regenerable emitters. However, the tungsten wires were only DC etched, resulting in an emitter tip such as the one shown in Figure 22.



Figure 22. Image of a typical bare tungsten needle shape and smooth surface structure after being electrochemically etched under DC conditions in a 2M NaOH solution.

The roughened emitter tips were created so that the tips had geometry as close to the re-generable emitters as possible. The tungsten wires were electrochemically etched in a 2M NaOH solution using DC conditions and a final etch was performed using AC power to create the rough surface structure previously shown in Figure 13. A by-product of the AC etch was that multiple nano-features were created on the emitter surface that had the potential to be field emitting sites.

3.1.3 Micro-ammeter

All of the experiments that were performed required measuring current on the order of 1-30 μ A flowing through wires that were biased up to 10 kV. The easy way to measure the current is to implement a correctly sized resistor in-line on the extraction electrode wire and to measure the differential voltage across the resistor. The problem with that technique is that the differential signal voltage floats at whatever the extraction voltage is biased at. Most data acquisition systems can measure differential voltage of 10 V but not when the signal is floating at multiple kV. Also, recording data by-hand is tedious when performing experiments that last for 10's to 1000's of hours. Long duration experiments motivated Washeleski and Makela to design a custom ammeter. The ammeter was designed for measuring current ranging from 100's of nanoamps to 10's of milliamps. The unique characteristic of the device is that the ammeter can measure current as it flows through an electrical conductor that is floating at high voltages, up to 10 kV, and safely output an optically isolated analog signal from 0 to 10 volts that corresponds to the amount of current. To accomplish the current measurement, a resistor is placed in series with the high voltage wire and the voltage drop across the resistor is input into

the ammeter, as shown in the block diagram in Figure 23. The voltage is then converted to a corresponding current to be input to an optical isolator. The optical isolator safely outputs a current that is dropped across a resistor, where a signal ranging from 0 to 10 volts can be measured by a data acquisition system. The output voltage directly correlates to the amount of current flowing through the wire of interest. By implementing the optical isolator, the current is directly, and safely, input to existing data acquisition systems that are capable of measuring 10 V analog input signals.



Figure 23. Micro-ammeter block diagram.

The ammeter prototype was designed to measure 0 to 100 μ A flowing through a cable that is floating at 0 to 10 kV. Multiple calibration curves were recorded by sourcing current using a Keithley Model 2410. Current from 0 to 100 μ A was sourced across the input resistor and the output voltage on the optically protected side of the circuit was recorded, as shown in Table 1. Four sets of calibration data are shown that were taken at roughly 30 minute intervals to ensure that the output voltage of the circuit was consistent during multiple experiments taken over a two hour span. Taking these data also allowed for a curve fit to be established for real-time data conversion.

Sourced	Circuit Calibration Experiments (V)						
Current (µA)	Test 1	Test 2	Test 3	Test 4			
0	3.228	3.226	3.230	3.226			
0.1	3.243	3.243	3.247	3.243			
0.2	3.260	3.260	3.263	3.260			
0.3	3.276	3.277	3.281	3.277			
0.4	3.293	3.294	3.298	3.294			
0.5	3.310	3.310	3.314	3.311			
1	3.394	3.394	3.398	3.395			
1.5	3.478	3.480	3.482	3.480			
2	3.563	3.565	3.566	3.565			
2.5	3.649	3.650	3.651	3.650			
5	4.072	4.072	4.075	4.073			
10	4.909	4.910	4.912	4.910			
15	5.712	5.716	5.716	5.714			
20	6.463	6.467	6.473	6.471			
25	7.180	7.190	7.190	7.190			
30	7.820	7.830	7.830	7.830			
40	8.590	8.590	8.590	8.590			
50	8.850	8.850	8.860	8.860			
60	9.000	9.000	9.000	9.000			
70	9.110	9.110	9.110	9.110			
80	9.190	9.190	9.190	9.190			
90	9.260	9.260	9.260	9.260			
100	9.320	9.320	9.330	9.320			

Table 1. Experimental calibration data for the micro-ammeter.

By analyzing the data from Table 1, a curve fit was created for interpreting the amount of current flowing through a high voltage cable in terms of the optically isolated voltage. The experimental data, as well as the calibration curve are shown in Figure 24. The calibration curve is broken into two parts. The first corresponds to the linear region, 0 to 30 μ A, and the second corresponds to the exponential section, 30 to 100 μ A. For the experiments reported in this document, only the linear regime was used.



Figure 24. Experimental data from sourcing current to the micro-ammeter using a Keithley 2410 while measuring the optically isolated output voltage to determine a calibration curve.

The linear section, shown in black, ranges from 3.23 to 7.83 volts and is valid for sourced current from 0 to 30 μ A. The power region, shown in gray, ranges from 7.83 to 9.35 volts and is valid for current ranging from 30 to 100 μ A. The calibration curve was created from averaging the data in Table 1 to fit to the following piecewise function:

$$I(V) = \begin{cases} 6.36 \times V - 20.68 & \text{if } 3.23 < V \le 7.83, \\ 28.81 + 2.92 \times 10^{-23} (V^{25.82}) & \text{if } 7.83 < V < 9.35. \end{cases}$$

The linear fit has an R^2 of 0.999 which shows that the linear fit is a great representation of the calibration data. As mentioned, for the experiments reported in this dissertation the emission currents of interest were in the linear region of the micro-ammeter output. The ammeter is shown in its final configuration in Figure 25.



Figure 25. Image of the custom ammeter as-packaged with high voltage input/output banana terminals and optically isolated 0 to 10 volt analog output via DB-9 connector.

The analog signal from the micro-ammeter was input to a 16 bit National Instruments buspowered multifunction DAQ module, Model NI PXI-6259. The signal was then recorded using Labview software. The extraction voltage was also controlled and recorded using Labview software to control the analog input and monitoring voltage output of the Glassman High Voltage power supply.

3.1.4 Vacuum Facilities

There were three vacuum facilities that were used for this research. An ultra high vacuum (UHV) chamber was used in the Isp Lab, the specimen chamber of a Field Emission Scanning Electron Microscope, and another UHV chamber was used in the Isp Lab and also on NASA's microgravity C-9B aircraft.

3.1.4.1 Isp Lab UHV Chamber

All of the experiments reported in Chapters 4 and 5 were performed in the Ultra High Vacuum (UHV) chamber shown in Figure 26, which is located in the Michigan Tech Ion Space Propulsion lab. The UHV chamber is approximately 0.5 meters in diameter by 0.5 meters deep and has a base pressure of 10^{-9} Torr, which is achieved by pumping with a single 280-I/s turbo-molecular pump that is backed by a 110-I/min dry scroll pump. The tank is also equipped with a 300-I/s combination ion-sublimation pump to reach ultra-high vacuum. With the addition of the titanium sublimation pump (TSP) to the ion pump, higher pumping speeds are possible due to the TSP pump's ability to handle getterable gases. An operating pressure of at least 5×10^{-8} Torr was maintained while long-duration testing of the re-generable field emission cathodes and the pure tungsten field emitters.



Figure 26. Image of the UHV chamber in the Isp Lab and the stereo microscope.

One additional feature of the UHV facility is a trinocular stereo microscope. The microscope is situated outside the front viewing port of the chamber and has a focal length that allows viewing within the chamber. The microscope has an optical magnification up to 90x and it is equipped with a color digital camera. The camera provides the ability to capture *in situ* images of an operating LMIS for diagnostic purposes, as well as the ability to record video directly through USB 2.0.

3.1.4.2 Specimen Chamber of the Field Emission Scanning Electron Microscope

Investigation of emitter tip surface topography that is reported in Chapter 6 was performed in the Field Emission Scanning Electron Microscope at Michigan Technological University's Applied Chemical and Morphological Analysis Laboratory. Research was performed in the specimen chamber of the FE-SEM, shown in Figure 27. The chamber is evacuated using a series of ion pumps that are backed by a diffusion pump. Vacuum pressure of 10⁻⁷ Torr was maintained throughout testing.



Figure 27. Field emission scanning electron microscope in the Applied Chemical and Morphological Analysis Laboratory.

3.1.4.3 Microgravity UHV chamber

All of the experiments reported in Appendix D were performed in the Ultra High Vacuum (UHV) chamber shown in Figure 28. The UHV chamber is primarily comprised of an 8-inch ConFlat cross and has a base pressure of 10^{-9} Torr, which is achieved by pumping with a single 280-I/s turbo-molecular pump that is backed by a 110-I/min dry scroll pump. An operating pressure of 5 x 10^{-7} Torr was maintained while performing all of the experiments.



Figure 28. Image of the microgravity UHV test facility showing key components.

3.2 Experimental Procedure

For each set of experiments that were performed in the Isp Lab's UHV chamber, the FE-SEM, and the microgravity UHV chamber, the following general procedure was followed to regenerate emitter tips and to establish electron field emission. The electrical schematic and experimental apparatus shown previously in Figure 16 is repeated in Figure 29 for reference.



Figure 29. Electrical schematic for the re-generable electron source.

To re-generate emitter tips the following procedure was used:

- 1. Secure a tungsten wire to the Teflon fixture using a stainless-steel bolt.
- 2. Secure another tungsten wire to the Teflon fixture using a second stainless-steel bolt.
- 3. Carefully wrap the two wires, as shown in the FE-SEM micrograph in Figure 30, to form a Y-shape with the tungsten electrodes.



Figure 30. A FE-SEM micrograph of the wrapped section of tungsten electrodes – completing the electric circuit used for resistive heating to re-generate emitter tips.

- 4. Electrochemically etch the emitter tip and coat with indium, per the procedure described in Section 3.1.1.
- 5. Fasten the Teflon fixture that holds the emission electrode to the larger Teflon fixture that holds the stainless-steel extraction electrode.
- 6. Adjust the extraction electrode so that the gap spacing between the emitter tip and the extraction electrode is approximately the same distance as the width of the emission electrode ($250 \mu m$).

Two sets of instructions will be used from this point forward. The first set is for the Isp Lab UHV chamber and the microgravity UHV chamber. The second set is for the FE-SEM chamber.

Isp Lab and microgravity UHV chambers:

- 7. Place the entire assembly into the UHV chamber and bolt the assembly to the stainlesssteel bracket inside the chamber.
- 8. Attach the appropriate wires to the stainless-steel bolts; two wires for the resistive heater and one high-voltage wire for the extraction electrode.
- 9. Position a new copper gasket on the knife-edge of the UHV Conflat flange and secure the flange using proper torque specifications sealing the vacuum chamber.
- 10. Start the vacuum pumps and wait until the vacuum pressure is reduced to 10^{-8} Torr or less. If desired the UHV chamber can be pumped with a 280 L/s Turbopump or a 300 L/min ion pump to maintain 10^{-8} Torr or less.
- 11. To re-generate emitter tips, increase the heater power to approximately 4 Watts (at 3 to 4 A, about 1 V).
- 12. Increase the extraction voltage (biased negative with respect to the emission electrode) until ion emission current is established.
- 13. Adjust the extraction voltage to achieve the desired magnitude of ion emission current.
- 14. Allow the emission current to stabilize, which requires 1's to 10's of minutes.
- 15. Turn off heater power to quench the emitter.

- 16. Reverse the extraction electrode polarity to achieve electron field emission.
- 17. Record electron emission data (exact procedures described in Chapters 4, 5, and 7)
- 18. Turn off power supplies
- 19. Turn off vacuum pumps.

Field Emission Scanning Electron Microscope Specimen Chamber

- 7. Vent the airlock on the FE-SEM.
- 8. Attach the re-generable field emitter assembly to the insertion rod.
- 9. Evacuate the airlock to the same vacuum pressure as the specimen chamber.
- 10. Open the airlock valve and insert the field emitter assembly into the specimen chamber until the emitter assembly mates with the internal electrical connections.
- 11. Use the chamber scope to visually inspect the electrical connections and test the connections on the air-side electrical feedthrough to ensure continuity.
- 12. Remove the insertion rod and close the airlock valve.
- 13. To re-generate emitter tips, move the specimen stage to 20 mm away from the objective and increase the heater power to approximately 4 Watts (at 3 to 4 A, 1 V).
- 14. Increase the extraction voltage (biased negative with respect to the emission electrode) until ion emission current is established.
- 15. Adjust the extraction voltage to achieve the desired magnitude of ion emission current.
- 16. Allow the emission current to stabilize, which requires 1's to 10's of minutes
- 17. Turn off heater power to quench the emitter.
- 18. Reverse the extraction electrode polarity to achieve electron field emission.
- 19. Take electron emission data and then move the specimen stage back to 10 mm setting to acquire micrographs (exact procedures described in Chapter 6).
- 20. Open airlock and remove emitter apparatus.
- 21. Close airlock valve to seal the FE-SEM specimen chamber and vent airlock.
- 22. Remove emitter apparatus from insertion rod.

References for Chapter 3

- [1] Driesel, W. and C. Dietzsch, "In situ HVTEM observation of the tip shape of tin liquid metal ion sources", *Applied Surface Science*, 93, 179-190, 1996.
- [2] Driesel, W., C. Dietzsch and M. Moser, "In situ HV TEM observation of the tip shape of lead liquid metal ion sources", *J. Phys. D: Appl.Phys.*, 29, 2492-2500, 1996.
- [3] Driesel, W., C. Dietzsch and R. Muhler, "In situ observation of the tip shape of AuGe liquid alloy ion sources using a high voltage transmission electron microscope", *Journal of Vacuum Science and Technology B*, 14, 5, 3367-3380, 1996.
- [4] Driesel, W., C. Dietzsch, H. Niedrig and B. Praprotnik, "HV TEM in situ investigations of the tip shape of a gallium liquid-metal ion/electron emitter", *Ultramicroscopy*, 57, 45-58, 1995.
- [5] Praprotnik, B., W. Driesel, C. Dietzsch and H. Niedrig, "HV-TEM in-situ investigations of the tip shape of indium liquid metal ion emitter", *Surface Science*, 314, 353-364, 1994.
- [6] NASA Johnson Space Center, "Microgravity University Reduced Gravity Student Flight Opportunities Program." <<u>http://microgravityuniversity.jsc.nasa.gov/></u>, 2009
- [7] Ibe, J. P., J. P.P. Bey, S. L. Brandow, R. A. Brizzolara, N. A. Burnham, D. P. DiLella, K. P. Lee, C. R. K. Marrian and R. J. Colton, "On the electrochemical etching of tips for scanning tunneling microscopy", *Journal of Vacuum Science Technology A*, 8, 4, 3570-3575, 1990.
- [8] Bell, A. E. and L. W. Swanson, "The Influence of Substrate Geometry on the Emission Properties of a Liquid Metal Ion Source", *Applied Physics A*, 41, 335-346, 1986.

Chapter 4

4 Emitter Tip (Re-) Generation

This chapter is focused on investigating the current-voltage (I-V) characteristics of the nano-structures that are formed after quenching an operating liquid metal ion source. The nano-structures were quenched at multiple ion emission currents ranging from 1 to 25 μ A and the nano-structures were quenched multiple times at each ion current (in a randomized order). After quenching and recording the I-V characteristics, the I-V data were applied to the Fowler-Nordheim model to estimate the emitter tip radii. Rather than reference each paragraph throughout this entire chapter, it should be noted that most of this chapter has been reproduced and expanded upon from a journal publication written in 2009 for the *Journal of Propulsion and Power* by Makela, Washeleski, and King.¹

4.1 Procedure and Results

A detailed procedure for etching and coating the emitter tips was provided in Section 3.1.1 and an operating procedure was presented in Section 3.2. However, the basic procedure for re-generating the emitter tips will be outlined here. To obtain ion emission it was necessary to first liquefy the layer of indium on the tungsten electrode. To adequately liquefy the indium, the emitter heating supply, shown in Figure 31, was powered and increased to achieve a temperature greater than the melting point of indium, which is 156.6°C.



Figure 31. Re-generable electron source experimental apparatus, shown previously in Section 3.1.

For the experiments reported in this Chapter, the heater power during ion emission remained between 1 and 1.25 Watts which corresponded to 2.4 to 2.5 A of heater current. The heater supplied enough power to keep the indium liquefied, without heating it to significant evaporation. The extraction electrode was then biased negatively with a high-voltage power supply and the emitter electrode was grounded. The extraction voltage was increased (negative

with respect to the emitter) until ion emission was achieved, typically requiring -3 to -4 kV for the onset of emission. The high-voltage power supply was current-limited at an ion discharge current of 2 μ A, allowing the voltage to float where necessary to maintain ion emission. The current-limited operation was maintained for the first 30 minutes for conditioning; during this period ion emission stabilized at constant voltage. After the 30-minute period, the ion discharge current was then switched to voltage-limited mode. The extraction voltage could then be adjusted to obtain a desired ion emission current. When the emission current of interest was reached, the emitter heater power was quickly turned off, causing the emission current to decrease to 0 μ A in 2 to 3 seconds.

Once a sharp emitter was formed by quenching, the extraction electrode was biased with a positive potential to obtain electron field emission from the cold needle. The extraction voltage was swept between 0 and the voltage necessary to obtain 0.5 μ A of emission current, which will be referred to from this point forward as V_{max} . The extraction voltage was swept by using the analog input controls on the high-voltage power supply. A step function was input to sweep the extraction voltage at 50 V intervals every second up to V_{max} while recording 1000 samples/s of the emission current through the analog output of a micro-ammeter.

Sharpened field emission tips were quenched at ion emission currents between 1 and $25 \,\mu\text{A}$ to determine what effect the ion current prior to quenching had on the electron emission characteristics after quench. The procedure for the first 25 experiments was to obtain ion emission, then quench the emitter to solidify and preserve the nano-structure, and then use the quenched nano-structure to produce an electron emission I-V curve by sweeping the extraction voltage while recording the emission current. After the electron emission characteristics were established, the emitter heater was increased to 2 Watts for 25 to 30 seconds with no applied extraction voltage in an effort to flash heat the emitter tip and, hopefully, destroy the indium tip to prepare for re-generation in subsequent tests. It was initially thought that melting the indium on the emitter tip in the absence of an electric field would allow the surface tension of the liquefied indium to destroy any sharp protrusions so that additional testing could be done to form new sharp tips at different quenching currents. Upon further investigation using the optical microscope, it was obvious that merely increasing the amount of heat to the emitter wasn't removing the sharpened protrusions: subsequent tests were likely inducing emission from the same protrusion each time. This prevented a study of protrusion shape as a function of ion guench current, so a new method was conceived to destroy the emission site prior to regrowth. The new method was to increase the extraction voltage to approximately two to three times what was necessary to achieve 0.5 µA of emission current, which was defined earlier as V_{max} . Increasing the extraction voltage to $3V_{max}$ caused the emitter to briefly arc to the extraction electrode, a process that exploded the protrusion off of the emitter apex, giving off a micro-scale flash that was visible using the optical microscope that was equipped outside the viewport on the UHV chamber. A microscopic image acquired prior to removing the microstructure and the instant after removing the micro-structure is shown in Figure 32.



Figure 32. Microscopic images of an indium-coated tungsten electrode with a) nano-structure solidified and b) after the structure has been removed.

Once the new method of 'resetting' the emitter tips was discovered, a new set of experiments were conducted ranging the ion emission current at quench between 1 and 25 μ A. Just as previously done, the heater power was increased to approximately 1-2 Watts and then the extraction voltage was increased until ion emission began. Then the ion source was current-limited at 2 μ A for 30 minutes. After the emission stabilized, the ion discharge current was switched to voltage-limited mode. The extraction voltage was then adjusted to achieve the desired emission current and then the heater power was decreased to 0 Watts, solidifying the nano-structure. The polarity of the extraction electrode was then reversed to obtain electron field emission and swept from 0 to V_{max} . Multiple electron I-V curves are shown in Figure 33.



Figure 33. A sample of electron emission current vs. extraction voltage data for emitter tips quenched at ion currents of 1.0, 3.2, 7.8, 10.4, and 15.0 μ A.

Each curve was taken after quenching nano-structures at a different ion emission current. Between each successive test the tips were exposed to an increased electric field to cause tip explosion, followed by the application of 2-3 Watts of heater power in an attempt to smooth out any additional protrusions. After the emitter tip surface smoothness was 'reset' the process of tip re-generation was repeated multiple times at various ion emission currents. The re-generated emitter tips were each used to acquire an electron I-V curve. Most of the emitters that were quenched at higher ion emission currents had a lower onset voltage, as shown in the sample data in Figure 33. The electron emission data were all applied to the Fowler-Nordheim model. The results from applying the Fowler-Nordheim model to each electron I-V data set are shown in Figure 34. Numbers were placed next to each data point in the plot showing the randomized order in which the data were collected.



lon Current at Quench (μA)

Figure 34. Estimated electron emitter radius using Fowler-Nordheim modeling at quenching currents ranging from 1 to 25 μ A. The number next to each data point shows the sequential order in which the data were collected.

It is clear from looking at Figure 34 that the estimated emitter tip radius decreases as the ion current at quench is increased. This was expected from an examination of Figure 33. Sharper protrusions had much lower onset voltages, where onset voltage refers to the extraction voltage necessary for electron field emission to begin. From this data set it appears that once an ion quenching current of 15 μ A was exceeded, the emitter tip radius reached a minimum. A more detailed statistical analysis using sample data is included in the following section, which provides an explanation of how the data were taken from raw form through the Fowler-Nordheim model to obtain a tip radius estimate.

4.2 Analysis of I-V Characteristics

This section includes examples of how the data that are reported in this dissertation have been analyzed. The raw data that were collected from an electron I-V sweep from a quenched nano-structure is shown and then each step of the analysis process. This section also includes how the experimental error bars were calculated.

4.2.1 Fowler-Nordheim Modeling:

The raw data were acquired by sweeping the extraction voltage using the analog input controls of the high voltage power supply. While the extraction voltage was swept from 0 to V_{max} at 50-100 V increments, the analog voltage from the micro-ammeter was recorded, which

corresponded to the magnitude of electron emission current that was being emitted. Data from a sample electron emission sweep is shown in Figure 35.



Figure 35. Raw data recorded from sweeping the extraction voltage while recording electron emission current using the analog output voltage from the custom micro-ammeter.

Gomer's technique of applying the Fowler-Nordheim model to predict the emitter tip radius was then applied to the data.² Gomer's technique claims to be valid for predicting emitter tip radii to within 20%. To verify that each set of data had an error of less than 20%, the following methods were employed to analyze the data. Using the raw electron emission current, *I*, and the extraction voltage, *V*, data the Fowler-Nordheim plot of $\ln I^2/V$ vs. 1/V was produced and was linear (indicative of electron field emission), as shown in Figure 36. A linear regression was performed to determine the slope of the Fowler-Nordheim plot. Recalling from Chapter 2, the slope of the Fowler-Nordheim plot can be used to estimate the nano-structure size using the Fowler-Nordheim *b*' coefficient, which was shown in Equation 2.5 but is repeated in Equation 4.1 below. In addition to performing Fowler-Nordheim modeling, the R² and standard error were calculated to interpret the radii estimations and to ensure the estimations were within 20%.



Figure 36. Fowler-Nordheim plot using data from an electron emission I-V sweep.

The coefficient of determination, more commonly known as R^2 , was calculated to determine how well the regression line on the Fowler-Nordheim plot approximated the raw data points. The closer the R^2 value was to 1, the better the regression line represented the data. An R^2 value of 1 means that the regression line fits all of the raw data points perfectly. So what value of R^2 indicates that the data is acceptable? The answer to that is open to interpretation so each set of data is reported, along with its R^2 number.

To determine the error in the slope, and thus the emitter radii error, the standard error was calculated using the Data Analysis Toolkit in Microsoft's Excel 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,

$$m-2s_m \le m \le m+2s_m$$
.³ Equation 4.1

For the example in Figure 36 the standard error that was generated using the Data Analysis Toolkit was 0.64. Results from the Data Analysis Toolkit are shown in Table 2. The standard error was then used to bound the data.

Regression Statistics				
Multiple R	0.950			
R Square	0.902			
Adjusted R Square	0.896			
Standard Error	0.641			
Observations	19			

Table 2. Regression results from Microsoft Excel's Data Analysis Toolkit for the Fowler-Nordheim data shown in Figure 36.

When the standard error was input to Equation 4.1, the slope of the linear regression line was -41.20 ± 1.28 . The Fowler-Nordheim model could then be applied to obtain an estimate of the nano-structure radius as follows;

$$m = b' \phi^{1.5}$$
 Equation 4.2
where
 $b' = 6.8 \times 10^7 \alpha k r_t$. Equation 4.3

Substituting 1 in for α , 5 in for k, and 3.8 eV in for ϕ (the work function of indium), an estimate of the nano-structure radius could be made. For the example in Figure 36, a slope of -41.20 \pm 1.28 was used to approximate the emitter tip radius. When the slope was inserted into Equation 4.2 and combined with Equation 4.3, the resulting radius approximation was 16.4 ± 0.5 nm. It is important to note that using 1 for α and 5 for k with the Fowler-Nordheim model, the model provides an approximation of the emitter tip radius to within 20%. Recalling from Chapter 2, k varies with the natural log of the gap spacing and emitter tip radius. For the geometry used in the experiments reported in this dissertation, the image-correction factor, α_{i} could vary from 0.9 - 1.0 and the field voltage proportionality factor, k, was $5 \pm 1.^2$ For the reported experiments an α of 1 and k of 5 were used, which allowed for the estimated emitter tip radii to be within the 20% that Gomer's method of Fowler-Nordheim modeling predicts. For the example, 20% of the estimated tip radius of 16.4 nm is about 3.3 nm, so Gomer's technique results in a radius estimate of 16.4 ± 3.3 nm. The error that was experimentally calculated from the regression estimation was \pm 0.5 nm, so the experimentally determined value was within 20%. For all of the experiments, the data was analyzed to determine the experimental error to verify that the error bars were within 20% and then \pm 20% of the emitter tip radius was used for the error bars.

4.2.2 Comparing Estimated Tip Radii Against Each Other:

Another statistical analysis was performed to determine if the emitter tip radii that were approximated using the Fowler-Nordheim model and plotted versus the ion current at quench showed any statistically meaningful trends. The plot is repeated from Figure 34 and shown in Figure 37. To determine if the estimated emitter tip radii were larger at lower ion quench currents the emitter tip radii were grouped and then groups were statistically tested against one another using the two-sample t-test.



Figure 37. Estimated electron emitter radius using Fowler-Nordheim modeling at quenching currents ranging from 1 to 25 μ A.

The data were separated into five groups. Any emitters quenched at currents less than 5 μ A were grouped, emitters quenched at currents from 5 to 9 μ A were grouped, 10 to 14 μ A were grouped, 15 to 19 μ A were grouped, and anything over 20 μ A was grouped as shown in Table 3.

Ion Quench Current (µA)	nated Emitter Tip Radii (nm)				
< 5	30.0	45.8	16.9	36.9	27.4
5-9	18.3	27.1	12.5	21.7	14.0
10-14	26.9	23.8	16.0		
15-19	4.6	16.8	14.2	13.9	17.5
> 20	19.6	18.7	18.2	17.1	

Table 3. Grouped emitter tip radii estimations used to gather a statistically meaningful comparison between ion quench current and emitter tip size.

Using a confidence interval of 95%, two-sample t-tests were performed using the Data Analysis Toolkit in Microsoft Excel to compare each group of data against each other group and are included in Appendix A. The two-sample t-test results showed that emitters that were quenched at less than 5 μ A were statistically larger in radius than those quenched at 5-9 μ A, 15

to 19 μ A, and > 20 μ A, but no statistical difference was apparent between emitters quenched at >5 μ A and those quenched at 10 to 14 μ A, when looking at the two-tailed p-value. When the other groups were tested against one another there were no statistical differences between them. Therefore, quenching at lower emission current appears to generate less sharp nano-structures than quenching at larger currents but there appears to be a statistically relevant plateau of minimal nano-structure size. For instance, quenching the emitter at 15 to 19 μ A should produce the same size nano-structure as quenching at greater than 20 μ A, but quenching an emitter at a current of less than 5 μ A should produce less sharp nano-structures.

4.3 Summary

It was found that the ion quenching current had an important role in the geometry of the nano-structures that were formed. By applying the Fowler-Nordheim model to the data that were collected it is apparent that higher quenching currents form sharper emitters, which is what was expected when looking at the electron I-V curves after each quench. The electron I-V curves showed that electron emission onsets at much lower extraction voltages from emitters that were quenched at higher currents, which implies that the local electric field enhancement at the tip apex was greater – supporting the fact that the emitter tips that were formed at higher ion currents before quenching were sharper than the nano-structures that were formed at lower ion currents before quenching.
References for Chapter 4

- [1] Makela, J. M., R. L. Washeleski and L. B. King, "Re-generable Field Emission Cathode for Spacecraft Neutralization", *Journal of Propulsion and Power*, 25, 4, 2009.
- [2] Gomer, R., <u>Field Emission and Field Ionization</u>. New York, NY: American Institute of Physics, 1961.
- [3] Coleman, H. W. and W. G. Steele, <u>Experimentation, Validation, and Uncertainty Analysis for</u> <u>Engineers</u>. Third. Hoboken, NJ: John Wiley & Sons, Inc., 2009.

Chapter 5

5 Re-generable Emitter Comparison with Tungsten Emitters

This Chapter describes extended duration and elevated pressure experiments to evaluate the performance of re-generable emitter tips and then to compare the performance to pure tungsten emitters. The goal was to investigate the lifetime of a quenched ion source in an ultra high vacuum environment and to determine the robustness of quenched ion sources against elevated neutral pressure conditions. The data were then compared with pure tungsten field emitters that were subjected to the same experiments. 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.^{1, 2}

The Chapter begins with a description of the long-duration experiments to determine if re-generable indium emitter tips are capable of sustaining long-lasting emission at relatively low extraction voltages. Emitter tips were quenched at 20 μ A of ion emission current and then they were operated in electron emission mode for about 1,750 hours. For each experiment the extraction electrode was held at a constant voltage during electron emission and the emission current was observed. Extended duration experiments were performed using bare smooth tungsten emitters and bare roughened tungsten emitters. In addition, electron emission from each type of emitter was observed while operating in vacuum pressure of 10⁻⁸ Torr and then while increasing the vacuum pressure to 10⁻⁴ Torr.

5.1 Lifetime and Elevated Pressure Results

5.1.1 Smooth Bare Tungsten Emitters

The first experiment was performed with a smooth bare tungsten field emitter, as shown in Figure 38. The emitter was electrochemically DC etched using the procedure described in Section 3.1.1. The smooth bare tungsten emitter was tested in the UHV chamber at a vacuum pressure of 10^{-9} Torr.



Figure 38. Image shown in Section 3.1.2 of the typical bare tungsten needle shape and smooth surface structure after being electrochemically etched under DC conditions in a 2M NaOH solution.

A Fowler-Nordheim sweep was taken before the lifetime experiment and yielded a tip radius estimate of 13.9 \pm 1.2 nm. Electron emission was then achieved by increasing the extraction electrode to 1.4 kV to obtain an electron emission current of 3 μ A. The extraction voltage was then held constant and emission current was observed for 625 hours. The test was concluded after 625 hours due to the necessity to perform additional experiments in the UHV facility. To conclude the experiment, at t_e = 625 hours the ion pump on the UHV chamber was turned off to increase the chamber pressure. At vacuum pressure of 10⁻⁷ Torr, and after less than two minutes, the emission ceased, as shown in Figure 39. A Fowler-Nordheim analysis was not performed after the 625 hour test because electron emission could not be acquired at an extraction voltage of 4 kV. For the elevated 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.



Elapsed Time (hr)

Figure 39. Electron emission current from an electrochemically etched tungsten field emitter, showing a sudden decrease in emission current when background pressure was increased from 10⁻⁹ to 10⁻⁷ Torr. Emission current could not be established in subsequent testing (at an extraction voltage of 4 kV) indicating that the smooth bare tungsten emitter tip had been permanently damaged when exposed to an increased vacuum chamber pressure.

A new smooth bare tungsten emitter was used for the next experiment. The tungsten emitter was electrochemically etched using the exact procedure as the previous emitter. The new tungsten emitter experiment was also performed in the UHV chamber, at a starting vacuum pressure of 10^{-8} Torr. Before exposing the emitter tip to increased vacuum pressure a Fowler-Nordheim analysis was performed and yielded a tip radius estimate of 3.1 ± 0.7 nm. After the Fowler-Nordheim sweep, the extraction voltage was increased to 4 kV to obtain about 4 μ A of electron emission current. The emission current increased to about 6 μ A and then remained relatively constant for the remainder of operation at 10^{-8} Torr.

After about two hours of stable operation, at $t_e = 2.1$ hr, the ion pump on the UHV chamber was turned off to increase the neutral vacuum pressure. After a few minutes the vacuum pressure increased to 10^{-7} Torr. Electron emission ceased when the chamber pressure was increased, as shown in Figure 40.



Figure 40. Electron emission current from an electrochemically etched smooth bare tungsten field emitter. At $t_e = 2.1$ hr background pressure was increased from 10^{-8} to 10^{-7} Torr and emission ceased.

After the emission ceased, electron emission could not be re-established at 4 kV. Since emission couldn't be established, the emitter tip must have been catastrophically damaged by the increase in vacuum pressure. Exposure to elevated pressure rendered the bare tungsten emitter useless as a field emission electron source.

5.1.2 Roughened Bare Tungsten Emitters

Two roughened bare tungsten emitters were tested for comparison. The roughened emitters were electrochemically DC and AC etched following the procedure in Section 3.1.1 to create surface grooves, as shown in Figure 41. Etching the surface grooves on the emitter was intended to make the electrode as close as possible to the indium-coated re-generable electrodes. A possible indirect benefit of adding surface grooves was that the number of field-emitting nano-structures on the surface may have also increased.



Figure 41. Image shown in Section 3.1.1 of an electrochemically etched tungsten electrode with longitudinal micro-grooves. The grooves were intended to make the tungsten electrode geometry as close as possible to the indium-coated electrodes.

5.1.2.1 Roughened Bare Tungsten Tip 1

An electron I-V sweep was acquired on the new roughened bare tungsten emitter at 10^{-8} Torr, as shown in Figure 42. 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.



Figure 42. Electron I-V sweep performed on a new roughened bare tungsten field emitter, yielding a tip radius estimate of 4.6 ± 0.9 nm when applied to the Fowler-Nordheim model.

The roughened bare tungsten emitter was then operated for about 20 hours to ensure the emission current was stabilized. Holding the extraction voltage constant, at $t_e = 21.2$ hours the vacuum chamber pressure was increased. The pressure was increased by turning off the ion pump. As the vacuum pressure increased the electron emission current was observed. Electron emission sustained for about 7 hours at background pressure ranging between 10^{-5} and 10^{-3}

Torr, as shown in Figure 43, which was about 7 hours 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. Extrapolating the pressure data and analyzing data from other elevated pressure experiments, an approximate vacuum pressure of 5×10^{-5} Torr was estimated when emission ceased.



Figure 43. Electron emission current from a roughened bare tungsten emitter held at a constant extraction voltage of 2.8 kV from $t_e = 3.1$ hr to $t_e = 28.9$ hr 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.

Once emission ceased, the vacuum 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 ± 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 vacuum pressure.



Figure 44. Electron I-V sweep taken from a new roughened bare tungsten field emitter and an electron I-V sweep acquired after the first exposure of the emitter tip to elevated vacuum pressure. The I-V sweep from after the first exposure was applied to the Fowler-Nordheim model and the tip radius was estimated at 9.9 ± 2.0 nm.

The roughened bare tungsten emitter was then used to establish electron emission, as shown in Figure 45. Emission was sustained for about 4 hours at a vacuum pressure in the 10^{-9} Torr range and then the vacuum pressure was increased while emission was observed. During the first 4.5 hours of the experiment, the emission current gradually decreased when vacuum pressure was constant in the 10^{-9} Torr regime, implying degradation of the tip. After about 1.5 hours, emission ceased when vacuum pressure reached $2x10^{-5}$ Torr.



Elapsed Time (hr)

Figure 45. Electron emission current from the same roughened bare tungsten emitter used in the test of Figure 43. The emitter was held at a constant extraction voltage of 1.6 kV from t = 0.4 hr to t = 6 hr as background pressure was increased for the second time.

During the second exposure to elevated pressure, emission from the roughened bare tungsten emitter lasted longer than electron emission from a smooth bare tungsten emitter. After emission ceased the vacuum pressure was decreased back to 10^{-9} Torr and an electron I-V sweep was performed to apply the Fowler-Nordheim model, as shown in Figure 46. The model yielded a tip radius estimate of 6.3 ± 1.3 nm, which was sharper than the estimated tip radius before exposing the emitter to increased pressure. A few possible reasons for a decrease in the tip radius after exposure to elevated pressure are discussed at the end of this section.



Figure 46. Electron I-V sweep taken from a roughened bare tungsten field emitter after the first exposure to elevated pressure and an electron I-V sweep acquired after the second exposure of the emitter tip to elevated vacuum pressure. The I-V sweep from after the second exposure was applied to the Fowler-Nordheim model and the tip radius was estimated at 6.3 ± 1.3 nm.

The roughened bare tungsten emitter tip was then used to achieve electron emission again. Emission was established at an extraction voltage of about 2.1 kV and the electron emission was then left to stabilize for about 4 hours. At $t_e = 3.8$ hours vacuum pressure was increased and the emission current was observed, as shown in Figure 47. The emission sustained for about 2.5 hours into the pressure range of 10^{-6} to 10^{-3} Torr. Extrapolating the pressure data, the pressure when emission ceased was approximately 6.3×10^{-6} Torr.



Elapsed fillie (iii)

Figure 47. Electron emission current from the same roughened bare tungsten emitter used in the test of Figure 43 and Figure 45. The emitter was held at a constant extraction voltage of 2.1 kV from $t_e = 0$ hr to $t_e = 6.5$ hr as background pressure was increased for the third time.

After emission ceased the vacuum chamber pressure was restored to 10^{-9} Torr and an electron I-V sweep was taken, as shown in Figure 48, so the Fowler-Nordheim model could be applied. The F-N model yielded a tip radius estimate of 4.6 ± 1 nm. Since numerous I-V sweeps have been acquired and the Fowler-Nordheim model yielded emitter tips that have been approximately the same size, it is possible that the roughened bare tungsten emitter may have multiple emission sites with similar nano-structures. Each time the roughened bare tungsten emitter was exposed to increased vacuum pressure, it is possible that a single emission site was damaged but the next sharpest nano-structure could be used for subsequent testing.



Figure 48. Electron I-V sweep taken from a roughened bare tungsten field emitter after the second exposure to elevated pressure and an electron I-V sweep acquired after the third exposure of the emitter tip to elevated vacuum pressure. The I-V sweep from after the third exposure was applied to the Fowler-Nordheim model and the tip radius was estimated at 4.6 \pm 1 nm.

For the fourth time, electron emission was established from the roughened bare tungsten emitter tip that had been exposed to increased pressure. Emission current was stable around 4 μ A for about an hour and then the vacuum pressure was increased at t_e = 1 hr, as shown in Figure 49. Emission current sustained for about 5 hours at increased vacuum pressures ranging from 10⁻⁵ and 10⁻³ Torr. Extrapolating the pressure data, the vacuum chamber pressure when the emission failed was approximately 2 x 10⁻⁵ Torr.



Elapsed Time (hr)

Figure 49. Electron emission current from the same roughened bare tungsten emitter used in the test of Figure 43 to Figure 47. The emitter was held at a constant extraction voltage of 3.1 kV from $t_e = 0$ hr to $t_e = 6.3$ hr as background pressure was increased for the fourth time.

Once emission ceased, an electron I-V sweep was acquired, as shown in Figure 50, and the Fowler-Nordheim model was applied – yielding an emitter tip radius of about 7.1 ± 1.4 nm. Although the emitter tip radius estimations were within 10 nm of each other, the extraction voltage required to achieve electron emission after successive exposures to increased vacuum pressure had been increasing and decreasing randomly, indicating that the emitter tip was being damaged after some of the exposures and was getting sharper during other exposures to increased pressure. A discussion is included at the end of this Chapter that includes some ideas concerning this phenomenon.



Figure 50. Electron I-V sweep taken from a roughened bare tungsten field emitter after the third exposure to elevated pressure and an electron I-V sweep acquired after the fourth exposure of the emitter tip to elevated vacuum pressure. The I-V sweep from after the fourth exposure was applied to the Fowler-Nordheim model and the tip radius was estimated at 7.1 \pm 1.4 nm.

After the Fowler-Nordheim sweep, the extraction voltage was increased to 4 kV to obtain electron emission. The initial emission current was 4 μ A but quickly increased to about 6 μ A. After staying constant at 6 μ A for about two hours the vacuum pressure was increased. Emission sustained for about 20 hours at increased pressure as shown in Figure 51.



Elapsed Time (hr)

Figure 51. Electron emission current from the same roughened bare tungsten emitter used in the test of Figure 43 to Figure 49. The emitter was held at a constant extraction voltage of 4 kV from $t_e = 0$ hr to $t_e = 24$ hr as background pressure was increased for the fifth time.

After emission ceased the vacuum pressure was decreased to 10^{-8} Torr and an electron I-V sweep was performed, as shown in Figure 52. The Fowler-Nordheim model was applied and resulted in an emitter tip radius of about 10.0 ± 2.0 nm. Thus far the roughened bare tungsten emitter tip was exposed to an increase in vacuum pressure five times and the estimated emitter tip radii have only varied by 5-6 nanometers. The interesting thing is that the emitter tip radii have are still increasing and decreasing randomly after each exposure to elevated pressure. As mentioned a discussion concerning the emitter tip radii variability is included at the end of this Chapter.



Figure 52. Electron I-V sweep taken from a roughened bare tungsten field emitter after the fourth exposure to elevated pressure and an electron I-V sweep acquired after the fifth exposure of the emitter tip to elevated vacuum pressure. The I-V sweep from after the fifth exposure was applied to the Fowler-Nordheim model and the tip radius was estimated at 10.0 \pm 2.0 nm.

After the tip estimate was calculated the extraction voltage was increased to 3.9 kV and the emission current was observed while exposing the roughened bare tungsten emitter to increased vacuum pressure for the sixth time. Emission lasted for about 4 hours as the pressure was increased, as shown in Figure 53.



Elapsed Time (hr)

Figure 53. Electron emission current from the same roughened bare tungsten emitter used in the test of Figure 43 to Figure 51. The emitter was held at a constant extraction voltage of 3.9 kV from $t_e = 0$ hr to $t_e = 6$ hr as background pressure was increased for the sixth time.

When emission ceased the vacuum pressure was returned to 10^{-8} Torr and another I-V sweep was performed, resulting in a Fowler-Nordheim emitter tip radius estimate of 7.8 ± 1.6 nm. Again, the emitter tip radius estimate was on the same order as the past couple of Fowler-Nordheim analyses, but that is expected since the extraction voltage required for emission for the past couple of experiments has been relatively consistent.



Figure 54. Electron I-V sweep taken from a roughened bare tungsten field emitter after the fifth exposure to elevated pressure and an electron I-V sweep acquired after the sixth exposure of the emitter tip to elevated vacuum pressure. The I-V sweep from after the sixth

exposure was applied to the Fowler-Nordheim model and the tip radius was estimated at 7.8 \pm 1.6 nm.

After the Fowler-Nordheim analysis the same roughened bare tungsten emitter tip was exposed to increased vacuum pressure a seventh time. The emission current sustained for about 5 hours as shown in Figure 55. Emission ceased at about 10⁻⁵ Torr.



Elapsed Time (hr)

Figure 55. Electron emission current from the same roughened bare tungsten emitter used in the test of Figure 43 to Figure 53. The emitter was held at a constant extraction voltage of 4.0 kV from $t_e = 0$ hr to $t_e = 6.5$ hr as background pressure was increased for the seventh time.

After emission ceased, the vacuum chamber pressure was decreased to 10^{-8} Torr and an electron I-V sweep was acquired, as shown in Figure 56. A post-test Fowler-Nordheim analysis yielded an emitter tip radius estimate of 14.0 ± 2.8 nm. After seven exposures to elevated vacuum pressure conditions, emission from the roughened bare tungsten emitter tip could not be established at an extraction voltage of 4 kV.



Figure 56. Electron I-V sweep taken from a roughened bare tungsten field emitter after the sixth exposure to elevated pressure and an electron I-V sweep acquired after the seventh, and final, exposure of the emitter tip to elevated vacuum pressure. The I-V sweep from after the seventh exposure was applied to the Fowler-Nordheim model and the tip radius was estimated at 14.0 ± 2.8 nm.

In summary, the first roughened bare tungsten emitter that was tested survived 7 exposures to elevated vacuum 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 between 1.5 and 20 hours of electron emission after the increase in pressure began for each experiment.

5.1.2.2 Roughened Bare Tungsten Tip 2

A second roughened bare tungsten emitter was electrochemically etched and subjected to multiple exposures at elevated pressure conditions. A pre-test Fowler-Nordheim sweep wasn't acquired so an estimation of the original emitter tip radius isn't reported. The first experiment with the roughened bare tungsten emitter was operated for about 22 hours at an extraction voltage of 2.4 kV, as shown in Figure 57. Electron emission lasted for about 0.6 hours as the vacuum pressure was increased to 10^{-5} Torr.



Figure 57. Electron emission current from a roughened bare tungsten emitter held at a constant extraction voltage of 2.4 kV from $t_e = 0$ hr to $t_e = 21.7$ hr as background pressure was increased for the first time.

After the emission current ceased due to the first exposure to elevated pressure, the extraction voltage was turned off and the vacuum pressure was restored to UHV conditions. An electron I-V sweep was acquired, as shown in Figure 58. Applying the Fowler-Nordheim model resulted in an emitter tip radius estimation of 7.3 ± 1.5 nm.



Figure 58. Electron I-V sweep taken from a roughened bare tungsten field emitter after the first exposure to elevated pressure. The I-V sweep from after the first exposure was applied to the Fowler-Nordheim model and the tip radius was estimated at 7.3 ± 1.5 nm.

After the Fowler-Nordheim sweep was taken, the extraction voltage was increased to 3.5 kV and emission current was observed for over 15 hours. After 15 hours, the emitter was exposed to elevated pressure conditions. Emission lasted for about 1.4 hours as pressure was increased to 10^{-5} Torr, as shown in Figure 59.



Figure 59. Electron emission current from the same roughened bare tungsten emitter used in the test of Figure 57. The emitter was held at a constant extraction voltage of 3.5 kV from $t_e = 0$ hr to $t_e = 18.5$ hr and background pressure was increased for the second time

After emission ceased, the extraction voltage was turned off and the tank pressure was decreased back to UHV conditions again. An electron I-V sweep was performed, as shown in Figure 60. Applying the Fowler-Nordheim model resulted in a tip radius approximation of 10.4 \pm 2.1 nm.



Figure 60. Electron I-V sweep taken from a roughened bare tungsten field emitter after the first and second exposures to elevated pressure. The I-V sweep from after the second

exposure was applied to the Fowler-Nordheim model and the tip radius was estimated at 10.4 \pm 2.1 nm.

After the sweep, the extraction voltage was increased to 3.7 kV and emission current was observed for about 22 hours. After 22 hours, the emitter was exposed to elevated pressure, as shown in Figure 61. Emission current lasted for about 2.2 hours as vacuum pressure was increased to 10^{-5} Torr.



Elapsed Time (hr)

Figure 61. Electron emission current from the same roughened bare tungsten emitter used in the test of Figure 57 and Figure 59. The emitter was held at a constant extraction voltage of 3.7 kV from $t_e = 0$ hr to $t_e = 24.1$ hr and the background pressure was increased for the third time.

After emission ceased the extraction voltage was turned off and the chamber pressure was restored to 10^{-8} Torr. An electron I-V sweep was acquired, as shown in Figure 62. Applying the Fowler-Nordheim model to the sweep indicated that the tip radius was approximately 21.8 ± 4.4 nm.



Figure 62. Electron I-V sweep taken from a roughened bare tungsten field emitter after the second and third exposures to elevated pressure. The I-V sweep from after the third exposure was applied to the Fowler-Nordheim model and the tip radius was estimated at 21.8 ± 4.4 nm.

Then the extraction voltage was increased to 3.9 kV while emission was observed. Over the course of this experiment the emission current increased slightly, but that was expected since vacuum pressure was decreasing. After operating in UHV conditions for about 70 hours the vacuum pressure was increased and the emission lasted for about 20 hours, although it nearly extinguished after about five hours, as shown in Figure 63.



Elapsed Time (hr)

Figure 63. Electron emission current from the same roughened bare tungsten emitter used in the test of Figure 57 to Figure 61. The emitter was held at a constant extraction voltage of 3.9 kV from $t_e = 0$ hr to $t_e = 92.2$ hr and the background pressure was increased for the fourth time.

After the fourth exposure to elevated pressure, the vacuum was restored to 10^{-8} Torr and an electron I-V sweep was acquired, as shown in Figure 64, and the tip radius estimation was 13.9 ± 2.8 nm. After the electron I-V sweep, emission could not be established and stabilized at an extraction voltage of over 4 kV so experiments with the second roughened bare tungsten emitter were terminated. The electron I-V sweeps that were acquired show that an increase in extraction voltage was necessary to achieve electron emission after each exposure to increased pressure.



Figure 64. Electron I-V sweep taken from a roughened bare tungsten field emitter after the third and fourth exposures to elevated pressure. The I-V sweep from after the fourth exposure was applied to the Fowler-Nordheim model and the tip radius was estimated at 13.9 \pm 2.8 nm.

5.1.2.3 Roughened Bare Tungsten Emitter Summary

In summary, the second roughened bare tungsten emitter survived 4 exposures to elevated pressure and emission sustained from about 1.4 to 20 hours while pressure was increased. The maximum pressure that was measured before emission ceased was 9.9×10^{-5} 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 4.

Emitter Type	# of Tested Emitters	Emission Duration at Elevated Pressure	# of Exposures Before Failure	Maximum Tank Pressure Reached
Smooth Bare Tungsten	2	1 - 5 min	1 - 2	10 ⁻⁷ Torr
Roughened Bare Tungsten	2	1.4 - 20 hr	4 - 7	10 ⁻⁵ Torr

Table 4. Emitter tip comparison table displaying how long the smooth and roughened bare tungsten emitter tips lasted at elevated pressure, how many exposures to increased pressure caused permanent failure, and the maximum vacuum chamber pressure that emission could sustain.

5.1.3 Re-generable Field Emitters

5.1.3.1 Re-generable Field Emitter Lifetime

To investigate the lifetime of re-generable emitters, a dual ion/electron source was placed in the UHV chamber and the tank was evacuated to a vacuum pressure of 10^{-9} Torr. The emitter was then heated and operated at 20 μ A of ion emission current for one minute 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 μ A. The voltage required for 4 μ 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 μ A as a target was because operating a single needle electron source at emission currents of 10's of μ A 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 μ A for a few minutes and then quickly decreased down to about 3 μ A as shown in Figure 65. The emission current fluctuated between about 2 and 4 μ A after the first hour until about t_e = 600 hours. At 600 hours into the experiment the emission quickly increased to about 5 μ A and then slowly decreased down to about 3 μ A again. At t_e = 950 hours of operation the experiment was voluntarily shut off for 5 hours to observe if the emission current would return to the same magnitude when turned back on. Shutting off the experiment was achieved by simply decreasing the extraction voltage from 4.1 kV to 0 kV.



Figure 65. Electron emission lifetime experiment from a single-needle LMIS quenched at ion emission current of 20 µA, showing locations of voluntary shutdowns.

The experiment was turned off for 5 hours and then the extraction electrode was increased back to the original setpoint of 4.1 kV. The emission current increased to almost 10 μ A after the onset of emission but quickly decreased to about 5 μ A at t_e = 952 hours. Then the electron emission current remained at 5 μ A until approximately t = 975 hours and then rapidly decreased to 2 μ A. Once reaching 2 μ A the emission current slowly increased up near 3 μ A until t_e = 1025 hours. The experiment was then voluntarily shut down for a span of 4 hours. During the 4 hour period of downtime, an electron I-V sweep was taken to estimate the emitter tip radius, as shown in Figure 66. Applying the Fowler-Nordheim model to the I-V data yielded a tip radius estimate of 9.5 ± 1.9 nm



Figure 66. Electron I-V sweep taken after 1025 hours into a field emission electron lifetime experiment. The I-V sweep at 1460 hours corresponds to an emitter tip radius of 9.5 ± 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 3 μ A ± 1 μ A for 435 hours of operation, bringing the total duration of the lifetime test thus far to 1460 hours. At t_e = 1460 hours, the experiment was voluntarily shut down again to obtain another electron emission I-V sweep, as shown in Figure 67. 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 model was applied to the electron I-V data acquired during the third shutdown and yielded a tip radius estimate of 16.4 ± 3.3 nm.



Figure 67. Electron I-V sweeps taken after 1025 and 1460 hours into a field emission electron lifetime experiment. The I-V sweep at 1460 hours, plotted on the right, corresponds to an emitter tip radius of 16.4 ± 3.3 nm.

Following the I-V sweep, the extraction voltage was again increased to 4.1 kV. The emission current gradually increased back to near 3 μ A as the extraction voltage was increased. The lifetime experiment was continued for 300 additional hours, bringing the total lifetime test to about t_e = 1750 hours. The lifetime experiment was ended after 1750 hours 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 nano-structure tip radius, as shown in Figure 68. 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.



Figure 68. Electron I-V sweeps taken after 1460 and 1750 hours into a field emission electron lifetime experiment. The I-V sweep at 1750 hours, plotted on the right, corresponds to an emitter tip radius of 16.2 ± 3.2 nm.

With so few electron I-V curves taken it is difficult to determine whether the emitter tip radius from which Fowler emission was occurring was actually degrading over time. However, the trend that is apparent from the set of data that was acquired showed that the electron emitting tip slightly increased in tip radius over time. An increasing emitter tip radius was expected due to the nature of operating a field emission cathode for a long duration of time. Heating of the emitter tip due to electron emission current was expected, as well as a minimal amount of ion bombardment from the small amount of background neutrals that could have been present in vicinity of the emitter tip in the UHV chamber.

5.1.3.2 Re-generable Field Emitters at Elevated Pressure

To compare with the single needle bare tungsten field emitters that were exposed to increased vacuum pressure, similar experiments were performed with a quenched LMIS. The LMIS was first operated as an ion source for 1 min at 20 μ A, 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, as shown in Figure 69. The Fowler-Nordheim model was applied and the model resulted in a tip estimate of about 13.7 ± 2.8 nm.



Figure 69. Electron I-V sweep taken from a field emitter that was re-generated at an ion current before quench of 20 μ A. Applying the Fowler-Nordheim model to the I-V data yielded a tip radius estimate of 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 μ A so the extraction voltage was decreased to 2.1 kV in an attempt to keep the emission current around 5 μ A. After the emission current remained relatively constant for approximately 1 hour 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 couldn't 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 Figure 70, along with a bound of the possible background pressure. Extrapolating the pressure data, the vacuum chamber pressure was estimated at 8 x 10^{-5} Torr. As shown, electron emission from the quenched LMIS sustained for over 15 hours, which was 15 hours longer than the smooth bare tungsten emitters.



Figure 70. Electron emission current from a quenched LMIS held at a constant extraction voltage of 2.1 kV from $t_e = 0.3$ hr to $t_e = 20$ hr as background pressure was increased.

Once emission ceased, the ion pump on the vacuum facility was turned back on and vacuum pressure was decreased to 10^{-8} Torr. Then an electron I-V sweep was performed to obtain an estimate of the nano-structure radius that was emitting electrons, as shown in Figure 71. Applying the Fowler-Nordheim model resulted in a tip radius estimate of 12.8 ± 2.6 nm.



Figure 71. Electron I-V sweep taken from a field emitter that was re-generated at an ion current before quench of 20 μ A and the same emitter after one exposure to elevated pressure. Applying the Fowler-Nordheim model to the I-V data take after the first exposure yielded a tip radius estimate of 12.8 ± 2.6 nm.

The extraction voltage was then increased to 6.1 kV to obtain an electron emission current of about 4 μ A. The extraction voltage was adjusted during the first 30 minutes because the emission current kept increasing. At t_e = 30 minutes the extraction voltage was set to 3.4 kV to return the emission current to about ~5 μ A. The extraction voltage was held at 3.4 kV for the remainder of the experiment, which was an increase of 1.3 kV in the extraction voltage that was necessary to obtain the same magnitude of emission current as before the emission ceased. Once the emission current remained stable for 2 hours, the ion pump on the UHV system was turned off again to increase the background pressure. Electron emission current was maintained for nearly 50 hours after turning off the ion pump and increasing the vacuum pressure from 10⁻⁸ Torr to between 10⁻⁵ and 10⁻³ Torr, as shown in Figure 72. Once again, electron emission was sustained for 10's of hours longer than with a single bare tungsten field emitter.



Elapsed Time (hr)

Figure 72. Electron emission current from the same quenched LMIS used in the test of Figure 70. The emitter was held at a constant extraction voltage of 3.4 kV from $t_e = 0.5$ hr to $t_e = 50$ hr as background pressure was increased.

After electron emission ceased, the ion pump on the UHV chamber was turned back on and the chamber pressure was decreased back to 10^{-8} Torr. An electron I-V sweep was acquired, as shown in Figure 73. The Fowler-Nordheim model was applied to the sweep and resulted in a tip radius estimate of about 15.5 ± 3.1 nm, which was sharper than the previously exposed emitter tip. Looking at the I-V sweeps in Figure 73, it is apparent that the emitter tip after the first exposure should be sharper than the emitter tip after the second exposure, however, the error bars on the data could account for it.



Figure 73. Electron I-V sweeps taken from the same field emitter used in Figure 69 and Figure 71. The sweeps were taken after the first and second exposures to elevated pressure. Applying the Fowler-Nordheim model to the I-V data taken after the second exposure yielded a tip radius estimate of 15.5 ± 3.1 nm.

After emission ceased, the vacuum pressure was restored to 10^{-8} Torr and electron emission couldn't be achieved at an extraction voltage of up to 4 kV. The tip must have sustained damage from the two consecutive exposures to elevated background pressure, so the field-emitting nanostructures were re-generated through a sequence of heating, ion-emission, and quenching. The tip was operated at an ion emission current of 20 μ A at 10^{-9} Torr for 1 min while supplying 2.25 A and 0.3 V of heater power. The emitter tip was then quenched and an electron I-V sweep was acquired, as shown in Figure 74. The Fowler-Nordheim model was applied to the electron I-V sweep and resulted in a re-generated nano-structure radius of about 14.3 ± 2.9 nm, which was sharper than the emitter tip before re-generating it.



Figure 74. Electron I-V sweeps taken from the field emitter that had been exposed to elevated pressure twice, shown in Figure 73, and an electron I-V sweep taken after re-generating the

emitter at an ion current before quench of 20 μ A. Applying the Fowler-Nordheim model to the I-V data taken after tip re-generation yielded a tip radius estimate of 14.3 ± 2.9 nm.

The freshly quenched emitter tip was then operated as an electron source for over 150 hours at a background pressure of 10^{-9} Torr to demonstrate reliable operation from an emitter tip that had been exposed to increased background pressure multiple times and then had been re-generated, as shown in Figure 75. The extraction voltage was held at 4.1 kV from $t_e = 0$ to $t_e = 1.5$ hr and then the extraction voltage was decreased to 2.5 kV due to a rapid rise in emission current. The extraction voltage was held at 2.5 kV for the remainder of the experiment.



Elapsed Time (hr)

Figure 75. Extended duration experiment for an electron emitter that was re-generated by heating, acquiring ion emission, and then re-quenching after being damaged in the test sequences of Figure 70 to Figure 74. The extraction voltage was held constant at 2.5 kV from $t_e = 1.5$ hr to $t_e = 175$ hr and the test was ended by increasing the vacuum chamber pressure after over 150 hours of operation.

At $t_e = 166$ hr, the ion pump on the UHV chamber was turned off and the emission current was observed as tank pressure increased. At $t_e = 175$ hr, the emission ceased. Once electron emission ceased, the ion pump on the UHV chamber was turned back on and vacuum pressure was decreased to 10^{-9} Torr. A final electron I-V sweep was performed, as shown in Figure 76, resulting in a nano-structure radius estimation of 9.7 ± 1.9 nm after applying the Fowler-Nordheim model.



Figure 76. Electron I-V sweeps taken after re-generating the emitter at an ion current before quench of 20 μ A and then after operating the emitter for 175 hours at UHV followed by exposing the emitter to elevated pressure. Applying the Fowler-Nordheim model to the I-V data taken after tip re-generation yielded a tip radius estimate of 9.7 ± 1.9 nm.

5.1.3.3 Emitter Tip Radii Variation Discussion

Some of the emitter tips that were exposed to an increase in vacuum chamber pressure that were discussed in the previous two subsections demonstrated better electron emission performance after exposure. While no definitive argument is proposed some of the possible reasons for that phenomenon are discussed in this section.

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 μ A 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 actually sputtered away a few of the atoms on the field-emitting nano-structures, leaving an even sharper emitter.

5.2 Summary

The results from the comparison between the smooth bare tungsten, the roughened bare tungsten, and the re-generable emitter are summarized in Table 5. The table includes the length of time that emission could sustain at elevated pressure, the number of times the emitter was exposed to an increased vacuum pressure before permanent failure was observed, and the magnitude of the maximum vacuum pressure that was recorded while emission sustained.

Emitter Type	# of Tested Emitters	Emission Duration at Elevated Pressure	# of Exposures Before Failure	Maximum Tank Pressure Reached
Smooth Bare Tungsten	2	1 - 5 min	1 - 2	10 ⁻⁷ Torr
Roughened Bare Tungsten	2	1.4 - 20 hr	4 - 7	10 ⁻⁵ Torr
Re-generable Emitter	2	6 - 45 hr	Unlimited ¹	10 ⁻³ to 10 ⁻⁵ Torr

Table 5. Emitter tip comparison table displaying how long the emitter tips lasted at elevated pressure, how many exposures to increased pressure caused permanent failure, and the maximum vacuum chamber pressure that emission could sustain.

The series of experiments performed with a re-generable emitter demonstrated longer life operation than single bare tungsten field emitters at increased vacuum pressures. While the quenched LMIS required an increase in extraction electrode voltage after each exposure to increased vacuum pressure, the quenched LMIS emitters survived more harsh conditions than bare tungsten emitters and they appear to be more robust field emission electron sources. The smooth bare tungsten field emitters were irreversibly destroyed after only cycling them at increased vacuum pressures a couple of times, while the indium-coated emitters demonstrated the ability to be restored to their original performance by re-quenching them under ion-emitting conditions.

When comparing electrochemically etched bare tungsten field emitters with quenched LMIS emitters, the quenched emitters demonstrated 10's of hours of operation longer than the tungsten field emitters when exposed to increased vacuum pressure. While the tungsten field emitters would irreversibly fail after two to three cycles at background pressures greater than 10^{-7} Torr, the quenched LMIS emitters could be re-generated to their original performance after damage at elevated pressure. Furthermore, the indium-coated and quenched emitters proved to be more robust, sustaining electron emission at pressures greater than 10^{-5} Torr.

Lifetime experiments demonstrated that the re-generable 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 sites on the order of 10's of nm. The sharp nano-structures successfully demonstrated electron emission at an emission current of about 3 μ A for 1,750 hours from a single-needle that was quenched at an ion emission current of 20 μ A. Emission current from the re-generable emitters stayed relatively constant for the long duration tests while the current degraded over time for both types of bare tungsten emitters.

¹ where 'Unlimited' refers to the capability to re-generate the tip as long there is a sufficient metal supply available on the emitter tip.
References for Chapter 5

- [1] Marrese, C. M., "Compatibility of Field Emission Cathode and Electric Propulsion Technologies", *Doctoral Dissertation*, University of Michigan, 1999.
- [2] Park, Y. J., J. H. Choi, H. W. Lee, N. S. Lee, J. W. Kim, S. S. Hong, N. S. Park, J. E. Jung and J. M. Kim, "Fabrication of Spindt-type tungsten microtip field emitter arrays with optimized aluminum parting layers", J. Vac. Sci. Technol. B 18, 2, 2000.

Chapter 6

6 Surface Morphology of Re-generable Field Emitters

The primary goal of the research reported in this Chapter was to examine the nanostructures that are formed during quenching an operating ion source. To accomplish that goal, experiments were performed inside of a Field Emission Scanning Electron Microscope (FE-SEM). A secondary goal was to understand if the morphology of the structures depends on the ion emission parameters during the generation process. The experiments reported in this Chapter show micrographs of quenched emitter tips and include Fowler-Nordheim plots that were taken from the quenched emitters. At the end of this Chapter, the micrographs and Fowler-Nordheim data are compared with the data that were presented on re-generated emitter I-V performance in Chapter 4.

6.1 Custom FE-SEM apparatus

The emitter fixture that was used for the FE-SEM experiments is shown in Figure 17. As shown, some modifications were made to the general apparatus that allowed a liquid metal ion source (LMIS) to be placed in the specimen chamber of the FE-SEM for *in situ* emitter regeneration experiments. The custom fixture was equipped with electrical connections to operate the resistive heater and extraction electrode that are necessary to operate the LMIS. The electrical connections were made by three stainless steel discs that were connected to the heater and extraction electrode and were located underneath the top Teflon surface, shown in Figure 17. The three discs were used to mate with the internally mounted electrical interface. Implementing the custom fixture and electrical connections allowed the dual ion/electron source to be operated in the specimen chamber of the FE-SEM.



Figure 77. Dual electron/ion source apparatus and the custom electrical interface mounted inside the Field Emission Scanning Electron Microscope.

The chamber was evacuated using a series of three ion pumps and vacuum pressure of 10^{-7} Torr was 10^{-7} Torr was maintained throughout testing. The emitter tip and extraction electrode configuration

configuration remained the same as the previous experiments. An electrical schematic of the completed re-generable emitter apparatus is shown in

Figure 78.



Figure 78. Electrical schematic of the FE-SEM specimen fixture showing the heater supply, the extraction supply, and the ammeter placement.

For UHV 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. The internal electrical interface was installed permanently inside of the FE-SEM specimen chamber as shown in Figure 79. Electrical connections were made by inserting the custom specimen fixture so that the stainless steel electrodes that were connected to the heater and extraction electrode on the specimen fixture were made continuous with the internally mounted electrical interface by surface contact.



Figure 79. Internal specimen chamber of the FE-SEM showing the custom electrical interface that was installed to mate with the experimental apparatus' electrical contacts.

To achieve ion emission, the resistive emitter heater, shown previously in

Figure 78, was supplied with 2.75 A, 1.3 V with the purpose of maintaining the indiumcoated electrode above the melting temperature of indium, which is 156.6°C. The high-voltage extraction supply was then increased to obtain ion emission. For each set of experiments the extraction supply was increased until the desired ion emission current was reached and then the extraction electrode voltage was held constant while a pre-determined time was allowed to elapse, t_e . Leaving the extraction electrode constant, the heater power was turned off to quench the operating ion source. After the LMIS was allowed to cool for 30 seconds, the extraction supply was also turned off. Once the heater and extraction power supplies were off, the electron optics on the FE-SEM were engaged and micrographs of the emitter tip were acquired. An explanation of why the optics weren't used for *in situ* imaging is located in Appendix B. After imaging the emitter tip, the electron optics were turned off and for some of the experiments an electron I-V sweep was performed to apply to the Fowler-Nordheim model for tip radii estimations. The process of tip re-generation with subsequent imaging and electron I-V acquisition was repeated multiple times at a range of ion emission currents from 2 to 40 μ A and a range of t_e from 10 to 240 seconds.

For the temporal nano-structure formation experiments the emitter tip wasn't 'reset' between successive tip re-generations but when a new ion current before quench was chosen, the emitter tip was 'reset' using extreme electron emission conditions. To 'reset' a smooth surface, the extraction power supply was current-limited at 100 μ A while increasing the extraction voltage to about 7 kV. Using 100 μ A of emission current was great enough to heat the sharp tips and the extraction voltage was great enough to cause arcing to destroy any locally

sharp points on the apex. After resetting the emitter, the extraction polarity was reversed, the heater turned on, and the ion emission current was adjusted to the desired magnitude by adjusting the extraction voltage. Once the desired emission current was reached, the extraction voltage was held constant for a length of time, t_e . The heater was then turned off, quenching the emitter tip, and the tip was imaged using the FE-SEM.

6.2 Experimental Results

Two sets of experiments were performed within the FE-SEM and are reported in this Section. Section 6.2.1 describes the temporal evolution of a re-generable emitter tip. The regenerable emitter was operated as an ion source for 10 second intervals and then quenched and imaged using the FE-SEM. After imaging, the emitter was operated at the same ion emission current (*sans* resetting) for another 10 second interval and then quenched and imaged. The process was repeated for seven 10 second intervals (*sans* resetting between each test) and the acquired micrographs are presented to show the evolution of the emitter tip during subsequent quenches. Section 6.2.2 shows micrographs and Fowler-Nordheim analysis that were acquired after 2 minute quenches at ion emission currents ranging from 2 to 20 μ A. The data presented in the section is then compared with the data that was reported in Chapter 4. Appendix C contains an experiment where anomalous behavior was observed from one of the many regenerable emitters that were tested.

6.2.1 Temporal Nano-structure Formation

Prior to this investigation it was thought that a single jet-like protrusion would form when operating a liquid metal source. It was hypothesized that the jet-like protrusion is what would be solidified upon quenching the operating ion source and subsequently used for stable electron emission. To investigate the surface morphology, the re-generable source was operated at ion emission currents of 10, 20, and 30 μ A for 10 second intervals and imaged at each interval. The first experiment was performed at 10 μ A of ion emission current, quenching the source every 10 seconds to show the evolution of the emitter tip after seven consecutive quenches at a constant ion emission current, as shown in Figure 80. For the 10 μ A quenches, successive quenches created surface modification after about 40 seconds of ion emission.



Figure 80. FE-SEM micrographs taken after consecutive quenches at ion emission current of 10 μ A.

Micrographs that were acquired at higher magnification than those shown in Figure 80 are presented in Figure 81. It is apparent that slight surface modification occurs after the first

10 s quench at 10 μ A. After the next quench for 10 seconds the surface roughness appears to get more defined and not much changes between the second and third quench. After the fourth quench the emitter tip appears to have grown a Taylor cone type structure. After the fifth quench of 10 seconds at 10 μ A, the Taylor cone becomes more pronounced. After the sixth and seventh quenches, surface texture starts to become visible on the surface of the Taylor cone.



Figure 81. Higher magnification images of the same emitter tip shown Figure 80, taken after consecutive quenches at ion emission current of approximately 10 μA.

A question that arose and had to be addressed was if the micrographs were showing the evolution of the emitter tip or did the emitter tip relax each time it was heated to establish ion

emission, and then form a new structure? Before presenting the micrographs of an emitter that was quenched at consecutive intervals at 20 and 30 μ A, another experiment was performed to answer the proposed question. An emitter tip was 'reset' and then operated at 20 μ A for 20 seconds and quenched to form a Taylor cone. The Taylor cone was imaged with the FE-SEM and then heated in the absence of an electric field to observe if the emitter tip relaxed, as shown in Figure 82.



Figure 82. Micrographs of a 'reset' emitter, after operating the emitter for 20 seconds at 20 μ A, and then two micrographs acquired after heating the emitter for 20 second intervals in the absence of an electric field, showing that the emitter tip doesn't completely relax between consecutive quenching experiments.

The same emitter was then operated for 20 seconds at 20 μ A (*sans* resetting) and quenched. After quenching, the emitter was imaged and heated for 20 seconds in the absence of an electric field. Following the heating experiment, the emitter was imaged and heated again for an additional 20 seconds, as shown in Figure 83.



Figure 83. Micrographs of the same emitter shown in Figure 82 and then after operating the emitter for 20 seconds at 20 μ A, followed by two micrographs acquired after heating the emitter for 20 second intervals in the absence of an electric field, showing that the emitter tip doesn't relax between consecutive quenching experiments.

Since the emitter tip didn't completely relax between subsequent tests, the images in this section show the evolution of an emitter quenched at ion emission currents of 10, 20 and 30 μ A. The same emitter was then reset and used to observe nano-structure formation at higher ion quenching currents. The experiment was performed by operating the re-generable source at an ion emission current of 20 μ A with about 3 W of heater power for a quantity of seven 10 second intervals and imaging the emitter at each interval. Just as with the previous experiment the images are intended to show the temporal evolution of the emitter tip after consecutive quenches at a constant ion emission current of 20 μ A. After the first quench, at t_e = 10 s, a large cone formed at the center of the emitter apex, as shown in Figure 84.



Figure 84. FE-SEM micrographs taken after consecutive quenches at ion emission current of approximately 20 μ A.

After 10 additional seconds of operation the cone appeared to retract and some nanostructures began to form. At an elapsed time of 30 s a very well structured Taylor cone formed with some nano-structure along the surface of the apex. Additional ion quenches didn't generate many more nano-structure beyond the first 30 s. Higher magnification images of the micrographs shown in Figure 84 are presented in Figure 85 to show the nano-structure better.



Figure 85. Higher magnification images of the same emitter tip shown in Figure 84, a regenerable field emitter that was subjected to multiple 20 μ A quenches at 10 s intervals.

The higher magnification images show an interesting depression at the apex that resembles a "micro-volcano" that formed after about 40 seconds of operation. 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, In_2O_3 , 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 beneath the oxide layer that was able to break through the layer, resulting in the "micro-volcano" structure due to depletion of indium. However, that is simply a theory and the "micro-volcano" structure is not understood at this time.

After seven quenches, totaling 70 seconds of ion emission, the same re-generable source was then exposed to an electron extracting field to reset the surface. The extraction supply was current-limited at 100 μ A while increasing the extraction voltage to 'reset' the surface smoothness of the emitter. The re-generable source was operated at 30 μ A of ion emission current and then quenched after 10 s of operation. The emitter tip was imaged and operated at 30 μ A of ion emission current for six additional quenches at 10 second intervals, as shown in Figure 85.



Figure 86. FE-SEM micrographs taken after consecutive quenches at ion emission current of approximately 30 μ A.

The apex of the emitter began to form a cone after the first 10-second-quench. The cone became more defined after 20 seconds of operation and some nano-structure began to

form along the tip's surface. It wasn't until after the third 10-second-quench that the nanostructure really started to become defined, as shown in the higher magnification micrographs in Figure 87. Also, the "micro-volcano" structure appeared after the fourth quench, which was at the same point as in the 20 μ A series when the structure formed.



Figure 87. Higher magnification images of the same emitter tip shown in Figure 86, a regenerable field emitter that was subjected to multiple 30 μ A quenches at 10 s intervals.

6.2.2 Nano-structure Formation with Fowler-Nordheim Modeling

This experiment was intended to overlap with some of the data points that were acquired in Chapter 4, only within the FE-SEM rather than the UHV chamber. The purpose was to use the FE-SEM to visually verify the trends that were observed via Fowler-Nordheim modeling of data in Chapter 4. The data in Chapter 4 relied on the nano-structure of the quenched emitters physically changing shape, but there wasn't any way to verify nano-structure changes in the UHV chamber. The optical microscope that is equipped on the UHV chamber only has 90X magnification, which wasn't high enough magnification to resolve the nano- and micro-structure of quenched emitters. Obtaining higher resolution micrographs motivated this series of experiments.

These experiments were performed by quenching the re-generable emitter at multiple ion emission currents, comparable to those in Chapter 4, inside the FE-SEM. The FE-SEM was then used to image the nano-structure formation after quenching. Also, electron emission I-V sweeps were performed after each quench to estimate the emitter tip radii that were formed 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 μ A, operating each for 2 minutes exactly as in Chapter 4. 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 destroy surface features. The quenching experiments were performed in a randomized order but will be 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 μ A. After showing the individual results, a summary will be provided with all of the compiled data, including data from Chapter 4.

For each experiment the re-generable emitter was first exposed to electron 'reset' conditions as shown in Figure 88. For the 1st reported experiment, the emitter was operated at 2 μ A of ion emission current (at 3.3 kV) for 2 minutes, the same amount of time as the experiments in Chapter 4, and quenched. The other three images in Figure 88 show postquench images at increasing magnification from 2500X up to 9000X, the highest resolution that was possible to obtain during testing. As shown, a cone-type structure formed with nano-scale structures on the surface. Also, the volcano-type structure that was observed in Section 6.2.1 was also present in this test.



Figure 88. Re-generable emitter tip showing the pre-quench 'reset' and multiple images of the nano-structure after a 2 μ A quench for 2 minutes at magnification of 2500X, 5000X, and 9000X.

After quenching and imaging the emitter tip, the electron I-V sweep was performed by increasing the extraction voltage up to 2.9 kV at 50 volt increments. A Fowler-Nordheim plot was created from the sweep data, as shown in Figure 89, 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.



Figure 89. Fowler-Nordheim plot from a re-generable emitter quenched after 2 minutes at 2 μ A, yielding a tip radius estimate of 7.0 ± 1.4 nm.

The process that was just described was then repeated multiple times. For each new experiment, the ion current at quench was changed to a value between 2 and 20 μ A and then the quenched nano-structures were imaged using the FE-SEM and subjected to an electron I-V analysis. Table 6 shows the test conditions that were explored and includes the ion current before quench and the extraction voltage at quench.

Ion Current at Quench	Extraction Voltage at Quench	Chronological Order of Data	Extraction Voltage Max. for I-V Sweep
(μA)	(kV)		(kV)
2	3.3	10	2.9
3	3.4	8	3.0
5	3.8	3	3.7
6	3.2	6	4.6
10	3.5	2	4.9
10	3.4	9	3.7
15	3.9	4	3.5
16	3.4	7	4.7
20	3.7	1	2.7
20	3.8	5	3.5

Table 6. Settings that were explored for tip re-generation experiments. The ion emission current and extraction voltage at quench are presented, as well as the chronological order that the data points were acquired and the extraction voltage that was required for the post-quench electron I-V sweeps taken on the nano-structures, V_{max} .

In an effort to present the data as straightforward as possible, the FE-SEM micrographs from each experiment will be presented first. Following the complete set of micrographs, the electron I-V sweep and Fowler-Nordheim model that were acquired from the nano-structures after each quench will be reported. The data above that were used to describe the experimental procedure, in Figure 88 and Figure 89, are repeated below so that all of the data is presented together. At the end of this section, the ion current at quench and the Fowler-Nordheim emitter tip estimations will be summarized in Figure 110.



Figure 90. Micrographs repeated from Figure 88 of a re-generable emitter tip showing the prequench 'reset' and multiple images of the nano-structure after a 2 μ A quench for 2 minutes at magnification of 2500X, 5000X, and 9000X.



Figure 91. Re-generable emitter tip showing the pre-quench 'reset' and multiple images of the nano-structure after a 3 μ A quench for 2 minutes at magnification of 2500X, 5000X, and 9000X.



Figure 92. Re-generable emitter tip showing the pre-quench 'reset' and two images of the nano-structure after a 5 μ A quench for 2 minutes at magnification of 2500X and 3500X. For unknown reasons the indium didn't form a Taylor cone during this quench.



Figure 93. Re-generable emitter tip showing the pre-quench 'reset' and multiple images of the nano-structure after a 6 μ A quench for 2 minutes at magnification of 2500X, 5000X, and 9000X.



Figure 94. Re-generable emitter tip showing the pre-quench 'reset' and multiple images of the nano-structure after the first of two 10 μ A quenches for 2 minutes at magnification of 2500X, 5000X, and 9000X.



Figure 95. Re-generable emitter tip showing the pre-quench 'reset' and multiple images of the nano-structure after the second of two 10 μ A quenches for 2 minutes at magnification of 2500X, 5000X, and 9000X.



Figure 96. Re-generable emitter tip showing the pre-quench 'reset' and two images of the nano-structure after a 15 μ A quench for 2 minutes at magnification of 2500X and 3500X. Just like the 5 μ A quench, for unknown reasons the indium didn't form a Taylor cone during this quench.



Figure 97. Re-generable emitter tip showing the pre-quench 'reset' and multiple images of the nano-structure after the 16 μ A quench for 2 minutes at magnification of 2500X, 5000X, and 9000X.





Figure 99. Re-generable emitter tip showing the pre-quench 'reset' and multiple images of the nano-structure after the second of two 20 μ A quenches for 2 minutes at magnification of 2500X, 5000X, and 9000X.

As mentioned previously, all of the electron I-V data and the Fowler-Nordheim plots were grouped together. The caption for each Figure includes the ion current at quench and the emitter tip estimation results from applying the Fowler-Nordheim model. All of the data are summarized in Figure 110 at the end of this Section.



Figure 100. Electron I-V sweep and the Fowler-Nordheim plot from a re-generable emitter quenched after 2 minutes at 2 μ A, yielding a tip radius estimate of 7.0 ± 1.4 nm.



Figure 101. Electron I-V sweep and the Fowler-Nordheim plot from a re-generable emitter quenched after 2 minutes at 3 μ A, yielding a tip radius estimate of 28.9 ± 5.8 nm.



Figure 102. Electron I-V sweep and the Fowler-Nordheim plot from a re-generable emitter quenched after 2 minutes at 5 μ A, yielding a tip radius estimate of 22.6 ± 4.5 nm.



Figure 103. Electron I-V sweep and the Fowler-Nordheim plot from a re-generable emitter quenched after 2 minutes at 6 μ A, yielding a tip radius estimate of 9.0 ± 1.8 nm.



Figure 104. Electron I-V sweep and the Fowler-Nordheim plot from the first of two regenerable emitters quenched after 2 minutes at 10 μ A, yielding a tip radius estimate of 5.5 ± 1.1 nm.



Figure 105. Electron I-V sweep and the Fowler-Nordheim plot from the second of two regenerable emitters quenched after 2 minutes at 10 μ A, yielding a tip radius estimate of 14.1 ± 2.8 nm.



Figure 106. Electron I-V sweep and the Fowler-Nordheim plot from the re-generable emitter quenched after 2 minutes at 15 μ A, yielding a tip radius estimate of 29.6 ± 5.9 nm.



Figure 107. Electron I-V sweep and the Fowler-Nordheim plot from the re-generable emitter quenched after 2 minutes at 16 μ A, yielding a tip radius estimate of 9.1 ± 1.8 nm.



Figure 108. Electron I-V sweep and the Fowler-Nordheim plot from the first of two regenerable emitters quenched after 2 minutes at 20 μ A, yielding a tip radius estimate of 27.3 ± 5.5 nm.



Figure 109. Electron I-V sweep and the Fowler-Nordheim plot from the second of two regenerable emitters quenched after 2 minutes at 20 μ A, yielding a tip radius estimate of 20.4 ± 4.1 nm.

Compiling the estimated tip radii data from the 10 experiments yielded the following plot, Figure 110. As shown, there is a lot of scatter in the data and as the quench current increases there isn't a lot of variation in the emitter tip radii. Within the error bars it is difficult to note any trends.



Figure 110. Data from experiments performed inside the FE-SEM showing estimated emitter tip radius using Fowler-Nordheim modeling at various ion currents before quenching.

Looking at the FE-SEM micrographs shown previously, and one that is repeated in Figure 111, it is possible to visually make an approximation on the upper limit of the emitter tip radii. Looking at the nano-structures that are near the scale-bar it is apparent that many of the nano-structure's bases are on the order of 100's of nanometers. Therefore, the sharpest parts of the nano-structures must be smaller than the bases, which would be on the order of 1's to 10's of nanometers and correspond with the Fowler-Nordheim modeling of emitter tip radii.



Figure 111. Micrograph showing the nano-structure size on the surface of an emitter tip that was quenched at 20 μ A.

When the data acquired in the FE-SEM were combined with the data taken in the UHV chamber, reported in Chapter 4, the following plot was generated, Figure 112. All of the data points that were obtained by Fowler-Nordheim modeling of electron I-V curves taken from quenched field emitters were in the same range. As shown, there is a lot of scatter in the data but all of the emitter tip radii estimations were between 5 and 50 nm.



Ion Current at Quench (μA)

Figure 112. Data from experiments performed inside the FE-SEM and experiments performed in the UHV chamber that were reported in Chapter 4 showing estimated emitter tip radii, using Fowler-Nordheim modeling, at ion currents before quenching of 2 to 25 μ A.

6.3 Summary

The surface topography of a quenched liquid metal ion source plays an important role in the surface's potential to be used for electron field emission. Investigation of the surface structure of a quenched ion source using a Field Emission Scanning Electron Microscope resulted in the observation of multiple nano-structures that were solidified on the emitter tip rather than a single jet-like protrusion. The structures were formed from quenching a liquid metal ion source at ion emission currents ranging from 2 to 30 μ A. It was shown that a welldefined Taylor cone substructure forms after 20 to 30 seconds of ion emission and that the number of nano-structures that are formed on the surface of the Taylor cone tends to reach a maximum after 30 to 40 seconds of ion emission. The exceptions were two of the quenches, for unknown reasons a 5 and 15 µA quench didn't produce a Taylor cone substructure or multiple nano-structures, however, they still had some sharp nano-structure. Another interesting observation was that the Taylor cones didn't completely melt by adding heat in the absence of an electric field. The Fowler-Nordheim model approximations in this Chapter had comparable tip radii estimations to those reported previously in Chapter 4. Visually inspecting the nanostructures concluded that the nano-structure tip radii are on the order of 1's to 10's of nm at all quenching conditions since the bases of the nano-structures were 100's of nm, while the Fowler-Nordheim model estimations ranged from about 5 to 50 nm radii.

Chapter 7

Conclusions

7 Contributions of this Work

The purpose of the research that is described in this dissertation was to evaluate a regenerable electron source to determine the feasibility of using the re-generable source as a neutralizer for electric propulsion devices. Since the emitter tip sharpness is the most important factor influencing local electric field enhancement, for re-generable emitters to ever be implemented in space propulsion devices it was important to demonstrate that re-generable emitter tips could be formed as small as the other emitters. Emitter tip robustness was also examined to ensure that re-generated emitters could operate in elevated vacuum conditions and for long durations of time. A variety of experiments were performed in an ultra high vacuum chamber and in a Field Emission Scanning Electron Microscope to evaluate the regenerable emitter performance. The results from each set of experiments allow several conclusions to be drawn to support development of re-generable electron sources for electric space propulsion. Also, at the end of this Chapter some potential applications for re-generable electron sources are discussed as well as some recommendations for future work.

7.1 Re-generation of Emitter Tips

The capability for any space-bound piece of hardware to repair itself when its performance degrades or when the device functionality ceases to work altogether is very advantageous. Chapter 4 of this dissertation discussed how micro-scale sharpened tungsten electrodes that were coated with a thin film of indium (also known as liquid metal ion sources) could be used as re-generable field emission electrodes. Where current state-of-the-art field emission electron sources lack the ability to be re-generated, the device described in this dissertation can be repaired *in situ* an almost infinite number of times.

Through extensive performance testing it was shown that quenching an operating liquid metal ion source at different ion emission currents created nano-structures that could then be used to obtain electron field emission. Not only could sharp nano-structure be formed, but a small amount of control over the structure geometry was also demonstrated. By quenching the liquid metal ion source at higher ion emission currents, sharper nano-structures were formed. The data suggested that quenching at an ion emission current of 10-15 μ A was great enough to generate the smallest emitter tips without having to increase the emission current to a magnitude great enough for droplets of indium to be expelled.

Using Fowler-Nordheim modeling of the electron I-V data that were gathered from each of the nano-structures, it was shown that emitter tips of radii from about 45 ± 9 nm to 15 ± 3 nm could be formed, which are on the same order as the size of current state-of-the-art Spindt-type and carbon nanotube field emitters.
7.2 Re-generable Emitter Comparison with Tungsten Emitters

In order to determine how robust re-generable emitters were, lifetime experiments were performed using three types of field emitters; smooth bare tungsten, roughened bare tungsten, and the re-generable emitters. Single-needle tungsten field emitters were used for comparison rather than Spindt-type emitters to avoid complicated micro-fabrication 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 performance.

The main purpose of the comparison experiments was to determine if re-generable field emitters had similar emission characteristics to bare tungsten emitters and to determine what, if any, advantages re-generable 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 vacuum pressure environments. The smooth bare tungsten emitters ceased to function after 1-3 cycles from 10^{-8} to 10^{-7} Torr. In addition, emission would cease after only a few minutes at increased pressure.

The experiments that were performed with the electrochemically DC and AC etched roughened bare tungsten emitters exhibited better performance than the smooth bare tungsten emitters. When exposed to increased vacuum pressure, the roughened emitters could sustain emission for up to 22 hours at pressure up to 10⁻⁵ Torr. Also, the roughened emitters survived 4-7 exposures to increased pressure before the emitters were permanently damaged. As mentioned, the smooth bare tungsten emitters lasted a maximum of 3 exposures. The increased lifetime was most likely due to the multiple nano-scale sharp ridges that were created during the AC electrochemical etch. Where the DC etched emitters were smooth and had a single sharp nano-structure, the AC etched emitters had multiple longitudinal nano-structures that were capable of field emission, which allowed the roughened bare tungsten emitters to last longer than smooth bare tungsten. However, emission current from the roughened bare tungsten emitters decreased over the duration of most experiments in UHV conditions, much like the Spindt-type arrays and carbon nanotube emitters in literature, indicating emitter tip degradation over time.

The re-generable emitters demonstrated the longest lasting performance at elevated pressure of the three emitters. The re-generable emitters survived up to 45 hours at vacuum pressures up between 10⁻³ to 10⁻⁵ Torr. While the re-generable 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 re-generated and subsequently operated for 100's of hours or longer. Therefore the re-generable emitters could potentially be cycled to increased pressure an almost unlimited number of times. Not only did the re-generable 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 re-generable emitters have estimated that the re-generable emitters can be used to create the same size nano-structures as state-of-the-art field emitters, 1's to 100's of nm.

To help determine the reason for re-generable emitters lasting longer than tungsten emitters the sputter yield of indium and tungsten were calculated using TRIM-2008 software,

which is a free and downloadable program that uses the Monte Carlo simulation method. TRIM is a useful tool that is one of the most trusted and widely used sputtering applications to estimate the sputter yield of elements under a wide range of environmental and sputtering conditions and has been used for over 25 years.¹⁻³ For the experiments reported in this dissertation it was assumed that incident ions arriving at the field emitting tips were from nitrogen and were at an ion energy of 2 keV. The sputtering yield of indium resulted in 1.90 atoms/ion. Using the same simulation parameters for tungsten resulted in a sputter yield of 0.80 atoms/ion so the sputter yield of indium is over two times greater than that of tungsten.

In spite of the sputtering yield being greater for indium, the experimental lifetime and elevated pressure results showed that re-generable indium emitters lasted longer and had more stable emission than tungsten emitters. A possible reason for re-generable indium emitters lasting longer than tungsten emitters could be due to the multiple field emission sites that were on the apex of re-generable emitters. Therefore, even with the sputter yield of indium at over twice the sputter yield of tungsten there were more sites capable of field emission on the regenerable emitters. With more field emission sites as the sites were sputtered and damaged there were additional sites capable of sustaining emission. Another reason that re-generable emitters lasted longer and had more stable emission than tungsten field emitters could be due to statistical variation of the data that were collected. The comparison experiments were only performed a couple of times. Had the experimental sample been increased additional trends could have been observed. The most important thing to remember from this Chapter is that regenerable field emitters can be continually be repaired, where traditional field emitters cannot be fixed when they fail.

7.3 Surface Morphology Experiments in the FE-SEM

The main purpose of the FE-SEM experiments was to observe what happened to the surface morphology after quenching a liquid metal ion source and to compare the results with Fowler-Nordheim modeling. From literature, it was expected that a single jet-like protrusion would form at the tip of a Taylor cone. It was also expected that the geometry of the single protrusion would change depending on the ion emission current that was being emitted upon quenching. The latter did occur, but instead of a single jet-like protrusion forming many nano-structures of similar geometry were formed all over the apex of the emission electrode. As the quenching process was repeated, the protrusions that were formed became more and more defined and between subsequent quenches the overall structure of the Taylor cones maintained their shape, even when tested solely with heat in the absence of an electric field.

Formation of multiple protrusions from a quenched liquid metal ion source was a phenomenon that had never been observed but is very advantageous for re-generable cathodes. Multiple nano-structures could be responsible for the more stable emission that was observed from a re-generable source when compared to the emission that was observed from bare tungsten emitters, which degraded while operating. Since all of the nano-structures had very similar electric field enhancement, emission could have occurred from multiple nano-structures simultaneously. The multiple nano-structures that were formed on the re-generable field emitters could even act as a field emitting array, like Spindt-type arrays and carbon nanotube mesh. However, where the emission current from Spindt-type arrays and carbon nanotube mesh decreases over 10's to 1000's of hours, a single-needle re-generable emitter

didn't exhibit a decrease in emission current when held at a constant extraction voltage during a 1750 hour experiment.

An interesting feature was also present in many of the micrographs that were taken after quenching an operating liquid metal ion source multiple times. A hollow depression that looked like a "micro-volcano" formed at the emitter tip apex after subsequent 10 second quenches and appeared to form after the fourth 10 second 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 from depleting indium at the apex.

7.4 Potential Applications

An exciting possibility for implementing a re-generable electron source is with Field Emission Electric Propulsion (FEEP) systems. FEEP thrusters are created from an array of liquid metal ion sources. Potentially, an operating FEEP array could be quenched to generate an array of sharp nano-structures. Employing two FEEP arrays could then allow for one ion-emitting array to be used for thrust and another adjacent electron-emitting array as an electron source for spacecraft neutralization, as shown in Figure 113.



Figure 113. Thrust-producing FEEP-type array on the left and a quenched array on the right that could be used for electron field emission for spacecraft neutralization.

Whenever the electron field emission performance decreased, the ion emitting FEEP array could be quenched to generate new nano-structures. The array that was being used as an electron source could then be used to obtain ion emission by simply heating the array and reversing the polarity of the extraction electrode. Each array could switch roles as often as necessary to achieve optimum performance.

Arrays of re-generable emitters aren't limited to use with FEEP systems. An array of regenerable emitters could also be employed with other types of thrusters. Table 7 contains information on various low- and high-power electric propulsion thrusters. The Table includes the power level of each thruster and the amount of electron current that is required for each device. The theoretical cathode array information that is presented was taken under the assumption that 10 μ A of emission current was available from each emitter tip.

Thruster Details			Cathode Array Details		
Thruster label	Various	Thruster	Required	Estimated	Estimated
for Figure 114	Thruster	Power	Current	Emitter	Power
& Figure 115	Types	(W)	(A)	Quantity	(W)
1	FEEP/Colloid ^{4, 5}	10	0.01	1000	0.1
2	50 W Hall Thruster ⁶	50	0.5	50000	5
3	BTHT-200 ⁷	200	0.7	70000	7
4	SPT-100 ⁸	1350	4.5	450000	45
5	BPT-4000 ⁹	4500	15	1500000	150
6	10 kW Hall Thruster ¹⁰	10700	23	2300000	230
7	Low-Power Ion ¹¹	300	0.2	20000	2
8	NSTAR ¹²	2300	1.8	180000	18
9	NEXT ¹³	6900	3.52	352000	35.2
10	NEXIS ¹⁴	25000	4	400000	40

Table 7. Table showing information on existing low and high power thrusters, along with the electron current that is required from a cathode. Theoretical array details are also shown, assuming 10 μ A of electron emission current is available per emitter.

In order to achieve the required emission current, arrays of 1000's to 1,000,000's of emitters are required. If the array size was built on the same order as the thruster size, tip-to-tip spacing of 100 μ m or less would be required, which is a packing density of 10⁴ emitters/cm². Micro-fabrication techniques for Spindt-type devices have demonstrated 10⁷ emitters/cm²,¹⁵ so building the arrays could be possible. However, there are additional considerations that must be investigated – the amount of power that a large emitter array requires, how replacing traditional cathodes with field emission cathodes affects the thruster's specific impulse, and what conditions cause the emission current from the tips to be space-charge limited.

Recalling from Section 2.1, state-of-the-art field emitter arrays require on the order of 10's of volts to establish electron emission current. Assuming that each emitter tip can safely supply 10 μ A of emission current, approximately 0.1 mW of power is required for a single emitter to operate. Looking at Table 7 for the SPT-100 thruster, using a neutralizer with 450,000 emitters would be necessary to supply the required electron current for the SPT-100, which corresponds to 45 Watts of power for the neutralizer. That amount of power is on the same order of magnitude as thermionic cathodes. Using a field emission array for the BPT-4000 would require 1,500,000 emitters, or about 150 Watts of power, which is more power than some thermionic cathodes use. With that in mind, using arrays of re-generable emitters would require the same order of magnitude, or more, power than thermionic cathodes at the point where the arrays had to be produced larger than a few hundred thousand emitters.

To determine which types of thrusters would benefit from using a field emission array the fraction of cathode power to thruster power was calculated and plotted for each thruster, as shown in Figure 114. As stated previously, the field emission array power was estimated assuming each emitter tip could provide 10 μ A of emission current at an extraction voltage of 10

V. Also included in Figure 114 is a dashed line representing a 'typical' cathode (such as a thermionic cathode) operating at a constant 50 W as a reference.



Figure 114. Plot showing the fraction of cathode power to thruster power vs. total thruster power. Each data point represents the estimated power that would be required if a field emission array were used to provide the thruster's electron current and the number next to each point corresponds to a thruster from Table 7. The dashed line represents a cathode operating at 50 W of power to establish a baseline for comparison, which is an approximate power level required for a thermionic cathode.

Using a 50 W cathode as a reference allows for a couple of conclusions to be drawn concerning the power consumption of cathodes for a variety of electric propulsion devices. For FEEP and Colloid thrusters the fraction of cathode power to thruster power using a field emission array is much lower than if a 50 W thermionic cathode were used. Therefore, implementing a field emission array for FEEP and Colloid thrusters would be advantageous.

For Hall-effect thrusters there is a clear division between where field emission arrays benefit and where using traditional thermionic cathodes are beneficial. At a Hall-effect thruster power under about 1 kW field emission arrays would use less power. In fact, some of the electron sources that were used for the low-power Hall-effect thrusters that are listed in Table 7 were thermionic and operated at higher power than the thruster discharge so implementing a field emission array would be very advantageous. However, at thruster power just over 1 kW it would be more beneficial to use a thermionic cathode.

The electron sources used for ion thruster neutralization appear to fall into more of a gray area since ion thrusters are typically operated at high voltage and relatively low beam currents. At low power (< 1 kW) it is apparent that using a field emission array requires less power than a thermionic cathode. At ion thruster power levels over 1 kW, field emission arrays

and a 50 W electron source are nearly equivalent in terms of power consumption so either device could be used.

In addition to investigating power consumption when using a thermionic or a field emission cathode, the mass flow requirement of each electron source has a large impact on a thruster's specific impulse, or I_{sp} , which is

$$I_{sp} = \frac{F_T}{\dot{m}g_o} = \frac{F_T}{(\dot{m}_a + \dot{m}_c)g_o},$$
 Equation 7.1

where F_{τ} is thrust, \dot{m}_a is anode mass flow, \dot{m}_c is cathode mass flow, and g_o is the acceleration due to gravity. When the I_{sp} that is reported in literature for the thrusters listed in Table 7 are adjusted to exclude the mass flow contribution from a cathode there is an increase in I_{sp} , as shown in Figure 115.



Figure 115. Plot showing actual specific impulse of Hall-effect and ion thrusters from Table 7 with a number next to each point corresponding to a thruster in Table 7. Also included is the approximate specific impulse at each power level if a field emission electron source were used to replace the existing cathode. Since field emission electron sources do not require propellant there is an increase in specific impulse for each thruster.

Since field emission arrays do not require propellant flow, implementing the arrays in place of thermionic cathodes for any electric propulsion system increases the specific impulse. For Hall-effect thrusters the increase is relatively small but for higher power ion thrusters there appears to be a large increase in I_{sp} when using electron sources that do not require propellant.

The final consideration in implementing field emission arrays with electric propulsion systems is space-charge current limit. Fortunately, Marrese focused much of her doctoral work on the topic. Marrese researched various Spindt-type field emitter arrays, including silicon and

molybdenum, and modeled space-charge current limits when operating the arrays near various thrusters. A critical factor that was investigated in order to operate field emission arrays near electric propulsion thrusters was the maximum extraction voltage that could be used to provide a reasonable length of time for field emitters to survive. Marrese found the maximum extraction voltage for silicon and molybdenum field emission arrays was 30 and 19 volts, respectively, when operated in Hall-effect and ion thruster environments.¹⁶

Through modeling Marrese also found that if field emission electron sources were carefully dimensioned and positioned the arrays could supply enough current without being limited by space-charge effects. Marrese used a 1-D planar sheath model to estimate the space-charge limitations of field emission cathodes operating in different thruster environments. The planar model was used since the sheath of a field emission cathode is geometry and environment dependent and because the physical size of the field emitters were larger than several Debye lengths.¹⁶ If the field emitters were smaller than the sheath thickness, the spherical sheath model should yield a closer space-charge current limit approximation.

Using the planar sheath model Marrese made conservative estimates of space-charge limitations and found that field emission arrays could provide a maximum current density for Hall-effect and ion thrusters of 1.7 A/cm² and 8 A/cm², respectively, when the arrays were placed within the discharge chamber of an ion thruster and at the center of a 1.4 kW Hall-effect thruster.¹⁶ When a 1.44 cm² array was modeled outside of ion and Hall-effect thrusters the array was able to produce approximately 68 mA/cm² so multiple arrays or larger arrays would be necessary to provide enough current for thruster operation.¹⁶ For micro-propulsion applications, electron emission current requirements are lower and space-charge limitations are not predicted to be a problem.

To summarize, implementing field emission arrays into thruster environments definitely limits the emission current of the arrays but doesn't appear to be a show-stopper. By carefully controlling the cathode dimensions and physical placement of field emission arrays the arrays have the potential to supply enough current to be used for micro-propulsion, Hall-effect thrusters, and ion thrusters.

7.5 Future Work

The experimental data and the conclusions that were reported within this document show potential for re-generable electron sources to be applied to electric propulsion systems. The following additional work would be beneficial to further characterize the re-generable electron source:

- Operating a re-generable electron source near a plasma source. If re-generable electron sources are ever to be used on a spacecraft, they would have to be characterized in an environment similar to the environment that will be present near a spacecraft in orbit. Other field emission cathodes have already been characterized near a Hall-effect thruster with promising results.¹⁶
- Scaling up the number of emission electrodes and increasing the electron emission current from each emitter. While a lot was learned on the micro- and nano-scale about what is occurring at the apex of an operating liquid metal ion source and during the quenching process, it would take a large array of emission electrodes to supply enough emission current to be used in conjunction with a space propulsion device.

References for Chapter 7

[1] Ecke, G., R. Kosiba, V. Kharlamov, Y. Trushin and J. Pezoldt, "The estimation of sputtering yields for SiC and Si", *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 196, 1-2, 39-50 2002.

[2] Biersack, J. P., "Computer simulation of sputtering", *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 27, 1, 21-36, 1987.

[3] Eckstein, W. and J. Biersack, "Sputtering investigations with the Monte Carlo program TRIM SP", *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 2, 1-3, 550-554, 1984.

[4] Ziemer, J. K. and S. M. Merkowitz, "Microthrust Propulsion for the LISA Mission", *40th AIAA/ASME/SAE/ASEE Joint Propulsion Conference and Exhibit*, AIAA-2004-3439, Fort Lauderdale, FL, 2004.

[5] Hruby, V., M. Gamero-Castano, P. Falkos and S. Shenoy, "Micro Newton Colloid Thruster System Development", *27th International Electric Propulsion Conference*,, IEPC-01-281, Pasadena, CA, 2001.

[6] Khayms, V. and M. Martinez-Sanchez, "Fifty-Watt Hall Thruster for Microsatellites", 2000.

[7] Hruby, V., J. Monheiser, B. Pote, C. Freeman and W. Connolly, "Low Power Hall Thruster Propulsion System", *International Electric Propulsion Conference*, IEPC-99-092, 1999.

[8] Garner, C. E., J. R. Brophy, J. E. Polk and L. C. Pless, "A 5,730-Hr Cyclic Endurance Test Of The SPT-100", *Proceedings of the 24th International Electric Propulsion Conference*, 1995.

[9] deGrys, K., N. Meckel, G. Callis, D. Greisen, A. Hoskins, D. King and F. Wilson, "Development and Testing of a 4500 Watt Flight Type Hall Thruster and Cathode", *27th International Electric Propulsion Conference*, IEPC-01-011, Pasadena, CA, 2001.

[10] Jankovsky, R. S., C. McLean and J. McVey, "Preliminary Evaluation of a 10 kW Hall Thruster", *37th Aerospace Sciences Meeting & Exhibit*, AIAA–99–0456, Reno, Nevada, 1999.

[11] Patterson, M. J., "Low-Power Ion Thruster Development Status", *34th Joint Propulsion Conference and Exhibit*, AIAA-98-3347, Cleveland, Ohio, 1998.

[12] Polk, J. E., J. R. Anderson, J. R. Brophy, V. K. Rawlin, M. J. Patterson, J. Sovey and J. Hamley, "An Overview of the Results from an 8200 Hour Wear Test of the NSTAR Ion Thruster", AIAA 99-2446, 1999.

[13] Soulas, G. C., H. Kamhawi and M. J. Patterson, "NEXT Ion Engine 2000 Hour Wear Test Results", *40th Joint Propulsion Conference and Exhibit*, AIAA–2004–3791, Fort Lauderdale, FL, 2004.

[14] Goebel, D. M., J. Polk and A. Sengupta, "Discharge chamber performance of the NEXIS ion thruster", *40th AIAA/ASME/SAE/ASEE Joint Propulsion Conference*, Fort Lauderdale, FL, 2004.

[15] Spindt, C. A., C. E. Holland, A. Rosengreen and I. Brodie, "Field-Emitter Arrays for Vacuum Microelectronics", *IEEE Transactions on Electron Devices*, 38, 10, 2355-2363, 1991.

[16] Marrese, C. M., "Compatibility of Field Emission Cathode and Electric Propulsion Technologies", *Doctoral Dissertation*, University of Michigan, 1999.

Appendices

Appendix A – Statistical t-test Results for Chapter 4

To compare the tip radius estimations that were made in Chapter 4 using Gomer's technique of Fowler-Nordheim modeling,¹ a two-sample t-test was performed (assuming equal variances) with the radii approximations. The groups of data were compared against each other to see if there were any statistically meaningful trends between the estimated emitter tip radii and the ion current at quench. The data were grouped as shown in Table 8.

Ion Quench Current (µA)	Estimated Emitter Tip Radii (nm)				
< 5	30.0	45.8	16.9	36.9	27.4
5-9	18.3	27.1	12.5	21.7	14.0
10-14	26.9	23.8	16.0		
15-19	4.6	16.8	14.2	13.9	17.5
> 20	19.6	18.7	18.2	17.1	

Table 8. Grouped emitter tip radii estimations from Chapter 4 that were used to gather a statistically meaningful comparison between ion quench current and emitter tip radius.

Each group was compared against each other group, as shown in the following tables. The null hypothesis was that one group has the same emitter tip radii as the group that it is being tested against. Assuming a 95% confidence interval, to accept the null hypothesis the two-tail p-value must be greater than 0.05. To accept the alternative hypothesis, that one group has different tip radii than the group it is being tested against, the two-tail p-value must be less than 0.05. The results from the t-tests were reported in Chapter 4.

Groups <5 and 5-9		
t-Test: Two-Sample Assuming Equal Variances		
	Variable 1	Variable 2
Mean	31.46	18.72
Variance	116.26	35.06
Observations	5.00	5.00
Pooled Variance	75.66	
Hypothesized Mean Difference	0.00	
df	8.00	
t Stat	2.32	
P(T<=t) one-tail	0.02	
t Critical one-tail	1.86	
P(T<=t) two-tail	0.05	
t Critical two-tail	2.31	

Table 9. Two-sampled t-test using Microsoft Excel's Data Analysis Toolkit to compare the group of emitters quenched at less than 5 μ A with the group quenched from 5 to 9 μ A.

Groups <5 and 10-14		
t-Test: Two-Sample Assuming Equal Variances		
	Variable 1	Variable 2
Mean	31.46	22.23
Variance	116.26	31.54
Observations	5.00	3.00
Pooled Variance	88.02	
Hypothesized Mean Difference	0.00	
df	6.00	
t Stat	1.35	
P(T<=t) one-tail	0.11	
t Critical one-tail	1.94	
P(T<=t) two-tail	0.23	
t Critical two-tail	2.45	

Table 10. Two-sampled t-test using Microsoft Excel's Data Analysis Toolkit to compare the group of emitters quenched at less than 5 μ A with the group quenched from 10 to 14 μ A.

Groups <5 and 15-19		
t-Test: Two-Sample Assuming Equal Variances		
	Variable 1	Variable 2
Mean	31.46	13.41
Variance	116.26	26.54
Observations	5.00	5.00
Pooled Variance	71.40	
Hypothesized Mean Difference	0.00	
df	8.00	
t Stat	3.38	
P(T<=t) one-tail	0.00	
t Critical one-tail	1.86	
P(T<=t) two-tail	0.01	
t Critical two-tail	2.31	

Table 11. Two-sampled t-test using Microsoft Excel's Data Analysis Toolkit to compare the group of emitters quenched at less than 5 μ A with the group quenched from 15 to 19 μ A.

Variable 1	Variable 2
31.46	18.40
116.26	1.09
5.00	4.00
66.90	
0.00	
7.00	
2.38	
0.02	
1.89	
0.05	
2.36	
	Variable 1 31.46 116.26 5.00 66.90 0.00 7.00 2.38 0.02 1.89 0.05 2.36

Table 12. Two-sampled t-test using Microsoft Excel's Data Analysis Toolkit to compare the group of emitters quenched at less than 5 μ A with the group quenched from 15 to 19 μ A.

Groups 5-9 and 10-14		
t-Test: Two-Sample Assuming Equal Variances		
	Variable 1	Variable 2
Mean	18.72	22.23
Variance	35.06	31.54
Observations	5.00	3.00
Pooled Variance	33.89	
Hypothesized Mean Difference	0.00	
df	6.00	
t Stat	-0.83	
P(T<=t) one-tail	0.22	
t Critical one-tail	1.94	
P(T<=t) two-tail	0.44	
t Critical two-tail	2.45	

Table 13. Two-sampled t-test using Microsoft Excel's Data Analysis Toolkit to compare the group of emitters quenched from 5 to 9 μ A with the group quenched from 10 to 14 μ A.

Groups 5-9 and 15-19		
t-Test: Two-Sample Assuming Equal Variances		
	Variable 1	Variable 2
Mean	18.72	13.41
Variance	35.06	26.54
Observations	5.00	5.00
Pooled Variance	30.80	
Hypothesized Mean Difference	0.00	
df	8.00	
t Stat	1.51	
P(T<=t) one-tail	0.08	
t Critical one-tail	1.86	
P(T<=t) two-tail	0.17	
t Critical two-tail	2.31	

Table 14. Two-sampled t-test using Microsoft Excel's Data Analysis Toolkit to compare the group of emitters quenched from 5 to 9 μ A with the group quenched from 15 to 19 μ A.

Groups 5-9 and >20		
t-Test: Two-Sample Assuming Equal Variances		
	Variable 1	Variable 2
Mean	18.72	18.40
Variance	35.06	1.09
Observations	5.00	4.00
Pooled Variance	20.50	
Hypothesized Mean Difference	0.00	
df	7.00	
t Stat	0.11	
P(T<=t) one-tail	0.46	
t Critical one-tail	1.89	
P(T<=t) two-tail	0.92	
t Critical two-tail	2.36	

Table 15. Two-sampled t-test using Microsoft Excel's Data Analysis Toolkit to compare the group of emitters quenched from 5 to 9 μ A with the group quenched >20 μ A.

Groups 10-14 and 15-19		
t-Test: Two-Sample Assuming Equal Variances		
	Variable 1	Variable 2
Mean	22.23	13.41
Variance	31.54	26.54
Observations	3.00	5.00
Pooled Variance	28.21	
Hypothesized Mean Difference	0.00	
df	6.00	
t Stat	2.28	
P(T<=t) one-tail	0.03	
t Critical one-tail	1.94	
P(T<=t) two-tail	0.06	
t Critical two-tail	2.45	

Table 16. Two-sampled t-test using Microsoft Excel's Data Analysis Toolkit to compare the group of emitters quenched from 10 to 14 μ A with the group quenched from 15 to 19 μ A.

Groups 10-14 and >20		
t-Test: Two-Sample Assuming Equal Variances		
	Variable 1	Variable 2
Mean	22.23	18.40
Variance	31.54	1.09
Observations	3.00	4.00
Pooled Variance	13.27	
Hypothesized Mean Difference	0.00	
df	5.00	
t Stat	1.38	
P(T<=t) one-tail	0.11	
t Critical one-tail	2.02	
P(T<=t) two-tail	0.23	
t Critical two-tail	2.57	

Table 17. Two-sampled t-test using Microsoft Excel's Data Analysis Toolkit to compare the group of emitters quenched from 10 to 14 μ A with the group quenched at >20 μ A.

Groups 15-19 and >20		
t-Test: Two-Sample Assuming Equal Variances		
	Variable 1	Variable 2
Mean	13.41	18.40
Variance	26.54	1.09
Observations	5.00	4.00
Pooled Variance	15.63	
Hypothesized Mean Difference	0.00	
df	7.00	
t Stat	-1.88	
P(T<=t) one-tail	0.05	
t Critical one-tail	1.89	
P(T<=t) two-tail	0.10	
t Critical two-tail	2.36	

Table 18. Two-sampled t-test using Microsoft Excel's Data Analysis Toolkit to compare the group of emitters quenched from 15 to 19 μ A with the group quenched at >20 μ A.

Appendix B – Electron Optics and in situ Imaging

The first attempt at *in situ* observation of a liquid metal ion source in the FE-SEM went surprisingly well for observing unique results but not very well for observing Taylor cone formation. Operating the dual ion/electron source in ion emission mode proved to work perfectly with all of the electron optics turned off. That was expected since it is simply a vacuum chamber when the optics are disengaged. However, when the microscope's electron beam was turned on for imaging (using the secondary electron detector) some interesting micrographs were acquired. Understanding how the secondary electron detector works will help to understand the micrographs so the discussion will begin with electron optics. A diagram showing the incident electron beam from a scanning electron microscope interacting with a sample specimen is shown in Figure 116. Most of the incident electron beam has some sort of interaction with the atoms inside the sample causing the electrons to scatter in a variety of ways.





The secondary electron detector is the detector that is used for most imaging applications. The detector functions by collecting the low energy secondary electrons that are ejected from a specimen due to interactions with the energetic beam electrons. The collected electrons are used to create a detailed image of their source sample by using an Everhart-Thornley detector. The detector was named after the two designers who created it in 1960 and the detector is used to create an image of a sample specimen by combining a scintillator and photomultiplier.² A scintillator is a substance that exhibits luminescence when it is struck by an incident high energy particle. For electron microscopy, the detector is electrically biased at a positive potential to attract and accelerate the sample specimen's ejected secondary electrons

towards the scintillator. The scintillator then produces flashes of visible light with intensity that corresponds to the incident electron energy. The light intensity is then amplified by the photomultiplier and can be output as a two dimensional intensity distribution. The two dimensional display is then converted from an analog signal to a digital signal that is rastered using the microscopy software.

For the *in situ* experiment, as the potential between the liquid metal ion source and the extraction electrode was increased, the equipotential lines between the two electrodes could be rendered using the FE-SEM and became visible to the naked eye. Due to the interaction of the microscope's electron beam with the applied electric field, the electron detector rendered images showing the electrons that were deflected along the equipotential lines. Micrographs were captured when the potential difference between the emitter and the extraction electrode was 3.0, 3.5, 4.0, and 4.5 kV, as shown in Figure 117.



Figure 117. Micrographs showing the electric field contour caused by electron beam deflection from the applied electric field at extraction electrode voltages of a) -3.0, b) -3.5, c) - 4.0, and d) -4.5 kV with a gap spacing of 0.55 mm.

Maxwell SV electrostatic –field simulation software was used to model the emitter tip and extraction electrode geometry as closely as possible to the images acquired for Figure 117. Simulated boundary conditions were used with a grounded emitter tip and a negative potential applied to the extraction electrode of -4.5 kV, as in Figure 117 d). An interesting, but not surprising, result was that the simulated equipotential lines were nearly identical to the *in situ* observations that were made, as shown in Figure 118.



Figure 118. Equipotential line comparison between a) a Maxwell SV electrostatic model and b) *in situ* observation. In both images the emitter tip is at ground potential and the extraction electrode is biased at negative 4.5 kV.

Since the actual emitter tip isn't a perfectly smooth needle with a single sharp apex like the model in Figure 118 a), the jagged lines on the left side of the emitter tip in Figure 118 b) could be due to small protrusions and imperfections on the surface of the indium coating which is causing a small deviation in the electric field symmetry. However, the vast majority of the electric field contour corresponds closely in the remainder of the figure.

As interesting as it was to watch the electric field contour change as the potential between the source and extraction electrodes was increased, it proved to be impossible to observe the emitter tip in situ while the apex was emitting ion current using the secondary electron detector. At the instant that emission current was onset, the secondary electron detector became completely saturated, which caused the observed image to be washed out. After adjusting the electron optics for quite some time, a method of viewing the emitter tip at extremely low magnification was discovered. By using the backscatter electron detector rather than the secondary electron detector, viewing the apex of the emission electrode (while the electrode sustained a discharge current of ions) was possible. A backscatter electron detector is positioned above a sample specimen within the microscope and collects high energy beam electrons that are reflected away (or backscattered) from the sample. Backscatter detectors are able to collect higher energy electrons due to the precise positioning of the detector. The placement of a secondary electron detector is typically to one side of a specimen which isn't the optimal location to collect reflected electrons. Also, the relatively low potential that the secondary electron detector is biased at isn't great enough to attract the high energy electrons that the backscatter detector can collect. Therefore backscatter detector placement is very important and must be at the most favorable angle with respect to the specimen. Since the backscatter electron detector is capable of collecting higher energy electrons than the secondary electron detector, it was possible to resolve a low-magnification image of the operating liquid metal ion source as displayed in Figure 119b).



Figure 119. Image taken a) using the FE-SEM's secondary electron detector before operating as a liquid metal ion source and b) using the backscattered electron detector during Taylor cone formation.

Even though *in situ* observation of an operating liquid metal ion source proved to be possible, the resolution necessary to distinguish nanometer-scale features was unobtainable. As the magnification was increased on the microscope, the optics that control and focus the electron microscope's incident electron beam must be constantly adjusted to maintain a decent resolution image due to deflection of the incident electron beam from the electric field that is created when operating a liquid metal ion source. That is probably why a Transmission Electron Microscope (TEM) is what has been used to image an operating LMIS in the past.³⁻⁶ Most TEMs are capable of accelerating voltages of over 1 MV, where the FE-SEM used for this investigation is only capable of 50 kV.

Appendix C – Anomalous FE-SEM Experiment

One indium-coated emitter, out of the 10's of emitters that were tested, had surprising results. Rather than forming a single Taylor cone with sharp nano-structures by quenching an operating ion source, something during the ion emission process caused the formation of multiple crystal-like structures all over the surface of the emitter tip, as shown in Figure 120. Multiple protrusions were formed upon quenching and as sequential quenches were performed, the protrusions that were created become more and more pronounced.



Figure 120. Micrographs obtained a) after destroying the emitter tip using electron emission and then b) after operating the emitter as an LMIS for one minute and quenching at 20 μ A and then c) operating as an LMIS for one additional minute and quenching at 20 μ A, and finally d) operating as an LMIS for one more minute followed by an ion quench at 20 μ A. The inset square shows the location where the magnified images in Figure 121 were acquired.

A higher-magnification examination of the surface topography indicated that the sharp protrusions had the appearance of crystal facets as shown in Figure 121. Furthermore, it was evident that the surface layer did not melt between tests, despite the application of a considerable amount of heat producing an approximate surface temperature of 1250°C, which is some 1090 degrees above the melting temperature of indium.



Figure 121. Magnified micrographs of the sharp nano-structures shown in Figure 120 b), c), and d) highlighting the crystal-like structures that have been formed at the emitter tip apex after a series of 20 μ A quenches.

The quenching experiment was then performed again to determine if crystals could be re-formed. To reset the smooth surface, the emission electrode was operated as an electron source by current-limiting the extraction supply at 100 μ A and then increasing the voltage to approximately 7 to 10 kV for one minute. After smoothing the tip and acquiring a micrograph, the extraction electrode polarity was reversed to obtain ion emission, the heater power was increased to about 4 W, and the extraction voltage was increased to obtain ion emission. Just as with the first quenching experiment in this section, the extraction voltage was adjusted to obtain the desired emission current and then it was left constant for one minute. After one minute of stable ion emission the heater power was turned off which quenched the emitter.

The emitter tip was imaged with the FE-SEM and then the heater was increased back to 4 W, the extraction voltage was increased to obtain the desired emission current, the current was maintained for one minute, and then the heater was turned off to quench the nanostructure. The same process was repeated a third time at the same emission current and the acquired images can be seen in Figure 122. Also, a strange feature resembling coral formed on the underside of the emitter after 120 s of operation.



Figure 122. Micrographs obtained a) after destroying the emitter tip using electron emission and then b) after operating the emitter as an LMIS for one minute and quenching at 10 μ A and then c) operating as an LMIS for one additional minute and quenching at 10 μ A, and finally d) operating as an LMIS for one more minute followed by an ion quench at 10 μ A. The inset square shows the location where the magnified images in Figure 123 were acquired.



Figure 123. Magnified micrographs of the nano-structures shown in Figure 122 b), c), and d) highlighting the crystal-like structures that have been formed at the emitter tip apex after a series of 10 μ A quenches.

The entire quenching process, starting with the electron emission 'reset' to smooth the tip and then performing three consecutive heating/quenching cycles at the same ion emission current, was then performed at ion emission currents of 30 and 40 μ A. The micrographs that 145

were obtained are shown in Figure 124 and Figure 125 for the 30 μA quenches and Figure 126 and Figure 127 for the 40 μA quenches.



Figure 124. Micrographs obtained a) after destroying the emitter tip using electron emission and then b) after operating the emitter as an LMIS for one minute and quenching at 30 μ A and then c) operating as an LMIS for one additional minute and quenching at 30 μ A, and finally d) operating as an LMIS for one more minute followed by an ion quench at 30 μ A. The inset square shows the location where the magnified images in Figure 125 were acquired.



Figure 125. Magnified micrographs of the sharp nano-structures shown in Figure 124 b), c), and d) highlighting the structures that have been formed at the emitter tip apex after a series of 30 μ A quenches.



Figure 126. Micrographs obtained a) after destroying the emitter tip using electron emission and then b) after operating the emitter as an LMIS for one minute and quenching at 40 μ A and then c) operating as an LMIS for one additional minute and quenching at 40 μ A, and finally d) operating as an LMIS for one more minute followed by an ion quench at 40 μ A. The inset square shows the location where the magnified images in Figure 127 were acquired.



Figure 127. Magnified micrographs of the sharp nano-structures shown in Figure 126 b), c), and d) highlighting the structures that have been formed at the emitter tip apex after a series of 40 μ A quenches.

The formation of multiple crystal-like pyramids brought up the question of why a single Taylor cone wasn't forming. Since the global features on the emitter apex seem unchanged after each quench, it was initially thought that the indium wasn't getting hot enough to 147

completely liquefy. If the indium was completely melting, the global features would be melted and reformed into completely different shapes during each quench. A crude heat transfer calculation which assumed a heater power of 4 W resulted in an estimated tip temperature of about 1750°C, which should be more than sufficient to melt indium (melting temperature is 156.6°C). A more in-depth thermal analysis was then performed to confirm the predicted temperature, since 1750°C would have resulted in complete vaporization of the indium film. Using thermal modeling software, the emitter tip temperature was estimated at a range of heater power from 0.25 to 8 W during ion emission. At the same magnitude of heater power used for the hand-calculation, 4 W, the thermal model estimate was a tip apex temperature of 1250°C, which is still much higher than desirable. A plot of the heater power vs. tip temperature from the thermal model is shown in Figure 128.



Figure 128. Emitter tip temperature vs. heater power estimated using thermal modeling software with inset picture showing the probe location where temperature was modeled.

Assuming that all of the boundary conditions in the thermal model were accurate, all of the indium would evaporate from the emitter tip within a few minutes due to the vapor pressure of indium at 1250°C. It was clear from the micrographs that a layer of metal was definitely coating the underlying tungsten electrode and hadn't been evaporated, and it was also clear from the thermal analyses that this layer could not be indium. The (now) natural conclusion was that the layer was likely indium oxide. Indium oxide, In₂O₃, has a melting temperature of 1910°C and tends to form tetragonal pyramid-like crystals that are very similar to the structures that were observed throughout the experimentation reported in this document.⁷ Indium oxide has a molecular weight of 82.7% indium and 17.3% oxygen so an Energy Dispersive X-ray Spectroscopy (EDS) analysis was performed on the emission electrode to determine the molecular make-up, as shown in the energy spectrum plot in Figure 129. It should be noted that the unlabeled peaks were elements that made up the microscopy adhesive that is used to adhere small SEM samples to the specimen holder.



Figure 129. Energy spectrum analysis of a dual ion/electron source using Energy Dispersive Xray Spectroscopy. The bottom axis is in units of keV and the vertical axis shows relative peak magnitudes. The unlabeled peaks were from the electron microscope adhesive used to fix the LMIS to the sample holder.

The results were conclusive that the LMIS was coated with indium oxide rather than pure indium. EDS analysis of two different locations on the LMIS, at the emitter apex and about 0.5 mm down the shaft, yielded weight percents of 83.1% indium, 16.9% oxygen and 80.7% indium, 19.3% oxygen, respectively. The relative weight percentages of indium and oxygen confirmed that the indium liquid metal ion source was definitely coated with an indium oxide layer, which could explain why a higher temperature than 156.6°C at a modest 4 W of heater power was necessary to obtain ion emission. An EDS analysis was also performed on the bulk indium that was used to coat the tungsten electrode to determine if a large amount of oxygen was present before coating the tungsten. The analysis was performed on two separate samples and resulted in 99% indium, 1% oxygen in the first sample and 98.6% indium, 1.4% oxygen in the second sample so it is safe to say that the LMIS used for testing had, at minimum, an outer shell of indium oxide.

While the confirmed presence of a layer of indium oxide explained why the structure did not melt during heating, it raises other questions relating to the ion emission process, i.e. what is being emitted during operation as an LMIS if there is no liquid metal? Two simple experiments were performed to try to isolate the source of emission; 1) heating the LMIS without an applied extraction potential and 2) applying an extraction potential without heating the LMIS. For the first experiment the LMIS was resistively heated at 4 W, which was the same heater power as the previous experiments, for one minute intervals. After applying heat for one minute and quenching the LMIS, a micrograph was acquired. The process was repeated and the LMIS was heated a total of three times for one minute intervals. The acquired images are shown in Figure 130. As shown, no crystal-like pyramids were observed by solely heating and quenching the LMIS.



Figure 130. Micrographs taken after heating and quenching the LMIS for one minute intervals without extraction voltage applied, where a) is after the first one minute heating, b) is the second one minute interval, and c) was taken after applying heat for the final one minute interval.

By applying a potential between the LMIS and the extraction electrode (without heating the LMIS) no ion emission current was measured and the micrographs didn't show any changes. Coupling the information gathered from both experiments, it is evident that both heat and the extraction potential are important factors to achieve ion emission and nano-structure formation. Further work is necessary to explore the phenomena responsible for positive current emission from an indium-oxide coated needle.

Once concluding that some sort of electrostatic modification of indium oxide was responsible for the crystal formation, an investigation of why indium oxide formed pyramid-like crystals was begun. Although a literature search didn't yield many results, a group from Peking University in China, Jia et al, were able to synthesize tetragonal indium oxide pyramids by using a chemical vapor deposition process.⁷ Although the process that Jia et al used to form In₂O₃ crystals was different from what is reported here, their research demonstrated the low-energy crystal structure that indium oxide naturally wants to relax into – a tetragonal pyramid-like structure that was observed in this research. More similar research to what is reported here was performed in 1999 by a group from the Muroran Institute of Technology in Muroran, Japan. Saito et al showed single pyramid formation from a quenched indium LMIS.⁸ The group also investigated emission patterns from a quenched indium LMIS and observed a circular electron emission pattern on a phosphor screen rather than a single point were emission occurred, implying that electron emission was taking place from multiple points around the emitter apex.

Whatever the reason for crystal formation, creating multiple field emitting nanostructures rather than a single Taylor cone has the potential to increase the lifetime of an emitter when operating as an electron source. When one pyramid wears out and 'dies', electron emission can continue from one of the neighboring pyramids, since each pyramid has a very similar geometrically enhanced electric field.

Appendix D – Microgravity Experiment

An exciting opportunity presented itself when a group of undergraduate students were searching for an experiment to perform aboard NASA's microgravity jet (the C-9B). Even though gravity shouldn't play a role in emitter tip formation, with such a delicate balance of forces present at the apex of an operating liquid metal ion source it was worth investigating the quenching of nano-structures in a microgravity environment. The experiment consisted of operating ten liquid metal ion sources aboard the microgravity aircraft and quenching the ion sources at emission currents ranging from 4 to 25 μ A while the aircraft was experiencing the zero-gravity segment of the flight. The emitter tips that were quenched in 0-g were then compared with emitter tips that were quenched in the same vacuum facility using the identical quenching process in 1-g. The aircraft that the experiments were performed aboard achieves a microgravity environment by flying in a parabolic pattern. The plane first climbs altitude at approximately a 45 degree angle, as shown in Figure 131.



Figure 131. A NASA public access image of the microgravity jet ascending during a parabola.⁹

The aircraft then noses-over and flies back toward the Earth at about a 45 degree angle. While the aircraft is nosing-over, everything inside the plane experiences a period of weightlessness. It is during the weightless period that microgravity experiments can be performed. Since the microgravity window is very small, the experiments must be on a timescale of approximately 30 seconds.

7.6 Experimental Apparatus

The microgravity experimental apparatus was designed so that 12 liquid metal ion sources could be fixed within the vacuum chamber. Each LMIS could be operated independently of the others by using an electrical switchboard made up of toggle switches that was located outside of the chamber. The switchboard allowed the operator to perform ion and electron emission experiments with an individual emitter while electrically isolating the remaining 11. An

image showing a series of four emitters, along with some of their key components, is shown in Figure 132.



Figure 132. Image showing a portion of the internal components for the microgravity experiment.

7.7 Procedure and Results

The microgravity experiments were performed on two separate flights that were backto-back on the same day. The first flight consisted of operating ten emitter tips for three microgravity parabolas each. The goal was to operate the emitters to achieve stable ion emission over the course of two parabolas and then to quench the ion emission on the third parabola so that a sharp jet-like protrusion could be solidified in a microgravity environment. The emitters were quenched at ion currents ranging from 4 to 25 μ A.

7.7.1 Flight 1 – Microgravity Nano-structure Formation

Once altitude had been reached aboard the microgravity jet, the experiments were conducted. Using the switchboard to isolate all but one LMIS, the single LMIS was resistively heated using 2 to 2.5 A of heater current at approximately 1 V. After a few seconds, the extraction electrode was biased negatively with respect to the emitter tip. The extraction potential was then increased to obtain ion emission. The extraction potential was then adjusted until the desired ion emission current was reached (ranging from 1 to 25 μ A) and then the extraction potential was held constant to allow the ion emission current to stabilize. The ion emission current was monitored over the course of two parabolas, each consisting of two 30 second intervals in zero-g and two 120 second intervals at roughly 1.8 g's. After the two

parabolas were completed and the microgravity jet continued into a third parabola, the heat was removed from the LMIS causing the emitter tip to quench in a zero gravity environment. This process was repeated for the remaining nine liquid metal ion sources at ion emission currents between 4 and 25 μ A.

7.7.2 Flight 1 – Ground Characterization

Immediately after the microgravity jet landed, ground experiments could be performed. The ground experiments consisted of operating the quenched nano-structures as field emission electron sources. By reversing the polarity of the extraction electrode so that is was positively biased with respect to the emitter tips, each tip could be used to obtain electron emission. The extraction voltage was swept from zero to the voltage that was required to obtain approximately 0.5 to 1 μ A of electron emission current. The electron emission I-V data could then be used for Fowler-Nordheim modeling to estimate the nano-structure radii.

Due to unforeseen circumstances, electron I-V data had to be taken by sweeping the extraction voltage from zero to the desired voltage by slowing increasing the extraction voltage by hand. The data presented in this section should be interpreted accordingly. The same method of Fowler-Nordheim modeling that was described earlier was used to analyze the microgravity data. Figure 133 shows the data points that were collected from the first microgravity flight.



lon Current at Quench (μA) Figure 133. Estimated emitter tip radii from microgravity data using Fowler-Nordheim modeling.

Data from Flight 2 had to be discarded due to inclement weather conditions when the microgravity airplane landed. The entire UHV facility had to remain in the plane until the weather cleared up, at which point the UHV facility had to be transported back to MTU's campus via the back of a truck before analyzing the emitter tips. Due to the emitter tips being

so close to the extraction electrode and the lack of a vibration isolation system for the facility, each of the emitter tips came into contact with the extraction electrode so the sharp nano-structures were ruined.

Once the experimental apparatus was brought back to Michigan Tech, it was reassembled in the Isp lab. The ten emitters that were operated in the microgravity flights were then used to obtain experimental ground testing data (in 1-g) to compare to the experimental microgravity data (in 0-g). The LMISs were quenched at emission currents ranging from 1 to 25 μ A and then they were used to obtain electron field emission. The electron field emission sweeps were analyzed using the Fowler-Nordheim model and then both sets of data were plotted, as shown in Figure 134.



Ion Current at Quench (μA)

Figure 134. Estimated tip radii from jet-like protrusions formed in microgravity and during ground testing using Fowler-Nordheim modeling.

As reported in Chapter 4, an investigation of the nano-structure radius concluded that there was a decrease in tip radius with an increase in ion quench current. The data collected in the microgravity environment supported the trend of decreasing tip radius as the ion quench current increased. As expected, quenching nano-structures in microgravity had no effect on the emitter tip radius. The force of gravity has a much less significant effect than the electrostatic and surface tension forces.

7.8 Summary

The experiments that were performed in a microgravity environment had nearly identical results as when the emitters were ground tested. The nano-structures that were quenched in microgravity had estimated tip radii of 3.0 ± 0.6 nm to 13.0 ± 2.6 nm when quenched at ion currents ranging from 4 to 25 μ A. The microgravity data were very comparable to the ground data of tip radii ranging from 6.0 ± 1.2 nm to 13.0 ± 2.6 nm at ion quench currents

of 1 to 28 μ A, supporting that gravity has a minimal contribution, if any, when compared to the electrostatic and surface tension forces on the emitter apex.

References for Appendices

- Gomer, R., <u>Field Emission and Field Ionization</u>. New York, NY: American Institute of Physics, 1961.
- [2] Goldstein, J., <u>Scanning Electron Microscopy and X-Ray Microanalysis</u>, <u>3rd Ed.</u> New York, NY: Springer, 2003.
- [3] Driesel, W. and C. Dietzsch, "In situ HVTEM observation of the tip shape of tin liquid metal ion sources", *Applied Surface Science*, 93, 179-190, 1996.
- [4] Driesel, W., C. Dietzsch and M. Moser, "In situ HV TEM observation of the tip shape of lead liquid metal ion sources", *J. Phys. D: Appl.Phys.*, 29, 2492-2500, 1996.
- [5] Driesel, W., C. Dietzsch, H. Niedrig and B. Praprotnik, "HV TEM in situ investigations of the tip shape of a gallium liquid-metal ion/electron emitter", *Ultramicroscopy*, 57, 45-58, 1995.
- [6] Praprotnik, B., W. Driesel, C. Dietzsch and H. Niedrig, "HV-TEM in-situ investigations of the tip shape of indium liquid metal ion emitter", *Surface Science*, 314, 353-364, 1994.
- [7] Jia, H., Y. Zhang, X. Chen, J. Shu, X. Luo, Z. Zhang and D. Yua, "Efficient field emission from single crystalline indium oxide pyramids", *Applied Physics Letters*, 82, 23, 4146-4148, 2003.
- [8] Saito, Y., H. Nakane and H. Adachi, "Field electron emission from W covered with In", *Journal of Vac. Sci. Technol. B*, 18, 2, 1093-1096, 1999.
- [9] NASA's Johnson Space Center, "Zero-G Experiments" http://zerog.jsc.nasa.gov/RGO/public/subviewer.cgi