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Cesium iodide (CsI) coated carbon fiber cathodes have shown promise as a cold cathode for microwave and x-ray devices. In particular, the cathodes have demonstrated over 1 000 000 shots lifetime at operating voltages at or in excess of 165 kV and current densities greater than 25 A/cm^2. While the vacuum emission characteristics have been well studied, the materials characteristics of the cathodes themselves, particularly after operation, have received little attention. Furthermore, while researchers at University of Wisconsin have demonstrated a reduction in work function of carbon due to the CsI coating, the details of the emission mechanism remain poorly understood. This article gives results of a series of materials diagnostics investigating the cathode surface morphology as well as the changes in the carbon fiber structure with cathode shot history. We demonstrate that the cathode surface undergoes several changes in relation to the bond line along the fiber-substrate interface as well as at the fiber tips. While the exact mechanisms leading to these changes have not been determined, we offer several possible explanations for the changes, as well as the means by which these mechanisms can be ascertained. © *2008 American Institute of Physics*.
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I. INTRODUCTION

Cesium iodide (CsI) coated carbon fiber cathodes have shown promise as a high current density electron emitter.[1,2] Such cathodes find application in a wide variety of areas, ranging from high power microwave sources, to x-ray tubes, to conventional microwave sources and possibly field emission displays. Briefly,[1,2] these cathodes consist of an array of carbon fibers pyrolytically bonded to a carbon substrate. The fibers then receive a CsI coating using either a vapor deposition technique or an aerosol spray technique. The cathodes discussed in this article have been coated using the vapor deposition technique.[2] These cathodes have been operated for slightly over 1 000 000 pulses at 165 kV and 25 A/cm^2 current density with a 64 cm^2 cathode area. Generally, researchers[3–5] have attributed the emission from cold cathodes at these current densities and voltages to explosive field emission, in which heating causes individual fibers to vaporize, leading to plasma formation along the cathode surface. Such plasmas then form a low work function material, which emits copious electrons as well as copious amounts of neutral gas. The neutral gas causes numerous difficulties for the operation of many devices, leading to effects such as pulse shortening and excessive loading of vacuum pumps. In the case of the CsI coated carbon velvet cathode, researchers developed the cathode in a successful attempt to overcome the emission of large amounts of neutral gas. Indeed, previous work demonstrated that little plasma existed at the cathode surface and, if it did exist, it had a very low temperature.[2] This article furthers the arguments presented previously for low plasma formation and contributes additional knowledge to the physical changes attendant at the cathode surface during operation. This work lays the basis for future work on the CsI-on-carbon emission process and indicates as well possible failure modes for the CsI coated carbon velvet cathode.

This article begins with a review of both the experimental apparatus used to test the cathodes as well as a description of the diagnostics. We then review the scanning electron microscopy (SEM) and Raman microscopy. After this review we proceed to discuss the experimental results, focusing on the changes of the carbon fibers as well as the changes in the fiber-substrate bond line. In some scenarios, these changes may well share a close relationship. We discuss the implications of these results for the lifetime of the cathode as well as for the CsI coating.

II. EXPERIMENTS

Figure 1 shows a schematic of the cathode test chamber. The system operates at a cold base pressure (no electron beam or high voltage on the cathode) of 5.0×10^{-9} torr with a cryogenic pump providing the vacuum. The pressure can be raised above this level, using a "calibrated leak," to the 1.0×10^{-5} torr level, with background gases of laboratory air, nitrogen, or argon. The vacuum system contains optical

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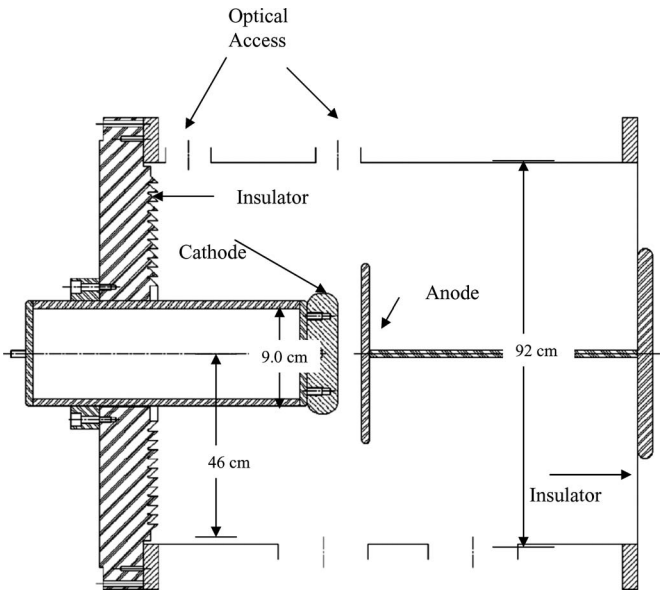

FIG. 1. Experimental schematic. The anode-cathode gap typically remains at 4 cm, but can be varied.

access, used for spectroscopy and other measurements.[2] Capacitive and resistive dividers provide voltage measurements from the oil side of the vacuum interface along the cathode shank. Current transformers provide current measurements of the cathode current, again measured along the cathode shank, and anode current, measured along the current return path. The anode, which is cooled to 20 °C using a chiller with ethylene glycol, can be retracted away from the cathode along the second vacuum interface. All of the data described here occurred at an anode-cathode (*A-K*) gap of 4 cm and a diode voltage of 165 kV. A pulse forming network (PFN) based modulator provides the high voltage to the cathode. The system pulse charges a type E PFN through a resonant charging transformer. When the PFN reaches full voltage, a blown gas switch then discharges the PFN to the cathode load. The results presented here depict experiments at a 1 microsecond (μs) pulse length at 165 kV and 2 Hz operation. Figure 2 shows current and voltage traces from the experiment taken at shots (a) 100, (b) 200 000, (c) 400 000, (d) 600 000, (e) 800 000, and (f) 1 000 000. While the ripple on both traces stems from mismatches between the PFN and diode and appears also when the pulser operates into a pure resistive load, we note that the shot to shot uniformity and reproducibility remains excellent throughout the testing.

For elementary materials diagnostics, we performed Raman microscopy using a Renishaw confocal Raman spectrometer using an Ar^+ laser operating at an excitation wavelength of 514.5 nm with a depth sensitivity of approximately 1 μm. The system utilized an 1800 lines per millimeter grating with spectra accumulated from 100 to 3200 cm^{-1}. For SEM measurements we employed a Jeol SEM model capable of energy dispersive spectroscopy and operating at voltages ranging from 5 to 15 kV.

Cathode testing proceeded as follows. Immediately after cathode substrate manufacture, but before the CsI coating process, several fibers were plucked from the center of the cathode. These fibers formed the control set; i.e., no shots

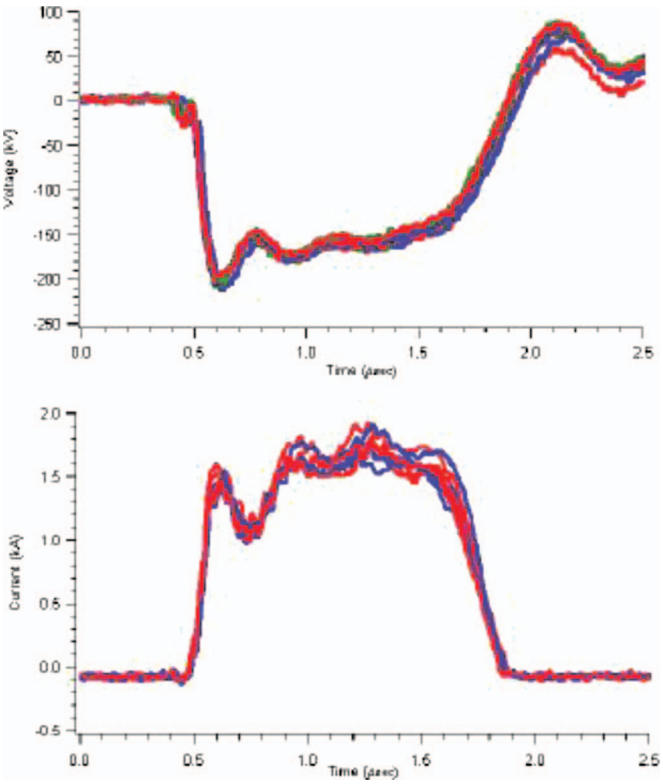

FIG. 2. (Color) Current and voltage traces during standard operation of the experiment. The ripple on current and voltage traces stems from mismatches in the PFN and diode, and occurs even with a resistive load.

and no CsI application. Due to excessive impedance collapse of the diode current with uncoated carbon velvet cathodes, we then coat the cathodes with CsI using the vapor deposition process referenced above. After coating, we fired the cathode at 165 kV and 1.6 kA with a 1 μs pulse length and 2 Hz repetition rate. Every 200 000 pulses we vented the system to atmosphere using a grade 5 dry nitrogen purge and removed (plucked) fiber samples from the cathode. After reinstalling the cathode and pumping the diode to a base pressure of at least 5×10^{-8} torr, we then resumed pulse testing on the cathode. After applying 1 000 000 pulses, we examined the fiber/substrate bond line using the SEM, a process that necessitated cleaving the cathode in order to perform the measurements. The cleaving process resulted in "fallen" fibers along the cut. We performed energy dispersive spectroscopy (EDS), SEM, and Raman spectroscopy at the bond line and along the length of the individual fibers from the control samples and those that were plucked at 0 pulses and successive sets of 200 000 to 1 000 000 pulses. Both the EDS and the confocal Raman spectroscopy have a depth sensitivity of approximately 1 μm.

III. RESULTS AND DISCUSSION

Several results hold particular interest for the cathode. First, we consider aspects of the CsI coating. Note that the current and voltage waveforms showed very little change (see Fig. 2) after 1 000 000 pulses. Further, EDS revealed the presence of Cs and I (approximately 1:1) on the coated fiber tips and within 10 μm along the length of the fiber. Neither Cs nor I was present in the EDS spectra of the bond line or

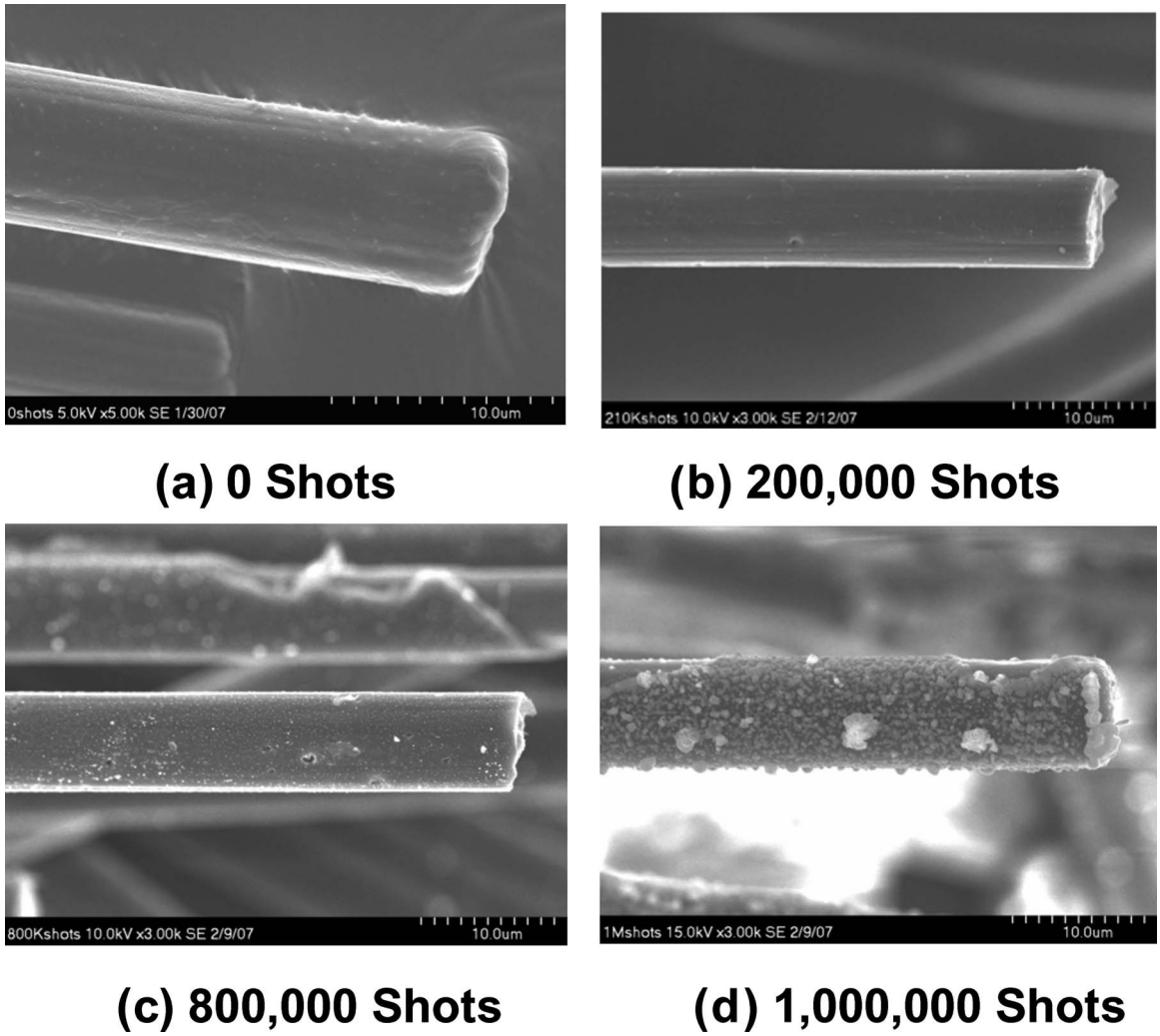

FIG. 3. SEM images of the several fiber tips, at various points during a shot sequence. Note that the fibers show no evidence of explosive emission and that CsI leaches out of the fiber during the cathode evolution.

on the fiber beyond 10 μm from the tip. The initial lack of CsI beyond 10 μm of the tip is expected because the cathode fiber density would effectively mask the vapor deposition of CsI much beyond a few micrometers depth. In addition, EDS showed that the CsI did not migrate along the length of the fiber beyond 10 μm from the tips or to the cathode surface, even after 1 000 000 pulses. Figure 3 shows SEM images of individual fibers at no shots, 200 000 shots, 800 000 shots, and 1 000 000 shots. First, we note that the as deposited CsI coated fibers look the same as uncoated fibers [see Fig. 3(a)]. However, as the count increases, CsI crystals begin forming on the surface of the fibers. These are particularly evident between 800 000 and 1 000 000 shots [Figs. 3(c) and 3(d)]. We hypothesize that the CsI intercalates into the fiber during the coating process, and begins to leach out of the fibers as the cathode accumulates more shots, forming localized CsI crystals on the surface. Finally, we note that even at 1 000 000 shots, the fibers show no evidence of the extensive damage associated with explosive field emission; i.e., no exploded fiber tips.

Next, Fig. 4 shows a cross sectional view of the substrate/fiber bond line. The fibers that have fallen in these images stem from cleaving of the cathode in order to insert it into the SEM. In these images, one sees an initially rough cathode substrate that becomes smoother by 1 000 000 shots. The original bonding process introduces submicron sized carbon soot to the substrate. These particles clearly appear in Fig. 4(a), the image taken with no shots on the cathode. However, by the time the cathode reaches 1 000 000 shots, the nature of the bond line clearly changes, as shown in Fig. 4(b). At this point, the carbon substrate appears glassy and wets the base of the carbon fibers. Recall no CsI was found at the cathode base using EDS, only carbon. No clear interpretations exist for this result, requiring further research.

Figure 5(a) shows the typical Raman spectra of a CsI coated fiber with no shots as a function of distance along the length of the fiber where 0 μm is the tip of the fiber and 1560 μm is its base. The spectra are essentially the same along the length of the fiber other than the slightly larger fluorescence background present in the spectrum taken near the base of the fiber. The prominent features of these spectra are the broad peaks at 1350 and 1590 cm^{-1} referred to as the *D* (disordered) and *G* (graphite) peaks commonly observed in primarily sp^2 bonded disordered forms of carbon. The very broad bands in the range of 2000–3000 cm^{-1} are the second order spectra.

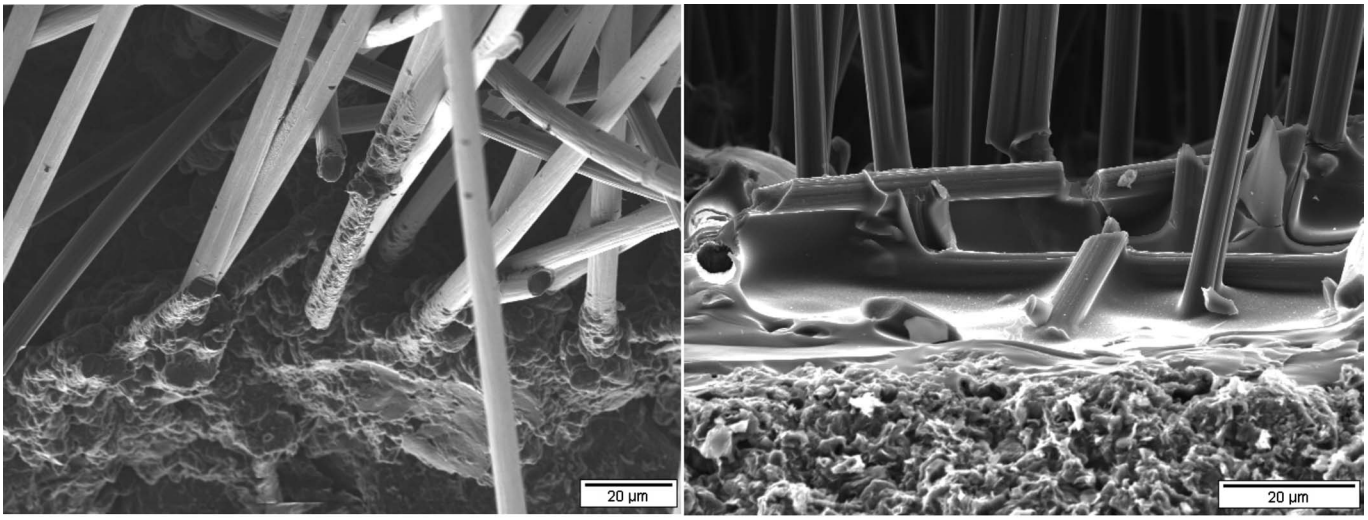

FIG. 4. SEM images of the bond line with (a) showing a cathode with no shots and (b) showing a cathode at 200 000 shots. Note the difference in the texture and appearance of the surface.

After 220 000 shots, there are distinct changes in the Raman spectra near the tips of the fibers [see Fig. 5(b)]. Within $5-7$ μm from the fiber tips, the Raman spectra remain essentially the same as that of the fibers with no shots. However, as one progresses nearer to the fiber tip, the *D* and *G* peaks become narrower and separate. The peak positions also shift to higher wave numbers. The narrowing of the *D* and *G* peaks along with the emergence of more distinct second order peaks indicate that the carbon atoms rearrange into a more ordered "graphitelike" structure with the largest degree of ordering occurring within the first 1.5 μm of the fiber tips. The shift to higher wave numbers indicates a change in the stress/strain in the fiber. The source of this change cannot be conclusively stated without further study. However, the change in stress/strain in the fiber is consistent with our hypothesis that CsI intercalates into the carbon fiber and leaches out as the shot count increases.

The results shown in this article indicate several interesting properties of the CsI coated carbon emission process. First, the SEM images show no evidence of explosive field emission, a fact consistent with previous experimental results[6,7] and with modeling and simulation efforts to be presented in future publications.[8] (Overheated or exploded fiber tips appear to have mushroomlike structure that will be discussed in Ref. 8.) Thus, we posit that the emission consists of field emission without explosive processes that generates sufficient electrons for space charge limited flow.[9] Second, we posit that the CsI intercalates into the carbon fiber and then leaches out of the fiber as the cathode is shot. In a sense, this mechanism resembles that of barium in a dispenser cathode.[10] In fact, Booske *et al.*[11] have demonstrated using *ab initio* calculations that a thin layer, perhaps as little as a monolayer, of CsI reduces the work function of carbon from 4.8 to 2.0 eV, a significant decrease. This leaching process could well be a manifestation of this effect.

Next, we note that both the cathode substrate at the bond line and the cathode tips, as evidenced by Raman spectroscopy, undergo significant structural changes as the cathode evolves in its lifetime. Due to constraints of the Raman spectroscopy system and the cathode test apparatus, one cannot look at the Raman spectrum of the bond line during the cathode shot history. However, SEM images indicate significant change in the carbon at this point, with the Raman spectrum showing significant changes at the cathode tips. While the source of these changes may be due to different mechanisms, we note that several sources for this variation exist. First, the changes could be due to heating of the bond line and the fiber tips. This mechanism will be explored both experimentally with single fiber cathodes and computationally with magneto-hydrodynamic codes. Second, the diode geometry shown here suffers from significant ion back bombardment, typically in the form of protons, leading to full bipolar flow in the diode. The impact of these protons could also lead to the observed changes. This mechanism can also be explored by direct proton bombardment of a substrate. Finally, the observed changes in the Raman spectrum as one moves along the carbon fiber could stem from heating, proton bombardment, and the leaching of the CsI from the fiber during the shot history. The fibers clearly show a transition from a more disordered to a more graphitic state. Finally, further research clearly dictates the use of transmission electron microscopy (TEM) in order to determine whether our hypothesis of CsI intercalation is correct. In this measurement, one would hope to actually see the rows of Cs and I atoms interspersed with the C atoms. This feature should be clearly obvious, since Cs and I are much larger than C and the regions where only CsI and only C atoms reside should be obvious as well. Further, we could determine the thickness and arrangement of the interfacial area (where Cs, I, and

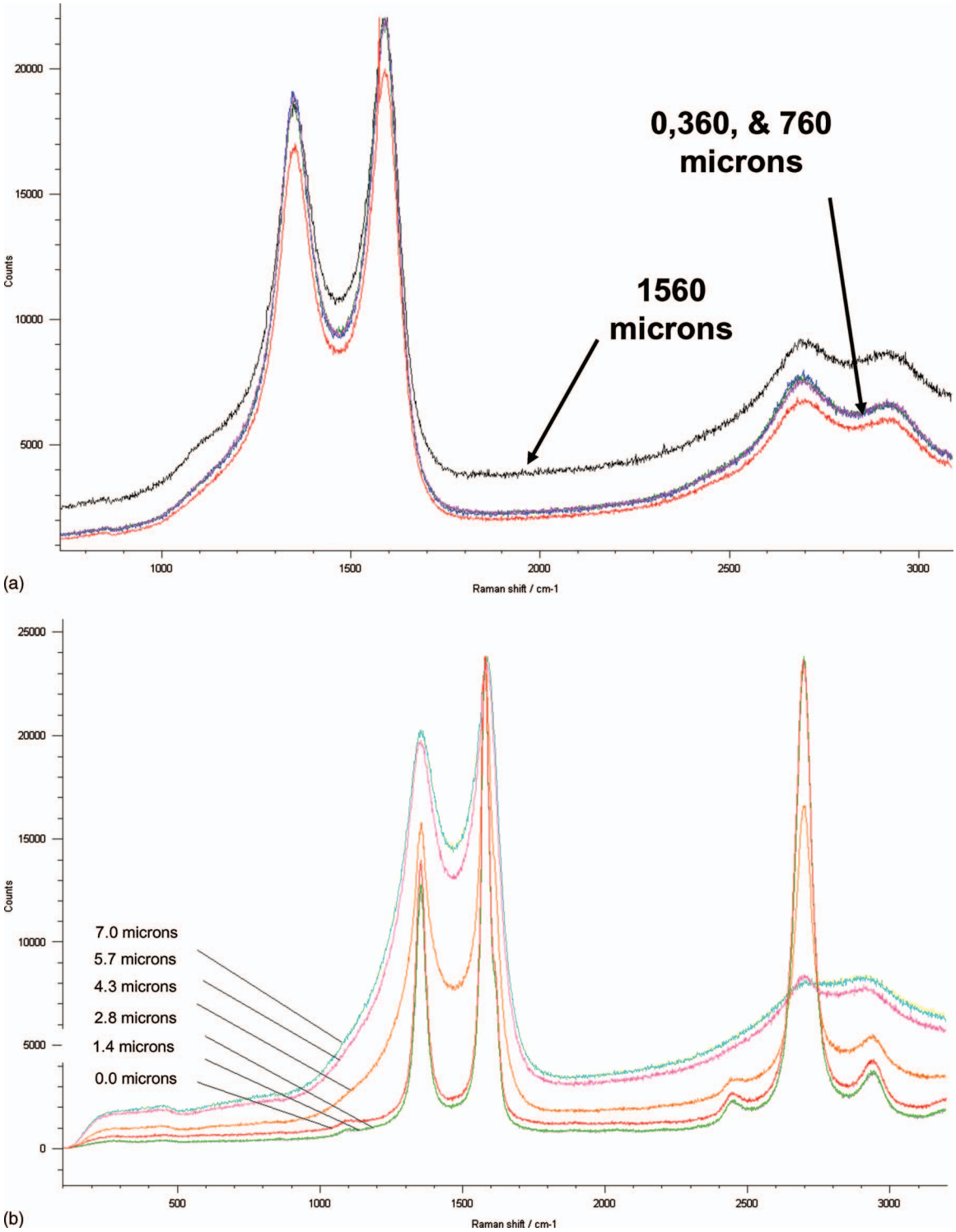

FIG. 5. (Color) Raman spectra. (a) Uncoated fibers with no CsI coating. (b) Coated fiber with CsI coating at 200 000 shots.

C are all present), leading to a better understanding of the emission process. However, these measurements remain beyond the scope of this article.

Finally, we note that the observed changes in the cathode surface morphology appear to have no impact upon cathode performance given the number of shots to which the cathode has been operated. We have not operated significantly past the 1 000 000 mark due to the time constraints of the low repetition rate operation. Nevertheless, given this limit, we can state the following. *After conditioning*, the cathode operates stably and reproducibly for the entire life history (1 000 000 shots at this time). The only changes evident consist of the physical changes noted in this article. One must accept these statements with some degree of caution, however. The observed changes could well signal the onset of failure modes for this technology, if allowed to proceed further. For example, the leaching of CsI could well lead to loss of CsI, causing degradation of the cathode. Further, the struc-

tural changes in the carbon fiber could have similar effects. Unfortunately, until data exist to dictate otherwise, we can only speculate about the effects of these changes.

IV. SUMMARY

These results, taken as a whole, give some indications about the electron emission process for this class of materials. The emission apparently depends sensitively upon the coating process. The modeling work by Booske appears to confirm this basic result, with additional facts awaiting determination. Further, the leaching of the CsI must play some role in the emission, the nature of which has not yet been determined. Finally, the lack of exploded fibers remains remarkable and gives further credence to the low out-gassing nature of these cathodes. The attainment of space charge limited flow under these conditions also remains noteworthy.

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[1]D. Shiffler, M. LaCour, M. Ruebush, K. Golby, D. Zagar, M. Haworth, T. Knowles, and R. Umstattd, Rev. Sci. Instrum. **73**, 4358 (2002).
[2]D. Shiffler, U.S. Patent No. 6,683,399 (27 January 2004).
[3]R. B. Miller, J. Appl. Phys. **84**, 3880 (1998).
[4]D. D. Hinshelwood, IEEE Trans. Plasma Sci. **PS-11**, 188 (1983).
[5]J. Nation, IEEE Trans. Plasma Sci. **87**, 185 (1999).
[6]D. Shiffler, J. Heggemeier, M. LaCour, K. Golby, and M. Ruebush, Phys. Plasmas **11**, 1680 (2004).
[7]D. Shiffler, J. Luginsland, R. Umstattd, M. LaCour, K. Golby, M. Haworth, M. Ruebush, D. Zagar, A. Gibbs, and T. Spencer, IEEE Special Issue on High Power Microwaves, June 2002.
[8]J. Luginsland, D. Shiffler, Y. Y. Lau, S. Heidger, and P. Adamson, "Modeling of cathode tip failure," Phys. Plasmas, Nov. 2007.
[9]I. Langmuir, Phys. Rev. **2**, 250 (1913).
[10]R. T. Longo, J. Appl. Phys. **94**, 6966 (2003).
[11]J. Booske, private communication, June 2007.