## Operation of a gated field emitter using an individual carbon nanofiber cathode

M. A. Guillorn,<sup>a)</sup> A. V. Melechko, V. I. Merkulov, E. D. Ellis, C. L. Britton, M. L. Simpson, and D. H. Lowndes

Molecular-Scale Engineering and Nanoscale Technologies Research Group, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831 and University of Tennessee, Knoxville, Tennessee 37996

L. R. Baylor

Fusion Energy Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831

(Received 18 June 2001; accepted for publication 10 September 2001)

We report on the operation of an integrated gated cathode device using a single vertically aligned carbon nanofiber as the field emission element. This device is capable of operation in a moderate vacuum for extended periods of time without experiencing a degradation of performance. Less than 1% of the total emitted current is collected by the gate electrode, indicating that the emitted electron beam is highly collimated. As a consequence, this device is ideal for applications that require well-focused electron emission from a microscale structure. [DOI: 10.1063/1.1419038]

Field emission (FE) of electrons from nanostructured graphitic carbon-based materials including single<sup>1-3</sup> and multiwalled<sup>4,5</sup> carbon nanotubes (CNT) and carbon nanofibers<sup>6,7</sup> (CNF) has been an area of intense investigation in recent years. This body of research indicates that these materials have several advantages over other candidate materials for FE applications, namely very low threshold voltages,  $V_{\rm th}$ , for the initiation of electron emission and extraordinary environmental stability.<sup>8</sup> Most of the work in this field has focused on measurements of the FE properties of these materials deposited or grown onto a variety of flat substrates using a vast array of different deposition and measurement techniques. However, very little work has been presented on integrated gated cathode structures using these materials as FE elements. Xu and Brandes<sup>9</sup> presented an operating CNTbased gated cathode device in 1998 employing disordered mats of multiwalled CNTs (MWNT) grown within electrostatic gating structures by thermal chemical vapor deposition (CVD). Wang et al.<sup>10</sup> reported the operation of a similar device fabricated by a technique using a paste of CNT material and conductive epoxy deposited into microfabricated well structures. Lee et al.<sup>11</sup> also have recently reported on the operation of gated cathode structures similar to those demonstrated by Xu and Brandes<sup>9</sup> with minor improvements in the structure fabrication process and increased control of the in situ MWNT growth step.

The CNT material in the aforementioned gated cathode structures is likely to contain numerous FE sites; there are multiple CNT tips in each cathode and there is evidence that FE can occur from sites located along the walls.<sup>11</sup> While all of these devices possess operating characteristics desirable in any FE device (i.e., low  $V_{\text{th}}$  and high FE currents) these fabrication processes offer no way to precisely control the location or density of the emission sites within the device. These factors complicate the construction of FE devices that

produce a well-focused electron beam as required in applications such as electron microscopy or electron-beam lithography (EBL).

Recently, we reported a technique for fabricating gated cathode structures that uses a single *in situ* grown vertically aligned CNF (VACNF) as a FE element.<sup>12</sup> This technique offers a way to produce gated cathode structures that take full advantage of the FE properties inherent to nanostructured graphitic carbon materials, while providing a deterministic way to control the point of emission. In this letter, we present the operation of a gated cathode structure fabricated using a variation of the previously reported process.<sup>12</sup>

A diagram of the fabrication process is shown in Fig. 1. The details of this process are discussed in another publication.<sup>13</sup> Briefly, EBL was used to define catalyst sites for deterministic growth of VACNFs and alignment marks for subsequent lithography steps on low resistivity *n*-type Si substrates [Fig. 1(a)]. Individual VACNFs were grown using dc plasma enhanced CVD (PECVD) as we have described in earlier work<sup>14</sup> [Fig. 1(b)]. The VACNFs produced for this work were conical in shape possessing a base diameter of ~200 nm, a height of ~1  $\mu$ m, and a tip radius of curvature of ~15 nm. A 2.5  $\mu$ m thick conformal layer of SiO<sub>2</sub> was then deposited onto the substrates using a silane-based rf PECVD process [Fig. 1(c)].



FIG. 1. Process flow for the fabrication of the VACNF-based gated cathode structures.

3506

Downloaded 15 Nov 2001 to 128.219.65.64. Redistribution subject to AIP license or copyright, see http://ojps.aip.org/aplo/aplcr.jsp

<sup>&</sup>lt;sup>a)</sup>Author to whom all correspondence should be addressed; electronic mail: guillornma@ornl.gov



FIG. 2. (a) Scanning electron micrograph of a completed gated cathode structure taken at a  $35^{\circ}$  angle from normal incidence and (b) schematic of the FE measurement setup.

In order to perform further processing on these substrates, chemical mechanical polishing was used to planarize the structures [Fig. 1(d)]. The SiO<sub>2</sub> layer was polished until all gross surface nonuniformities were removed, producing a final oxide thickness of approximately 1.5  $\mu$ m. A 50 nm thick layer of Mo was then deposited onto the substrates using electron gun physical vapor deposition [Fig. 1 (e)]. Photoresist was applied to the substrates and the gate electrode pattern was exposed [Fig. 1(f)]. This pattern consisted of macroscopic bonding pads connected to 10  $\mu$ m square electrodes with 2  $\mu$ m diameter apertures aligned to the VACNF buried beneath the SiO<sub>2</sub> layer. Using the resist as an etch mask, the substrates were subjected to a CF4/O2 reactive ion etch to pattern the Mo gate layer followed by a  $CHF_3/O_2$  SiO<sub>2</sub> etch [Fig. 1(g)] to release the buried VACNF. We have found that this process inflicts minimal structural damage on the buried VACNF structures while improving their FE properties.<sup>7,13</sup> The structures were dipped into a dilute HF solution (10:1, ammonium fluoride:HF) for 1 min to create undercut in the well sidewall profile [Fig. 1(h)] to help prevent charging of the well sidewalls during device operation. Finally, the photoresist was removed in acetone [Fig. 1(i)], completing the structure. A micrograph of a finished device taken at a 35° angle from normal incidence is shown in Fig. 2(a)

FE measurements were carried out in a chamber evacuated to a pressure of  $10^{-6}$  Torr producing a test environment similar to potential, less than ideal, operating environments. A flat Cu anode was placed 700  $\mu$ m directly above the Si substrate containing the VACNF-based FE device. A Hewlett Packard 4156A Precision Semiconductor Parameter Analyzer containing four dc source measure units (SMU) was connected to the structure and anode as shown in Fig. 2 (b). All of the data presented in this work was obtained with the gate at ground potential, and a 100 V positive bias on the anode. Anode and gate currents were measured as the cathode potential was varied between ground and -100 V.

An initial FE current–voltage (I-V) curve is show in Fig. 3, displaying a  $V_{\text{th}}$  80 V.<sup>15</sup> The apparent lack of a monotonic increase in the anode current with an increasing gate– cathode bias after FE has been initiated can possibly be attributed to the initial condition of the tip. The structure was not cleaned in any way prior to operation, ensuring the presence of an adsorbate layer on the VACNF. Constant bias measurements of the emission current were then conducted using a cathode bias of -90 V. The device was operated with and without a ballast resistance of 22 M $\Omega$  in series with the cathode and SMU 3 (60 s time intervals are shown in Figs. 4(b) and 4(a) respectively). These tests were conducted con-



FIG. 3. Initial FE I-V curves for a 2  $\mu$ m aperture structure showing the measured anode and gate current.

tinuously for 10 min each. In both of these tests, the average gate current remained below 3 nA, while the anode current averaged above 400 nA with the ballast resistor and 700 nA without it. This shows that less than 1% of the emitted current was collected by the gate during device operation. This result is in stark contrast to the device presented by Wang where more than 30% of the total anode current is seen at the gate<sup>10</sup> and indicates that the emitted current from the VACNF cathode is coming from a well-focused point source, presumably the fiber tip. A final FE I-V curve with the ballast resistor in place is shown in Fig. 5. This data was taken after the 20 min of constant bias testing and indicates a reduction in  $V_{\rm th}$  to approximately 65 V; this may be attributed to emitter conditioning during FE operation. The FE I-V data fit



FIG. 4. Constant bias time plots of FE current from a 2  $\mu$ m aperture well structure with a cathode bias of -90 V (a) without a ballast resistor and (b) with a 22 M $\Omega$  ballast resistor in series between the cathode and SMU 3.

Downloaded 15 Nov 2001 to 128.219.65.64. Redistribution subject to AIP license or copyright, see http://ojps.aip.org/aplo/aplcr.jsp



FIG. 5. FE I-V curve taken with a 22 M $\Omega$  ballast resistor in series between the cathode and SMU 3 following 20 min of constant bias operation. Inset: Fowler–Nordheim plot of the FE I-V data.

well to a Fowler–Nordheim plot and is shown inset in Fig. 5. Postemission characterization of the VACNF tip was conducted using scanning electron microscopy and revealed no significant morphological changes.

In this letter, we have demonstrated the operation of a gated field emitter using a single VACNF as the FE cathode. This device displays operating characteristics inherent to other nanostructured graphitic carbon-based FE devices: it is capable of operation in less than ideal environments and can achieve significant operating currents for extended periods of time without causing degradation to the VACNF tip. Although  $V_{\rm th}$  may appear somewhat high for these devices when compared to similar CNT-based devices, it is in reasonable agreement with the values expected from the VACNF aspect ratio<sup>7,13</sup> and the geometry of the well. By decreasing the diameter of the gate aperture and increasing the aspect ratio of the VACNF, it should be possible to lower the operating voltage of these devices into a regime comparable to the CNT-based devices that have been reported previously. The low percentage of the emitted current measured at the gate electrode indicates that the emitted electron beam is highly collimated. These promising results show that VACNF-based FE devices are ideal for cold cathode FE applications that require well-focused electron emission from a microscale structure.

The authors wish to thank P. H. Flemming, G. J. Bordonaro, and D. Woodie for their assistance in sample preparation. This work was funded by DARPA under Contract No. 1868HH26X1 and by the Laboratory Directed Research and Development Program of the Oak Ridge National Laboratory (ORNL). ORNL is managed by UT-Battelle, LLC, for the U.S. Department of Energy under Contract No. DE-AC05-00OR22725. This work was performed in part at the Cornell Nanofabrication Facility (a member of the National Nanofabrication Users Network) that is supported by the National Science Foundation under Grant No. ECS-9731293, its users, Cornell University, and industrial affiliates.

- <sup>1</sup>J. M. Bonard, J. P. Salvetat, T. Stochli, W. A. de Heer, L. Forro, and A. Chatelain, Appl. Phys. Lett. **73**, 918 (1998).
- <sup>2</sup>Y. Saito, K. Jamaguchi, T. Nishino, K. Hata, K. Tohji, A. Kasuya, and Y. Nishina, Jpn. J. Appl. Phys., Part 2 36, L1340 (1997).
- <sup>3</sup>K. Matsumoto, S. Kinosita, Y. Gotoh, T. Uchiyama, S. Manalis, and C. Quate, Appl. Phys. Lett. **78**, 539 (2001).
- <sup>4</sup>A. G. Rinzler, J. H. Hafner, P. Nokolaev, L. Lou, S. G. Kim, D. Tomaneck, P. Nodlander, D. T. Colbert, and R. E. Smalley, Science **269**, 1550 (1995).
- <sup>5</sup>X. Xu and G. R. Brandes, Appl. Phys. Lett. **74**, 2549 (1999).
- <sup>6</sup>V. I. Merkulov, D. H. Lowndes, L. R. Baylor, J. Appl. Phys. 89, 1933 (2001).
- <sup>7</sup>L. R. Baylor, V. I. Merkulov, E. D. Ellis, M. A. Guillorn, D. H. Lowndes,
- M. L. Simpson, and J. H. Whealton (unpublished).
- <sup>8</sup>K. A. Dean and B. R. Chalmala, Appl. Phys. Lett. **75**, 3017 (1999).
- <sup>9</sup>X. Xu and G. R. Brandes, Mater. Res. Soc. Symp. Proc. 59, 107 (1998).
- <sup>10</sup>Q. H. Wang, M. Yan, and R. P. H. Chang, Appl. Phys. Lett. **78**, 1294 (2001).
- <sup>11</sup> Y. H. Lee, Y. T. Jang, D. H. Kim, J. H. Ahn, and B. K. Ju, Adv. Mater. 13, 479 (2001).
- <sup>12</sup>M. A. Guillorn, M. L. Simpson, G. J. Bordonaro, V. I. Merkulov, L. R. Baylor, and D. H. Lowndes, J. Vac. Sci. Technol. B **19**, 573 (2001).
- <sup>13</sup> M. A. Guillorn, E. D. Ellis, M. L. Simpson, A. V. Melechko, V. I. Merkulov, G. J. Bordonaro, L. R. Baylor, and D. H. Lowndes, Proceedings of the 45th International Conference on Electron, Ion and Photon Beam Technology and Nanofabrication (EIPBN), Washington D.C. (2001) (in press).
- <sup>14</sup> V. I. Merkulov, D. H. Lowndes, Y. Y. Wei, G. Eres, and E. Voekl, Appl. Phys. Lett. **76**, 3555 (2000).
- <sup>15</sup>Defined in this work as the gate-to-cathode bias voltage required to extract 10 nA of FE current.