Carbon nanotubes as electron source in an x-ray tube

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Field emitters comprised of aligned carbon nanotubes are shown to be promising as a primary electron source in an x-ray tube working in a nonultrahigh vacuum ambience. At a pressure of 2×10^{-7} Torr, the nanotube emitters continue to emit electrons for more than 1 h, and yield better resolved x-ray images than do thermionic emitters, independently of whether the sample is biological or nonbiological. The near-uniformity in energy distribution of electrons emitted from carbon nanotubes might be related to the improved image quality in the field-emission mode.
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Since its theoretical formulation by Richardson,[1] thermionic emission (TE) has been a key concept in electron-beam techniques. In thermionic electron emission, the solid electron source (i.e., the cathode) is heated above 2000 °C to allow free electrons to escape from the surface. The greatest advantage of this so-called ''hot cathode,'' usually a heated tungsten (W) filament, is that it works even in nonultrahigh vacuum (non-UHV) ambiences, which contain vast numbers of gaseous molecules. Underlying this is the fact that the gas-impinging rate is inversely proportional to the square root of surface temperature.[2] However, hot cathodes are prone to chemically react with residual water and oxygen to produce tungsten oxides, and get thinner and thinner over a long duration through the sublimation of the oxides. In addition, hot cathodes require a power supply for heating, thus making it difficult to construct a compact electron-beam tool.

It was suggested in the mid-1950s that these disadvantages of hot cathodes may be overcome by replacing them with field emission (FE), or cold, cathodes.[3] Unfortunately, the electron emission from a FE cathode is exponentially affected by the chemical and morphological states of the electron-emitting area,[4] resulting in instability of emitted currents in non-UHV ambiences. This is particularly true of metallic cathodes, which strongly interact with residual gaseous molecules. Thus, no attempt to use field emitters in non-UHV has been of practical significance.

Chemically, carbon is far more stable, and hence more robust in non-UHV, than metals. Indeed, several groups have claimed that carbon nanotubes (CNTs)[5] are promising as multiple field electron sources.[6–10] Most studies of this kind are directed to the application of CNTs to flat-panel displays, but in our view they are still far from practical.

Presumably, the most important application of x rays is x-ray radiography (XR).[11] The traditional way to generate x rays is to bombard a metal target with accelerated thermionic electrons. In recent years, the demand is increasing in high-tech communities for compact, portable x-ray tubes that can be set up in a narrow space, e.g., between the fan blades of jet engines. What meets this demand would be a FE x-ray tube, and its construction would be possible with the aid of CNTs. Here we report our first step toward a practical x-ray tube equipped with a CNT field emitter. We also demonstrate that FE, compared with TE, provides a better-resolved x-ray image for both biological and nonbiological samples.

We recently developed a technique to align CNTs on a cobalt-coated W wire.[12] These CNTs are topped with a metallic crystallite, amounting to $\sim 6\times10^{7}\ \mathrm{mm}^{-2}$ in site density. The metallic crystallites are not bare but covered with a thin graphite layer, ensuring that the tips of the respective CNTs will function as a graphitic electron source.[12]

We modified a nonbankable metal vacuum chamber, which was previously used for a different purpose, into a simple x-ray tube [Fig. 1(a)]. The tube was pumped down to $\sim 2\times10^{-7}$ Torr with the aid of two turbo-molecular pumps. The electron current emitted from the cathode (CNTs) was controlled by manually varying the potential of the counter electrode placed just before the cathode. By replacing the CNTs with a mere W wire (0.3 mm diameter), the tube could also be operated in the TE mode. For some CNT emitters, the intensity of x rays passing through the beryllium (Be) window was measured as a function of target potential, with total electron current as the parameter. Very roughly, the x-ray intensity increased exponentially with the linear increase in target potential [Fig. 1(b)].

Since a major industrial application of XR is the nondestructive inspection of electronic device, we chose a large-scale integrated (LSI) circuit as the first sample. Its x-ray image recorded with a CNT emitter was so sharp as to disclose every gold wire ($\sim 30\ \mu$m across) for electrical conduction [Fig. 2(a)]. Because of a low electron current (1.5 μA), the exposure time was as long as 14 min, but the second to fourth imaging could be done by this emitter with no appreciable deterioration in image resolution.

The above LSI was also x-ray imaged in the TE mode, and the result is shown in Fig. 2(b). Compare these x-ray images, and the superiority of the FE mode becomes clear. For example, the arrowhead-indicated lead wires in Fig. 2(a) are missing or barely perceptible in Fig. 2(b). The vast majority of field electrons are emitted through a tunneling process from the Fermi level, which is a function of electronic

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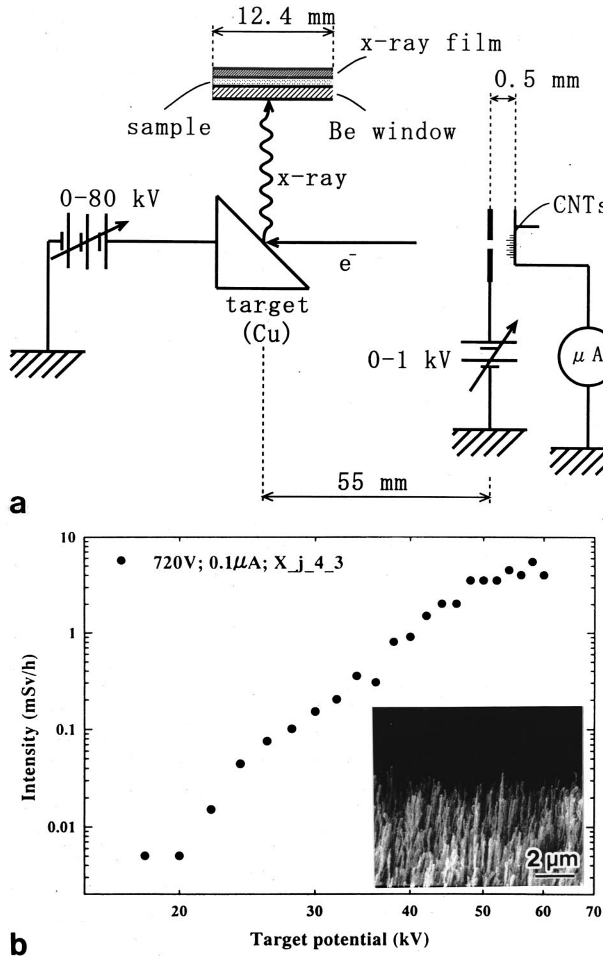

FIG. 1. (a) Diagram of the x-ray producing circuit. The electron beam drawn out from CNTs was not focused because of the preliminary nature of the experiments. (b) X-ray intensity vs target potential curve at an electron current of 0.1 μA, plotted with the aid of a Geiger–Müller tube placed just outside the Be window. Inset in (b) shows the CNTs used in this measurement, imaged by scanning electron microscopy (SEM). Emitter-operating pressure: 2×10^{-7} Torr.

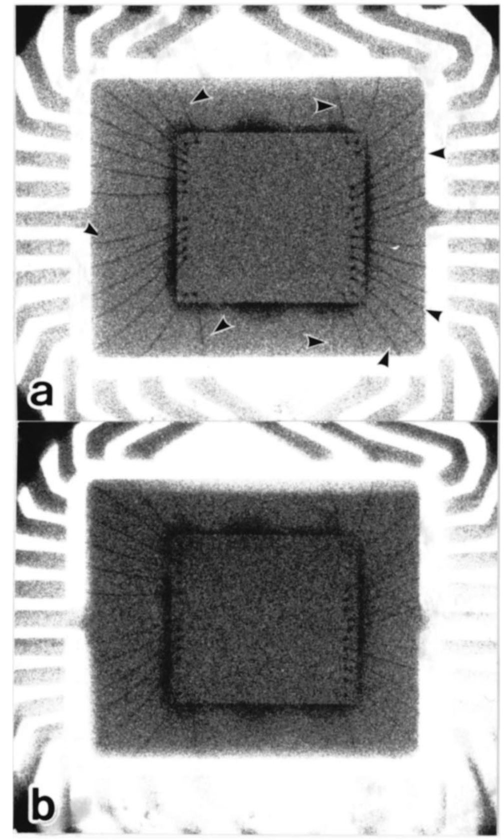

FIG. 2. X-ray images of a LSI circuit recorded in the FE, (a), and TE, (b), modes, respectively. The gold lead wires ($\sim 30\ \mu$m in diameter) indicated by the arrowheads in (a) are not well recognized in (b). The imaging was done through the plastic packaging. The imaging conditions for (a) and (b) were target voltage: 60 kV, exposure time: 14 min, total x-ray dose: 0.2 mSv.

conductivity. The electronic conductivity of CNTs has been predicted to depend on their diameter, as well as their lattice helicity.[13–15] Thus, field electrons from different tubes with different diameters possess different energies, and hence cause a Fermi level distribution. For our CNT emitters, however, very few prominent tubes well uniform in field-enhancement factor actually emit electrons,[12] so the emission energy distribution should be far more uniform than for thermal electrons. This near uniformity in electron energy might be responsible for an improved image resolution in the FE mode.

For soft samples like plants, the energy of x rays must be lowered to the soft x-ray region. In terms of electron energy, the upper limit for the soft energy region is several keV; a prolonged exposure time is needed for soft x-ray imaging. Unfortunately, the lifetime of our CNT emitters is 60–80 min in 2×10^{-7} Torr, making them unable to operate in the true soft x-ray region. (A prolonged application of electric field led to a local removal of the nanotube film from the substrate, whereupon the emitter ceased to work.) This forced us to image biological samples at 10 keV. Figure 3(a) shows a leaf imaged at 10 kV, revealing leaf veins for nutrition transportation. The exposure time was around 1 h, reflecting the robust nature of CNTs in non-UHV. This kind of observation at low energies is completely impossible with metallic cathodes, due to their limited lifetime in non-UHV. It is emphasized that this leaf was snatched from its tree just before imaging and hence was still not dehydrated.

Other biological samples that we examined in the FE mode included insects, each of which presented a clear x-ray image at 20–40 keV.

As noted already, the controlled operation of a field electron emitter is very difficult in a non-UHV ambience, mainly because of cathode ''sputtering,'' i.e., residual gas molecules ionized through the collision with field-emitted electrons bombard the emitter and damage it.[12] To completely eliminate this troublesome effect, the UHV is essential. At a pressure in the 10^{-7} Torr region, a single-point field emitter of tungsten, for instance, is morphologically damaged by a few minutes of operation. Although our CNT emitters are much more resistant to sputtering, they were not completely stable but fluctuated at an amplitude of $\pm10\%$ at the pressure employed [Fig. 3(b)]. Fortunately, the imaging on photoplates involves an integrated detection of x rays, so this amount of current fluctuation has no negative effect on imaging the subject as is. If an x-ray image intensifier is used, however, the exposure time will be dramatically shortened, perhaps by two orders of magnitude, and therefore the current fluctuation would make a one-shot image less reliable. To minimize

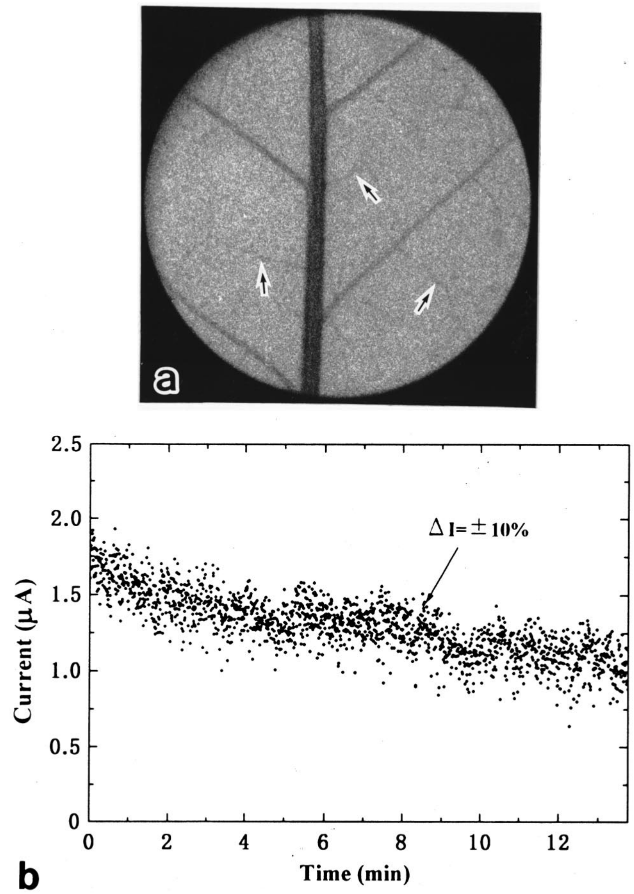

FIG. 3. (a) Fresh leaf imaged at 10 kV, and (b) variation in electron current at the early stage of the imaging. The electron current decreased gradually while fluctuating with an amplitude of around 10% at a constant counter-electrode potential. The arrows in (a) indicate capillary veins.

such current fluctuation, the electron-gun chamber would have to be differentially pumped down to UHV. Also, the electron current could be automatically stabilized in a non-UHV ambience by electronically controlling the counter-electrode potential.

The present work was stimulated by a technological demand for a non-TE x-ray tube, but it led to the unexpected finding that field electrons might improve the image resolution in XR.

A more developed version of contact XR is projection x-ray microscopy. In this technique, the shadow image of the specimen is formed on the screen by radially propagating x rays. Theoretically, the more the x-ray emitting area is reduced, the more the image resolution is improved.[16] Finely focusing the electron beam emitted from a single-point field emitter might thus produce highly resolved x-ray micrographs. Also, some industrial groups are planning to develop x-ray endoscopes, for which the use of a field electron source is indispensable. The present work may pave the way to these next generations of x-ray technology, which may be termed the ''field-emission x-ray radiography.''

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