

Study of Carbon-Nanotube Web Thermoacoustic Loud Speakers

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Received June 23, 2010; accepted August 2, 2010; published online January 20, 2011

Thermoacoustic carbon nanotube (CNT) speakers were fabricated using CNT webs spun from a multiwalled carbon nanotube (MWNT) array of 0.8–1.6 mm height. The generated sound pressure level (SPL) showed a linear relationship with frequency over a wide range from 10 Hz to 40 kHz with a slope of 6 dB/octave. In addition to this, significantly broad and flat SPLs were obtained in the ultrasonic region, ranging from 40 to 100 kHz. The distance from the speaker to the microphone was 0.5 m. The high SPL is due to the good heat radiation property of the MWNT web. Herein, we showed an acoustical property for the MWNT web thermoacoustic speaker from the viewpoint of the structural web morphology. The role of heat radiation behavior and the effects of the length of individual MWNTs are discussed. © 2011 The Japan Society of Applied Physics

1. Introduction

Since their discovery in 1991,¹⁾ carbon nanotubes (CNT) have been investigated as a key nanomaterial for diverse applications.²⁾ Their small tubular structure is responsible for their diverse features, including high mechanical strength,³⁾ good electric property,⁴⁾ good heat conductance,⁵⁾ and high electron emission,⁶⁾ which are of interest to both academic and industrial researchers.^{7,8)} Recently, their good thermal property has attracted much attention because it can radically improve the performance of devices and instruments such as high-performance computers and high-power optical components.⁹⁾ Bulk samples of well-aligned CNTs represent an interesting material with highly anisotropic electrical and thermal transport properties. An extremely high thermal conductivity of $\kappa = 6600 \text{ W}/(\text{m}\cdot\text{K})$ has been theoretically predicted for individual CNTs, which suggests potential improvement in heat management systems.^{10,11)} However, the extremely high thermal and electrical properties of CNT have not yet been achieved for macroscopic industrial applications. CNTs have been used as passive materials that provide only charge and/or heat transfer channels, such as polymer composite materials. Meanwhile, Xiao *et al.* have reported that, with its good thermal and electrical properties, a piece of a CNT thin film can be a practical magnet-free loudspeaker by simply passing an audio frequency current through it. A rapid temperature oscillation causes pressure oscillation in the air surrounding the film, and the thermally induced pressure oscillation changes to sound.¹²⁾ Because of their very small heat capacity per unit area (HCPUA), these sheets possess outstanding thermoacoustic properties and can be used in thermally driven loudspeakers. Xiao *et al.* demonstrated that their CNT loudspeakers can generate sound in a wide frequency range with a low total harmonic distortion. However, the acoustical property of CNT webs from the viewpoint of thermoacoustic phenomenon is still not well understood.

To date, we have reported a simple and easy growth method for an ultralong, vertically aligned multiwalled CNT (MWNT) array.¹³⁾ We refer to the proposed method as “chloride-mediated chemical vapor deposition” (CM-CVD). This method enables the synthesis of highly drawable

MWNT array samples. The MWNTs are easily drawn simply by pinching and pulling out from the array edge. The bundles form a continuous network, called a web, and are preferably oriented along the drawing direction.

In this work, we studied the thermoacoustic properties of CNT speakers made from MWNT webs. Hereafter, we describe the MWNT web speaker as the CNT speaker. To understand the mechanism of the thermoacoustic effect in more detail, the frequency response in a wide range of what has been investigated in this study. The effects of the length of individual MWNTs and web morphology on the acoustic output were analyzed using a standard acoustic measurement commonly used to evaluate commercial audio systems. The heat dynamics of the MWNT web surface was also measured using thermography. The MWNT web speaker output high sound pressure levels (SPLs), i.e., 100 Hz to 100 kHz, in other words, from the zone of audibility to the ultrasonic region.

2. Experimental Procedure

2.1 Synthesis of MWNT arrays and webs

A vertically aligned MWNT array was synthesized using a conventional thermal CVD system. Figure 1 schematically shows the experimental setup. A smooth quartz substrate was placed at the center of a horizontal quartz tube with iron chloride (FeCl_2) powder (99.9%) using a quartz boat. In a conventional method, a thin metallic film as the catalyst is deposited on a substrate. However, no predeposition treatment is necessary in our method. While heating, the growth chamber was maintained at a vacuum of 1×10^{-3} Torr. Once the optimal growth temperature was reached, acetylene (98%) was introduced into the furnace tube through a mass flow controller. CVD growth was carried out at a furnace temperature of 820 °C. The MWNT had lengths of up to 2.4 mm and a growth rate of $>100 \mu\text{m}/\text{min}$. The length of MWNT was controlled by adjusting growth time.

Our MWNT array sample has a highly drawable feature. A MWNT web was easily drawn using tweezers or other appropriate tools, as shown in Fig. 2(a). A MWNT web of over 60 m was obtained. Figure 2(b) shows the transition from a vertically aligned MWNT array to a horizontally aligned MWNT web. The as-drawn web is an anisotropically conductive material.

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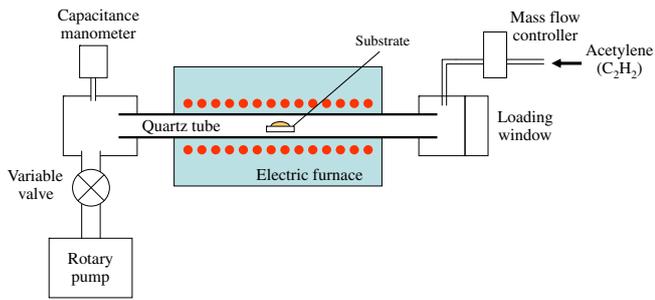


Fig. 1. (Color online) Schematic of chloride-mediated chemical vapor deposition (CM-CVD) system.

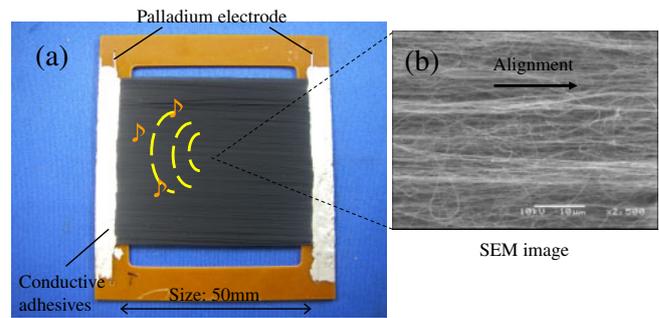


Fig. 3. (Color online) (a) Thermoacoustic carbon nanotube (CNT) speaker and (b) SEM image of aligned MWNT.

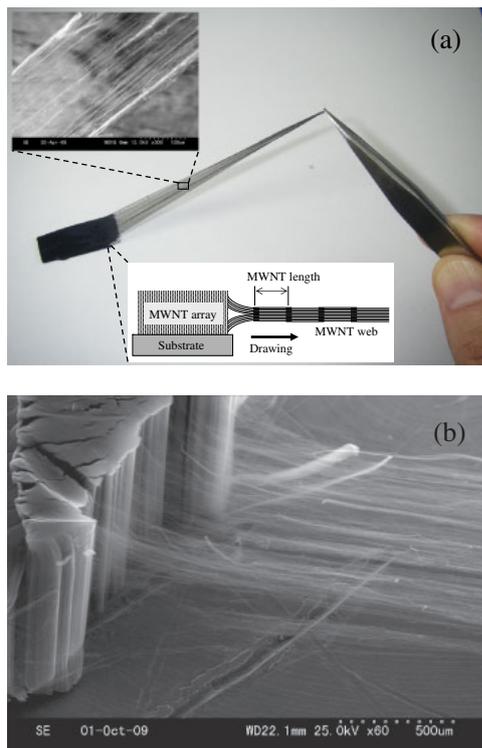


Fig. 2. (Color online) (a) Drawing of multiwalled carbon nanotube (MWNT). The inset is a drawing model of the web. (b) Scanning electron microscopy (SEM) image of MWNT web from MWNT array.

2.2 Fabrication of CNT speakers

To fabricate a CNT speaker, MWNT webs were waved between two palladium electrodes. Conductive paste was used for the electrical contacts. The speaker area was $50 \times 50 \text{ mm}^2$ as shown in Fig. 3(a). In each MWNT web, most of the MWNTs were aligned in the drawing direction, as shown in Fig. 3(b). To investigate their thermoacoustic properties, CNT speakers were fabricated as shown in Fig. 4. Since several MWNT webs were stacked to form the active area, in the drawing direction, our CNT speakers contain multi-web layers. The impedances between the electrodes were precisely adjusted to 19Ω by changing the number of layers. To study the heat dynamics of the MWNT web, three types of CNT speakers with different MWNT lengths, i.e., 0.8, 1.2, and 1.6 mm, were examined. In addition, another set, in which webs were condensed, was fabricated. The condensation process was performed as

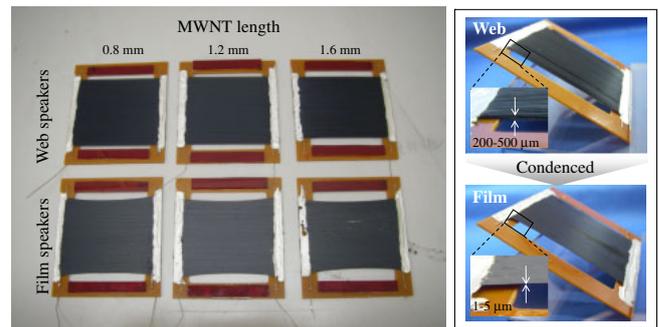


Fig. 4. (Color online) Samples of MWNT speakers with varying length of individual MWNTs and morphological differences: the web and the film.

follows: a volatile liquid (for example, ethanol) was sprayed on the MWNT webs, and the wet webs were naturally dried in air. By densification, a MWNT web was converted from the aerogel state with a thickness range of $200\text{--}500 \mu\text{m}$ to a film with a thickness range of $1\text{--}5 \mu\text{m}$. The spray and dry process caused shrinkage mainly in the thickness direction by a factor of $100\text{--}200$. The shrinkage reduced the speaker impedance from 19 to 17Ω . We called the as-waved and densified speaker the web speaker and film speaker, respectively.

2.3 Acoustic measurements and thermal emissive imaging

Acoustic measurements were conducted in an anechoic room. Figure 5(a) schematically shows the experimental setup. A sinusoidal alternating current (AC) signal was used as an input signal. The frequency response was measured by an audio analyzer (Audio Precision SYS-2712). The distance between the CNT speaker and the microphone (B&K 4138) was set to 0.5 m . These measurement configurations are the same as those for a standard acoustic evaluation system. Applying a simple AC signal with a frequency of f onto the CNT speaker generated an acoustic pressure with a frequency of $2f$. Since one cycle of the surface temperature oscillation resulted from one current cycle from zero to peak, the AC current generated a double temperature oscillation, as shown in Fig. 5(b). To avoid this modulation problem, the direct current (DC) was biased to change the AC current into a one-way current. By DC biasing, the frequency of sound changed to f .

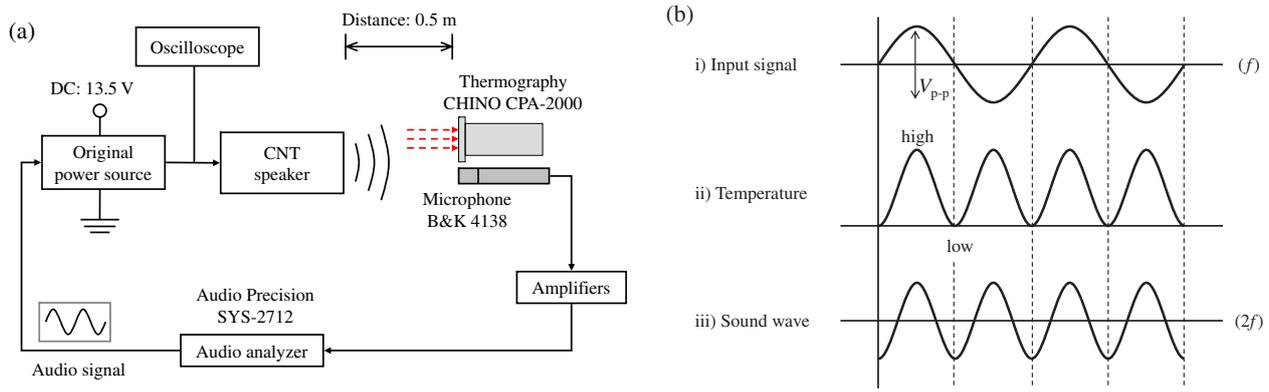


Fig. 5. (Color online) (a) Experimental setup for acoustic measurements. (b) Waveforms of signals, (i) input signal current (ii) temperature oscillation, and (iii) acoustic signal. The output frequency is two times higher than the input frequency.

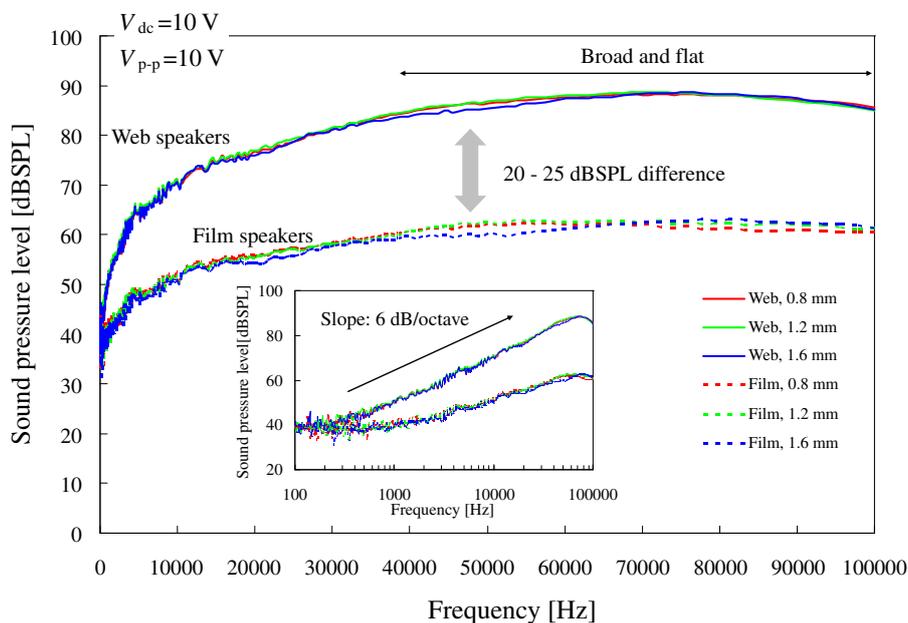


Fig. 6. (Color online) SPL vs frequency. The inset shows SPL vs logarithmic behavior of frequency. The slope of SPL in the audible region is 6 dB/octave. The frequency range shows broad and flat characteristics in the ultrasonic region, from 40 to 100 kHz.

The thermal images of the speakers were acquired using an infrared camera (CHINO CPA-2000) with a temperature resolution of 0.1 °C. Joule heating caused by a thermoacoustic effect was monitored. Every measurement was carried out after 3 min operation to ensure the steady state. The distance between the MWNT web surfaces and the infrared camera was set to 0.5 m. The emissivity of the MWNT web used here was 0.96 which is common for the carbon material.

3. Results and Discussion

Figure 6 shows the relationship between SPL and frequency for the CNT speakers. The input signal voltage, V_{p-p} , was kept constant at 10 V, including the DC biases (V_{dc} : 10 V). High and broad acoustic pressures were observed over a wide frequency range up to 100 kHz. The high-frequency limit was determined by the system sampling frequency of 192 kHz, and the spectrum tendency remained constant in a higher-frequency region. It was presumed that the sound generating performance was maintained above 100 kHz.

The slope of SPL in the audible region is 6 dB/octave (inset in Fig. 6). Since the sound pressure in a sound wave is proportional to the frequency when the volume velocity of a sound wave is constant, the constant volume velocity of a sound wave represents a constant temperature difference in the thermoacoustic effect.

It is notable that unlike resonant devices, such as piezoelectric elements, the CNT speakers show broad and flat characteristics in the ultrasonic region. In addition, the CNT speaker frequency can be made tunable in a higher ultrasonic region simply by changing the driving frequency. Meanwhile, piezoelectric devices are used at narrow resonant frequencies. These advantages would provide new features for a broad range of application fields.

It was found that the SPLs from the film speakers were extremely low compared with those of web ones. The SPL difference was 20–25 dB SPL. The SPL of the thermoacoustic speaker depends on the HCPUA.¹²⁾ From the viewpoint of the thermal source in the speaker, air surrounding MWNTs plays an important role. In the web,

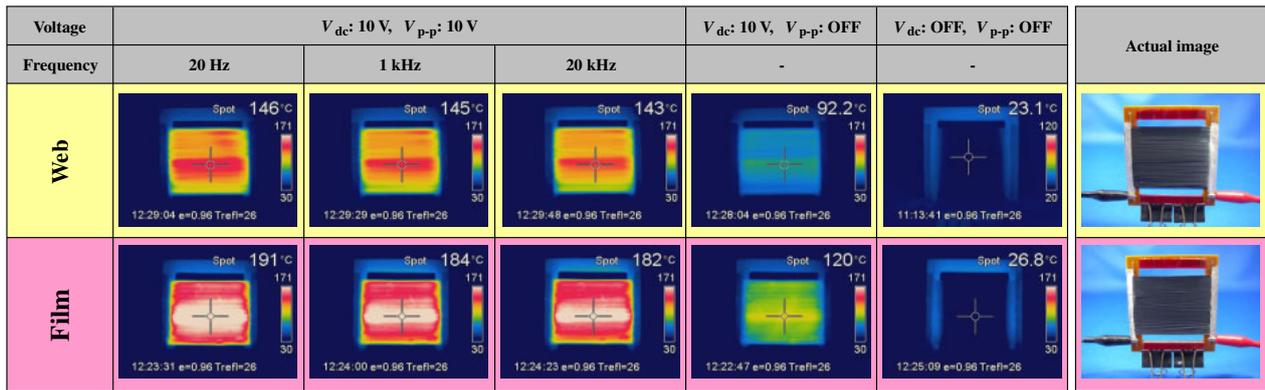


Fig. 7. (Color online) Thermal images of speaker samples in DC/AC driving mode (input AC frequency: 20 Hz, 1 kHz, and 20 kHz, respectively). The upper right of the thermal images shows the spot temperature of the cross-shaped point in the images.

MWNTs are aligned in the longitudinal direction connected by van der Waals force.^{14,15} The MWNT webs are fluffy layer materials where most of the MWNTs are isolated from each other. Therefore the heat generated at the MWNT–MWNT connections are easily quenched into air. On the other hand, in the film speakers, since MWNTs are condensed, Joule heat is likely to be accumulated inside the film. Thus, it is considered that the heat capacity of the film speaker is greater than that of the web one, resulting in lower SPLs.

To clarify the thermoacoustic properties further, thermal images were compared for the two types of speakers, as shown in Fig. 7. The thermal images show a sharp thermal contrast with a macroscopically uniform heat distribution over the speaker area. There is no frequency dependence of the surface temperature. The temperature of the web speakers is lower than that of the film speakers. Heated air diffuses from inside to outside the web. On the other hand, since MWNTs of the film speakers are rather tightly connected to each other, air thermal diffusion is suppressed and Joule heat accumulates mainly inside the film. This comparison clearly indicates that the film speaker structure enhances heat accumulation, as we expected. Furthermore, this thermal image also reveals that a high operation temperature does not necessarily imply a high SPL generation. Because of the heat insulation structure of the present film speaker, heat oscillation is reduced and then SPL markedly decreases.

It is interesting to note that SPL frequency characteristics are quite identical among the two types of speakers, independent of MWNT length. From this result, it is suggested that the sound is generated not in entire MWNT but mainly at MWNT–MWNT interconnections. The present results suggest that the CNT web speaker is regarded as a nanosized sound source ensemble.

4. Conclusions

High-performance thermoacoustic CNT speakers were demonstrated. The MWNT speakers were fabricated using long MWNT webs. A strong acoustic output was observed in a wide frequency range from 100 Hz to 100 kHz. SPLs were extremely broad and flat in the ultrasonic region from 40 to 100 kHz. It was found that high heat-diffusion structures such as web speakers are advantageous for thermoacoustic devices, especially in the ultrasonic region. The present study demonstrates the potential of MWNT webs for MWNT-based industrial applications.

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