

## Monolithic Integration of Micro-Capacitors by Lithographically Patternable, Wafer-Scale Single-Walled Carbon Nanotube Film

Kazufumi Kobashi, Karolina U. Laszczyk, Atsuko Sekiguchi, Fumiaki Tanaka, Chandramouli Subramaniam, Don, N. Futaba, Takeo Yamada and Kenji Hata

Nanotube Research Center, National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba Central 5, 1-1-1 Higashi, Tsukuba, Ibaraki 305-8565, JAPAN

Micro-supercapacitors have recently attracted much attention to develop self-powered systems from energy harvesters toward hand-held smart devices.<sup>1</sup>

Conventionally plural capacitors must be connected in a stacked manner to utilize high power delivery and high energy storage. However such a bulky system is not suitable to meet the demand for slimmness in electronic devices. We report first monolithic integrated, in micro-scale, plural and compactness connection of capacitors, in-series or in-parallel, that, depending on the connection style, enables to reach different current and voltage level.

Lithographically processible carbon nanotube (CNT) wafers have been recently used to fabricate microelectromechanical devices.<sup>2,3</sup> Based on the microfabrication technology of CNT film, in this study the monolithic integration of CNT micro-capacitors was attempted.

Monolithic integrated micro-capacitors were fabricated using lithographically patternable, large area (4-inch wafer size) CNT film. The flat, structurally homogeneous CNT film enabled the use of lithography for patterning into integrated micro-capacitors. In series or in parallel connection of CNT micro-capacitors were successfully achieved in the in-plane direction, showing the tuned voltage and current. (Fig. 1)

Vertically aligned, single-walled carbon nanotube (SWNT) forest (hundreds of  $\mu\text{m}$  in height, average SWNT diameter: 3nm and carbon purity above 99.9%) was synthesized by Super-Growth method.<sup>4</sup> These SWNTs can be transformed into electrode sheets without any binders due to one-dimensional fibrous material.<sup>5,6</sup> We utilized the SWNTs with high specific surface area ( $> 1000 \text{ m}^2/\text{g}$ )<sup>5-7</sup> as a promising electrode material for electrochemical capacitors.

To fabricate the electrode sheet by lithographically patternable, large area CNT films, CNTs were first dispersed in water with surfactant at a concentration of 0.3 wt% by ultrasonic homogenizer. The dispersion process afforded high CNT loading which led to a facile thickness control for CNT film over a large area. Secondly, the CNT dispersion was sprayed to form CNT film with thickness up to 30  $\mu\text{m}$  on Si substrate with metal current collector and electrical paths. The CNT film was processed into interdigitated pattern (700 $\times$ 900  $\mu\text{m}$ ) by reactive ion etching. An electrolyte of ionic liquid was filled in each interdigital CNT electrodes by nano-litter scale droplets from micro inkjet machine. The precise application of electrolyte fulfilled isolation of each CNT electrode. Consequently, each electrode connected in series or in parallel was insulated from each other. Furthermore these CNT electrodes were sealed by parylene coating.

To characterize the performance of the CNT micro-capacitors, firstly the single capacitor was tested by the electrochemical measurements in a dry Ar environment.

The results showed the power density and energy density comparable to those on a macro scale.<sup>5,6</sup> On the basis of the performance of single capacitor, the monolithic, integrated capacitors were analyzed. Regarding in series connection of 2 micro-capacitors, the operational voltage attained at least 5 V beyond the voltage which the single capacitor can provide. Additionally in parallel connected 2 micro-capacitors clarified that the operational current for charging and discharging is twice as high as that of the single capacitor (Fig. 1).

These fundamental studies have been done to realize a very high power delivery at hundreds of volt from integrated CNT micro-capacitor circuit. Such a monolithic, integrated micro-capacitors would be feasible and useful for the energy applications.

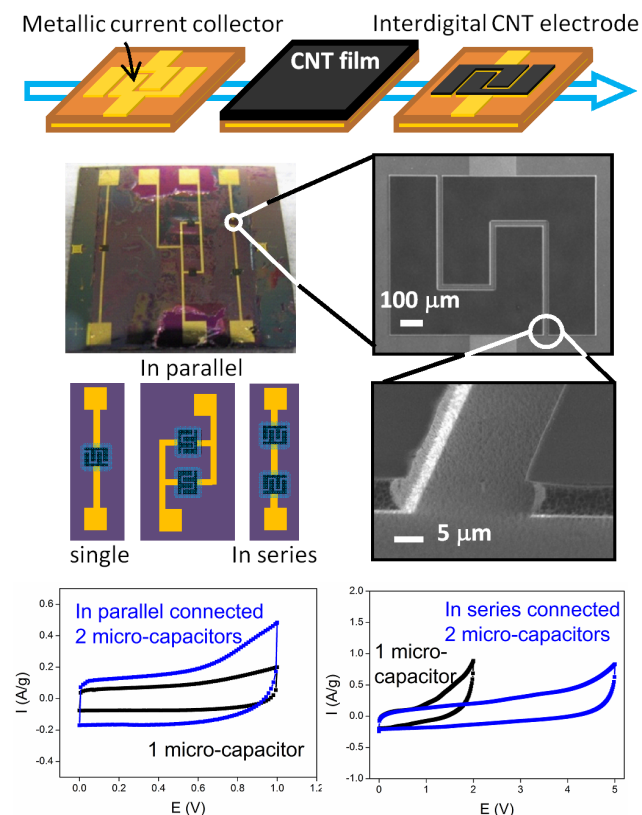


Fig. 1 Monolithic integration of CNT micro-capacitors.

### ACKNOWLEDGMENTS

Support by Core Research for Evolutional Science and Technology (CREST) from Japan Science and Technology Agency (JST) is acknowledged.

### REFERENCES

1. J. Chmiola, C. Largeot, P.-L. Taberna, P. Simon and Y. Gogotsi, *Science*, 2008, 328, 480-483.
2. Y. Hayamizu et al. *Nature Nanotech.* 2008, 3, 289-294.
3. T. Yamada et al. *Nano Lett.* 2012, 12, 4540-4545.
4. K. Hata, D. N. Futaba, K. Mizuno, T. Namai, M. Yumura and S. Iijima, *Science*, 2004, **306**, 1362-1364.
5. T. Hiraoka, A. Izadi-Najafabadi, T. Yamada, D. N. Futaba, S. Yasuda, O. Tanaike, H. Hatori, M. Yumura, S. Iijima and K. Hata, *Adv. Func. Mater.*, 2009, **19**, 1-7.
6. A. Izadi-Najafabadi, S. Yasuda, K. Kobashi, T. Yamada, D. N. Futaba, H. Hatori, M. Yumura, S. Iijima and K. Hata, *Adv. Mater.* 2010, 22, E235-E241.
7. D. N. Futaba, K. Hata, T. Yamada, T. Hiraoka, Y. Hayamizu, Y. Kakudate, O. Tanaike, H. Hatori, M. Yumura and S. Iijima, *Nat. Mater.*, 2006, **5**, 987-994.