## High-strength carbon nanotube buckypaper composite as applied to free-standing electrode for supercapacitors

Supporting information: SEM images of BP-PPy-CE infiltrated with CE solution of different concentrations, contact angel and Electrochemical Impedance Spectroscopy

Jianfei Che<sup>1,2</sup>, Peng Chen<sup>1</sup>, Mary B. Chan-Park<sup>1\*</sup>

 <sup>1</sup>School of Chemical and Biomedical Engineering, Nanyang Technological University, 62 Nanyang Drive, Singapore 637459, Singapore
<sup>2</sup>Key Laboratory of Soft Chemistry and Functional Materials, Ministry of Education, Nanjing University of Science and Technology, Nanjing, P.R. China

E-mail: MBEChan@ntu.edu.sg

It was found that infiltration in a butanone solution containing 2.5 % CE (comprising 95 wt% HF-3 and 5 wt % DBA) is optimal to form a coaxial packaging of CE over CNT bundle surfaces while preventing excess resin from blocking the BP pores. However, when the buckypaper sample was immersed into a butanone solution of 10 % CE, the solution was too viscous for CE resin to penetrate into the inner of the buckypaper. A resin-rich coating below buckypaper surface blocked the BP pores and prevented the resin infiltration.



(B)

S1 Cross section of BP-PPy immersed into a butanone solution containing 2.5 % CE with different magnifications



S2 Top view of BP-PPy immersed into a butanone solution containing 2.5 % CE with different magnifications



(A)



S3 Cross section of BP-PPy immersed into a butanone solution of 10 % CE with different magnifications



(A)



S4 Top view of BP-PPy immersed into a butanone solution of 10 % CE with different magnifications

The buckypaper is a porous material, so the droplets are prone to entering the paper and can not keep stable on the surface of BP. This is the reason that the contact angel changes over time. But from the dynamic analysis of the surface contact angles, contact angle of BP-PPy is lower than that of BP. The slope of contact angle curve of BP-PPy is also bigger than that of BP. It implies that the electrode with PPy is more hydrophilic. So that, the open cellular structures of BP-PPy-CE provide better accessibility of electrolyte to penetrate inside and provide a large electrochemically active surface to increase the capacitive performance of the electrode



S5 dynamic analysis of the surface contact angles

The impedance measurement (EIS) was carried out in 1 M KCl solution at an open-circuit potential in the frequency range from 10 mHz to 10kHz. The semicircular part at high frequency is associated with the charge transfer resistance of electrode. There is no significant change between BP and BP-PPy-CE in the semicircular parts, which indicates the very thin PPy and CE coatings are very important for BP-PPy-CE electrodes to remain low electrical resistance.



S6 Electrochemical impedance spectroscopy (EIS) of BP and BP-PPy-CE electrodes