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## **Electronic Supplementary Information**

## Improved supercapacitor performances by adding carbonized $C_{60}$ based nanospheres to PVA/TEMPO-cellulose hydrogel-based electrolyte

Han Jia,<sup>a</sup> Sabina Shahi,<sup>b</sup> Lok Kumar Shrestha,<sup>c,d,\*</sup> Katsuhiko Ariga,<sup>c,e</sup> and Tsuyoshi Michinobu<sup>a,\*</sup>

<sup>a</sup> Department of Materials Science and Engineering, Tokyo Institute of Technology, 2-12-1 Ookayama, Meguro-ku, Tokyo 152-8552, Japan. E-mail: michinobu.t.aa@m.titech.ac.jp
<sup>b</sup> Central Department of Chemistry, Tribhuvan University, Kirtipur, Kathmandu 44613, Nepal
<sup>c</sup> Research Center for Materials Nanoarchitectonics (MANA), National Institute for Materials Science (NIMS), 1-1 Namiki, Tsukuba 305-0044, Japan. E-mail: SHRESTHA.Lokkumar@nims.go.jp
<sup>d</sup> Department of Materials Science, Faculty of Pure and Applied Sciences, University of Tsukuba, 1-1 Tennodai, Tsukuba 305-8573, Japan
<sup>e</sup> Department of Advanced Materials Science, Graduate School of Frontier Sciences, The University

of Tokyo, 5-1-5 Kashiwanoha, Kashiwa 277-8561, Japan



Fig. S1. SEM images of  $C_{60}$ -based nanospheres fabricated with different  $C_{60}$  concentrations.



Fig. S2. Photos of TCCS aqueous suspension after (a) 0 h and (b) 24 h of storage.



**Fig. S3.** SEM images of the porous structure of freeze-dried TC films. The image (b) is a magnified one of the image (a).



Fig. S4. The I-V curve of a freeze-dried TCCS film at a voltage of -1 V to 1 V. The red line is the fitted curve. The sample size was  $0.8 \text{ cm} \times 0.1 \text{ cm} \times 0.03 \text{ cm}$  (length × width × thickness).



Fig. S5. TGA and DTG curves of freeze-dried TC and TCCS films from 20 to 1000 °C.

Table S1. Decomposition temperature  $(T_d)$  and residual amount after TGA measurements of the freeze-dried TC and TCCS films

Sample	$T_{d}$ (90%) (°C)	Residual amount (%)
TC	237	22
TCCS	247	57



Fig. S6. SEM images of TCCS films after TGA measurements. The image (b) is a magnified one of

the image (a).



Fig. S7. Strain-stress curves of (a) PVA-TC and (b) PVA-TCCS hydrogels.

The hydrogels were sandwiched and sealed between two stainless steel discs for the measurement. The ionic conductivities ( $\sigma$ ) of organogels were calculated according to the following equation:

$$\sigma = \frac{L}{RA} \tag{S1}$$

where L represents the thickness of the hydrogel, R represents the impendence value obtained by the intercepts of EIS curves with the x-axis, and A represents the contact area of the organogel between two stainless steel electrodes.



Fig. S8. The EIS plots of PVA-TC hydrogel and PVA-TCCS hydrogel after the solvent exchange with  $1 \text{ M H}_2\text{SO}_4$  aqueous solution measured at a frequency range from  $5 \times 10^5$  to  $10^{-1}$  Hz and 10 mV voltage.

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Sample	Area	Thickness	Ionic conductivity
	$(cm^2)$	(cm)	$(S \text{ cm}^{-1})$
PVA-TC	1	0.10	0.17
PVA-TCCS	1	0.07	0.09

Table S2. Area, thickness, and ionic conductivity of the hydrogels measured by EIS.



**Fig. S9.** (a) The Nyquist plots for the electrode. (b) CV curves vs. scan rate for the electrode. (c) GCD profiles vs. current density for the electrode. (d) Specific capacitance as a function of the current density of the electrode.



**Fig. S10.** (a) CV profiles of the PVA-TC system at different scan rates and (b) the corresponding CD profiles at different currents.