Supporting Information

Improving Charge/Discharge Properties of Radical Polymer Electrodes Influenced Strongly by Current Collector/Carbon Fiber Interface

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A SEM image of a composite electrode was obtained without any deposition using a 3D real surface view microscope (VE-9800 / Keyence Co.). The SEM image of the composite electrode prepared from W1 (Figure S1) showed a typical network of the carbon fibers, and the radical polymer was situated within the carbon network. The carbon fibers act as the electron transfer path to assist the repeatable oxidation/reduction processes of the radical polymer.

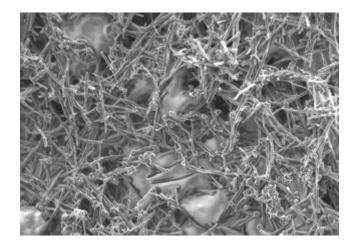


Figure S1. SEM image (x5,000) of the composite electrode prepared from W1. The fibrous network corresponds to the carbon fiber, and the clayey particle corresponds to the radical polymer.

The impedance spectra of composite electrodes recorded on various current collectors such as an ITO/glass plate, a platinum plate, and a glassy carbon plate are shown in Figure S2. The values of R_f obtained in the impedance spectra of the composite electrodes prepared from N1 and N3 depended on the current collectors. The composite electrodes on the ITO/glass plates gave smaller R_f than those on the platinum plates and the glassy carbon plates. The composite electrodes prepared from N1 gave larger R_f on the platinum plate than on the glassy carbon plate. However, the composite electrodes from N3 gave almost the same R_f values on the glassy carbon plate and on the platinum plate. This tendency was the same as observed in composite electrodes prepared from the water-based inks. These results may help us to clarify the dominating factor that influences the process (4).

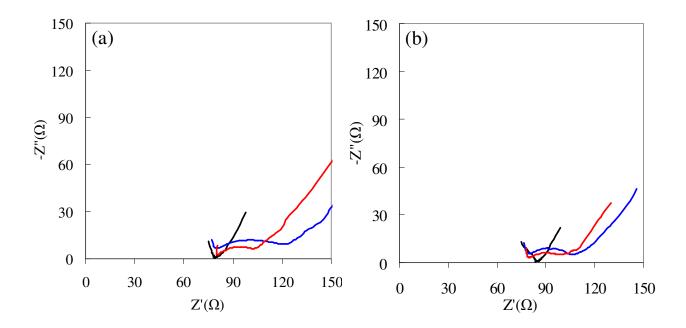


Figure S2. Nyquist plots of (a) composite electrodes prepared from N1 on various current collectors of an ITO/glass plate (black curve), a glassy carbon plate (red curve), and a platinum plate (blue curve), and (b) composite electrodes prepared from N3 on various current collectors of an ITO/glass plate (black curve), a glassy carbon plate (red curve), and a platinum plate (blue curve). The dry film thicknesses of the composite electrodes were about $20 \,\mu$ m.

Work functions of an ITO/glass plate, a glassy carbon plate, a platinum plate, and the several compositions of composite electrodes prepared from the NMP-based inks and the water-based inks were measured by a UPS spectrometer (Model AC-1 / Riken Keiki Co., LTD.) with an ambient ultraviolet photoelectron spectroscopy (UPS) technique. Observed work functions and theoretical volume resistivities of the ITO/glass plate, the glassy carbon plate, the platinum plate, and observed work functions of the composite electrodes were described in Table S1. The work function datum revealed that the energy barrier between the ITO/glass plate and the composite electrodes was higher than that between the platinum plate and the composite electrodes. And the theoretical volume resistivities

revealed that electrons could transfer in the platinum plate more smoothly than in the ITO/glass plate. These results could not explain that the composite electrode on an ITO/glass plate had shown the minimum overall resistance. Thus we assumed that the superiority of an ITO/glass plate as a current collector material would be originated by the surface roughness and adhesion to the composite layers, rather than their work functions and volume resistivities.

Table S1. Work functions and volume resistivites of an ITO/glass plate, a glassy carbon plate, a platinum plate, and composite electrodes.

	ITO/glass plate	glassy carbon plate	platinum plate	composite electrode
volume resistivity($\Omega \cdot cm$)	1.0E-04	4.5E-03	1.0E-05	-
work function(eV)	4.9	4.9	5.4	5.5~5.8