Effect of Meso- and Micro-Porosity in Carbon Electrodes on Atomic Layer Deposition of Pseudocapacitive V_2O_5 for High Performance Supercapacitors

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Figure S1. Comparison of G60 electrodes in a measured using cyclic voltammetry in both a 3-electrode and 2-electrode configuration. There is no significant difference in the performance of the electrodes.

Figure S2. Elemental analysis by EDS of (a) G60 and (b) Supra carbon shows more oxygen in the carbon after the acid treatment consistent with the IR results.

Figure S3. Raman spectra of (a) G60 and (b) Supra. (c) Band fitting for untreated G60 is shown; and (d) the comparison of the peaks is shown for all samples. The carbon powders were measured with a Raman spectrometer after different acid treatments to identify the disorder (D) and graphitic (G) nature of the powder.¹ The deconvolution of the Raman spectra shows that for the G60 powder, the high temperature acid treatment tends to decrease the relative intensity of the D-band (i.e. increasing graphitic structure) whereas the Supra shows a relative increase in the D-band. However, these changes are relatively small, and we do not see significant trends in electrode performance for these changes in D- and G- mode intensity.

Figure S4. (a) V_2O_5 growth per cycle at varying temperatures; and (b) growth with cycle number at 150°C . Film thickness was determined from ellipsometry under saturating conditions. The ALD temperature window and growth rate compare favorably with previous reports.²

Figure S5. (a) XPS and (b) XRD of $400 \text{ \AA}$ vanadium oxide films deposited at 175°C on Si indicate the vanadium oxide deposited is amorphous V_2O_5 .³⁻⁶

Figure S6. Mass gain of (a) G60 and (b) Supra powder that was treated with boiling nitric acid as a function of ALD cycles of V_2O_5 at 150°C and the corresponding thickness of V_2O_5 measured on monitor Si wafers. The resulting linear increase in mass gain fits for G60 carbon fits in the with the linear film growth seen on monitor silicon wafers. Similar trends were also seen for the limited number of Supra powder coated. The G60 and Supra carbons displayed different mass gains after similar number of ALD cycles. The variation could be a result of the Supra carbon powder that was coated having a surface area that was only about 1/3 of that of the G60 carbon powder that was coated. We would expect different carbon powders to have a linear mass gain as well, but with a different slope, which would vary as a result of different pore size distribution and surface area of the carbons.

Figure S7. Effect of acid treatment on capacitance measured in a 3-electrode cell on (a) G60 and (b) Supra in 1 M KCl reported at a scan rate of 200 mV/s.

Figure S8. Coulombic efficiency of V_2O_5 coated G60 electrodes as a function of ALD cycles charged/discharged from 0.0 to 0.8 V at a rate of 5 A/g.

Figure S9. EIS model results of G60 electrodes coated with V_2O_5 . (a) specific capacitance and (b) specific capacitance of deposited V_2O_5 .

Figure S10. Current response during cyclic voltammetry from either capacitive processes (varies with v) or diffusion processes (varies with $v^{-1/2}$) in the G60 EC coated with V_2O_5 ALD: (a) 0 cycles; (b) 75 cycles; (c) 200 cycles; and (d) 1000 cycles according to the relationship $i(V) = k_1v + k_2v^{1/2}$, where k_1 and k_2 are constants at a particular sweep rate. The capacitive current (dashed line) represents 67 to 93% of the total current (solid line).^{7,8}

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