## SUPPORTING INFORMATION

Manganese hexacyanoferrate/MnO<sub>2</sub> composite nanostructures as cathode material for supercapacitors

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The stoichiometry for the as-synthesized material could be written as  $K_{2-x}Mn^{3+}_{x}Mn^{2+}_{1-x}Fe^{2+}(CN)_{6}\cdot zMnO2\cdot 1.036zMnOOK\cdot mH2O$ , Irrespective of the impurities. For the quantitative determination of K, Mn and Fe, about 2 mg of powder was dissolved in 10 mL of de-ionized water. The total amount of K, Mn and Fe in the powder was determined as a K: Mn: Fe ratio of 1.95: 1.77: 1 by inductively coupled plasma mass spectroscopy. The O 1s core level XP spectra was used to determine the content ratio, MnO2: MnOOK=1: 1.036 (Table 1). The total  $K^+$  content was from KMnFe(CN)<sub>6</sub> nH<sub>2</sub>O,  $K_2Mn^{2+}Fe^{2+}(CN)_6$  and MnOOK. The high content of K indicates a high electrochemical capacitance. According to the above data, the final formula of our material could be determined as  $K_{1.56}Mn^{3+}_{0.44}Mn^{2+}_{0.56}Fe^{2+}(CN)_6 \cdot 0.38MnO_2 \cdot 0.39MnOOK \cdot mH_2O$ , Irrespective of the impurities. If all the mass of MnO<sub>2</sub> take participate in electrochemical reaction, theoretical specific capacitance for this powder would be 298 F g<sup>-1</sup>.

Unfortunately, as for the mass of active material (m), in fact, it decreased during the first a few segments of "Deep electro-oxidation", because the unstable components of electro-active material, such as  $K_2Mn^{2+}Fe^{2+}(CN)_6$ , tend to be dissolved. The decreased amount was not large and could hardly be evaluated. We use the origin mass of active material to calculate the specific capacitance and specific capacity, ignoring the material wastage.



Figure 1S. XRD pattern of the active materials in DOME. The three strong-lines correspond to (200), (220) and (400) planes of KMnFe(CN)<sub>6</sub>·nH<sub>2</sub>O, respectively. As MnHCF crystals broke apart into small crystals because of the intercalation or de-intercalation of Na<sup>+</sup> ions, the three peaks was not as strong as the original samples. Residual acetylene black and polyvinylidene difluoride dry powder (PVDF) stick to the active materials also affected the peak intensity.