† Electronic Supplementary Information (ESI)

XPS measurements were done at a PHI 5600 system (Physical Electronics, USA) using Mg-kα X-rays for excitation and 3.5 keV Ar^+ ions at 45° and 2 nm/min sputter rate for depth profiling. Similar to AES the concentration quantification was done using standard single element sensitivity factors.

The Fe 2p XPS spectrum reveals that iron is present in the iron (hydr)oxide CMK-3 nanocomposites only in the Fe^{3+} oxidation state. The peak splitting observed in the O1s spectrum as well as the calculated stoichiometric amount of oxygen in the detected iron oxide between 75 at.% ($Fe(OH)_3$) and 60 at.% (Fe_2O_3) points to an (hydr)oxide modification.

Comparison of AES and XPS results: A carbon-to-iron ratio [at.%] of 93.0:7.0 was calculated from averaging values gained by AES for 100 sputter cycles at three different sample areas. These values are comparable with the results of the XPS investigations. A carbon-to-iron ratio [at.%] of 93.5:6.5 was detected there. This clearly points at the homogeneous functionalization of the inner surface area of the nanocomposite structure, as XPS analysis gains an average over at least one powder particle and DP-AES measures the locally varying chemical composition within one nanocomposite powder particle.

The CV cycling stability with increased potential scan rate is shown in Figure ESI-1. Prior to the capacitance determination the symmetric two electrode cells were cycled 20 times at a potential scan rate of 10 mVs^{-1}. To determine the capacitance at each potential scan rate five cycles were measured. The results are presented here.

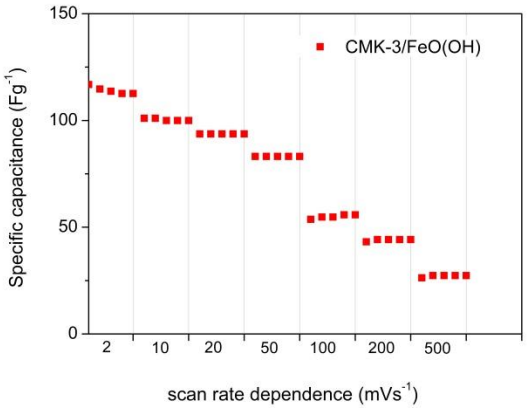

Fig. ESI-1 Potential scan rate dependence of the iron (hydr)oxide CMK-3 nanocomposite. (The integral specific capacitance of the electroactive material is shown for each of the five cycles at each potential scan rate.)

To compare published iron oxide carbon composite materials to our optimized nanocomposite structure the energy density of the materials is calculated according to the following equation (1). Results are listed at Table ESI-1.

$$E = \frac{1}{2} C_{spec} V^2 \qquad (1)$$

where E is the energy density (Wh kg^{-1}), C_{spec} is the integral specific capacitance (Ah $V^{-1}kg^{-1}$) based on the mass of electroactive material of both electrodes for a two electrode arrangement (2EA) and of one electrode for a three electrode arrangement (3EA) and V is the operating potential window (V).

Table ESI-1 Energy density of different iron oxide carbon composite materials.

Electrode material	V (V)	C_{spec} (Fg^{-1})	C_{spec} ($AhV^{-1}kg^{-1}$)	E ($Whkg^{-1}$)
C-FeO(OH), 3EA [1]	1.0	85	23.6	11.8
C-Fe_2O_3, 3EA [2]	0.85	151.8	42.2	14.8
C-Fe_3O_4, 3EA [3]	0.6	163.4	45.4	8.2
C-Fe_2O_3, 2EA [4]	1.4	25 (50)*	6.9 (13.9)*	6.8 (13.6)*
C-FeO(OH), 2EA, this study	1.8	56.5 (113)*	15.7 (31.4)*	25.4 (50.9)*

* Values in brackets based on the mass of one electrode for two electrode arrangement

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