Electronic Supplementary Information

Direct self-assembly of Fe$_2$O$_3$/reduced graphene oxide nanocomposite for high-performance lithium-ion battery

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**Fig. S1** Zeta potential measurements of GO and Fe$_2$O$_3$ NPs dispersions in pure water individually.

**Fig. S2** XRD patterns: a) GO and graphite and b) rGO.
Fig. S2b shows a broad peak around 23° and a weak narrow peak at around 43°, demonstrating the rGO possesses disordered sheets stacking structures. Unfortunately, in the XRD pattern of the Fe$_2$O$_3$/rGO nanocomposite (Fig. 2), these peaks overlap either with the diffraction peaks from Fe$_2$O$_3$ nanoparticles or with background diffraction. We would like to note that diffraction reflexes of well-exfoliated rGO in nanocomposites are often absent in materials reported in literature (e.g., ACS Appl. Mater. Interfaces, 2013, 5, 3764; ACS Nano, 2012, 6, 8349; J. Mater. Chem. A, 2014, 2, 13942; J. Power Sources, 2013, 239, 37).

Fig. S3 Raman spectrum of GO.

Fig. S4 XPS spectrum of GO.
Fig. S5 FTIR of GO, rGO, Fe$_2$O$_3$ and Fe$_2$O$_3$/rGO.

FTIR results of GO and rGO and Fe$_2$O$_3$/rGO show that after the reduction process the oxygen-containing groups have almost disappeared in the spectra of rGO and Fe$_2$O$_3$/rGO. New absorption bands centered at 1575 cm$^{-1}$ in rGO and Fe$_2$O$_3$/rGO spectra are attributed to the skeletal vibrations in the graphene sheets (J. Phys. Chem. C, 2011, 115, 20747; J. Mater. Chem., 2010, 20, 1722). These results indicate that the GO was successfully reduced to rGO after the low-temperature hydrothermal process (Carbon, 2012, 50, 1959; J. Mater. Chem. A, 2014, 2, 18480). Furthermore, a broad band at around 1540 cm$^{-1}$ in the spectra of rGO and Fe$_2$O$_3$/rGO is attributed to the vibration of N-H groups (J. Mater. Chem. A, 2014, 2, 18480; Carbon, 2012, 50, 1959). The Fe-O absorption band centered at 560 cm$^{-1}$ appears in the spectrum of Fe$_2$O$_3$/rGO, which further confirmed the loading of Fe$_2$O$_3$ nanoparticles on the rGO layers (Molecules, 2014, 19, 4997; J. Mater. Chem., 2012, 22, 895).
HR-TEM images of the Fe$_2$O$_3$/rGO nanocomposite reveal that the crystallized Fe$_2$O$_3$ NPs sit on the rGO layers. Even at the edge of the rGO, most of the Fe$_2$O$_3$ NPs are enwrapped by the rGO layers. The rGO layers are well exfoliated and each sheet has around three layers. The rings in FFT (Fig. S7a upper inset) are ascribed to (220) and (311) of Fe$_2$O$_3$ nanoparticles.
**Fig. S7** HR-TEM images of the Fe$_2$O$_3$/rGO nanocomposite at different view a) and b), upper inset in a) is the FFT image of the Fe$_2$O$_3$ nanoparticles in a).

**Fig. S8** Thermogravimetry measurement of the Fe$_2$O$_3$/rGO nanocomposite.
**Fig. S9** XRD analysis of the TG measurement residue of the Fe$_2$O$_3$/rGO nanocomposite.

**Fig. S10** Impedance spectra in Nyquist complex plane plots and respective equivalent circuits used for the fit for a) graphene oxide, b) Fe$_2$O$_3$ nanoparticles and c) the Fe$_2$O$_3$/rGO nanocomposite. d) specific conductivities of the materials calculated from their geometries (see text).
The impedance measurements were fitted to suitable equivalent circuits to exactly reproduce the impedance properties of the different materials. **Fig. S10a** shows the Nyquist plots (measurement and fit) of graphene oxide pellets and the respective equivalent circuit. It consists of a parallel circuit with a resistor and a Constant Phase Element (CPE). The impedance measurement shows a typical semicircle as it is expected for a parallel RC element, however, fitting to a RCPE element results in a much better agreement between measurement and fit. This might be attributed to a slight distribution around a mean distance between the graphene oxide sheets leading to some variation in capacitance. The impedance measurements of compacted iron oxide nanoparticles are shown in **Fig. S10b**. In contrast to the measurements of graphene oxide it is obvious that the impedance spectrum consists of two contributions. The right part can be attributed to an RCPE element while the left part can be fitted to an RC element. These two contributions can be assigned to the grain boundary and bulk conductivity, respectively. Due to the low resistance of the Fe$_2$O$_3$/rGO composite material (about 100 $\Omega$ compared to about $10^6$ $\Omega$ (Fe$_2$O$_3$) and $10^8$ $\Omega$ (GO)), inductive effects originating from the wiring and the composite itself have to be taken into account as can be seen from the spectrum shown in **Fig. S10c**. Therefore, an inductance was added to the equivalent circuit to consider this effect.

The specific conductivities of the compacted materials were calculated from the geometry of the pressed pellets (area and thickness) and the outcome of the equivalent circuit fits. **Fig. S10d** shows the respective results for the real part of the specific conductivities plotted against the frequency. It is obvious that GO was successfully reduced to rGO and that the conductivity of the composite is significantly enhanced compared to pure Fe$_2$O$_3$ nanoparticles thus promoting electron transport between the nanoparticles and the current collector.
Fig. S11 Galvanostatic cycling at different current densities: Delithiation capacity and Coulombic efficiency vs. cycle number for rGO.