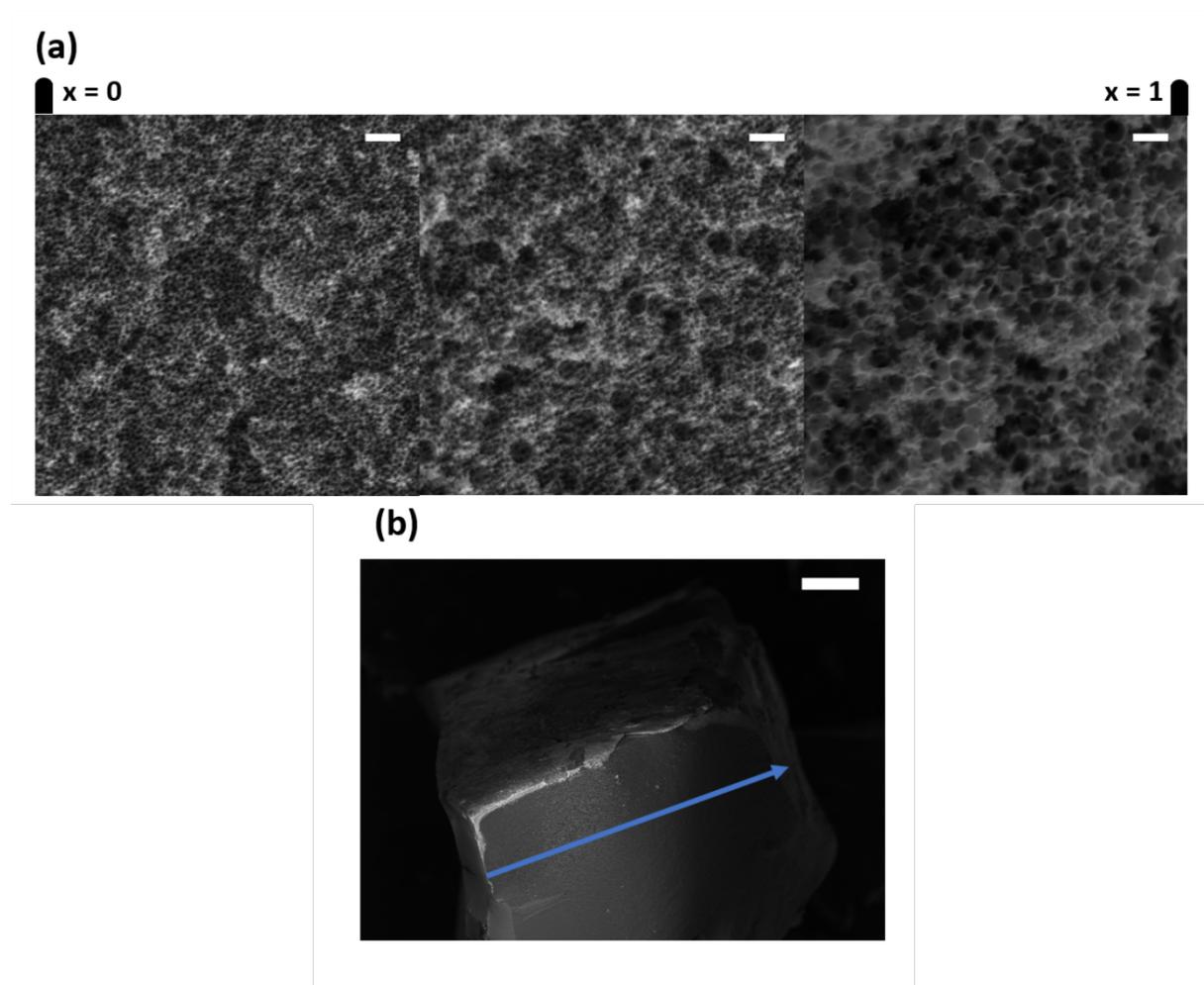


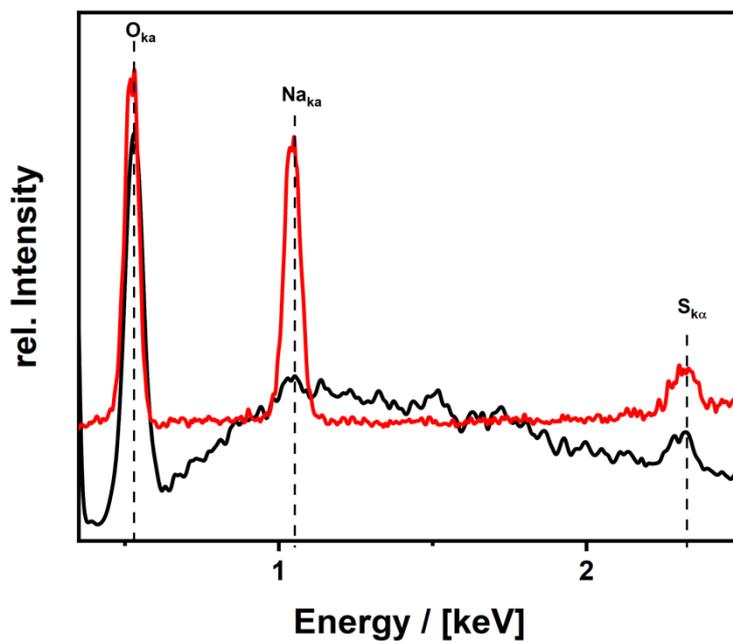
# Anisotropic Magnetism in Gradient Porous Carbon Composite Aerogels

*Jochen Bahner, Nicolas Hug and Sebastian Polarz*

**Figure S1.** SEM micrographs of a sulfur doped gradient carbon material



**Figure S2.** EDX measurement of a sulfur doped carbon material before and after washing.

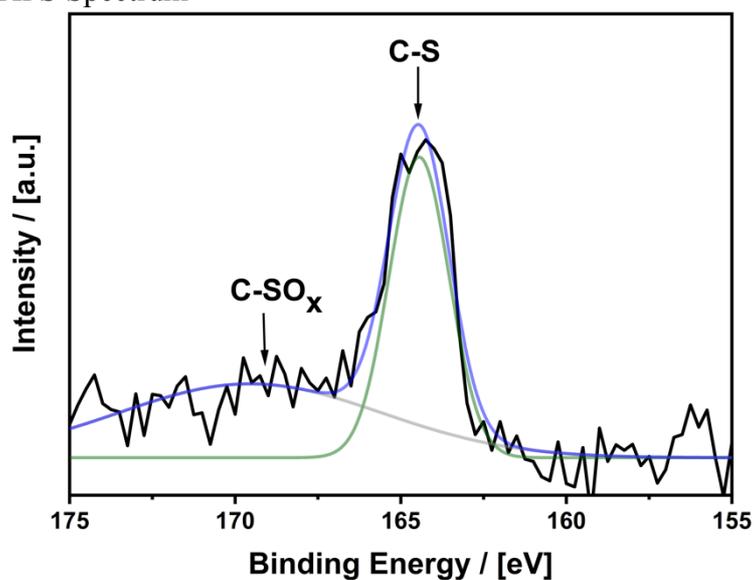


EDX-Spectra of a sulfur doped gradient porous carbon material before (red) and after (black) washing with diluted HCl.

To exclude the incorporation of inorganic sulfur salts into the gradient porous carbons, we treated the material with diluted hydrochloric acid. This leads to a complete removal of sodium species, while sulfur remains bound in the carbon network.

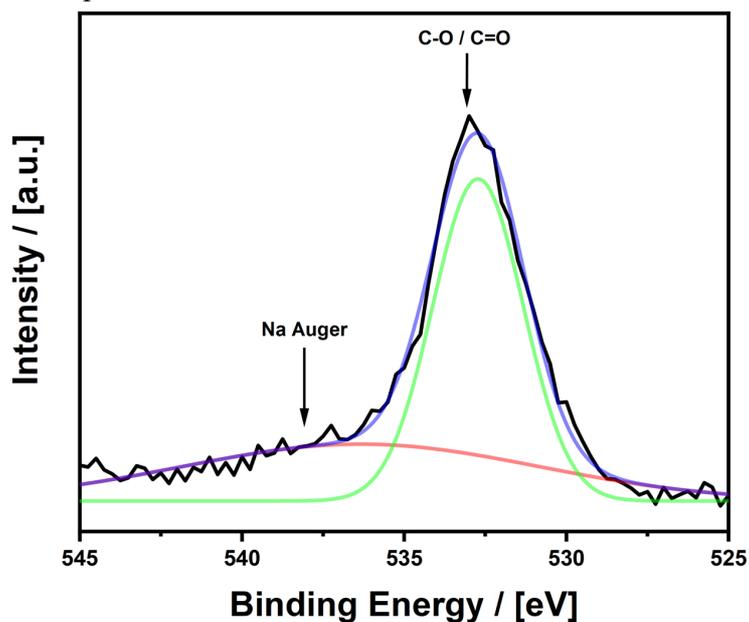
**Figure S3-XPS spectra of a sulfur doped gradient porous carbon material**

(a) Detailed S 2p XPS Spectrum



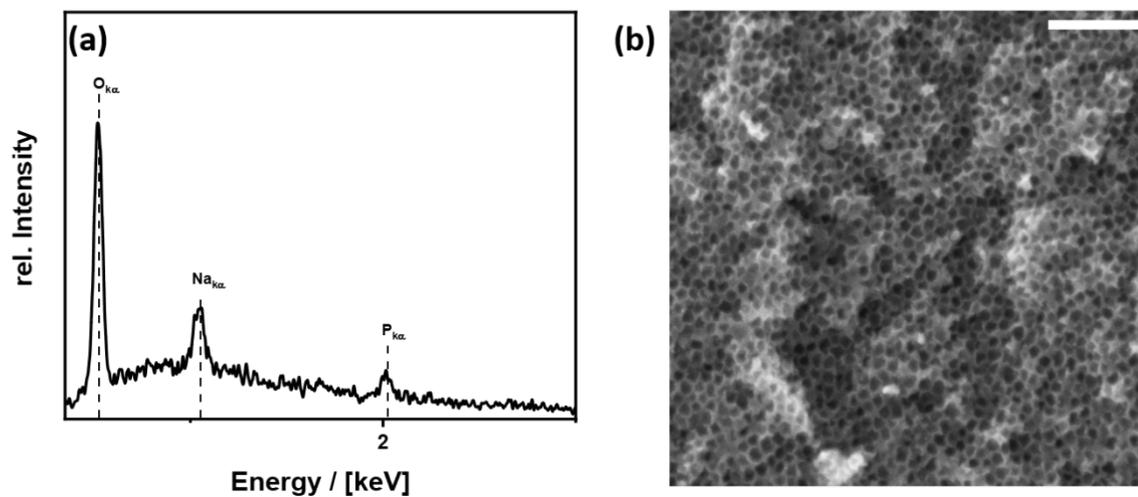
Blue: cumulative fit, Deconvolution given in green for C-S bonds and grey for C-SO<sub>x</sub>-species.

(b) Detailed O 1s XPS Spectrum



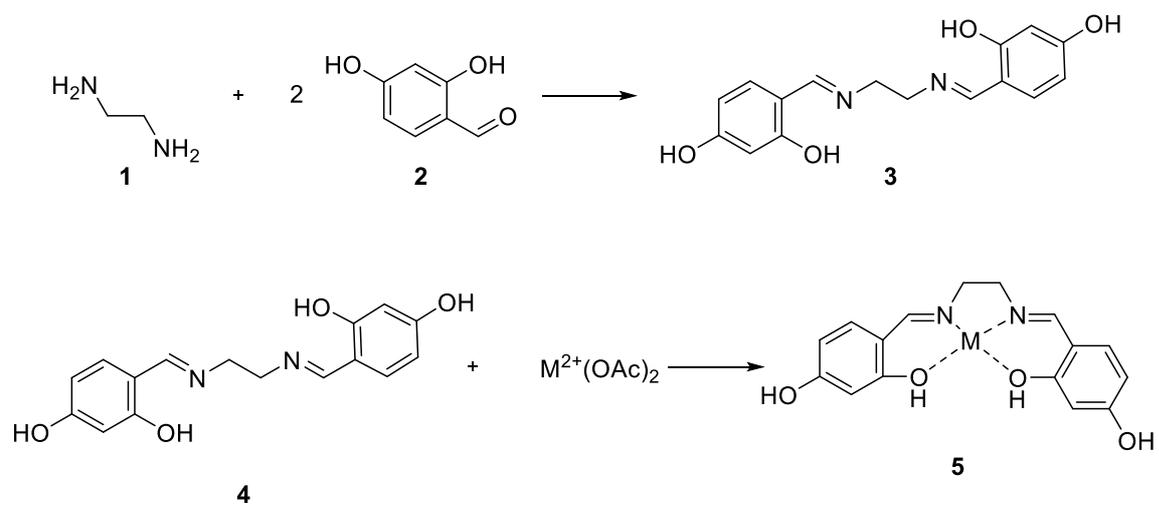
Blue: cumulative fit, red: deconvolution of the Na-Auger signal, green: deconvolution of the C-O/C=O signal.

**Figure S4.** Analytic data of a phosphorous doped carbon material.

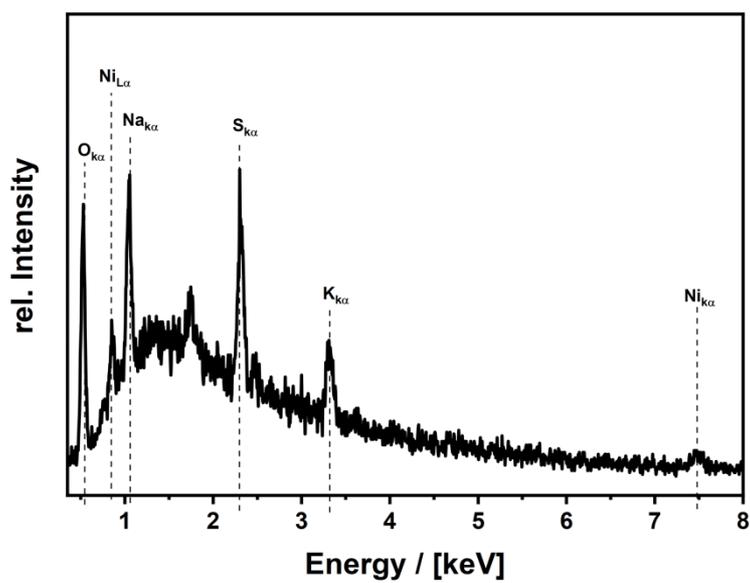


(a) EDX-spectra and (b) SEM micrograph (scale 500 nm) of a monomodal carbon material synthesized using pol(styrene-co-vinylphosphonicacid) templating particles.

**Scheme S1: Synthesis of modified metal salen complexes.**

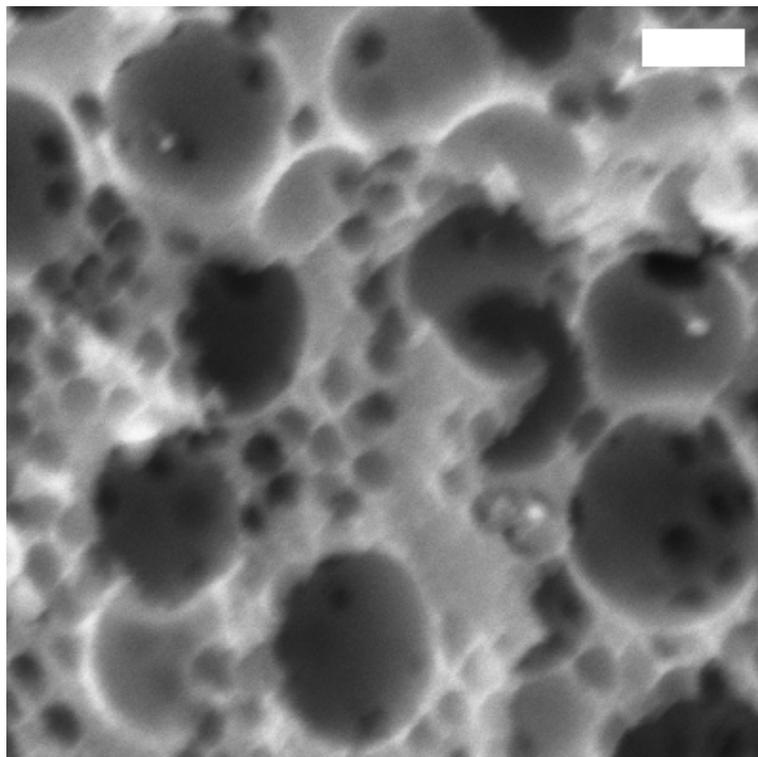


**Figure S5.** EDX spectrum of a homogenized Ni@C sample.



Sodium and potassium stemming from remaining salts bound to the surface after washing with water.

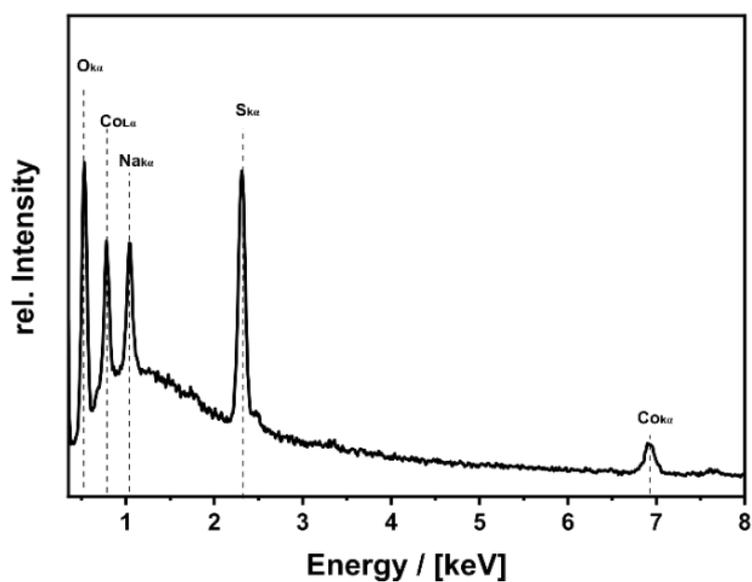
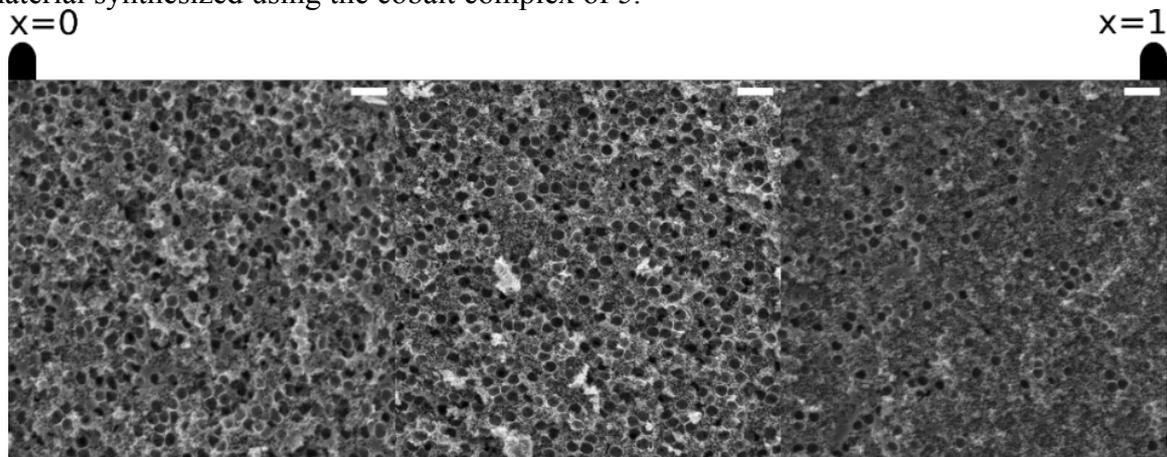
**Figure S6.** Enlarged SEM micrograph of a Ni@C sample.



Scale bar is 200 nm.

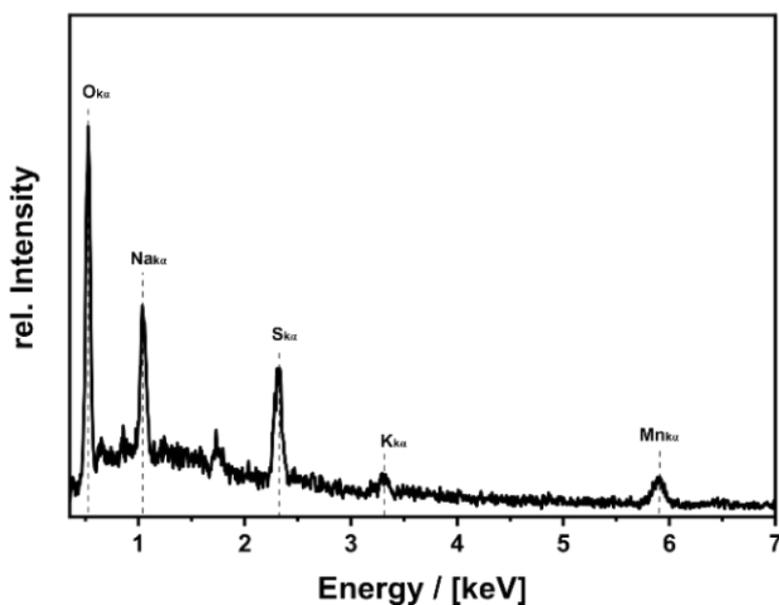
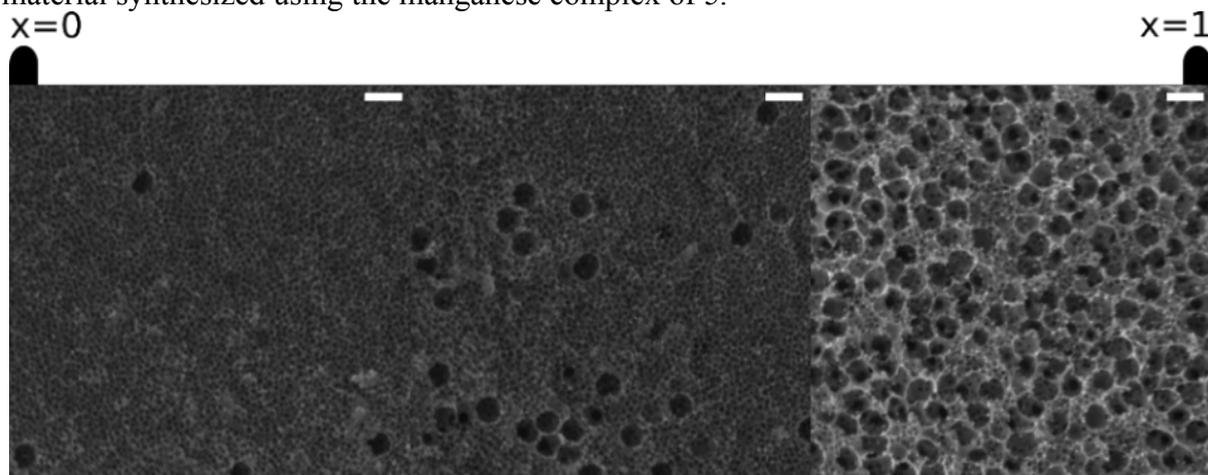
**Figure S7** Metal-Carbon composites synthesized using 5.

(a) SEM Micrographs (top, scale = 1000 nm) and representative EDX spectra (bottom) of a material synthesized using the cobalt complex of 5.



Sodium stemming from remaining salts bound to the surface after washing with water.

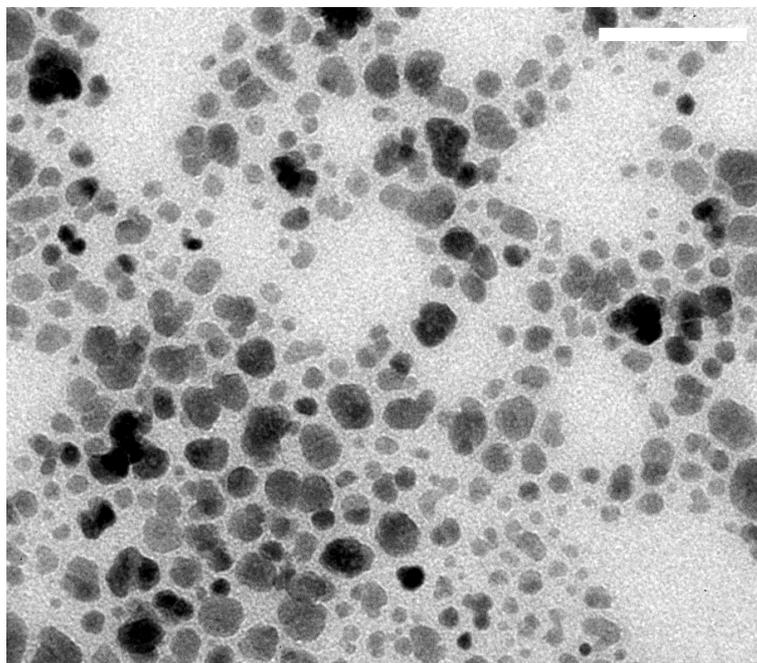
(b) SEM Micrographs (top, scale = 1000 nm) and representative EDX spectra (bottom) of a material synthesized using the manganese complex of 5.



Sodium and potassium stemming from remaining salts bound to the surface after washing with water.

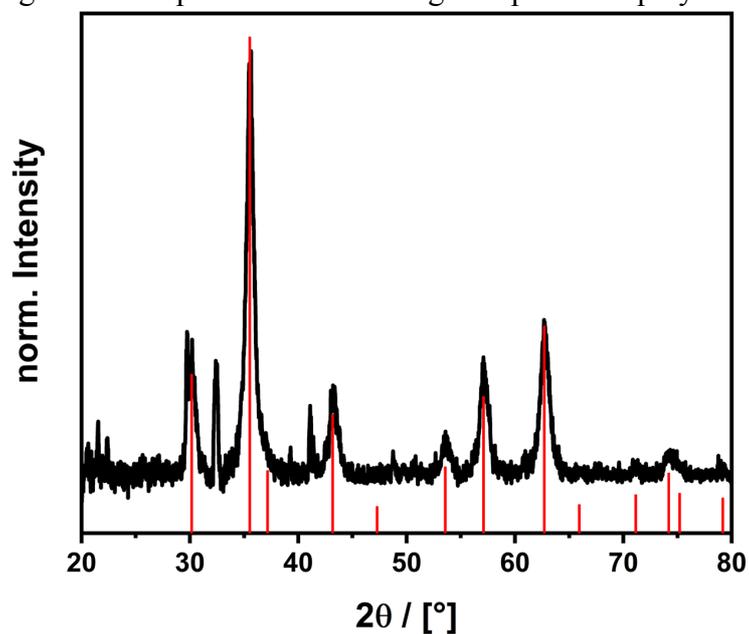
**Figure S8. Analytical data of the synthesized MagPS nanoparticles**

a) TEM micrograph of the synthesized magnetite nanoparticles used during encapsulation polymerization



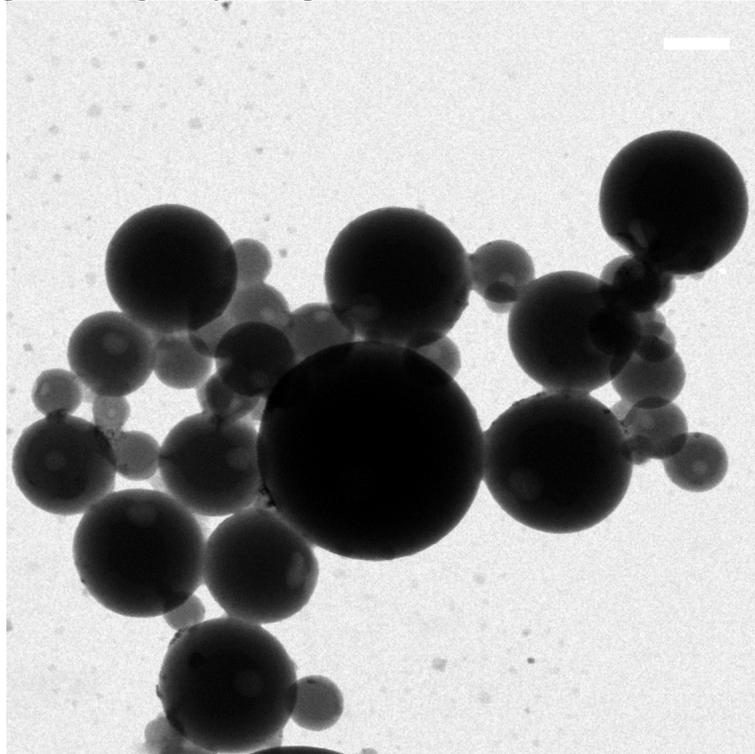
TEM micrograph, scale bar 50 nm

b) PXRD of the magnetite nanoparticles used during encapsulation polymerization



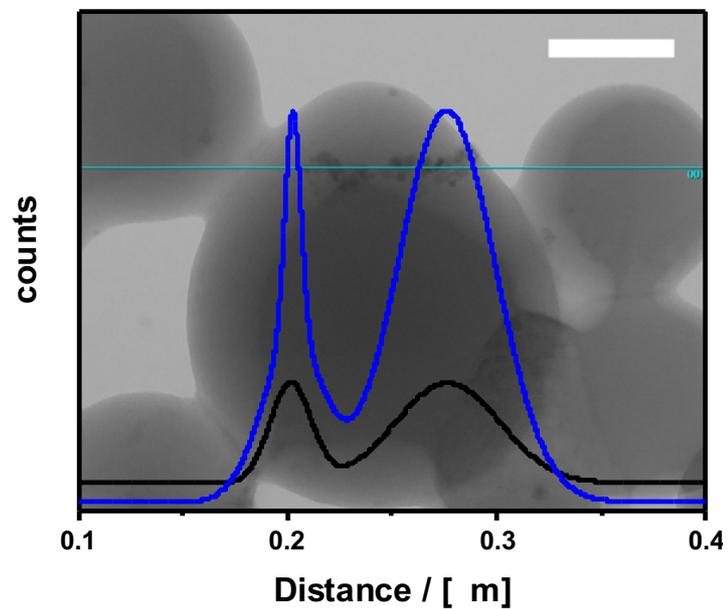
PXRD of the synthesized particles with magnetite reference (COD 2101926, red)

c) TEM micrographs of MagPS-hybride particles



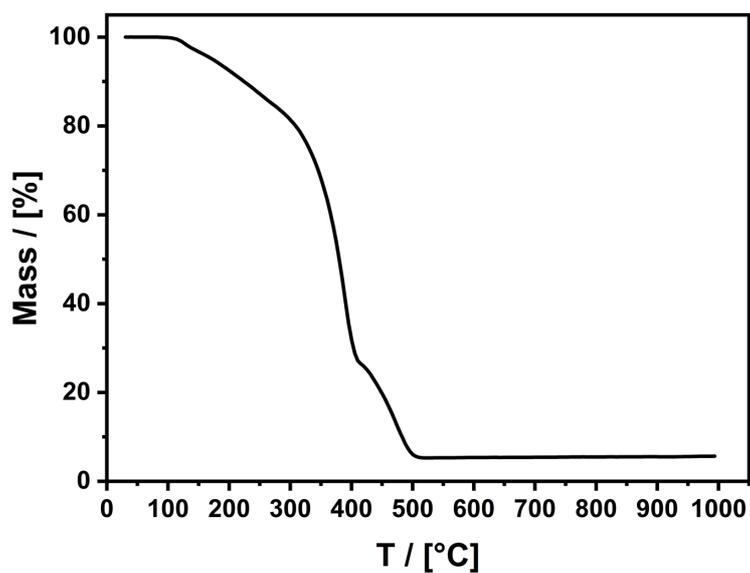
TEM micrograph, scale bar: 200 nm

d) enlarged TEM micrograph and EDX linescan along the indicated direction of Mag@PSS-hybride particles



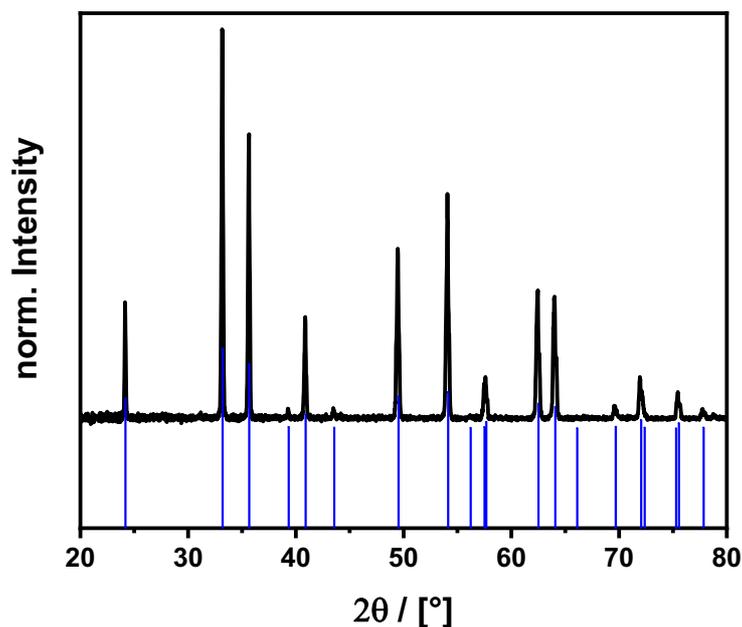
TEM micrograph with EDX Linescan measurement, blue: Fe<sub>Kα</sub>, black: O<sub>Kα</sub>, slight shift in maxima due to drifting during measurement

e) TGA trace of Mag@PSS-hybride particles conducted under oxygen atmosphere



Decomposition process under oxygen, showing a thermal depolymerization of polystyrene starting from 300 °C, followed by an oxidation of magnetite to hematite. Remaining mass is 5.4 %.

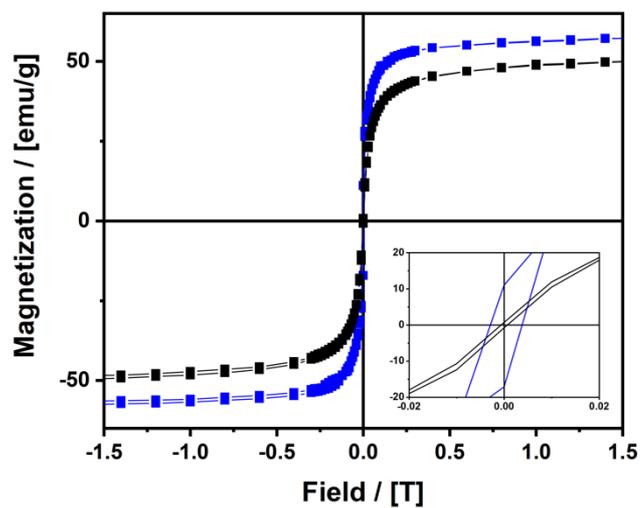
f) PXRD of the residue after TGA of Mag@PSS under oxygen



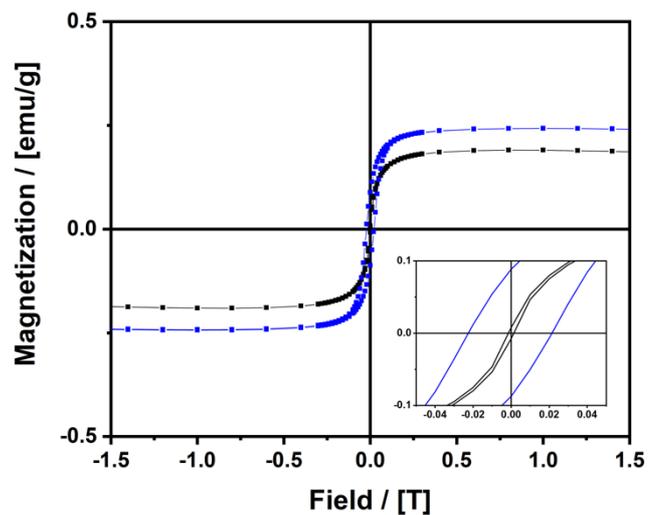
Black: measured data, blue: hematite reference (COD 9009782); hematite formed by oxidation of magnetite during TGA measurement.

**Figure S9. SQUID measurements of magnetite particles and MagPS Particles**

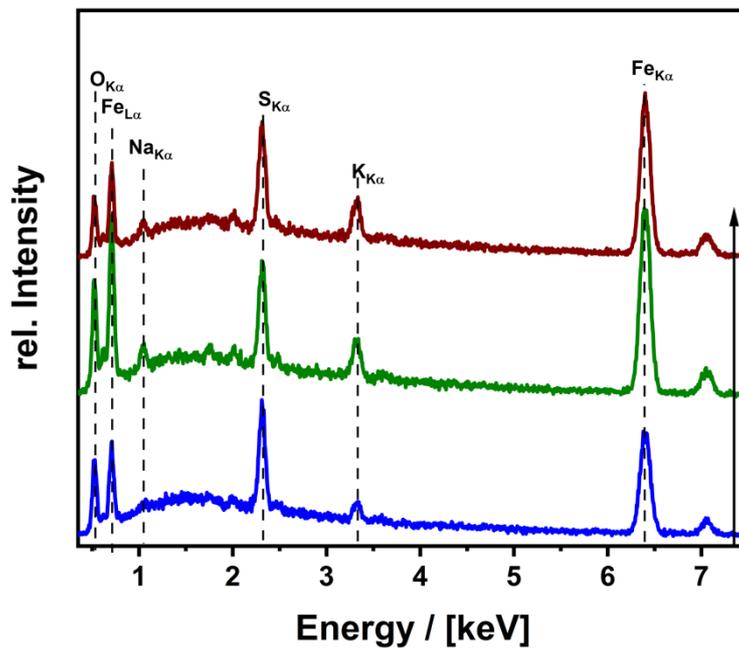
SQUID of the magnetite particles at 300 K (black) and 4 K (blue)



b) SQUID of Mag@PSS at 300 K (black) and 4 K (blue)

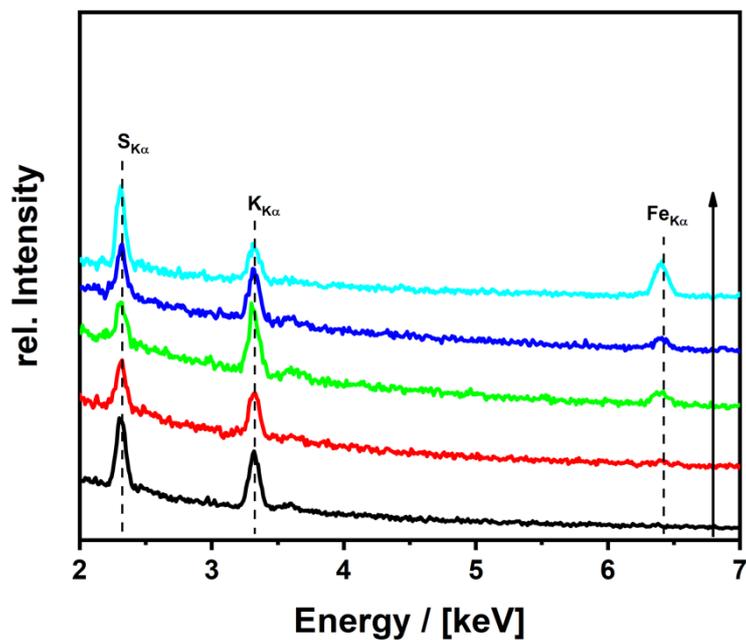


**Figure S10:** EDX spectra recorded along the direction of the centrifugal force of the material presented in Figure 2.



Arrow indicating the direction of the centrifugal force, color in accordance to Fig. 2. Sodium and potassium stemming from remaining salts bound to the surface after washing with water.

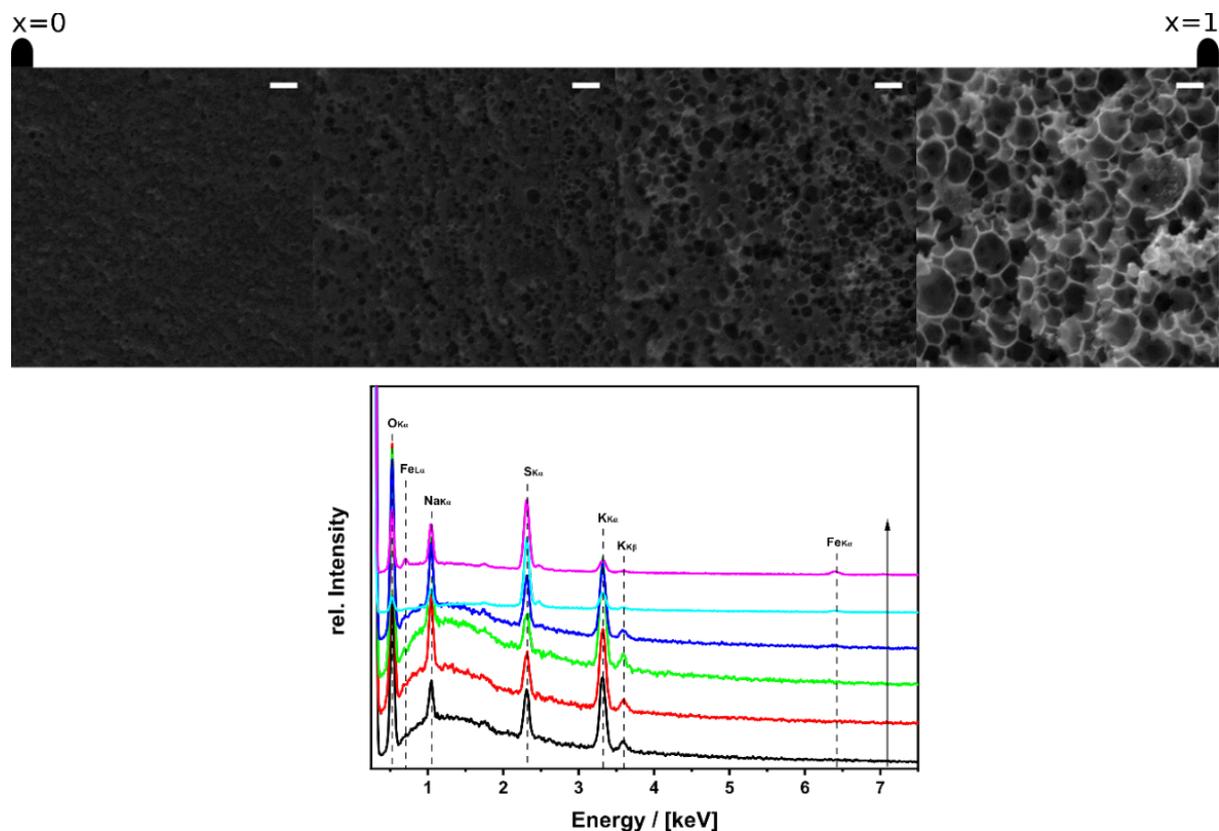
**Figure S11.** EDX spectra of a material prepared using PSS-137 and Mag@PSS



EDX spectra in direction of the centrifugal force (indicated by arrow). Potassium stemming from remaining salts bound to the surface after washing with water.

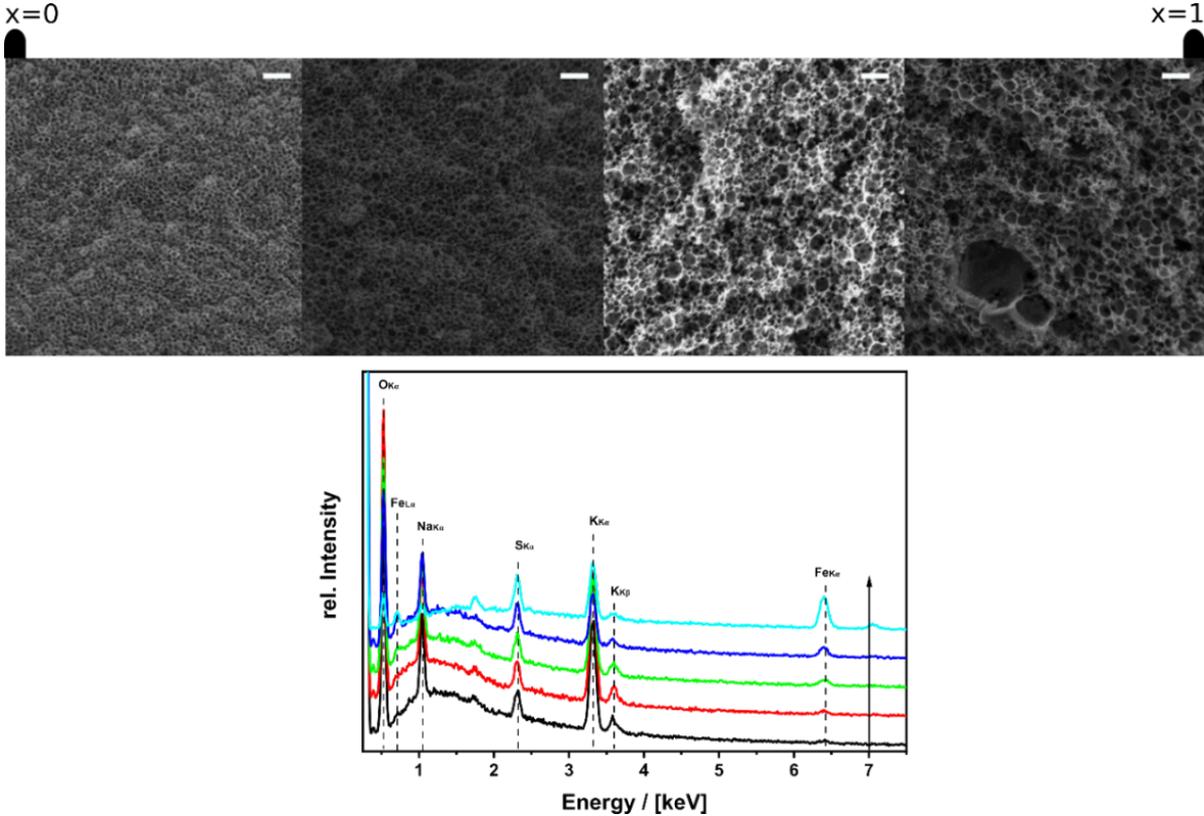
**Figure S12. Additional dual gradient materials**

(a) SEM Micrographs (top, scale = 500 nm) and EDX spectra (bottom) of a material synthesized using PSS-38 and Mag@PSS taken along the direction of the centrifugal force.



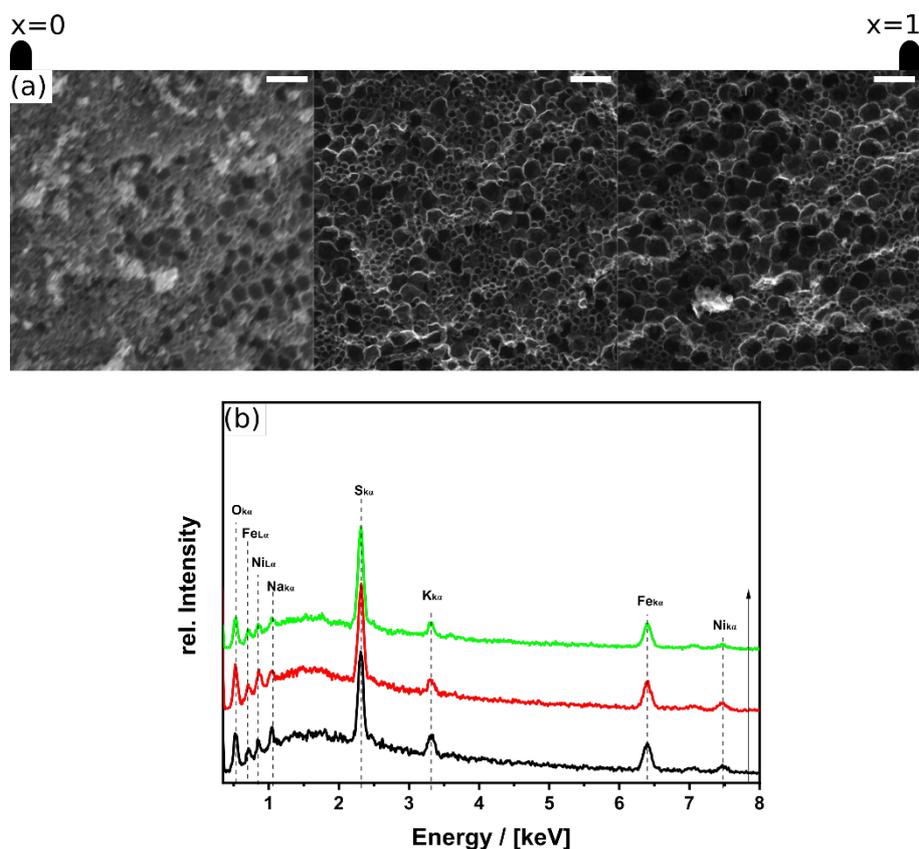
Sodium and potassium stemming from remaining salts bound to the surface after washing with water.

(b) SEM Micrographs (top, scale = 1000 nm) and EDX spectra (bottom) of a material synthesized using PSS-288 and Mag@PSS taken along the direction of the centrifugal force.



Sodium and potassium stemming from remaining salts bound to the surface after washing with water.

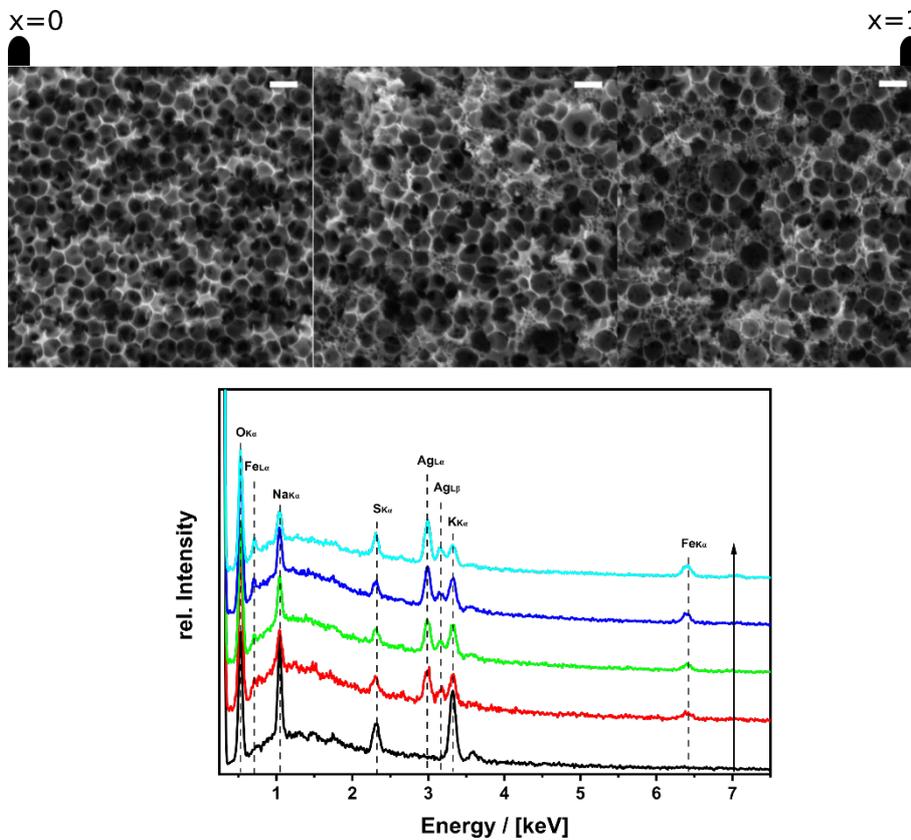
**Figure S13.** Analytical data for a material synthesized using PS-137, Mag@PSS and 5.



(a) SEM micrographs, scalebar: 500 nm; (b) EDX spectra of the material taken along the direction of the centrifugal force. Sodium and potassium stemming from remaining salts bound to the surface after washing with water.

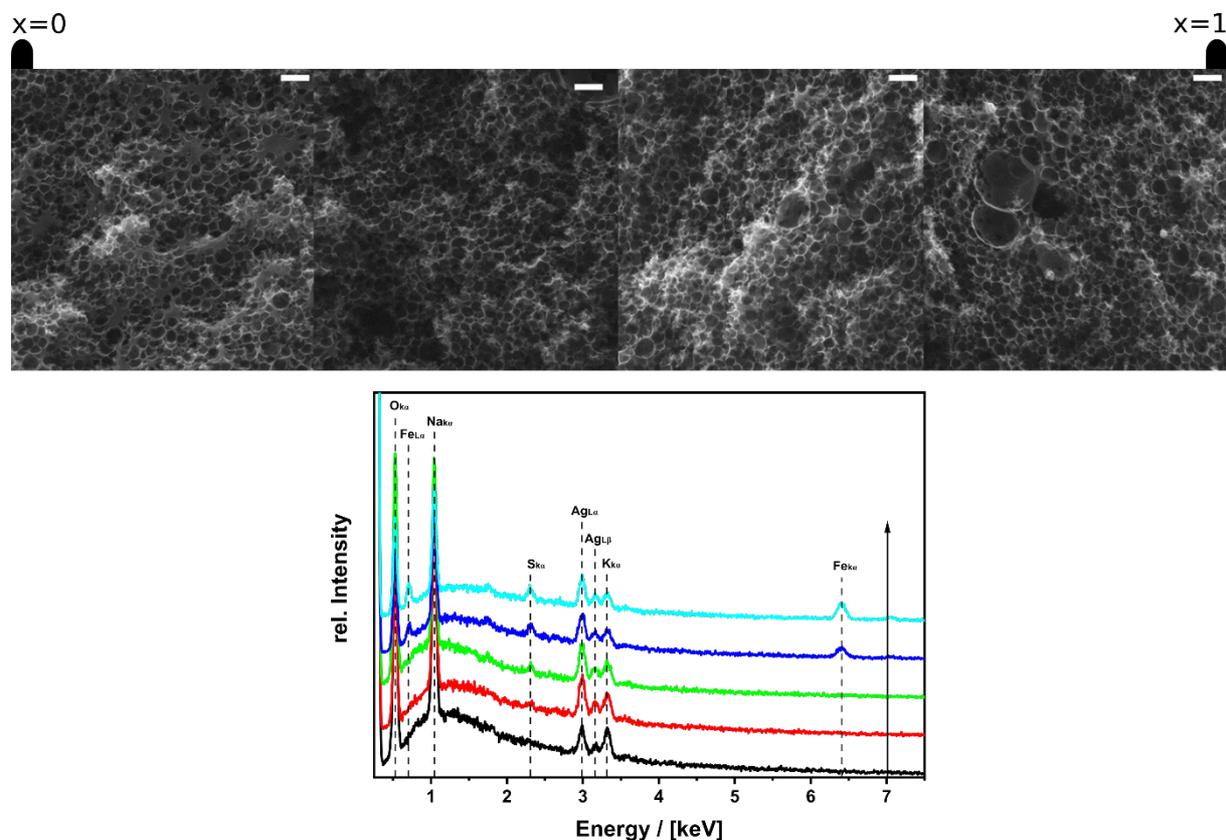
**Figure S14. Analytical data for Ag+Mag@C materials**

(a) SEM Micrographs (top, scale = 1000 nm) and EDX spectra (bottom) of a material synthesized using Ag@PSS-138 and Mag@PSS taken along the direction of the centrifugal force.



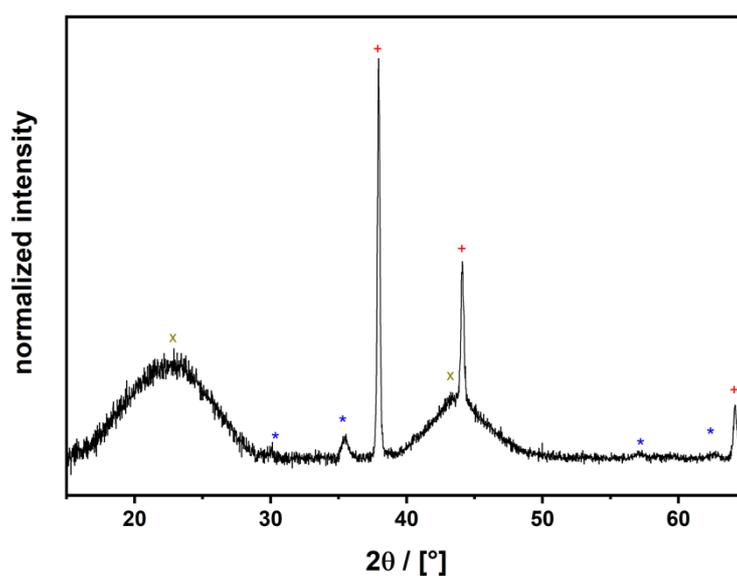
Sodium and potassium stemming from remaining salts bound to the surface after washing with water.

(b) SEM Micrographs (top, scale = 1000 nm) and EDX spectra (bottom) of a material synthesized using Ag@PSS-480 and Mag@PSS taken along the direction of the centrifugal force.



Sodium and potassium stemming from remaining salts bound to the surface after washing with water.

**Figure S15.** PXRD of a dual gradient magnetite/silver material.



Signals marked corresponding to carbon (olive), magnetite (blue) and silver (Ag).