**Supporting Information for:**

# High sulfur content composite materials from renewable fatty acid cellulose esters (FACE) via inverse vulcanization

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## Contents

# 1 Yield calculation for FACEs

The corresponding yields of the synthesized FACE were determined according to Eqn. S1 and S2:

$$\text{Yield} = \frac{\left(\frac{m_{CE}}{M_{repunit}}\right)}{\left(\frac{m_{cellulose}}{M_{AGU}}\right)} \quad \text{(S1)}$$

$$M_{repunit} = M_{AGU} + (M_S - 1{,}01\ \text{g mol}^{-1})\ \text{x}\ DS_{1H} \quad \text{(S2)}$$

$m_{CE}$: mass of cellulose ester
$m_{cellulose}$: mass of cellulose educt
$M_{repunit}$: average molar mass of CE repeating unit
$M_{AGU}$: molecular weight of anhydroglucose unit (162.14 g mol$^{-1}$)
$M_S$: molecular weight of the substituent (without the linking oxygen atom between the substituent and the cellulose backbone; 167.27 g mol$^{-1}$)

# 2 Structural characterization FACE

## 2.1 ATR-IR spectra of FACE

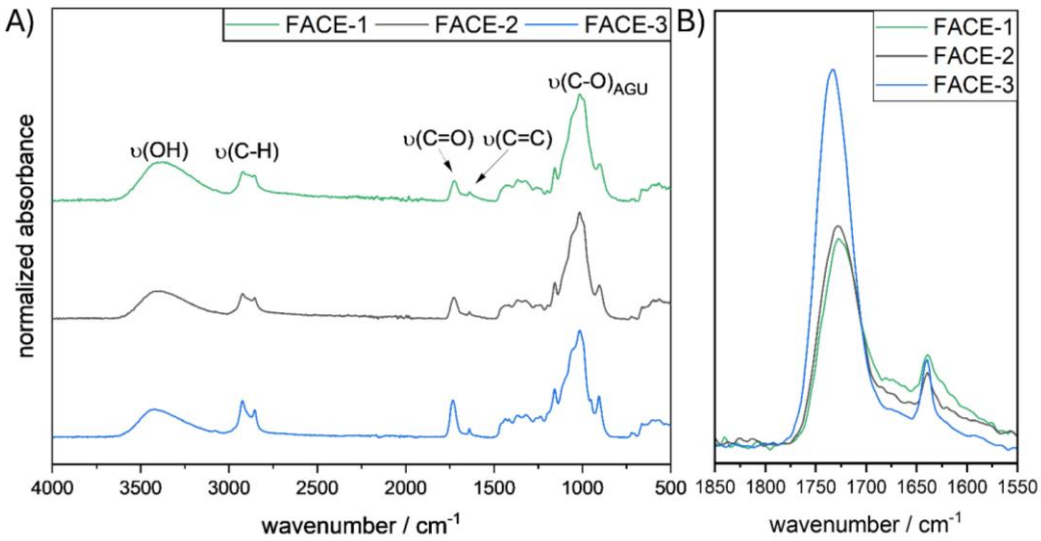


**Figure S1.** (A) ATR-IR spectra of FACE-1 (green line), FACE-2 (black line), and FACE-3 (blue line). (B) Expanded view of the C=O and C=C stretching vibration bands for FACE-1 (green line), FACE-2 (black line) and FACE-3 (blue line).

## 2.2 $^{1}$H NMR spectra of FACE

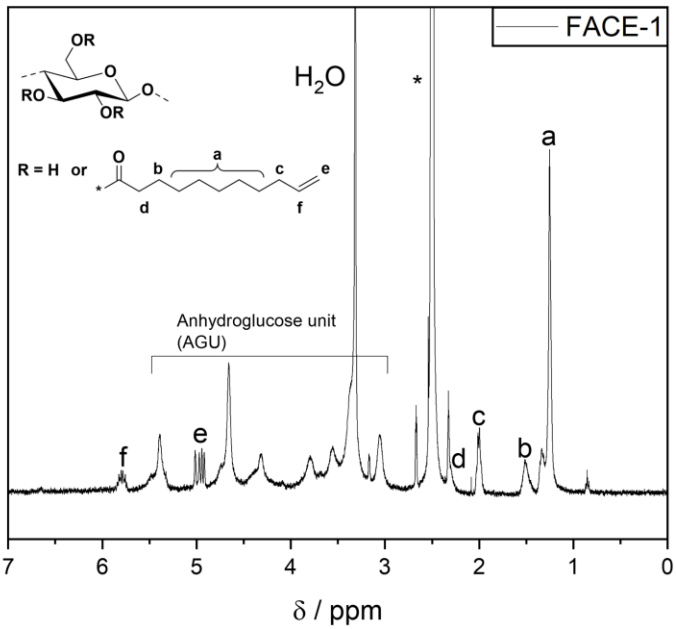


**Figure S2.** $^{1}$H NMR (400 MHz) of FACE-1 in DMSO-$d_6$ (*) at ambient temperature.

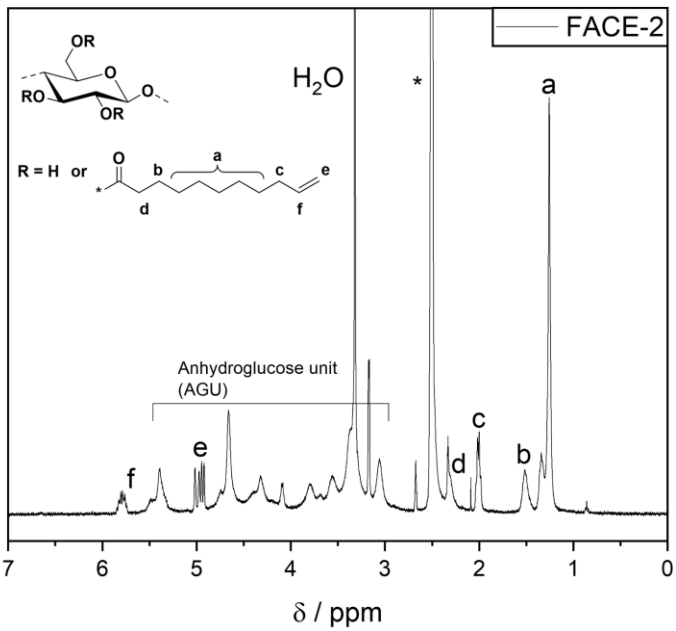


**Figure S3.** $^{1}$H NMR (400 MHz) of FACE-2 in DMSO-$d_6$ (*) at ambient temperature.

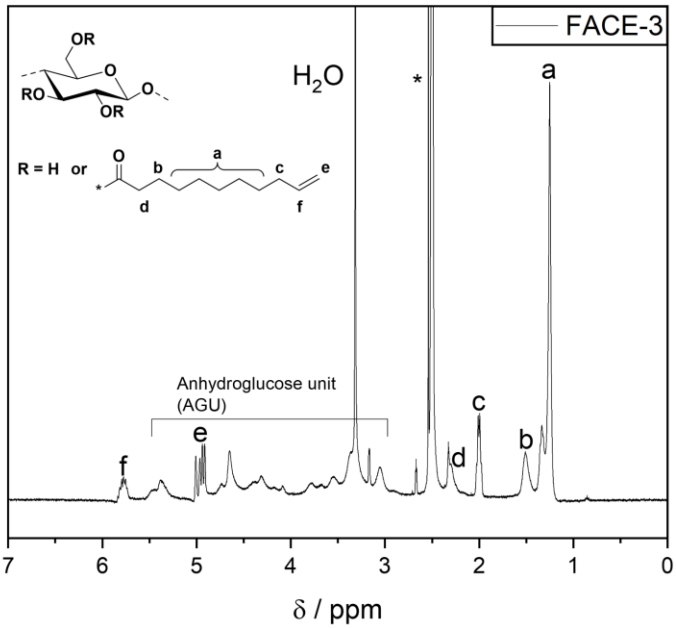


**Figure S4.** $^{1}$H NMR (400 MHz) of FACE-3 in DMSO-$d_6$ (*) at ambient temperature.

## 2.3 $^{31}$P NMR for DS determination

The DS of the synthesized FACEs were calculated as previously published by Kilpeläinen et al.[1] according to Equation S3 and S4:

$$\mathrm{DS} = \mathrm{DS_{max}} \times \frac{\frac{1}{\mathrm{OH_S}} - \frac{1}{\mathrm{OH_C}}}{\mathrm{M_S} + \frac{1}{\mathrm{OH_S}} - 1} \qquad \text{(S3)}$$

$$\mathrm{OH_s} = \frac{\mathrm{C_{Is}\ x\ V_{IS}\ x\ I_R}}{1000000\ \mathrm{x\ m_s}} \qquad \text{(S4)}$$

$DS_{max}$: highest achievable DS value (3 for unsubstituted cellulose)
$M_S$ : molecular weight of the substituent (without the linking oxygen atom between the substituent and the cellulose backbone; 167.27 g mol$^{-1}$)
$c_{IS}$: concentration of the internal standard (mmol L-$^{1}$)
$V_{IS}$: volume of the employed internal standard (μL)
$I_R$: integration ratio of remaining functionalized cellulose hydroxyl groups against internal standard
$m_S$ : sample mass (mg)
$OH_S$: free hydroxyl groups per weight unit of substrate (mol g$^{-1}$)
$OH_C$: free hydroxyl groups per weight unit of cellulose ($OH_C = DS_{max}/M_{AGU}$ = 3 /162.14 g mol$^{-1}$)

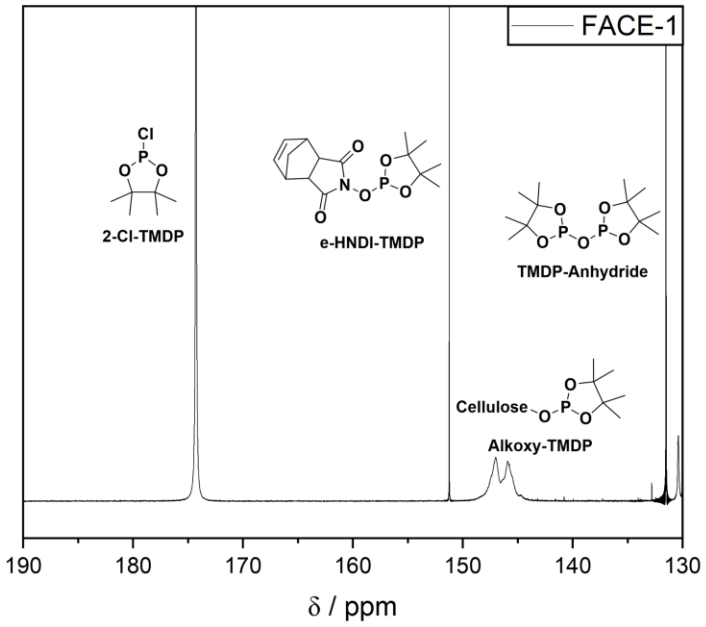


**Figure S5.** $^{31}P$ NMR of phosphorylated FACE-1 in $CDCl_3$ at ambient temperature.

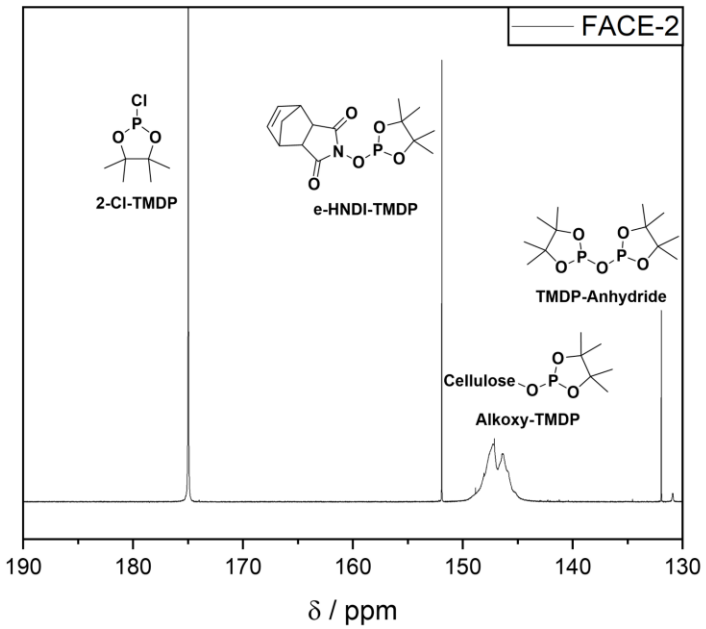


**Figure S6.** $^{31}P$ NMR of phosphorylated FACE-2 in $CDCl_3$ at ambient temperature.

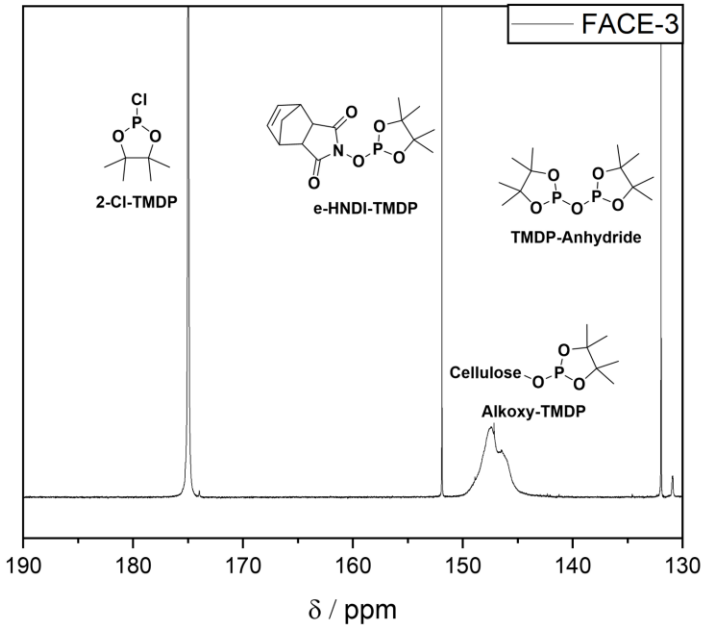


**Figure S7.** $^{31}$P NMR of phosphorylated FACE-3 in $CDCl_3$ at ambient temperature.

# 3 Material properties of FACEs

## 3.1 TGA traces of FACEs

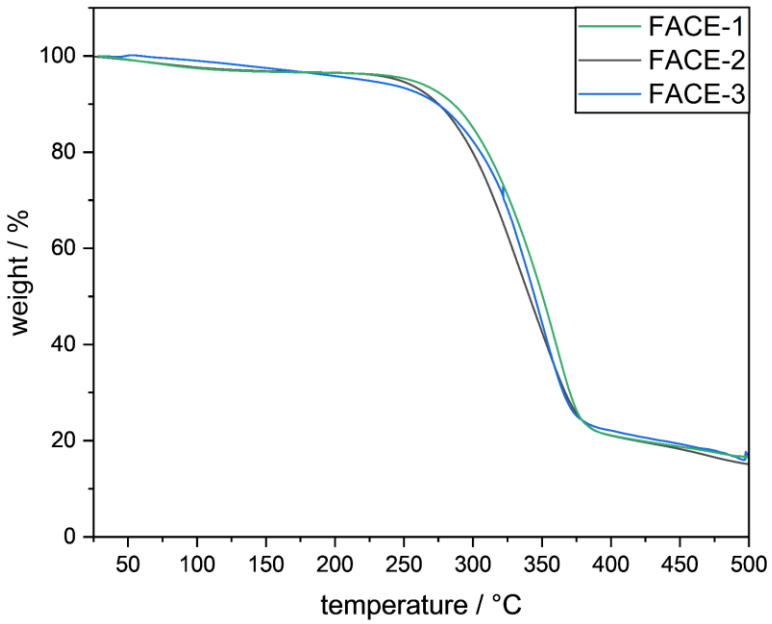


**Figure S8.** Thermogravimetric analysis (TGA) of FACE-1 (green line), FACE-2 (black line), and FACE-3 (blue line) from 25 to 500 °C with a heating rate of 10 K min$^{-1}$ under a nitrogen flow.

## 3.2 DSC traces of FACEs

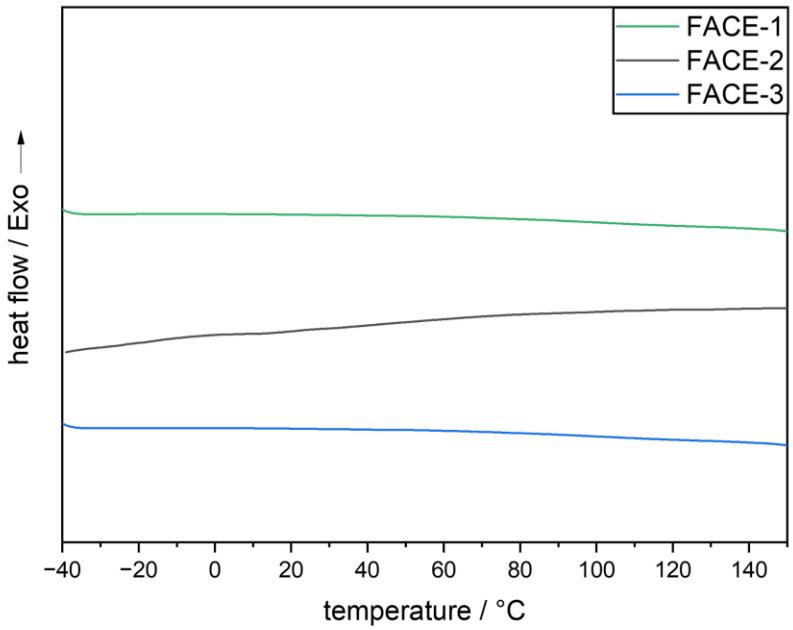


**Figure S9.** DSC studies (second heating run) of FACE-1 (green line), FACE-2 (black line), and FACE-3 (blue line) from −40 to 220 °C with a heating rate of 20 °C $min^{-1}$under a nitrogen flow.

## 3.3 SEC traces of FACEs

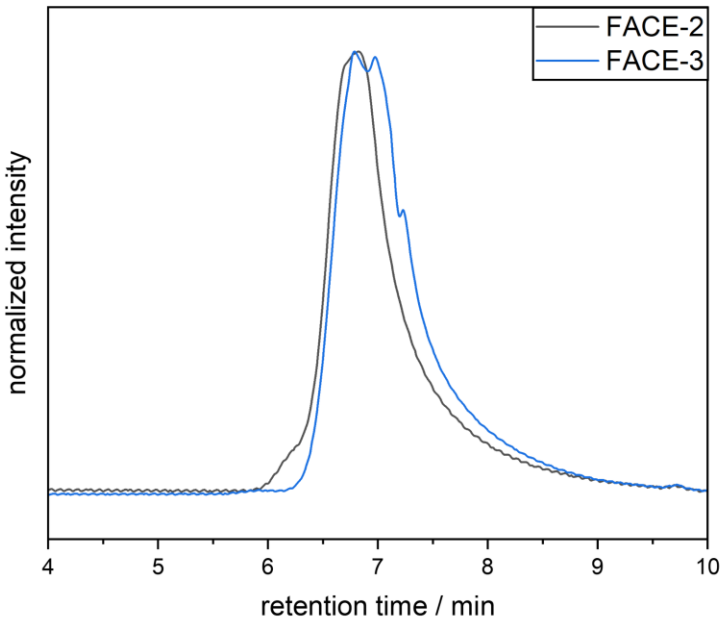


**Figure 10.** SEC traces of FACE-2 (black line) and FACE-3 (blue line) in in HFIP+ 0.1%w/v KTFA.

## 4 DSC traces of FACE-XS (X = 1, 2 or 3)

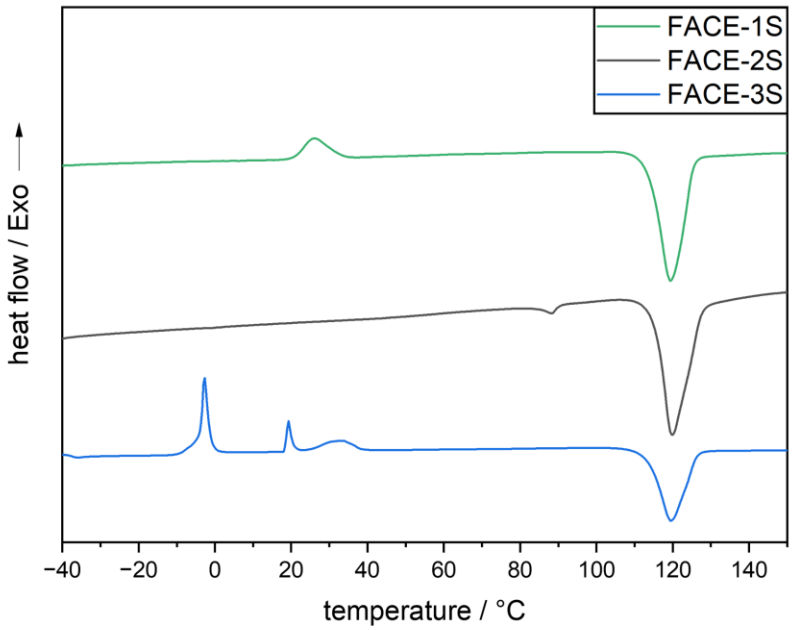


**Figure S11.** DSC studies (second heating run) of FACE-1S (green line), FACE-2S (black line), and FACE-3S (blue line) from −40 to 220 °C with a heating rate of 20 °C $min^{-1}$ under a nitrogen flow.

## 5 DSC traces of FACE-XS-washed (X = 1, 2 or 3)

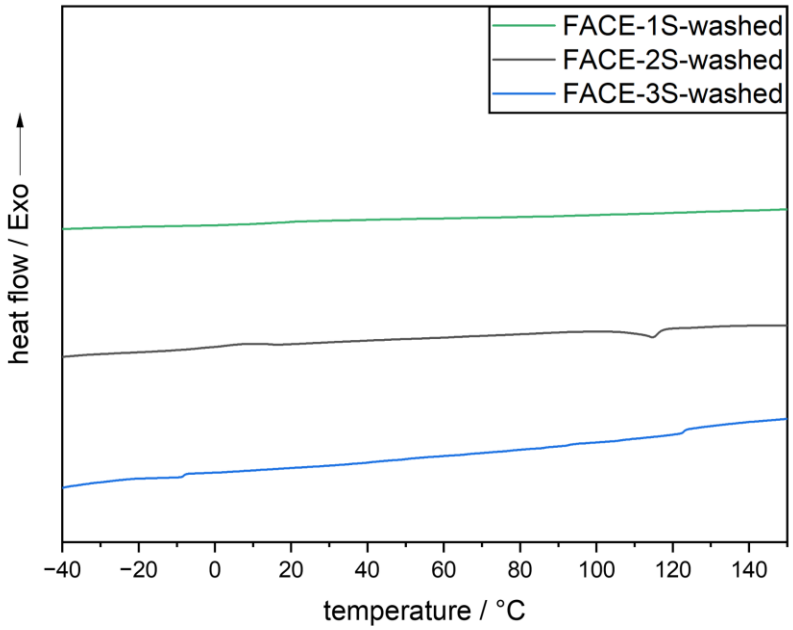


**Figure S12.** DSC studies (second heating run) of FACE-1S-washed (green line), FACE-2S-washed (black line), and FACE-3S-washed (blue line) from −40 to 220 °C with a heating rate of 20 °C $min^{-1}$under a nitrogen flow.

## 6 EDX spectra of FACE-XS-washed (X = 1, 2 or 3)

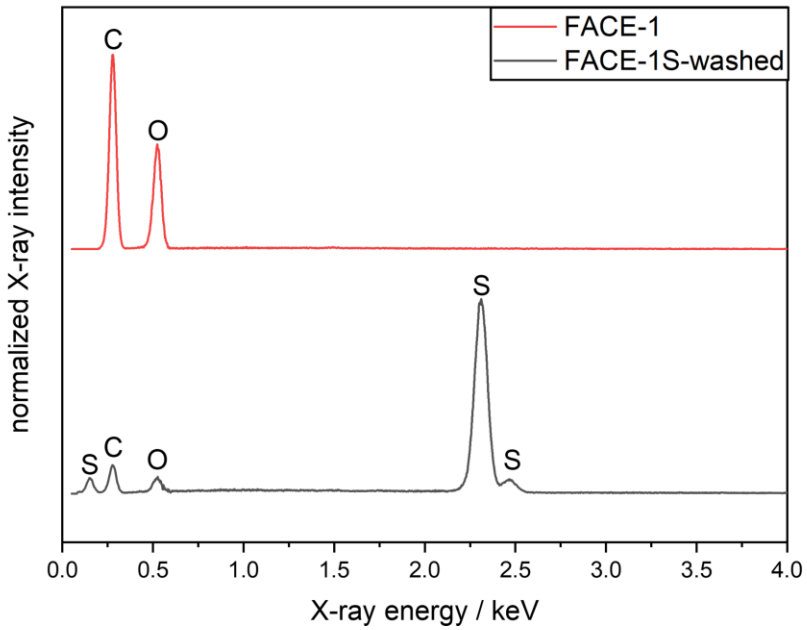


**Figure S13.** EDX spectra of FACE-1 (red line) and FACE-1S-washed (black line).

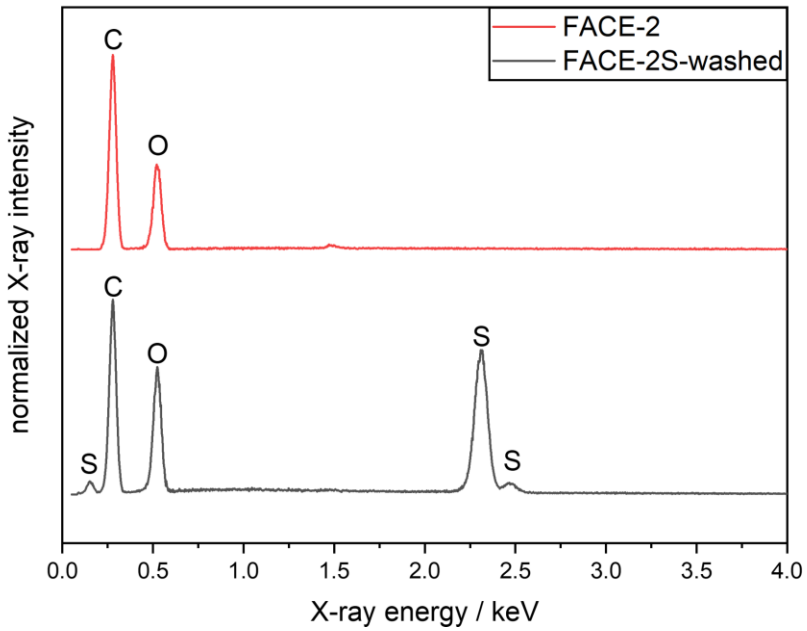


**Figure S14.** EDX spectra of FACE-2 (red line) and FACE-2S-washed (black line).

# 7 SEM images of FACE-XS-washed (X = 1, 2 or 3)

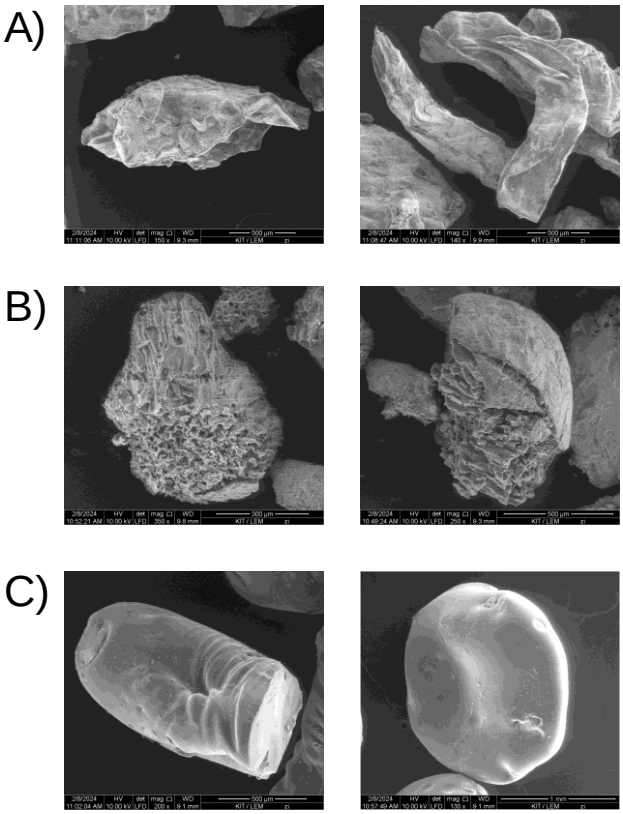


**Figure S15.** Additional SEM images of FACE-1S-washed (A), FACE-2S-washed (B), and FACE-3S-washed (C).

# 8 Mercury sorption studies

| Material | $C_i$ (µg $L^{-1}$) | $C_t$ (µg $L^{-1}$)[a] |
|---|---|---|
| FACE-1S | 1000 | 41.8 |
| FACE-2S | 1000 | 211 |
| FACE-3S | 1000 | 291 |

[a]after stirring for 24 h at ambient temperature.

# 9 Backscattering electron (BSE) images

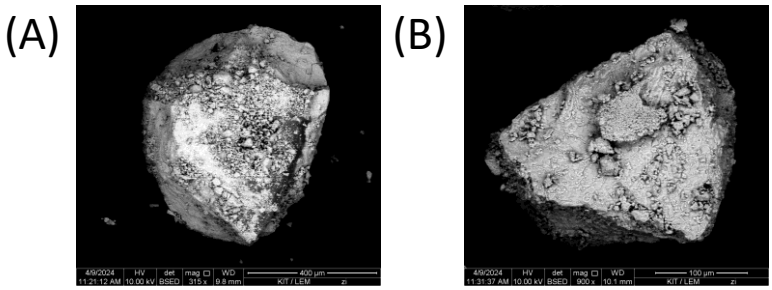


**Figure S16.** BSE-SEM images of FACE-2S (A) and FACE-3S (B).

References:

1 A. W. T. King, J. Jalomäki, M. Granström, D. S. Argyropoulos, S. Heikkinen and I. Kilpeläinen, *Analytical Methods*, 2010, **2**, 1499-1505.