X-ray Raman Scattering for Bulk Chemical and Structural Insight into Green Carbon

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XRS spectroscopy Data Treatment:

Data Treatment for XRS data collected at 20ID APS (Argonne, USA) using LERIX module of the XRStools software. XRStools is software written and maintained on GitLab by Alessandro Mirone and Christoph Sahle of the European Synchrotron (https://gitlab.esrf.fr/mirone/XRStools). LERIX is an additional module written by LJRH in order to treat data collected at ID-20 of the APS: https://github.com/LJRH/XRStools

Gaussian Fitting:

Gaussian fitting was performed using the LMFIT python software, which can be found at https://lmfit.github.io/lmfit-py/index.html. The model was based on three major functions taken from Stohr *et.al* [1,2]:

1) Gaussian Function

$$G = f(x;A,\mu,\sigma) = \frac{A}{\sigma\sqrt{2\pi}} e^{\left[-\frac{(x-\mu)^2}{2\sigma^2}\right]}$$

Parameters:

- Independent variable - x – Energy Loss (eV)
- A – Amplitude, μ – centroid position, σ – sigma (width)

2) Asymmetric Gaussian Function

$$AG = f(x;A,\mu,\sigma) = \frac{A}{(x*m-b)\sqrt{2\pi}} e^{\left[-\frac{(x-\mu)^2}{2(x*m-b)^2}\right]}$$

Parameters:

- Independent variable - x – Energy Loss (eV)
- A – Amplitude, μ – centroid position, $m = 0.57$, $b = 166$

3) Step Function

$$SF = f(x;A,\mu,\sigma) = \frac{A \cdot \left(1 + \mathrm{erf}\left(\frac{x-\mu}{\sigma}\right)\right)}{2 \cdot (e^{-d \cdot (x-\mu-\sigma)})}$$

Parameters:

- Independent variable - x – Energy Loss (eV)
- A – Amplitude, μ – centroid position, $d = 0.005$

Gaussian Fitting Statistics:

Table S1: Statistics for the Gaussian fitting performed collected for BC-Oak-450 and BC-Oak-650 XRS spectroscopy data.

Parameter	BC-Oak-450	BC-Oak-650
Fitting Method	Least Squares	Least Squares
Function Evaluations	121	91
Data Points	188	188
Chi-Square	0.116	0.237
Reduced Chi-Square	6.58×10^{-4}	1.32×10^{-3}

NEXAFS Spectra Homogeneity:

Figure S1 (below), shows averaged NEXAFS spectra for sub-regions (blue, green, purple) within the region averaged (red) to produce NEXAFS spectra for (a) BC-Oak-650 and (b) HTC-Oak-250 within the main text.

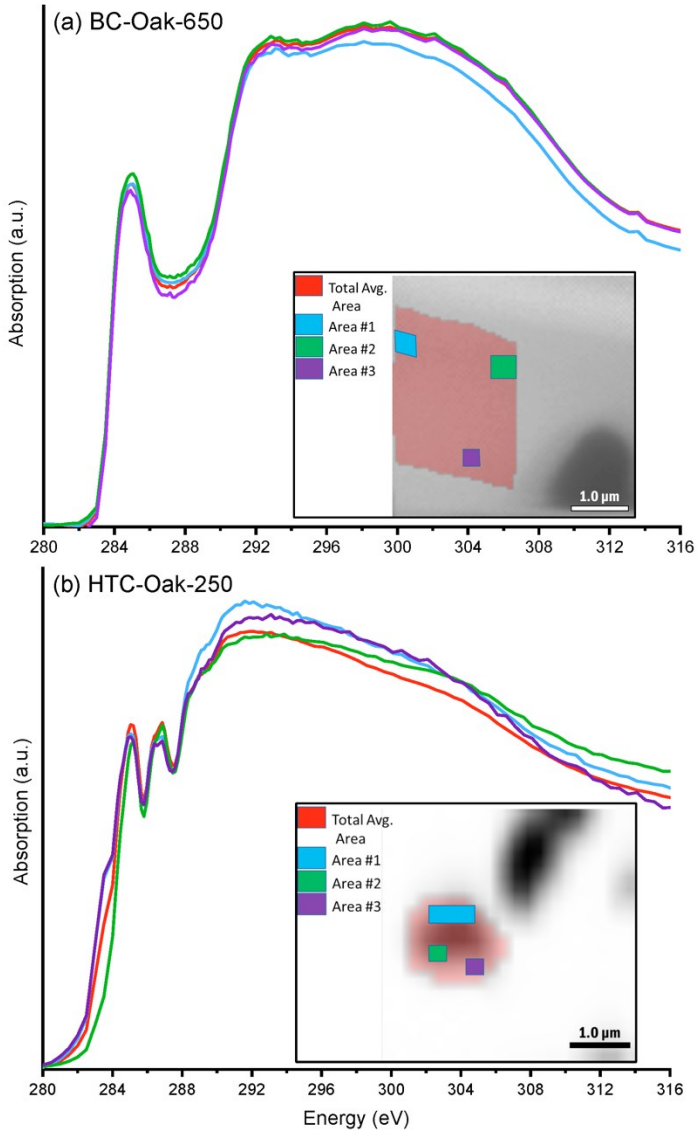

Figure S1: Scanning transmission X-ray microscopy images of (a) BC-Oak-650 and (b) HTC-Oak-650 with three areas and their corresponding average spectra highlighted in blue, green and purple. The average NEXAFS spectra used in this study and its corresponding selected area are shown in red.

***q*-Dependence Calculations:**

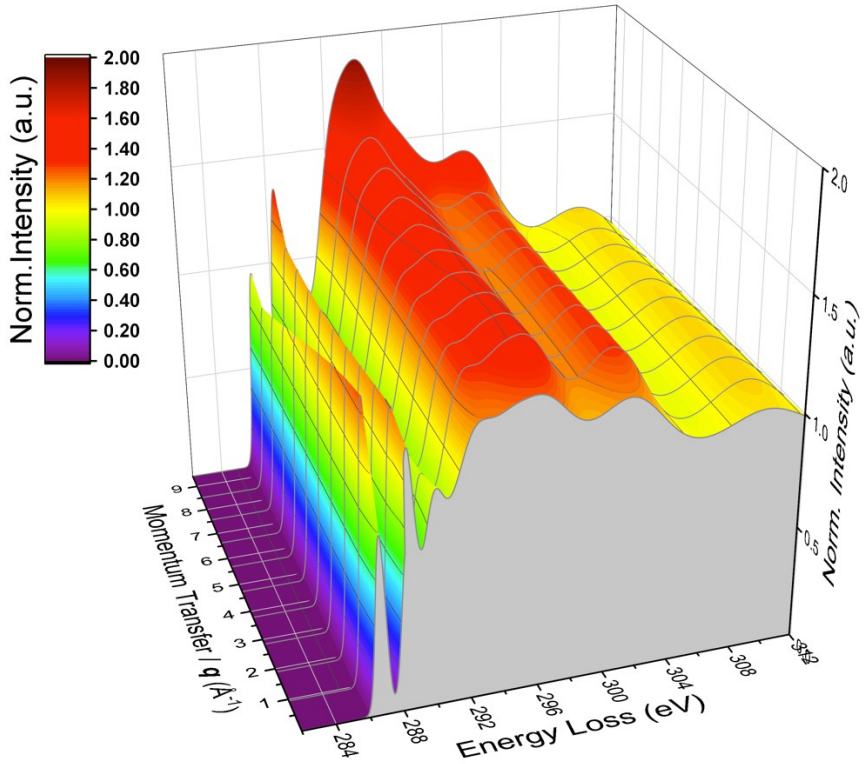

Figure S2: Momentum transfer dependent DFT calculations for levulinic acid.

Figure S2 shows the ***q***-dependence calculations for the levulinic acid standard. Calculations were performed over a range of momentum transfer values (0.02, 0.94, 1.89, 2.83, 3.78, 4.72, 5.67, 6.61, 7.56, 8.50, 9.45 $Å^{-1}$) using the method described in the main text (section 2.6.2.).

Calculated Spectra:

Calculated Spectra can be found in the attached .zip file "calculated_spectra.zip" or on LJRHs github repository https://github.com/LJRH/HTC-Structures.

References:

[1] J. Stöhr, NEXAFS Spectroscopy, 1st ed., Springer Berlin Heidelberg, Berlin, Heidelberg, 1992. https://doi.org/10.1007/978-3-662-02853-7.

[2] D.A. Outka, J. Stöhr, Curve fitting analysis of near-edge core excitation spectra of free, adsorbed, and polymeric molecules, J. Chem. Phys. 88 (1988) 3539–3554. https://doi.org/10.1063/1.453902.