This supplementary file includes the following:

A. Materials and Methods

B. Carbonization parameters

C. EDX

D. XPS

E. Raman

F. XRD

G. Ball-milling

A. Materials and Methods

- **Morphological characterization**

SEM

The morphology of the blue shark-based and commercial chondroitin sulfate-based carbon samples was assessed by scanning electron microscopy (SEM) associated with energy-dispersive X-ray (EDX, Oxford detector-analyzer) analysis (Hitachi SU 8230, Tokyo, Japan).

Transmission electron microscopy (TEM) (Hitachi model H8100, with LaB6 filament and an accelerating voltage of 200 kV) was also used.

Raman

Raman spectra was recorded with Raman spectrometer Ramos PA532 Ostec, using a 532 nm excitation wavelength. In addition, the crystallite size of the sp^2 lattice (La) was calculated according to the equation presented by Pimenta *et al.* [1]:

$$L_a = 2.4 \times 10^{-10} \lambda_L^4 \left(\frac{I_D}{I_G}\right)^{-1} \quad \textbf{Eq. 1}$$

Where λ_L is the excitation wavelength, I_D/I_G is the degree of graphitization of the carbon materials, related to the intensity of the D and G bands.

XRD

X-ray diffraction measurements of all samples were carried out in the 5 – 90° range on a Rigaku Smart Lab X-Ray Diffractometer (Rigaku Corporation, Tokyo, Japan) using Cu Kα radiation (λ=0.154060 nm) operating at room temperature. The phase was identified using the International Center for Diffraction Data – ICDD (PDF-2) database.

UHR-STEM

Scanning Transmission Electron Microscopy (STEM) system equipped with EDX Oxford detector-analyzer was used to perform sample analysis. For the analysis, the samples in powder form were dispersed in ethanol using a probe type ultrasonic homogenizer and deposited on standard Cu TEM grids with Formvar and Lacey Carbon polymeric films. Structural studies were performed and SEM, Z Contrast (HAADF – High Angle Annular Dark Field) and transmission (Bright Field) images were obtained.

STEM was used to perform the ultrahigh-resolution images (UHR-STEM). This system is able to acquire co-localized images with three different detectors. The SEM image is collected with secondary electron detector and provides topographic information about the sample, the Z – Contrast (atomic mass contrast) image is collected with annular dark field detector (HAADF – High Angle Annular Dark Field Mode) providing the compositional contrast of the specimen and

STEM images (transmitted electrons signal) are collected from the bright field detector providing internal structure information of the analyzed sample. The STEM is a cold field emission microscope and for images acquisition was operated at 200kV acceleration voltage leading to 2.51 pm electrons wavelength and 10 μA emission current.

XPS

The chemical composition of the obtained materials was evaluated by X-ray Photoelectron Spectroscopy (XPS) system (Kratos Analytical Ltd, Manchester, UK) with a 15 kV X-ray source. Attenuated total reflectance infrared Fourier Transform Infrared spectroscopy (ATR-FTIR) measurements were performed using a Bruker FT−IR System Tensor 27 spectrophotometer (Massachusetts, USA) from 4000 to 600 cm^{-1}.

ATR-FTIR

Attenuated total reflectance infrared (ATR-FTIR, Bruker FT-IR System Tensor 27 spectrophotometer, Massachusetts, USA) investigations were achieved in the range of 4000 to 600 cm^{-1}.

- **Electrochemical studies**
  - Preparation of DES electrolyte

Choline chloride (ChCl, Sigma-Aldrich, 99%) was dried at 60 °C in the oven overnight before use, and ethylene glycol (Sigma-Aldrich, 99%) was used as received. The eutectic mixture was prepared by stirring and heating at 60 °C the ChCl with ethylene glycol, as HBD, in the molar ratio of 1:2 until a homogeneous and clear liquid was formed. This liquid will be referred to from now on as ethaline.

  - Aqueous electrolytes

Alongside the DES preparation, two aqueous electrolytes (1 M) were also prepared, H_2SO_4 (95%, Sigma-Aldrich) and KOH (pellets, 98%, Sigma-Aldrich).

  - Glassy Carbon (GC) electrode preparation

The immobilization of the carbon material on the electrode was successfully achieved using the method previously described by Brandão *et al.* [2]. Briefly, a dispersion of 5 mg of carbon in 950 μL N, N-Dimethylformamide (DMF, 99.8%, Sigma Aldrich) and 10 μL Nafion® 117 (~5 %, Sigma Aldrich) was prepared. Ultrasonication was used for 2 h to obtain a homogeneous dispersion of the material. The weight of the carbon-coated on the GC electrode was obtained as an average of several measurements, considering that the material and the Nafion dispersion in DMF are

homogeneous. The suspension was drop cast on the GC electrode surface using a micropipette. It was dried at room temperature before combining with the electrochemical cell and immersed in the eutectic mixture.

- Electrochemical characterization of half-cell setup

The electrochemical measurements were performed in a three-electrode cell using a computer-controlled VSP-300 multichannel potentiostat from BioLogic, controlled with EC-Lab V11.26 software.

The three-electrode configuration consisted of a GC electrode as a working electrode, a graphite counter electrode, and a silver wire pseudo-reference electrode. The working electrode was polished following the method previously presented by several authors [2–4].

The experiments were performed at 30 °C for both eutectic and aqueous electrolytes. Voltametric experiments were carried out at 50 mV s^{-1}, starting at 0 V towards the positive side to 1 V.

Galvanostatic charge/discharge curves were obtained at a current density of 1 A g^{-1}. The specific capacitance in a three-electrode configuration was calculated from the galvanostatic discharge curves using **Equation 2**, according to Stoller *et al.* [5]:

$$C = \frac{I\,\Delta t}{m\,\Delta V} \qquad \textbf{Eq. 2}$$

Where I is the discharge current (A), Δt is the discharge time (s), ΔV is the potential window (V), and m is the weight of the carbon material in the electrode (g).

The differential capacitance (F cm^{-2}) can be obtained from the Electrochemical Impedance Spectroscopy (EIS) measurements at fixed potentials from 0 V to 1 V.

The extraction of the capacitance can be obtained from the Z″ – Z′ plot, from the parameters of CPE (Y_0, n) obtained from the adjustment of the equivalent circuit (R-Q), according to **Equation 3**, and (R-R-Q), according to **Equation 4**, in which the presence of a slight curvature on the impedance Nyquist plot, using **Equation 3** and **Equation 4** [6]:

$$C = Y_0^{1/n} \left(\frac{1}{R_S}\right)^{(n-1)/n} \qquad \textbf{Eq. 3}$$

$$C = Y_0^{1/n} \left(\frac{1}{R_S} + \frac{1}{R_f}\right)^{(n-1)/n} \qquad \textbf{Eq. 4}$$

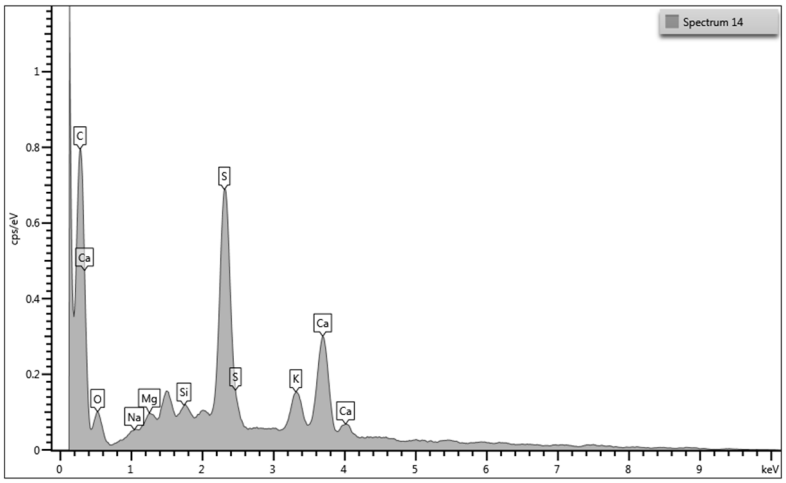
The quality of the fitting was judged by the value of χ^2 ($<10^{-3}$).

B. Carbonization parameters

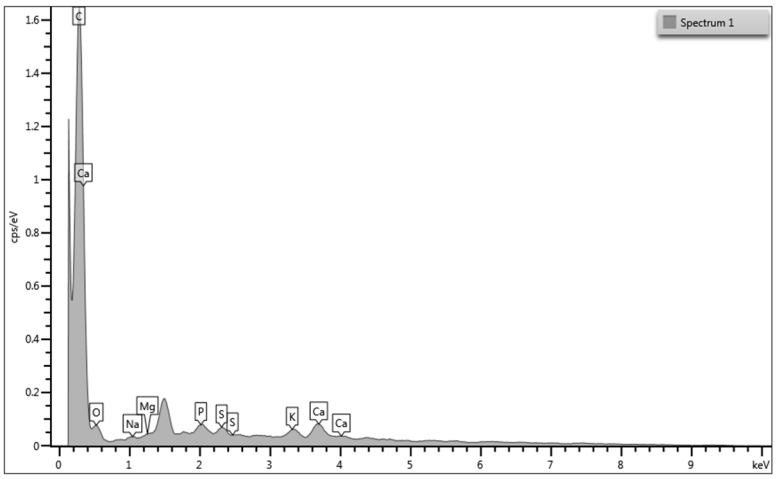
Table S1 Carbonization time and temperature for blue shark (chondroitin sulfate and gelatine) and commercial chondroitin sulfate with associated S_{BET}, capacitance, and % retention.

Carbon precursor	Temperature	Carbonization time	S_{BET} ($m^2\ g^{-1}$)	C ($F\ g^{-1}$) 1st cycle	% retention After 5000th cycle
Blue shark chondroitin sulfate	1000 °C	10 min	45.21	27	68
		30 min	77.56	31	73
		1 h	**135.24**	**40**	**71**
		2 h	134.89	38	71
		3 h	135.01	35	72
		4 h	90.77	26	70
	900 °C	1 h	70.11	34	64
	500 °C		23.12	16	68
Blue shark gelatine	1000 °C	10 min	13.54	5	65
		30 min	22.44	5	70
		1 h	**30.32**	**7**	**71**
		2 h	26.53	4	72
		3 h	27.54	4	70
		4 h	21.12	5	68
	900 °C	1 h	21.14	3	68
	500 °C		16.55	3	69
Chondroitin sulfate commercial	1000 °C	10 min	43.21	20	79
		30 min	64.18	22	80
		1 h	**76.11**	**25**	**86**
		2 h	76.03	22	81
		3 h	75.12	23	83
		4 h	59.44	23	80
	900 °C	1 h	67.11	24	75
	500 °C		23.59	21	73

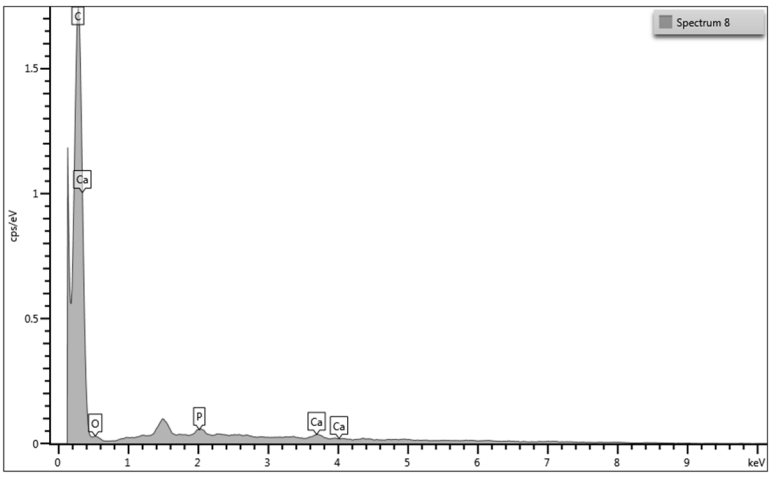
C. EDX



(a)



(b)



(c)

Figure S1 EDX analysis of (a) Blue shark chondroitin sulfate, (b) commercial chondroitin sulfate, and (c) blue shark gelatine.

D. XPS

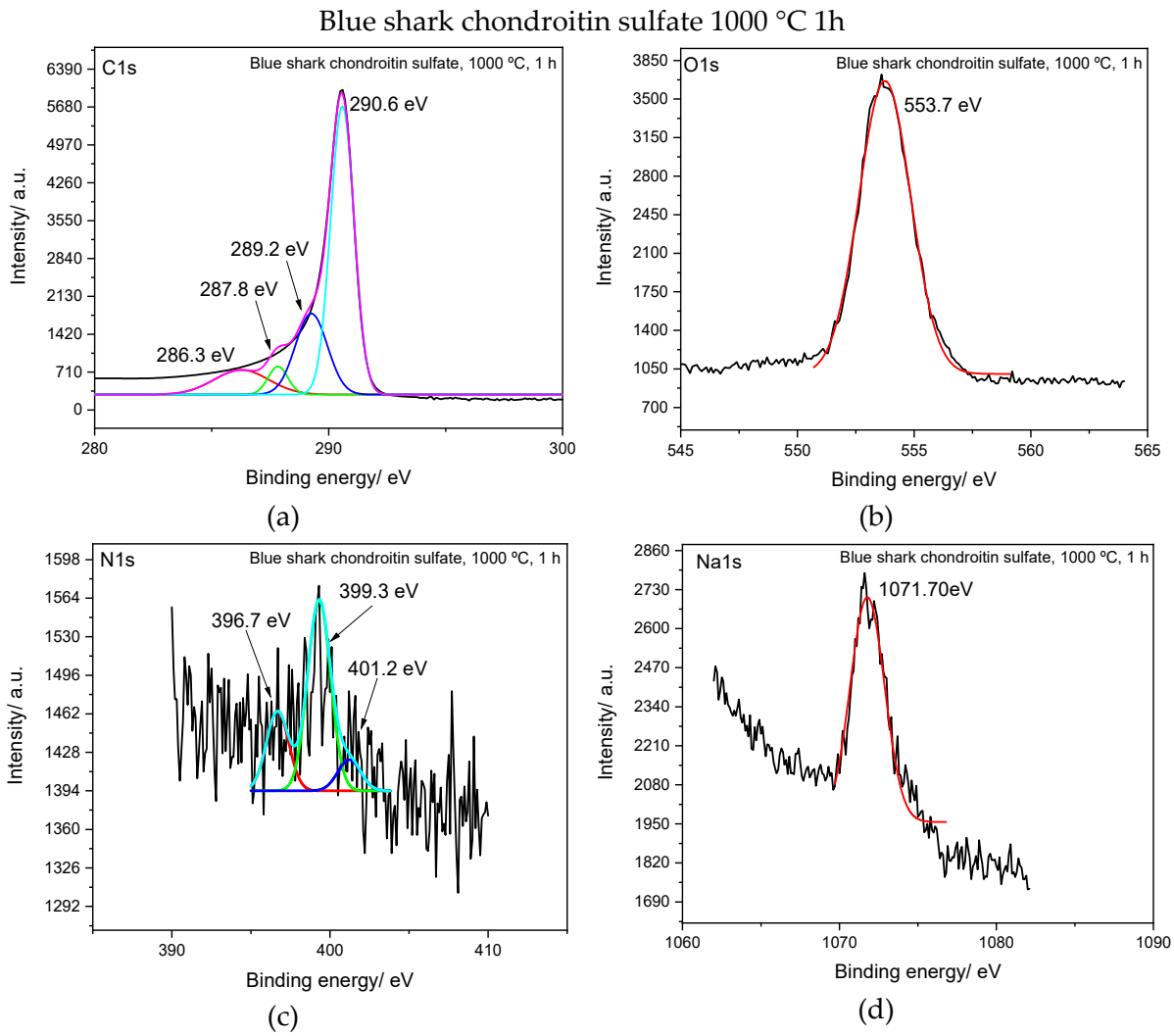


Figure S2 Deconvolution of the peaks of the XPS survey spectra for blue shark chondroitin sulfate carbonized for 1 h at 1000 °C: C1s (a), O1s (b), N1s (c) and Na1s (d).

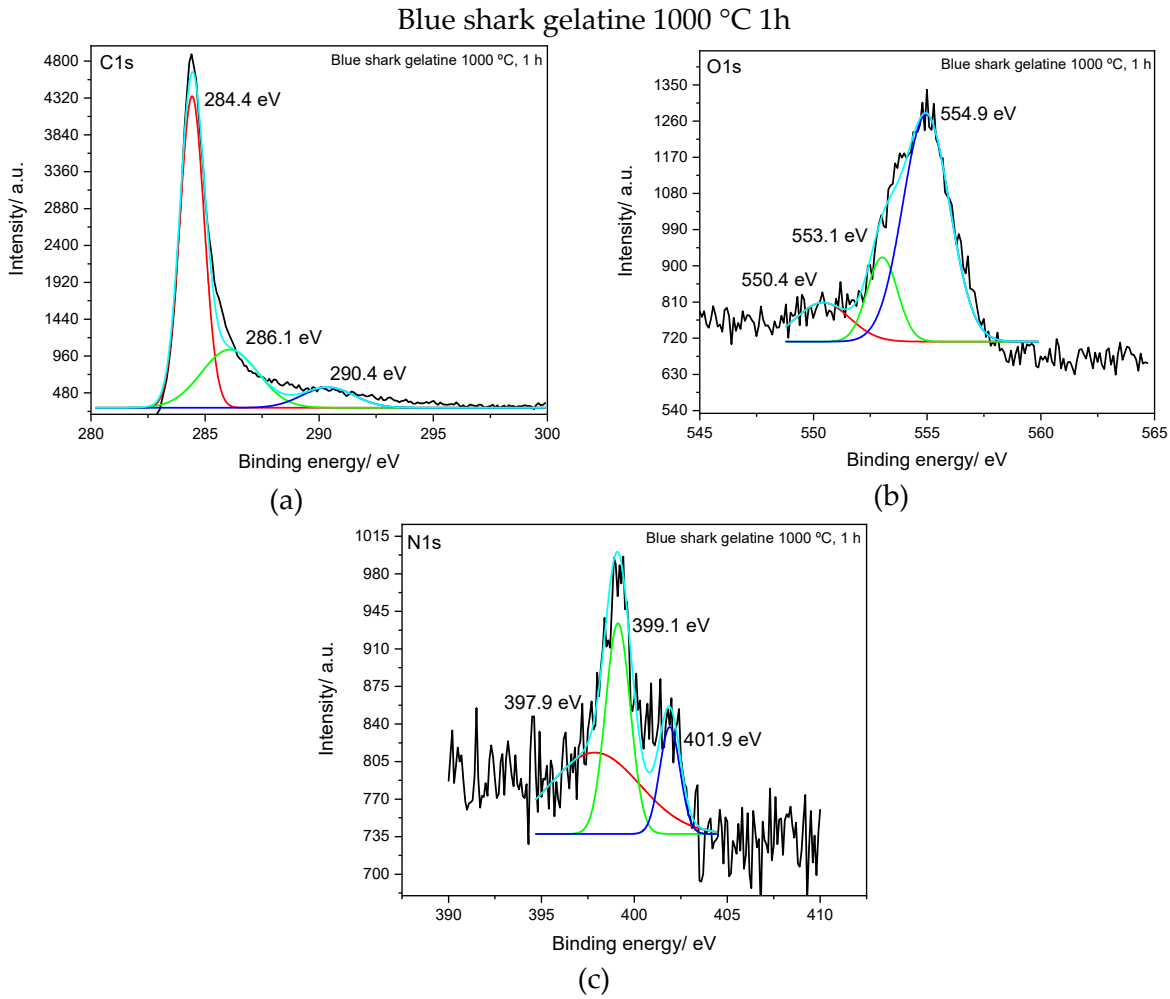


Figure S3 Deconvolution of the peaks of the XPS survey spectra for blue shark gelatine carbonized for 1 h at 1000 °C: C1s (a), O1s (b), and N1s (c).

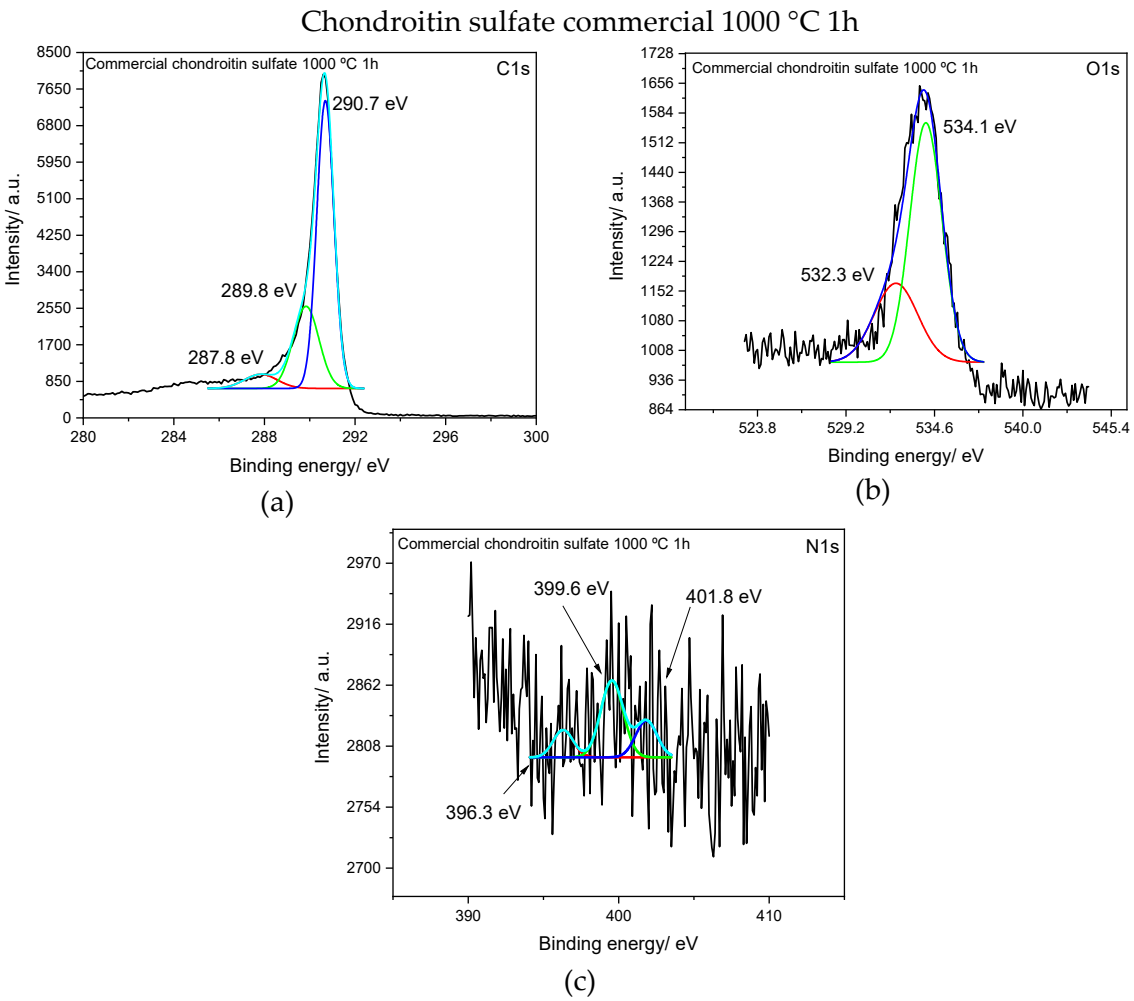


Figure S4 Deconvolution of the peaks of the XPS survey spectra for commercial chondroitin sulfate carbonized for 1 h at 1000 °C: C1s (a), O1s (b), and N1s (c).

E. Raman Analysis

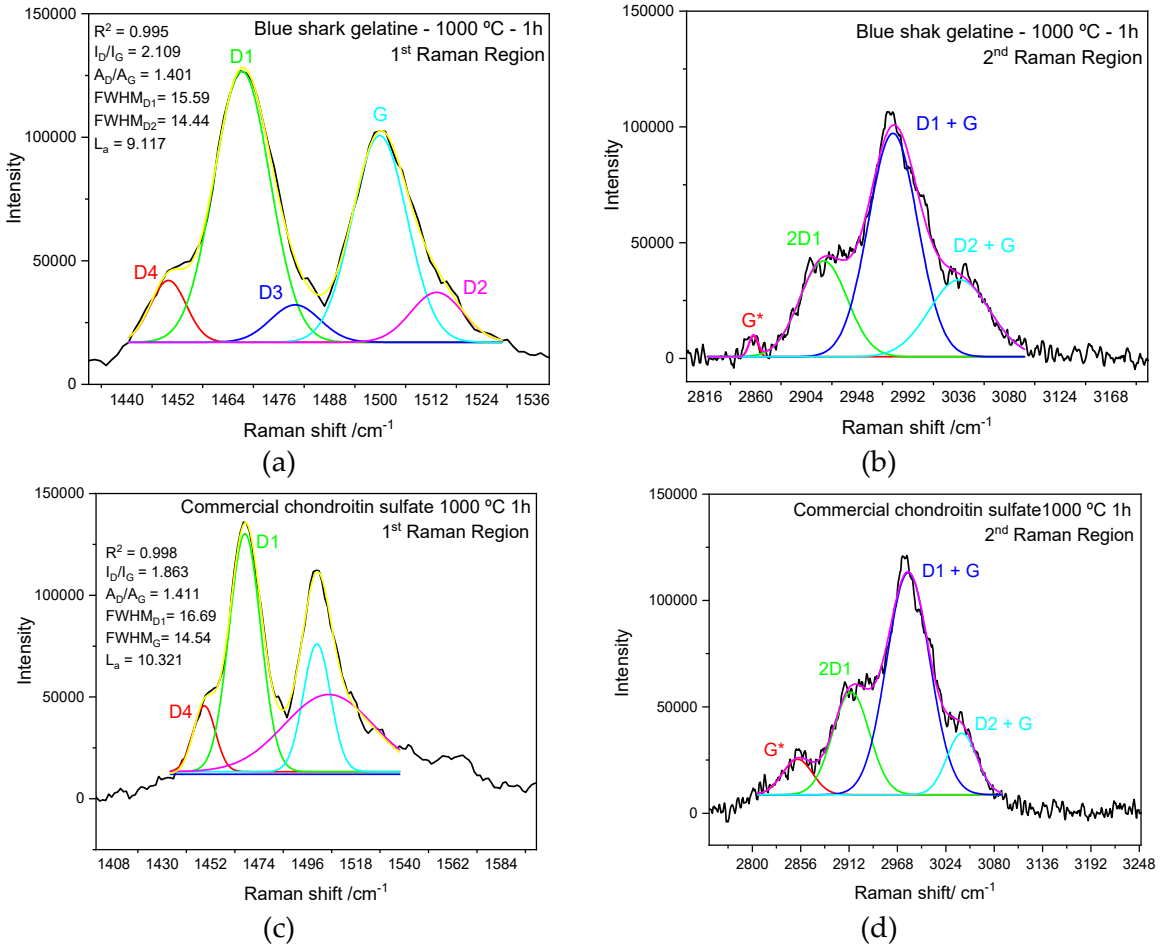


Figure S5 1st Raman region (**a-c**) and 2nd Raman region (**b-d**) of the Raman spectra of the blue shark gelatine and commercial chondroitin sulfate-based carbons, respectively.

F. XRD

Table S2 Peaks extracted from the diffraction patterns in Figure 10.

		2θ (deg)	**d (Å)**	FWHM (deg)	**Int. I (counts deg)**	**Int. W (deg)**
Blue shark chondroitin sulfate	1	8.35	10.58927	7.1474	2425.5	11.0841
	2	31.8	2.81403	2.9237	154.33	5.7018
	3	45.56	1.99107	7.1474	508.69	2.8254
Blue shark gelatin	1	8.55	10.34201	0.1	139.18	0.0002
	2	25.32	3.51758	0.1	72.5	0.3797
	3	43.86	2.06422	0.1	8.38	1.8838
Chondroitin sulfate commercial	1	8.21	10.76952	10.76952	1889.76	12.6699
	2	25.41	3.50532	3.50532	1163.25	14.8233
	3	44.6	2.03167	2.03167	140.79	7.7984

G. Ball milling

Table S3 S_{BET}, V_{micro}, V_{meso}, and D_p parameters for blue shark chondroitin sulfate-based carbon at different ball milling times.

Blue shark chondroitin sulfate-based carbon	S_{BET}	V_{micro}	V_{meso}	V_{total}	D_p
Ball milling time (h)	($m^2\ g^{-1}$)	($cm^3\ g^{-1}$)	($cm^3\ g^{-1}$)	($cm^3\ g^{-1}$)	(nm)
0	135.24	0.023	0.044	0.067	1.44
0.25	144.11	0.029	0.046	0.075	1.44
0.5	156.54	0.035	0.051	0.086	1.46
1	171.99	0.037	0.055	0.092	1.47
2	199.45	0.04	0.052	0.092	1.49
3	**260.12**	**0.044**	**0.052**	**0.096**	**1.55**
4	244.12	0.043	0.05	0.093	1.52
5	214.1	0.044	0.049	0.093	1.53

Table S4 S_{BET}, V_{micro}, V_{meso}, and D_p parameters for blue shark gelatine-based carbon at different ball milling times.

Blue shark gelatine-based carbon	S_{BET}	V_{micro}	V_{meso}	V_{total}	D_p
Ball milling time (h)	($m^2\ g^{-1}$)	($cm^3\ g^{-1}$)	($cm^3\ g^{-1}$)	($cm^3\ g^{-1}$)	(nm)
0	30.32	0.011	0.019	0.03	0.87
0.25	**33.56**	**0.022**	**0.021**	**0.043**	**0.91**
0.5	31.25	0.022	0.022	0.044	0.89
1	28.44	0.021	0.024	0.045	0.89
2	25.11	0.02	0.019	0.039	0.86
3	26.01	0.021	0.02	0.041	0.81
4	25.66	0.019	0.019	0.038	0.82
5	24.88	0.015	0.021	0.036	0.83

Table S5 S_{BET}, V_{micro}, V_{meso}, and D_p parameters for commercial chondroitin sulfate-based carbon at different ball milling times.

Chondroitin sulfate commercial-based carbon	S_{BET}	V_{micro}	V_{meso}	V_{total}	D_p
Ball milling time (h)	($m^2\ g^{-1}$)	($cm^3\ g^{-1}$)	($cm^3\ g^{-1}$)	($cm^3\ g^{-1}$)	(nm)
0	76.11	0.019	0.024	0.043	1.11
0.25	89.45	0.021	0.026	0.047	1.22
0.5	96.12	0.022	0.031	0.053	1.25
1	**125.22**	**0.024**	**0.032**	**0.056**	**1.26**
2	119.45	0.026	0.032	0.058	1.27
3	110	0.025	0.035	0.06	1.23
4	98.11	0.024	0.029	0.053	1.22
5	77.1	0.026	0.031	0.057	1.17