# Microbubble synthesis of hybridised bacterial cellulose-gelatin separators for multifunctional supercapacitors

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**1. Calculation**

**Specific surface area**

The specific surface area estimated via the adsorption of methylene blue ($S_{MB}$)

$$S_{MB} = \frac{N_g \times a_{MB} \times N \times 10^{-20}}{MW_{MB}} \quad \text{(S1)}$$

Where N is Avogadro's number (6.02 x $10^{23}$ $mol^{-1}$), $a_{MB}$ is the surface area which one molecule of methylene blue is occupied, $N_g$ is the number of methylene blue molecules which are adsorbed, and $MW_{MB}$ is the molecular weight of methylene blue (319.85 g/mol).

**The crystallinity**

$$CI = \frac{A_c}{A_t} \times 100\% \quad \text{(S2)}$$

Where $A_t$ is the total area of the diffraction including crystalline and amorphous diffraction and $A_c$ is the area of the crystalline diffraction.

**The tensile test**

Stress ($\sigma_t$) is calculated at the maximum applied force ($F_{max}$) where the sample is broken normalised by the cross-sectional area of sample ($A_{CSA}$) through Equation S3. The strain (ε) is calculated from the change in length of the sample from the original length to the failure length (Δx) dependent on gauge length (L) which is used.

$$\sigma_t = \frac{F_{max}}{A_{CSA}} \tag{S3}$$

$$\epsilon = \frac{\Delta x}{L} \tag{S4}$$

Young modulus of elasticity (E) is obtained through the proportional of average stress by average strain of samples (S5).

$$E_t = \frac{\sigma}{\epsilon} \tag{S5}$$

**The specific capacitance of the as-fabricated symmetric supercapacitors**

Gravimetric capacitance ($C_{CV}$) of the symmetric devices, which is determined via the Cyclic voltammetry (CV) technique, is calculated by the equation below.

$$C_{cv} = \frac{4 \int idv}{\Delta V_{cv} \times m \times v} \tag{S6}$$

Where v is the scan rate, $\Delta V_{cv}$ is a voltage window (V), $\int$ idv is an integral under CV discharge curve, and m is the mass of electrode active materials.

Gravimetric capacitance ($C_{GCD}$) of the symmetric devices, which is investigated via the galvanostatic charge discharge (GCD) technique using S7 equation to calculate.

$$C_{GCD} = \frac{4 i_{GCD}}{(\frac{dV}{dt})} \tag{S7}$$

where $i_{GCD}$ is the applied current density (A/g), which is the applied current normalised by the mass of electrode active materials. $\frac{dV}{dt}$ is calculated from the slope obtained by fitting the discharge curve.

**Gravimetric energy and power density of as-fabricated symmetric supercapacitors**

Gravimetric energy density ($E_{cell}$) and gravimetric power density ($P_{cell}$) were calculated by S8 and S9 equations respectively.

$$E_{cell} = i \int V dt \tag{S8}$$

$$P_{cell} = \frac{E_{cell}}{t_{discharge}} \qquad (S9)$$

Where $t_{discharge}$ is the discharging time, i is the applied current density (A/g) and $\int Vdt$ is the area under discharge curve.

**The Coulombic efficiency**

Coulombic efficiency (*η*) is amount of discharging time divided by amount of charging time at the same applied current density:

$$\eta = \left(\frac{t_{discharge}}{t_{charge}}\right) \times 100\% \qquad (S10)$$

Where $t_{charge}$ is charging time and $t_{discharge}$ is the discharging time.

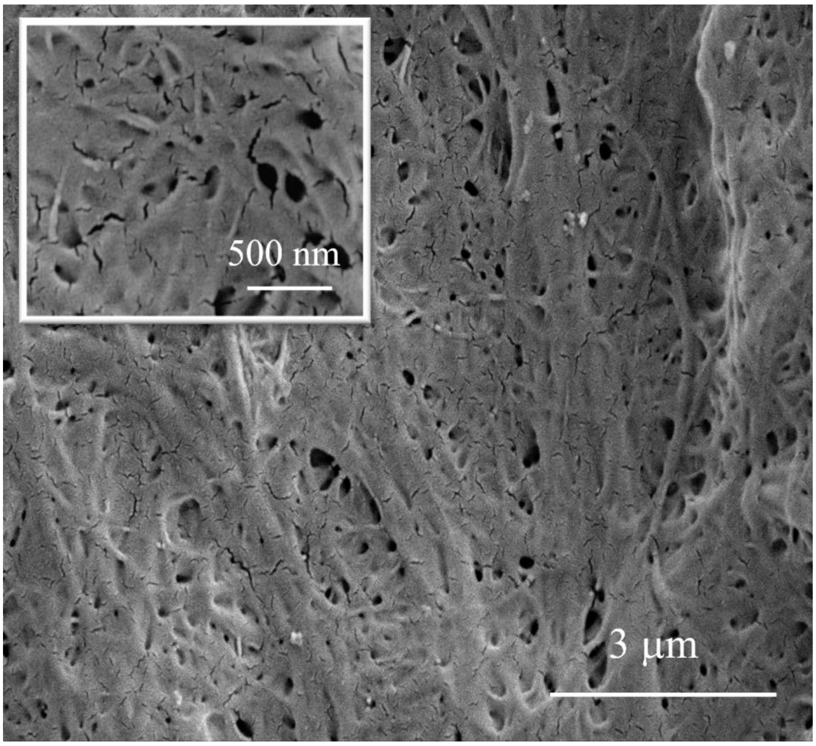


**Fig. S1** FE-SEM images of 2BC2GT_R200

**Table S1.** The specific surface areas determined by methylene blue adsorption ($S_{MB}$) using equation S1 and the amount of methylene blue molecules which are adsorbed to as-synthesised BC and hybridised BC-gelain separators ($N_g$).

| Sample | $N_g$ $10^{-3}$ kg/kg | $S_{MB}$ $10^{-3}$ $km^2/kg$ |
|---|---|---|
| 2BC | 0.00473 | 17.58 |
| 2BC1.5GT | 0.00483 | 17.95 |
| 2BC_R200 | 0.00508 | 18.87 |
| 2BC1.5GT_R200 | 0.00539 | 20.03 |

**Table S2.** Thickness of as-synthesised BC and hybridised BC-gelain separators.

| Sample | Thickness (μm) |
|---|---|
| 2BC | 15.3±0.9 |
| 2BC1.5GT | 22.6±2.7 |
| 2BC_R200 | 16.4±2.8 |
| 2BC1.5GT_R200 | 38.8±3.1 |

**Table S3.** Contact angle information.

| | | contact angle Θ [°] | | | | |
|---|---|---|---|---|---|---|
| **Sample** | **Drop** | **left** | **right** | **average (left and right)** | **SD** | **%RSD** |
| 2BC | 1 | 83.8 | 78.7 | 79.7 | 3.4 | 4.3 |
| | 2 | 75.7 | 76.0 | | | |
| | 3 | 80.9 | 82.9 | | | |
| 2BC1.5GT | 1 | 72.7 | 70.4 | 71.6 | 0.8 | 1.2 |
| | 2 | 71.9 | 72.0 | | | |
| | 3 | 70.9 | 71.3 | | | |
| 2BC_R200 | 1 | 52.8 | 51.6 | 53.1 | 1.0 | 2.0 |
| | 2 | 53.2 | 54.8 | | | |
| | 3 | 53.4 | 52.8 | | | |
| 2BC1.5GT_R200 | 1 | 27.7 | 26.9 | 27.5 | 0.3 | 1.1 |
| | 2 | 27.6 | 27.6 | | | |
| | 3 | 27.5 | 27.5 | | | |

**Mechanical performance**

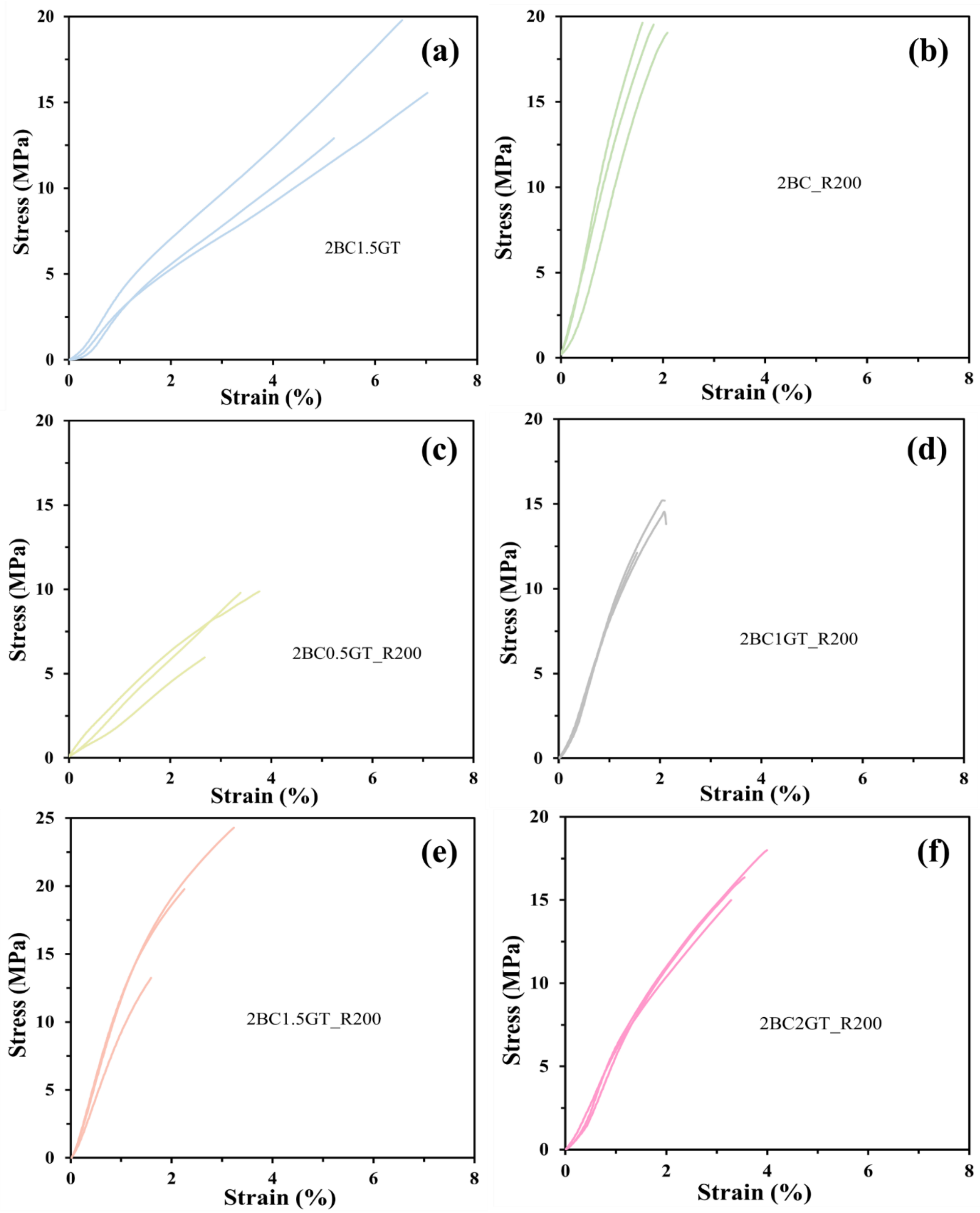


**Fig. S2** Stress–strain curves of (a) 2B1.5GT, (b) 2BC_R200, (c) 2BC0.5GT_R200, (d) 2BC1GT_R200, (e) 2BC1.5GT_R200 and (f) 2BC2GT_R200.

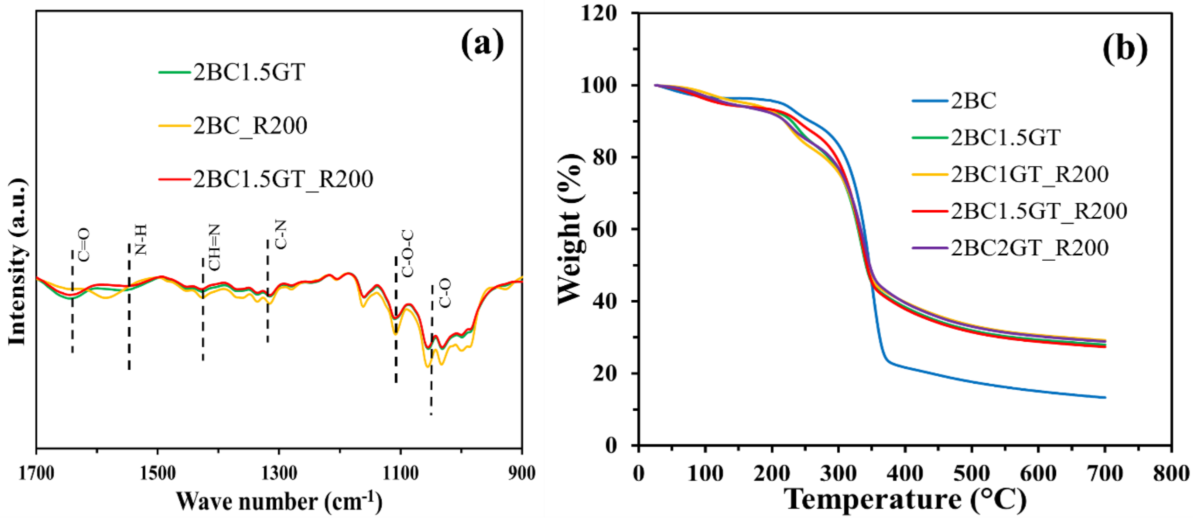


**Fig. S3** (a) FTIR spectrum of 2BC1.5GT,2BC_R200 and 2BC1.5GT_R200 and (b) TGA ($N_2$) curves of as-synthesised BC and hybridised BC-gelatin separators.

**The electrochemical characteristic of hybridised BC-gelatin separators via EIS**

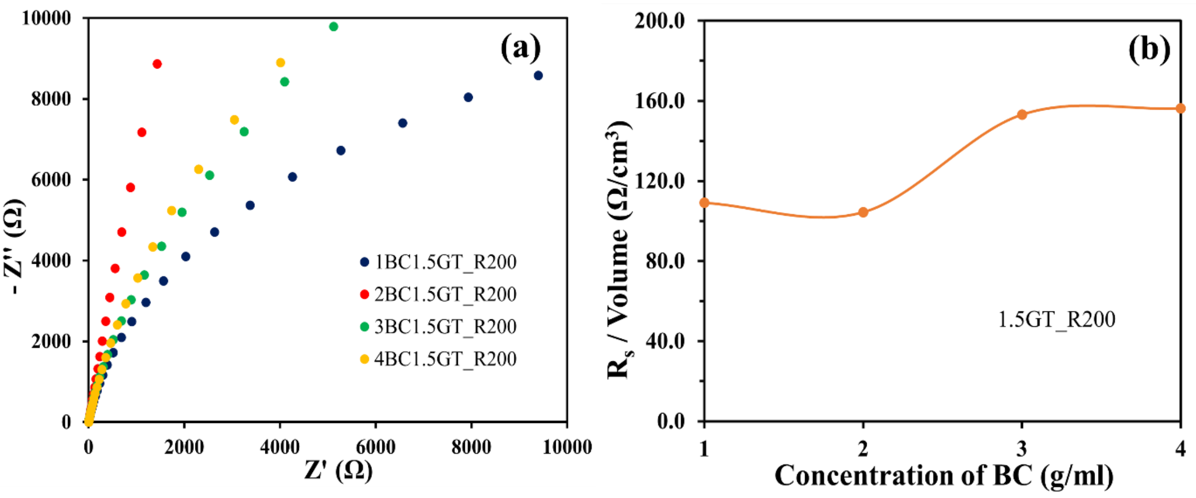


**Fig S4.** (a) Nyquist plots and (b) $R_s$/volume of of as-prepared BC and hybrisided BC-gelatin separators at different concentration of BC.