

Electronic Supplementary Information (ESI)

Experimental details:

The sulfonation of the carbon materials was carried out in a 100 ml dilute sulfuric acid solution of 0.1, 0.5 and 1M for 30 minutes. 1g of commercial carbon black (CB) (Meijo carbon, corp. particle size ranged between 30 - 40 nm) was chosen as a precursor for carbon catalysts. The carbon loading was 0.01 g/ml. A pair of 1 mm tungsten electrodes (Nilaco Corporation) shielded with an insulating ceramic tube was placed at the centre of a glass reactor with a gap distance of 0.5 mm. A bipolar high voltage pulse of 2.0 kV was applied to the tungsten electrodes using a high-voltage bipolar pulse generator (Pekuris MPP-HV04). After treatment, the solution temperature was slightly increased from 20 to 60 °C. All catalysts were washed with ethanol, followed by deionized water until the pH of the filtrate water reached neutral.

Definition and calculations of total conversion of cellulose, glucose yield and glucose selectivity:

- (1) Total conversion of cellulose (C %): mol of water soluble organic carbon / mol of carbon in ball-milled cellulose determined by CHNS analyzer) X100
- (2) Glucose yield (%): mol of glucose determined by HPLC X 6 / mol of carbon in ball-milled cellulose determined by CHNS analyzer X 100
- (3) Glucose selectivity (%): glucose yield(%) / total conversion of cellulose (%)

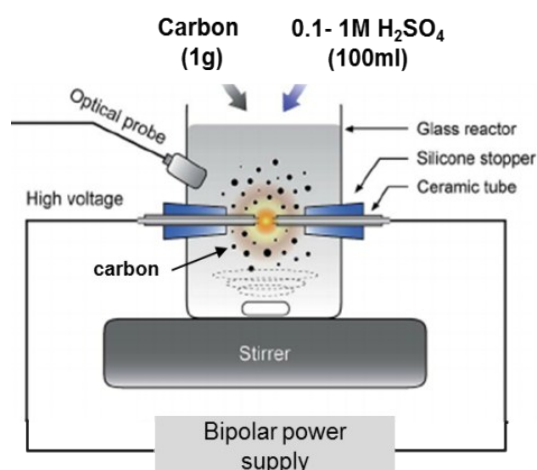


Fig. S1 Schematic image of sulfonation process of carbon materials by plasma process in dilute sulfuric acid solution of 0.1M, 0.5M and 1M

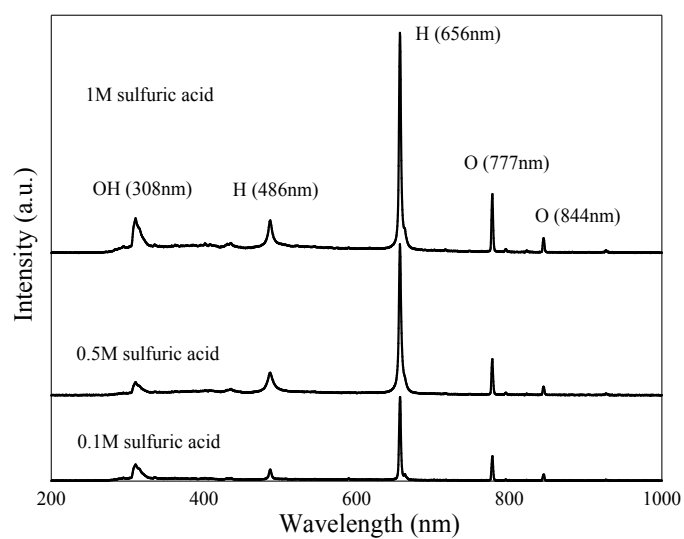


Fig. S2 Optical emission spectrum during plasma discharge at 0.1M, 0.5M and 1M H₂SO₄

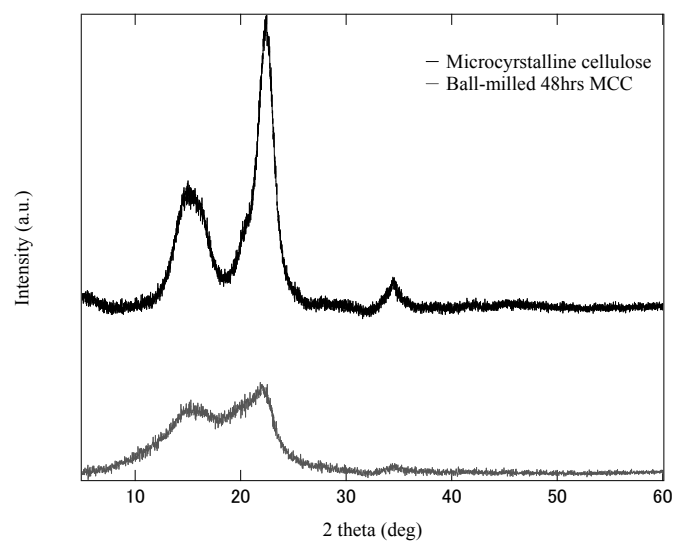


Fig. S3 X-ray diffraction of original microcrystalline cellulose and cellulose after ball-milling at 48 hours.

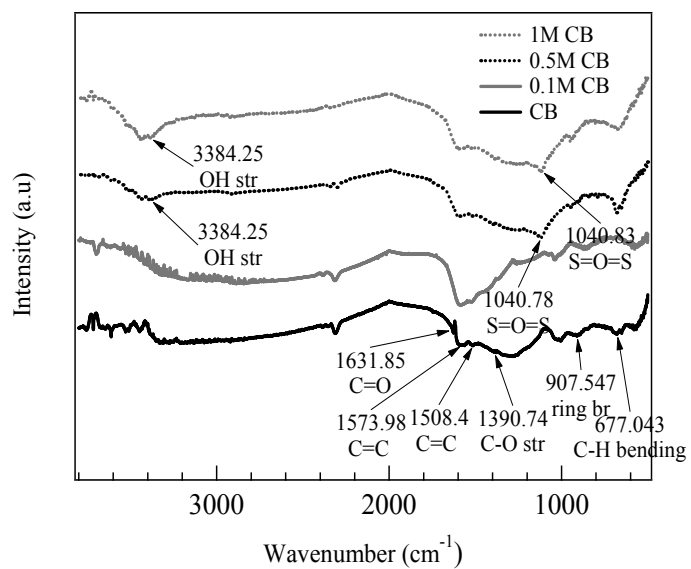


Fig. S4 Fourier transform infrared spectra (FTIR) of original CB, 0.1M CB, 0.5M CB and 1M CB

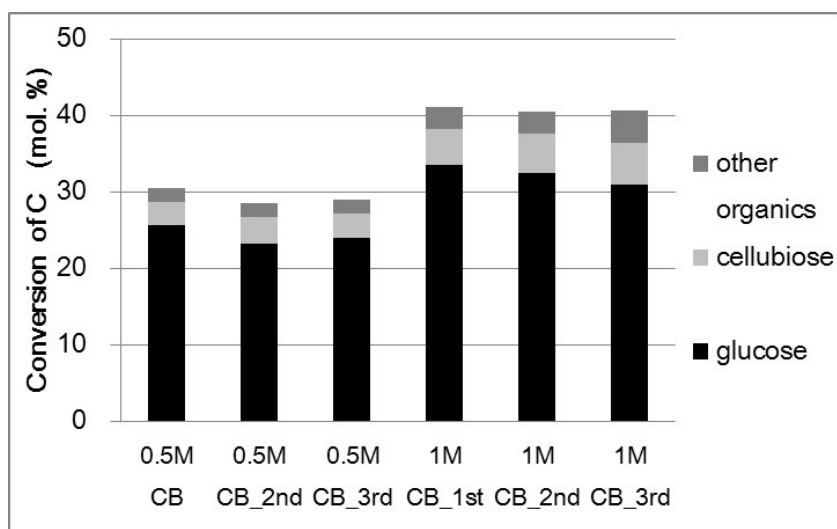


Fig. S5 Cellulose conversion percentage of 0.5M CB and 1M CB in hydrolysis reactions for three continuous trials

Table S1. Thermal stability of $-SO_3H$ in 1M CB under hydrothermal treatment at 120 - 210 °C (24 hours)

Temperature °C	$-SO_3H$ (mmol g ⁻¹)
1M CB (as synthesized)	2.0 - 2.2
120	1.6 - 1.8
150	1.5 - 1.7
180	1.5 - 1.7
210	1.5 - 1.7