Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information

Production of Cellulose Nanocrystals via a Scalable Mechanical Method

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Isolation of CNCs via ultrasonication: The cellulose suspension obtained after ultrasonication was left untouched for 24 h. Upon visual observation, two distinct layers of solution were identified; the top layer was used to isolate CNCs. The suspension from the bottom layer showed aggregates or bundles of cellulose particles.

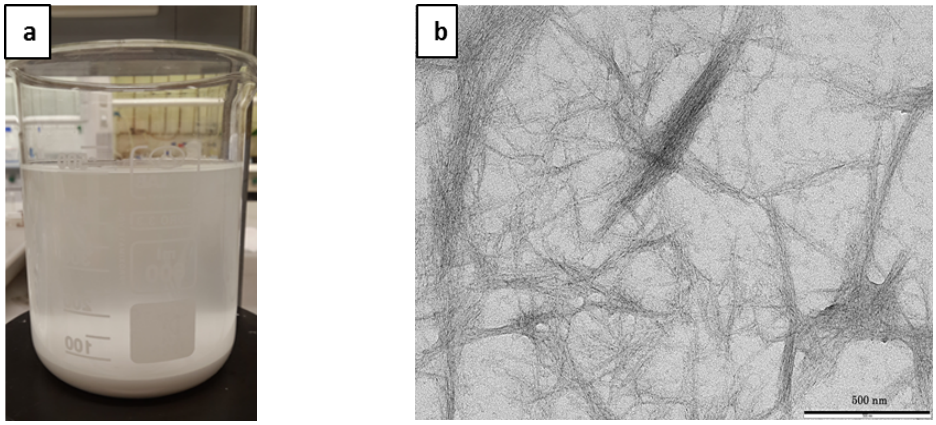

Fig. S1.a) CNC suspension produced via ultrasonication which was then left untouched for 24 h and b) TEM image obtained from the bottom layer of the suspension showing aggregates of particles ranging from 100 to 500 nm.

Isolation of CNCs via HEBM: This photograph was taken approximately a month after the milling process. The CNC-MC3 suspension is uniform and shows good dispersibility.

Fig. S2. Photograph of cellulose dispersion (2 wt. %) after milling which was allowed to stand for one month.

Dispersibility of CNCs from the HEBM process: Freeze-dried samples were suspended in deionised water at a concentration of 2 mg/mL and then sonicated in a bath sonicator for 4 hours. Significantly, the CNC-MC3 suspension shows higher dispersibility after 2 days compared with the other CNCs obtained via HEBM.

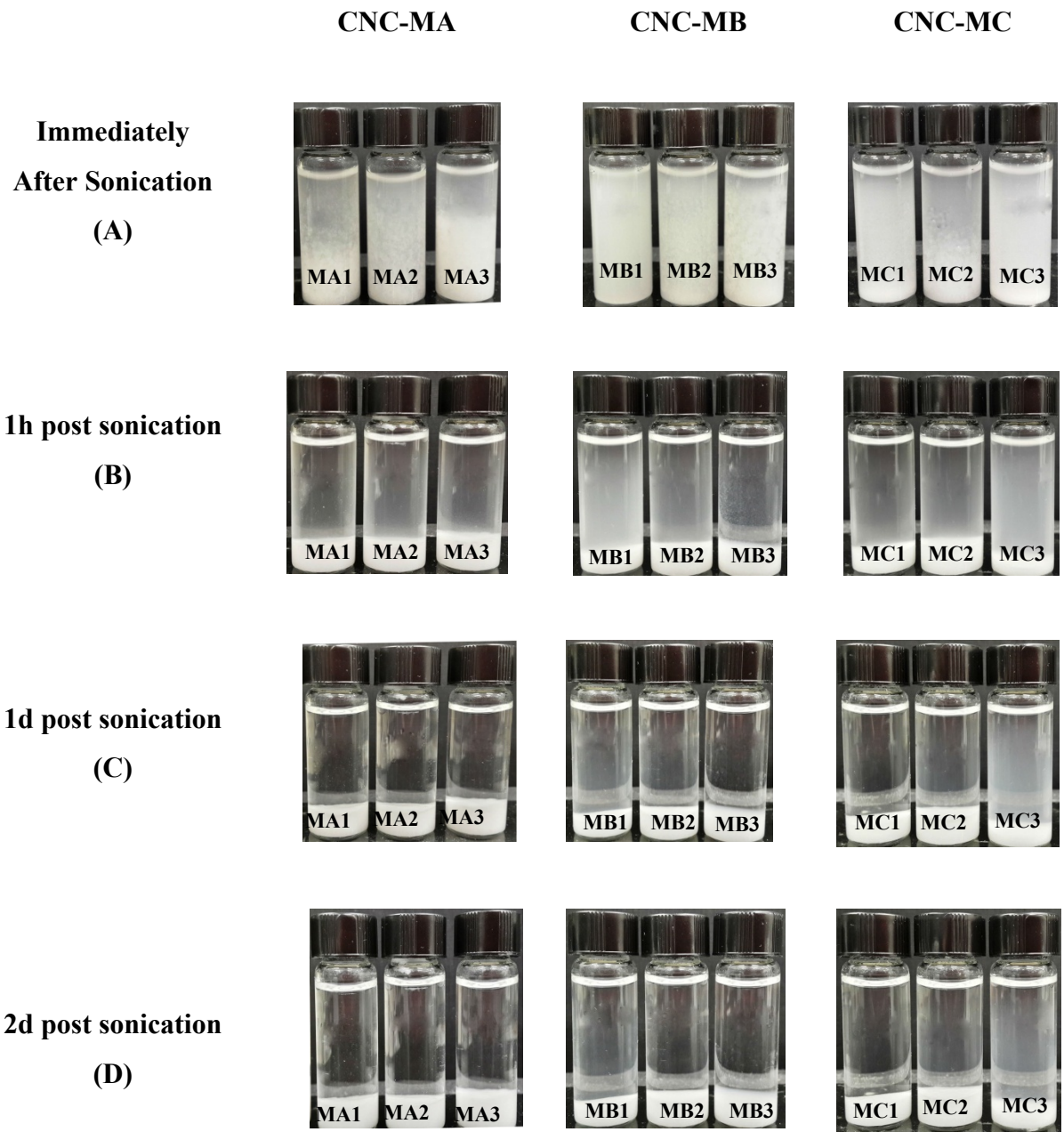

Fig. S3. Photograph of CNC dispersions in deionised water. These samples were obtained from HEBM method followed by sonication A) immediately after sonication B) 1 hour post sonication C) 1 day post sonication and D) 2 days post sonication.

Isolation of CNCs via HEBM with H_3PO_4.TEM micrographs of samples with a low concentration of MCC and high milling duration (CNC-MPA3) and high concentration of MCC with low duration of milling (CNC-MPC1 and CNC-MPC2). CNC-MPA3, CNC-MPC1 and CNC-MPC3 were unable to produce distinctive shape of rod-like /rice-like CNC particles.

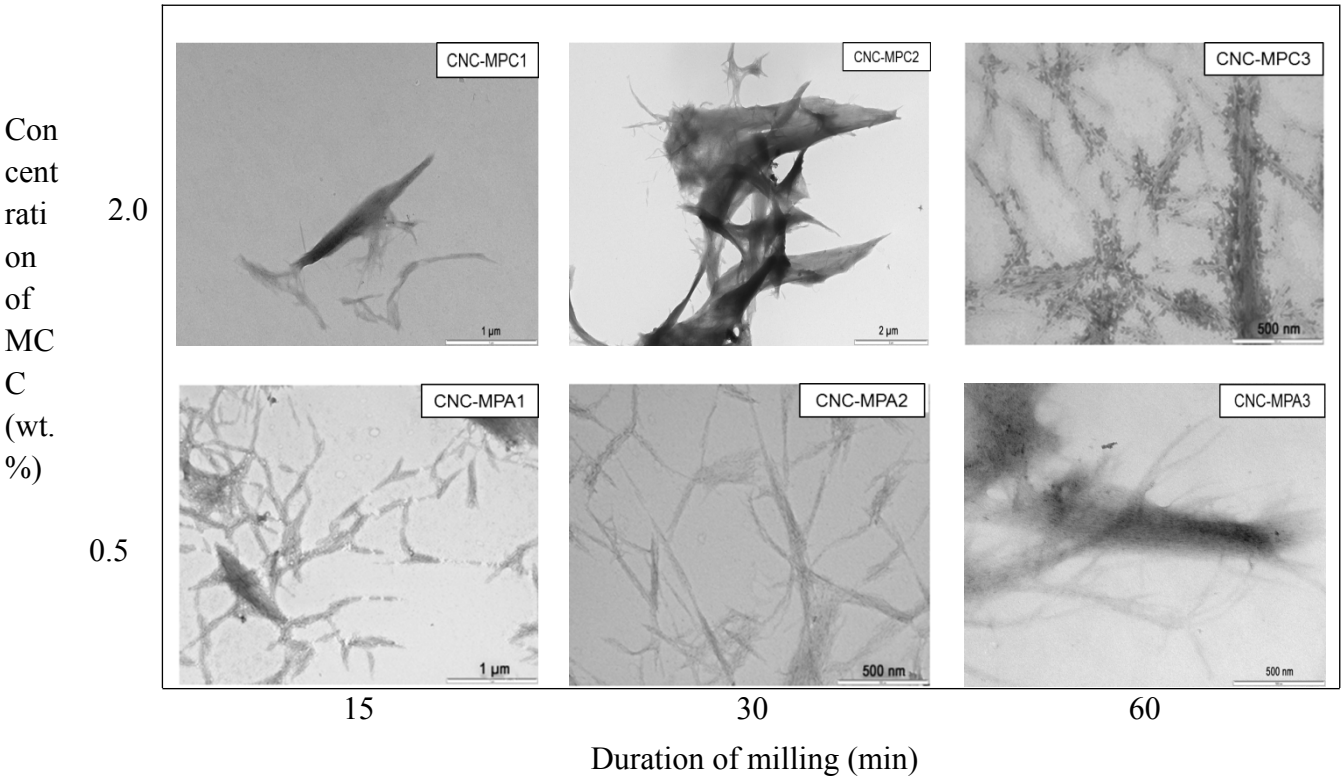

Fig. S4.TEM images CNCs milled with 1wt.% of H_3PO_4

Dispersibility of CNCs isolated in presence of dilute H_3PO_4. Freeze-dried samples were suspended in deionised water at 2 mg/mL concentration and then sonicated in a bath sonicator for 4 hours. As can be seen in the Fig. S5 CNC-MPA2 has the lowest dispersibility immediately after the sonication. The remaining CNCs, CNC-MPA1 and CNC-MPC3 have settled after 1 day.

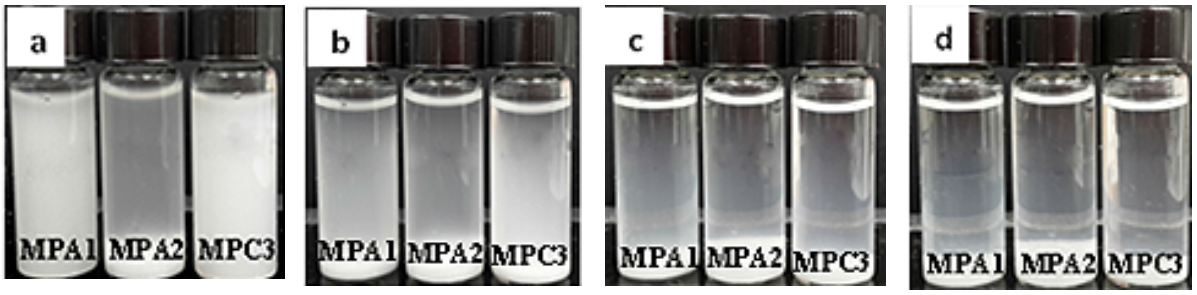

Fig. S5. Photograph of CNC dispersion in deionised water obtained from HEBM method with phosphoric acid. a) after sonication b) after 1 hour c) after 1 day and d) after 2 days.

XRD pattern. The crystallinity of CNCs obtained were calculated using formula described in in the body of the article. XRD peaks at 2θ= 15 (101), 16.5 (10$\bar{1}$), 20.8 (021), 22.5 (002) and 34.3 (040) (stars) were seen for the raw material. These peaks were also evident after milling with acid.

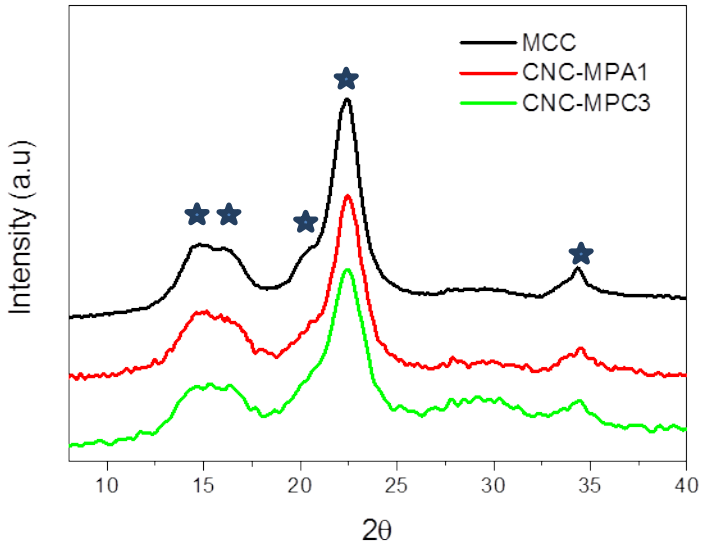

Fig. S6. XRD pattern of CNC obtained via HEBM with dilute phosphoric acid

Comparison of energy consumption for CNC isolation:

Energy consumption was calculated based on time and voltage of equipment used in a typical lab-scale isolation process. Table S1 shows the calculated amount of energy for both HEBM process (with initial input 4 g of raw material) and acid hydrolysis (with initial input of ~ 5.2 g of raw material).

Table S1 Comparison of energy consumption for both HEBM and acid hydrolysis

HEBM process				**Acid hydrolysis process**			
Equipment	**Voltage equipment used (Watt)**	**Time (h)**	**Energy (MJ)**	**Steps**	**Voltage of equipment used (Watt)**	**Time (h)**	**Energy (MJ)**
Stirring	15	12	0.7	Heating and stirring (Hot plate)	650	2	4.7
				Centrifugations	2704	1.7	16.6
Milling (Netzsch, *Labstar*)	3000	1	10.8	Dialysis	15	120	6.5
				Ultrasonication	500	0.5	0.9
Total			11.5	Total			28.7