Electronic Supplementary Material (ESI) for Soft Matter.

Supporting Information

Tailoring percolating conductive network of natural rubber composites for flexible strain sensors via cellulose nanocrystals templated assembly

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The components of experimental vulcanization system are shown in Table S1.

Table S1 The experimental vulcanization formula for preparation of NR based nanocomposites

Component	Zinc oxide	Stearic acid	Sulfur	CBS	OP emulsifier
Content (phr[a])	3	1.8	1.68	0.9	0.25

[a] parts per hundred parts of natural rubber

In order to determine whether the level of reductions in percolation threshold is linked to the CNC aspect ratio, a contrast research was conducted. The CNC with smaller aspect ratio was prepared by extending the time of sulfuric hydrolysis from 45 min to 3 h. The morphologies of the CNC with two different aspect ratios were characterized by TEM as shown in Fig. S1. As expected, the CNC in Fig. S1b exhibited a much smaller aspect ratio than that of Fig. S1a.

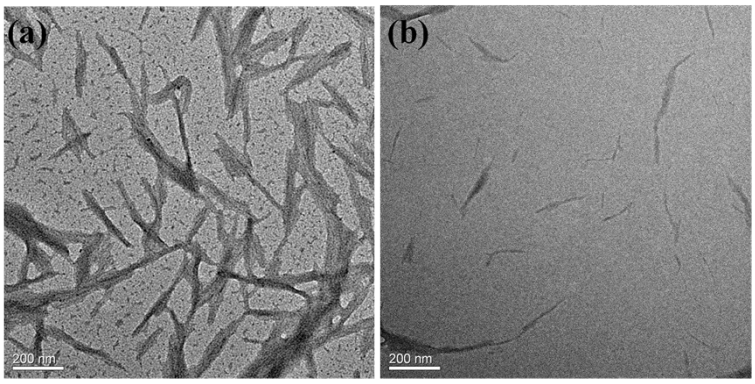

Fig. S1 TEM images of CNC prepared by sulfuric hydrolysis at 45 °C for (a) 45min and (b) 3h.

According to the results, there was no significant change in the electrical conductivity and percolation threshold of the sample with different CNC aspect ratios (as shown in Fig. S2). In this study, the main role of CNC is to assist assembly of CNTs for the construction of 3D conductive network, which is based on the colloidal-size structure and unique amphiphilic nature of CNC. Therefore, the level of reduction in percolation threshold is not linked to the CNC aspect ratio.

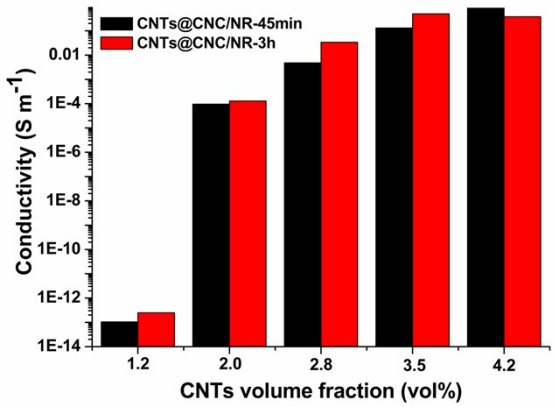

Fig. S2 Electrical conductivity of CNTs@CNC/NR nanocomposites with two different CNC aspect ratios as a function of CNTs volume fractions.

As a proof-of-concept demonstration for the application of strain sensors, the sample of CNTs@CNC/NR-2.8 was integrated into a simple circuit with a 9 V battery to light a light-emitting diode (LED), as shown in Fig. S3. As expected, the sample cannot maintain conductance when it was stretched to 40% strain in an integrated circuit. Furthermore, the LED restored almost the same illumination intensity after being subjected to repetitive stretch/release cycles, which was in accordance with Fig. 6b.

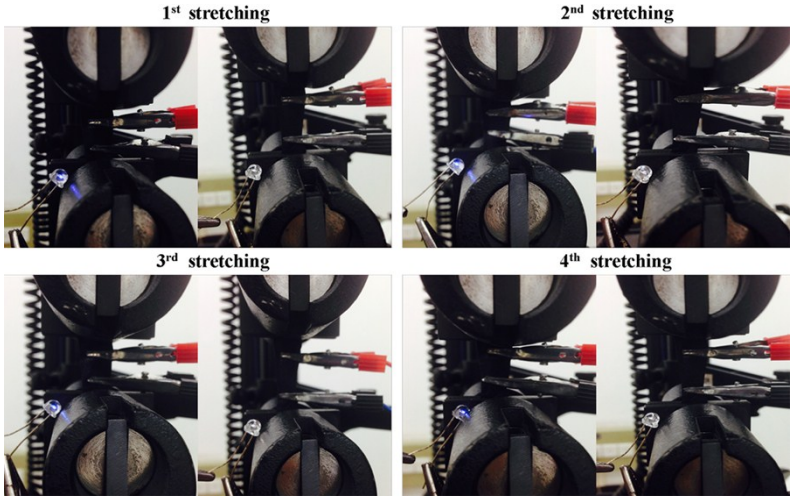

Fig. S3 Digital images of the LED integrated circuits with 3D structured strain sensor operating under repetitive stretch/ release cycles of 40% strain.

However, CNTs/NR composites with homogeneous dispersion morphology typically show irreversible changes in resistance when stretched to 100% strain levels (as shown in Fig. S4), which can be attribute to combinations of irreversible destroying of inter-nanotube interconnections and introduction of CNTs alignment.[1]

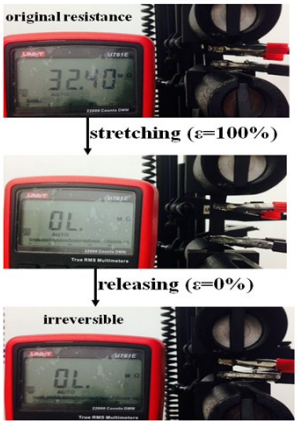

Fig. S4 Irreversible resistance change of the two-roll mill processed sample CNTs/NR-7.9.

REFERENCES

1 M. K. Shin, J. Oh, M. Lima, M. E. Kozlov, S. J. Kim and R. H. Baughman, *Adv. Mater.*, 2010, **22**, 2663.