Ultra-stretchable, sensitive and durable strain sensors based on polydopamine encapsulated carbon nanotubes/elastic bands

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Fig. S1 The Raman spectra of CNTs used in this work, the spectra were obtained using a WITec alpha300 R with 632 nm laser excitation at 1 cm\(^{-1}\) resolution.

Raman spectroscopy is performed to analyze the structure of CNTs. As shown in Fig. S1, three typical peaks at 1322, 1579, and 2643 cm\(^{-1}\) are presented, corresponding to the D, G, and 2D bands, respectively, which are the signatures of the CNTs structure. As is well known, the D band is related to the defects induced by disorder, whereas the G and 2D bands correspond to the in-plane vibration of the C-C bond.\(^1\)\(^,\)\(^2\)

Fig. S2 The TEM images of CNTs used in this work with different magnifications
The scheme of dopamine self-polymerization is illustrated in Fig. S4. Dopamine is easy to be oxidized into dopamine o-quinone form in aqueous solution. The oxidation reaction of phenolic hydroxyls in dopamine is reckoned to be the primary step of the cross-linking and formation of polydopamine (PDA) layer.\textsuperscript{3,4} The reverse dismutation reaction of dopamine and dopamine o-quinone directly results in cross-linking self-polymerization. During the oxidation and reverse dismutation process, the covalent and non-covalent interaction between dopamine and substrate are built up, making PDA layer tightly adhere to the surface of CNTs/EB.\textsuperscript{5,6}
**Fig. S5** Photograph of the electrodes and the PDA/CNTs/EB strain sensor.

**Fig. S6** The sticky tape images of CNTs/EB and PDA/CNTs/EB after peeling test.
Fig. S7 The change of $R/R_0$ for the first 10 cycles, 1001-1010 cycles and 9901-9910 cycles during loading-unloading cycles under 100% strain at 100mm/min for 10000 cycles.

Fig. S8 (a) The photos of PDA/CNTs/EB strain sensor with different stretching states. (b, c) The magnified view PDA/CNTs/EB strain sensor under 0% and 600%, respectively.

As shown in Fig. S8, the surface of PDA/CNTs/EB is balck due to the anchored CNTs (the strain is 0%, Fig. S8b). With the increasing of the strain, the PDA/CNTs/EB fiber becomes long and thin, resulting in the increasing distance between CNTs (black) on the EB (yellow) surface. When the strain is 600%, the surface of PDA/CNTs/EB become pale yellow due to the generation of gaps (Fig.
We investigate the evolution of conductive channels under different levels of strain by optical microscope (OM) (as shown in Fig. S9). At the beginning (Fig. S9a), the CNTs conductive networks are intact and no crack or gap appears on the surface. When the applied strain is increased to 200% (Fig. S9b), some minor white gaps are observed owning to the increased distance between emulsion particles on the surface of EB, resulting in a slight increase of RCR value. The numbers and areas of the white gap increase gradually with the further increasing of the strain (400%, as shown in Fig. S9c). With continually increasing the strain to 600% (Fig. S9d), a large amount of white gaps appear and the emulsion particles carrying CNTs separate with each other greatly. At the same time, CNTs conductive networks in the inner regions of PDA/CNTs/EB are destroyed significantly, these two damaged effects work together, leading to the rapid increase of RCR value (as shown in Fig. 10). In a word, the sensing mechanism of PDA/CNTs/EB strain sensor is mainly related to the evolution of the conductive paths both at the surface and the inner regions of the PDA/CNTs/EB.

After a pre-stretching (600%), as shown in Fig. S10, it is found the surface of PDA/CNTs/EB has some obvious gaps after the tension.
**Fig. S10** The SEM of PDA/CNTs/EB after a pre-stretching at a strain of 600%.

**References**