Supporting Information

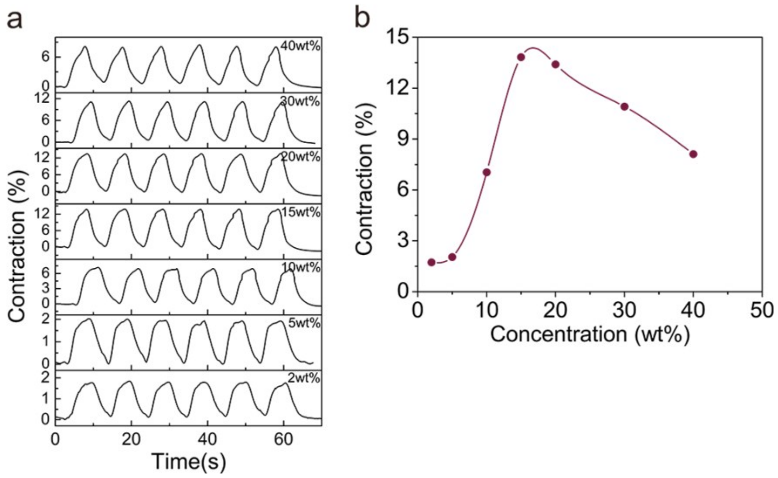

Fig.S1 a) and b) Contraction strain versus time for coiled CNT/TPU with different infiltrated TPU/DMF solution concentration from 2 wt% to 40 wt% when the 0.0.95 A~0.175 A with 0.1 Hz square wave pulse current was applied.

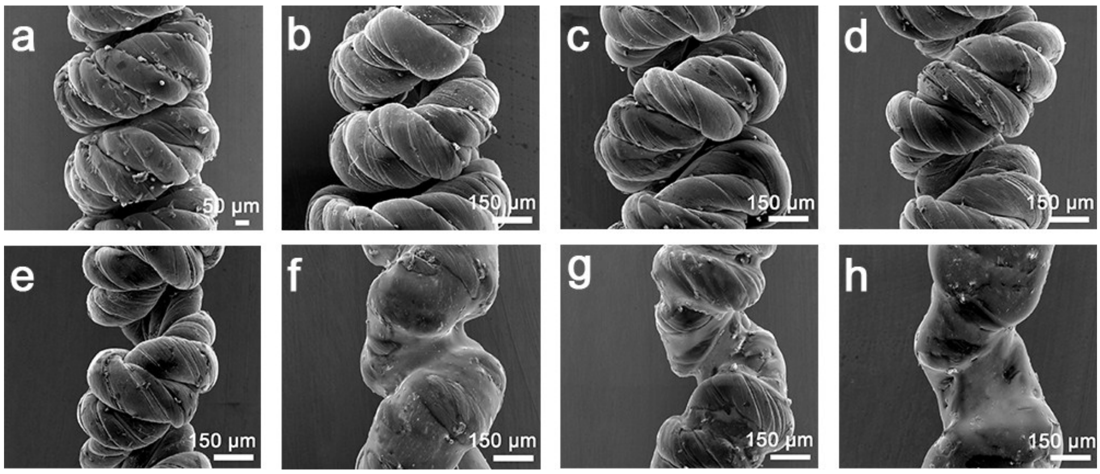

Fig.S2 SEM images of coiled CNT/TPU composite yarn treated with annealing and contraction training process, at different infiltrated TPU/DMF solution concentration from 0 wt% to 40 wt%. (a)~(h):0 wt%， 2 wt%, 5 wt%, 10 wt%, 15 wt%, 20 wt%, 30 wt%, 40 wt%.

It has been confirmed that volume expansion of a guest material within a coiled CNT yarn could cause reversible untwist which generated torque actuation driving large-stroke tensile actuation of coiled yarn. Nevertheless, the relationship between the amount of guest materials twisted in the coiled structure and contraction strain has not been discussed. The driven testing was firstly conducted on the coiled hybrid yarn under the same applied load (0.1 N) and current (0.175 A) to analyze the effect of concentration of infiltrated polyurethane solution on the tensile actuation and showed that with the rise of concentration from 2 wt% to 40 wt%, the contractive strain (defined as the percentage of contraction to initial yarn length) increased firstly from 1.7 % to 13.8 % and then decreased to 8.1 wt% (Fig.S1).

As could be seen from the morphology of coiled yarns after the treatment of annealing and contraction training (Fig.S2), coil diameter decreased and coil pitch increased with increasing concentration from 0 wt% to 40 wt%. With TPU/DMF solution consistence increasing from 0 wt% to 15 wt%, the increasing coil pitch and the content of TPU adhered to the internal structure of the fiber gave rise to a continuous improvement on lengthwise contraction strain. Whereas when the concentration of impregnating TPU solution exceeded 15 wt%, the actuation performance of composite yarns declined, probably ascribed to few TPU resin influx into the internal structure of CNT fiber and a large quantity enrichment on the surface of CNT fiber, resulted from the great viscosity of highly concentrated solutions (Fig.S2f~h). And furthermore, outsized coil pitch of coiled CNT/TPU composite yarns treated with annealing and contraction training process may also be responsible for driving performance degradation. From the above, the coiled composite yarns synthesized at the TPU/DMF solution concentration of 15 wt% were selected as the main focus in this work, given that the desired giant contraction was crucial for the coiled yarn actuator which could be extensively employed as the self-deployable structure in aerospace field.

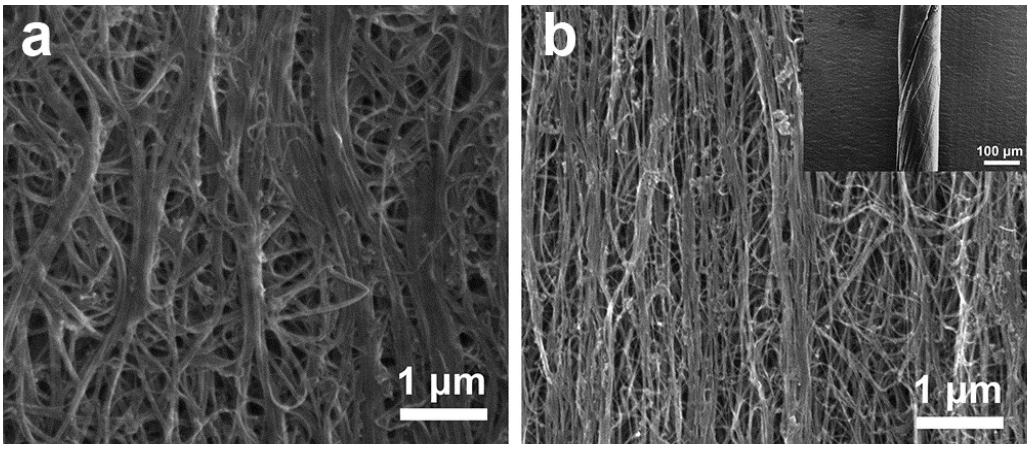

Fig.S3 Scattering patterns of CNT ribbon (a), CNT yarn-2k r/m (b), neat coiled CNT yarn (c), hybrid coiled CNT/TPU yarn (d).

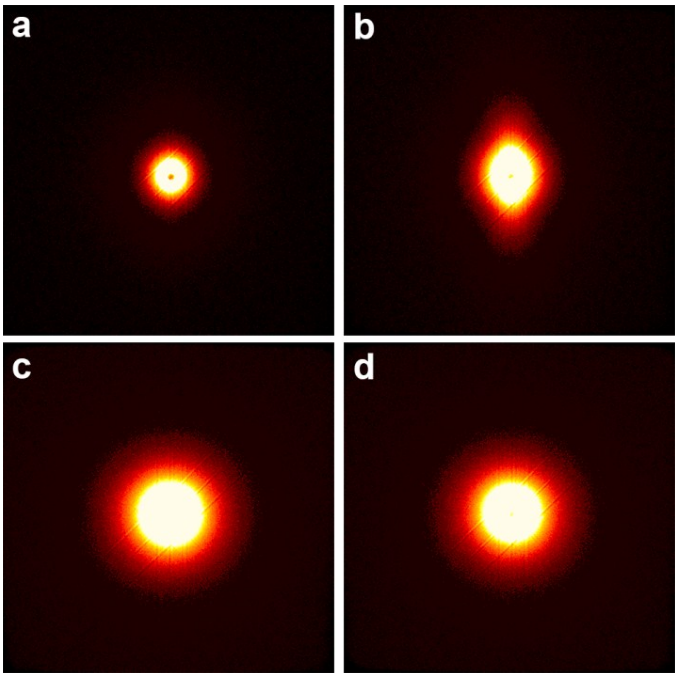

Fig.S4 Scattering patterns of CNT ribbon (a), CNT yarn-2k r/m (b), neat coiled CNT yarn (c), hybrid coiled CNT/TPU yarn (d).

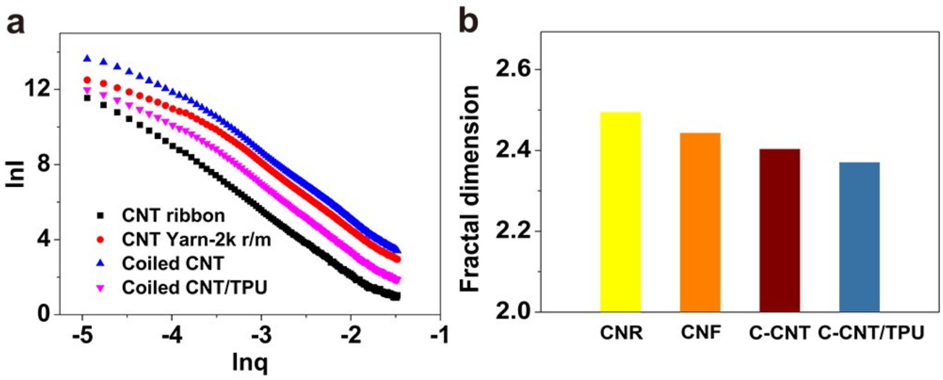

Fig.S5 a) Scattering vector dependence of the scattering intensity for the samples presented in Fig.S4. I stands for intensity, q stands for scattering vector. b) Fractal dimension of CNT ribbon (CNR), CNT fiber-2k r/m (CNF), coiled CNT (c-CNT), coiled CNT/TPU (C-CNT /TPU).

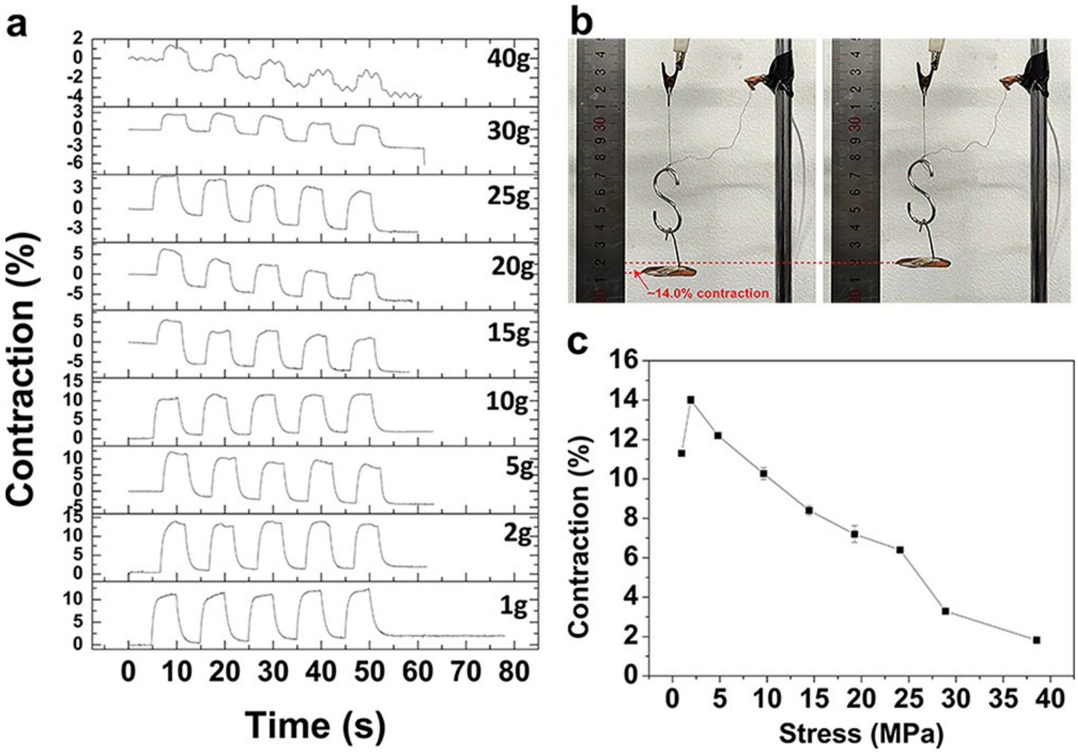

Fig.S6 a) Contraction of single-ply coiled CNT/TPU yarn at different loads from 0.01 N to 0.4 N. b) Photograph showing the single-ply coiled CNT/TPU yarn actuator can reversibly lift up 2 gram weight at ~14.0 % contraction strain. C) The stress dependence of contraction strain of single-ply coiled CNT/TPU yarn.

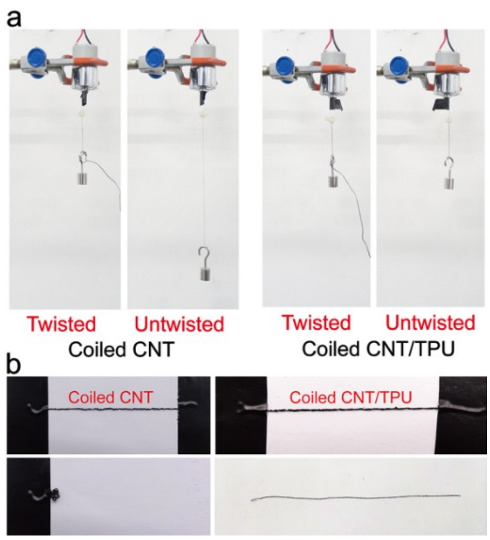

Fig.S7 a) Photograph of coiled CNT (left), coiled CNT/TPU (right) before and after untwisting under 0.2 N load, respectively. b) Photograph of coiled CNT (left), coiled CNT/TPU (right) before and after heat-treated, respectively.

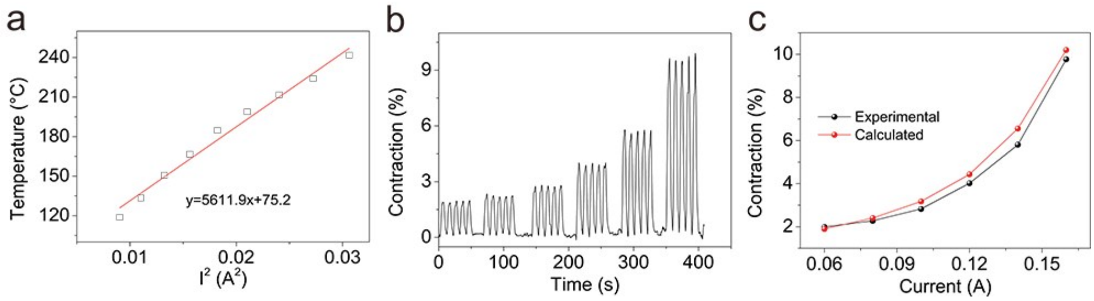

Fig.S8 a) Experimental and fitting curve of temperature versus the square of the current for coiled CNT/TPU. Contraction strain versus time for coiled CNT/TPU with the length of 3.5 cm and applied load of 0.1 N. b) The applied square wave pulse current was 0.06A, 0.08A, 0.10A, 0.12A, 0.14A, 0.16A with 0.1Hz. c) Experimental and calculated contraction strain at different input electrical current.

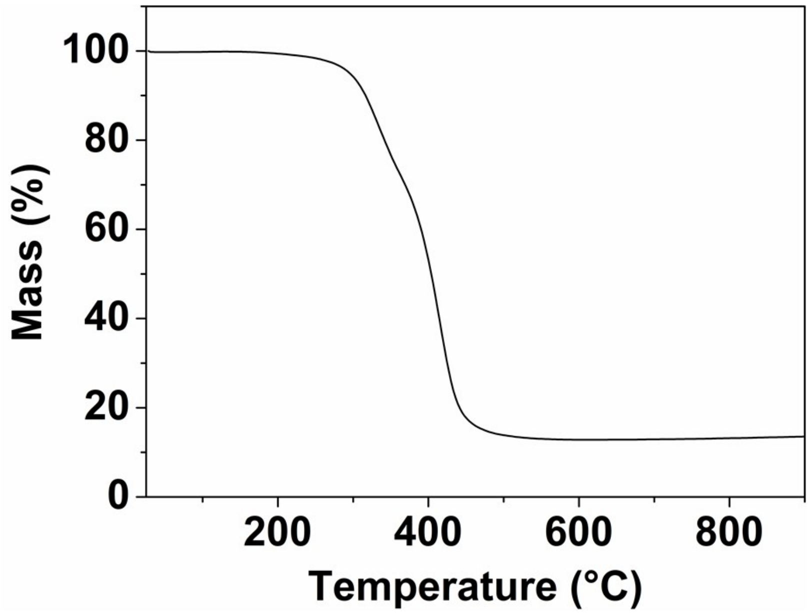

Fig.S9 TGA result of TPU

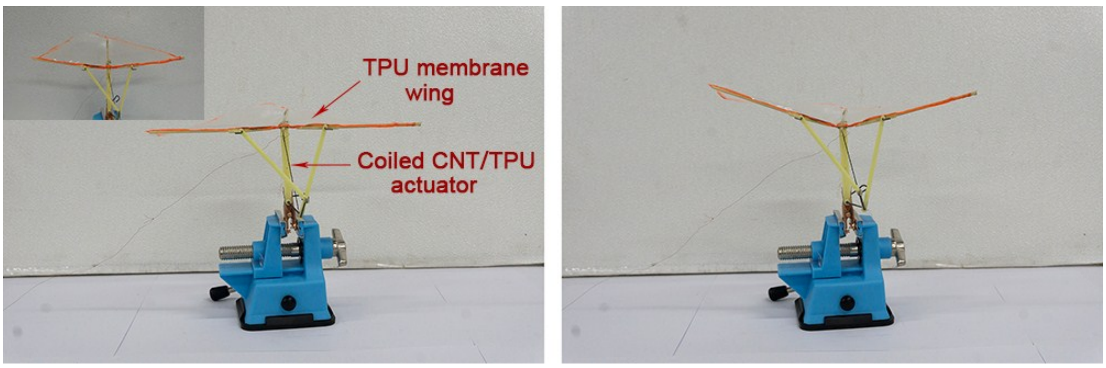

Fig.S10 Setup of morphing aircraft structures before (left) and after (right) being applied a square-wave current

(0.175 A, 0.1 Hz).

Video S1 showing tensile actuation of coiled CNT/TPU hybrid yarn at a applied 0.175 A, 0.1 Hz square-wave current.

Video S2 showing the untwist of the coiled neat yarn annealed at 120 °C under the applied load of 0.2 N.

Video S3 showing the untwist of the coiled CNT/TPU yarn annealed at 120 °C under the applied load of 0.2 N.

Video S4-S7 showing the unfold and fold process of morphing aircraft structures when applied a square-wave current of 0.175 A with current frequency of 0.1 Hz, 0.5 Hz, 1 Hz, 2.5 Hz, respectively.