Scratch-Resistant, Highly Conductive, and High-Strength Carbon Nanotube-Based Composite Yarns

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Textiles composed of high-strength and conductive yarns are very attractive for people's daily life, which allow wearers multifunctions ranging from listening to audio devices to controlling temperature and are also indispensable in many military and aerospace applications, such as bullet-proof vests, radiation protection suits, and spacesuits. Historically, the progress of yarns has depended on the novelty of the materials. Carbon nanotubes (CNTs), an ideal one-dimensional flexible nanomaterial, possess remarkable mechanical properties and great electrical conductivity.[1] Individual CNTs are one of the strongest materials known with high tensile strengths between 10 to 100 GPa and a Young's modulus up to 1 TPa[2−4] and are ideal one-dimensional conductors with a quantum conductance of $2G_0$.[5] To fully utilize the excellent properties of individual CNTs, many methods have been developed to make macroscale CNT yarns composed of pure CNTs or a large fraction of CNTs.[6−25] However, the strength and the conductivity of these yarns are reduced greatly compared to individual CNTs. The main reason is that there are weak CNT joints and weak intertube interactions in a yarn,[18,24,26] inducing a poor load transfer efficiency and a large contact resistance.

Researchers have developed some approaches to enhance the strength and the conductivity of CNT yarns. However, there is a problem when trying to simultaneously enhance both of them. To obtain high-strength CNT yarns, some polymers, such as polyvinyl alcohol (PVA) or epoxy, were used to make CNT/polymer composite yarns, sometimes with the aid of a surfactant.[6−8,17,25] The tensile strength of the as-produced composite yarns can reach

ABSTRACT   High-strength and conductive carbon nanotube (CNT) yarns are very attractive in many potential applications. However, there is a difficulty when simultaneously enhancing the strength and conductivity of CNT yarns. Adding some polymers into CNT yarns to enhance their strength will decrease their conductivity, while treating them in acid or coating them with metal nanoparticles to enhance their conductivity will reduce their strength. To overcome this difficulty, here we report a method to make high-strength and highly conductive CNT-based composite yarns by using a continuous superaligned CNT (SACNT) yarn as a conductive framework and then inserting polyvinyl alcohol (PVA) into the intertube spaces of the framework through PVA/dimethyl sulphoxide solution to enhance the strength of yarns. The as-produced CNT/PVA composite yarns possess very high tensile strengths up to 2.0 GPa and Young's moduli more than 120 GPa, much higher than those of the CNT/PVA yarns reported. The electric conductivity of as-produced composite yarns is as high as 9.2×10^4 S/m, comparable to HNO_3-treated or Au nanoparticle-coated CNT yarns. These composite yarns are flexible, lightweight, scratch-resistant, very stable in the lab environment, and resistant to extremely humid ambient and as a result can be woven into high-strength and heatable fabrics, showing potential applications in flexible heaters, bullet-proof vests, radiation protection suits, and spacesuits.

KEYWORDS: carbon nanotube · yarn · array · composite · superaligned

up to more than 1 GPa, but the insulating polymer and the possibly residual surfactant reduces the conductivity of the composite yarns. On the other hand, to obtain highly conductive CNT yarns, CNTs should be treated using superacid, HNO_3, or $HAuCl_4$,[9,10,21] but these treatments would induce many defects of CNTs and thus greatly reduce the tensile strength of CNT yarns. Therefore, it is of vital importance to develop a method to produce both high-strength and highly conductive CNT yarns.

To achieve this goal and make the spinning process continuous, our basic idea is to first produce a pure CNT yarn continuously as a framework to ensure a good conductivity and then insert a polymer into the small spaces inside the framework, forming a strong CNT−polymer interface coupling to enhance the strength of yarns. In this process, any acid or surfactant treatment should not be involved. Here we use a
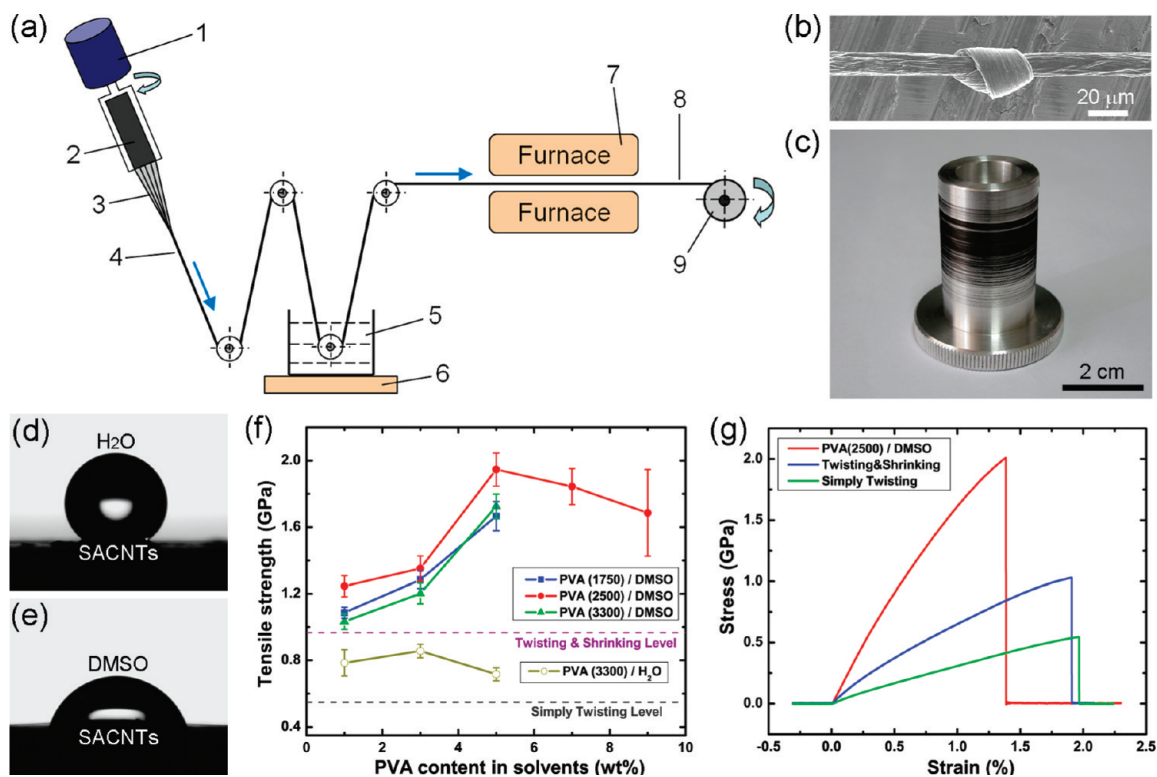
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Figure 1. Experimental setup, as-produced yarns, and their mechanical properties. (a) Illustration of setup. The arrows indicate the moving direction of a yarn in a spinning process: (1) high-speed rotating motor for twisting; (2) SACNT array; (3) SACNT film; (4) twisting yarn; (5) vessel filled with PVA/DMSO solution; (6) heating plate; (7) tube furnace; (8) SACNT/PVA yarn; and (9) rotating motor for yarn collection. **(b)** SACNT/PVA yarn flexibly tied with a kink. **(c)** 100 m long SACNT/PVA yarn with a diameter of 15 μm collected on a metal winder. **(d)** 10 μL water droplet on a cross-stacked SACNT film. **(e)** 10 μL DMSO droplet on a cross-stacked SACNT film. **(f)** Dependence of tensile strengths of SACNT/PVA yarns on the concentration of PVA/DMSO and PVA/H₂O solutions. The polymerization degrees of PVA are indicated in the brackets. The two dashed lines indicate referenced tensile strengths of two types of pure SACNT yarns: simply twisting and twisting and shrinking (by acetone) yarn. **(g)** Stress−strain curves of a typical SACNT/PVA yarn made by the method developed herein and the two types of pure SACNT yarns. All of the yarns are drawn out from a 425 μm high and 1 cm wide SACNT array with the twisting speed of 1050 rpm (round per minute) and the collecting speed of 20.4 cm min^{-1}.

simple twisting yarn made from superaligned CNT (SACNT) arrays as the framework and PVA/dimethyl sulphoxide (DMSO) solution to insert the PVA into the yarn. Based on these ideas, we can therefore obtain SACNT/PVA yarns possessing very high tensile strengths up to 2.0 GPa and Young's moduli more than 120 GPa, much higher than the reported values of CNT/PVA yarns. The electrical conductivity of SACNT/PVA yarns is as high as 9.2×10^4 S m^{-1}, comparable to HNO$_3$-treated or Au-coated SACNT yarns. These composite yarns are flexible, lightweight, scratch-resistant, very stable in the lab environment, and resistant to an extremely humid environment and as a result can be woven into high-strength and heatable fabrics.

PVA is a flexible polymer material that has excellent adhesive property and is used to enhance the strength of CNT yarns.[6−8,17,25] Currently, PVA is introduced into CNT yarns *via* PVA/H$_2$O solution. The major problem with using PVA/H$_2$O solution is that this solution cannot wet CNTs very well, as CNTs are hydrophobic. Thus, some surfactants, such as sodium dodecyl sulfate (SDS), are used to chemically modify CNTs to make them hydrophilic.[6−8,25] However, the additional surfactant is

usually left in the produced CNT/PVA yarns, which has a negative influence on the strength and the conductivity of the composite yarns. To overcome this problem, our idea is to find a solvent that can not only dissolve PVA but also wet CNTs. As will be discussed in the following text, DMSO is such a solvent.

CNT yarns are prepared using a dry method that draws CNTs out from SACNT arrays.[16,18,27] This method has many advantages for preparing long CNT yarns as desired.[23] First, the process is continuous, which is convenient for producing a yarn tens to hundreds of meters long; second, this method is cost-effective because the raw material is cheap multiwalled CNTs (MWCNTs); and third, it is easy to control the diameter of a yarn by varying the width of a SACNT array and to produce ultrathin yarns with the diameter less than 10 μm. Currently, there are two approaches to spinning yarns from SACNT arrays: twisting by a motor[17,19−22] and shrinking by solvents.[18]

Our spinning method combines the two spinning approaches. The experimental setup is shown in Figure 1a. The detailed spinning process is described in the Methods Section. Briefly, a SACNT yarn is first twisted

from a SACNT array, then passes through PVA/DMSO solution, and is finally dried. This spinning process is continuous, simple, and cost-effective. The as-produced SACNT/PVA yarn is flexible (Figure 1b). Figure 1c shows a 100 m long continuous SACNT/PVA yarn with a diameter of 15 μm enwound on a winder which is spun from a 15 cm (length) × 1 cm (width) array.

The key point to obtain high-strength SACNT/PVA composite yarns is the usage of DMSO for dissolving PVA. DMSO is a polar solvent which has excellent solvating power, large surface tension, and is less toxic than other members of this class, such as dimethylformamide and *N*-methyl-2-pyrrolidone. It is extensively used as a solvent for chemical reactions and as an extractant in biochemistry and cell biology and is also used for dissolving CNTs and PVA to make CNT-enhanced PVA tapes and films.[28,29] In our experiment, PVA can be dissolved in DMSO well within the weight concentration required, ranging from 1 to 9 wt %, which indicates that DMSO is an effective solvent for dissolving PVA. The merit of DMSO over water as the solvent used is that DMSO wets hydrophobic CNTs. The contact angle between water and SACNTs is about 135° (Figure 1d) but that between DMSO and SACNTs is about 70° (Figure 1e). This wettability between DMSO and CNTs makes PVA/DMSO a solution capable of infiltrating the intertube spaces and coating CNTs with PVA more effectively than PVA/H_2O solution.

We used three kinds of PVA with different polymerization degrees (1750, 2500, and 3300) in the PVA/DMSO solution. The polymerization degree indicates the average number of repeat units in a polymer chain. The PVA with a higher polymerization degree usually possesses a longer molecular chain, higher strength, and better stickiness. As shown in Figure 1f, we found that the PVA, with the moderate polymerization degree of 2500, gives rise to the best enhancement for the strength of SACNT/PVA yarns. This may be due to the following reason: When the polymerization degree is not very high, the strength of the composite yarn increases with increasing the polymerization degree of PVA used because of the increase of the strength and stickiness of PVA. However, when the polymerization degree is very high, the very long molecular chain of PVA makes it hard to infiltrate the intertube spaces inside a yarn in the treating process using PVA/DMSO solution to form strong CNT−PVA couplings. Therefore, too low and too large polymerization degrees of PVA are not favorable for obtaining high-strength CNT/PVA composite yarns.

Figure 1f also shows that with increasing the PVA (2500) weight concentration of the PVA/DMSO solution, the tensile strengths of as-produced SACNT/PVA yarns first increase and then decrease but are always much higher than those of pure SACNT yarns made by the twisting and shrinking (by acetone) and simply twisting methods (dashed lines in Figure 1f). As reference samples, SACNT/PVA yarns treated with PVA/H_2O solution were also prepared; the tensile strengths of these yarns are much lower than the twisting and shrinking SACNT yarns, though still higher than the simply twisting SACNT yarns (Figure 1f). These results show that DMSO, as the solvent of PVA solution, is more effective than H_2O to make high-strength composite yarns.

As shown in Figure 1f, the peak value of the tensile strengths of SACNT/PVA yarns is 1.95 GPa when using 5 wt % PVA/DMSO solution, which is 255% higher than the tensile strength of a simply twisting SACNT yarn (0.55 GPa) and 103% higher than that of a twisting and shrinking (by acetone) SACNT yarn (0.96 GPa). Compared with these SACNT yarns, the maximum strain at fracture of the SACNT/PVA yarn reduces to 1.4%, but the Young's modulus abruptly increases to 120−200 GPa (Figure 1g). The SACNT/PVA yarn is lightweight with a density of about 0.98 g cm^{-3}. The specific strength of the composite yarn is about 2.0 GPa (g cm^{-3})$^{-1}$, and the specific modulus is 120−205 GPa (g cm^{-3})$^{-1}$. These values of strengths and moduli are higher than the reported values of the CNT/PVA yarns in literatures.[6−8,17,25]

The good mechanical properties of SACNT/PVA yarns should be attributed to the combined action of PVA and DMSO, which acts in two ways. First, the wettability between DMSO and CNTs makes PVA fill the intertube spaces inside a yarn (Figure 2b vs f), drawing CNTs close and sticking CNTs together, which then decreases the diameter of the yarn (Figure 2a vs e). Second, the strong PVA−CNT interface coupling improves the load-transfer efficiency and thereby increases the tensile load of a yarn (Figure 3a). Thus, generally speaking, both the reduction in yarn diameter and the enhancement in tensile load result in remarkable improvements in strength and modulus of the SACNT/PVA yarns. Next we will further study the effect of DMSO and PVA, with a purpose to distinguish the individual function of DMSO and PVA.

It is found that many solvents, whether wetting or not wetting CNTs, such as ethanol, acetone, and water, can shrink CNT yarns.[14,15,18,23] The extent of shrinking is related to the wettability and the surface tension of the solvent.[23] For example, acetone shows a good shrinking effect due to its good wettability with CNTs,[14,15,23] while water is capable of shrinking CNT yarns, largely because of its huge surface tension.[15,23] In our spinning method, the solvent DMSO has the advantage of wetting CNTs, as mentioned above. Here we should note that the other merit of DMSO is its very high surface tension (43.54 mN m^{-1} at 20 °C), which is nearly twice the surface tension of ordinary organic solvents, such as ethanol and acetone.[30] After a simply twisting yarn is shrunk by DMSO, the diameter of this yarn is reduced from 19.0 to 14.4 μm (Figures 2a and c and 3a), and the tensile load is increased from 152 mN to 188 mN (Fig-
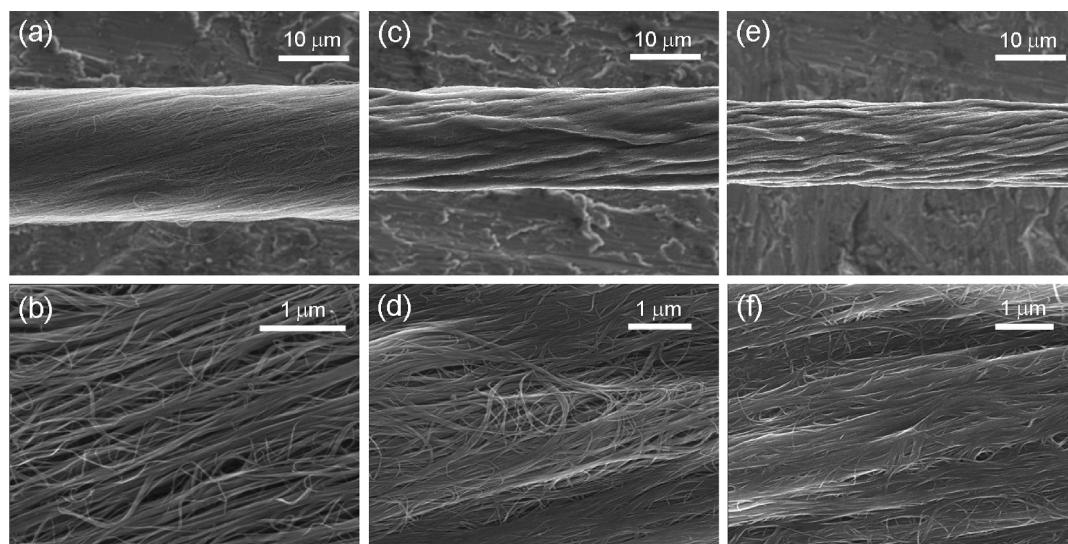
Figure 2. SEM images of yarns. (a and b) SACNT yarn made by the simply twisting method. It can be seen that there are many large intertube spaces. (c and d) SACNT yarn made by the twisting and shrinking (by DMSO) method. The intertube space of this yarn as well as the yarn diameter is reduced greatly compared with the simply twisting yarn. (e and f) SACNT/PVA yarn made from 5 wt % PVA/DMSO solution. Its intertube space is filled by PVA, and its diameter is further reduced compared with the twisting and shrinking (by DMSO) yarn.

ure 3a). As a result, the tensile strength increases from 0.54 to 1.16 GPa, a value greater than that of a twisting and shrinking (by acetone) yarn (0.96 GPa, as shown in Figure 1f and g). These results indicate that DMSO itself has a very good shrinking effect (Figure 2c and d), which is even better than acetone, and greatly enhances the strength of yarns, due to its wettability with CNTs and high surface tension.

When PVA is dissolved in DMSO, PVA and DMSO will cofunctionate on SACNT yarns. Because DMSO wets CNTs, DMSO taken along with PVA can infiltrate the intertube spaces in a yarn. The density of a simply twisting yarn is measured to be $0.4-0.55$ g cm^{-3}, and thus the intertube spaces in the yarn are calculated to be more than 70 vol% (taking the graphite density $2.09-2.23$ g cm^{-3} as the density of SACNTs without intertube spaces). Filling these huge intertube spaces would require a large amount of PVA greater than 60 wt % (PVA density $1.19-1.31$ g cm^{-3}), but the actual PVA content is measured to be only about 19 wt % according to the thermogravimetric analysis (TGA) mea-

surements in the strongest SACNT/PVA yarn when using 5 wt % PVA/DMSO solution (Figure S1, Supporting Information). Therefore, the shrinking effect of DMSO, as mentioned previously, should play a vital role on prereducing the intertube spaces in a yarn, which draws CNTs close and benefits the further action of PVA. In this process, increasing the temperature of PVA/DMSO solution will reduce the surface tension of DMSO as well as its shrinking effect and as a result increase the yarn diameter and weaken the yarn (Figure 3b). This implies that the strongest SACNT/PVA yarns made by our method can be obtained at room temperature (*i.e.*, $20-28$ °C in our experiments), which is favorable for reducing the production cost.

While DMSO shrinks a SACNT yarn to a great extent, PVA sticks CNTs together and draws them closer after the DMSO is vaporized due to the heating process in the furnace, further reducing the yarn diameter and increasing the tensile load. To investigate the effect of PVA, we studied the properties of SACNT/PVA yarns made from PVA/DMSO solution with various concentra-
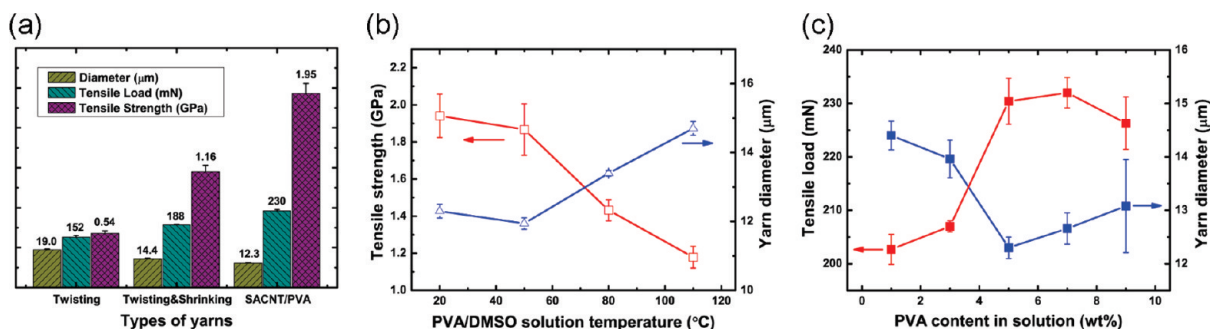

Figure 3. Comparison of different types of yarns based on SACNTs and the dependence of mechanical properties of SACNT/PVA on solution parameters. (a) Comparison of a simply twisting, a twisting and shrinking (by DMSO), and a SACNT/PVA yarn. (b) Dependence of tensile strengths and diameters of SACNT/PVA yarns on the temperature of PVA solution. (c) Dependence of tensile loads and diameters of SACNT/PVA yarns on the concentration of PVA solution.
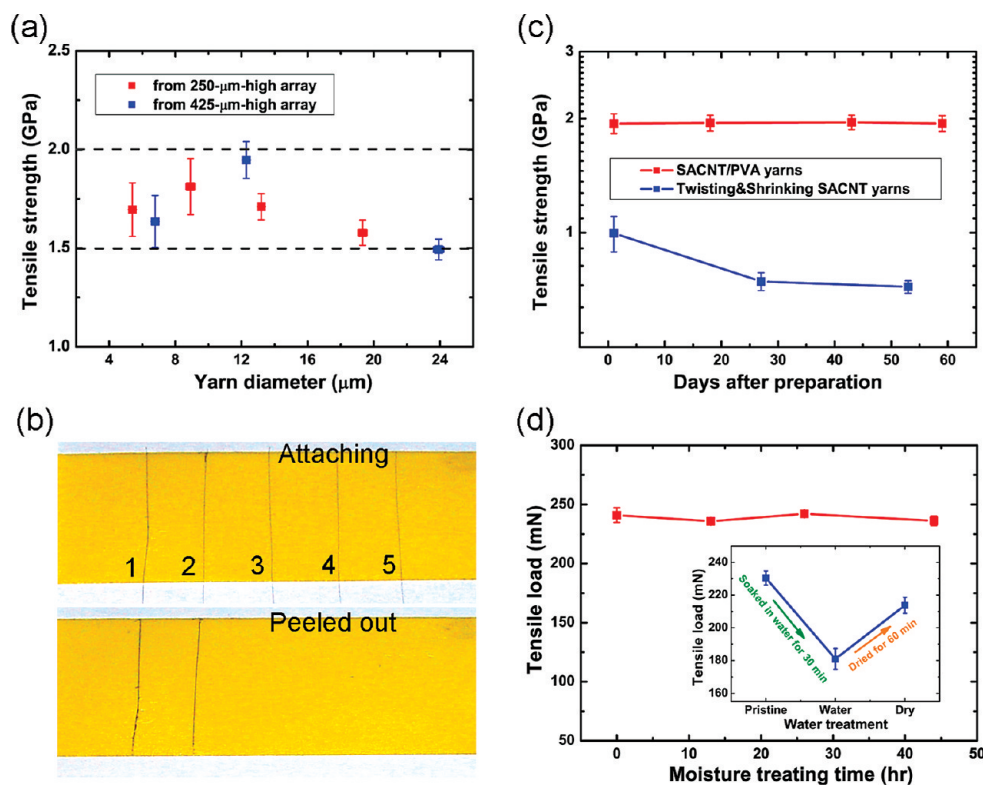
Figure 4. Merits of SACNT/PVA yarns. (a) Tensile strengths of SACNT/PVA yarns with various diameters made from two arrays with different heights. The dashed lines indicate the tensile strength values of 1.5 and 2.0 GPa, respectively. **(b)** Comparison of a simply twisting SACNT yarn (labeled by "1"), a twisting and shrinking (by acetone) SACNT yarn (labeled by "2"), and SACNT/PVA yarns made from 1, 3, and 5 wt % PVA/DMSO solutions (labeled by "3", "4", "5", respectively). These yarns were first attached on a 3 M adhesive tape and then peeled out. **(c)** Changes of tensile strengths of a SACNT/PVA yarn and a twisting and shrinking (by acetone) SACNT yarn with the time after preparation. **(d)** Antimoisture property of SACNT/PVA yarns. The panel shows the tensile loads of SACNT/PVA yarns after being treated under an extreme humid ambient at 20 °C and RH 80% for different times; the inset shows the change of the tensile load of a SACNT/PVA yarn first soaked in water for 30 min and then dried for 60 min.

tions. With increasing the concentration of the PVA/DMSO solution, the PVA content in the produced SACNT/PVA yarn increases accordingly (Figures S2 and S3, Supporting Information), but the yarn diameter and the tensile load do not vary monotonously. As shown in Figure 3c, the yarn diameter first decreases as the PVA concentration increases in the range of 1−5 wt %, while further increasing the PVA concentration increases the yarn diameter. Compared with the yarn diameter, the tensile load of yarn shows an adverse dependence on the PVA concentration. With increasing the PVA concentration, the tensile load first increases when the PVA concentration is lower than 5 wt % and then decreases when the PVA concentration is greater than 7 wt %. These results show that it requires a certain amount of PVA to draw CNTs close enough to make PVA and CNTs effectively couple for strengthening a yarn (*i.e.*, the case of <5 wt %). However, as more PVA occupies more spaces in a SACNT/PVA yarn, the yarn becomes weakened, because PVA is not a strong component in the composite yarn (*i.e.*, the case of >5−7 wt %).

In our experiments, therefore, the strongest SACNT/PVA yarn is obtained by using a 5 wt % PVA/DMSO solution at room temperature. The excellent mechanical properties of SACNT/PVA yarns can be summarily attributed to three advantages of PVA/DMSO solution: (1) the wettability between DMSO and CNTs makes the PVA/DMSO solution capable of infiltrating the spaces inside a yarn; (2) the large surface tension of DMSO makes it shrink a yarn to a large extent, favoring the action of PVA; and (3) the sticking property of PVA further reduces the cross-sectional area of the yarns, and the forming PVA/CNT interface coupling increases the tensile load of yarns. These advantages make PVA/DMSO solution a unique system for enhancing the strength of yarns.

Our method is widely applicable for enhancing the strength of SACNT yarns. We tested two SACNT arrays with heights of 250 and 425 μm and spun yarns with various diameters from the two arrays. As shown in Figure 4a, the diameters of as-produced composite yarns have a wide range from 4 to 24 μm, but the tensile strengths of these yarns all fall in the range of 1.5−2.0 GPa, and their moduli are more than 70 GPa. Therefore, by our method, it is easy to produce high-strength SACNT/PVA yarns with a desired diameter. Our method can also be expected to have a good strength enhancement for other kinds of CNT yarns.

These SACNT/PVA composite yarns are quite different from the pure SACNT yarns in their micro structures. In the SACNT yarns, CNTs interact with each other *via* weak van der Waals forces. In the SACNT/PVA yarns, however, CNTs are assembled together by PVA. The PVA/CNT interface coupling is very strong, and thus SACNT/PVA yarns have much higher load-transfer efficiency than SACNT yarns. Due to this property, SACNT/PVA yarns not only possess excellent strength and modulus but also have some other merits. First, SACNT/PVA composite yarns are more scratch-resistant than pure SACNT yarns. This is because in SACNT yarns, outmost CNTs are easily broken by other surfaces, such as human hands and an adhesive tape, due to the weak interaction among CNTs, but in SACNT/PVA yarns, PVA wraps and sticks CNTs closely enough to protect CNTs from being destroyed (Figure 4b). Second, in the lab environment (~25 °C, relative humidity 10−50%), SACNT/PVA yarns retain their excellent mechanical properties, while SACNT yarns weaken gradually. We measured samples made from a 3 m long SACNT/PVA yarn in a period of about 2 months. The tensile strengths of the samples, in the range of 1.87−2.01 GPa, are unchanged during this long time period (Figure 4c). As a contrast, the tensile strengths of a SACNT yarn reduce evidently (Figure 4c), and the diameter of the yarn increases slightly (Figure S4, Supporting Information) within the two months after preparation. This indicates the composite structure in a PVA/SACNT yarn is very stable in the lab environment, while the weak van der Waals interaction among the CNTs in a SACNT yarn is not stable, and thereby the micro structure in a SACNT yarn is easily changeable.

Because PVA and CNTs are insoluble in most organic solvents, SACNT/PVA yarns should resist ordinary organic solvents, such as ethanol, methanol, acetone, and benzene. However, because PVA is water soluble, one question is that if the yarns can resist moisture or water. We put a SACNT/PVA yarn in an extremely humid environment (40 °C and relative humidity 80%) and then measured its tensile strength approximately every 8−10 h. As shown in Figure 4d, the yarns do not have any evident change in tensile load, showing that they are quite resistant to moisture. This antimoisture property may be because CNTs are hydrophobic and thus protect the PVA from being taking away by moisture. However, if we further soaked a SACNT/PVA yarn in water for half an hour and measured it immediately, we found that the tensile load of the yarn reduced by about 20%. After the yarn was dried, its tensile load recovered to some extent though still not to the original value (the inset of Figure 4d). These results show that the SACNT/PVA yarns cannot work well under water but can in a humid environment. This shortcoming can be overcome with coating an antiwater layer on the surface of yarns.

The SACNT/PVA yarns are also highly conductive. Compared with a twisting and shrinking (by acetone) pure SACNT yarn, the electrical resistivity decreases by 14% to $(1.09 \pm 0.04) \times 10^{-5}$ Ω m (corresponding to an electrical conductivity $\sim 9.2 \times 10^4$ S m^{-1}) due to the reduction of the yarn diameter (Figure S5, Supporting Information), which is comparable to those of HNO$_3$-treated or Au-coated SACNT yarns ($9.6-11 \times 10^4$ S m^{-1}).[21] These results show that a small amount of PVA does not have a negative influence on the framework of

(a)

0.5 mm

(b)

E

E=0.21 V/cm, T=30.6 °C

E=0.42 V/cm, T=36.4 °C

E=0.63 V/cm, T=50.2 °C

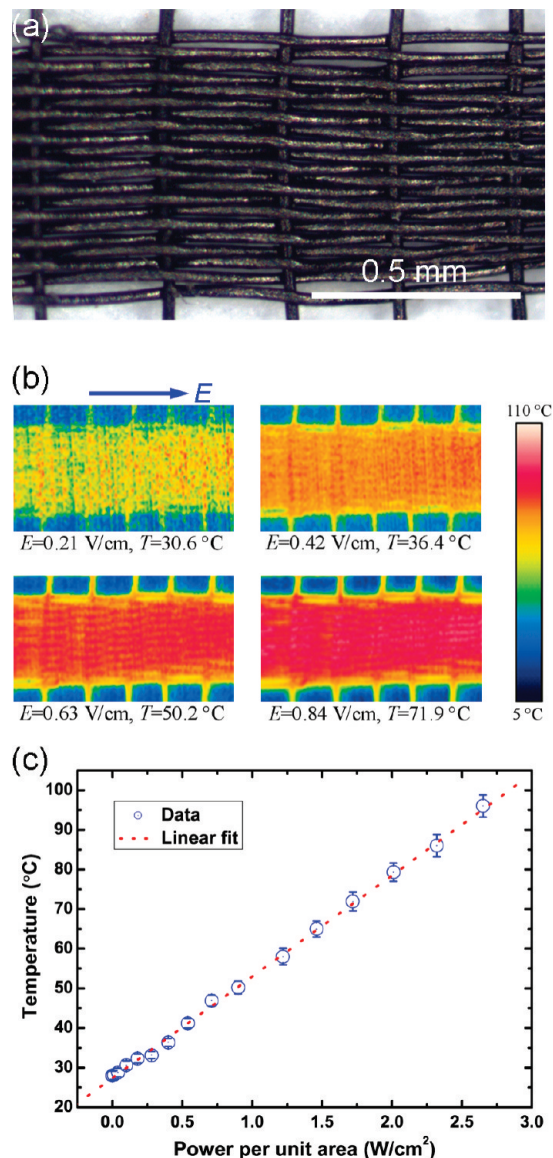E=0.84 V/cm, T=71.9 °C

110 °C

5 °C

(c)

○ Data
⋯⋯ Linear fit

Figure 5. Conductive fabric woven using SACNT/PVA yarns. (a) Optical image of part of a woven fabric which consists of 21 densely aligned yarns at horizontal direction and 5 sparsely aligned yarns at perpendicular direction in the visual field. The yarn diameter is about 30 μm. (b) Photos of temperature distribution of the woven fabric under different voltages along the horizontal direction, recorded by a thermal infrared imaging camera. The corresponding electric field E and average temperature T of the woven fabric are indicated. (c) Relationship between the temperature of the woven fabric and the input power per unit area. The room temperature when carrying out these measurements is about 28 °C.

SACNT yarns; on the contrary, because of the great reduction of the sectional area of yarns, the SACNT/PVA yarns possess a better electrical conductivity.

The advantages of good conductivity, excellent mechanical properties, scratch-resistance, lightweight, flexibility, and stability make SACNT/PVA yarns quite unique. Compared to carbon fibers which are strong and rigid, the SACNT/PVA yarns are superior in flexibility; while compared to some strong polymer fibers, such as aramid fiber (Kevlar) and poly p-phenylene benzobisthiazole (PBO), they have much better conductivity. These good properties of SACNT/PVA yarns make them capable of being woven into conductive fabrics. Figure 5a shows a small piece of fabric prototype. If a small voltage is applied on it, the fabric is heated with a uniform temperature distribution for the most part (Figure 5b). A small electric field 0.42 V cm^{-1} can raise the temperature of the fabric to 36.4 °C, which is near to the human body temperature; further increasing the electric field to 0.84 V cm^{-1} will cause the temperature of the fabric to increase to more than 70 °C (Figure 5b). The temperature of the fabric increases with an increasing applied voltage and linearly relates to the calculated input power per unit area (Figure 5c). Because SACNT/PVA yarns are strong and scratch-resistant, the woven fabric consisting of SACNT/PVA yarns can be used as high-strength and heatable fabrics, which shows potential applications in flexible heaters, bullet-proof vests,

and spacesuits. And because CNTs are an ideal blackbody that absorbs a wide range of electromagnetic waves, this kind of woven fabric is also expected to be used in radiation protection suits.

In summary, by using a continuous twisting SACNT yarn as a conductive framework and inserting PVA into the intertube spaces of the SACNT framework through PVA/DMSO solution to enhance the strength of the yarn, we are able to produce flexible, lightweight, high-strength, and highly conductive SACNT/PVA composite yarns. The key point to obtain such high-performance composite yarns is the usage of DMSO, which has a high surface tension and good wettability with CNTs, for dissolving PVA. The as-produced yarns possess very high tensile strengths up to 2.0 GPa, high Young's moduli of more than 120 GPa, and good electrical conductivity of 9.2×10^4 S m^{-1}. Because of the strong CNT/PVA interface coupling, the SACNT/PVA yarns are more scratch-resistant and stable than the pure SACNT yarns in the lab environment and retain their excellent mechanical property even in an extremely humid environment. Our spinning method is simple, continuous, and cost-effective. The SACNT/PVA yarn can be woven into high-strength and heatable fabrics with a large rise in temperatures even under small voltages, showing the potential applications in flexible heaters, bullet-proof vests, radiation protection suits, and spacesuits.

METHODS

Spinning Process. Before spinning, a laser was used to etch two parallel lines on a long strip of SACNT array at a desired separation distance to ensure a uniform effective width of the array.[23] As shown in Figure 1a, the long strip of SACNT array was attached to and rotated with a high-speed rotating motor (labeled by "1" and "2"), so that the SACNT film drawn out from the array was twisted into a yarn by the rotating motor ("3" and "4"). The twisting yarn then passed through the PVA solution ("5") on a heating plate ("6"), and was dried at 150 °C by a tube furnace ("7"), and finally collected on a winder ("8" and "9"). The yarn diameter is controlled by tuning the effective width of the SACNT array after laser etching, and the twisting angle of a yarn is controlled by varying the ratio of the twisting speed to the collecting speed. The spinning setup is slightly different from that of spinning pure SACNT yarns in our previous study[23] (see Section 1 in Supporting Information), but it can also be used to produce pure SACNT yarns, including simply twisting yarns and twisting and shrinking yarns. If there is no solution in the vessel (labeled by "5" in Figure 1a), i.e., no solution treating process, then the produced yarn is simply twisting. If the vessel is filled by some solvents, such as acetone or DMSO, then the produced yarn is twisting and shrinking (by acetone or DMSO).

Characterizations. The diameter of each yarn was obtained statistically by observing at least five positions under a scanning electron microscope (SEM, FEI Tecnai 100). The stress–strain curves were recorded by an INSTRON 5848 MicroTester at a gauge length of 20 mm and a stretch rate of 0.4 mm min^{-1}; at least five samples taken from the same yarn were measured to obtain the tensile strength and Young's modulus of the yarn. TGA measurements were carried out using a NETZSCH STA 449C by heating samples from 50 to 900 °C at a rate of 10 °C min^{-1} in air. The antimoisture experiment was carried out by first treating yarns under a KSON LabTester in which the temperature

and humidity is set as desired and then measuring their stress–strain curves. The thermal infrared imaging for measuring the temperature distribution of the woven SACNT/PVA fabric was carried out using a FLIR A40.

Supporting Information Available: Additional information for the spinning setup; thermogravimetric analysis (TGA) of PVA and yarns; SACNT/PVA yarns made from different concentrations of PVA/DMSO solution; change of the diameter of a twisting and shrinking (by acetone) yarn with time; and electrical resistivity of SACNT/PVA yarns. There are five figures (i.e., Figure S1–S5). This material is available free of charge via the Internet at http://pubs.acs.org.

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