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Triboelectric nanogenerators and power-boards from cellulose nanofibrils and recycled materials

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ABSTRACT

This paper reports the implementation of renewable, biodegradable, and abundant cellulose nanofibrils (CNFs) in triboelectric nanogenerator (TENG) development. Flexible and transparent CNF thin films are triboelectric positive material with nanoscale surface roughness. They are paired with FEP (fluorinated ethylene propylene) to assemble TENG devices, which exhibit comparable performance to the reported TENG devices built on synthetic polymers. CNF-based TENG is further integrated within a fiberboard made from recycled cardboard fibers using a chemical-free cold pressing method. The fiberboard produces up to ~30 V and ~90 μA electric outputs when subjected to a normal human step. This development shows great promises in creating large-scale and environmentally sustainable triboelectric board for flooring, packaging and supporting infrastructures from CNF and other natural wood-extracted materials.

1. Introduction

Mechanical energy from the environment (e.g. ambient vibrations or activities of human body) [1,2] represents a unique and sustainable energy source that perfectly suits for powering portable electronics and unattended devices [2–4]. In 2012, the triboelectric nanogenerator (TENG) was first reported [5] as a new type of technology that converts environment mechanical energy into electrical energy [3]. Compared to other technologies, TENG is advantageous in terms of high efficiency, high power density, light weight, low cost, and great manufacturability [2,6–8]. TENG operates based on the coupling effects of contact electrification and electrostatic induction. The working principle requires two dissimilar surfaces to be oppositely charged upon contact. The common positive materials in TENG are polyamides, [9] metals, [2] indium tin oxide (ITO), [3] and zinc oxide, [10] and negative materials include fluorinated ethylene propylene (FEP), [11,12] polytetrafluoroethylene (PTFE) [9] and polyvinylidene fluoride (PVDF), [2] polydimethylsiloxane (PDMS), [3,10,13,14] and polyethylene terephthalate (PET) [8,13–15]. The positive and negative materials are typically used in pairs in TENGs due to their opposite tendencies to gain or lose charges upon contact.

Cellulose, the most abundant natural polymer on Earth, can also be a good candidate as a triboelectric material. Cellulose nanofibrils (CNF) thin films, which are assembled from nanosized cellulose fibrils, are highly transparent, [16–18] biodegradable, [18] flexible, [19] and possess desirable surface roughness and electrical properties [18,20]. As a result, CNF thin films are increasingly explored in modern electronic and optoelectronic devices [3,7,8,13,18]. Due to the inclusion of abundant oxygen atoms, natural cellulose exhibits high tendency of losing electrons and thus becomes positively charged [21]. Therefore, it can be paired with a wide range of triboelectric negative materials, such as FEP, PTFE, and PET to assemble TENGs. However, although recent work has predicted the promise of using cellulose as an actively TENG material, [22] no practical TENG development on cellulose has been reported so far. In addition, cellulose can also be chemically modified to further tune their electronegativity. This paper reports the implementation of CNF materials in TENG development. Flexible and transparent CNF thin films with nanostructured surface were paired with FEP (fluorinated ethylene propylene) and operated in the vertical contact-separation mode. The TENG device exhibited comparable performance to the reported TENG counterparts built on synthetic polymers. The CNF-based TENG was also integrated within fiberboard made from recycled cardboard, creating a power board with high electric outputs using environmental-friendly wood-extracted materials.

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2. Experimental section

2.1. Fabrication of CNF film

First, CNF hydrogel was prepared from wood pulp by tetramethylpiperidine-1-oxy (TEMPO)-mediated oxidation and subsequent mechanical homogenization following the method from Saito et al. [23] Commercially supplied, bleached kraft eucalyptus pulps were oxidized in a mixture of TEMPO, sodium hypochlorite (NaClO), and sodium chlorite ($NaClO_2$) under pH 6.8. The oxidation was carried out at 60 °C for 48 h. Oxidized pulps were thoroughly washed in distilled water and refined in a disk refiner to break apart the residual fiber bundles. The refined fibers were then separated by centrifuge to remove the supernatant fraction, and concentrated to 1 wt% using ultrafiltration. Finally, this suspension was subjected to high pressure mechanical homogenization by passing through a series of 200- and 87-μm chambers on a microfluidizer for three times (M-110EH-30 Microfluidizer, Microfluidics, Newton, MA, USA). The resulting mixture of nanofibrils and water formed a transparent, stable aqueous colloid system (with a cellulose solid weight of 1%), i.e., CNF hydrogel. To obtain the flexible and transparent CNF film, the as-processed CNF hydrogel was diluted with DI water and then filtered under approximately 0.55 MPa air pressure in a filtration system (Millipore Corporation, USA). In the filtration chamber, water within the slurry passed through a polytetrafluoroethylene membrane (0.1 μm pore sizes), leaving the CNF filter slab. After separation from the membrane, the CNF slab was sandwiched between layers of waxy coated paper, filter paper, and caul plates for room temperature drying followed by 65 °C oven drying. During drying, pressure was applied on top by weights to prevent warping and wrinkling. Consequently, we obtained the flat, transparent and flexible CNF film.

2.2. Assembly of CNF film TENGs

The TENG used in demonstrating the design and performance of CNF-based TENGs was fabricated as follows: a pure CNF film with a size of 1 cm×1 cm was attached to the center of an ITO/PET substrate (2 cm×5 cm), which was considered as the top electrode. Bottom electrode comprised fluorinated ethylene propylene (FEP) with the same size and location on the other ITO/PET substrate. The two electrodes were separated by 1 mm spacers, and connected to the external circuit through copper tapes. Thickness of the CNF was 320 μm; while thickness of the FEP was 60 μm. TENG devices with various active areas from 1 to 40 cm^2 were used in the performance characterization.

2.3. Manufacturing of power fiberboard

Fibers recovered from cardboard were dispersed in water and formed a uniform mixture by mechanical stirring. The mixture was forced through a screen by vacuum sucking, leaving a wet mat. An 8 cm×8 cm CNF-based TENG with all peripheries sealed was embedded in the wet mat during vacuum sucking. Subsequently, the mat was sandwiched between release papers and coil plates, and sent to

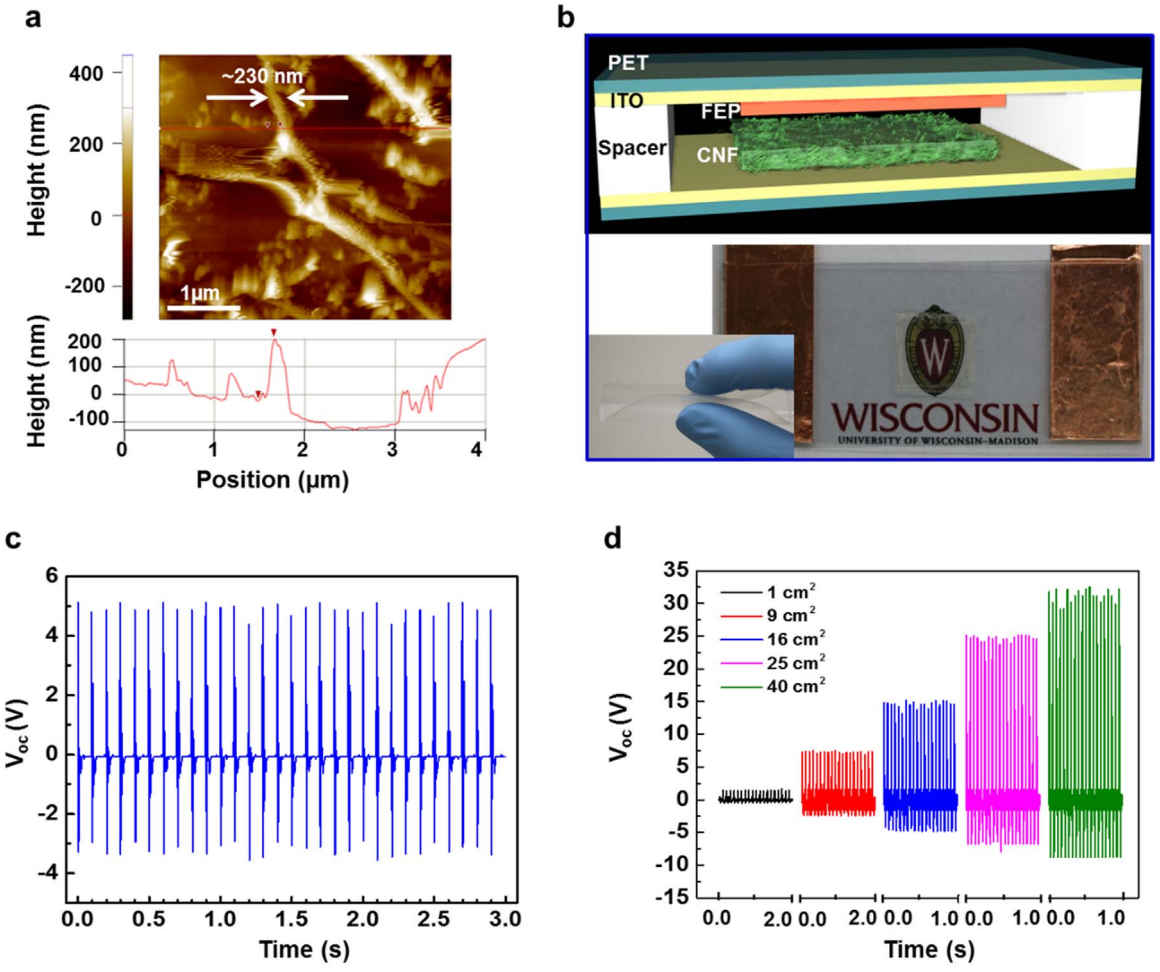

Fig. 1. Design and performance of CNF film TENG. (a) AFM topography image of a CNF film surface. **(b)** Schematic drawing (top) and a digital photo (bottom) of the CNF film-based TENG, inset shows the flexibility of CNF film. **(c)** Open circuit voltage (V_{oc}) from the CNF film-based TENG. **(d)** V_{oc} measured from CNF-based TENG with various active surface areas.