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Featured Letter

Electroluminescent nanocellulose paper

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ABSTRACT

Surface light-emitting transparent nanocellulose papers operating by alternating current voltages were demonstrated, where phosphors were embedded in the paper and silver nanowire electrodes were formed on both side of the paper. Silver nanowire electrodes were first coated on membrane filter with vacuum filtration using wet wiper tissues and then successfully transferred to the paper surface by a mechanical compression. The power consumption of a light-emitting paper was small in less than 0.36 W/cm^2. We could control the luminescent color and intensity from light-emitting papers by adjusting electrical frequency and the applied voltage level. White light emission was also demonstrated by using phosphors with a broad luminescence spectrum. The results develop into a versatile approach for commercializing the nanofibrillated cellulose and pave the way towards the realization of commercially viable, flexible and transparent light-emitting papers.

1. Introduction

Cellulose is very familiar in our lives because we are surrounded by cellulose products, including cosmetics, medicine, construction materials, and so on. In particular, cellulose from wood is used as the main materials forming newspapers, books, copy paper, tissue, clothes, etc. Cellulose paper consists of fibers with diameters of tens of micrometers. Using chemical pretreatments followed by high-pressure homogenization, micrometer-sized cellulose fibers can be disintegrated into nanofibrillated cellulose with a diameter of less than 20 nm [1]. Over and over passed through a high-pressure homogenizer, the diameter of cellulose fiber is decreased. By compressing the nanofibrillated cellulose pulp with the right composition in a sheet-former, highly transparent nanocellulose paper (NCP) or film with a large light scattering in the forward direction can be produced [2,3]. Recently, NCP has been widely studied for use in flexible devices as a recyclable, environmentally friendly, and sustainable material [4–12] in which NCP was mainly used as a passive component such as a substrate. In this work, we demonstrated surface light-emitting transparent papers operated by alternating current (AC) voltages. This work is the first to apply NCP as a light-emitting active component. For this, the main issue was to uniformly coat electrodes on both side of the NCP. To overcome this problem, we developed a vacuum filtration of silver nanowire (Ag NW) ink using wet wiper tissues. Because the wet tissue uniformly dispersed Ag NW ink, we were able to uniformly coat Ag NWs on a 80-mm dia. polyvinylidene difluoride (PVDF) membrane filter, which was transferred to the NCP surface.

2. Experimental section

Cellulose powder (about 20 μm), potassium hydroxide, and sodium chloride were purchased from Sigma-Aldrich. All chemical were used without further purification. PVDF membrane filters (diameter 90 mm and pore size 0.65 μm) were purchased from Durapore. 0.05 wt.% Ag NW ink (Nanopyxis, Korea) with diameters of 20–40 nm and lengths of 20–30 μm was diluted about one 20th with DI water. Green (GG45) and white (GG84) phosphors were purchased from Global Tungsten & Powders.

Nanofibrillated celluloses were made from 1 wt.% cellulose powders dispersed in DI water which were chemically treated by potassium hydroxide (2 wt.%) and sodium chloride (2 wt.%) at 60–70 °C for 2 h in order to remove lignin and hemicelluloses. After the chemical treatment, the samples were rinsed with DI water until the residues were neutralized. A centrifuge (2500 rpm for 3 min) reduced the rinse time because of no waiting time until the residues sank to the bottom. This water slurry was passed 10 times through a high-pressure homogenizer under 1,500 bar (Nano

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Disperser, Suflux) and then cellulose was nanofibrillated with diameters of 10–20 nm and lengths of 1–2 μm (Fig. S1). The diluted Ag NW ink of 3 ml was coated on PVDF filter placed on a glass filter of the vacuum filtration equipment (KG-90, Advantec). To uniformly coat Ag NWs, we covered a piece of wet wiper tissue (Kimtech) on the PVDF filter (Fig. S2a-b). The coated Ag NW electrodes showed a sheet resistance of 30–40 Ω/sq.

Nanofibrillated cellulose suspension was dispersed in DI water at a fiber content of 0.1 wt.% and the dispersed suspension of 250 ml was mixed with luminescent phosphors. The concentration of phosphors was the weight percent against the pure cellulose weight. The mixed suspension of 250 ml was vacuum filtered for 30 min at room temperature using Ag NW coated PVDF membrane filter. The filtered nanocellulose sheet with luminescent phosphors was covered with Ag NW coated PVDF membrane filter and pressed under 7 MPa at 30–40 °C for 12 h. After that, Ag NWs were transferred onto both side of the nanocellulose sheet, showing the same resistance as that of Ag NWs coated on PVDF filter.

The transmittance of NCPs was measured with an ultraviolet–visible spectrometer (100 Conc, Cary) and scanning electron microscopic images were obtained using an FEI Sirion 400 operating at 10 keV. Luminescence was characterized by a spectrometer (USB 2000+, Ocean Optics). For photoluminescence (PL), He-Cd laser (Kimmon) with 325 nm was used as an excitation source. For electroluminescence (EL), a function generator (AFG 3022C, Tektronix) with a power amplifier (4010, NF Electronic Instruments) was used as an electric power source and a digital multimeter (34461A, Agilent) was also used to monitor root-mean-square AC voltage and current. Conductive fabric tape (RGW series, MTC) was used as electrical contact pads on both side of NCP. Luminance was measured using a chroma meter (CS-100, Minolta).

3. Results and discussion

A vacuum filtration using wet wiper tissues is fast due to using a vacuum pump and reduces the ink drying time in less than 30 s. These Ag NW electrodes were perfectly transferred onto the NCP after compression and Ag NWs are not nearly remained on a PVDF filter after transferred to NCP (Fig. S2c). The direct coating of Ag NWs on a substrate or a conductive composite of Ag NW/cellulose nanofibril using a vacuum filtration was previously reported [13–15]. However, because a paper (or filter) easily absorbs solutions, the coating of Ag NWs in a solution on a wide paper is very difficult. Using a vacuum filtration can solve this problem, but a direct filtration on NCP is unable to uniformly coat Ag NWs on both surface sides of the NCP. We solved the problems by using direct transfer of Ag NW electrode from PVDF filter to NCP.

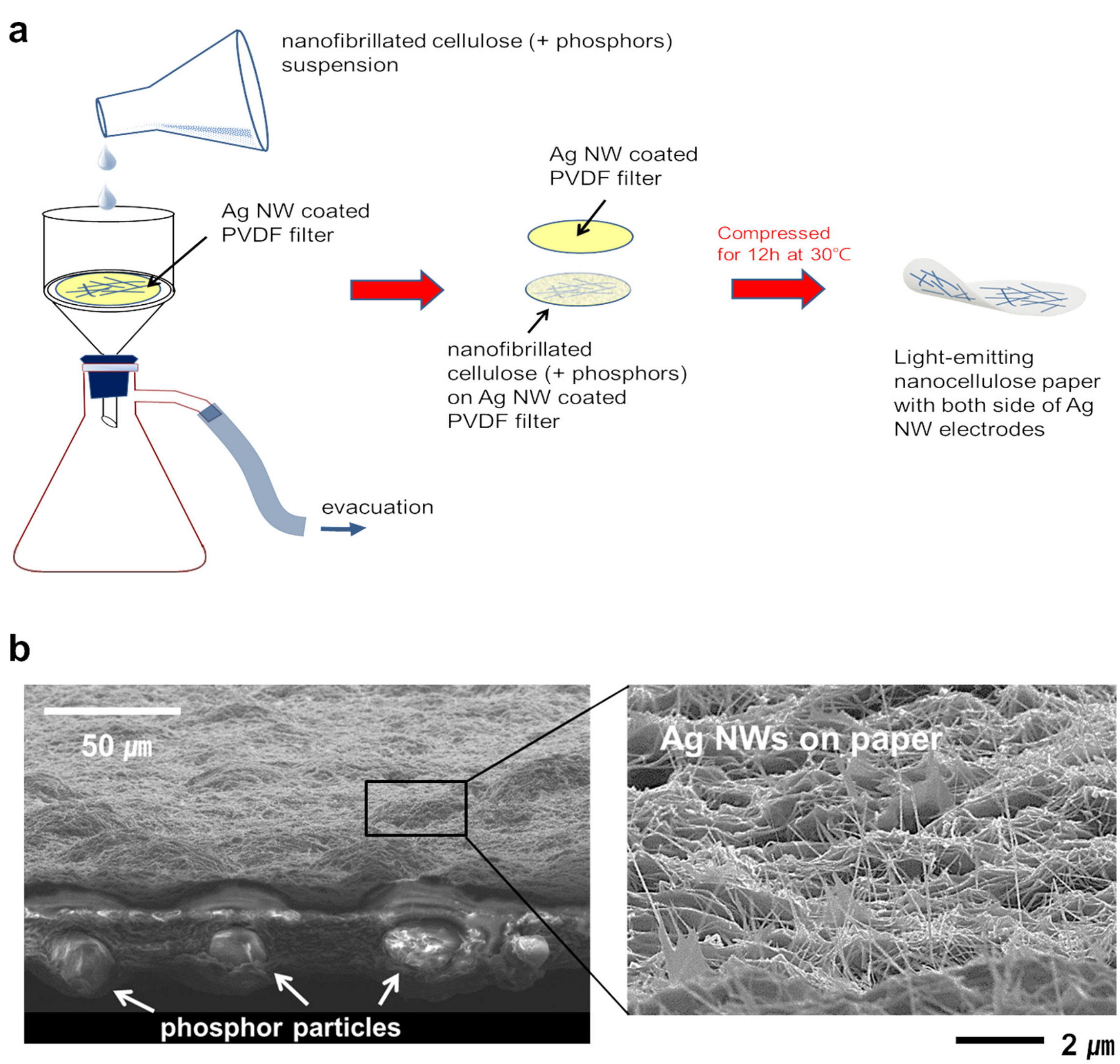

Fig. 1. (a) Schematic illustrations of the fabrication of a light-emitting transparent NCP. (b) Scanning electron microscopic images of the cross-section and the surface high-resolution of light-emitting transparent NCP.