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Cellulose optical fiber for human health monitoring

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ABSTRACT

Cellulose, as a fully renewable, biodegradable, and biocompatible material, creates new possibilities for optical fiber (OF) sensor applications. Cellulose OFs are highly hygroscopic, exhibiting rapid wetting and drying properties with water and moisture, easily functionalized, and can be either made water-resistant or water-soluble. These fibers are not aimed towards replacing the existing glass or polymer OFs in telecommunication or in current sensing applications, rather cellulose OF sensors can open new application areas where the reactive materials are required. For example, compared to glass or polymer OFs, cellulose OFs are porous and allow liquid transport in and out of the fiber. Moreover, the cellulose waveguide material itself can be chemically functionalized. Such cellulose OFs fit well with human health monitoring where the new possibilities that cellulose offers can be utilized. Here we demonstrate a face mask that contains regenerated cellulose (RC) OF with a 1.8 dB/cm attenuation constant for respiratory rate monitoring. RC is a class of man-made cellulose materials that includes commonly materials like rayon textiles and cellophane films. The cellulose fiber inside the face mask rapidly absorbs moist and dries between each breath, which causes a periodic change in optical power transmitted through the fiber. A face mask does not prevent fast drying of the fiber. Such RC fiber fits well to respiratory rate monitoring because it exhibits good mechanical performance in both dry and wet states. Cellulose OF length was about 5 cm long with a loop-type sensor structure. Measured respiratory rates varied between 16-54 breaths per minute.

Keywords: Cellulose, regenerated cellulose, optical fiber, respiratory rate, sensor

1. INTRODUCTION

Optical fibers (OFs) are not only the backbone of modern telecommunications but they are applied in various sensing applications such as strain, displacement, temperature and bending. OFs have capability for humidity [1-4] and pressure measurements [5]. OF pressure sensors are demonstrated in respiratory rate monitoring [6] and they are often connected to smart textiles [7-10]. There are also non optical sensors for respiratory rate monitoring like electrical moisture measurement with a paper based sensor [11] and a piezoresistive strain sensor [12]. Optical respiratory rate sensors are needed for example in magnetic resonance imaging (MRI) applications where electrical sensors cannot be used due to electrical interference [13, 14].

Commonly used glass OFs and polymer OFs are typically employed in sensor applications but novel materials are being explored to fabricate OFs targeting specific new sensing applications. Biocompatible and implantable OFs have been presented for human health applications [15]. One such material is also cellulose, the most abundant biopolymer on Earth and the structural backbone of all plants and trees. Cellulose is a fully renewable, biodegradable, and biocompatible material which offers exciting properties for sensing applications aimed at human health monitoring such as high hygroscopicity, rapid wetting and drying kinetics with water and moisture, and it can be either made water-resistant or water-soluble.

The first cellulose OF was reported by Orelma et al. in 2020 where regenerated cellulose (RC) fibers were prepared using wet spinning of cellulose dissolved in ionic liquid (IL) [16]. The RC fiber core was cladded with a layer of cellulose acetate to realize an optical fiber. This work was followed by Reimer et al. who developed an OF based on a RC fiber core and cellulose acetate butyrate as a cladding material [17].

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Subsequent works by Hynninen et al. and Jaiswal et al. showed the preparation of OFs from cellulose derivates such as methyl cellulose and carboxymethyl cellulose via the hydrogel route, i.e., using fully water-based systems [18, 19]. Cellulose OFs can also be prepared in a cladding-free architecture where the fiber core is exposed to the environment where it can act as a sensing element. Moreover, cellulose OFs are porous and allow liquid and gas transport in and out of the fiber. Sensing capability of cellulose OFs has been demonstrated in the form of water sensing [16], mercury ion sensing [18], touch and respiratory rate sensing [19], pressure and bending sensing [20], and spectroscopic sensing [21].

In this work, we report the fabrication of RC OFs via dissolution of pure cellulose into an IL followed by wet spinning of the cellulose solutions into water. Two different cellulose raw materials have been used for the fiber preparation and the OFs are used without any cladding. Furthermore, we investigate the waveguiding properties of the prepared OFs in terms of their signal attenuation and the transmission window. In our earlier work, carboxymethyl cellulose OFs have been demonstrated for respiratory rate measurements with loop-type and reflection-type measurement modes [19]. Herein, we employ the same rapid moisture-desorption kinetics of the RC OFs in respiratory rate sensing which was demonstrated by embedding the RC OF in a face mask.

2. METHODS

2.1 Fabrication of optical fibers

RC optical fibers were prepared via the dissolution and regeneration route. Two different cellulose sources were used, namely microcrystalline cellulose (MCC) and dissolving pulp (DP) (Figure 1a). The MCC was purchased from Merck GmbH (product code: 22182) and the DP was provided by Domsjö Fabriker, Sweden). The DP material was provided as a once-dried pulp sheet which was grinded to powder form using a Fritsch mill. The cellulose solvent used in this study was an IL, namely 1-ethyl-3-methylimidazolium acetate (EMIMOAc), purchased from IoLiTec GmbH, Germany (>95% purity). EMIMOAc is a direct dissolution solvent for cellulose, meaning that no intermediate cellulose derivatives are required to enable dissolution. The intrinsic viscosity of the cellulose raw materials was measured using ISO 5351:2004 protocol.

Cellulose dissolution

Both MCC and DP celluloses were dissolved in EMIMOAc using the same protocol. A calculated amount of IL was added to a 250 ml round bottom flask which was installed on a carousel reactor (Radleys Co.). A calculated mass of the dry cellulose powder was then added to the IL under constant stirring. The reactor was sealed and reaction temperature set to 80°C with Teflon impeller stirring for 18 hours. The concentration of cellulose in the dissolved system (termed as ‘dope’) was 5% (w/w). The prepared dopes were then removed from the reactor flask and poured into glass containers where they were degassed and dried inside a vacuum oven at 80 °C for 12 h and then stored in closed glass bottles in room temperature until use.

Fiber spinning

RC OFs were prepared using wet spinning. The regeneration (often called spinning) bath was filled with pure reverse osmosis water. The cellulose dopes were pre-heated in an oven to 80°C and then filled in a syringe with a Luer lock-type tip, carefully avoiding bubble formation. The syringe was heated using an electrically heated jacket (New Era Pump Systems Inc.) to maintain a constant dope temperature during fiber spinning. The dope temperature was set to 80 °C for MCC-15G and DP-15G samples and 90°C for the MCC-17G sample. Then a stainless steel spinning needle (Hamilton Co.) was attached to the tip of the syringe. The entire assembly was then loaded on a syringe pump (New Era Pump Systems Inc.) which was placed vertically, and the height of the apparatus was adjusted such that the spinning needle was submerged into the water bath and the extrusion of the liquid dope into the regeneration bath was initiated (Figure 1b). The volumetric flow rate of the dope was fixed at 5 ml/min.

As the extruded dope filament came in contact with water (a non-solvent to cellulose), the cylindrical shape of the filament was immediately preserved while the IL solvent started to migrate out of the filament. The spun cellulose fibers were kept in the regeneration bath for 10 min during which sufficient IL had migrated out of the fibers and the fibers could be mechanically handled. Approximately 2 m long fibers were then transferred to a washing bath which contained fresh water and kept submerged in the bath for 2 hours during which all the remaining IL diffused out of the fiber, resulting in a RC
fiber which was water-swollen. The next step was to dry the RC fibers to obtain solid OFs. The fiber samples were dried under tension where both ends of the fiber were fixed to a solid support to avoid shrinkage in the longitudinal direction while the drying shrinkage in the radial direction was possible. The fibers were dried in room temperature overnight. The final fiber length was 20-30 cm (Figure 1c) in the batch process but longer continuous fibers can be produced on a roll-to-roll fiber spinning machine. The RC fibers had uniform circular cross-sections which are shown in Figure 1c inset. The fiber diameter was measured using an optical microscope. The sample description is given in Table 1 where ‘G’ in the spinning needle description denotes the standard needle gauge. The 15G needle has an outer diameter of 1.829 mm, an inner diameter of 1.372 mm, and wall thickness of 0.229 mm, and corresponding values for 17G needle are 1.473 mm, 1.067 mm, and 0.203 mm.

<table>
<thead>
<tr>
<th>Sample name</th>
<th>Cellulose type</th>
<th>Spinning needle</th>
<th>Avg. fiber diameter (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MCC-15G</td>
<td>MCC</td>
<td>15G</td>
<td>375</td>
</tr>
<tr>
<td>DP-15G</td>
<td>DP</td>
<td>15G</td>
<td>500</td>
</tr>
<tr>
<td>MCC-17G</td>
<td>MCC</td>
<td>17G</td>
<td>256</td>
</tr>
</tbody>
</table>

Table 1. Description of different cellulose OF samples. ‘G’ in the spinning needle description denotes the standard needle gauge.

2.2 Attenuation and transmission spectrum measurements

Attenuation and transmission spectrums were measured as was shown in our previous works [1, 4, 6]. The attenuation was measured with 637 nm laser light (Thorlabs, S4FC637) and with 1050 nm SLED (Thorlabs, S5FC1050P). Light sources were coupled to RC fiber with a single-mode (SM) fiber (9/125μm core/cladding diameters). Transmitted signals were collected with a power meter interface (Thorlabs, PM101) and a silicon photodetector (Thorlabs, 120C) using a multimode (MM) optical fiber (105/125 μm core/cladding diameters).

The transmission spectrum was measured with halogen lamp (Ocean Optics, HK-2000-HP) that was coupled to RC fiber with MM fiber (105/125 μm core/cladding diameter). Transmitted light from RC fiber was collected to Optical Spectrum Analyzer (OSA) (Ando, AQ-6315A) with MM fiber (400/425 μm core/cladding diameter). Transmission spectrum from 350 to 1750 nm was defined using 10 nm resolution.

2.3 Respiratory rate sensor embedded in face mask

As a proof-of-concept demonstration, a cellulose OF respiratory rate sensor was embedded inside a commercial nonwoven polypropylene face mask (Figure 2a). Respiratory rates were measured with a wireless measurement setup with a battery operated laser (Tuscon Optic, TC-10-50, 650 nm, 50 mW) and a silicon photodetector (Thorlabs, PM160, 400-1100 nm). The signal from the detector was collected with Bluetooth connection using an Android mobile phone application (Thorlabs, Optical Power Meter). Laser light was coupled to RC fiber MCC-17G (Figure 2b) with MM glass optical fiber (105/125 μm core/cladding diameters) using 1.25 mm diameter sleeves and ferrules from Thorlabs (Figure 2c). Transmitted
light was collected with the same 1.25 mm diameter sleeves/ferrules and MM glass optical fiber to the photodetector. By combining cellulose fiber with standard MM glass optical fibers with low attenuation, long optical connections are possible.

Figure 2. (a) RC OF-based respiratory rate sensor embedded in a face mask, (b) operating principle of the sensor, c) zoomed image of the RC fiber with MM glass optical fiber connections.

3. RESULTS

3.1 Attenuation measurements

The cross-sections of the RC OFs were well-defined and circular which indicated isotropy in the fibers in the radial direction (Figure 1c). The cellulose dope was extruded directly into the water bath in the form of a circular filament (circularity controlled via the shape of the spinning needle) which was immediately immobilized into a circular shape. The fibers were also dried with their ends attached which allowed drying shrinkage developed uniaxially in the radial direction and thus preserving the cross-section shape and isotropy.

Attenuation coefficient of the RC OFs was measured using the ‘cut-back’ method. In our previous work, we reported an attenuation coefficient of 6.3 dB/cm for RC fibers which were cladded with cellulose acetate [1]. In this work, we improved the fiber fabrication process and tested different cellulose raw materials for OF preparation. This led to improvement in the optical quality of the OFs as the attenuation coefficient reduced to 1.8 dB/cm at 1050 nm (Figure 3b). The reduction in attenuation can be attributed to the use of MCC as the raw material for fiber preparation. MCC has a much lower degree of polymerization as compared to the DP (300 vs. 2500). The shorter polymer chain length leads to complete and easier dissolution of the solid cellulose into the IL solvent. Moreover, the MCC dope has a much lower intrinsic viscosity as compared to the DP dope (200 ml/g and 450 ml/, respectively). The lower viscosity helps in easier shearing of the dope during the spinning process and aligning the polymer chains in the longitudinal direction of the fiber, leading to more dense fibers with lower attenuation coefficient (Figure 3a-b). The two MCC fibers, spun with different spinning needle gauges (15G and 17G) while all other parameters were kept constant, showed a difference in attenuation values. The thinner MCC-17G fibers showed a higher attenuation value than the MCC-15G fibers.
3.2 Transmission spectrum measurements

Transmission spectra for all three RC fibers were in the same range that we have shown earlier for other cellulose OFs [1, 4, 6]. The transmission range for these RC fibers was from 500 nm to 1400 nm (Figure 4). MCC-15G and MCC-17G have lower attenuation than DP-15G as was observed in the attenuation measurements in Figures 3a and 3b. Here, transmission spectra have been defined just for the shortest fiber length after the ‘cut-back’ attenuation measurement: 3.6 cm for MCC-
15G, 3.1 cm for DP-15G and 3.8 cm for MCC-17G. The absolute transmission loss curve height depends on the final RC fiber sample cutting and preparation. For instance, polishing of the fiber ends decreases light coupling variations between samples. However, polishing for the cut fiber ends was not used because in this study, we were interested in characterizing just the transmission ranges. Nonetheless, the transmission loss curves were in the same order than in the attenuation measurements shown in Figures 3a and 3b.

![Transmission spectra of the RC fiber samples.](image)

**3.3 Respiratory rate measurements**

Cellulose is an intrinsically hydrophilic and hygroscopic material in nature. RC fibers are composed of chemically unmodified cellulose (with small amounts of hemicelluloses) which readily interacts with liquid water and moisture. During application in respiratory sensing, a thin layer of moisture contained in the breath is adsorbed on the surface of the RC fiber, creating a transient ‘cladding’ on the fiber (Figure 2b). This moisture then rapidly evaporates and desorbs from the fiber surface. Therefore, the transmission signal from the OF shows a peak when the breath moisture is adsorbed and the signal returns to the ground level as the moisture desorbs. The kinetics of this entire cycle were found to be quick enough to measure respiratory rate in practical applications.

Respiratory rates were measured using the wireless measurement system and Optical Power Meter application on a mobile phone. A volunteer wore the face mask with the embedded OF respiratory rate sensor and performed three basic actions: sitting, walking at normal pace, and climbing stairs. All measurement results are shown here without any signal filtering or processing. The laser wavelength of 650 nm was selected to have a small visible portable light source in the respiratory rate sensor demonstration. It must be noted that infrared wavelengths would give better signal quality than visible ones because the attenuation is smaller in the near infrared range as can be seen from Figures 3a, 3b, and 4.

Resting state respiratory rate of 16 breaths/minute was measured after a few minutes of sitting (Figure 5a). The respiratory rate during walking was collected after a few minutes of walking at normal pace to get a stable signal (Figure 5b). During walking, the respiratory rate signal increased only to 18 breaths/minute because of the slow indoor walking pace. The maximum signal rate comfortable for the volunteer wearing the face mask was measured while walking on stairs (Figure 5c) where 54 breaths/minute were measured during quick ascent and descent on stairs through four office floors. There was again a couple of minutes of walking before the signal recording was started. Rapid breathing during climbing on stairs decreased the absolute signal change, but it was also the maximum comfortable rate with face mask for a few minutes sampling.
Figure 5. Respiratory rates measured with fiber RC MCC-17G in the face mask during a) rest state, b) walking at normal pace, and c) walking on stairs. Raw data is shown without any signal processing.
4. CONCLUSIONS

Regenerated cellulose (RC) optical fibers (OFs) were fabricated by dissolving and regenerating two different cellulose raw materials, namely microcrystalline cellulose (MCC) and dissolving pulp (DP). The RC OFs were characterized for their optical properties in terms of signal attenuation and transmission window in the visible and near infrared range. Using a low molecular weight cellulose raw material i.e., MCC, the attenuation of the RC OF was decreased to 1.8 dB/cm (at 1050 nm wavelength) from 6.3 dB/cm that we reported earlier. The RC OF was then embedded into a face mask to realize a respiratory rate sensor based on rapid wetting and drying of RC OFs from the moisture present in breath. The respiratory measurement was performed with a wireless setup that makes movements like walking possible during sampling. The slowest respiratory rate that was measured at the resting state was 16 breaths/minute and the highest value of 54 breaths/minute was measured during walking on stairs.

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