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Environmentally-friendly cellulose nanofibre sheets for humidity sensing in microwave frequencies

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ABSTRACT

This paper investigates environment friendly cellulose nanofibres (CNF) as new and sensitive material for humidity sensing in RF/microwave frequencies. CNF sheets were fabricated by sonocatalyzed TEMPO process and physically characterized. Humidity sensing investigation was performed with CNF sheets taped on the top of circuits in coplanar waveguide (CPW) technology. This investigation includes sensitivity and dynamic range analysis with reflected waves along the CPW circuit through resonant frequency shift, and transmitted waves through S21 phase shift. Moreover, sheets with various amounts of CNF were used to study the influence of CNF weight on humidity sensing performances. Regarding the resonant frequency shift, the best sensitivity was measured with the weightier CNF film (71 g/m^2), that is 2.82 MHz/%RH from 55%RH to 100%RH. Regarding the phase shift, the same film sensitivity is 0.7°/%RH from 70%RH to 100%RH, with a figure of merit of 7.43°/dB as a phase shifter.

1. Introduction

Humidity sensing has gained increasing interest in terms of becoming an essential assessment tool for a wide range of commercial and industrial applications including environment control, industrial processing and lifestyle improvement [1]. To meet industrial demand, the humidity sensors must satisfy the following requirements: low cost, high sensitivity, high selectivity and wide dynamic range. Given the high dependence of humidity detection systems on sensitive material properties, sensitive materials investigation has become a major field of research for the purpose of improving the commercial competitiveness of humidity sensors [2]. Various materials have been investigated using different technologies for humidity detection including ceramics, organic polymer and organic/inorganic materials [3]. Non-ionic but very polar polyimides and esters are suitable candidates for microwave humidity sensors whose sensing mechanism is based on the variation of dielectric properties as a function of humidity [4]. The effects of moisture on the dielectric constant of some organic polymers films such as Poly(vinyl cinnamate) (PVCi) or Poly(methyl methacrylate) (PMMA) have been investigated through resistive-type or impedance-type sensors in low frequencies [5,6].

Kapton polyimide and polyvinyl alcohol (PVOH) have been investigated for humidity sensing in RF/microwave frequencies, with the benefits of passive operation as well as integration with RFID technology [7,8]. The key advantage of Kapton is its compatibility with printing technologies [9,10] whereas PVOH has shown high sensitivity performances when used alone or when compared to Kapton [11,12]. A customized polymeric film synthetized from oxidianiline (ODA), *m*-pyromellitic dianhydride (*m*-PMDA), and *N*-methly-2-pyrrolidone (NMP) was also reported [13]. However, due to their fossil nature, all these materials may cause contamination when decomposing in nature. Therefore, it is essential to find more environmentally-friendly remedies to address this problem while maintaining attractive performances for microwave humidity sensing.

Cellulose is the most abundant organic polymer on earth and stands out as a good candidate for environmentally-friendly humidity detection. Cellulose was used as the host-matrix for ceramic particle encapsulation, or blended with organic conducting polymers such as polypyrole and polyaniline (PANI) to form humidity-sensitive composites [14–16]. Cellulose derivatives such as cellulose acetate butyrate (CAB) and Carboxymethyl Cellulose (CMC) were also studied [17,18]. CAB corresponds to cellulose

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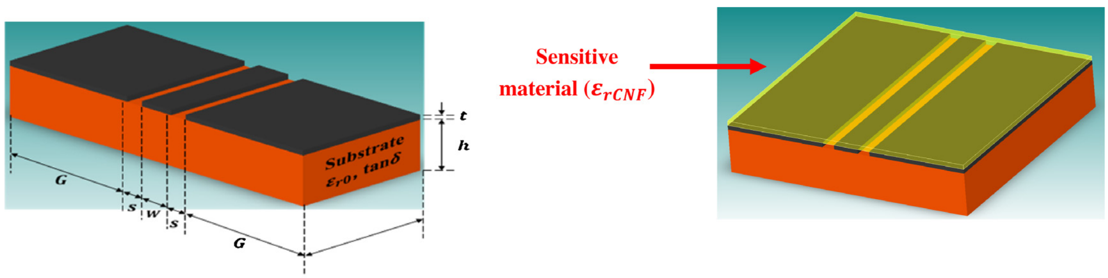

(a) (b)

Fig. 1. CPW structure (a) CPW line with substrate (b) CPW line covered by the sensitive material.

acetate derivative by grafting of the butyrate group, which makes this material non-biodegradable. For its part, CMC is a matrix that is usually prepared through the reaction of alkali cellulose with monochloroacetate or its sodium salt in and organic medium. CMC was used to improve the humidity response with PANI and Ag-nanocomposite [19,20]. The PEL Nano P60 paper was studied in printed electronics but required an inorganic coating to achieve absorption of solvents and dispersion agents in the conductive ink [21]. To the best knowledge of the authors, no investigation on pure cellulose and no investigation on fully biodegradable materials for humidity sensing in microwave frequencies has been reported.

This paper presents the cellulose nanofibers (CNF) as new and environmentally-friendly sensitive materials for humidity sensing in microwave frequencies. CNF are nanoscale cellulose with fiber diameter from 5 to 50 nm [22]. Here, CNF were processed through sonocatalysed 2,2,6,6-Tetramethylpiperidinyloxy (TEMPO) oxidation to increase the hydroxyl groups at their surface and improve their affinity to water [23]. No grafting is required. As a final product, CNF are available in the form of foldable sheets of approximately 50 μm thick, making them suitable materials for microwave sensing mechanisms through transmission line technologies, such as the coplanar waveguide (CPW).

This paper investigates the humidity sensing performances of CNF sheets through CPW transmission line technology, well-known for its sensitivity to the surrounding dielectrics. Additional advantages of the CPW structures include low losses compared to microstrip and relatively easy integration with other planar devices. The proposed sensor consists of a CNF sheet taped on the top of the CPW circuit to sense humidity variations in the surrounding environment. The microwave study includes sensitivity analysis through reflected signals with resonant frequency and transmitted signals with phase shift. Furthermore, this paper investigates the influence of the CNF weight by comparing the sensitivity and the dynamic range of sheets with different amounts of CNF.

2. Humidity sensing principle

The proposed microwave structure for humidity detection in this paper is a coplanar waveguide (CPW) circuit covered by a CNF sheet on the top as shown in Fig. 1.

The CPW consists of two ground planes surrounding the center signal strip to ensure microwave propagation. The center strip is separated from each ground plane by a narrow gap. The theory of CPW circuits using conformal mapping techniques is presented in [24]. The effective dielectric constant of the circuit presented in 1-b is given in (1). The terms q_0 and q_{CNF} are the partial filling factors of the substrate and the sensitive material respectively. They depend on the substrate thickness (t), the gap dimension (S) and the trace width (W). The terms ε_{r0} and ε_{rCNF} represent the dielectric properties of the substrate and the CNF sheet respectively.

$$\varepsilon_{eff} = 1 + q_0 (\varepsilon_{r0} - 1) + q_{CNF} (\varepsilon_{rCNF} - 1) \quad (1)$$

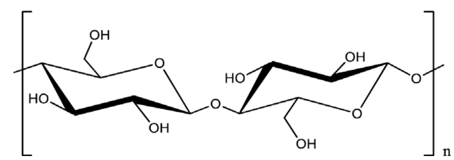

Fig. 2. Molecular structure of cellulose.

From (1), the humidity sensing capability can be integrated into the CPW circuit thanks to the hydrophilic or the hydrophobic character of the sensitive material. In this paper, the substrate of the CPW structure is insensitive to humidity so that the effective dielectric constant ε_{eff} closely depends on the dielectric properties of the CNF sheet (ε_{rCNF}) in case of humidity variation. The molecular structure of cellulose is shown in Fig. 2. The polymer backbone contains functional alcohol groups in carbon position 2, 3 and 6. The TEMPO reaction oxidizes the primary alcohol in the carboxyl group as shown in Fig. 3. The abundance of hydroxyl groups and oxygen atoms allows the cellulose to form an extensive network of intra and intermolecular hydrogen bonds, which confers its remarkable chemical stability. The polar groups exert high attraction on water molecules by establishing hydrogen bonds and confer to the cellulose its hydrophilic character.

The humidity absorption by the CNF sheet has two effects. The first effect is the change in its dielectric and conductive properties as a function of humidity. Indeed, humidity absorption modifies the internal electrical state of the CNF and this change is reported to the microwave substrate parameters [4]. The second effect of humidity absorption is the swelling of the CNF films. The presence of beta 1-4 linkage between glucose monomers makes the cellulose insoluble in water so that the sensitive material swells but does not dissolve after humidity absorption. From (1), the change in the dielectric properties and/or the dimensions of the CNF sheet generated by these two effects will be reported on the effective dielectric constant, leading to the change of the characteristic impedance of the CPW circuit as a function of humidity. As a result, reflected and transmitted electromagnetic waves traveling across the circuit will be modified as a function of humidity. Based on this principle, the CPW circuit can be used to monitor humidity variations in the surrounding environment with RF/microwave parameters.

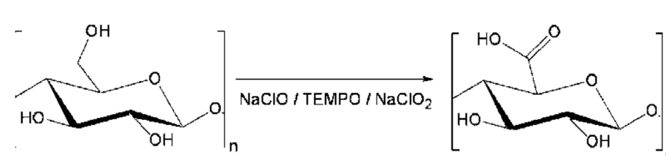

Fig. 3. TEMPO oxidation of cellulose.