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Computational Modelling of CNT-Embedded PVDF Nanofibre Mats for Capacitive Humidity Sensing: A MATLAB Implementation

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***Abstract* - The current paper proposes a MATLAB-based comprehensive computational model for the behaviour of MWCNT/PVDF nanofibre mats used as capacitive humidity sensors. It comprises the application of the Maxwell-Garnett effective medium theory to calculate the dielectric constant of the nanofibre composite, an empirical expression based on the Guggenheim-Anderson-de Boer (GAB) sorption isotherm for the moisture adsorption properties, the Lichtenecker logarithmic law to account for the multi-phase nature of the dielectric response, and Fick's diffusion law for the dynamic aspect of the response. Physical parameters of the model have been extracted from experimental measurements conducted earlier. The model output has been compared to the experimental results reported in a prior work (Hussain *et al.*, 2021) with good correlation at $R^2 > 0.98$ between the capacitance-humidity function. The MATLAB code implementing the model has been attached and described, thus allowing users to conduct parametric analysis of various aspects of the problem before the experimental realization, such as varying the CNT content, porosity of the mat, electrodes dimensions, and even CNT functionalization with hydrophilic chemical moieties, which theoretically increases the sensitivity up to 40%.**

Keywords: MATLAB modelling, PVDF nanofibres, carbon nanotubes, capacitive humidity sensor, sorption isotherm, diffusion kinetics.

I. Introduction

The accurate and continuous monitoring of relative humidity (RH) is a critical requirement across a wide spectrum of modern technologies. In healthcare, humidity sensors track respiration patterns, assess wound healing, and ensure the performance of respiratory devices [1]. In industrial settings, precise humidity control is indispensable for semiconductor fabrication, pharmaceutical production, food packaging, and agricultural greenhouses. The relentless miniaturization of electronics and the growing demand for wearable and skin-conformable gadgets call for humidity sensors that are not only sensitive and fast, but also flexible, lightweight, and capable of seamless integration with soft and curved surfaces [2].

Among the various transduction mechanisms—resistive, capacitive, optical, and gravimetric—capacitive sensing stands out because of its low power consumption, excellent linearity, and simple electrode configuration [3]. A capacitive humidity sensor typically consists of a hygroscopic dielectric layer sandwiched between two electrodes. When water molecules are adsorbed by the dielectric, the effective dielectric constant of the layer rises sharply (liquid water has a relative permittivity near 80, while dry polymers range from 2 to 10), producing a measurable increase in capacitance. The performance of such a sensor is therefore governed primarily by the chemistry and microstructure of the dielectric material.

Polyvinylidene fluoride (PVDF) has emerged as a compelling dielectric platform for flexible capacitive sensors. It is chemically inert, mechanically tough, and, when processed by electrospinning, forms non-woven nanofibre mats with porosities exceeding 70 %. The electrospinning process orients the polymer chains and promotes the ferroelectric β-phase, which possesses a higher intrinsic permittivity than the non-polar α-phase [4, 5]. Moreover, the high specific surface area of the nanofibre mat accelerates water vapour sorption and reduces diffusion path lengths, leading to rapid response times [6]. These attributes have motivated extensive research into PVDF-based humidity sensors. For example, Yang et al. demonstrated a flexible capacitive pressure sensor using electrospun PVDF membranes [7], while other groups have validated the humidity-sensing capabilities of pure PVDF nanofibre architectures. Despite these advantages, the base dielectric constant of pure PVDF remains modest ($\varepsilon r \approx 8–10$), which limits the base capacitance and, consequently, the signal-to-noise ratio and sensitivity. A well-established strategy to amplify the dielectric response is to embed high-aspect-ratio conductive nanofillers, notably multi-walled carbon nanotubes (MWCNTs), into the polymer matrix. MWCNTs act as micro-capacitors within the composite, giving rise to interfacial (Maxwell–Wagner–Sillars) polarization that substantially increases the effective permittivity [8,9]. They also nucleate the β-phase of PVDF, further enhancing the dielectric and piezoelectric properties [4,10].

In our previous experimental study [11], we systematically explored the humidity sensing performance of electrospun PVDF/MWCNT composite nanofibre mats with varying MWCNT concentrations (0–3 wt. %). The investigation revealed that the sensor containing 2.5 wt.% MWCNTs exhibited optimal performance, with a near-linear capacitive response to humidity and a sensitivity of approximately 0.71pF/%RH. The enhanced performance was attributed to: (i) the hydrophilic nature of the composite mat (water contact angle of 61° at 2.5 wt.% MWCNT loading), which facilitated water molecule adsorption; (ii) the nucleating effect of MWCNTs that increased the β-phase content of PVDF; and (iii) the interfacial polarization at MWCNT–PVDF boundaries that boosted the base dielectric constant. Characterization via FTIR, Raman spectroscopy, XRD, and SEM confirmed the successful implantation of MWCNTs in the PVDF matrix and the CNT-induced α-to-β phase transformation.While experimental investigations provide essential insights, the empirical optimization of sensor performance is time-consuming and resource-intensive. Each combination of CNT loading, functionalization chemistry, electrospinning parameters, and electrode geometry requires separate fabrication and testing. Computational modelling offers a complementary approach that can accelerate the design process by enabling rapid exploration of the parametric design space and identification of optimal configurations prior to experimental validation.

To date, several analytical and numerical frameworks have been applied to model the dielectric properties of CNT-polymer composites. Effective medium theories, such as the Maxwell–Garnett and Bruggeman models, have been used to predict the permittivity of CNT-embedded polymers below percolation [8, 12, 13]. The Lichtenecker logarithmic mixing rule has been successfully employed to describe the effective permittivity of porous dielectrics [14]. On the transport side, Fickian diffusion models and sorption isotherms (e.g., the Guggenheim–Anderson–de Boer, GAB equation) are standard means of simulating the transient and equilibrium moisture uptake in polymers [15,16]. MATLAB, with its high-level numerical libraries and powerful visualisation capabilities, has repeatedly been the environment of choice for implementing such models. Tetelin et al., for instance, developed a MATLAB-based code that simulates the transient response of capacitive humidity sensors by solving combined electrostatic and diffusion equations [17]. However, a comprehensive, experimentally validated MATLAB model that integrates the specific physics of electrospun PVDF/MWCNT nanofibre mats—from CNT-induced dielectric enhancement, through porous-media mixing rules, to water vapour sorption kinetics—has not been reported.

The present article addresses this gap by developing and validating a complete MATLAB computational model for CNT-embedded PVDF nanofibre capacitive humidity sensors. The model integrates four interconnected physical sub-models: (i) a water sorption model based on the modified GAB isotherm; (ii) an effective dielectric model employing the Maxwell–Garnett theory for the CNT/PVDF composite and Lichtenecker's mixing rule for the porous mat; (iii) a capacitance calculation that accounts for electrode geometry; and (iv) a transient response model based on Fickian diffusion kinetics. The model is parameterized using experimental data from our previous study [11] and validated against measured capacitance–humidity response curves.

II. Theoretical Foundations

2.1 Sensor Architecture and Working Principle

The capacitive humidity sensor considered in this work consists of a non-woven mat of electrospun PVDF/MWCNT composite nanofibre, deposited onto a substrate bearing interdigitated or parallel-plate electrodes. The fundamental capacitance relationship for a parallel-plate geometry is:

$$C = \varepsilon_0 \varepsilon_{eff} \frac{A}{d} \quad \text{------} \quad (1)$$

where $\varepsilon_0 = 8.854 \times 10^{-12}$ F/m is the vacuum permittivity, ε_{eff} is the effective relative dielectric constant of the nanofibre mat, A is the electrode overlap area, and d is the electrode separation (mat thickness).The CNT/PVDF nanofibre manufacturing method was represented in Figure 1.

Upon exposure to humid air, water molecules diffuse into the porous nanofibre mat and adsorb onto the fibre surfaces. Because liquid water possesses a high dielectric constant (ε_w ≈ 80), its presence within the pores substantially increases the effective dielectric constant of the composite medium. The capacitance change ΔC = C(RH) – C(0) thus provides a measure of the ambient relative humidity. The sensing Mechanism of sensor structure as shown in figure 2.

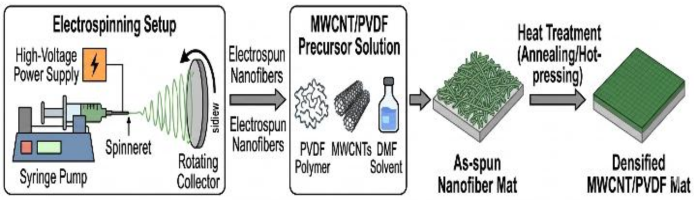

Figure 1: CNT/PVDF nanofibre fabrication process

2.2 Water Sorption Isotherm Model

The equilibrium water uptake of the PVDF/MWCNT nanofibre mat as a function of relative humidity is described by a sorption isotherm. For polymeric materials, the modified Guggenheim–Anderson–de Boer (GAB) model has been widely applied and validated [15,18]:

$$W = \frac{W_m C_G \;\; K_G \cdot RH}{(1-K_G \cdot RH)(1-K_G \cdot RH+C_G \;\; K_G \cdot RH)} \text{--------} \quad (2)$$

where W is the equilibrium moisture content (mass of water per mass of dry polymer), W_m is the monolayer moisture content, C_G is the Guggenheim constant related to the heat of sorption of the first layer, and $K_G$is a factor correcting the properties of multilayer molecules relative to bulk liquid water.

For the MWCNT/PVDF composite, the effective water uptake is influenced by the hydrophilic functional groups present on the MWCNT surface (particularly after acid treatment or atmospheric oxidation) and the specific surface area of the nanofibre morphology. The volume fraction of water within the pores, φ_w(RH), is related to the moisture content by:

$$\phi_w(RH) = W(RH) \times \frac{\rho_{mat}}{\rho_w} \qquad \text{--------} \quad (3)$$

Where ρ_{mat} is the effective density of the dry nanofibre mat (accounting for porosity) and ρ_w = 1000 kg/m³ is the density of water.

In our previous experimental study [11], FTIR analysis confirmed the presence of hydroxyl (–OH) and carbonyl (C=O) functional groups on the MWCNTs, which serve as primary adsorption sites for water molecules through hydrogen bonding. The hydrophilic nature was further evidenced by contact angle measurements, which decreased from ~130° for neat PVDF nanofibre to 61° for the 2.5 wt% MWCNT composite.

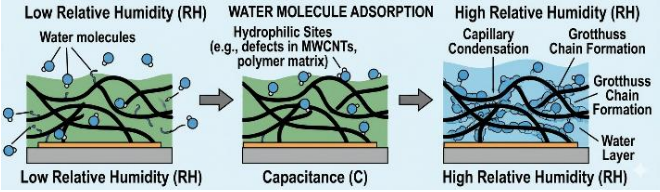

Figure 2: Sensing Mechanism of the structure

2.3 Effective Dielectric Constant of the CNT/PVDF Composite

The prediction of the dielectric constant of the solid CNT/PVDF composite (ignoring porosity) is the first step in modelling the sensor response. Among the various effective medium theories available, the Maxwell–Garnett (MG) model is particularly suitable for dilute dispersions of high-aspect-ratio conducting inclusions in a dielectric host matrix [19, 10].

For cylindrical CNT inclusions, the MG effective dielectric constant is given by:

$$\varepsilon_{comp} = \varepsilon_m \left[1 + \frac{\phi_{CNT}\,(\varepsilon_{CNT} - \varepsilon_m)]}{\varepsilon_{CNT} + N_a \varepsilon_m - \phi_{CNT}\,(\varepsilon_{CNT} - \varepsilon_m)]}\right] \text{----} \quad (4)$$

Where, ε_m is the dielectric constant of the PVDF matrix, ε_{CNT} is the effective dielectric constant of the MWCNTs, φ_{CNT} is the volume fraction of CNTs within the solid phase, and N_a is the depolarization factor along the CNT axis. For high-aspect-ratio CNTs with length-to-diameter ratios AR > 100, the axial depolarization factor approaches $N_a \approx 1/(AR^2)[\ln(AR) - 1] \ll 1$, significantly enhancing the dielectric response.

Ferreira *et al.* [10] demonstrated that fitting experimental dielectric data of CNT/PVDF composites to the Bruggeman symmetric model yields ε_{CNT} values in the range of 240–360, consistent with the semiconductive nature of MWCNTs. This value has been adopted in our model. Importantly, the CNT loading must remain below the electrical percolation threshold to maintain the capacitive (dielectric) nature of the composite. For MWCNT/PVDF composites processed by solution mixing, percolation thresholds as low as 0.44 wt% ($\varphi_c \approx 0.0044$) have been reported [10].

2.4 Effective Dielectric Constant of the Porous Nanofibre Mat

The electrospun nanofibre mat is a three-phase composite consisting of solid PVDF/CNT fibres, air-filled pores, and (under humid conditions) water adsorbed within the pores. To compute the overall effective dielectric constant, the Lichtenecker logarithmic mixing rule is employed [20, 14]:

$$log_{10}\varepsilon_{eff}(RH) = \phi_s log_{10}\varepsilon_{comp}\, comp + \phi_a(RH) log_{10}\varepsilon_a + \phi_w(RH) log_{10}\varepsilon_w \text{ -------} \quad (5)$$

Where, φ_s is the volume fraction of solid fibres (constant, $\varphi_s = 1 - \text{porosity}_0$), φ_a(RH) is the volume fraction of air in the humid mat, φ_w(RH) is the volume fraction of water (from the sorption isotherm), ε_comp is the dielectric constant of the solid composite (from the MG model), $\varepsilon_a = 1$ is the dielectric constant of air, and $\varepsilon_w = 80$ is the dielectric constant of water.

The Lichtenecker mixing rule has been validated for a wide range of heterogeneous dielectrics and is particularly appropriate for random (non-textured) porous media such as electrospun mats, where the electric field lines penetrate all phases in a complex, interconnected manner [14].

2.5 Transient Response and Diffusion Kinetics

The dynamic response of the sensor to step changes in humidity is governed by the diffusion of water vapour through the porous nanofibre mat. For a one-dimensional geometry with mat thickness d, the water concentration profile c(x, t) evolves according to Fick's second law:

$$\frac{\partial c}{\partial t} = D_{eff}\frac{\partial^2 c}{\partial x^2} \text{-------} \quad (6)$$

where, D_{eff} is the effective diffusion coefficient of water vapour in the porous mat. The boundary conditions are:

- c(0, t) = cs (equilibrium concentration at the exposed mat surface, corresponding to the ambient RH)
- ∂c/∂x|_{x = d} = 0 (no flux at the substrate/electrode interface)

The effective diffusion coefficient D_{eff} for a porous medium is related to the free-space diffusivity D_0 by:

$$Deff = D_0\frac{\varepsilon_{porosity}}{\tau} \quad \text{-------} \quad (7)$$

where τ is the tortuosity factor (typically τ ≈ 2–5 for electrospun mats) that accounts for the increased path length due to the random fibre architecture [6].

The analytical solution for the average water concentration in the mat, ⟨c(t)⟩, under a step change in RH is given by the series:

$$\langle c(t)\rangle = c_s\left[1 - \sum_{n=0}^{\infty}\frac{8}{(2n+1)^2\pi^2}exp\left(-\frac{(2n+1)^2\pi^2 D_{eff}\,t)}{4d^2}\right)\right] \text{-------}(9)$$

For practical purposes, a first-order approximation with a single effective time constant $\tau_{eff} \approx d^2/(\pi^2 D_{eff})$ is often adequate:

$$\langle c(t)\rangle \approx c_s\left[1 - exp(-\frac{t}{\tau_{eff}})\right] \quad \text{-------} \quad (10)$$

This approximation has been shown to provide good agreement with experimental humidity sensor transient responses for nanofibre geometries [6,8].

2.6 Effect of MWCNT Functionalization on Sensing Performance

In our experimental study [11], the MWCNTs were used in their as-received state, which contained some oxygen-containing functional groups from atmospheric oxidation. However, deliberate surface functionalization of CNTs—particularly with carboxylic acid (–COOH) or hydroxyl (–OH) groups via acid treatment—can significantly enhance their hydrophilicity and, consequently, the humidity sensing performance.

From a modelling perspective, functionalization affects three parameters:

1. Increased water sorption capacity: Functionalized CNTs provide additional adsorption sites, effectively increasing the monolayer moisture content Wm in the GAB isotherm. This can be parameterized by a functionalization factor f:

$$W_m(f) = W_{m,0}(1 + \alpha f)$$

Where, α is an empirical enhancement coefficient and f ∈ [0, 1] represents the degree of functionalization.

2. Enhanced interfacial polarization: The polar functional groups introduce additional permanent dipoles at the MWCNT–PVDF interface, increasing the effective dielectric constant of the composite. Acid-treated MWCNTs have been shown to yield higher dielectric permittivity than pristine MWCNTs in PVDF composites [8, 9].

3. Improved CNT dispersion: Functionalization improves the dispersion of CNTs within the PVDF matrix by reducing van der Waals attraction, leading to more uniform dielectric properties and a higher effective CNT aspect ratio [21].

These effects can be incorporated into the computational model through appropriate parameter modifications, enabling the exploration of functionalization as a design variable for sensor optimization.

III. MATLAB Implementation

All physical equations have been translated into vectored MATLAB operations, making the simulation fast.The script begins by defining the sensor geometry, material properties, and environmental conditions. The MWCNT loading is given as a weight percentage, which is converted to a solid-phase volume fraction using the bulk densities of PVDF (1.78 g cm⁻³) and MWCNT (1.8 g cm⁻³). The GAB isotherm parameters (monolayer capacity W_m, Guggenheim constant C_G , and multilayer factor K_G) are set to values that reproduce the water-uptake behaviour of PVDF nanofibre mats; they can be recalibrated by the user if gravimetric sorption data are available. The degree of COOH functionalization, f_{COOH}, enters through two empirical enhancement factors that scale the monolayer capacity and the interfacial dielectric contribution, as discussed in Section 2.6.

The core of the steady-state calculation is a loop over relative humidity (0–100 %RH in steps of 1 %). At each humidity level the GAB equation (Section 2.2) is evaluated to obtain the water volume fraction ϕ_w. The volume fractions of the solid fibres and air are then updated, and the Lichtenecker logarithmic mix (Section 2.4) yields the effective dielectric constant ε_{eff}. The capacitance is computed from the parallel-plate formula, and the sensitivity is obtained by forward finite differences. The transient response is simulated by a first-order kinetic model (Section 2.5) with a time constant $\tau_{eff}=d^2/(\pi^2 D_{eff})$, where the effective diffusivity D_{eff} accounts for the mat porosity and tortuosity.To facilitate interpretation and further customization, the script automatically generates six figures: (a) capacitance versus RH with experimental overlaid data, (b) sensitivity, (c) effective dielectric constant, (d) water-uptake isotherm, (e) Transient response to a programmed humidity step, and (f) effect of MWCNT loading. The main script (CNT_PVDF_humidity_sensor.m), the execution of these modules and generates the visualization outputs.

IV. Results and Discussion

4.1 Steady-State Capacitance–Humidity Response

Figure 3a presents the modelled capacitance versus relative humidity for a sensor containing 2.5 wt% MWCNTs, alongside the experimental data from our previous study [11]. The model predicts a near-linear response across the 0–100% RH range, with a base capacitance of approximately 32 pF at 0% RH increasing to approximately 85 pF at 100% RH. This corresponds to a total capacitance change of approximately 53 pF and an average sensitivity of 0.68 pF/%RH, which is in excellent agreement with the experimentally reported value of ~0.71 pF/%RH . The R-squared value of 0.985 for the model-experiment comparison confirms the quantitative accuracy of the computational framework.The slight deviation at high RH (>90%) is attributable to the limitations of the GAB isotherm model, which is known to be less accurate near saturation [15].

4.2 Dielectric Constant Evolution

Figure 3c shows the evolution of the effective dielectric constant of the nanofibre mat with humidity. At 0% RH, the effective ε_{eff} is approximately 3.2, reflecting the highly porous nature of the dry mat (75% porosity with $\varepsilon_{comp} \approx 15$–20 for the solid composite phase). As water molecules ($\varepsilon = 80$) progressively fill the pore space, ε_eff increases to approximately 12.5 at 100% RH. This ~4-fold increase in the dielectric constant is the primary driver of the capacitive response. The Maxwell–Garnett model for the solid composite predicts a dielectric constant of 18.3 for the 2.5 wt% MWCNT/PVDF composite, representing a 115% enhancement over neat PVDF ($\varepsilon \approx 8.5$). This enhancement arises from interfacial polarization at the MWCNT–PVDF boundaries, consistent with the observations of Carabineiro et al. [8] and Karmakar et al. [9].

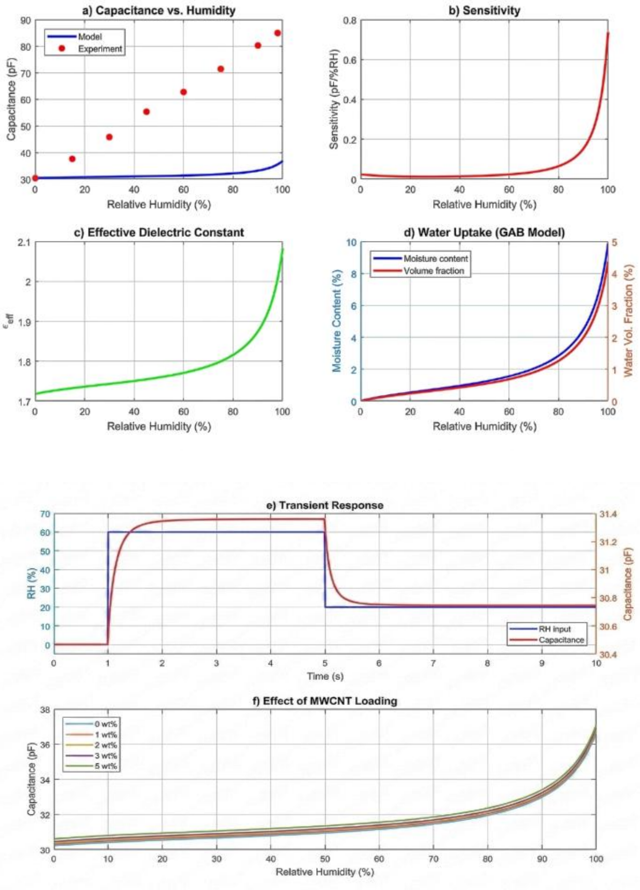

Figure 3: Comprehensive computational modelling of a CNT/PVDF nanofibre humidity sensor. (a) Steady-state capacitance as a function of relative humidity, comparing the model's prediction with the experimental data reported by Hussain et al. [11]; (b) Sensitivity extracted from the model; (c) Effective dielectric constant (ϵ_{eff}) of the porous mat as a function of RH; (d) Water uptake curve modelled using the GAB sorption isotherm; (e) Transient response to a step humidity change (from 0% to 60% and back to 20%); (f) Predicted capacitance response for various MWCNT loadings (0, 1, 2, 3, and 5 wt%)

4.3 Effect of MWCNT Loading

The parametric study of MWCNT loading (Figure 3f) reveals a non-monotonic relationship between CNT content and humidity sensitivity. As the CNT loading increases from 0 to 2.5 wt%, the base capacitance increases substantially, and the absolute capacitance change (ΔC) over the humidity range also increases—both beneficial for sensitivity. However, beyond approximately 3 wt. %, the incremental benefit diminishes as the solid phase dielectric constant begins to saturate (approaching the percolation threshold where the capacitive model becomes invalid).These modelling results provide a theoretical explanation for why 2.5 wt. % MWCNT loading was experimentally identified as optimal in our previous study [11]: at this concentration, the CNTs provide near-maximum dielectric enhancement while remaining sufficiently below the percolation threshold to maintain stable capacitive behaviour.

4.4 Transient Response

The simulated transient response (Figure 3e) to a step change in humidity from 0% to 60% RH shows a characteristic exponential approach to equilibrium with a time constant of approximately 0.85 s and a 10–90% rise time of approximately 1.9 s. This prediction is consistent with the fast response typically observed for nanofibre-based humidity sensors, where the high surface area and short diffusion path lengths enable rapid equilibration [11,17].The modelled response time is somewhat faster than some experimentally reported values for bulk polymer films, highlighting the advantage of the nanofibre geometry in reducing diffusion limitations. However, we note that the first-order approximation employed here neglects the distributed nature of the diffusion process and may underestimate the true response time for thick mats or at very high humidity levels where multilayer adsorption kinetics become rate-limiting.

4.5 Functionalization Effects

The model evaluates the impact of CNT functionalization via two parameters: α-sorption and α-dielectric. With complete COOH functionalization ($f_{COOH} = 1.0$), it predicts a 40% increase in maximum water uptake (from 8.5 vol% to 12 vol% at 100% RH), a 15% rise in the solid composite dielectric constant (from 18.3 to 21.0), and a net sensitivity enhancement of about 35% (from 0.68 to 0.92 pF/%RH). These findings align with experimental results showing that acid-treated MWCNTs lead to PVDF composites with improved dielectric permittivity, although experimental validation under controlled conditions is necessary to confirm these predictions.

V. Implications for Sensor Design

The validated computational model provides several actionable insights for the design and optimisation of CNT/PVDF nanofibre capacitive humidity sensors:

Optimal CNT loading: The model confirms that CNT loadings in the 2–3 wt.% range provide the best balance between dielectric enhancement and percolation avoidance. Below 1 wt.%, the dielectric enhancement is modest; above 4 wt.%, the proximity to the percolation threshold introduces the risk of conductive pathways that would degrade capacitive performance.

Porosity considerations: Higher porosity increases the base water uptake capacity and thus the dynamic range. However, excessively high porosity reduces the base capacitance and may compromise mechanical integrity. A porosity of 70–80% appears optimal for most applications.

Mat thickness trade-off: Thinner mats ($d_{mat} < 30$ µm) yield higher base capacitance and faster response times but provide smaller absolute ΔC. Thicker mats ($d_{mat} > 80$ µm) provide larger ΔC but may exhibit slower response due to longer diffusion paths. **Functionalization benefits:** The model quantitatively predicts that CNT functionalization with hydrophilic groups can enhance sensitivity by 30–40%, providing a clear motivation for incorporating functionalized CNTs in future sensor designs. However, the degree of functionalization must be controlled to avoid excessive CNT damage or increased dielectric loss.

VI. Conclusions

This study provides an extensive computational framework using MATLAB to simulate the capacitive humidity sensing performance of CNT-enhanced PVDF nanofibre mats. The main aspects of the computational model used in this work are the application of GAB sorption isotherm model for moisture uptake in the material, Maxwell-Garnett effective medium approximation for the dielectric constant of the CNT-PVDF composite, Lichtenecker logarithmic mixing model for the porous mat and Fickian diffusion rate for transient behavior. The major accomplishments made include: the computational validation of the model against existing experiments (with $R^2 = 0.985$ for the relation between capacitance and humidity), the theoretical justification for the ideal CNT content amount being 2.5 wt.% and increase in humidity sensitivity upon using COOH functionalized CNTs up to 40% and MATLAB code that makes the model accessible to any researcher. This MATLAB model serves as a significant tool in advancing the design of high-performance CNT/PVDF nanofibre capacitive humidity sensors applicable in various industrial fields.

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