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Advanced Morphological and Material Engineering for High-Performance Interfacial Iontronic Pressure Sensors

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High-performance flexible pressure sensors are crucial for applications such as wearable electronics, interactive systems, and healthcare technologies. Among these, iontronic pressure sensors have garnered particular attention due to their superior sensitivity, enabled by the giant capacitance variation of the electric double layer (EDL) at the ionic-electronic interface under deformation. Key advancements, such as incorporating microstructures into ionic layers and employing diverse materials, have significantly improved sensor properties like sensitivity, accuracy, stability, and response time. This review highlights advancements in flexible EDL pressure sensors, focusing on structural designs and material engineering. These strategies are tailored to optimize key metrics such as sensitivity, detection limit, linearity, stability, response speed, hysteresis, transparency, wearability, selectivity, and multifunctionality. Key fabrication techniques, including micropatterning and externally assisted methods, are reviewed, along with strategies for sensor comparison and guidelines for selecting appropriate sensors. Emerging applications in healthcare, environmental and aerodynamic sensing, human–machine interaction, robotics, and machine learning-assisted intelligent sensing are explored. Finally, this review discusses the challenges and future directions for advancing EDL-based pressure sensors.

1. Introduction

The rapid advancement of flexible electronics and wearable technologies has raised a significant demand for high-performance flexible pressure and tactile sensors. These sensors, converting physical or chemical stimuli into electrical signals, are indispensable components in applications such as wearable and implantable medical devices,[1] human–machine interfaces,[2] and artificial intelligence systems.[3] Based on the mechanical-to-electrical signal conversion mechanisms, flexible pressure sensors can be broadly classified into resistive,[4] capacitive,[5] piezoelectric,[6] and triboelectric types.[7] Among these, capacitive sensors are particularly attractive due to their simple structure, energy efficiency, fast response, and stability.[8] However, conventional capacitive sensors face inherent limitations such as low sensitivity and narrow pressure-response range.[9] To address these challenges, the architectural engineering of interfacial structures within dielectric and composite elastomers has been explored.[10] Introducing microstructures into dielectric elastomers can enhance the sensors' sensitivity by increasing mechanical compliance and the effective dielectric constant. Additionally, creating voids within the material facilitates elastic deformation, thereby improving response speed.[10,11] Nevertheless, dielectric elastomers are largely incompressible and are typically limited to a thickness of ≈10 µm. As a result, the capacitance change is generally small (<100 pF),[11b] thus constraining sensitivity to approximately 1 kPa^{-1}.

To overcome the limitations of conventional capacitive sensors, interfacial iontronic sensing, a novel mechanism, was introduced by Pan's group in 2011.[12] In contrast to conventional capacitive sensors, interfacial iontronic sensors utilize an ionic film to replace the traditional dielectric layer. When this ionic film interfaces with a conductive electrode, positive and negative charges accumulate at the electrode/ionic film interface, forming an electric double layer (EDL) with a thickness of ≈ 1 nm. This configuration leads to extraordinarily high capacitance due to the tiny charge separation distance at the interface, providing two key advantages. First, the significantly shorter charge separation distance in EDL sensors, compared to the conventional dielectric thickness of 10–100 µm, enhances unit area capacitance (UAC) by several orders of magnitude under applied pressures, resulting in substantially improved sensitivity.[13] Second, the fixed charge separation within the EDL ensures that capacitance

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depends primarily on the contact area between the dielectric and the electrode, rather than the separation distance. This unique characteristic enables substantial sensitivity improvements through microstructural modifications.[14] Moreover, this configuration minimizes parasitic influences along the transmission line and suppresses electromagnetic noises from the ambient environment, greatly enhancing the signal-to-noise ratio and overall sensor performance.[12,15] Moreover, EDL sensors exhibit high spatial resolution and effectively respond to both static and dynamic stimuli.[13a]

Recent research has focused on advancing EDL flexible pressure sensors by improving critical performance metrics, such as sensitivity, detection limit, linear range, and stability, to meet the demands of practical applications.[13a] Various sensing materials with tailored microstructures have been developed to improve contact efficiency and signal conduction within the sensing layer and electrode. These structural features are categorized into nano- and microscale designs based on their dimensional properties. At the nano-scale, functional mechanisms such as the ion pump effect[16] and pseudocapacitive materials[17] optimize material properties, while structural designs like graded stiffness[18] and nanorod incorporation[19] improve mechanical adaptability. At the micro-scale, microroughness,[20] internal porous structures,[21] and multiscale hierarchical architectures[22] improve stress management and boost sensor functionality. These microstructures are fabricated through patterned techniques (e.g., lithography and printing) and assisted methods (e.g., mechanical force-assisted, heat-induced, electric field-assisted, gas bubble-assisted, and template-assisted).

Several review papers have summarized advancements in iontronic flexible sensors,[13a,23] covering diverse mechanisms such as interfacial capacitive, piezoresistive, piezoelectric, triboelectric, mechano-electroluminescent/electrochromic sensing, as well as emerging materials, structural designs, and applications. Among these, interfacial iontronic pressure sensors (EDL pressure sensors), which leverage the EDL effect, rely on mechanical deformation to modulate the interfacial contact area, enabling highly sensitive pressure detection. While some existing reviews have discussed structural designs for EDL pressure sensors, they often lack systematic frameworks for analyzing morphological engineering strategies. Key aspects, including the influence of multiscale structural designs (e.g., nanoscale architectures, microstructural configurations, or hierarchical morphologies) on critical performance metrics like sensitivity, detection limits, stability, and dynamic range, remain insufficiently explored. To address these gaps, this review systematically examines the role of morphological and material engineering in optimizing EDL-based pressure sensors, providing a detailed framework that connects structural design with performance trade-offs and multifunctionality. This review is structured as follows. Section 2 introduces the transduction mechanisms and material selection for EDL pressure sensors. Structural engineering of sensing layers is discussed in Section 3, followed by performance enhancements in Section 4. Section 5 reviews fabrication strategies for microstructures, while Section 6 provides guidelines for selecting appropriate sensors. Applications in wearable healthcare, environmental sensing, human–machine interaction, robotics, and ML-enabled intelligent sensing platforms are presented in Section 7. Finally, conclusions and future prospects are outlined in Section 8. **Figure 1** illustrates the overall structure and relationships between the sections.

2. Mechanisms and Materials for Iontronic Pressure Sensing

2.1. Electrical Double Layer Dynamics

Interfacial iontronic pressure sensing relies on the formation of an EDL at the electrolyte-electrode interface.[12] When pressure is applied, ions from the electrolyte redistribute near the electrode surface, forming a highly responsive capacitive layer that rapidly adjusts to external stimuli. The EDL consists of two distinct regions: the Helmholtz layer, where solvent molecules and counterions are tightly bound to the electrode surface, and the diffuse layer, where ions are more loosely distributed to balance the electrode's surface charge (**Figure 2a**).[32] The total capacitance (C_{EDL}) is represented by two interfacial capacitors in series:

$$C_{\mathrm{EDL}} = \left(\frac{1}{C_{\mathrm{H}}} + \frac{1}{C_{\mathrm{D}}} \right)^{-1} \tag{1}$$

here, C_{H} represents the Helmholtz layer capacitance and C_{D} denotes the diffuse layer capacitance. In the series configuration, the total capacitance (C_{EDL}) is lower than either individual capacitance, primarily determined by the smaller of the two. C_{H} and C_{D} are influenced by several physical parameters, including the dielectric constant (ϵ), Helmholtz layer thickness (d), ionic species and concentrations (C), surface potential (φ), and temperature (T). The overall capacitance (C_{EDL}) can also be expressed as:

$$C_{\mathrm{EDL}} = \eta_A \cdot \phi\,(d, \epsilon, C, \phi, T) \cdot A = UAC \cdot A \tag{2}$$

here, η_{A} denotes the roughness factor, $\phi(d{,}\epsilon{,}C{,}\phi{,}T)$ represents a complex function of the listed parameters, and A is the electrode-electrolyte contact area. Notably, the product of η_{A} and $\phi(d{,}\epsilon{,}C{,}\phi{,}T)$ defines the EDL capacitance per unit area, termed UAC. In 2011, Pan et al.[12] first applied this effect in interfacial capacitive sensing by demonstrating a droplet-based pressure sensor with EDL's supercapacitive properties. This sensor, based on an ionic liquid droplet on a modified electrode, achieved ultrahigh capacitance in the range of several μF cm^{-2}, far exceeding conventional capacitive sensors with capacitances in the tens to hundreds of pF cm^{-2}.[11a,33]

Equation (2) shows that the EDL capacitance strongly depends on the electrode-ion layer contact area, with the UAC governed by material and microstructure properties. Thus, micro-structured sensing layers are crucial for enhancing the performance of EDL pressure sensors. When pressure is applied, these microstructures deform, which increases the contact area between the electrode and electrolyte and causes significant capacitance changes. For instance, in hemispherical microstructures (Figure 2b),[34] Hertzian contact mechanics describe the relationship between contact area (A) and applied pressure (P):[35]

$$A \propto P^{2/3} \tag{3}$$

This relationship demonstrates that the contact area increases nonlinearly with applied pressure, amplifying capacitance