A MICRODROPLET-BASED CAPACITIVE SENSING MATRIX FOR TACTILE APPLICATIONS

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ABSTRACT

In this paper, we present a highly sensitive microdroplet-based interfacial capacitive sensor matrix device, achieving ultrahigh sensitivity of 0.4nF/kPa and minimal detectable pressure of 33Pa at a 3×3×0.2mm³ packaging. The microdroplet-based sensor is comprised of an array of nanoliter ionic droplets sandwiched between two flexible substrates with patterned transparent electrodes. The sensing principle primarily relies on high elasticity of the sensing droplet and large capacitance presented at the electrode-electrolyte interface. Theoretical analyses and experimental investigations on several design parameters are thoroughly conducted to characterize. Finally, the microdroplet-based pressure sensor is successfully utilized for tactile sensing applications, such as Braille characters scanning and non-invasive cardiovascular pressure recording.

INTRODUCTION

Artificial tactile sensing devices have been extensively researched for their potential utilities in medicines and robotics, in which a wide spectrum of sensing mechanisms have been introduced and investigated, including resistive, piezoelectric and capacitive sensing [1-4]. Among those, elastomeric resistive materials have been frequently employed in building such arrays for their low cost and easy manufacturability [1]. Alternatively, piezoelectric nanomaterial has been incorporated into a pressure sensitive matrix with a sub-millimeter resolution for tactile imaging [2]. Capacitive sensing leads another trend in the development of artificial skins. Conventional capacitive tactile array, using a parallel plate configuration, has been devised for both pressure and force mapping [3]. In more recent effort, a flexible capacitive pressure sensor has been implemented with a microstructured elastomer layer as the sensing element for rapid mechanical responses (in the range of milliseconds) [4].

Latest development on iontronic materials (e.g., ionic liquids) renews long interest in utilizing ionic solutions to regulate the electronic transport at the electrolyte-electrode interface. Upon the solid-liquid contact, an electric double layer (EDL) immediately establishes with an ultrahigh unit-area capacitance on the order of 10μF/cm² at the atomic interface, which is more than thousand times greater than that of the solid-state counterpart (up to 2nF/cm²) [5,6]. It has been shown that the iontronic device can be utilized in transistor or sensors, where the EDL exhibits ultrahigh-density charge accumulation while avoiding reactions by its electrochemical inertness within the potential windows [7-9].

This report present a novel microdroplet-based iontronic sensor array (MISA), utilizing the ultrahigh EDL capacitance at the solid-liquid interface, for tactile sensing applications. Figure 1 illustrates the transparent MISA device of 12×12 elements with the spatial resolution of 1mm. Benefiting from the ultrahigh unit-area capacitance of the EDL, we can achieve a device sensitivity of 0.4nF/kPa and minimal detectable pressure of 33Pa at a 3×3×0.2mm³ packaging, which to our best knowledge is the most sensitive capacitive pressure sensor at its dimension. As the demonstrations of utilities of the iontronic devices, the MISA has successfully resolved the surface topology and detected the dynamic wrist pulses throughout cardiovascular cycles.

Fig. 1: Photography of the sensor array of 12×12 elements with the spatial resolution of 1mm.

THEORY

The MISA device consists of two flexible films with micropatterned transparent electrodes and one spacing layer hosting an array of nanoliter ionic droplets. Figure 2 exhibits the interfacial capacitive sensing principle. The direct ionic droplet-electrode contact immediately establishes the EDL, which possesses a remarkable interfacial capacitance (on the order of 10μF/cm²). Under external loads, the flexible surfaces experience mechanical deformation, resulting in circumferential expansion of the droplet-electrode contact. The corresponding capacitive change over the increased contact area can be detected electronically. Device sensitivity can be modeled both mechanically and electrically. The measured EDL capacitance can be directly related to the area of the droplet-electrode contact, as the invariant unit-area capacitance can be experimentally determined. On the other hand, the mechanical deformation of the membrane can be well defined in the classic mechanic theory [10]. It is worth noting that the interfacial capacitive sensing principle offers an ultrahigh capacitive sensitivity, which is more than thousand times greater than that of the solid-state counterpart, contributed mainly from the nanoscopic charge separation in EDL. The relationship between the measurable capacitive change (ΔC) and the contact pressure applied (ΔP) can be derived from the capacitive sensing principle:

$$\Delta C \approx C_0 \times \left[\frac{\Delta P}{\alpha \times H \times T^3 / L^4} + \left(\frac{\Delta P}{\alpha \times H \times T^3 / L^4} \right)^2 \right]$$

where C_0 and H indicate the initial capacitance and height of the sensing cell, respectively, and the constant $\alpha = 5 \times E \times (1 - \nu^2)^{-1}$ represents the mechanical deformation of the sensing membrane, influenced by the Young's modulus E and Poisson ratio ν . As can be seen, the relationship between the capacitive change (ΔC) and the contact pressure (ΔP) is highly dependent on the spatial resolution L and the membrane thickness T . In addition, the gravitational effect has been neglected in our consideration, as the microdroplet dimensions are considerably less than that of the capillary length (of ~1.8mm).

Fig. 2 The interfacial capacitive sensing principle. The EDL capacitance is established at the ionic droplet-electrode contact (top). The contact area expands circumferentially, which is caused by the membrane deformation under external loads and results in an increase in the EDL capacitance (bottom).

METHODS

Device fabrication

The fabrication process starts with micropatterning of conductive ITO electrodes on the flexible PET films using standard photolithography followed by wet etching. In the subsequent step, dry-film photoresist is laminated and photopatterned into a micropillar array on the electrode surface, serving as the supporting structure. To accurately position the microdroplets, surface wettability patterning is applied. A hydrophobic oligomer layer of PDMS is transfer-printed onto both electrode surfaces. Subsequently, using a microfluidic impact printing technique, nanoliter droplets of ionic liquid will be sequentially deposited into a grid matched to the hydrophilic spots formed by the wettability patterning. Prior to the final assembly, two electrode films are aligned face-to-face with the conductive patterns positioned orthogonally to each other, forming a grid of capacitances at the crossover points where the ionic droplet array sits in. The top and bottom layers are then bonded together through the plasma activated hydroxyl groups of the PDMS oligomer layers.

Electrical and mechanical characterization

Experimental investigations of the device sensitivity have been conducted on individual sensing units of the iontronic microdroplet array. The measurement stage is comprised of a force gauge with 1 mN resolution (DFS, Chatillon) amounted on a computer-controlled step motor (VT-80, Plmicos) with a spatial resolution of 400nm, from which mechanical loads and displacements can be controlled and monitored simultaneously. The corresponding capacitive changes are directly recorded through an LCR meter (4284A, Agilent). In the characterization of the responsive time, an electromagnetically driven pin actuator (Panasonic KX-PI150), powered by a pulsed voltage signal, has been used to apply the periodic contact pressure to the sensor surface. The output signals are measured by a custom circuitry.

RESULTS

Sensing Fluid

As a vital part of the interfacial sensing device, physical properties of the sensing droplet have to satisfy following criteria: high ionic concentration (ensuring high electrical conductance and interfacial capacitance), polarized molecular structure (reversible elasticity on the hydrophobic surfaces) and low fluidic viscosity

(allowing rapid mechanical response). Three types of ionic fluids have been considered, such as aqueous electrolytes (e.g., NaCl electrolyte solution), organic solvent solutions (e.g. KClO₄/PEO), and ionic liquids, which are commonly investigated in electrochemical processes [11]. Aqueous and solvent-based electrolytes are typically highly evaporative under room conditions and it becomes extremely challenging to maintain the constant electrical performance as both the volume and the physical properties change over time. As an emerging alternative, ionic liquids, consisting of an organic anion or cation, exhibit high electrical conductivity, low volatility, and tunable viscosity [12]. In the report, IL of 1-ethyl-3-methylimidazolium tricyanomethanide has been selected as the sensing fluid for the MISA.

Mechanical-to-capacitive sensitivity

As aforementioned, the overall mechanical-to-electrical sensitivity ($\Delta C/\Delta P$) can be determined by the geometrical confinements (the spatial resolution L and height H of each sensing unit), and the material properties of the membrane, e.g. the thickness T , Young's modulus E and Poisson ratio ν . Among those, the spatial resolution of the sensing unit play the most important role, followed by the thickness of the sensing membrane. Experimental investigations have been conducted to verify the above theoretical predications.

Fig. 3 Influences of spatial resolution **a** and membrane thickness **b** on the device sensitivity.

Figure 3 shows the experimental measurements on the device sensitivity with various geometrical designs of spatial resolutions and membrane thicknesses, in which the measurement results (dots) are plotted against the theoretical predications from Equation 1 (curves). As shown in Fig. 3a, the sensitivity exhibits a strong dependence (minor 4th power) on the spatial resolution. By varying the spatial resolution from 1mm to 3mm with a constant membrane thickness of 75μm, the sensitivity can be improved from 3.9pF/kPa to 433.7pF/kPa (more than 100-fold increase), and the device achieves the highest sensitivity with the largest initial capacitance (of 2.3nF) among all the capacitive sensors at the same dimension. In addition, the minimal detectable pressure of 33 Pa is

characterized on the sensor with the highest sensitivity.

Moreover, the membrane thickness plays another notable role in the device performance, as the sensitivity is inversely related to the 3rd power of the thickness. As plotted in Fig. 3b, by adjusting the membrane thickness from 75 μm to 175 μm , with a fixed spatial resolution of 2mm, the thinner membrane (of 75 μm -thick) shows a higher sensitivity of 77.7pF/kPa, while the thicker devices (of 175 μm -thick) membranes exhibit a lower sensitivity of 7.8pF/kPa. Furthermore, the targeted dynamic range can be tuned by the geometrical constrains. For instance, in the most sensitive design (3mm in resolution and 75 μm in thickness), the maximal pressure is around 7kPa, while the design of 2mm in resolution and 175 μm in thickness can extend the measurement range up to 450kPa. Overall, the spatial resolution and the membrane thickness of the MISA could be the determinant factors in the sensor performance (i.e., device sensitivity and dynamic range), allowing highly customizable sensors for a wide range of specifications and applications.

Response Time

Experiments have been conducted to characterize the response time of the MISA devices. A pulsed contact pressure (of ~ 1.4 kPa) in the frequency ranging from 10Hz to 100Hz has been applied to the device surface through an electromagnetically driven pin actuator. Both the driving voltages to the actuator and the capacitive changes are recorded. As shown in Fig. 4, the capacitive changes of the sensor repeats in the same frequency to the corresponding voltages applied to the pin actuator, suggesting that the sensor can response to the pressure in the frequency up to 100Hz. It is worth noting that the distortion of the recorded pressure signals likely attributes to the open-loop operation of the load applied by the actuator (i.e., the rapid rise edge and slow recovery phase of the capacitive readings).

Fig. 4 The time-resolved sensor response measurements under repetitive mechanical loads in the frequency of 10Hz and 100Hz (red curves indicate the input voltage to drive actuator, and the blue

curves are the output capacitance measured from a single sensing unit of the MISA device).

Surface Topology Mapping

To demonstrate the flexibility and adaptability of the MISA devices to artificial tactile applications, we have configured a sensor array to detect fine surface topology, such as Braille letters. As can be seen in Fig. 5, the custom MISA consists of 2×3 pixels with the spatial resolution of 2.3mm (to match with the standard Braille letters), and can be worn in a fingertip set up. For the fingertip reading of Braille texts, a gentle contact pressure has been applied to the text surface by the finger. The raised dotted impressions of each Braille character cause the membrane deformation in the corresponding droplet sensing units, which can be subsequently detected by the change of the interfacial capacitance. As shown in bottom panel of Fig. 5, using the finger-mounted MISA device, the letter of “BRAILLE” has been successfully resolved, in which each pressure reading is converted to a digital colorimetric scale. Digital recording of the tactile sensing can be further processed and transmitted into audible readings, and thus, it can be of potential use for Braille education for visually impaired patients.

Fig. 5 2×3 MISA device, amounted onto a fingertip, is configured to resolve raised dots of the “BRAILLE” characters in a Braille textbook.

Wrist Pulse Recording

Furthermore, we have utilized the ultrahigh device sensitivity and rapid response time of the iontronic droplet sensors to non-invasive cardiovascular pressure recording. An MISA device of 5×5 array with the spatial resolution of 3mm has been positioned in contact with the skin above the radial artery and fixed by a plastic wristband (Fig. 6a). Real-time pulse recording has been performed by scanning all the sensing elements covering the skin area of $15 \times 15 \text{ mm}^2$ at the sampling frequency of 1kHz in each unit. The MISA device enables two important functions in the pulse recording. First of all, the sensor spatially maps the pulse on the skin surface, from which the sites of the maximal pressure variations can be located. Comparing the pressure mapping results (Fig. 6b) with the sensor position (Fig. 6a), the pressure sensing units right above the radial artery provide the highest capacitive recordings (marked as I, II, III) as expected, and thus, closely reflects the cardiovascular pressure readings using the tonometry principle. In the following step, the pressure wave forms are continuously tracked from these optimal sensing positions. Fig. 6c shows the continuous pulse recordings from the three pixels, respectively. As can be seen, the

maximal pulse variation is around 1.2kPa recorded by Sensor II. Fig. 6d provides a close-up analysis of a single cardiac cycle, which has been characterized into three peaks (P_1 , P_2 and P_3). These maxima are caused by traveling waves of the systolic phase and diastolic phase of the blood pressure conducted in the elastic cardiovascular vessels [13]. Clinically significant parameters, such as the radial augmentation index, e.g. $AI (=P_2/P_1)$, can be directly extracted and computed from the maximal pulse recordings, which can be potentially used to screen the arterial compliance [14]. Moreover, the radial pulse waveforms recorded at the optimal sites can be further processed to estimate the central aortic pressure and cardiac output, which reflects the important cardiovascular events and the health states. Though a similar measurement has been conducted recently through a single-channel capacitive sensor, the IMA offers the combined advantage of simultaneous pressure mapping of the optimal recording area and continuous tracking of the blood pressure waveform, in addition to its flexible transparent packaging [4]. In this fashion, the MISA can serve as a flexible sensing device that is highly attractive for the emerging wearable health monitoring applications, in comparison with the conventional invasive cardiovascular monitoring.

Fig. 6 Real-time wrist pulse measurements. *a*, illustration of a transparent MISA device of 5×5 array with each pixel size of $3 \times 3 \text{ mm}^2$ embedded in a wrist band for pressure wave recording. *b*, the spatial distributions of the pulse intensities mapped by the MISA. The highest pressure variation readings, marked as I, II and III, are located above the radial artery, in corresponding to the positions of the three units in *a*. *c*, time-resolved strongest pulses recorded by the marked sensing units in *a* and *b*, respectively. *d*, a close-up view of one pulse signal recorded by sensor II in *c*, in which P_1 , P_2 and P_3 represent the three consecutive peaks of the pressure wave in each cardiovascular cycle.

CONCLUSIONS

In this paper, we have presented a novel microdroplet-based iontronic sensor array (MISA) with a very simple architecture and tunable sensitivities. The interfacial sensing principle utilizes the presence of large unit-area EDL capacitance at the elastic droplet-electrode contact. A theoretical model has been proposed to analyze the influences of the geometrical parameters (e.g. spatial resolution and membrane thickness) on the device sensitivity. Taking advantage of ultrahigh sensitivity, fast mechanical response, simple fabrication, mechanical flexibility and optical transparency, the iontronic microdroplet sensors are expected to be employed in a wide range of applications, including robotics, medical prosthetics, surgical instruments, video gaming and wearable computing

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