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A Textile-Based Stretchable Multi-Ion Potentiometric Sensor

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Recent trends in personalized healthcare have led to growing demands for real-time monitoring of the physiological status of human subjects. Detecting disorders in electrolyte levels, such as hyperkalemia, hypernatremia, hypokalemia or hyponatremia is very important and challenging.[1,2] Electrolyte imbalance represents a potential risk of fatal abnormal heart rhythms.[3,4] Electrolyte loss is a major concern in disorders such as cystic fibrosis[5] and hyperhidrosis.[6] Monitoring key electrolytes may also alert to latent cardiac problems[7] and diagnose apparent life-threatening events.[8,9] Overall, real-time measurements of electrolyte concentrations could indicate the patient, doctor, coach or athlete to potential electrolyte loss,[10] dehydration status[11] and the associated need for electrolytes replenishment.[12] Cumbersome methods to measure the sodium and potassium imbalance, such as electrocardiogram and sweat patch, have been reported.[13,14] Recent activity have led to wearable electrochemical (potentiometric) devices for noninvasive electrolyte monitoring.[15,16] However, the success of wearable sensing devices for health monitoring requires proper attention to key challenges concerning their mechanical resilience and large-scale manufacturing. Recent efforts to address these issues have relied on stretchable printable electrochemical devices.[17]

This article reports for the first time on a highly stretchable and printable textile-based potentiometric sensor array for simultaneous multi-ion sweat analysis using variety of fabric materials toward diverse healthcare and fitness applications. Screen-printing has been applied recently for fabricating amperometric sensors[18] and biofuel cells[19] on common textiles, but not in connection to potentiometric sensors or stretchable textile devices.[20] Textiles are attractive components of wearable sensing devices and offer rich elastomeric properties toward achieving conformal contact between the sensor and the body. Integrating chemical sensors directly into fabrics offer major advantages for future healthcare monitoring systems. Yet, a key issue involving textile-based sensing devices is the ability to operate under extreme mechanical tensions (that reflect daily activity) without compromising their analytical performance. In this work, we realized highly stretchable textile-based potentiometric sensors by combining polyurethane (PU)-based ion-selective membranes and inks with a serpentine sensor pattern and recently developed stretch-enduring printed electrodes.[21] The compositions of the selective potentiometric membrane and of the printed inks have thus been tailored for ensuring selectivity, electrical conductivity, reproducible printing, and strong adherence to conventional textiles. To provide the necessary biocompatibility and further resistance to mechanical stress we relied on polyurethane as replacement to the common polyvinyl chloride (PVC) matrix of ion-selective sensing membranes as well as the binder of the printed carbon nanotubes (CNT) trace. Polyurethanes have attractive mechanical and biocompatibility properties that make them suitable for many wearable devices. These materials are known to minimize unwanted inflammation, fouling, and other adverse physiological effects,[22] while providing exceptional analytical performance using potentiometric technique.[23] As illustrated in **Figure 1**A,B, such PU-based membrane and CNT ink have been coupled to an Ecoflex-containing Ag/AgCl ink and combined with a solid-contact reference electrode, leading to highly stretchable textile-based potentiometric sensors that transduce their potential response under extreme mechanical stress. As shown in Video S1 (Supporting Information) the resulting textile-based potentiometric sensors sustain extreme conditions, including large stretching and bending deformations. Such resiliency is crucial for realizing high-performance wearable potentiometric sensing devices.

The resilience and analytical performance of the novel dual-electrolyte textile-based printable wearable potentiometric sensor was examined using open-circuit potential measurements (Figure 1B). A calibration curve was executed by recording the electromotive force versus the time and changing the concentration of NaCl and KCl. Sensor array in Figure 1C exhibited a Nernstian response of 59.4 mV/log [Na^+], linear range from 10^{-4} M up to 10^{-1} M and limit of detection (LOD) of $10^{-4.9}$ M for its sodium selective electrode (Na^+ISE) and a Nernstian response of 56.5 mV/log [K^+] from 10^{-4} M up to 10^{-1} M and a LOD of $10^{-4.9}$ M for the potassium selective electrode (K^+ISE). Both results are comparable to the values reported previously using similar ion-selective membranes.[24] These dynamic ranges cover the physiological sodium and potassium levels in sweat before, during, and after prolonged exercise.[25] Electrolyte concentrations progressively increase during such exercise activity to provide a useful indicator of the dehydration status.[26] Moreover, understanding the amount of ionic species loss during exercise could facilitate recovery of the ionic balance during and after exercise.[27]

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