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Wearable Sensors

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This virtual issue of *ACS Sensors* on wearable sensing devices gives a taste of the current state of the art in this exciting field. Papers selected from the period 2020−2022 showcase the interdisciplinary nature of wearable sensors, where engineering, materials chemistry, analytical chemistry, electrochemistry, applied spectroscopy, microfluidics, bioanalysis, physics, data science, and medicine all form important elements of progress. *ACS Sensors* has published a number of reviews and perspective articles on wearable sensing systems that are not included in this virtual issue.[1−7] It is a productive and developing research field and for this reason this virtual issue can unfortunately not be comprehensive. The 29 publications shown here have been selected to showcase the breadth of methodologies, materials, and analytical targets reported in this journal.

To date, wearables have been developed for a diversity of reasons. Perhaps the most active application area has been in health monitoring, with a wide range of wearables being used to observe activity, physiology, and environment in real time. Innovative advances in the recognition and sensing of a wide range of target analytes have been reported, often using electrochemistry as a low power method. In beautiful work by Ganguly and co-workers, aptamer-modified electrodes were used to detect cortisol in sweat by electrochemical impedance spectroscopy, suggesting that continuous on-body measurements for up to 8 h are possible.[8] Electrochemical detection of nicotine in sweat was demonstrated by Javey and colleagues, coupling a nicotine oxidizing enzyme to a nanodentrite-modified electrode.[9] In addition, vitamin C in sweat was detected by Joseph Wang's team through enzymatic degradation and the use of an oxygen electrode to monitor oxygen loss upon enzyme conversion.[10] Lastly, Crespo and co-workers presented an enzymatic electrochemical lactate sensor, where the analyte flux was restricted with a plasticized polymeric membrane covering the enzyme layer, thereby improving robustness and ensuring that the signal is less prone to enzyme loss.[11]

Elegant studies have also leveraged spectroscopy for readout. For example, Mitsubayashi and colleagues were able to optically image ethanol emanating from the skin with nicotinamide adenine dinucleotide-dependent alcohol dehydrogenase embedded in a mesh. The reaction yields fluorescent NADH that provides real-time, spatially resolved ethanol concentration information about various locations on the skin.[12] Variations in glucose on the skin were also detected using fluorescence by Wu and co-workers, using two luminescent nanomaterials, of which one is degraded by the hydrogen peroxide product of the glucose oxidase enzyme reaction, thereby turning the fluorescence on.[13] Subcutaneous measurements of pH and lactate were measured continuously by Nguyen et al. using fluorescence and a simple LED light source.[14] For lactate, an oxygen sensor was used that tracked diminished oxygen upon enzymatic conversion of lactate, which is in some way analogous to Wang's work described above. In other exciting studies, Quan Liu's group demonstrated the enzyme-free detection of subcutaneous glucose by surface enhanced Raman Spectroscopy using monolayer-covered silver coated arrays, with results on rat models being successfully validated.[15] Finally, a completely wireless glucose sensing approach was reported by Christopher Reiche's group, where boronic acid modified hydrogels were implanted and observed by a routine medical ultrasound transducer.[16] Variable glucose concentration was found to swell the hydrogel in real time at a specific tunable frequency, with the results also being successfully cross-correlated.

From an engineering perspective, wearable sensors come in a wide variety of forms. These include patches, tattoos, facemasks, contact lenses, fabrics, bandages, spectacles, and watches. While the choice of format is often dictated by the analyte or property to be measured and the environment in which the sensor should function, other factors such as operational lifetime, discreteness, and the complexity of the decision-making process are equally as important. That said, the careful selection of substrate material is always key to creating a successful wearable sensor, since the material must facilitate sensing, while also possessing a range of other features, which may include flexibility, toughness, processability, biodegradability, and transparency. In this regard, Luo and co-workers have described a flexible patch for continuous blood glucose level monitoring that integrates biodegradable microneedles.[17] In a similar vein, Müller et al. fabricated acrylate-based microneedle arrays to measure oxygen within the interstitial fluid of the skin.[18] In both studies, the biocompatibility of the sensor is a critical issue in engendering long-term *in vivo* use. Conversely, Zhu and colleagues have used commercial contact lens materials to fabricate smart contact lenses able to perform wireless intraocular pressure measurements, using a spectacle-integrated impedance-based reader.[19] Highly stretchable substrate materials for the realization of head-band integrated potentiometric sensors

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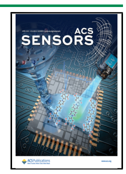

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ACS Sens. 2023, 8, 1368−1370

for sweat monitoring were reported by Xu and co-workers, demonstrating minimal signal change upon stretching the device 200%.[20] Interestingly, Bae and colleagues recently introduced a simple method for making stretchable optical waveguides from elastomers. These form the basis of wearable optical sensors (integrating LEDs, heaters, and photodetectors) and have been successfully used to monitor heart rate, breathing, and blood oxygen saturation.[21] Due to its low cost and availability, paper is particularly interesting for single use colorimetric applications, where test outcome can be read by eye or a smartphone. That said, integrating flow control in an efficient and low-cost manner can be challenging. To address this issue, Vaquer et al. integrated dissolvable polymer valves within a wearable urea biosensor that detects pH of sweat over extended time periods.[22]

While many wearable sensors have been developed to monitor basic physical properties, such as temperature, pressure, and motion, the analysis of bodily fluids (e.g., sweat, urine, interstitial fluid, and breath) requires more sophisticated processing and thus necessitates the integration of functional components within the sensor construct. Fluidic manipulations are perhaps foremost in this regard, with the sensor needing to manipulate fluids in an efficient but passive manner. Some excellent examples of fluidic integration have been published in *ACS Sensors* over the past two years. For example, Vinoth and co-workers fabricated wearable microfluidic sensors that enable the electrochemical monitoring of sweat biomarkers during exercise.[23] Here, fluidic channels are used to rapidly capture and direct sweat to sensing electrodes, allowing the electrochemical detection of ions and pH. In a similar manner, Hozumi and colleagues developed a flexible sensor for the simultaneous monitoring sweat glucose, heart activity, and skin temperature.[24] The ability to perform such measurements in real time was enabled by the use of a fluidic channel that refreshes sweat over the sensor surface. Additionally, Choi et al. reported the development of a multilayer capacitive sweat rate sensor, integrating a central microfluidic channel to allow real-time sweat rate monitoring without the need for microfabricated electrodes.[25] In a related study, Saha and colleagues presented a continuous sweat lactate sensor, integrating osmotic sweat extraction, paper-based microfluidics (to control sweat transport), and a screen-printed electrochemical sensor. This work is notable since the sensor enables the continuous monitoring of sweat lactate during both rest and exercise.[26] More recently, Zhang and associates described a clever fabric-based microfluidic wearable for calcium monitoring in sweat.[27] Here, the authors infused laser-cut thermoplastic films into fabrics to form integrated microfluidic circuits, that were effective in delivering sweat toward screen-printed electrodes. This work is especially exciting, since it suggests a simple route toward mass production of smart clothing in the future. Finally, it is interesting to see activity in the development of smart bandages for wound status monitoring. Two excellent examples in this regard have been reported by Charkhabi and Liu. In the former study, an LC-resonator is embedded in a commercial dressing and used to monitor wound healing in a rat cohort.[28] In the latter, a multiplexed sensing bandage was used for the real-time monitoring of sodium, potassium, calcium, pH, uric acid, and temperature to provide an early diagnostic of infection and inflammation.[29]

From a materials perspective, recent years have seen the incorporation of a range of advanced functional materials within wearable sensors. Compelling examples in this regard include the use of copper-based MOFs (metal organic frameworks) on freestanding titania nanochannels for the chemiresistive sensing of nitric oxide at ppb levels,[30] a bifunctional gas sensor incorporating gold nanoparticle-modified Au InSe nanosheets for real-time monitoring of atmospheric ammonia and nitrogen dioxide,[31] a neuron-mimic gas sensor (comprising gold quantum dots on Bi_2S_3 nanosheets) for the sensitive detection of nitrogen dioxide,[32] a porous PDMS capacitive pressure sensor for head trauma assessment,[33] and MXene-based resistive tattoos as sensitive strain sensors for the continuous monitoring of pulse rate, respiration rate, and muscle response.[34]

Finally, a desirable feature of any wearable sensor is that it should be small and ideally unobtrusive. While the sensor itself may fit the bill, the ability to miniaturize the power supply is often more problematic. One solution to this problem is to harvest (and store) energy from the wearer or environment using thermoelectric materials. Using such an idea, Li and co-workers have recently presented a self-powered, fabric-based temperature sensor.[35] The sensor comprises layers of compressible spacer fabric and thermoelectric material (PEDPT:PSS), and is able to sense both temperature and pressure variations with excellent resolution and response times. More importantly, the power needed to drive the sensor is generated by the temperature difference between the wearer and the environment. Moreover, Zhang and colleagues have developed biodegradable facemasks for respiratory disease diagnosis.[36] Their sensor, based on a polylactic acid electret fabric and carbon paper electrode layer, is able to successfully diagnose individuals suffering from asthma, bronchitis, and chronic obstructive pulmonary disease within a few minutes. Importantly, the facemasks are simple to make, comfortable to wear, and integrate portable readout circuitry.

To conclude, we hope that you will agree that wearable sensor research continues to thrive and evolve. Indeed, our only wish was that we could have included more papers in this virtual issue. Regardless, we hope that you will enjoy the collection and appreciate the exceptional science and technology.

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ACS Sens. 2023, 8, 1368−1370

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