Made available by Hasselt University Library in https://documentserver.uhasselt.be

Monitoring Body Fluids in Textiles: Combining Impedance and Thermal Principles in a Printed, Wearable, and Washable Sensor Peer-reviewed author version

JOSE, Manoj; OUDEBROUCKX, Gilles; BORMANS, Seppe; Veske, Paula; THOELEN, Ronald & DEFERME, Wim (2021) Monitoring Body Fluids in Textiles: Combining Impedance and Thermal Principles in a Printed, Wearable, and Washable Sensor. In: ACS Sensors, 6 (3), p. 896 -907.

DOI: 10.1021/acssensors.0c02037 Handle: http://hdl.handle.net/1942/34144 * Unknown * | ACSJCA | JCA11.2.5208/W Library-x64+tmanuscript.3f (R5.0.i3:5004 | 2.1) 2020/02/05 13:43:00 | PROD-WS-116 | rq_6161757 | 1/23/2021 09:24:31 | 13 | JCA-DEFAULT



pubs.acs.org/acssensors

Article

¹ Monitoring Body Fluids in Textiles: Combining Impedance and ² Thermal Principles in a Printed, Wearable, and Washable Sensor

3 Manoj Jose, Gilles Oudebrouckx, Seppe Bormans, Paula Veske, Ronald Thoelen, and Wim Deferme*

Cite This: https://dx.doi.org/10.1021/acssensors.0c02037



ACCESS Metrics & More Article Recommendations Supporting Information

4 **ABSTRACT:** This work explores the feasibility of coupling two 5 different techniques, the impedance and the transient plane source 6 (TPS) principle, to quantify the moisture content and its 7 compositional parameters simultaneously. The sensor is realized 8 directly on textiles with the use of printing and coating technology. 9 Impedance measurements use the fluid's electrical properties, while 10 the TPS measurements are based on the thermal effusivity of the 11 liquid. Impedance and TPS measurements show equal competency 12 in measuring the fluid volume with a lowest measurable quantity of 13 0.5 μ L, enabling ultralow volume passive measurements for sweat 14 analysis. Both sensor principles were tested by monitoring the 15 drying of a wet cloth and the measurements show perfect



16 repeatability and accuracy. Nevertheless, when the biofluid property changes, the TPS sensor does not reflect this information 17 on its readings, whereas, on the other hand, impedance can provide information on compositional changes. However, since the 18 volume of the fluid changes simultaneously, one cannot differentiate between a volume change and a compositional change from 19 impedance measurements alone. Therefore, we show in this work that we can apply impedance to measure the compositional 20 properties; meanwhile, the TPS measurements accurately carry out volume measurements irrespective of the interferences from its 21 compositional variations. To prove this, both of these techniques are applied for the quantification and composition monitoring of 22 sweat, showing the capability to measure moisture content and compositional parameters simultaneously. TPS measurements can 23 also be an indicator of the local temperature of the medium confined by the sensor, and it does not influence the fluid parameters. 24 Compiling both impedance and thermal sensors in a single platform triggers smart wearable prospects of metering the liquid volume 25 and simultaneously analyzing other compositional changes and body temperature. Finally, the repeatability and stability of the sensor 26 readings and the washability of the device are tested. This device could be a potential sensing tool in real-life applications, such as 27 wound monitoring and sweat analysis, and could be a promising addition toward future smart wearable sensors.

28 **KEYWORDS**: textile, biofluid, printed, moisture content, composition analysis, sweat monitoring, ionic concentration, 29 temperature measurement, washability

harles Darwin wrote in the 1800s "It is not the strongest 30 of the species that survives, nor the most intelligent, but 31 32 the one most responsive to change". Nowadays, the health care system has been transiting from physical hospitals toward 33 34 virtual hospitals where the patients themselves carry mini-35 aturized versions of medical labs, i.e., wearable sensors that do 36 not interrupt the human being's normal life. Physical, chemical, 37 and biological human body responses become the primary 38 analytes for future health monitoring in medical systems. 39 Although health-monitoring smartwatches and activity-tracking 40 health bands have been exhibiting growing potential for 41 commercialization, biofluid-based sensor patches are yet to be 42 matured for health markets. Peripheral body biofluids such as 43 sweat and wound swabs are analytes containing human body 44 responses that assist the vital learning of the physiological and 45 psychological aspects of human health in a noninvasive 46 manner. Hydration monitoring,¹ perspiration analysis,² stress 47 monitoring,³ early detection of cystic fibrosis,⁴ and thermal-

comfort detection are some of the major health-tracking 48 proposals of sweat volume and analyte-based wearables. 49 McColl et al. utilized moisture-level tracking to optimize 50 wound dressing management,⁵ and many other studies report 51 on wound exudate analysis to make effective wound-healing 52 monitoring.^{6,7} There are also reports for numerous feasibility 53 studies employing wound discharge pH to monitor the healing 54 progress,^{8,9} and other researches have shown the exudate 55 impedance changes as a pointer for infection detection.^{10,11} 56 The analytical studies based on the biofluids largely depend on 57

Received: September 30, 2020 Accepted: January 14, 2021



58 the quantification of biomarkers such as volume,^{12,13} ionic 59 concentration,^{14–16} bacterial presence,^{10,17} pH,^{9,18} and similar 60 parameters.

Most of the reported studies in the literature took advantage 61 62 of monitoring the electrical properties for determining the 63 aforementioned biomarkers, whereas these biomarkers are 64 generally tracked individually without any cross-sensitivity in 65 laboratory conditions. Bacterial detection in (simulated) 66 wound exudate is conducted on a defined volume of the test 67 liquid,¹⁹ and the perspiration analysis assumes a uniform sweat 68 rate. Wearable sweat analyte-monitoring studies have been 69 reported, focusing on the sportsman's biochemical and 70 physiological account to optimize the performance. However, 71 recent studies emphasize the necessity of volume measure-72 ments for comprehensiveness.^{20,21} Such errors can also occur 73 during the practical measurements of pH of the biofluids.²² 74 Many wearable sensors in the literature have been originally 75 designed to sense a predefined volume of the test fluid but may 76 lose accuracy when exposed to an unknown volume. Moisture 77 content tracking on wearable applications suffers from 78 measurement errors due to the variations in electrical 79 properties of the body fluids.^{23–25}

To avoid the cross-sensitivity on sweat rate measurements in 80 81 thermal-comfort monitoring, Sim et al. developed a watchlike 82 device that uses expensive and high-power-consuming complex 83 methodologies to perform accurate sweat rate sensing. This 84 system consists of heaters, humidity sensors, actuators, and a 85 diaphragm-enclosed frame, which are limited to be a wrist 86 watchlike instrument and are not suitable for any other 87 wearable textile applications mentioned before.²⁵ Similarly, 88 sweat rate measurements performed with the help of 89 microfluidic channels integrated with the heater and two 90 thermocouples at the sides are reported elsewhere. The sweat 91 in the channels flow through the heater, and the temperature 92 difference between the thermocouple indicates the sweat 93 rate.¹³ There were also attempts to carry out sweat rate 94 measurements with the help of humidity sensors. Two 95 identical humidity sensors were arranged at a differential 96 distance from the skin, where a gasket was used to maintain 97 definite spacing from the skin.²⁶ There are also sweat 98 collection devices built to quantify sweat loss and they use 99 microchannels reported in previous studies.^{27,28} A recent study 100 developed a novel methodology for early detection of cystic 101 fibrosis, and here the researchers use potentiometric measure-102 ments to monitor the sweat chloride content in the biofluid. 103 This work also studied sweat rate measurements with the help 104 of an additional sweat collecting device to attain error-free 105 measurements.¹⁵ These kinds of electrochemical potential 106 quantifications on wearables are not affected by the fluid 107 volume above the required threshold to perform measure-108 ments. Current approaches utilize the active stimulation of the 109 sweat glands with exercise to produce an adequate volume of 110 sweat (more than 10 μ L) to be able to perform the analysis. 111 This, however, is often not feasible for wearable functionalities 112 especially for patients, infants, and elderlies. Besides, many of 113 these sweat measurement/collecting devices use channels, 114 pumps, and relatively complex poly(dimethylsiloxane) 115 (PDMS)-based microfabrication strategies, the downside of 116 which are high power consumption, long-time heating, high 117 cost, and not being feasible to be integrated into textiles. On 118 the other hand, laboratory-based testing applied for wearable 119 systems often assumes a definite volume of the wound exudate 120 or sweat rate to quantify its biomarkers. Contrarily, the

moisture content (volume) measurements of the biofluids are 121 carried out assuming identical electrical properties of the 122 liquids, which is not always the case. Therefore, erroneous 123 measurements are achieved, which can only be solved when 124 both volume sensing and ionic concentration measurements 125 are independent of each other. 126

The selectivity toward biomarkers, their reproducibility, and 127 difficulties in integrating conformal fluid monitoring sensors 128 into textiles still have bottlenecks, spanning from the sensor 129 fabrication to sensing principles and the optimal integration of 130 multiple techniques in a wearable platform. Conventional 131 sensor fabrication uses glass or ceramic substrates applying 132 microfabrication techniques, such as photolithography, sputter- 133 ing, and etching, and the processing involves high-temperature 134 curing above 300 °C.^{29,30} These sensors are unconformable to 135 the human body, and mechanical mismatch with the skin elicits 136 motion artifacts and end-user inconvenience. Multistimulant 137 sensible skinlike devices,^{31,32} wearable human emotion 138 monitoring patches,³ breathable and stretchable temperature 139 sensors,³³ thermal principle-based moisture monitoring 140 sensors, and many more have been developed over the last 141 few years to surpass the shortcomings of conventional systems. 142 All of these major studies are promising ones but focus on 143 clean room-based microfabrication of flexible devices on 144 polyamides, PDMS, etc. However, complicated sensor 145 fabrication steps, limited substrate selectivity, and the necessity 146 for huge investment in the production facilities are certain 147 disadvantages. Development of low-temperature processable 148 functional inks and compatible high-speed printing techniques 149 launched opportunities for mass production of conformable 150 functionalities onto flexible substrates. Although microfabrica- 151 tion techniques are superior for feature size, printing opens up 152 adaptability in terms of substrate, cost, and production speed.³⁴ 153 Printing is an additive manufacturing technique that simplifies 154 the production steps and reduces production waste. The 155 compatibility of printing the sensors on textiles shows massive 156 opportunities for textile-based wearable applications. A few 157 studies on multiparameter-sensing smart bandages,³⁵ activity 158 trackers,³⁶ and imaging modality on brassiere for cancer 159 detection³⁷ have been reported. However, it still remains for 160 printed sensors on textiles to explore in detail the wearable 161 biophysical and biochemical measurements for health and 162 comfort.

This article addresses the research problems related to 164 wearable biofluid monitoring as specified before. Generally, the 165 wearable biofluid tracking sensors encounter uncertainty 166 between the volume of the liquid and the concentration of 167 the ions/biological contents present in it, especially in 168 electrical property-based sensors. When it comes to the 169 volume-based electrical parameter-detecting moisture sensors, 170 the accuracy in measurement is substantially affected due to 171 the presence of ions/metabolites. Similarly, electrical property- 172 based health-monitoring wearables are adversely affected due 173 to the unknown volume of the body fluid. To circumvent these 174 shortcomings, the proposed work in this article attempts to 175 club two measurement techniques for accurate and precise 176 quantification of biofluid and its biomarkers without 177 disruption. Impedance-based health monitoring in wearables 178 has been reported in previous studies.^{1,38} It is a widely 179 accepted fluid-sensing technique adopted as one of the 180 measurement techniques in this work. Prof. Gustafsson 181 developed the transient plane source (TPS) method in 1979, 182 as an alternative technique to measure the thermal 183



(1)Screen printing(2) Silver paste(3) Heater pattern on screen (4)Textile substrate(5) Coating head(6) Tubiguard Spray mist(7) masking (8) blade coating(9) Tubicoat ink(10) silver paste(11) screen with Interdigitate electrode design(12) Tubicoat spray mist(13) masking

Figure 1. Sensor production flow. (A) Screen printing heater design on textiles. (B) Ultrasonic spray coating of the Tubiguard layer. (C) Blade coating Tubicoat. (D) Screen printing interdigitated electrodes (IDEs). (E) Ultrasonic spray coating of Tubicoat. (F) Final sensor on textiles.

184 conductivity of materials.³⁹ Based on this work, Schönfisch et 185 al. presented the moisture content assessment on textiles 186 applying the TPS method and the sensor consisted of a 187 cleanroom microfabricated heater on polyamide foil.⁴⁰ The 188 initial part of this article focuses on the inclusion of the TPS 189 and impedance sensors into a single wearable textile platform. 190 Selection of textile as the sensor substrate ensures conform-191 ability and feasibility for all range of wearable applications. As 192 textile-compatible fabrication techniques, screen printing⁴¹ and 193 ultrasonic spray coating^{42,43} for sensor fabrication ensured the conservation of textile wearability attributes. The final printed 194 sensor system on textile combines the impedance and TPS 195 196 technique, and both are compared for the moisture content (biofluid volume) measurement over a wide range, even with a 197 198 smallest measurable quantity of 0.5 μ L, which allows for 199 passive ultralow volume sweat analysis. This work investigated 200 the prospect to use sweat volume and composition analysis 201 without interference. The sensor system is designed and 202 applied for multiple biomarker detection, and it results in 203 accurate computations without any interference. Coupled with 204 biofluid monitoring, the human body's temperature is a highly 205 pertinent biomarker to be tracked for wearables. Besides the 206 fluid measurements, the TPS sensor system could manage to 207 perform passive temperature tracking, which is also shown in 208 this work. Finally, as the proposed sensors are fabricated on 209 textiles, the washability of the sensors is desirable. Therefore, 210 washing tests are performed, showing this printed sensor 211 system's potential for long-term applications in wearables.

212 MATERIALS AND METHODS

Sensor Fabrication. The textile substrate used in this work is a 214 polyester woven fabric (100% PES, washed and fixated, kw11401) 215 from Concordia Textiles (Valmontheim, Belgium) with an average 216 roughness of 6 μ m. Silver paste (flexible silver paste) was purchased 217 from Gwent group (Pontypool, U.K.), which has already been proven 218 for its flawless performance in flexible device applications.⁴¹ A water-

based dielectric ink Tubicoat and the insulation ink Tubiguard was 219 supplied by the CHT group, Tübingen, Germany. 220

TPS Moisture Sensor Fabrication. The TPS moisture sensor is 221 directly fabricated on the textile substrate without a planarization 222 layer. A coil heater is designed with track width and an interturn 223 spacing of 500 μ m and is screen-printed onto the textile surface (see 224 Figure 1B). The heater design is printed using silver paste and cured 225 f1 at 160 °C for 20 min in a box oven to make it conductive and to 226 remove all of the ink additives from the printed silver. The resultant 227 heater structure has resistance in the range of 75–150 Ω at room 228 temperature. Following this, a thin layer of Tubiguard formulation is 229 ultrasonically spray-coated on the top of the printed silver heater. 230 During spray coating, the heater area was left exposed and the textile's 231 remaining area was masked with a foil. After coating, the Tubiguard 232 layer is cured at 160 °C for 20 min in the box oven. The heater design 233 used for sweat monitoring was smaller than the one mentioned here. 234 The track width and spacing is set to 400 μ m for the smaller heater. 235

Impedance Moisture Sensor Fabrication. The impedance sensor 236 is fabricated on the textile substrate next to where the TPS sensor is 237 printed. The textile substrate is blade-coated with a Tubicoat 238 formulation layer to obtain a better feature size of printed electrode 239 fingers. This sandwich architecture enhances the dielectric properties 240 of the sensor. The coated layer was cured at 130 °C for 15 min in a 241 box oven. Interdigitated electrodes of area $20 \times 10 \text{ mm}^2$ are designed 242 for the moisture sensor (see Figure 1A). The electrodes with a finger 243 width and an interspacing of 400 μ m are prepared by screen printing 244 onto the coated side of the textile substrate, and silver paste is applied 245 as the conductive electrode material. The printed silver electrode is 246 cured in a box oven at 120 °C for 10 min. As a top layer for the 247 sensor, the Tubicoat material is deposited by ultrasonic spray coating. 248 The Tubicoat formulation is diluted to 5 wt % in water and kept in an 249 ultrasonic bath for 5 min to make a homogeneous and fine dispersion. 250 Ultrasonic spray coating of the diluted Tubicoat material underwent 251 multipass coating for adequate thickness. During this step, the TPS 252 sensor was masked with a foil to avoid the deposition of Tubicoat 253 over it. 254

Agilent 4284A precision LCR meter from Hewlett Packard and 255 PXIe-4139, a source measure unit from National Instruments, are 256 used for the impedance and TPS sensor measurements, respectively. 257



Figure 2. SEM cross section of impedance (A) and TPS sensors (B) and EDX and FTIR for the sensing layer of the impedance sensor (C and D, respectively).

Theory of Operation.^{39,40,44-46} Impedance Sensor. The 258 259 electrical impedance measurement of the moisture is based on tracking the changes in the sensor's electrical resistance and reactance 260 when its surface comes in contact with the moisture. The proposed 261 sensor consists of conductive interdigitated electrodes (IDEs) covered 2.62 with a moisture-responsive dielectric medium. The moisture sensor's 263 electrical model suggests that the moisture layer's intrinsic 264 conductivity contributes to a change in the electrical resistance of 265 266 the sensor.⁴⁷ Many moisture-sensing devices reported in the literature 267 thus use fluid resistivity as the sensing parameter to quantify the fluid volume. However, the presence of moisture also produces variations 268 269 in the permittivity of the dielectric sensing layer, resulting in a 270 proportional impedance change.⁴⁸ In addition to this, the changes in 271 the impedance occurring at the interface of the liquid and the sensing 272 layer also add up to the total impedance, Z, and are summed up as in 273 eq 1

$$Z = \frac{1}{2\Pi fC} + R \tag{1}$$

27

Here, *R* is the resistance, *C* is the capacitance, *Z* is the impedance, and $_{275}$ *f* is the frequency. $_{276}$

TPS Sensor. The sensor's thermal measurements are established 277 around a metallic heating element driven to Joule heating. As the 278 heating element heats up, the temperature-induced resistance change 279 is related by the renowned equation 280

$$R_{(t)} = R_0 (1 + \alpha \Delta T) \tag{2}_{281}$$

where R_0 is the initial resistance, $R_{(t)}$ is the resistance at any instant t, 282 and α is the temperature coefficient of resistance (TCR). The graph 283 plotted with resistance against temperature is a straight line, and the 284 slope is the product of α and R_0 . The proposed work investigates 285 moisture sensing using a thin-film heater, applying the transient plane 286 source method for the measurements. The measurement is performed 287 using a system that precisely monitors the change in temperature as 288 the heater is triggered for a short heating pulse. When the heating 289 sensor attains a higher temperature, it forms a thermal interface with 290 the surroundings and the corresponding interfacial heat transfer is 291 explained by the heat equation. The solution for the one-dimensional 292 293 heat equation relates the change in temperature (ΔT) of the heater as 294 a function of thermal effusivity (*e*) and time, as can be seen in eq 3⁴⁹

$$\Delta T = f(e, t) \tag{3}$$

296 where the thermal effusivity (e) is defined as the ability of a medium 297 to exchange heat with its surroundings and is determined as the 298 square root of the product of volumetric heat capacity and thermal 299 conductivity, as shown in eq 4

$$_{300} \qquad e = \sqrt{C_{\rm v}\lambda} \tag{4}$$

301 where C_v and λ stand for volumetric heat capacity and thermal 302 conductivity, respectively.

A short pulse of a defined power heats up the heater. The 303 304 subsequent rise in temperature is in inverse proportion to the thermal effusivity of the contact medium. When the dry heater plane becomes 305 306 wet, the heat energy is quickly transported from the sensor plane to 307 the surrounding medium. As the heat energy is rapidly extracted by 308 the water content, the sensor's rise in temperature exhibits less 309 steepness in contrast to the sensor under dry conditions. The thermal 310 effusivity of water is approximately 250 times higher than that of air, 311 facilitating high sensitivity toward the presence of moisture of the 312 sensor. The magnitude of the abovementioned effusivity of the water-313 content-induced temperature drop can be obtained from eq 5 that 314 relates resistance and temperature. The variations in resistance 315 produce changes in the applied voltage as can be seen from the 316 equation below.⁴

$$\Delta V = I \Delta R_{(t)} \tag{5}$$

318 **RESULTS AND DISCUSSION**

 f_2

319 Flexible and Printed Sensors on Textiles. As described 320 in the Materials and Methods section, the impedance sensor is 321 composed of IDEs and a sensing medium. The sensor 322 performance in terms of sensitivity and resolution is dependent 323 on the spacing between the interdigitates and the feature size 324 of the electrode fingers. The interdigitated electrode spacing 325 and feature size are limited mainly due to the printing process, 326 substrate, and ink composition. Such sensor fabrication on 327 textiles is often hindered due to the difficulties in fabricating 328 IDEs on textile substrates using conventional microfabrication 329 techniques. This work employed screen printing as a method 330 for the deposition of IDEs on textiles. Screen printing is known 331 for its reproducibility and good aspect ratio of the printed 332 structures. The architecture of the sensor layer buildup is 333 clearly observed from the scanning electron microscopy 334 (SEM) cross section of the sensor given in Figure 2. The 335 SEM images show that the IDEs printed on the textile 336 substrate are rather well reproduced and the feature size of 400 337 μ m line width is attained on the textile substrate. The presence 338 of organic compounds and silicon in the sensing material is 339 confirmed using energy-dispersive X-ray (EDX) spectroscopy (Figure 2C). Fourier transform infrared (FTIR) spectroscopy 340 341 (Figure 2D) confirms that the sensing material consists of a 342 polyurethane matrix and that the silicon dioxide is in the 343 dispersed phase. Polymers, especially polyurethane, are 344 moisture-sensitive materials and have been applied before for 345 humidity-sensing applications.^{50,51} Ceramic oxides alone serve 346 as moisture-sensing materials, and their polymer composites 347 improve their dielectric properties.

The intrinsic moisture conductivity, absorbed and adsorbed moisture on the polymer composite, changes the sensor's big dielectric and resistive properties. The sensor's exposure to moisture results in a thin conductive moisture film on the top to the sensing layer, which contributes to impedance changes. The sensing layer's absorption phenomena lead to the emergence of the changes in interface impedance and 354 capacitive and resistive changes on the dielectric sensing 355 layer. The abovementioned electrical quantities contribute to 356 the sensor impedance drop, which is proportional to the 357 volume of the test liquid. These sensing mechanisms can be 358 explained using the Cole–Cole plot for impedance-based 359 moisture sensors, as shown in Figure 3A. This plot is in 360 fb



Figure 3. Cole–Cole plot for impedance sensor for different fluid volumes (A) and transient heating of a sensor at two conditions (B).

agreement with that of the sensor reported in the literature, 361 which uses a polymer–ceramic composite layer for humidity 362 sensing.⁴⁴ A low volume of the moisture content and high 363 humidity levels have shown nearly the same Cole–Cole plots, 364 paving a way for predicting the different mechanisms 365 contributing to the impedance change. 366

Also, printed heaters can be used as thermal sensors for the 367 TPS-based moisture volume measurements. The plot (Figure 368 3B) of the normalized voltage change vs the square root of 369 time shows a sheer difference in the slope for wet and dry 370 sensor surfaces. This slope is used as a parameter to quantify 371 the water content on the surface of the sensor. Here, the TPS 372 measurement is envisaged in such a way that spatial sensitivity 373 is limited to the regime of the biofluid content on worn clothes 374

f4

375 and external ambient influences are needed to be segregated. 376 This is achieved with the help of restricting the penetration 377 depth of the heat pulse by setting the transient heating time (more details can be found in the Supporting Information). 378 379 This work used a transient heating time of 1 s. This can be 380 further optimized based on the end application. Prolonged 381 transient heating leads to noise disturbances, consumes more 382 power, and the sensor takes a long time to return to its initial 383 condition. The printed heater has a circular coil-like structure made of conductive material. As the width of the heater coil 384 385 and the spacing between them decreases, the initial resistance 386 of the heater increases. The impact of the initial resistance on 387 the sensitivity of the sensor is mentioned in the previous section. The heater coil feature size and the spacing between 388 389 them are desired to be small to enhance the sensor sensitivity and repeatability. The cross-sectional image of the sensor given 390 391 in Figure 2B shows the layer formation on the textile substrate. 392 The printed heater sensor is encapsulated with a thin layer coating that acts as a protection layer for the conductive tracks, 393 394 hindering the shunting between heater tracks during fluid testing. 395

Moisture Content Monitoring. This section investigates moisture volume measurements in textiles with the help of two different techniques. The sensors are characterized and tested for their performance in terms of work, sensitivity, precision, repeatability, and comparison of on with the other.

A moisture content measurement is performed using both 402 403 the thermal- and impedance-based sensing approaches, and a 404 comparison is made between them. In both approaches, the 405 experiments are initiated in a dry condition. Briefly, $1-250 \ \mu L$ 406 of the test liquid is dropped onto the sensor surface in discrete 407 volumes. The impedance measurements exhibit a drop at lower 408 frequencies, as shown in Figure 4A, whereas at higher 409 frequencies, the impedance remains almost constant, which is 410 as expected for resistive-capacitive circuits. As the volume of 411 the test liquid increases, the corresponding impedance drops 412 from a few megaohms to a few hundred ohms. Likewise, the 413 thermal principle (TPS) was also assigned for moisture volume 414 measurement. The printed heater is allowed to transiently heat 415 up for 1 s, and the presence of moisture induces variations in 416 the temperature rise of the heater. The presence of moisture 417 segregates the heat quickly from the heater surface, and this 418 heat transfer in turn reduces the heat buildup on the moist 419 heater surface. The transient heating time of 1 s and the 420 applied power are kept constant for the experiments. After 421 every transient heating cycle, the heater remains idle for 30-422 100 s while it returns to its initial condition (which depends on ⁴²³ how high the heating temperature is). The slope of the heating 424 curve is observed to drop with increasing moisture content, as 425 seen in Figure 4B, and this can be explained based on the 426 section theory of operation.

⁴²⁷ The relationship of the sensor output signal as a response to ⁴²⁸ the stimulus needs to be characterized, and this is known as the ⁴²⁹ sensor transfer function. The impedance sensor transfer ⁴³⁰ function is defined by measuring the impedance variations ⁴³¹ with moisture volume at a frequency of 100 Hz. At the same ⁴³² time, the TPS principle-based sensor is characterized by ⁴³³ measuring the slope of the normalized voltage vs the square ⁴³⁴ root of the time (\sqrt{t}) heating curve. The slope is calculated ⁴³⁵ between a transient heating time of 0.5 and 0.8 s^{0.5}. The curve ⁴³⁶ plotted between impedance and the liquid volume is best fitted ⁴³⁷ to a third-order exponential decay function with an adj R^2



Figure 4. Moisture volume measurement from 1 to 250 μ L. Impedance sensing (A) and TPS sensing (B).

value of 0.999, whereas the plot of the TPS sensor is best fitted $_{438}$ to a first-order exponential decay function with an adj R^2 value $_{439}$ of 0.991 (Figure 5). The sensitivity of the sensor can be $_{440}$ fs calculated from the derivative of the transfer functions (F1 for $_{441}$ impedance, F2 for thermal) and are given by eqs 6 and 7. $_{442}$

$$dF1/dx = -11956.9e^{-(x/4.6)} - 300390.6e^{-(x/0.86)} - 315.38e^{-(x/42.21)}$$
(6) 443

$$dF2/dx = 0.000598e^{-(x/4.6)}$$
(7) 444

where x is the volume of the test fluid. Both the proposed 445 impedance and TPS sensors have a least measurable volume of 446 1 μ L for the test fluid. The impedance-based moisture sensor 447 however has the potential to measure even smaller volumes, as 448 is illustrated in Figure 4A, where the impedance drops a few 449 orders of magnitude when the dry sensor becomes wet by 1 μ L 450 volume of the test liquid. The stability of the sensor readings 451 can be evaluated by repeatedly measuring the value of the 452 impedance for a particular volume of the test liquid. The 453 accuracy of the sensor is evaluated from recurring readings for 454 each test volume shown from 0 to 250 μ L volume in Figure 6. 455 for

F



Figure 5. Moisture-sensing characteristics for the impedance sensor at 100 Hz (A) and TPS sensor (B).



Figure 6. Impedance and TPS sensors' multiple readings for each liquid volume tested ranging from 0 to 250 μ L.

456 Slight deviations in the sensor readings mostly arise from the 457 dynamic spread of the liquid drop and its evaporation. The rate 458 of evaporation increases with spreading due to the increase in 459 the surface-to-volume ratio of the liquid drop. The accuracy of the measurements points out the necessity of another relevant 460 attribute, which is the reproducibility of the sensor measure- 461 ments in different cycles. 462

The reproducibility of the sensor readings over repeated 463 measurements is also investigated. This is an inevitable feature 464 for wearable sensors as it is directly correlated to health 465 diagnosis. Some minor misfits are observed between the sensor 466 readings of the repeated cycles, even at the regime of larger 467 volumes of the test liquid. The root cause for these variations is 468 attributed to the difference in the spreading of the liquid on the 469 sensor surface. It is observed that both impedance and thermal 470 measurements are influenced by the area of the liquid surface 471 coverage. To overcome this limitation, an even surface 472 distribution of the test fluid is a mandate. The improved 473 wearable sensor system could be designed in such a way that 474 the sensor is implemented to measure the volume of the liquid 475 contained in a textile or a similar material layer coated or 476 spread over the sensor surface, which is also shown in the next 477 section. This modified sensor system can thus eliminate the 478 aforementioned systemic errors, which are shown as drying of 479 the wet textile sample.

Monitoring the Drying of a Textile Sample and the 481 Sensors' Reproducibility. The primary goal of the proposed 482 sensors is to measure the volume of the moisture content 483 present in the wearables, which are wet by the secreted body 484 fluids, and is further quantified and analyzed. The moisture 485 volume assessment is made by monitoring the variations in the 486 impedance and thermal measurements over a period of time 487 where a piece of textile placed over the sensor surface is made 488 wet and set to dry. Here, phosphate saline buffer (PBS) is 489 applied for biofluids as it is widely recognized and used in 490 biosensor testing.^{2,11,35,52-54} Figure 7 shows the temporal 491 f7 evolution of (A) the impedance measurements of the sensor at 492 a constant frequency and (B) the slope of the transient heat 493 curve for thermal measurements. The sensor readings in the 494 below demonstrated experiment for tracking the drying of a 495 wet textile exhibited excellent reproducibility over repeated 496 cycles of measurements, as is evident in the results. This 497 feature counts as proof for the sensors' functionality to 498 measure moisture volume, its accuracy, and reliability for real- 499 life applications. 500

It can be noted here that both the sensor systems are tested 501 and verified for over a wide input full scale, even with capacity 502 for ultralow volumes. Both sensors are highly sensitive to 503 moisture changes and have good repeatability irrespective of 504 the fact that there are minor mismatches due to nonuniform 505 wetting issues. 506

Synergetic Action of Impedance and Thermal (TPS) 507 Sensors for Sweat Monitoring. As shown in the above 508 section, both sensors show consistent and reliable sensing 509 characteristics for volume measurements. However, wearable 510 moisture sensors coupled with biomarker detection would be a 511 pivotal novel functionality for comprehensive personal health 512 management. For example, bacterial detection in wound 513 exudate and chloride ion monitoring of sweat along with its 514 accurate volume measurements lead to an all-inclusive fluid 515 assessment. Figure 8 shows the sensor impedance dependency 516 f8 on different test liquid compositions like demi water, tap water, 517 and phosphate buffer (PBS). The impedance readings show a 518 completely unique sensing behavior for the test fluid where the 519 impedance changes in response to both volume and its 520 composition. This drives the impedance sensor into the 521 perspectives of biofluid composition analysis to understand 522



Figure 7. Drying of a wet textile monitored with impedance (A) and TPS sensors (B) shown over repeated cycles.



Figure 8. Impedance analysis for different fluids.

body health responses. However, this also raises a question 523 about the uncertainty arising from the simultaneous influence 524 of the biofluid composition and volume on the impedance 525 measurements. As the sensor testing goes beyond the 526 laboratory conditions from having defined volumes of the 527 test liquid to continuously varying and unknown volumes of 528 the biofluids, the uncertainty in impedance measurement 529 comes into action. 530

The composition and volume of biofluids such as sweat vary 531 depending on physical activities and physiological and 532 psychological factors. In wearables, it is vital to closely monitor 533 both the ionic composition and the volume of sweat. To 534 address this matter of contention, the sensor system should be 535 competent to quantify the volume of the sweat without any 536 considerable interference from its composition and vice versa. 537 The sweat used is prepared on the basis of European standard 538 EN1811.^{55,56} The sweat pH is adjusted to 6.5 with dilute 539 sodium hydroxide. The sweat ionic concentration is varied in 540 the physiologically relevant range. The concentration of 541 sodium chloride (NaCl) is varied to different molarities, and 542 different volumes of each are tested to see the feasibility of the 543 sensor to be used for such applications. A few millimolar 544 concentrations of sodium chloride ions in the test sweat 545 produced a significant change in the impedance of the sensor 546 (Figure 9A). Similarly, a magnificent change in the impedance 547 f9 is observed when the volume of the test sweat changes from 548 0.5 to 100 μ L, whereas different fluid compositions are 549 observed to have no significant influence on the TPS 550 measurements of the sweat (Figure 9B). Relative to previous 551 experiments, a more compact heater is used for the sweat- 552 monitoring experiments. Smaller and closer tracks are used for 553 the sweat experiments to reduce the errors from nonuniform 554 heating. Minor misfits in the TPS readings of different 555 concentrations, especially at higher volumes, are due to the 556 difference in the fluid dropping and its spreading. To prove 557 this, the sensor is tested with different concentrations of sweat 558 in equal volumes and is dropped on a thin tissue placed on the 559 heater surface. There was no difference observed, and all of the 560 readings were identical (Supporting Information, Figure S-2). 561 Compiling both impedance and TPS sensors in a single 562 platform develops more accurate and smart wearable prospects 563 of metering liquid volume and the simultaneous analysis of 564 other electrical properties. The proposed sensor platform has a 565 sensitivity to respond to a fluid volume of 0.5 μ L or possibly 566 less. Therefore, the synergistic operation of the two sensors in 567 a combined platform enables the impedance sensor to track 568 any compositional changes, such as ionic composition, in the 569 sweat and the thermal sensor to determine their volume. 570

Temperature Monitoring as a Complementary 571 **Functionality.** Temperature variations of the body or a 572 specific body part guide disease diagnosis, activity tracking, and 573 infection detection. This is considered one of the most 574 important biomarkers for wearables. Metals have a positive 575 temperature coefficient of resistance (TCR), and their 576 resistance changes based on the temperature and its TCR. 577 Wearable temperature sensors employing metal-coil-like 578 structures have shown good sensitivity and accuracy.⁵⁷ The 579 printed silver structures in this work exhibit a TCR of 580 0.001628, which is comparable to previously reported values.⁵⁸ 581 The heater structure's initial resistance is proportional to the 582 temperature of its medium, and Figure 10A indicates that the 583 f10 resistance increases in proportion to the temperature of the 584 sensor-enclosed oven. Figure 10A shows that the sensor has a 585



Figure 9. Experimental results of sweat monitoring. The response to volume and NaCl concentrations in sweat tested with the impedance sensor (A) and the TPS sensor (B). The impedance measurement is affected by both the NaCl concentration and the moisture content, whereas the thermal measurement is only influenced by the moisture content.

see sensitivity of 0.158 $\Omega/^{\circ}C$ and excellent linear transfer function 587 characteristics. As illustrated in Figure 10B, for the TPS 588 measurement, $H_{(t)}$ represents the duration for which a 589 transient heat pulse is applied and $C_{(t)}$ is the cooling time 590 where $C_{(t)} \gg H_{(t)}$. The initial resistance (R_0) of the heater at 591 the time of applying the heat pulse is proportional to the 592 ambient temperature (T_0) . Therefore, the difference in the 593 initial resistances of the heater measured at heat pulses 594 indicates sensor medium-temperature variations.⁵⁹ Thus, the TPS measurement mode can be beneficially exploited to 595 596 estimate an additional functionality, namely, the monitoring of 597 the body temperature, if the body is cohesive enough to the sensor. Body fluid volume, electrical properties, and temper-599 ature measurements together pave the way for a synergic effect 600 applicable for a myriad of wearable entreaties. Such synergetic 601 action of the textile sensor patch could address the possibility 602 of a smart bandage for wound monitoring. The wound exudate



Figure 10. Resistance reading of the sensor inside an oven with the help of a multimeter, where the box oven temperature is set to vary from 22 to 28 $^{\circ}$ C (A) and illustration of TPS measurements while ambient temperature varies over time (B).

detection could help with the wound dressing removal and 603 redressing, reducing the chances of infections. Infections, 604 however, need to be detected at the earliest and wound 605 exudate bacterial growth can be identified with impedance 606 spectra analysis.^{60,61} In conjunction with this, the temperature 607 measurements at the wound site could guide health 608 practitioners about the wound-healing progress.^{62–64} Similarly, 609 it is a highly germane model for sportswear to have a 610 temperature indicator along with the sweat rate and sweat 611 analysis.^{2,59}

Washability of the Sensor. The washability of the 613 wearable sensor is a vital aspect that makes the sensor system 614 applicable in real-life scenarios. The sensor design architecture 61s and material selection are crucial for the withstandability of the 616 sensor during washing cycles. The impedance sensor is 617 designed in such a way that the interdigitated electrodes are 618 sandwiched in between the sensing layer medium. The sensing 619 layer is based on polyurethane, which is compatible with 620

639

621 washing. The TPS sensor buildup is made in a way that the top 622 face of the heater structure is spray coated with Tubiguard, to 623 form a nonabsorbent layer.

Additionally, a few TPS sensors are also fabricated with both 625 sides of the sensor having Tubiguard layers. The washing tests 626 are conducted based on the ISO 6330 standards, where each 627 washing cycle is carried out at 40 $^{\circ}$ C and has a duration of 70 628 min each. Every sensor undergoes five such washing cycles. 629 Washability results in Figure 11A show almost identical



Figure 11. Impedance sensor monitoring the drying of a paper tissue before and after five washing cycles (A) and resistance change of the heater structures compared before and after washing cycles (B).

630 responses for the impedance measurements of the sensor 631 before and after performing the washing tests. However, there 632 is a slight offset that is supposed to arise from minor damages 633 that occurred during washing, which can be addressed by 634 increasing the thickness of the dielectric layer and adding cross 635 linking-agents to this polymeric ink. Heaters having encapsu-636 lation layers on both sides show better performance after 637 washing tests than those with the protective coating only on 638 one side, as shown in Figure 11B.

CONCLUSIONS AND FUTURE STUDIES

In this article, an approach for a wearable biofluid monitoring 640 system is described, combining two concurrent principles for 641 the first time to the best of our knowledge. The article 642 emphasizes the development of wearable techniques to carry 643 out precise quantification of peripheral body biofluid and its 644 biomarkers in a nonintrusive manner. Wearable sensors based 645 on electrical parameters often suffer from cross-sensitivity of 646 different biomarkers. This work applies two different measure- 647 ment techniques: one based on electrical impedance and the 648 other, the TPS principle, on thermal properties, combined for a 649 comprehensive independent sensing node. The sensor system 650 demonstrates consistent and repeatable moisture volume 651 measurements and is also deployed to monitor the drying of 652 wet textiles. The sensor system demonstrates the smallest 653 measurable volume of 0.5 μ L sweat, which assures the method 654 to be promising for health monitoring. Along with volume, the 655 sweat's compositional variations also induced changes in the 656 impedance readings. The TPS sensor measures the volume of 657 sweat accurately without any interference from the composi- 658 tional variations. The sweat solution volume was varied with 659 simultaneous variation in the NaCl concentration over a range 660 of 0.005-0.1 M, and the proposed sensor system accurately 661 tracked and measured both without any disruption. The sensor 662 system is conceived such that it can work reciprocally toward 663 better wearable applications. 664

The printed sensors on textiles incorporating this dual- 665 sensing technique exclusively address some of the critical 666 challenges in terms of the selectivity of biomarkers, moisture 667 content measurement, wearability, and economic viability. 668 Additionally, the TPS measurement system possesses the 669 capacity to track the temperature of the medium and the textile 670 sensor with a sensitivity of 0.158 $\Omega/^{\circ}C$. Besides the 671 possibilities toward biofluid volume measurement, biomarker 672 detection, and passive body temperature measurement, this 673 work also accounts for promising results toward washing 674 compatibility. The selection of the right materials and smart 675 device engineering ensure the sensor's withstandability during 676 washing and provide a promising feature. As most of the 677 research articles focusing on wearable sensors utilize flexible 678 foil as the substrate, this work ensures the direct fabrication of 679 sensors on textiles. Screen printing and ultrasonic spray coating 680 were applied to deposit different material formulations, and 681 this enables the mass production of the conformable sensors at 682 a reduced cost.

The developed dual sensor system can be worthwhile in real- 684 life applications such as smart wound dressings, wearable 685 health bands, and sports wearables. To address such end- 686 application requirements, the sensor system needs to be ready 687 for on-body measurements. In future studies, the authors 688 would address body measurements where the sensors also 689 need to be integrated together with a wearable readout system. 690 Depending on the end-application, the sensor buildup 691 architecture may change from side-by-side placement to a 692 stack type. Additionally, for the TPS sensor, transient heating 693 needs to be optimized in such a way that it measures only the 694 moisture in textiles and does not get influenced by that from 695 the human body. The sensor system needs considerations in its 696 design and material selection in such a way that the errors that 697 could be induced by body movements and other variabilities 698 need to be kept as minimal as possible. Similarly, the 699 experimental results also show the possibilities of the sensor 700

701 system to handle volumes even less than 0.5 μ L. Future studies 702 must also check the viability to test even lower volumes by 703 improving the printing line-width resolution that could make 704 the heater coil and the electrode fingers even closer and 705 thinner. This helps to get better sensitivity and repeatability of 706 the sensor measurements. In addition, it would be beneficial if 707 the sensor interface layer to fluids has a more efficient wetting 708 behavior, which could be made possible with a thin hydrophilic 709 coating or by integrating a textile layer on the sensor surface. 710 This would add a more interesting perspective for more 711 accurate and repeatable measurements. To conclude, this work 712 demonstrated an innovative approach for the accurate analysis 713 of human body fluids in wearable textiles and can progressively 714 evolve as a step forward in smart wound monitoring and sweat 715 analysis applications.

716 **ASSOCIATED CONTENT**

717 S Supporting Information

718 The Supporting Information is available free of charge at 719 https://pubs.acs.org/doi/10.1021/acssensors.0c02037.

- 720 Description and explanation of TPS sensor's transient
- heating time selection; transient heating curve; and TPS
- 722 measurements of sweat with different ionic concen-
- 723 trations tested on thin tissue (PDF)

724 **AUTHOR INFORMATION**

725 Corresponding Author

- 726 Wim Deferme Hasselt University, Institute for Materials
- 727 Research (IMO-IMOMEC) 1, 3590 Diepenbeek, Belgium;
- 728 IMEC, Division IMOMEC, B-3590 Diepenbeek, Belgium;
- ⁷²⁹ orcid.org/0000-0002-8982-959X;
- 730 Email: wim.deferme@uhasselt.be

731 Authors

- 732 Manoj Jose Hasselt University, Institute for Materials
- Research (IMO-IMOMEC) 1, 3590 Diepenbeek, Belgium;
 IMEC, Division IMOMEC, B-3590 Diepenbeek, Belgium;
- 735 orcid.org/0000-0003-1635-1131
- 736 Gilles Oudebrouckx Hasselt University, Institute for
- 737 Materials Research (IMO-IMOMEC) 1, 3590 Diepenbeek,
- Belgium; IMEC, Division IMOMEC, B-3590 Diepenbeek,
 Belgium; ⊙ orcid.org/0000-0001-6278-3547
- Seppe Bormans Hasselt University, Institute for Materials
 Research (IMO-IMOMEC) 1, 3590 Diepenbeek, Belgium;
- 742 IMEC, Division IMOMEC, B-3590 Diepenbeek, Belgium
- Paula Veske Centre for Microsystems Technology (CMST),
 IMEC and Ghent University, 9052 Gent, Belgium
- 745 Ronald Thoelen Hasselt University, Institute for Materials
- 746 Research (IMO-IMOMEC) 1, 3590 Diepenbeek, Belgium;
- 747 IMEC, Division IMOMEC, B-3590 Diepenbeek, Belgium
- 748 Complete contact information is available at:

749 https://pubs.acs.org/10.1021/acssensors.0c02037

750 Notes

751 The authors declare no competing financial interest.

752 **REFERENCES**

(1) Yao, S.; Myers, A.; Malhotra, A.; Lin, F.; Bozkurt, A.; Muth, J. F.;
754 Zhu, Y. A Wearable Hydration Sensor with Conformal Nanowire
755 Electrodes. *Adv. Healthcare Mater.* 2017, *6*, No. 1601159.

756 (2) Gao, W.; Emaminejad, S.; Nyein, H. Y. Y.; Challa, S.; Chen, K.; 757 Peck, A.; Fahad, H. M.; Ota, H.; Shiraki, H.; Kiriya, D.; Lien, D.-H.; 758 Brooks, G. A.; Davis, R. W.; Javey, A. Fully integrated wearable sensor arrays for multiplexed in situ perspiration analysis. *Nature* **2016**, *529*, *759* 509–514. 760

(3) Yoon, S.; Sim, J.; Cho, Y.-H. A Flexible and Wearable Human 761 Stress Monitoring Patch. *Sci. Rep.* **2016**, *6*, No. 23468. 762

(4) Morello, R.; Capua, C. D.; Lugarà, M.; Lay-Ekuakille, A.; Griffo, 763 G.; Vergallo, P. In *Design of a Wearable Sweat Sensor for Diagnosing* 764 *Cystic Fibrosis in Children*, 2013 IEEE International Symposium on 765 Medical Measurements and Applications (MeMeA), 4–5 May 2013; 766 pp 268–273. 767

(5) McColl, D.; Cartlidge, B.; Connolly, P. Real-time monitoring of 768 moisture levels in wound dressings in vitro: An experimental study. 769 *Int. J. Surg.* **2007**, *5*, 316–322. 770

(6) Ochoa, M.; Rahimi, R.; Ziaie, B. Flexible Sensors for Chronic 771 Wound Management. *IEEE Rev. Biomed. Eng.* **2014**, *7*, 73–86. 772

(7) Brown, M. S.; Ashley, B.; Koh, A. Wearable Technology for 773 Chronic Wound Monitoring: Current Dressings, Advancements, and 774 Future Prospects. *Front. Bioeng. Biotechnol.* **2018**, *6*, No. 47. 775

(8) Sharp, D. Printed composite electrodes for in-situ wound pH 776 monitoring. *Biosens. Bioelectron.* **2013**, 50, 399–405. 777

(9) Qin, M.; Guo, H.; Dai, Z.; Yan, X.; Ning, X. Advances in flexible 778 and wearable pH sensors for wound healing monitoring. *J. Semicond.* 779 **2019**, 40, No. 111607. 780

(10) Farrow, M. J.; Hunter, I. S.; Connolly, P. Developing a real time 781 sensing system to monitor bacteria in wound dressings. *Biosensors* 782 **2012**, *2*, 171–188. 783

(11) Ciani, I.; Schulze, H.; Corrigan, D. K.; Henihan, G.; Giraud, G.; 784 Terry, J. G.; Walton, A. J.; Pethig, R.; Ghazal, P.; Crain, J.; Campbell, 785 C. J.; Bachmann, T. T.; Mount, A. R. Development of immunosensors 786 for direct detection of three wound infection biomarkers at point of 787 care using electrochemical impedance spectroscopy. *Biosens. Bio-* 788 *electron.* **2012**, *31*, 413–418. 789

(12) Milne, S. D.; Seoudi, I.; Al Hamad, H.; Talal, T. K.; Anoop, A. 790 A.; Allahverdi, N.; Zakaria, Z.; Menzies, R.; Connolly, P. A wearable 791 wound moisture sensor as an indicator for wound dressing change: an 792 observational study of wound moisture and status. *Int. Wound J.* **2016**, 793 *13*, 1309–1314. 794

(13) Brueck, A.; Iftekhar, T.; Stannard, A. B.; Yelamarthi, K.; Kaya, 795 T. A Real-Time Wireless Sweat Rate Measurement System for 796 Physical Activity Monitoring. *Sensors* **2018**, *18*, No. 533. 797

(14) Chung, M.; Fortunato, G.; Radacsi, N. Wearable flexible sweat 798 sensors for healthcare monitoring: a review. J. R. Soc., Interface **2019**, 799 16, No. 20190217. 800

(15) Choi, D.-H.; Kitchen, G. B.; Jennings, M. T.; Cutting, G. R.; 801 Searson, P. C. Out-of-clinic measurement of sweat chloride using a 802 wearable sensor during low-intensity exercise. *npj Digital Med.* **2020**, 803 3, No. 49. 804

(16) He, W.; Wang, C.; Wang, H.; Jian, M.; Lu, W.; Liang, X.; 805 Zhang, X.; Yang, F.; Zhang, Y. Integrated textile sensor patch for realtime and multiplex sweat analysis. *Sci. Adv.* **2019**, *5*, No. eaax0649. 807

(17) Ward, A. C.; Hannah, A. J.; Kendrick, S. L.; Tucker, N. P.; 808
MacGregor, G.; Connolly, P. Identification and characterisation of 809 *Staphylococcus aureus* on low cost screen printed carbon electrodes 810
using impedance spectroscopy. *Biosens. Bioelectron.* 2018, 110, 65–70. 811
(18) Manjakkal, L.; Dervin, S.; Dahiya, R. Flexible potentiometric 812

pH sensors for wearable systems. RSC Adv. 2020, 10, 8594–8617. 813

(19) Miller, C.; Stiglich, M.; Livingstone, M.; Gilmore, J. 814 Impedance-Based Biosensing of Pseudomonas putida via Solution 815 Blow Spun PLA: MWCNT Composite Nanofibers. *Micromachines* 816 **2019**, *10*, No. 876. 817

(20) Seshadri, D. R.; Li, R. T.; Voos, J. E.; Rowbottom, J. R.; Alfes, 818 C. M.; Zorman, C. A.; Drummond, C. K. Wearable sensors for 819 monitoring the physiological and biochemical profile of the athlete. 820 *npj Digit. Med.* **2019**, *2*, No. 72. 821

(21) Heikenfeld, J.; Jajack, A.; Rogers, J.; Gutruf, P.; Tian, L.; Pan, 822 T.; Li, R.; Khine, M.; Kim, J.; Wang, J.; Kim, J. Wearable sensors: 823 modalities, challenges, and prospects. *Lab Chip* **2018**, *18*, 217–248. 824 (22) Farooqui, M. F.; Shamim, A. Low Cost Inkjet Printed Smart 825 Bandage for Wireless Monitoring of Chronic Wounds. *Sci. Rep.* **2016**, 826 6, No. 28949. 827 828 (23) Kubiak, P.; Lesnikowski, J.; Gniotek, K. Textile Sweat Sensor
829 for Underwear Convenience Measurement. *Fibres Text. East. Eur.*830 2016, 24, 151–155.

(24) Amano, T.; Gerrett, N.; Inoue, Y.; Nishiyasu, T.; Havenith, G.;
Kondo, N. Determination of the maximum rate of eccrine sweat
glands' ion reabsorption using the galvanic skin conductance to local
sweat rate relationship. *Eur. J. Appl. Physiol.* 2016, *116*, 281–290.

835 (25) Sim, J. K.; Yoon, S.; Cho, Y.-H. Wearable Sweat Rate Sensors
836 for Human Thermal Comfort Monitoring. *Sci. Rep.* 2018, *8*, No. 1181.
837 (26) Salvo, P.; Francesco, F. D.; Costanzo, D.; Ferrari, C.; Trivella,
838 M. G.; Rossi, D. D. A Wearable Sensor for Measuring Sweat Rate.
839 IEEE Sens. J. 2010, 10, 1557–1558.

840 (27) Jain, V.; Ochoa, M.; Jiang, H.; Rahimi, R.; Ziaie, B. A mass-841 customizable dermal patch with discrete colorimetric indicators for 842 personalized sweat rate quantification. *Microsyst. Nanoeng.* **2019**, *5*, 843 No. 29.

844 (28) Reeder, J. T.; Choi, J.; Xue, Y.; Gutruf, P.; Hanson, J.; Liu, M.; 845 Ray, T.; Bandodkar, A. J.; Avila, R.; Xia, W.; Krishnan, S.; Xu, S.; 846 Barnes, K.; Pahnke, M.; Ghaffari, R.; Huang, Y.; Rogers, J. A. 847 Waterproof, electronics-enabled, epidermal microfluidic devices for 848 sweat collection, biomarker analysis, and thermography in aquatic 849 settings. *Sci. Adv.* **2019**, *5*, No. eaau6356.

850 (29) Hierlemann, A.; Brand, O.; Hagleitner, C.; Baltes, H.
851 Microfabrication techniques for chemical/biosensors. *Proc. IEEE*852 2003, 91, 839–863.

(30) Betancourt, T.; Brannon-Peppas, L. Micro- and nanofabrication
methods in nanotechnological medical and pharmaceutical devices. *Int. J. Nanomed.* 2006, 1, 483–495.

856 (31) Park, J.; Lee, Y.; Hong, J.; Lee, Y.; Ha, M.; Jung, Y.; Lim, H.; 857 Kim, S. Y.; Ko, H. Tactile-Direction-Sensitive and Stretchable 858 Electronic Skins Based on Human-Skin-Inspired Interlocked Micro-859 structures. *ACS Nano* **2014**, *8*, 12020–12029.

(32) Zhang, F.; Zang, Y.; Huang, D.; Di, C.-a.; Zhu, D. Flexible and
self-powered temperature-pressure dual-parameter sensors using
microstructure-frame-supported organic thermoelectric materials. *Nat. Commun.* 2015, *6*, No. 8356.

(33) Chen, Y.; Lu, B.; Chen, Y.; Feng, X. Breathable and Stretchable
Temperature Sensors Inspired by Skin. *Sci. Rep.* 2015, *5*, No. 11505.
(34) Sreenilayam, S. P.; Ahad, I. U.; Nicolosi, V.; Acinas Garzon, V.;
Brabazon, D. Advanced materials of printed wearables for
physiological parameter monitoring. *Mater. Today* 2020, *32*, 147–
177.

(35) Pal, A.; Goswami, D.; Cuellar, H. E.; Castro, B.; Kuang, S.;
Martinez, R. V. Early detection and monitoring of chronic wounds
using low-cost, omniphobic paper-based smart bandages. *Biosens. Bioelectron.* 2018, *117*, 696–705.

874 (36) Krykpayev, B.; Farooqui, M. F.; Bilal, R. M.; Vaseem, M.; 875 Shamim, A. A wearable tracking device inkjet-printed on textile. 876 *Microelectron. J.* **2017**, *65*, 40–48.

877 (37) Hong, S.; Lee, K.; Ha, U.; Kim, H.; Lee, Y.; Kim, Y.; Yoo, H. A 878 4.9 m Ω -Sensitivity Mobile Electrical Impedance Tomography IC for 879 Early Breast-Cancer Detection System. *IEEE J. Solid-State Circuits* 880 **2015**, *50*, 245–257.

881 (38) Ganguly, A.; Prasad, S. Passively Addressable Ultra-Low 882 Volume Sweat Chloride Sensor. *Sensors* **2019**, *19*, No. 4590.

(39) Gustafsson, S. E. Transient plane source techniques for thermal
conductivity and thermal diffusivity measurements of solid materials. *Rev. Sci. Instrum.* 1991, *62*, 797–804.

(40) Schönfisch, D.; Göddel, M.; Blinn, J.; Heyde, C.; Schlarb, H.;
Deferme, W.; Picard, A. New Type of Thermal Moisture Sensor for
in-Textile Measurements. *Phys. Status Solidi* (a) 2019, 216,
No. 1800765.

(41) Verboven, I.; Stryckers, J.; Mecnika, V.; Vandevenne, G.; Jose,
M.; Deferme, W. Printing Smart Designs of Light Emitting Devices
with Maintained Textile Properties. *Materials* 2018, *11*, No. 290.

893 (42) Samanta, A.; Bordes, R. Conductive textiles prepared by spray 894 coating of water-based graphene dispersions. *RSC Adv.* **2020**, *10*, 895 2396–2403. (43) Khattab, T. A.; Rehan, M.; Hamdy, Y.; Shaheen, T. I. Facile 896 Development of Photoluminescent Textile Fabric via Spray Coating 897 of Eu(II)-Doped Strontium Aluminate. *Ind. Eng. Chem. Res.* **2018**, *57*, 898 11483–11492. 899

(44) Wang, J.; Lin, Q.; Zhou, R.; Xu, B. Humidity sensors based on 900 composite material of nano-BaTiO3 and polymer RMX. *Sens.* 901 *Actuators, B* **2002**, *81*, 248–253. 902

(45) Farahani, H.; Wagiran, R.; Hamidon, M. N. Humidity Sensors 903 Principle, Mechanism, and Fabrication Technologies: A Comprehen-904 sive Review. *Sensors* 2014, 14, 7881–7939. 905

(46) Grammatikos, S. A.; Ball, R. J.; Evernden, M.; Jones, R. G. 906 Impedance spectroscopy as a tool for moisture uptake monitoring in 907 construction composites during service. *Composites, Part A* **2018**, *105*, 908 108–117. 909

(47) Li, X.; Wang, X.; Zhao, Q.; Zhang, Y.; Zhou, Q. In Situ 910 Representation of Soil/Sediment Conductivity Using Electrochemical 911 Impedance Spectroscopy. *Sensors* **2016**, *16*, No. 625. 912

(48) Caballero, A. C.; Villegas, M.; Fernández, J. F.; Viviani, M.; 913 Buscaglia, M. T.; Leoni, M. Effect of humidity on the electrical 914 response of porous BaTiO3 ceramics. *J. Mater. Sci. Lett.* **1999**, *18*, 915 1297–1299. 916

(49) C-THERM. Modified Transient Plane Source (MTPS), Theory 917 of Operation. C-THERM, 2020. 918

(50) Bae, Y.-M.; Lee, Y.-H.; Kim, H.-S.; Lee, D.-J.; Kim, S. Y.; Kim, 919 H.-D. Polyimide-polyurethane/urea block copolymers for highly 920 sensitive humidity sensor with low hysteresis. *J. Appl. Polym. Sci.* 921 **2017**, *134*, No. 11225. 922

(51) Bosch, P.; Fernández, A.; Salvador, E. F.; Corrales, T.; Catalina, 923 F.; Peinado, C. Polyurethane-acrylate based films as humidity sensors. 924 *Polymer* **2005**, 46, 12200–12209. 925

(52) Currano, L. J.; Sage, F. C.; Hagedon, M.; Hamilton, L.; 926 Patrone, J.; Gerasopoulos, K. Wearable Sensor System for Detection 927 of Lactate in Sweat. *Sci. Rep.* **2018**, *8*, No. 15890. 928

(53) Farooqui, M. F.; Shamim, A. Low Cost Inkjet Printed Smart 929 Bandage for Wireless Monitoring of Chronic Wounds. *Sci. Rep.* **2016**, 930 *6*, No. 28949. 931

(54) Manjakkal, L.; Dang, W.; Yogeswaran, N.; Dahiya, R. Textile- 932 Based Potentiometric Electrochemical pH Sensor for Wearable 933 Applications. *Biosensors* **2019**, *9*, 14. 934

(55) Callewaert, C.; Buysschaert, B.; Vossen, E.; Fievez, V.; Van de 935 Wiele, T.; Boon, N. Artificial sweat composition to grow and sustain a 936 mixed human axillary microbiome. *J. Microbiol. Methods* **2014**, *103*, 937 6–8. 938

(56) Midander, K.; Julander, A.; Kettelarij, J.; Lidén, C. Testing in 939 artificial sweat – Is less more? Comparison of metal release in two 940 different artificial sweat solutions. *Regul. Toxicol. Pharmacol.* **2016**, *81*, 941 381–386. 942

(57) Ali, S.; Hassan, A.; Bae, J.; Lee, C. H.; Kim, J. All-Printed 943 Differential Temperature Sensor for the Compensation of Bending 944 Effects. *Langmuir* **2016**, *32*, 11432–11439. 945

(58) Molina-Lopez, F.; Vásquez Quintero, A.; Mattana, G.; Briand, 946 D.; Rooij, N. Large-area compatible fabrication and encapsulation of 947 inkjet-printed humidity sensors on flexible foils with integrated 948 thermal compensation. *J. Micromech. Microeng.* **2013**, 23, No. 025012. 949

(59) Nakata, S.; Arie, T.; Akita, S.; Takei, K. Wearable, Flexible, and 950 Multifunctional Healthcare Device with an ISFET Chemical Sensor 951 for Simultaneous Sweat pH and Skin Temperature Monitoring. ACS 952 Sens. 2017, 2, 443–448. 953

(60) Schröter, A.; Rösen-Wolff, A.; Gerlach, G. In *Impedance* 954 *Measurement of Wound Infection Status,* Proceedings SENSOR 2013. 955 AMA Service GmbH, Ed.; 2013; pp 628–632. 956

(61) Furst, A. L.; Francis, M. B. Impedance-Based Detection of 957 Bacteria. *Chem. Rev.* **2019**, *119*, 700–726. 958

(62) Derakhshandeh, H.; Kashaf, S. S.; Aghabaglou, F.; Ghanavati, I. 959 O.; Tamayol, A. Smart Bandages: The Future of Wound Care. *Trends* 960 *Biotechnol.* **2018**, *36*, 1259–1274. 961

(63) Mostafalu, P.; Tamayol, A.; Rahimi, R.; Ochoa, M.; Khalilpour, 962 A.; Kiaee, G.; Yazdi, I. K.; Bagherifard, S.; Dokmeci, M. R.; Ziaie, B.; 963 964 Sonkusale, S. R.; Khademhosseini, A. Smart Bandage for Monitoring
965 and Treatment of Chronic Wounds. *Small* 2018, *14*, No. 1703509.
966 (64) Schröter, A.; Walther, A.; Fritzsche, K.; Kothe, J.; Rösen-Wolff,
967 A.; Gerlach, G. Infection Monitoring in Wounds. *Proc. Chem.* 2012, *6*,
968 175–183.