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Biocompatible and Flexible Piezoelectric Thin Film Materials and Devices for Skin Compliant Transducers*

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Abstract — Skin electronics for health monitoring require materials and devices with unique properties such as biocompatibility, flexibility, and high sensitivity. In this work, we describe our recent work in exploiting piezoelectric thin films, based on either aluminum nitride (AlN) or chitosan biopolymers, for skin-compliant transducers to record biomechanical signals on skin.

I. INTRODUCTION

Nowadays, the monitoring of the individual's health conditions rests mainly on the use of traditional methods and technologies that require rather unwieldy equipment and, at the same time, very complicated operating procedures. These methods use routine physical checkups and post-morbid consultations. On one side, they allow for assessing most health problems. On the other side, they have the drawback of being often invasive, causing discomfort and pain to patients, and for these reasons, unsuitable for long-term real-time monitoring of the body state [1], [2]. On the contrary, continuous health monitoring allows for identifying diseases in the early stage, consequently minimizing risks and improving prognostics [3].

Wearable technologies and smart healthcare systems pave the way to different innovative approaches for detecting and diagnosing diseases early, taking advantage of real-time monitoring, non-invasive reliability, simplicity, and rapidity. Wearable sensors support doctors in decisions, for tracking and completing the rehabilitation training of patients even at home, or they can help athletes in training, optimizing practice and exercises.

In this respect, skin-compliant transducers are a class of skin-compliant devices that make possible the detection of pathophysiological biomechanical signals and symptoms of disorders and diseases in an unobtrusive way. The skin is an efficient and suitable interface between tissues and the external environment, allowing access to different physiological signals [4]–[6]. Skin-like single or multi-function wearable sensors can be designed to monitor respiration rate, body temperature,

blood pressure, pulse, and frequency during normal activity, rehabilitation, or exercise. As auxiliary devices, they can assist users in carrying out essential daily activities. Moreover, being often simple in shape, small, and light in weight, they do not affect normal activities [7], [8].

In general, wearable sensors can be categorized into devices for detecting movements, e.g., inertial sensors and devices for sensing physiological parameters. Wearable devices can be classified according to the adopted transduction mechanisms. Among transduction methods, piezoelectric transducers are the most effective in recording biomechanical cues, such as movements, vibrations, and sounds, directly on the skin, exploiting the direct piezoelectric effect. Only in recent years compliant and flexible piezoelectric thin film materials have enabled the demonstration of piezoelectric bio-interfaces, such as transducers positioned on skin or implanted in the body, for stimulating, sensing, and monitoring biomechanical signals [9], [10] and for harvesting energy for autonomous microsystems for medical devices [11]. The employment of materials containing lead often prevents translation in the body of piezoelectric technology, e.g., lead zirconate titanate (PZT) and lead magnesium niobate-lead titanate (PMN-PT). Lead-based materials are employed in ultrasound probes for ultrasonography thanks to their excellent piezoelectric properties. However, they are toxic, and even in small traces, they cause severe chronic poisoning, as stated by organizations such as FDA, CDC, and RoHS. This makes commonly used lead-based piezoelectrics not applicable to clinical settings and the human body because materials and devices are not compliant and safe and are not designed for healthcare applications on the body.

The most recent generation of flexible piezoelectric thin film materials for biomechanical transducers includes inorganic piezoelectrics on polymeric substrates [12], synthetic polymers, and biopolymers [13]. The current challenge relies on producing efficient, flexible piezoelectric materials to be safely applied to the human body and processed in functional

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devices for in-vivo experiments. In this work, material and device technologies of two classes of biocompatible piezoelectrics will be discussed: (i) aluminum nitride (AlN) deposited on polymeric substrates and (ii) chitosan-based piezoelectric biopolymers. Both material systems combine good mechanical and piezoelectric properties, safety, and biocompatibility and do not need poling (a critical process to align individual dipole moments, allowing these materials to exhibit piezoelectric features). These two innovative materials lead to various advantages and disadvantages when compared. Piezoelectric AlN has a higher piezoelectric coefficient, making it very efficient for fabricating transducers to convert mechanical stress into electrical signals and vice-versa. It is a stable, durable material with high chemical resistance, mechanical strength, and a wide operating temperature range without significant performance degradation. Also, it exhibits high-frequency response, making it suitable for applications requiring rapid signal generation and detection, such as ultrasound transducers and sensors. Despite these merits, being a ceramic material, it is brittle and can be damaged, especially under high mechanical stress, limiting its use in specific applications requiring flexibility.

On the other hand, piezoelectric chitosan offers some additional advantages, such as biocompatibility and sustainability, making it suitable for use in biomedical applications, especially as drug delivery devices and biosensors. Chitosan films are also highly flexible and can conform to irregular shapes, bypassing the limits of rigid materials. Moreover, chitosan is relatively inexpensive, although it exhibits a lower piezoelectric coefficient than the AlN when cast with the same thickness. It also has limitations in terms of operating temperature range. The high temperature typically changes the chitosan films' morphological structure, making it unsuitable for high-temperature applications. Finally, chitosan may not have as high a frequency response as other piezoelectric materials, limiting its use in high-frequency applications. In summary, the choice between piezoelectric AlN and piezoelectric chitosan depends on the specific application and requirements.

II. WEARABLE AND SKIN COMPLIANT ALN PIEZO-TRANSDUCERS

AlN piezoelectric thin films have shown good mechanical and piezoelectric properties on silicon and polymeric substrates (Kapton®, polyimide, and PEN) [14]. Flexible and biocompatible piezoelectric aluminum nitride (AlN) thin films can be deposited by reactive magnetron sputtering on polymeric substrates (Kapton®, polyimide, and PEN) [15], doped by scandium [16], and applied to wearable sensors, acoustic resonators, and US transducers, and energy harvesting [17]–[20]. AlN is known as one of the most suitable materials for developing compliant and flexible piezoelectric sensors for applications in biomedicine.

A. AlN-Based Sensors for Swallowing Disorder Evaluation

Wearable sensors based on ultrathin, compliant, and flexible AlN piezoelectric patches, grown on soft Kapton substrate, were designed and fabricated for tracking the laryngeal movements following the skin deformation driven by

the pressure of the hyoid bone [21]. The sensor is fabricated exploiting standard photolithography and microfabrication techniques (RIE etching and sputtering deposition), and it is composed of a thin film of AlN embedded between two molybdenum electrodes. Kapton foil (25 μm thick) was chosen as a structural substrate by virtue of its mechanical properties.

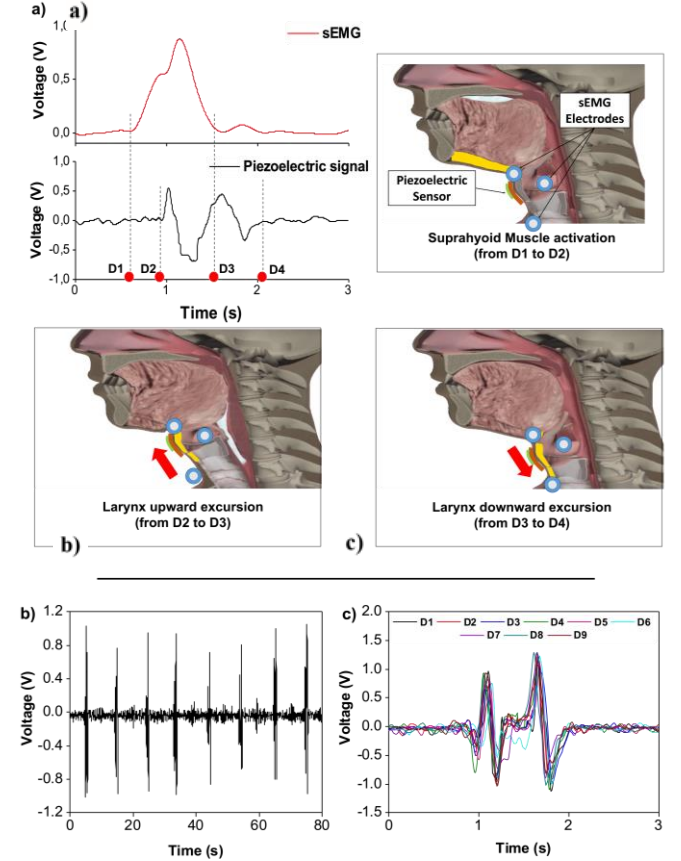


Fig. 1. (a) Comparison of the sEMG and the signal from the piezoelectric sensor, showing the correlation between the acquired signals and the swallowing events; (b) signal coming from the piezoelectric sensor; (c) segmentation and overlapping of all the acquired signals [22].

The piezoelectric sensor was used to generate a signal as an effect of rapid variations of dynamic pressure induced by the movement of the larynx, combined with electrophysiological and mechanical analysis. In this way, temporal events which occur during the deglutition act can be univocally identified. Voluntary subjects were tested, recording up to nine consecutive swallows when the sensor was attached directly to the neck in compliance with the laryngeal prominence (see Fig. 1(a)). Surface electromyography signals were used as triggers of the suprahyoid muscle activity and the thyroid cartilage movement. Signals from the sensor collected during the experiments revealed the repeating of two phases associated with the rising and descending motions of the thyroid cartilage, as shown in Fig. 1(a). Since the sensor is able to follow the larynx movement, the frequency of spontaneous swallowing f_d (measured in swallows per minute) and the time t_d to complete a swallow (it indicates the coordination of dedicated contraction muscles) can be identified (Fig. 1 (b) and (c)). For all the subjects under testing, the mean f_d values were between 1.5 ± 0.2 and 4.1 ± 0.6 swallows/min, whereas t_d was in a range from 0.9 ± 0.1 to 1.7 ± 0.5 s, in agreement with data recorded

on normal subjects. From these preliminary results, the doctor may objectively assess significant parameters, such as the length of the swallowing act and the frequency of spontaneous saliva deglutition, allowing the early detection of pathological diseases.

B. Smart Patch for Monitoring Heart and Blood Flow

By virtue of its high flexibility, the AIN-based sensor was also used to develop a lightweight and flexible smart microsystem for the continuous monitoring and examination of the blood flow hemodynamics in an artificial vessel [23] and the cardiovascular system status by direct estimation of the pulse wave velocity in real-time [24].

For this purpose, the sensor was integrated on the extraluminal surface of a vascular prosthesis, where it follows the wall deformation (Fig. 2a). An *ad hoc* measurement setup, pumping a liquid flow into the vessel, was used to mimic the sequence of systolic, diastolic notch and diastolic peaks. The corresponding output signals were then acquired in different hemodynamics conditions, replicating: (i) the variation of the peak-to-peak R-R interval, useful to identify the duration of the whole heart cycle and the corresponding flow rate; (ii) the variation of the Augmentation Index (AI) that is related to the changing of the vessels stiffness; (iii) the variation of the time interval between two consecutive peaks that provides information on the transit time of pressure wave.

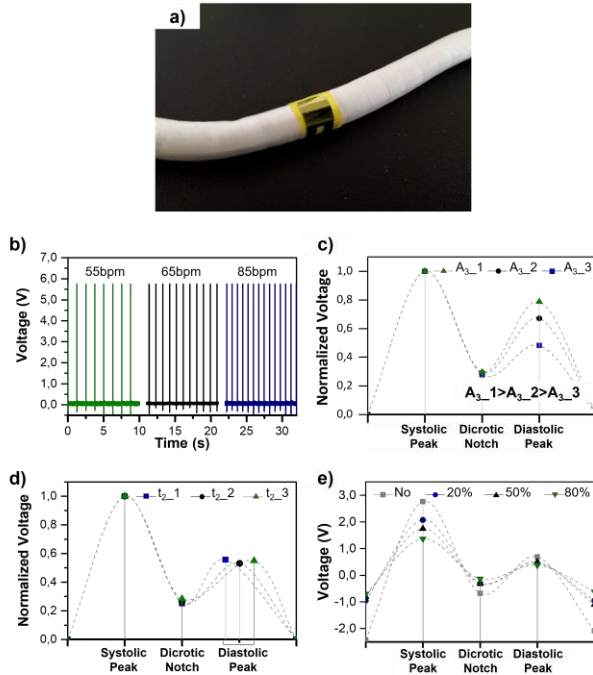


Fig. 2. (a) smart patch integrated on the vascular graft (b) output signals from the smart patch when different flow rates drive the graft; (c) Augmentation Index variation at the diastolic peak vs. the flow percentage of the pumped liquid inside the graft; (d) voltage response of the sensor vs. the variation of the time between the systolic and diastolic peak; (e) response of the sensor vs. different occlusion (in percentage) of the vascular graft [23].

The results demonstrated that, when coupled with the external surface of the vascular graft, the sensor can follow the motion of the graft wall, allowing it to extract information about the blood pulse wave in the vessel itself. Moreover,

pathological variation of the acquired signal is then correlated to the presence of stiffening or occlusion of the graft and vessel as a consequence. However, since the blood pulse wave propagates from the heart to the periphery through the artery, it is possible also to track the speed at which the blood flows by measuring the Pulse Wave Velocity (PWV). The sensor was placed on the carotid artery at the neck (Fig. 3a), and the output response (Fig. 3b) was acquired simultaneously with an ECG.

Therefore, the PWV was calculated as the ratio of this distance and the time delay between the R-peak of the ECG and the piezoelectric-generated signal (the highest peaks corresponding to the largest deformation of the vessel during the blood flow). From the test, a time delay of ~ 80 ms was successfully measured, corresponding to a PWV of 4.9 ms^{-1} (here, a distance of 40 cm between the sensor and the heart was considered), in good agreement with the values of the PWV reported in the scientific literature.

C. Application of AIN Sensors for Parkinson's Disease Evaluation

AIN wearable piezoelectric sensors are suitable for monitoring human body movements [25]. Mainly, the AIN piezoelectric sensors were effectively applied for monitoring and evaluating the conditions of patients affected by Parkinson's disease. A compact and discrete wearable device based on advanced and commercial sensors to monitor the course of the disease was developed.

Two thin-film piezoelectric sensors were integrated into a smart glove (Fig. 4): a half moon-shape ($25 \text{ mm} \times 15 \text{ mm}$) and a rectangular ($15 \text{ mm} \times 7 \text{ mm}$) sensors were installed into a flexible TPU (Thermoplastic Polyurethane) support achieved by 3D printing.

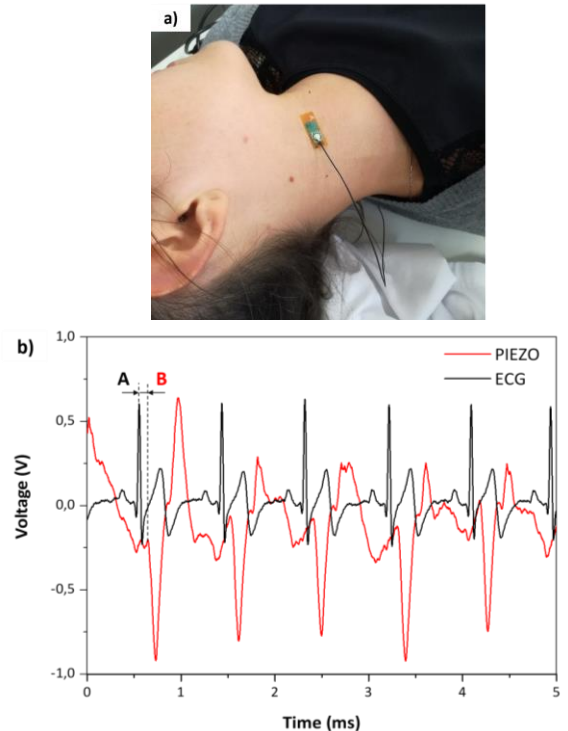


Fig. 3. (a) Smart sensor attached to the carotid artery (b) output voltage generated by the piezoelectric sensor and the ECG signal. A and B identify the peaks of the sensor and the ECG output; therefore, the PWV was calculated considering a distance of 40 cm between sensors [24].

The thin-film AlN sensors were interfaced with an electronic conditioning, acquisition, and processing section. The last unit comprises a dual-channel conditioning section, including charge amplifiers, level-shifting, and filtering modules, for making piezoelectric sensors' signals suitable for the following acquisition and processing. These tasks were entrusted to an nRF52840 System-on-Chip (Nordic Semiconductor), which acquires and processes data from the integrated piezoelectric and inertial sensors to extract features related to hand tremors, assigning scores to the patients according to the modalities defined by the UPDRS (Unified Parkinson Disease Rating Scale) (Fig. 5).

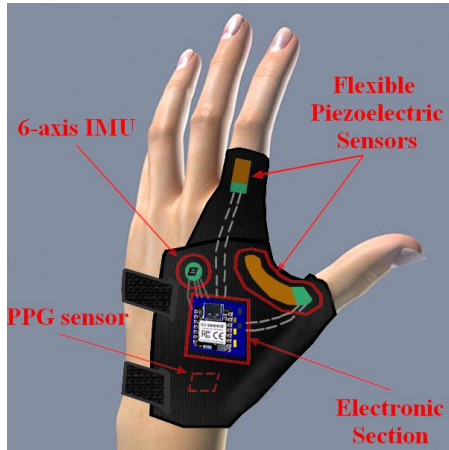


Fig. 4. Concept view of the smart glove for monitoring Parkinson's disease

The smart glove evaluates three standard tests involved by UPDRS to determine the patient's conditions, such as finger tapping, hand fist closure, and rest hand tremor. An embedded Machine Learning (ML) algorithm relying on an Artificial Neural Network (ANN) was deployed for each test. The Edge Impulse cloud platform was employed to gather datasets constituted by sample signals labeled according to the different scores of the UPDRS, train and test the ML models, and convert them to be executed on a microcontroller. A dataset consisting of 150 sample signals with a duration between 13 and 20 s was gathered during the execution of each test, reproducing the movements described by the UPDRS. Each dataset was split with an 80/20 train-test ratio. Spectral analysis was employed to extract features from FFT (Fast Fourier Transform) and PSD (Power Spectral Density) of the piezoelectric and inertial data, including statistical features (i.e., RMS-Root Mean Square, skewness, kurtosis, etc.) and spectral features (i.e., maximum value from FFT frames). Also, digital filtering stages were applied to remove undesired components from the input signals. Afterward, an ANN was trained and tested for each test; for each model, the ANN's architecture, training data size, obtained accuracy, and loss are summarized in Table I.

In addition, resting tremor is scored according to its amplitude and constancy over a 10 second time interval. This evaluation window was split into four 2.5 s sub-windows. The evaluation is performed on each of these windows, and the corresponding score is assigned. Then, the scores on the four windows are composed, extracting a single score considering the intensity and constancy of the tremor.

The test results demonstrated that the developed ML models obtain high accuracy in classifying piezoelectric and inertial data according to the UPDRS, reaching 95.12%, 98.39%, and 100% for finger tapping and hand fist closure exercises and rest tremor detection, respectively.

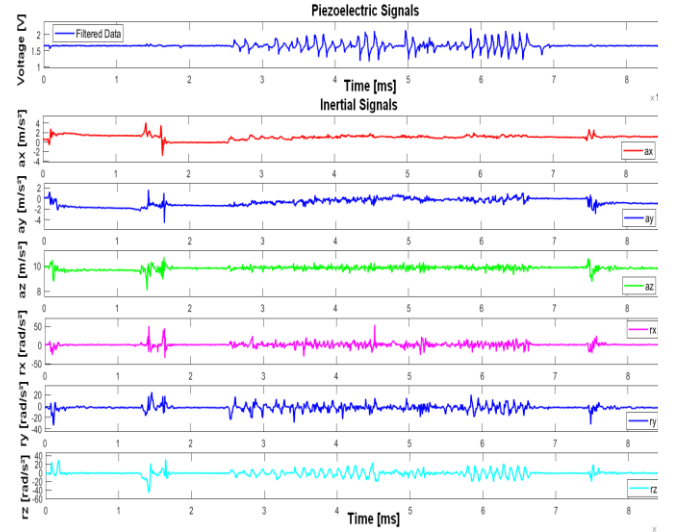


Fig. 5. Piezoelectric and inertial signals acquired by the smart glove.

TABLE I. Summarizing table with reported the architecture and performance of ANNs developed to classify each UPDRS test.

| UPDRS test | ANN architecture | Training Size [MB] | Accuracy [%] | Loss |
|-----------------------|---|--------------------|--------------|------|
| Finger-Tapping | 5 layers - Input Layer: 84 neurons; - 3 Dense Layers: 30, 20, 10 neurons; - Output layer: 6 classes (Score 0, Score 1, Score 2, Score 3, Score 4, Idle). | 1.76 | 95.12 | 0.11 |
| Hand Gesture | 5 layers - Input Layer: 112 neurons; - 3 Dense Layers: 60, 35, 20 neurons; - Output layer: 6 classes. (Score 0, Score 1, Score 2, Score 3, Score 4, Idle). | 2.40 | 98.39 | 0.05 |
| Resting Tremor | 5 layers - Input Layer: 105 neurons; - 3 Dense Layers: 70, 50, 30 neurons; - Output layer: 5 classes. (Score 0, Score 1, Score 2, Score 3, Score 4). | 1.54 | 100 | 0.03 |

III. SUSTAINABLE PIEZOELECTRIC CHITOSAN THIN FILM FOR HEALTHCARE APPLICATIONS

Piezoelectric biopolymers are innovative alternative materials for developing safe and compliant devices, being completely biocompatible and inherently biodegradable. Recently, highly piezoelectric and transparent chitosan, a biopolymer derivative of chitin, has been reported [26]. Chitosan thin films with piezoelectric coefficients ~ 3 times higher than previously reported (as high as 15.56 pC/N) were synthesized by a green and simple solvent casting method, enhancing the thin film's piezoelectric properties. Four types of wearable and implantable transducers were demonstrated,

enabling, for the first time, sustainable and flexible transducers based on thin chitosan film for health applications [26]–[28].

For this purpose, a 70 μm – thick chitosan film (hereafter CS-F), whose elementary crystal cell has an orthorhombic symmetry typical of the piezoelectric effect, was grown by drop casting method and characterized. The fabricated CS-F device is composed of an active rectangular area (2 cm x 1,5 cm) embedded between a bottom copper electrode onto a Kapton substrate and a top gold electrode (Fig. 6a). The bottom electrode was patterned exploiting ferric chloride wet etching, while the top electrode (80 nm – thick) was directly deposited by thermal evaporation.

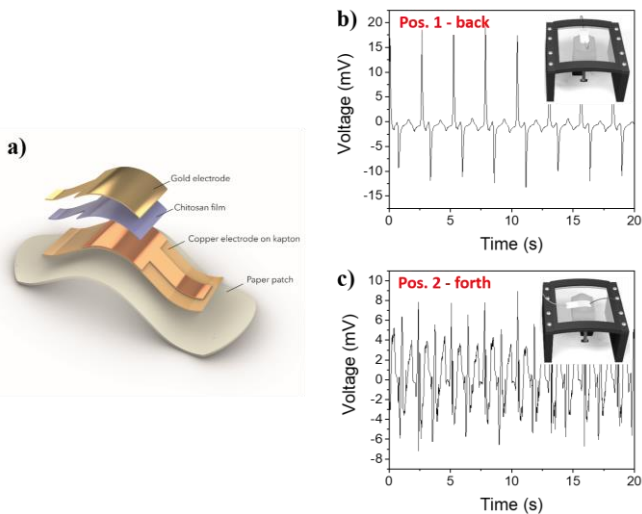


Fig. 6. a) Exploded schematic view of the chitosan-based patch; b) voltage output acquired when the sensor is in the Pos. 1 (back); c) output when the sensor is in the Pos. 2 (forth) [28].

The patch's ability to detect physiological strains was validated using an *ad hoc* setup, inducing the displacement of a thin PDMS (polydimethylsiloxane) membrane under the chitosan patch. The simulator applies cyclic pressure back and forth in one direction under the membrane while an oscilloscope collects the electric signal from the sensor. According to the position of the oscillator under the membrane, the sensor can generate different output signals (Fig. 6 b and c), whose amplitude varies from $V_{p-p} = 30$ mV (Pos. 1 – axis of the sensor in parallel with the movement's direction) to $V_{p-p} = 12$ mV (Pos. 2 – axis of the sensor perpendicular with the movement's direction). These preliminary results show that the sensor can distinguish the movements in different directions and is suitable for biocompatible wearable applications [27].

The enhancement of the piezoelectric features of the chitosan was achieved by applying a chemical treatment to tune the crystallographic alignment of the polymeric films [26]. After the chemical treatment, the sample with the highest piezoelectric coefficient of 15.5 pC/N⁻¹ was used to fabricate the wearable sensor (Fig. 7a). To test the properties of the highly piezoelectric chitosan film, miniaturized flags sandwiched between metal electrodes were fabricated (the final dimensions of the active area are 8 mm x 8 mm). The device was then calibrated, performing an impact test with different small plummets that differ in weight and fall from a trampoline at a height of 15 cm perpendicularly to the sensors. A metallic plate is positioned on top of the sensor to distribute the weight uniformly. Moreover, a wire whose one end is fixed on the

trampoline's edge and the second end to the metallic plate was used as a guide for the plummets to hit the exact center position of the sensor's active area under test. An oscilloscope records every impact, and each point on the corresponding plot was calculated as the average of at least three consecutive measurements. The device showed a linear output response in a range between 0.04 kPa and 0.5 kPa (as shown in Fig 7b).

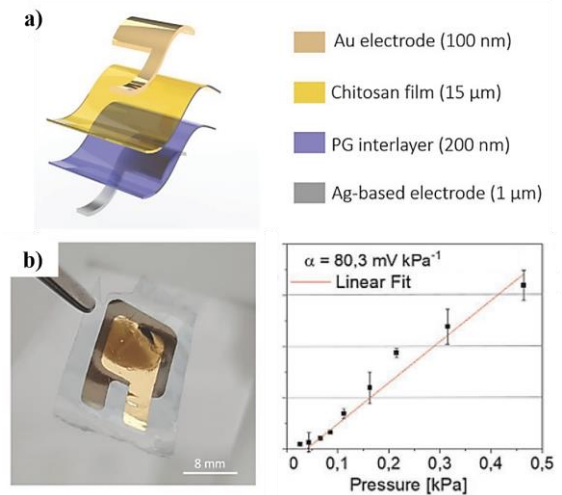


Fig. 7. a) Schematic of the fabricated sensor showing the composing materials; b) final sensor and the corresponding calibration curve achieved by impact test [26].

Moreover, the improved chitosan thin film was also used to fabricate piezoelectric micromachined ultrasonic transducers (pMUTs) to sense and detect ultrasound incoming waves in liquid (in particular water), exploiting the direct piezoelectric effect. Here, the waves are generated by a commercial immersion transducer driven by a pulse generator, whereas a hydrophone is used as a reference device.

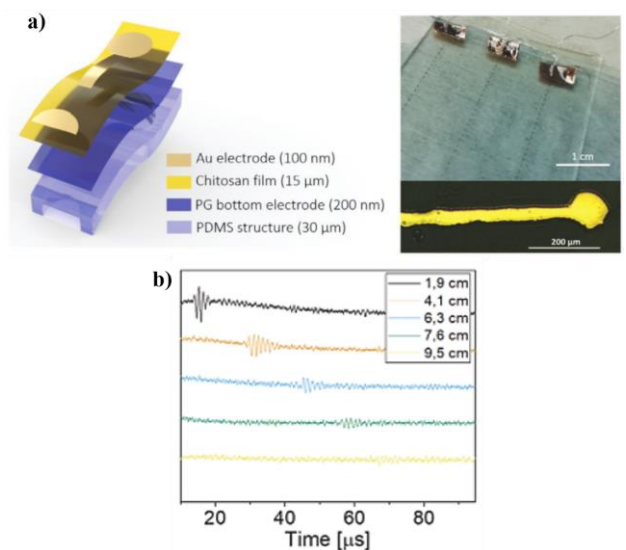


Fig. 8. a) Sketch of the chitosan pMUT device b) ultrasound waves acquired at 1 MHz when the pMUT is at a different position far from the commercial emitter [26].

An oscilloscope measured the output of the chitosan transducer and the reference hydrophone while the chitosan

device was moved back and forth from the source emitter using an X-Y translational stage. The preliminary results show that the chitosan-based pMUT can detect ultrasound waves up to 10 cm from the emitter – corresponding to a Time-Of-Flight of 70 μ s calculated considering the speed of sound in the water of 1428 m/s (Fig. 8).

IV. CONCLUSIONS

Thanks to their properties and performance, skin-compliant wearable sensors have a huge potential for health and wellness monitoring. Recently, they have gained increasing attention due to their advantages, which include transparency, high flexibility, strong stretchability, free folding and bending, biocompatibility, compliant features, etc. For these reasons, the range of possible applications of wearable sensors is widely increasing, especially in pre and post-care and medical treatment for patients. Exploiting the direct piezoelectric effect, piezoelectric transducers are efficient transduction devices for immediately detecting and tracking biomechanical stimuli and signals such as movements, vibrations, and body sounds directly from the skin and in an unobtrusive way.

Here, inorganic piezoelectrics on polymeric substrates and piezoelectric biopolymers exploited for skin-compliant biomechanical transducers have been reported. They are successfully applied to the human body to detect vital signs and physiological parameters, even in *in-vivo* experiments.

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