3D-Printable Metamaterial Integrated Piezopolymer-Based Sensing Platforms (3D-MIPS)

Hamdi Torun*[1], Olgac Ergeneman[2]

[1]Department of Mathematics, Physics and Electrical Engineering, Northumbria University, Newcastle upon Tyne, UK
[2]Institute of Robotics & Intelligent Systems (IRIS), ETH Zürich, Zurich, Switzerland

* hamdi.torun@northumbria.ac.uk

ABSTRACT

We have demonstrated flexible PVDF-based acoustofluidic devices integrated with electromagnetic metamaterials in this project. Acoustofluidics have been demonstrated effectively for liquid manipulation functionalities. Likewise, electromagnetic metamaterials have been employed as highly sensitive and wireless sensors. In this work, we introduced a new design combining the concepts of acoustofluidics and electromagnetic metamaterials on a single device realised on a flexible PVDF substrate. We characterise the operation of the device at acoustic and microwave frequencies. The demonstrated platform can be fabricated using 3D printing techniques and can be used in wearable biosensors with integrated liquid sampling and continuous wireless sensing capabilities.

Keywords: metamaterials; surface acoustic waves; PVDF

1. INTRODUCTION

In this project, we have developed a wearable sensing platform by integrating two emerging technologies: metamaterial sensors and piezoelectric polymers. The long-term goal of the project is to offer a technology that can be utilised for the realisation of versatile, cost-effective and personalised devices. This project highlights a road map for the integration of electromagnetic metamaterials and piezoelectric smart materials onto one mechanically flexible platform towards this goal.

Next generation of wearables beyond the detection of physiological biomarkers requires reliable and accurate biochemical detection. The project is designed towards a new approach to combine manipulation and actuation capabilities of piezoelectric layers with wireless and sensitive detection capabilities of electromagnetic metamaterials on a single, 3D-printable and flexible platform. This platform can be adapted for various applications where *in situ* and continuous monitoring are required enabling us to address three key challenges for future wearable devices: i) wireless and sensitive detection; ii) liquid manipulation and actuation; iii) personalisation.

In this project, we focused on polyvinylidene fluoride (PVDF) substrates for the fabrication of devices. PVDF is a functional piezopolymer that has found several applications, such as an electrical insulator, a binder in lithium ion batteries or as a membrane for protein blotting attributed to its distinct chemical, mechanical, and electrical properties. Additionally, the piezoelectric properties of PVDF have been exploited for manufacturing electroacoustic transducers, tactile sensors, and actuators. Unlike its state-of-the-art stiff and fragile piezoelectric material counterparts, PVDF can be easily shaped and is suitable for bending, a requirement for wearable sensors. Another advantage of PVDF is that its mechanical and electric properties can be tailored by copolymerizing PVDF with monomers such as trifluoroethylene (TrFE), chlorotrifluoroethylene (CTFE) and HFP (hexafluoropropylene). PVDF is also a piezoelectric building block in magnetically coupled piezoelectric devices and magnetoelectric composites. We designed, fabricated and characterised devices on PVDF using a single-layer metallisation combining surface acoustic waves and electromagnetic metamaterials on a single device. The device exhibits an acoustic resonance at 3.2 MHz and an electromagnetic resonance at 3.4 GHz. The acoustic resonance is used for liquid manipulation whereas the electromagnetic resonance is used for sensing functionalities.

2. STATE OF THE ART

The integration of electromagnetic metamaterials and piezoelectric smart materials onto one mechanically flexible platform has not been demonstrated in the literature. On the other hand, individual technologies have been recently explored and developed. Flexible piezoelectric and piezoresistive sensors have been developed for a variety of applications including force mapping and liquid manipulation. Surface and bulk acoustic wave transducers based on piezoelectric layers have been used for implementing microfluidic functions such as pumping, mixing and nebulisation [1,2]. Similarly, different approaches have been developed to perform both static and dynamic analysis of the biomechanical forces. The fundamental limitation for these devices is the need for unique wiring and individual readout circuitry for each sensor cell. The readout electronics for discrete sensor configurations is bulky, thus not feasible for miniaturised wearable applications. On the other hand, CMOS readout option is advantageous for scalable and standard implementation but does not offer any benefit for personalisation and customisation. Both options need conventional batteries to meet the energy requirements of the sensors and their readout electronics. In the context of the technological benchmark, our unique technology addresses these limitations. We combine piezoelectric transduction with metamaterial sensors on flexible platforms. Our sensor is electrically passive, fully customisable and wireless.

Metamaterials have been developed as a new sensing method, offering unprecedentedly high-quality factors and sensitivity. Metamaterials exhibit electromagnetic properties, which are not present in ordinary materials. Among different geometrical configurations, split-ring resonators (SRRs) are the most common structures for metamaterials. A conventional SRR is simply a ring with a split, which is preferably made of a highly-conductive metal and is fabricated on a dielectric substrate. In addition to their favourable electromagnetic characteristics, the SRR devices can be realised in a simple and cost-effective manner with various flexible configurations. Under an electromagnetic field excitation, a metamaterial structure electrically forms an LC resonator at its magnetic resonance when a circulating resonant current is induced along its surface with its resonant frequency determined by its geometry. SRR structures in centimetre to millimetre-scale are usually used for applications in microwave bands, which are the focus of this project. Unlike conventional types of passive resonators, an SRR structure typically exhibits very sharp resonant behaviour with quality factors larger than 1,000 at microwave frequencies [3]. Owing to high quality factors, the change in resonant frequency of an SRR structure can effectively be used as a sensing mechanism, which can be induced by a change in dielectric properties of the medium and the geometry of the structure.

3. BREAKTHROUGH CHARACTER OF THE PROJECT

Utilisation of PVDF-based flexible substrates with multi-functional devices with liquid manipulation and wireless sensing capabilities will fill the gap between fabrication of 3D functional materials and custom wearable sensors. A key requirement for wearable devices is a smooth and natural interaction with the human body. Therefore, sensors must exhibit flexibility and adaptability. Although customised wearable devices are in high demand, most of them such as wrist bands still rely on rigid sensors, thus limiting their use in many applications. In addition, current sensing technologies are not well optimised for biochemical sensing that requires liquid manipulation.

We identified three important aspects in biosensing technology that need to be addressed at a fundamental level so that the new generation of portable devices can eventually be used for real time and in vivo measurements of molecular interactions. First, we need fluid manipulation capabilities integrated into devices for efficient and accurate sample handling. Second, we need novel and high precision detection methods that are suitable for simultaneous, multiple and precision detection of analyte. Third, we need novel readout methods that are fast and wireless to enable convenient usages in electrically passive detection. In this project, we explored a new concept of integrated wireless sensing technology on PVDF films through the integration of electromagnetic metamaterials and thin film acoustic wave sensors, with capabilities of non-invasive, in-situ and continuous monitoring of environmental parameters and biomolecules wirelessly. The focus of our concept is to achieve a dual functionality using acoustic transduction and metamaterial-based sensing capable of providing non-invasive, portable, fast, affordable, and accurate biosensors.

4. PROJECT RESULTS

Design and Fabrication of the Devices

Surface acoustic wave (SAW) resonators are one of the building blocks of acoustofluidic devices where a metallised interdigitated transducer (IDT) is defined on a piezoelectric substrate.

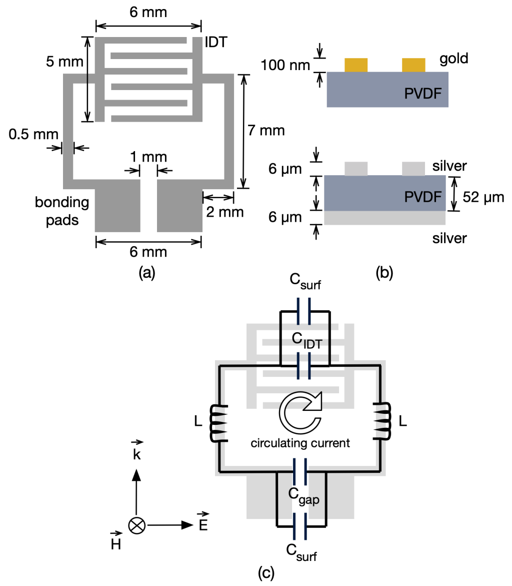

Fig. 1. (a) Schematics of the metallised layer on a PVDF substrate, (b) cross-sectional overview of the fabricated devices, (c) induced circulating current and the equivalent circuit of the device operating as an electromagnetic metamaterial.

The characteristics of the resonator are determined by the geometry of the IDT and are relatively independent to the geometry of the conductors connecting the IDT to the bonding pads where electrical connection is made. Careful design of the conducting line, bonding pad and the IDT can define an electromagnetic resonator at the resonant frequency of which induced current can circulate as we demonstrated recently. Fig. 1(a) shows a design where we designed the metallic layer to serve both functionalities. The IDT comprises 20 pairs of fingers with a wavelength of 300 µm to generate surface acoustic waves on the PVDF substrate. We fabricated two variations of devices using a single gold layer sputtered on a PVDF layer and double silver ink layers on another PVDF layer as shown in Fig. 1(b). Under an electromagnetic field excitation, the metallic structure electrically forms an *LC* resonator at its magnetic resonance as schematically shown in Fig. 1(c). A circulating resonant current is induced along its surface with its resonant frequency being determined by the geometry of the structure. The inductance (L) is given by the conductor geometry, the capacitance is determined by the gap (C_g), the IDT (C_{IDT}) and the surface (C_s) of the structure.

We investigated the electromagnetic resonances based on different excitations of the device as shown in Fig 3 using an electromagnetic simulator (CST Microwave Studio). When the electric field is along the gap (Fig. 3(a) and Fig. 3(b)), the field can polarise the opposing pads resulting in a circulating current. Likewise, when the magnetic field is normal to the device (Fig. 3(b) and Fig. 3(c)), a circulating current is induced. In addition, when the electric current is also perpendicular the fingers (Fig. 3(c)), the alternating fingers can be polarised resulting in complex current paths on the IDT area. This excites additional resonances as observed in Fig. 3(c). When neither the magnetic field is normal to the device nor the electric field polarises the gap, no resonance is observed within the range of 3-4.5 GHz (Fig. 2(d)).

Characterisation of the Devices

We characterised the acoustic and electromagnetic characteristics of the fabricated devices. First, we measured the reflection spectra (S_{11}) of the devices at acoustic frequencies by connecting the bonding pads to a vector network analyser. Fig. 2 shows the fundamental resonant frequency of the PVDF/silver-electrode device is 3.2 MHz. This corresponds to a speed of sound in the PVDF sample as 960 m/s.

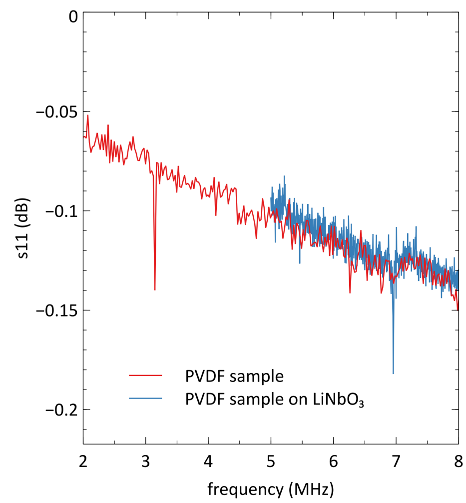

Fig. 2. S_{11} spectra of the PVDF/silver-electrode device measured between the bonding pads

We repeated this measurement after placing the PVDF sample on top of a $LiNbO_3$ plate. The resonant frequency is shifted to 7 MHz, corresponding to a speed of sound of 2100 m/s. The PVDF film is thin and travelling waves can be transmitted to the $LiNbO_3$ plate resulting in increased frequency and effective speed of sound.

In addition to acoustic characterisation, we excited the devices electromagnetically using a pair of monopole antennas to measure the electromagnetic resonant frequency of them. We used monopole patch antennas that are 26 mm in length and 3 mm in width, realised on a 35 µm-thick Cu layer on a PCB substrate. We connect the antennas to the ports of a vector network analyser to excite the device electromagnetically and to measure the transmission (S_{21}) spectra of the devices between the antennas. Fig. 4 shows the S21 spectrum of a device with a fundamental resonant frequency of 3.4 GHz and a quality factor of 300.

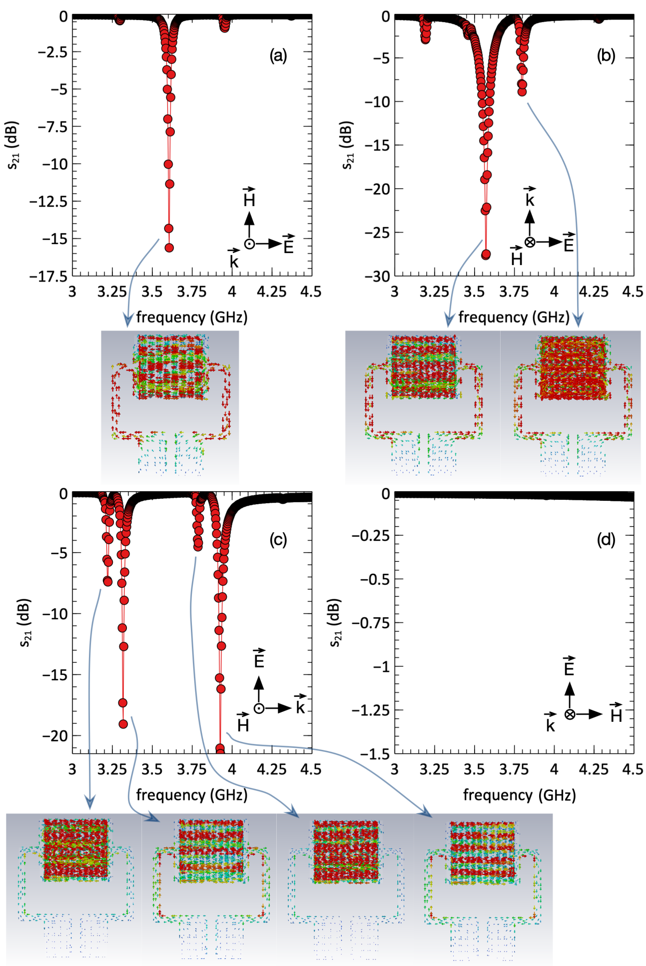

Fig. 2. Simulated electromagnetic resonances of the device with different excitation conditions.

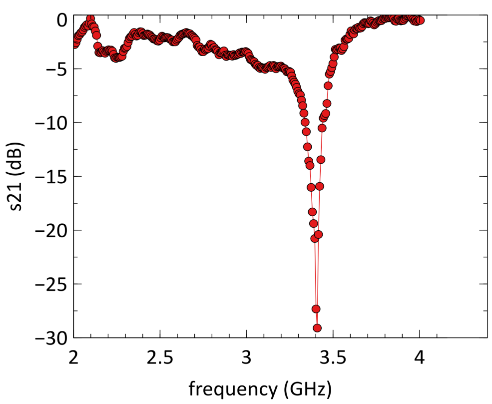

Fig. 3. S21 spectra of the PVDF/gold-electrode device measured using a pair of monopole antennas connected to a vector network analyser.

5. FUTURE PROJECT VISION

5.1. Technology Scaling

In 3D-MIPS we have demonstrated the technology and its capabilities for liquid handling and wireless sensing. The future work is more focused on the optimisation of this technology for biomedical applications. A key application is in sweat analysis for various biomarkers. The technological development towards this aim will be carried out following the steps of:

- Fabricating an array of devices on a single platform;
- Functionalising the surface of the devices with capture molecules of interests;
- Developing a readout unit for the wireless operation;
- Validating the measurements with clinical samples.

5.2. Project Synergies and Outreach

University of Northumbria at Newcastle and Magnes AG, as the organisations implementing this project, are part of a wider consortium that has been established to develop wearable devices able to detect and monitor stress markers in different body fluids wireless connected with a drug delivery system for an effective personalised medicine. The consortium brings together 10 partners located in 7 countries (France, Germany, Italy, Spain, Switzerland, Turkey, and United Kingdom). The results of this project will be made available for the further exploitation and to create synergy towards the aim of stress monitoring.

Stress evaluation is mainly based on survey responses on perceived stress. Commonly used biomarkers for stress (cortisol, chromogranin-A (CgA), alpha-amylase, oxitocin, and beta- endorphin) are not yet successfully validated. Our technology developed in 3D-MIPS offers an innovative platform for the detection of stress biomarkers especially in sweat.

5.3. Technology application and demonstration cases

Our aim for the ATTRACT Phase 2 is to propose an innovative scientific approach that will revolutionise the way to monitor and manage stress, combining objective and self-reported measures. Our approach combines: i) psychological-physiological stress evaluations for an effective stress biomarkers validation; ii) development of advanced sensing materials, microfluidics and wearable biosensors for an effective biomolecules detection and continuous monitoring; iii) design of wearable drug delivery systems for personalised medicine. Our goals are responding to the policy priorities of the Europe 2020 strategy (SC1 “Health, demographic change and wellbeing”) declaring that EU needs to invest in order to improve Europe ability to monitor health and to prevent, detect, treat and manage diseases.

5.4. Technology commercialization

We have already engaged with commercialisation efforts for the technology behind this project. Firstly, we filed a patent application (GB1814829.6 filed on 12.09.2019) to protect the core technology of integrating acoustofluidic and metamaterials devices on a single platform. Secondly, H. Torun was accepted to a commercialisation programme (“Future Founders”) offered by Northern Accelerator, an innovative collaboration between Durham, Newcastle, Northumbria and Sunderland Universities to accelerate commercialisation of research.

5.5. Envisioned risks

Major risks towards the final aim includes technological and clinical aspects. Technological risk is around the functionalisation of the sensors and the resolution of the measurements. We will consider these from the beginning of the project and will implement our project in phases to assess the risks. Different functionalisation strategies such as using aptamers, click chemistry, antibodies, etc. will be evaluated to mitigate the risk. The resolution of the devices can be improved by further miniaturisation. Our technology allows us to go towards this direction, if needed. The clinical risk is directly related to the composition of sweat samples and the presence of relevant biomarkers in sweat. We are assessing several biomarkers in terms of their reliable expression in sweat to mitigate this risk. We will also consider amplifying relevant markers once collected.

5.6. Liaison with Student Teams and Socio-Economic Study

The coordinator, H. Torun an associate professor at Northumbria University, will take the responsibility to create M.Sc. level projects based on the concepts of this project. Torun is responsible from the delivery of M.Sc. projects at Northumbria University and has access to 100+ students annually. A key aspect of our vision is to strengthen the link between fundamental engineering research and public healthcare. Our short- and mid-term goals, for an effective engagement with the public will be a dedicated website on a wiki space host updated on a monthly schedule. Our long-term goal is to exploit the link between engineering and healthcare to inspire young people into the STEM disciplines, building their enthusiasm for both the technology and the benefit it can provide to society. In particular, we will work together with NUSTEM (https://nustem.uk/, Northumbria University STEM project) to organise two demonstration events (end of the second and third years) to target school-age children.

6. ACKNOWLEDGEMENT

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7. REFERENCES

[1] Zhu, P. and Wang, L., 2017. Passive and active droplet generation with microfluidics: a review. Lab on a Chip, 17(1), pp.34-75.
[2] Ramadan, K.S., Sameoto, D. and Evoy, S., 2014. A review of piezoelectric polymers as functional materials for electromechanical transducers. Smart Materials and Structures, 23(3), p.033001.
[3] Torun, H., Cagri Top, F., Dundar, G. and Yalcinkaya, A.D., 2014. An antenna-coupled split-ring resonator for biosensing. Journal of Applied Physics, 116(12), p.124701.