Design, fabrication, and analysis of customized high-density MXene bioelectronics for upper and lower limb muscles

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ABSTRACT — High-resolution $Ti_3C_2T_x$ MXene wearable bioelectronics are a novel technology that shows great advantages in the recording of high-density surface electromyography (HD-sEMG) data, useful to determine muscle activation patterns in the desired area as the patient moves. MXenes are a new class of bioelectronic materials that prove to be highly functional in the creation of medical devices due to their biocompatibility with the human body. The goal of this investigation was to create multiple customizable MXtrodes to perform electromyography (sEMG) on the upper and lower limbs of amputees or patients with muscle movement difficulties to provide them with an accurate diagnosis. The interest of this technology is to provide patients with an economical, comfortable, and highly detailed alternative to conventional electrodes that require uncomfortable and irritating conductive gels. The versatility, simplicity, and scalability of the fabrication process of these new types of electrodes enable the creation of customized electrodes with different geometries for any part of the body with a low-cost manufacturing process. Multiple sets of high-resolution MXene wearable electrodes were designed and fabricated for upper and lower limb muscles and some of them were tested on human subjects. Spatial maps were generated with the obtained data to compare the muscle activation patterns at each point where the MXtrodes were placed to eventually use this data in future applications, like providing medical diagnostics, and rehabilitation to the patients with the use of assistive technologies like the control of prosthetics.

Index Terms— Ti3C2Tx MXene, high-density surface electromyography (HDsEMG), MXtrodes, spatial maps, electromyography (EMG).

I. INTRODUCTION

MXenes are a new class of bioelectronic materials that can be used in a variety of applications. Gel-free MXene wearable electrodes are one of the latest technologies that offer high-density neuromuscular monitoring and diagnostics with non-invasive technology. [1] Here, we introduce MXtrodes, a class of soft, high-resolution, large-scale bioelectronic interfaces enabled by $Ti_3C_2T_x$ MXene and scalable solution processing. In previous studies from the host lab of Dr. Vitale, they showed that the electrochemical properties of MXtrodes exceed those of conventional electrode materials and do not require conductive gels when used in epidermal electronics. Compared to conventional Ag/Agcl electrodes this new material allows the creation of new soft and flexible bioelectronic electrodes for diagnostics and medical applications like the use of prosthetics.

One of the main advantages of $Ti_3C_2T_x$-infused wearable sensors is that they can be customized as desired to study any part of the human body and rapidly fabricated for the patient's need. $Ti_3C_2T_x$ enables a low-cost manufacturing route for the fabrication of multielectrode arrays. These new types of wearable sensors offer a conductivity advantage and are more biocompatible compared to the commercially available sensors that require uncomfortable and irritating conductive gels. MXtrode arrays offer low electrode-skin impedance and maintain functionality for over 20 weeks. They also provide good skin contact for the study of muscles in static and dynamic conditions. These electrodes are highly compatible with clinical imaging technologies, like magnetic resonance imaging (MRI) and computed tomography (CT).

MXtrodes can be used in high-density surface electromyography (HDsEMG) which permits noninvasive muscle monitoring and diagnosis. [2] Surface electromyography (sEMG) is a technique used for noninvasive monitoring and diagnosis of neuromuscular pathologies, rehabilitation, as well as active control of limb prostheses.

In this project, a design of a customized set of MXtrode arrays to perform electromyography (sEMG) on the upper and lower limbs of amputees was created to analyze their muscle activation patterns for further applications like real-time control of prosthetics and assistive technologies. Along this paper we will be going more in-depth into what are MXenes, why are they useful in bioelectronics and learn about the fabrication process of our customizable electrodes.

II. BACKGROUND

Recent studies have discovered the potential use of $Ti_3C_2T_x$ in the manufacture of bioelectronics. MXenes are a class of two-dimensional (2D) materials that have been introduced in the fabrication of medical devices due to their unique properties. They are a family of transition metal carbides, nitrides, and carbonitrides that exhibit high surface area and excellent electrical conductivity. MXenes possess several electrochemical properties that make them desirable for energy storage applications. Their high electrical conductivity enables efficient charge transport within the electrode material. The high conductivity is attributed to the metallic nature of the transition metal carbide/nitride layers in the MXenes. Their accessible surface area also promotes increased electroactive sites for redox reactions and enhances ion/electron storage capabilities. The presence of surface terminations on $Ti_3C_2T_x$ MXene sheets influences electrochemical behavior by altering surface chemistry and charge transfer kinetics. Wearable devices, such as biosensors and energy harvesters, require flexible and conductive-gelled electrodes to establish efficient interfaces with the human body.

High-density MXene wearable electrodes have emerged as a promising candidate for such applications due to their unique electrical conductivity, biocompatibility, and mechanical flexibility. Their electrical conductivity is higher compared to commercially available electrodes, which allows them to transmit electrical signals and detect bioelectrical signals. They have also been shown to have excellent biocompatibility, meaning that they are well-tolerated by biological systems and do not cause adverse reactions if they come in contact with the human body. Another one of the main advantages of the use of MXenes in the fabrication of electrodes and bioelectronics is their mechanical flexibility which allows them to conform to the curved and dynamic surfaces of the human body without compromising their electrical performance. This property ensures the comfort and durability of the MXene-infused wearable sensors. One of the many applications of MXenes is the fabrication of textile electrodes. The process of infusing MXene into textiles allows the development of smart clothing with sensing capabilities.

The development of MXene-infused wearable electrodes has brought a new customizable alternative to conventional electrodes to perform high-density electromyography (HDsEMG) which is a non-invasive technique to measure electrical muscle activity with multiple (more than two) closely spaced electrodes overlying a restricted area of the skin [3].

III. METHODS

The fabrication process of customized high-density MXtrodes for epidermal sensing begins with the modeling of a set of arrays for the desired limb with the use of CAD software, then the process continues as follows with (i) laser patterning a porous absorbent substrate, (ii) infusing it with a water-based $Ti_3C_2T_x$ ink, and (iii) encapsulating the resulting conductive composite in flexible elastomeric films [1].

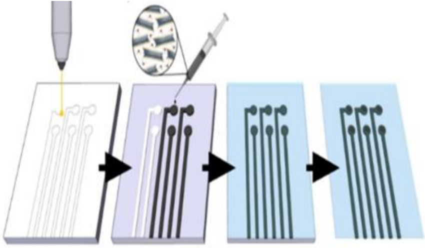
To begin with this process the desired array geometry must be designed and uploaded to a carbon dioxide (CO_2) laser to start patterning a nonwoven hydroentangled (60 to 40%) cellulose-polyester. An acrylic sheet containing a thin layer of polydimethylsiloxane (PDMS), a silicon-based organic polymer, must be prepared. The layer was created by preparing 15 g in a 1:10 ratio of curing agent to base. It was then degassed to remove air bubbles and cured at 70°C for 30 minutes.

After preparing the acrylic sheet and obtaining our desired laser-patterned cellulose-polyester the textile was placed over an acrylic sheet with medical bio adhesive.

A 20 mg mL^{-1} $Ti_3C_2T_x$ aqueous solution was loaded into the textile with the use of a syringe to create flexible and conductive fabric. After drying our MXene-infused substrates in a vacuum oven for approximately one hour at 70°C the substrates were ready for the next step. Flexible Flat Cable (FFC) connectors were attached by screen printing at the end of the arrays with a silver conductive epoxy as shown in Figure 2. The silver epoxy was cured at 70°C for 90 minutes.

Electrodes resistance was tested individually to prove their functionality and then a second layer of (PDMS) was applied to encapsulate and prevent textile degradation of the electrode arrays. The second layer was prepared by using 30 g of 1:10 (PDMS) and spread over the arrays to completely encapsulate them and result in a flexible and conformable film.

As a final step of the fabrication process of our high-density MXene wearable electrodes, the cured (PDMS) layers were cut into the desired shape and the electrode sites were exposed by using a biopsy punch to cut the head of the electrodes and expose the conductive MXene composite.



Driscoll N, Vitale F, et al., Sci. Trans. Med., 2021, 13(612): abf8629

Figure 1: MXtrode Fabrication process; (i) laser patterning a porous absorbent substrate, (ii) infusing it with a water-based $Ti_3C_2T_x$ ink, (iii) attaching flat cable connectors, and (iv) encapsulating the resulting conductive composite in flexible elastomeric films [1].

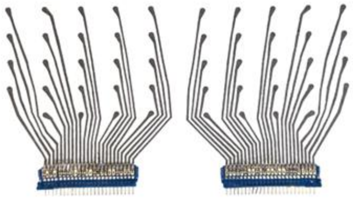
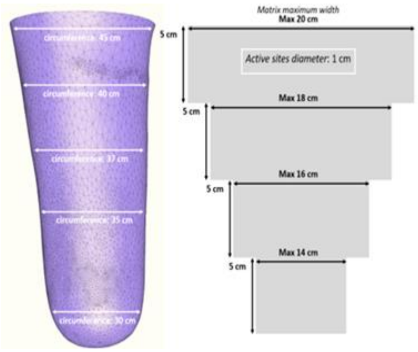


Figure 2: Customized design of MXtrode arrays created for upper arm muscles.



Adapted from: Micera lab, Scuola Superiore Sant'Anna

Figure 3: 3D scan of an amputee’s leg used for the creation of our customized MXene wearable electrodes.

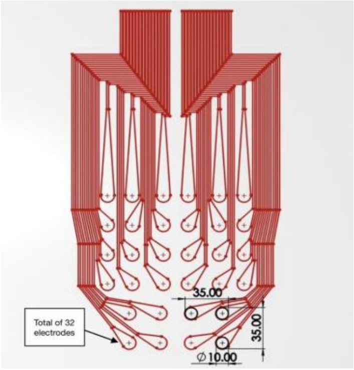


Figure 4: CAD design of the customized Leg MXtrode arrays that consist of 32 active electrodes.

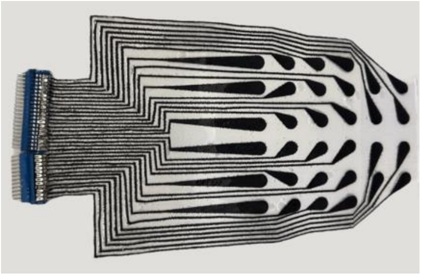
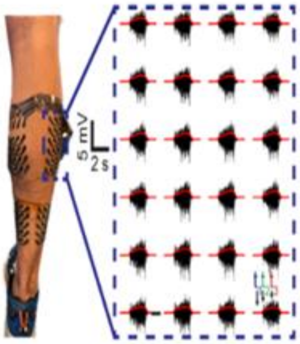


Figure 5: Finalized Leg MXtrode designed for the active control of prosthetics.

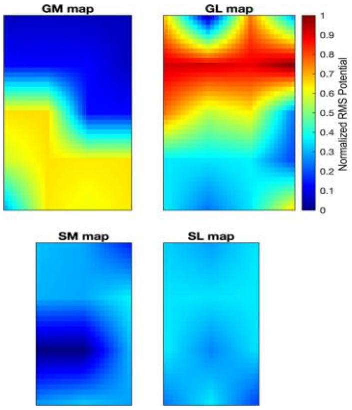
IV. MXTRODE APPLICATIONS

The mapping of spatial activation across muscles is a technique that allows us to visualize the (HDsEMG) muscle activation patterns across time during different motor tasks, such as walking, running, and jumping. Specific differences in activation patterns can be seen during a maximum voluntary isometric contraction. The scale is normalized between minimum and maximum activation potential (0-1).



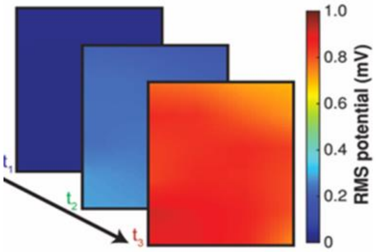
Adapted from: Garg et al., Small Methods., (2022)

Figure 6: Use of MXene bioelectronics for the recording and processing of (EMG) data from lower limb muscles.



Adapted from: Maggie Wagner, [Unpublished figure]

Figure 7: Spatial map with the visualization of three plantar flexor muscles across four MXtrode pads during a maximum voluntary isometric contraction.



Adapted from: Garg et al., Small Methods., (2022)

Figure 8: Colormaps illustrating the (EMG) activity at a specific time changing from no muscle movement at point t1 to maximum muscle voluntary contraction at t3.

V. CONCLUSION AND FUTURE STEPS

The objective of this research was to fabricate a customized set of MXene arrays to perform electromyography (sEMG) on the upper and lower limbs of patients to record and analyze high-density electromyography data. This technology will provide patients with a rapid, low-cost, and effective alternative to commercially available electrodes. During the summer term, we were able to work on the design of a set of customized MXene wearable bioelectronics for an amputee's leg and for the upper and lower arm muscles of multiple patients. The fabrication process and comfort provided by the MXene-infused arrays propound their use as a low-cost, gel-free, customizable, skin-conformable option for capturing (EMG) measurements across the muscles in any part of the body. The future directions are to test the fabricated MXtrode designs on multiple patients and create spatial maps to distinguish muscle activation patterns across time during a maximum voluntary contraction and eventually use this information to provide medical diagnostics, rehabilitation and apply the use of assistive technologies like real-time control of prosthetics.

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