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Exoskeletons for Aided Mobility

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À minha família.

To my family.

Abstract

The increasing number of people suffering from some kind of mobility impairments and disorders is a real concern for modern society. As a result, since a long time ago, basic solutions have already been available to mitigate the effects of this social problem, trying to include those people in the community. Wheeled vehicles, wheelchairs or wanderers offer some movement independence, which improves the quality of life and self-esteem of patients. However, those solutions are not enough to fight and prevent effectively this social problem, since they only help patients go through their daily routines instead of promoting their rehabilitation. Therefore, other alternatives need to be created.

In the last century, the scientific community, headed by researchers, doctors and engineers, has been trying to develop robust solutions to support those patients. Exoskeletons and exosuits are the strongest possibilities since they not only give support in daily tasks but also because they can be used in rehabilitation scenarios. These promising solutions represent the special motivation for the work proposed and developed in this dissertation.

Taking advantage of some properties of soft materials, in contrast to the traditional and rigid ones, an exoskeleton prototype was developed and studied to be used in re-educational movement therapies.

After an exhaustive literature review of all technical aspects related to the development of such devices (chapter 3), including materials, actuation and energy systems, control architecture and sensors, the exoskeleton prototype was conceptualized using soft solutions.

A PVDF-based soft actuator was produced using a novel experimental protocol, involving a non-toxic solvent and an ionic liquid. Firstly, the obtained piezoelectric films were characterized in terms of mechanical and chemical properties and observed under optical microscopy (chapter 4). Secondly, the electromechanical performance and viability of the PVDF-based actuation solution were scrutinized using different inputs (chapter 5), before claiming its effectiveness in real applications. The results of these studies showed to be very promising, in terms of displacement and haptic force.

With these significant outcomes, as proof of concept, a *finger sleeve* prototype was designed, being structurally printed with thermoplastic Polyurethane (TPU) (chapter 5). Two PVDF-based soft actuators were incorporated in the prototype to haptically feedback the user's finger, stimulating specific muscles in that region. This particular soft approach represents a more aesthetical and comfortable solution for users since the materials involved are light, biocompatible and easily respect the body's physiognomy and shape.

With the *finger sleeve* prototype completely developed, an *in silico* user study case was performed to investigate how the device would behave in real conditions (chapter 6). In the end, the device proved to exert a promising haptic sensation.

Furthermore, as a preliminary approach to the control actuation system, EMG sensors were used to capture the muscle contraction (chapter 7). This signal works as a trigger to actuate the device and, hence, support the user's movement.

The overall study contributed to enhancing the knowledge of soft technologies, emphasizing their promising potential to effectively treat and rehabilitate people with physical impairments. Nevertheless, more research and development are needed to achieve the ideal solution that gives back the best life to patients.

Keywords: Exosuit; Soft Materials; Rehabilitation Therapies; Haptic Feedback Device; *Finger Sleeve* Prototype

Resumo

O crescente número de pessoas que sofrem de algum tipo de limitação ou desordem motora é motivo de grande preocupação na sociedade moderna. Como resultado, desde há muito tempo, existem já soluções básicas que mitigam os efeitos causados por este problema, tentando assim incluir estas pessoas novamente na comunidade. Cadeiras motorizadas, cadeiras de rodas ou andarilhos oferecem já alguma independência, o que melhora a qualidade de vida e a auto-estima dos pacientes. No entanto, soluções deste tipo não são suficientes para combater e prevenir de forma eficaz este problema social, pois apenas ajudam nas tarefas diárias, em vez de promoverem a reabilitação. Em consequência, novas alternativas foram desenvolvidas.

Desde o século passado, a comunidade científica, encabeçada por investigadores, médicos e engenheiros, têm tentado desenvolver soluções robustas para apoiar estas pessoas. Entre as soluções mais promissoras, estão os exosqueletos e exofatos, uma vez que estes dispositivos também podem ser usados em cenários de reabilitação. Estas soluções promissoras representam uma motivação especial no desenvolvimento do trabalho proposta e apresentado nesta dissertação.

Tirando partido de algumas das propriedades dos chamados materiais suaves, em contraste com materiais tradicionais e rígidos, foi desenvolvido um protótipo de um exosqueleto para situações terapêuticas de reabilitação.

Após uma extensa revisão bibliográfica sobre os aspetos técnicos relacionados com o desenvolvimento de tais dispositivos externos (capítulo 3), que incluiu os materiais estruturais, energia e sistema de atuação, arquitectura de controlo e sensores relacionados,

o protótipo de exoqueleto foi conceptualizado servindo-se de soluções suaves.

Usando um protocolo experimental distinto, envolvendo um solvente não tóxico e um líquido iónico, foi desenvolvido um atuador suave à base de PVDF. Primeiramente, os filmes piezoelétricos obtidos foram caracterizados mecânica e quimicamente, e observados com microscopia ótica (capítulo 4). De seguida, o desempenho eletromecânico e a viabilidade dos mesmos foram escrutinados, usando diferentes estímulos (capítulo 5), antes de qualquer conclusão tendo em vista aplicações reais. Os resultados destes estudos mostraram-se bastante promissores.

Tendo isso em consideração, como prova de conceito, o protótipo *finger sleeve* foi estruturalmente concebido em poliuretano termoplástico (TPU) (capítulo 5). Dois atuatores suaves baseados em PVDF foram adicionados, para promover uma resposta háptica no dedo do utilizador, estimulando assim os músculos nessa região. Esta abordagem suave representa uma solução mais confortável e agradável visualmente, uma vez que os materiais usados são leves, biocompatíveis e respeitam a fisionomia do corpo.

Após o desenvolvimento do protótipo, foi realizado um estudo *in silico* para perceber o comportamento do dispositivo em situações de uso real (capítulo 6). Os resultados apontaram um estímulo háptico promissor.

Complementarmente, foi ainda desenvolvido um sistema de controlo, baseado em sensores EMG, para fazer a aquisição de sinais de contração dos músculos, funcionado como um gatilho para atuação do dispositivo (capítulo 7). Dessa forma, o suporte ao utilizador estaria garantido.

Em conclusão, o presente estudo contribuiu para uma melhor compreensão de tecnologias suaves, com enfâse no seu enorme potencial em aplicações de reabilitação. No entanto, são necessários mais estudos sobre este tipo de alternativas.

Keywords: Exofato; Materiais Suaves; Terapias de Reabilitação; Dispositivo de Feedback Háptico; Protótipo *Finger Sleeve*

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At this point, it would be impossible for me not to acknowledge the most important people in my life, who have supported me since the beginning of this already long journey. More important than having people who help you to get up from the ground, is to have people who do not let you fail.

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António Diogo Figueiredo André

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“Deus Quer, o Homem Sonha, a Obra Nasce.”

in ’Mensagem’, Fernando Pessoa

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List of Abbreviations

ABS	Acrylonitrile Butadiene Styrene
AFM	Atomic Force Microscopy
AFO	Ankle-foot orthosis
BCs	Boundary conditions
BM	Berlincourt method
BNNT	Boron nitride nanotube
CICECO	Aveiro Institute of Materials
CNS	Central nervous system
DEA	Dielectric elastomer actuator
DMAC	Dimethylacetamide
DMF	Dimethylformamide
DMSO	Dymethilsulfoxide
DOF	Degrees of freedom
DSC	Differential scanning calorimetry
EA	Electroactive
EAP	Electroactive polymer
ECG	Electrocardiogram
ECP	Electrically conducting polymer

EEG	Electroencephalogram
EMG	Electromyography
EU	European Union
FCT	Portuguese Foundation for Science and Technology
FDM	Fused Deposition Modeling
FEUP	Faculty of Engineering of University of Porto
FEA	Finite Element Analysis
FEM	Finite Element Method
FFF	Fused Filament Fabrication
FTIR	Fourier-transform infrared spectroscopy
i3A	Aragon Institute for Engineering Research
IL	Ionic liquid
ICL	Imperial College London
INEGI	Institute of Science and Innovation in Mechanical and Industrial Engineering
INL	Iberian Nanotechnology Laboratory
IR	Infrared
LAETA	Associated Laboratory of Energy, Transports and Aeronautics
LASI	Intelligent Systems Associate Laboratory
MEK	Methyl ethyl ketone
MEMS	Microelectromechanical systems
MCP	Metacarpophalangeal
MMG	Mechanomyography
MWCNTs	Multi-walled carbon nanotubes
PC	Personal computer

PFM	Piezoresponse Force Microscopy
PID	Proportional integral derivative
PLA	Polylactic acid
Pmim	1-propyl-3-methylimidazolium (IL cation)
PPy	Polypyrrole
PU	Polyurethane
PVDF	Polyvinylidene fluoride
PZT	Zirconate Titanate
QOL	Quality of life
RFID	Radio frequency identification
SC	Spinal cord
SEM	Standard error of the mean
SMA	Shape memory alloy
SMG	Sonomyography
SMM	Shape memory materials
SMP	Shape memory polymers
STD	Standard deviation
TEMA	Centre for Mechanical Technology and Automation
TGA	Thermal gravimetric analyser
TFSI	bis(trifluoromethanesulfonyl)imide (IL anion)
TPU	Thermoplastic Polyurethane
UCVE | UBS	Eng. Design & Experimental Validation Unit | Biomechanical and Health Unit
UN	United Nations
USA	United States of America

VR	Virtual reality
WHO	World Health Organization
XRD	X-ray diffraction

List of Symbols

A [mm^2]	Area
A_λ [dimensionless]	Absorbance coefficient at wavelength (λ)
d [mm]	Thickness
d_{kij} [pm/V; pC/N]	Piezoelectric constants
d_{33} [pm/V; pC/N]	Piezoelectric constant in direction 3 per unit stress applied in 3 axis
d_{31} [pm/V; pC/N]	Piezoelectric constant in direction 3 per unit stress applied in 1 axis
D_i [C/m^2]	Electric displacement
D_{exp} [mm]	Experimental displacement
D_{num} [mm]	Numerical displacement
C [F]	Capacitance
E [MPa]	Young modulus; Stiffness modulus
Err [%]	Error
E_k [MV/m]	Electric field
F [N]	Force
f [Hz]	Frequency
ΔH [J/mol]	Melting enthalpy
k [N/m]	Spring constant
K_λ [cm^2/mol]	Absorption coefficient at wavelength (λ)

l [mm]	Length
l_f [mm]	Free length
l_t [mm]	Total length
l [mm]	Free length of the soft actuator
p [Pa]	Contact pressure
R [Ω]	Ohmic Resistance
R [mm]	Radius
$s^E{}_{ijkl}$ [N/m]	Elastic compliance constants
S_{ij} [dimensionless]	Strain components
t [mm]	Thickness
T [s]	Period
T_λ [%]	Transmittance at wavelength (λ)
T_{kl} [Pa]	Stress components
T_m [°C]	Melting temperature
T_{onset} [°C]	Onset temperature
V_{AC} [V]	Voltage intensity
α	Crystalline phase of PVDF
β	Crystalline phase of PVDF
γ	Crystalline phase of PVDF
δ	Crystalline phase of PVDF
δ_P [$MPa^{1/2}$]	Coefficient of solubility (Hansen solubility)
ε	Crystalline phase of PVDF
ε_0 [F/m]	Permittivity of the vacuum

$\varepsilon^{T}{}_{ik}$ [F/m]	Permittivity constants
ε' [dimensionless]	Dielectric constant; Relative permittivity
ε_{Yield} [dimensionless]	Yield strain
θ [°]	Angle range
λ [°A]	Wavelength
ν [dimensionless]	Poisson ratio
ρ [kg/m^3]	Density
σ_{AC} [S/m]	AC conductivity
σ_{Yield} [MPa]	Yield stress
χ [%]	Degree of crystallinity

Chapter 1

Thesis Overview

1.1 Motivation and Aims

Mobility disorders have a high and long-term impact on the social, economic and financial sphere, affecting entire communities and healthcare systems worldwide, since in modern countries, the life expectancy has been increasing [1, 2]. Psychologically, locomotion problems are a source of concern, since they may be stressful, painful and, often, the cause of depression because the simplest and basic tasks are no longer easily achievable. Furthermore, the entire social person's life is negatively affected by these impairments, contributing even more to the depressive state of mind, seclusion and solitude [3–5]. These mental problems are prevalent during physical disabilities, affecting not only the person himself but also inner and outer circles, such as family, friends and even colleagues.

The lack of mobility also contributes to muscles' deterioration, which could result in bedridden or immobilized people [6], exacerbating even more some of the psychological problems already mentioned. In addition, the risk of other severe secondary medical conditions is higher, including obesity, coronary heart diseases and diabetes [7].

Scientists and engineers, whether from academic or industrial fields [8], have been increasing efforts to mitigate the consequences and the negative impact caused by ageing, injuries and diseases. For centuries, wanderers, wheeled vehicles and wheelchairs have been used as simple solutions to aid in mobility [9]. These auxiliary locomotion devices have the main purpose of restoring and giving back some personal freedom and independence to people. The most common solutions found in society are intended to only help patients in the movement of limbs, without going further in their rehabilitation. Precisely intending to fill in that void, exoskeletons and, more recently, the concept of exosuits started to be developed [10].

These external devices, in particular those conceptualized using soft materials, are considered promising alternatives since they have desirable intrinsic characteristics, such as an aesthetic appearance, lightness and biomimetism. They intend to be wearable, comfortable, user-friendly and faced as part of the body and not external and intrusive equipment, such as the rigid options.

Employing the concept of exosuits, the present work aims to develop a rehabilitative wearable soft device for re-educational movement purposes and study its soft actuation system, based on the use of piezoelectric materials as the main active elements. This choice confers to the actuation system the ability to be actuated when electrically stimulated.

1.2 Thesis Outline

The present dissertation was structured based on published and submitted scientific papers. It is composed of a total of 9 chapters.

Chapter 1, **Thesis Overview**, which contextualizes the thesis project, specifying the "Motivations and Aims" of the work for developing such a device. Moreover, the structure of the present dissertation ("Thesis Outline") is detailed in this chapter, as well as the "Personal Contribution and Conflict of Interest" statement.

Chapter 2, **Overall Context & State of the Art**, intends, firstly, to expose the demographics of people with mobility impairments, correlating it with the need to develop exo-supportive devices, which would benefit from the use of smart materials. In particular, the use of PVDF-based materials is exposed, focusing on the fields of medicine, bioengineering and rehabilitation. Moreover, the state of the art of exosuits using these soft materials for rehabilitation is described.

Besides a brief introduction to the context and the role of external devices in rehabilitation, Chapter 3, **Exo Supportive Devices: Summary of Technical Aspects**, focuses on the key technical aspects to develop an external supportive device. It includes the mechanical design, structural materials (rigid and soft), actuators and energy sources (traditional and soft actuators with their inherent energy sources) and, finally, the control system (describing the control architectures and sensors). In the end, the chapter describes some solutions for ankle/foot and hand/arm rehabilitation, addresses some ethical concerns and presents current and future perspectives.
This chapter is based on the following published paper: *A. D. André, P. Martins. Exo Supportive Devices: Summary of Technical Aspects. Bioengineering, vol.10, p.1328, November 2023. DOI: https://doi.org/10.3390/bioengineering10111328.*

Based on the literature review, a soft actuator was proposed in this work as a solution for a soft device to use in haptic feedback contexts. In chapter 4, called **Influence of DMSO non-toxic Solvent on the Mechanical and Chemical Properties of a PVDF Thin Film**, a soft material was developed and studied. A unique experimental protocol was designed to develop thin films, which comprises the actuation system, considering the combination of pure PVDF and an ionic liquid (IL) ([PMIM][TFSI]), dissolved in a non-toxic solvent (DMSO). This approach enhanced the properties of the thin film, being crucial for further characterization. An optical observation was carried out, to ensure the viability of the experimental protocol, in terms of PVDF dissolution and solvent evaporation. Moreover, the mechanical and chemical properties were studied to better understand the elastic limits of the samples and to be aware of the electroactive phases present.
This chapter is based on the following published paper: *A. D. André, A. M. Teixeira, P. Martins. Influence of DMSO Non-Toxic Solvent on the Mechanical and Chemical*

Properties of a PVDF Thin Film. Applied Sciences, vol. 14(8), April 2024, DOI: https://doi.org/10.3390/app14083356.

Following the characterization of the material, in chapter 5, **Piezo-Ionic Actuator for Haptic Feedback**, more studies were performed on PVDF-based actuators regarding the electromechanical properties and performance. As the PVDF-based thin films are intended to be used as soft actuators in a wearable rehabilitative device, it is extremely important to know the responsiveness of the material. Firstly, some electrical and piezo properties were determined. After that, the performance of the PVDF-based actuators was assessed by measuring the displacements and forces of samples subjected to different electrical inputs. In the end, a prototype of the external device was created, assuming an index finger as a model. It was called *finger sleeve* device and was structurally 3D printed using thermoplastic polyurethane (TPU). Its ultimate goal is to haptically feedback impaired muscles in rehabilitation therapies.

This chapter is based on the following published paper: *A. D. André, I. Coondoo, I. Bdikin, K.B. Vinayakumar, R.M.R. Pinto, P. Martins, M. Taghavi. Piezo-Ionic Actuator for Haptic Feedback. Sensors and Actuators A: Physical journal, vol.381, January 2025, DOI: https://doi.org/10.1016/j.sna.2024.116038*.

Considering all the properties of PVDF-based soft actuators previously achieved, in chapter 6, **In-Silico User Study Case: Wearable Feedback Haptic Device for Rehabilitation**, an *in silico* user study case of the wearable haptic device (presented chapter 5) was performed, considering a finite element analysis (FEA). Since it was not possible to experimentally measure all the piezo properties of the actuators, an inverse finite element approach was first developed to complete the characterization of the actuator, which is essential for the simulation. After that, the *in silico* user study case was carried out, using the *finger sleeve* device and a finger model based on literature, to simulate the interaction between the device and the user, assessing the effectiveness of the solution.

This chapter is based on the following publishedpaper: *A. D. André, M. Parente, P. Martins. In-Silico User Study Case: Wearable Feedback Haptic Device for Rehabilitation.*

Proceedings of the iMeche, Part L: Journal of Materials: Design and Applications journal, 0(0), October 2024. DOI: https://doi.org/10.1177/14644207241288391.

Chapter 7, **EMG Signals as a Way to Control Soft Actuators**, presents a preliminary practical example of how biological signals can be used as input for the actuation of PVDF-based soft actuators. Using electromyographic (EMG) sensors, strategically placed on the (impaired) muscles, the user movement intention is captured through the detection of the bioelectrical signals produced, which will be the trigger for the actuation of the device.

This chapter is based on the following published paper: *A. D. André, A. M. Teixeira, P. Martins. EMG Signals as a Way to Control Soft Actuators. In: Tavares, J.M.R.S., Bourauel, C., Geris, L., Vander Slote, J. (eds) Computer Methods, Imaging and Visualization in Biomechanics and Biomedical Engineering II. CMBBE 2021. Lecture Notes in Computational Vision and Biomechanics, vol 38. Springer, Cham. DOI: https://doi.org/10.1007/978-3-031-10015-4_4*.

In chapter 8, an overall **Discussion** of the findings and results achieved are under analysis, since a more detailed interpretation of the results is placed at the end of the correspondent chapters.

Finally, chapter 9 summarises the main **Conclusions** of the present dissertation, regarding the development of an exosuit for aided mobility using soft materials. Moreover, possible pathways for **Future Works** are synthesized.

Figure 1.1 summarizes and links all the chapters of the present dissertation.

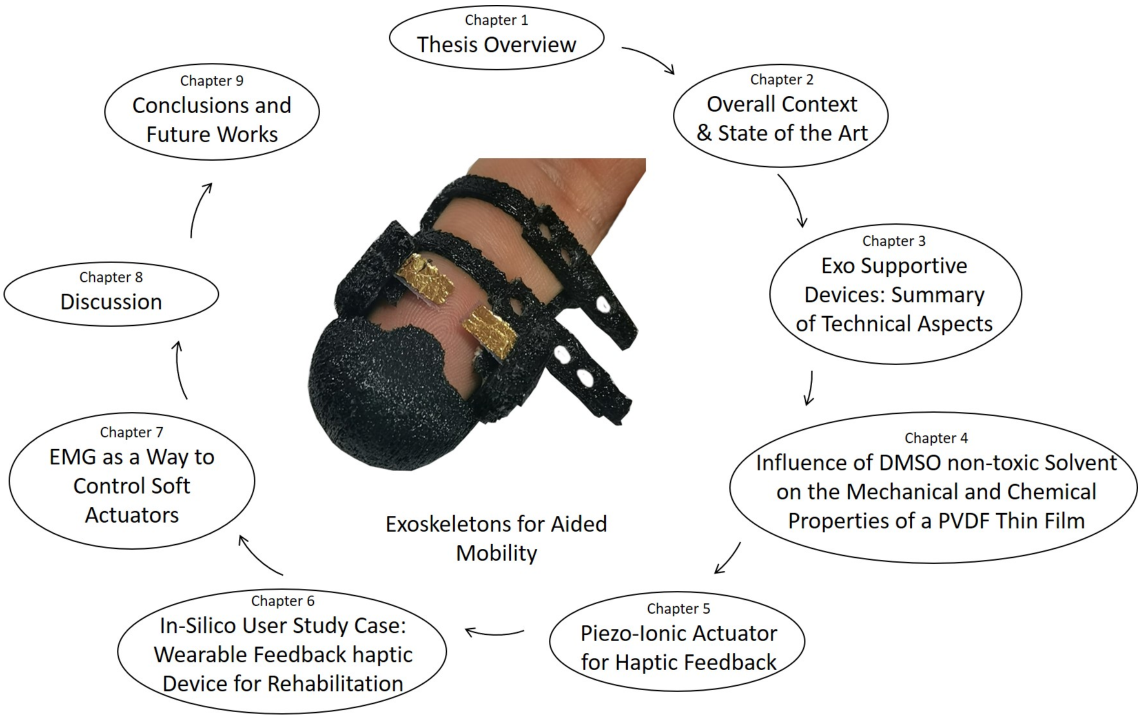

Figure 1.1: Schematic representation of the thesis outline.

1.3 Personal Contribution and Conflict of Interest

Personal contribution: Personal contribution and co-authors' contribution are described in each corresponding chapter.

Every chapter was conceptualized by the author, António Diogo André, and the experiments were carried out with the collaboration of other researchers, technicians and engineers mentioned in each chapter.

All chapters were written by António Diogo André and edited by the authors mentioned in each paper. Chapter 3 was edited by António Diogo André and Pedro Martins; Chapter 4 was edited by António Diogo André, Ana Margarida Teixeira and Pedro Martins; Chapter 5 was edited by António Diogo André, Indrani Coondoo, Igor Bdikin, Vinaya Basavarajappa, Rui Pinto, Pedro Martins and Majid Taghavi; Chapter 6 was edited by António Diogo André, Marco Parente and Pedro Martins; Chapter 7 was edited by António Diogo André, Ana Margarida Teixeira and Pedro Martins.

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Chapter 2

Overall Context & State of the Art

Mobility disorders are a social problem that has been intensified by the evolution and modernization of societies. From decades ago, demography has evolved, showing that people live longer, which exacerbates locomotion problems in society. To face and mitigate the situation, exo devices have been developed to support and rehabilitate impaired muscles. With the evolution of materials, new solutions have appeared. Those include the use of smart materials since they can smartly adapt their shape and size to different environments, triggered by external inputs.

The present chapter intends to expose the demography that led to the scenario of mobility disorders, the state of the art of exo devices in rehabilitation and smart materials, with a special focus on the electro-responsive ones (Figure 2.1).

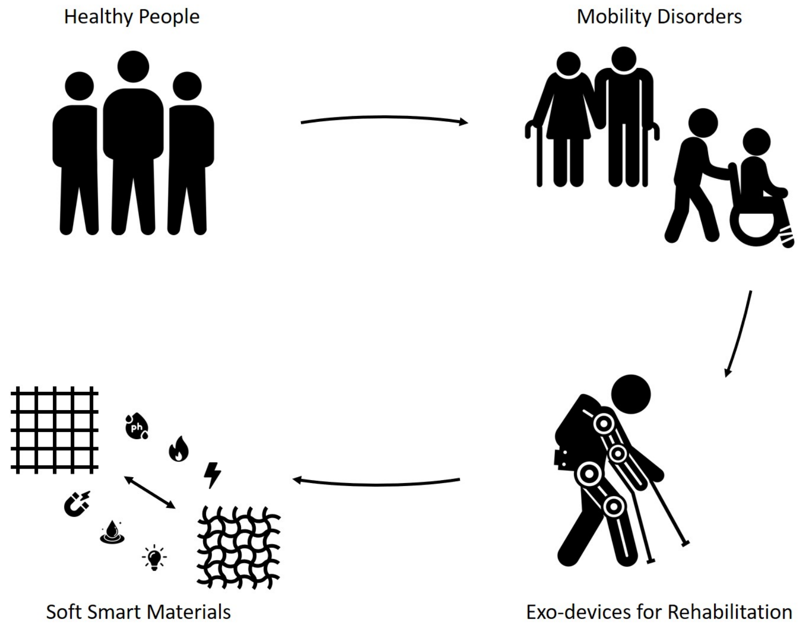

Figure 2.1: Overall context that leads to the development of exo-devices, which can be manufactured using soft smart materials.

2.1 Demographics Overview

According to the World Health Organization (WHO), at the beginning of the present decade, approximately 1.3 billion people worldwide experienced some physical disability [1]. Unfortunately, physical impairment limitations are a very common reality in all societies, despite the advances in technology and science, and the implementation of new legislation that imposes new rules for building facilities and city urbanisation.

Governments of several countries have already been implementing several laws to protect mobility-impaired people from inequality and to include them in society. Organizations, such as the United Nations (UN) and European Union (EU), have established some standards to be adopted for all their members, in terms of accessibility [2, 3]. In Portugal, for example, the decree law no. 163/2006, established technical requirements for accessibility in buildings and public spaces, such as ramps and elevators, as well as in public transportation, including the design of buses and trains [4].

The causes of physical impairments are essentially based on the normal ageing process, in consequence of the increasing life expectancy [5, 6]; on neurodegenerative diseases and disorders [7]; on daily accidents, such as falls, motor vehicle accidents or sports-related incidents [8]; and even on environmental factors [9].

The life expectancy of a country or a nation is directly related to its level of development. In those in which the quality of life (QOL) evidence higher indices, the average lifespan also tends to be higher than in countries with lower QOL indices. Some European countries, such as Norway and Germany, or Asian regions like Hong Kong and Japan, have an average lifespan of over 80 years. In contrast, African nations such as Central African Republic, Chad and Nigeria, have the lowest lifespan references, below 54 years old.

The high rates of the ageing population are becoming an involving phenomenon, frequently observed in developed countries. With a global population of over 8.1 billion people, approximately 17% live in modern countries [10]. Statistics show that, for instance, in the United States of America (USA), the total number of people with an age over 65 years old will be around 55 million in 2030 [11]. Similarly, in Europe, the percentage of elderly people already represents almost 20 % of the total population in the region [12]. Additionally, predictions from the UN estimate that, in 2050, 20% of the global population will be over 60 years old [13], in which 1.5 billion with an age over 65 years old [14]. These number predictions reflect twice the numbers of 2015 and 2019 [14, 15]. As an intrinsic consequence of an ageing population, mobility problems start becoming a concerning reality for those people [16]. However, locomotion disabilities, as previously mentioned, are not an exclusive effect of long lives. Neurological and degenerative diseases are also a recognised trigger for these problems.

According to the UN, 55 million people around the world suffer from some type of dementia, with millions of new cases every year [17–19]. As expected in situations where neurons are affected, the mobility of patients gradually declines as a consequence of the disease, which often leads to death within 2 years for many patients [20]. Despite ongoing research efforts, there are not yet effective treatments that restore the quality of life of the patients.

Injuries on the spinal cord (SC) can also be a source of physical impairments, for example, tetraplegia, quadriplegia or paraplegia, which lead to changes in muscular strength and body functions. These conditions can affect daily activities, such as walking and lifting simple objects. Common causes for these injuries include car accidents, falls, gunshots or sports-related incidents [21]. Additionally, some pathological problems, such as tumours [22], can also compress the SC, conditioning the control of muscles.

Moreover, mobility issues can impact negatively the psychological state of mind of patients, resulting in stress, pain and depression, since the simplest movements become challenging. In addition, the social life of the person can be severely affected by these mental illnesses and bad feelings [23, 24], having an impact on family and friends.

Indirectly caused by mobility disorders, different health conditions such as sarcopenia, obesity, coronary heart disease and diabetes can occur from the lack of consistent movement of the body and muscles [25, 26]. Aware of all potential negative consequences of the lack of mobility, society is putting efforts to minimize the effects of such infirmities [27]. Solutions from decades ago, such as wanderers and wheelchairs [28], are already available for everyone, allowing people to regain some movement independence. However, the goal of these assistive devices is only to support the daily locomotion needs of patients and not to rehabilitate impaired muscles. Therefore, distinct solutions, such as exoskeletons and exosuits, started to be developed not only to mitigate the effects of mobility disorders but also to promote muscle recovery [29].

2.2 Exo-Devices

The concept of an exoskeleton is inspired and based on nature. Invertebrate animals, like arthropods and molluscs, possess a rigid and hard outer layer, designated as an exoskeleton, which is used by those animals for different purposes. Protecting their fragile bodies from threats and external elements and acting as a barrier against dehydration and as a sensory element with the surroundings [30], are some of the functions of an external skeleton.

The definition of external devices applied to human beings emerged around the 50s of the last century [27]. Typically, they are mechanical systems, equipped with powerful actuators [31], with the main goal of expanding or enhancing the physical abilities of the user [30]. Originally, they were thought for military purposes, such as helping soldiers to carry heavy loads or improving their physical performance [30]. Nowadays, exo-devices are being developed for a wider range of applications, including medical therapies [32] and industrial tasks. As medical devices, exoskeletons can be used in a rehabilitation context after a trauma or surgical procedure as well as for muscle impairment therapies [33]. In industrial companies, they can be used by the employees to reduce their fatigue and, hence, increase their productivity [34]. Another example in which these external devices are a valuable help is for firefighters and rescue workers in which becomes easier to carry heavier loads and search for possible victims under rubble, when a tragic event happens.

Typically, these supportive devices can be controlled remotely by a third person or by the user himself. In cases of rehabilitation or supportive tasks in which the user is in charge, the control system can be implemented using distinct approaches (architectures), depending on the circumstances, to ease and help the movement. It will act as a bridge between the user and the device, capturing and interpreting the bioelectrical signals from the body (i.e. movement intentions) through sensors (such as electromyography - EMG) to activate the actuation system of the exoskeleton.

Despite the extensive range of current exoskeleton applications and their state-of-the-art, there are still undergoing strong academic and industrial developments to achieve even more efficient technologies, such as non-rigid devices. Regarding their actuation system and structural materials, those external devices are designated as exosuits, when soft and smart materials are used [35]. These special materials can be externally stimulated by different sources, such as heat or electrical inputs. In that case, soft solutions are characterized, for example, by flexibility, lightness and biomimetic properties [36, 37]. Exosuits tend to be used essentially in medical rehabilitation exercises, as haptic solutions for impaired muscles, since their abilities to assist are limited when compared to other actuation solutions, such as pneumatic actuators for gait assistance [38].

2.2.1 Exosuits for Rehabilitation

Exosuits, or soft exoskeletons, have several advantages in comparison to traditional rigid exoskeletons. As they are made from soft materials, such as polymers and composites, they are lighter, which reduces the weight of the global solution. In addition, they present biomimetic features, which will allow a more natural and free movement by the user [39].

However, these soft solutions also show some weaknesses when compared to traditional exoskeletons. The fact that they do not present external rigid frames, might make the interaction between the device and the user difficult [40].

Even so, researchers and companies have developed and reported exo solutions comprising soft and smart materials. For instance, Bartenbach et al [41] approached the use of a soft exosuit to give support in leg function for rehabilitation. They developed a device based on a driven system, in which cables are routed from actuators to the webbing segments, supporting the rehabilitation of the leg during gait. They concluded that the device assisted and supported considerably the movements of sit-to-stand and stair ascent.

Awad et al [42] trialled clinically a soft robotic exosuit, called ReWalk ReStore^{TM} [43], to be used in rehabilitation scenarios after stroke. It has implemented a cable-driven actuation system, actuated by batteries placed at the users waist. In the end, they concluded the safety and reliability of using the device for gait rehabilitation. Lessard et al [44] also developed a flexible upper-extremity exosuit for rehabilitation in after-stroke situations. They showed that the device has advantages in terms of portability, weight and ease of movement.

In a different line of rehabilitation devices, Bortone et al [45] developed a wearable haptic device for rehabilitation in children with neuromotor impairments (such as cerebral palsy and developmental dyspraxia), using virtual reality (VR). According to the authors, this approach can create a positive motivation, since the therapies are applied in the form of games. They assessed the viability of the immersive approach, which improved the upper extremity function.

Similarly, Camardella et al [46] developed a finger-type wearable haptic device to be combined with VR for a rehabilitation system. They observed that the effectiveness of the haptic feedback is an important feature of rehabilitation environments. Zhu et al [47] developed a smart, textile-driven, soft exosuit for spinal assistance. The light device was shown to be able to provide compression forces to assist stoop lifting through the use of multi-soft artificial muscles. Mazar et al [48] presented an artificial muscle based on PVDF and powered by glucose, which can be of great interest to soft robotics.

As exemplified previously, soft devices can make use of new materials, in particular smart materials, since the global solution can adapt itself to different environments, triggered and operated by external stimuli.

2.3 Smart Materials

Smart materials is a highly researched topic, with significant and distinct applications in various fields, such as aerospace and aeronautics [49, 50], electronics [51], architecture and design [52], bioengineering [53], and most importantly, in medicine and rehabilitation [54, 55]. This class of materials can respond smartly to the surrounding environment and conditions, by changing their properties and behaviour, including shape and stiffness.

In aviation, for example, smart materials are typically used in the wings of the aircraft (i.e. morphic wings). In that way, aeroplanes can adapt their flight to different outside conditions by changing the aerodynamics of the wings [56]. As a result, the aerodynamic performance is improved, as well as the fuel efficiency. Other examples of the use of smart materials are in microelectromechanical systems (MEMS) [57] in order to obtain flexible and adaptable electronic components, and in construction, where bio-smart materials are applied to repair crack in the concrete, by precipitation of calcite [58]. In healthcare, smart materials can be used to develop smart drug delivery systems, for example, in cancer treatments [59], and can be used in smart prosthetic solutions in rehabilitation therapies [60].

Focusing on rehabilitation scenarios, smart materials are specifically used to interact with patients and users and can be classified, accordingly to the input stimuli (i.e. exterior conditions) [61], as thermal [62], magnetic [62], light [63], pH [64], pressure [65], moisture [66] and electrical [62] triggers.

Electrically actuated smart materials, in particular piezoelectric-based solutions, are widely used for different purposes in healthcare scenarios [67]. Piezo materials have the interesting ability to produce an electrical signal when mechanically deformed (called direct effect) and also to change their shape and size in response to external electrical inputs (called converse effect) [68]. However, when used mostly in their pure state, these materials evidence some disadvantages and limitations for actuation systems, such as limited displacement [69]. Therefore, in literature, piezoelectric materials are rarely used alone due to the need for enhanced output responses. A promising route towards a significant improvement of the output is the addition of fillers during the fabrication process [70], which endows the resultant material with more desirable properties.

Polyvinylidene fluoride (PVDF) is a well-known electrical smart material, since evidences notable chemical and weather resistance [71], along with high flexibility, low density, biomimetic abilities and good biocompatibility, making it highly akin to muscle behaviour emulation [68]. Moreover, it exhibits high mechanical strength and high piezoelectric coefficient [72]. As a result, PVDF-based materials have been selected for a wide range of applications in medicine, bioengineering and rehabilitation, as highlighted in many studies.

Braga et al. [73] studied the mechanical properties and biocompatibility of a composite material based on PVDF to be used in bone restoration and filling, claiming it as a possible solution for medical and dental purposes. Chiu et al. [74] and Mahanty et al. [75] detailed the use of PVDF sensor patches for simultaneous heartbeat and breathing monitoring, obtaining similar signals to those registered by commercial devices. In a different vein of research, Foster et al. [76] used PVDF as a transducer for thyroid and breast imaging, as well as in ultrasound biomicroscopy. In the regenerative medicine field, Agueda et al. [77] synthesized and characterized PVDF scaffolds for renal bioengineering, and Guo et al. [78] investigated wound dressings based on electrical stimulation, which influences cellular behaviour.

Several fillers can be added to PVDF's polymeric matrix during the fabrication process to enhance the piezo behaviour of the resultant material [79, 80]. In particular, ionic liquids (ILs), such as 1-ethyl-3-methylimidazo-lium bis(trifluoromethylsulfonyl)imide ([$C_n$mim][TFSI]) [81], 1-Propyl-3-methylimidazolium iodide ([Pmim][I]) [82] or 1-(2-Amino-ethyl)-3-methylimidazolium bromide ([Aemim][Br]) [83], are widely used in combination with PVDF to improve its piezoelectric properties [84–86].

In consequence, PVDF or PVDF-based materials can be used as soft actuators [87–89] or integrate an actuation system as a sensor [90, 91], being promising approaches to help people with mobility disorders. Although the use of those alternatives still presents some drawbacks, such as limited actuation, it is expected a significant growth in the use of these materials in the future. Pan et al [92] claimed that the use of these piezo materials is in its infancy since they only represent 7% of soft actuation state-of-the-art for rehabilitation and assistance solutions.

In the present dissertation, a wearable exosuit using soft materials, in particular a PVDF-based material as a soft actuator, was developed and characterized in order to be used as a haptic device for rehabilitation scenarios. For that, a unique and optimized experimental protocol was developed, including the use of a non-toxic solvent and an ionic liquid. The influence of this approach on the chemical, physical and mechanical properties of the actuator was assessed. In the end, the integration of the final soft actuator into a real scenario was also investigated using *in silico* tools, proving the potential viability of the solution in biomedical applications.

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Chapter 3

Exo Supportive Devices: Summary of Technical Aspects

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Review

Exo Supportive Devices: Summary of Technical Aspects

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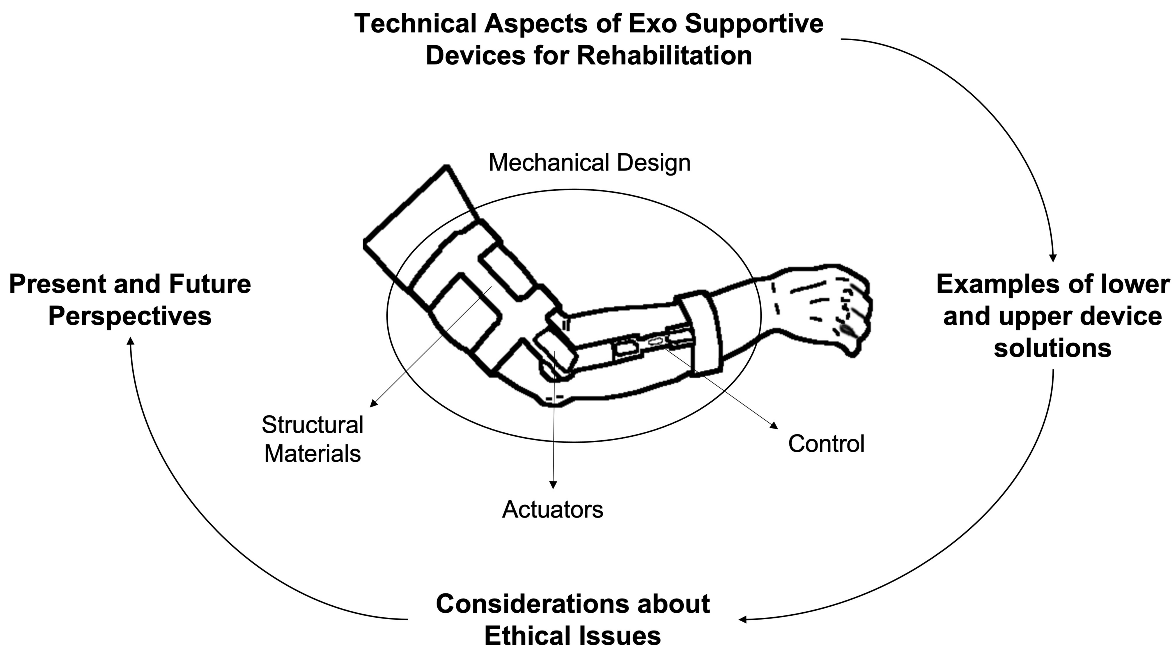
3.1 Abstract

Human societies have been trying to mitigate the suffering of individuals with physical impairments, with special effort from the last century. In the 50s a new concept arose with similarities with animal exoskeletons found in nature and with the goal to medically aid human movement (for rehabilitation applications). There have been several studies on using exosuits with this purpose in mind. So, the current review offers a critical perspective and a detailed analysis of the steps and key decisions, involved in the conception of an exoskeleton. Choices such as design aspects, base materials (structure), actuators (force and motion), energy sources (actuation), and control systems will be discussed, pointing out their advantages and disadvantages. Moreover, examples of exosuits (full-body, upper body and lower body devices) will be presented and described, including their use cases and outcomes. The future of exoskeletons as possible assisted movement solutions will be discussed - pointing to the best options towards rehabilitation.



Graphical abstract.

3.2 Introduction

Physical impairment limitations are still a common occurrence in today's society, despite the advancements in technology and science, and the implementation of new legislation in most countries, defining new rules for facilities. These physical impairments have many causes [1], such as the normal ageing process and increased average life expectancy [2, 3], neurodegenerative diseases [4], and daily accidents including falls, motor vehicle accidents or sports-related incidents [5].

3.2.1 Context and Demographics

It is clear that the average life expectancy of a nation is proportional to its level of development. Countries with higher quality of life (QOL) indices tend to have longer average lifespans than those with lower QOL indices. For example, Norway, Germany and Hong Kong all have an average lifespan of over 80 years whereas the Central African Republic and Nigeria both have an average lifespan below 54 years old. Therefore, countries should focus on improving their overall standard of living if they wish for their population to live longer lives beyond retirement age.

The ageing population is becoming a global phenomenon, mostly in developed countries. Statistics show that in the United States of America (USA), the number of people aged over 65 years old will be approximately 55 million in 2030 [6], similar to Europe, where the number of elderly people already represents almost 20% of the total population [7]. Complementary, surveys from United Nations (UN) reveal, that by 2050, around 20% of the global population will be over 60 years old [8] and from them, 1.5 billion over 65 years old [9]. These prevision numbers more than duplicate the 2015 and 2019 numbers, respectively [9, 10]. As an inherent consequence of an aged population, locomotion disorders became a reality for those people [11]. However, they are not aged exclusive results. Neurological pathologies, characterized by the progressive loss of structure and function of the central nervous system caused by neuron death, are also responsible for them. Without a healthy nervous system, sensory information (audition, vision, smell, tact and taste) and well as muscle coordination are compromised.

The global prevalence of dementia estimated by the UN points to more than 55 million people worldwide having some kind of dementia and 10 million new cases diagnosed each year [12–14]. Parkinson's disease (6.2 million in 2015 [15]) and Alzheimer's disease (60-70% of all cases [14]) are the two most common forms, along with amyotrophic lateral sclerosis. As these neurodegenerative disorders progress, they cause a gradual decline in patients' locomotion abilities, leading to death within 2 years for many patients [16]. Despite extensive research into treatments for these diseases, there is still no effective treatment available.

Spinal cord (SC) injuries are a major source of locomotion disorders. These changes in muscular strength or body functions can be either permanent or temporary and affect everyday activities such as walking and lifting a glass of water. These injuries can be caused by external traumas, such as car accidents (39.3%), falls (31.8%), gunshots (13.5%) or during sports (8%) [17]. They may also be caused by tumours (33-79%) [18], which compress the SC. However, depending on the SC damage extent, it is classified as complete (no messages are conveyed to body parts) and incomplete (some level of message transmission is still possible). Every year about 40 million people suffer from SC injuries, most of them between 20-35 years old [19].

In some scenarios, locomotion disorders are multi-factorial, making movement tasks much more challenging. An example, is the simultaneous occurrence of tumours and neurodegenerative disorders, since their risk increases with age [20]. All these adversities have a high and long-term impact on the social, economic and financial sphere, affecting communities and healthcare systems worldwide [3, 8].

Psychologically, locomotion problems may be a cause of stress, pain and depression since the simplest movements are no longer easily achievable. Besides, motion disorders also contribute to depressive states of mind, as they negatively impact a person's social life [6, 21, 22]. Assuming these bad feelings, mental illness is often found in people who are experiencing physical impairments, affecting not only the person but also family and friends.

Bedridden patients (or immobile patients) tend to develop a condition called "sarcopenia," which is the deterioration of muscle tissue that leads to immobility [23]. In addition, the risk of comorbidities, such as obesity, coronary heart diseases and diabetes increases [24].

Researchers, engineers and physicians, at universities, research institutes or companies [25], are working to address the consequences of ageing and injuries that affect human movement. Several solutions, such as wanderers, wheeled vehicles and wheelchairs [26], have been available for decades. These assistive technologies are meant to help the affected person regain some independence. However, these simple devices were not designed with rehabilitation in mind therefore, some exoskeletons were developed to fill that void [27].

3.2.2 Concept of an External Device

The concept of an exoskeleton has its roots in the natural world. Some animals, such as arthropods and molluscs, have a hard outer layer called an exoskeleton (distinct from the endoskeleton found inside the body of others), which serves to protect their bodies from the elements and provides a surface for muscle attachment and a barrier from dehydration, besides a sensory interface to the surrounding environment [28].

For humans, exoskeletons, which emerged in the 1950s [25], are systems that can expand or enhance a person's physical abilities [28]. These mechanical devices are fitted with powerful actuators at human joints, allowing for assisted movement [29]. Originally developed for military use, such as aiding soldiers with carrying heavier loads, running faster, jumping higher, or fighting better [28], exoskeletons are now being used and developed for different purposes, such as for medical applications (e.g. assisting physiotherapy [30]) and for industrial purposes.

Firefighters and other rescue workers have been using exosuits in their daily activities to help them carry heavier loads. Additionally, certain industrial companies have also been equipping their employees with passive external skeletons to help reduce fatigue and increase productivity [31]. Although the wide range of applications that exoskeletons and exosuits are already used for, they are still being actively developed and improved upon, as evidenced by the increasing number of publications on the subject. In 2014, the number of papers published on the topic was nearly double that of 1997 [32], demonstrating the remarkable progress being made in all aspects of exoskeleton and exosuit design.

3.3 External Devices in Rehabilitation Context

After a trauma or surgical procedure, continuous passive motion devices are typically used in rehabilitation to reduce oedema, bleeding, pain, and inflammation. These devices are the first step in the rehabilitation process. Active assistive movement is also used, which helps the patient perform desired movements with the help of a suit that assists in completing the movement. In cases of neurological rehabilitation, this method is the first choice to stimulate neuroplasticity and reduce common side effects such as muscle weakness. Active resistive motion involves applying an external resistive force against a dynamic or static muscle contraction and is an effective way to increase bone and muscle mass, making it essential for musculoskeletal rehabilitation. Exoskeleton usage can enhance the results of different physiotherapeutic approaches. Still, the final outcome depends on a range of rehabilitation factors, including timing, intensity, repetition, frequency, and task-specific training protocols [33].

Wearing an external device, such as an exoskeleton, can provide numerous advantages in a medical rehabilitation environment [34], not only for the patients but also for clinical centres. These devices can enable patients to perform intensive and repetitive movements with precision, minimizing the physiotherapist's intervention [35]. This can relieve therapists from fatigue and constant attention requirements. Additionally, this kind of technology can enable the rehabilitation of patients at their homes via video conference. Exoskeletons can also be used to evaluate recovery levels by measuring force levels and movement patterns [8]. This data can be collected from sensors [36] in the device itself and/or from motion capture devices that track motion patterns. This training can help people relearn lost motor functions and perform daily tasks.

The drawbacks of existing solutions should be object of careful consideration, taking into account the person and their particular circumstances. For example, some solutions may not be energy efficient, leading to high energy consumption [26], while others may make it difficult for the user to interact effectively with their surroundings [37].

When rehabilitating a patient using an exoskeleton, the need for a large empty room must be taken into account. Moreover, since a regular size cannot accommodate all users due to differences in body proportions, the creation of an adjustable device that can fit all sizes poses a great challenge due to its complexity. Thus, a disproportional device regarding the body may have a negative psychological impact on the user, leading to some reluctance in its use [38].

Despite all the challenges, researchers have already developed reliable solutions to rehabilitate or enhance various parts of the body, such as ankles [39–41], hands [33, 42], shoulders [43], lower limbs [44], upper limbs [45], arms [46], and back [47, 48].

3.3.1 Mechanical Design

Design is an imperative aspect to consider in exoskeleton development, as every detail affects user experience, with the final appearance being the first overall impression. During conception, several design considerations come into play during all stages of the project, from selecting structural materials to selecting control systems, with particular attention paid to key components such as batteries. An intelligent arrangement of actuators and energy sources (e.g. batteries) brings benefits beyond just aesthetics; it can improve weight distribution [27, 49] and in some cases even reduce power consumption

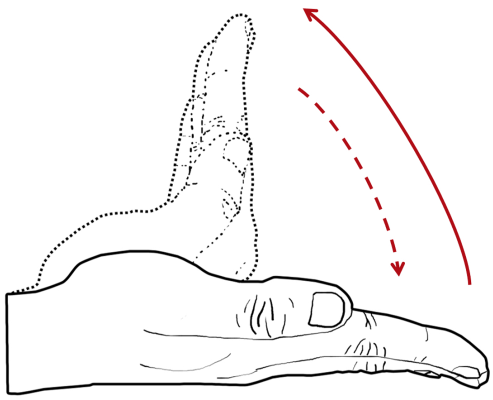
[50], which is directly linked to the choice of power source. Most importantly, a good design can feedback a positive first looking to the users, providing them with a sense of comfort, ergonomics, confidence, and convenience.

In addition to visual appearance, it is crucial to consider the technical aspects when designing the final solution. The movement's kinematic and dynamic degrees of freedom (DOF) found in the human body [51] based on anthropometry should be present, as a concept, throughout the projects. The range of motion, joint torque requirements, joint rotational velocity, and joint angular bandwidth [52] must also be factored in. The developed device aims to aid and follow human movement without constraint or interference with the natural freedom of movement [53].

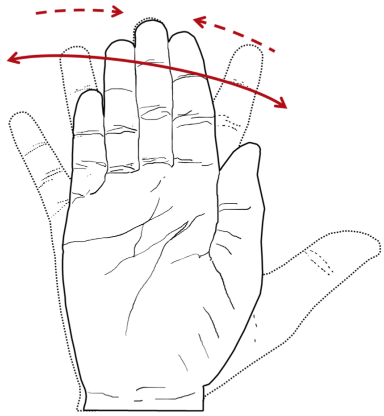
Based on the above-stated principles and keeping in mind the intended purpose, the wearable device should enable fundamental body movements, as described in [54]. These movements involve pairs of opposite gestures, such as *flexion* and *extension* of hand movements (depicted in Figure 3.1a), or *abduction* and *adduction* of the fingers (illustrated in Figure 3.1b). Additionally, *rotation* (medial or lateral), as shown by elbow rotation (Figure 3.1c), is another basic movement. By combining these basic movements, a person can perform complex movements like writing.

Creating and implementing practical solutions can be a challenging task due to the inherent complexity of the principles involved and their combination. A specific example of this complexity can be observed in the development of complete limb external skeletons. These particular devices are capable of an infinite number of combined movements, as they rely on seven distinct DOF [50] positioned along the limbs. These DOFs are vital for daily activities [55], with lower limbs having three DOFs at the hip, one at the knee, and three at the ankle, while upper limbs possess three at the shoulder (abduction-adduction, flexion-extension, and internal-external rotation), one at the elbow, one at the forearm, and two at the wrist.

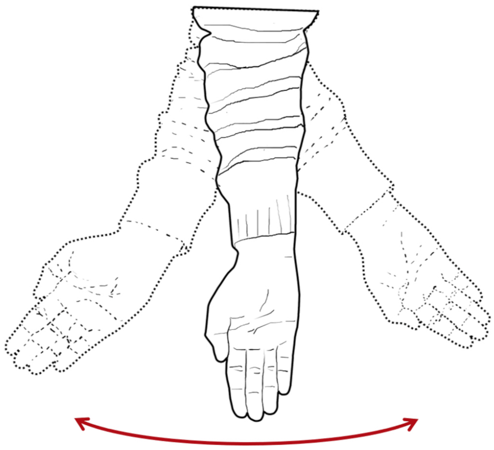
Providing the necessary DOFs for full-body applications becomes challenging with traditional exoskeletons, which often consist of rigid materials assembled in a series of



(a) Flexion (full line) and extension (dash line) movements.



(b) Adduction (dash line) and abduction (full line) movements.



(c) Arm rotation movement.

(d) Combined movements.

Figure 3.1: Basic human hand movements [54] and their combination towards complex movement.

fixed links. Their non-flexible characteristics can lead to problems of hyperstaticity [56] and can result in increased device complexity, which further complicates the design process. As an alternative, soft structures composed of mechanisms without rigid components, featuring elastic or elastomeric materials with softer (more flexible) mechanical properties, have emerged. As demonstrated by successful lightweight and flexible designs [57] and greater adaptability to both movement and the human body [58], they offer a promising alternative to their traditional rigid counterparts.

The primary function of the wearable device is not just to track human movement, but also to provide assistance by generating the necessary force or moment to hold the joints (e.g. elbow) in certain positions during daily activities or rehabilitation. Moreover, as per [59] guidelines, the device should be capable of generating the appropriate amount of auxiliary force or momentum to perform those daily tasks. However, it may be impossible to devise a solution that combines all the necessary DOFs with adequate motion generation, as illustrated by the challenge of creating a wearable finger device. This body limb is essential for performing basic daily tasks, such as typing or writing, and any solution must be practical and effective in addressing these needs. For example, opening a jar with a finger wearable device would require up to 120 N of force and 3 Nm of torque on the metacarpophalangeal (MCP) joint [60], (Figure 3.2), without neglecting other considerations such as overall aesthetic and having 4 DOFs [61, 62].

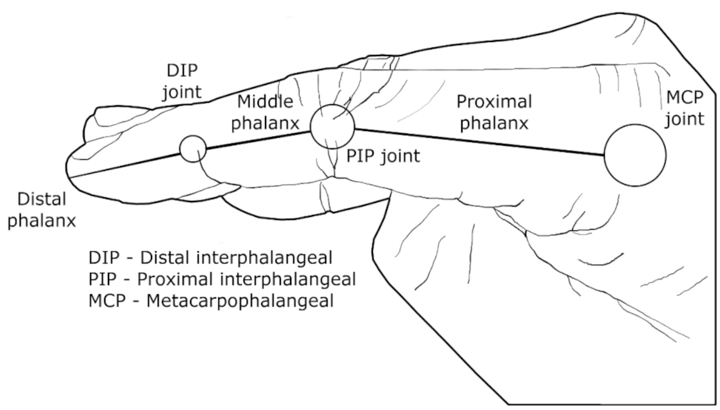


Figure 3.2: Finger joints and phalanges - the 4 DOF of a finger wearable [60].

The project must prioritize security measures, as an equal fundamental design factor. Following these safety standards avoid accidents and ensure the user's protection in unforeseen events, like power loss or current leaks. The probability of accidents using exoskeletons is real and remains a significant concern, since estimations suggest around 4 out of every 100 users may encounter issues [63]. The overall solution's appearance, functionality, safety, and ease of use are determined by the final design concept, which cannot disconnect design choices from fundamental design variables and options. Consequently, the final solution must represent a balance between design options, structural materials, actuators, energy sources, and control systems to achieve the best overall solution.

3.3.2 Structural Materials

While the terms exoskeleton and exosuit have often been used interchangeably, some argue that *exosuit* is a more accurate description for these devices albeit the general public is more acquainted with *exoskeleton*. Despite their similarities, those terms are not synonyms when the context involves structural materials. In reality, these words represent two distinct approaches for solving the same problem. Exoskeletons are typically constructed of rigid and metallic components [64, 65], while exosuits are designed using soft and flexible materials [66, 67]. Although they are classified differently, both solutions should be investigated together as they offer complementary features [68]. Regardless of the type of material used, it is essential that any solution designed for use in rehabilitation settings meets certain critical requirements that ensure safety, as mentioned in a study by Xiloyannis [49]. In particular, mechanical properties assume great importance since patients undergoing rehabilitation are often susceptible to minor accidents, such as small falls, and the material should be able to withstand and resist fatigue, as pointed out by Bogue [69]. These characteristics are vital for ensuring that the device has a long lifespan, even when deployed on a higher number of patients during rehabilitation. Additionally, the material should offer a warm and comfortable sensation to the wearer.

Rigid Vs Soft Materials

When it comes to a rigid approach, materials like stainless steel [69], aluminium [70], and titanium [65] are widely used. The final solution can involve one or multiple materials for example, with frames made from aluminium and joints made from stainless steel or titanium. This multi-material approach can offer several benefits, such as reducing weight and increasing mechanical strength at critical joints. In fact, using multiple materials is becoming popular in engineering because it provides a better balance between performance, cost, and durability.

Compared to exosuits, more rigid solutions offer some advantages but bring some disadvantages. Exoskeletons offer increased mechanical strength, making them an ideal solution when high levels of torque and strength are required. In fact, these devices can withstand up to 1 GPa of tension before experiencing plastic deformation and can endure up to 50% of strain before reaching a breakdown point, as shown in Figure 3.3 [71]. Such impressive performance metrics highlight the potential benefits of using exoskeletons in various settings.

However, the materials used are typically heavier, as shown in Figure 3.4a, which can limit their portability and cause discomfort for the user [72]. Additionally, achieving perfect alignment between the device and the user's joints can be a challenge, resulting in larger inertial loads that can lead to abnormal motion patterns [73]. Other common problems associated with rigid solutions include reduced usability and poor aesthetics, as noted by several authors [74, 75]. Despite these drawbacks, rigid solutions remain popular in many applications due to their mechanical reliability and stability.

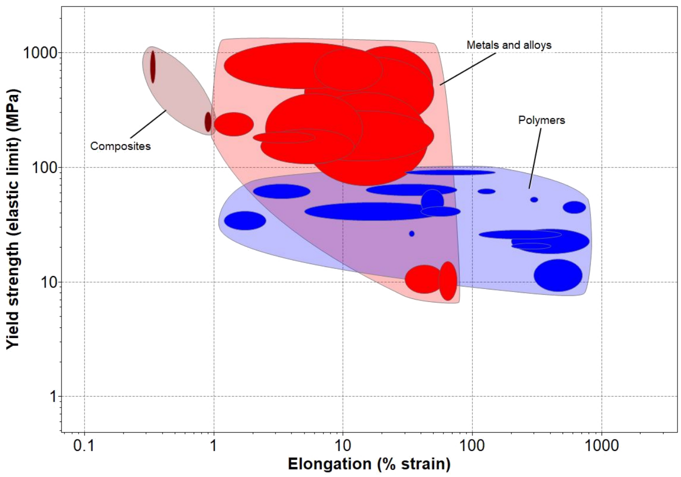
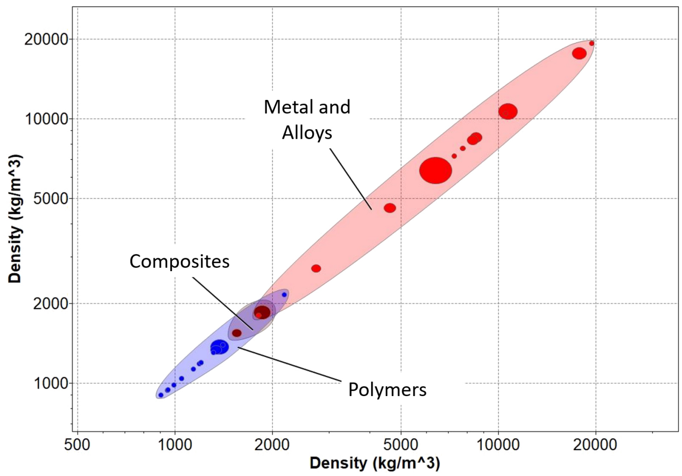
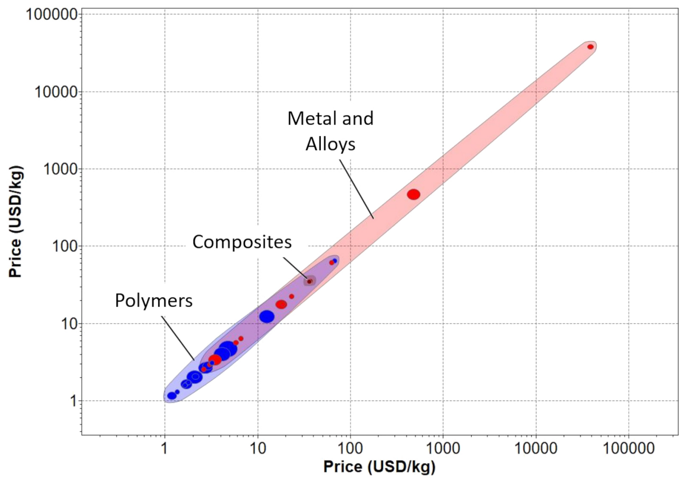


Figure 3.3: Yield strength (elastic limit) Vs Elongation of traditional and soft materials, Granta Edupack 2020 [71].

While exosuits and exoskeletons share some common characteristics, such as safety features (when applied in these devices) and price range (as depicted in Figure 3.4b), there



(a) Material density of traditional and soft materials.



(b) Price of traditional and soft materials.

Figure 3.4: Important considerations regarding traditional Vs soft materials, Granta Edu-Pack 2020 [71].

are clear differences in their design and construction. Exosuits typically have symmetric properties not being susceptible to misalignment, largely due to the materials used in their production. These materials primarily consist of polymeric or composite materials, including elastomers such as liquid crystal, dielectric, and acrylic elastomers [66, 68]; shape memory polymers (SMPs) such as those based on epoxy and polycaprolactone materials [66]; electroactive polymers (EAPs) like polyvinylidene difluoride (PVDF) [76, 77]; and conducting polymers such as polypyrrole [78, 79]. Their use in exosuits allows for greater flexibility and symmetry compared to their rigid exoskeleton counterparts.

Composite materials can be comprised of metallic and polymeric substances combined with carbon fibres [57, 69]. In some other cases, a solution made from chloroprene and polyurethane (PU) may also be used [57]. Additionally, textiles may also be utilized for certain applications [73, 79]. By combining these materials, it becomes possible to create lightweight and durable devices that can provide users with a wide range of benefits.

In general, these materials enable movement smoothness [80], comfort, portability, flexibility, lightweight (low density) [68, 73], adaptation to bioorganisms [66] and even the ability to emulate biological muscles [77]. Some of these materials can exceed their structural role and be used as actuators [77] since they are prone to deformations with associated large volume changes in response to external stimuli [66].

However, this approach presents some technical disadvantages. The amount of power that such actuators can transmit and their response in velocity, are highly diminished when compared to rigid solutions due to their (softer) mechanical properties [68]. Therefore, they are specially indicated for small assistance levels [68].

While rigid and flexible materials used in exoskeletons and exosuits possess distinctive properties, the most effective solutions typically involve a combination of both types of materials. Table 3.1 provides some essential information regarding the characteristics of these different materials and how they can complement each other. By leveraging the unique advantages of both rigid and flexible materials, it becomes possible to create devices that are both durable and comfortable, allowing users to benefit from the qualities of each material type.

3.3.3 Actuators and Energy Sources

Actuators play a critical role in wearable external devices, facilitating human movement by powering them and in this way enabling a better interaction with the surrounding environment. In a medical context, they can be particularly valuable for helping patients

Table 3.1: Advantages and disadvantages of Rigid Vs Soft materials.

Type of materials		Advantages	Disadvantages	References
Rigid	Aluminium Stainless steel Titanium	Higher mechanical strength Higher elastic limit Higher safety	Higher weight Diminished ergonomy and comfort Larger inertias Unnatural motion patterns Lead to higher power consumption	[65, 69–75]
Soft	Polymers Composites (e.g. SMPs, EAPs)	Safer Allow smoother movements Higher comfort Higher portability and flexibility Lightweight Biomimetic Accommodate large deformations Possible use as actuators Easy to process and mass produce	Lower yield strength Actuators with lower force/torque and velocity Adequate for smaller assistance levels	[66, 68, 76–78] [57, 69, 73, 79, 80]

undergoing rehabilitation by providing controlled motion patterns. As such, actuators are an indispensable component of many modern wearable devices, and their effectiveness can have a significant impact on user outcomes.

Actuators can be classified as either powered or unpowered, resulting in the creation of either active or passive external devices [81], respectively. Powered alternatives may be noisier and are generally costlier due to the need for additional components, as well as requiring users to carry bulky energy-supply systems [82]. On the other hand, passive devices do not require power units, making them lighter and weighing up to a fourth of their powered counterparts. A good example of this is the ankle exoskeleton developed by Mooney et al. [83] and Collins et al. [84], which aims to reduce the metabolic rate during walking [85].

Mooney et al. [83] achieved a weight of 2kg in their solution, while Collins et al. [44, 84] proposed an unpowered solution that was 1.5kg lighter and cheaper. The essential difference between these two approaches is the presence of either an autonomous system capable of producing its own energy, thus replacing human metabolic sources [83], or a passive system that makes body locomotion more efficient by reusing some of the energy already produced by the body [84]. Also, actuators can be categorized as either traditional or soft, depending on their constituent materials and energy-supply system type (see Table 3.2). Each alternative exhibits different advantages and disadvantages depending on their intended use.

Traditional Actuators

Traditional actuators typically are based on rigid systems, allowing them to generate higher forces [49], greater movement precision, and improved dynamic performance [68], and as a result, making them ideal for more complex tasks such as severe mobility disorders. However, it brings some disadvantages, such as leading to higher power consumption [68]. When a power supply is required to input the actuator, the user's freedom of movement can be limited. Additionally, elderly users may feel uncomfortable with the robotic aspect of the actuators, which can convey a detached and cold sensation and lead to their refusal to use the device.

Purely mechanical actuators such as springs [86], are commonly used in unpowered devices (which do not require any external source of energy) and convert the tension force from the actuators into torques at the joints [44]. This mechanical solution can help to reduce the metabolic consumption of energy [84] during walking or running activities [87]. However, the usability of such actuators has limited usefulness in rehabilitation cases, as they only provide passive assistance. For example, during walking, the user must first tense the actuator during flexion movement in order to receive assistance in the extension movement.

Mechanical servomotor-based actuators [88] are a simple and direct approach for achieving actuation through electrical stimulation. They provide motion and assistance when connected to the structural material (soft or rigid). However, due to the nature of the input type, they always require an external source of electrical energy, such as (portable) batteries. Plus, they are also rigid and bulky, which can limit the flexibility of the entire system [89].

Pneumatic-based actuators are a highly efficient and safe solution in terms of linear and rotational movement control since the actuator's motion is converted from pressurized air energy [89]. Also, they are particularly suitable for applications that demand repetitive opening and closing tasks, as well as in environments of extreme temperatures or even in industrial applications where other types of actuators are not viable alternatives. As air-compressed-based actuators, this type of solution is able to convert up to 6 bar of pressure into movement, if necessary. However, to perform all of this and enable movement, a connectivity to a rigid control and power system, such as a compressor, is a mandatory aspect requirement [89], which can occasionally lead to pressure drops and noise. Moreover, pneumatic actuators could be produced either considering rigid [90] or soft materials, such as latex or rubber tubes [91], which make them a feasible solution for exoskeletons [90] and exosuits [91].

Hydraulic actuators [92, 93] share similar advantages and disadvantages when compared to pneumatic actuators. Similarly, they require a hydraulic fluid to output linear, rotary or even oscillatory movements by the actuator, but as liquids are nearly incompressible, the force produced is considerably higher. The exoskeleton/exosuit movement is thus achieved by converting hydraulic into mechanical energy.

Soft Actuators

Actuation solutions based on soft actuators can be a comfortable alternative when used during the rehabilitation process [94] and unlike the traditional methods, they can be stimulated externally by different inputs. The direct incidence of light, heat, electric or magnetic fields results in mechanical movement performed by the actuators [77, 89]. They can be thus defined as mechanical and electrical elements whose output/operation varies under different physical, chemical and/or biological stimuli. Typically, these soft actuators can be built using a different range of materials (see Figure 3.5), from particles to polymers, such as EAPs [77] or SMPs [95], papers [89], fluids, shape memory alloys (SMAs) [89], hydrogels, liquid materials [66], 2D materials, carbon-based materials [66] or combinations thereof [89]. Despite the numerous alternatives, not all of these soft actuators are viable for rehabilitation cases. The pertinence of their applicability is based on performance parameters such as stress, strain, Young's modulus, power, energy, and force density [89].

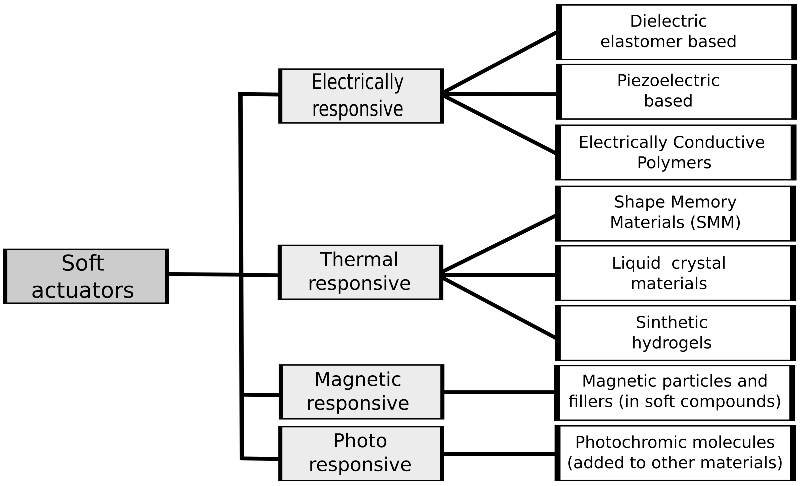


Figure 3.5: Soft actuators, responsiveness to stimuli and base materials.

Electrically responsive soft materials are flexible and stretchable materials able to convert external electric inputs into mechanical response outputs. Depending on the type of material, they are classified as dielectric elastomer actuators (DEAs), piezoelectric-based actuators, and electrically conducting polymers (ECPs). DEAs have their input-output conversion based on Coulombic attraction. Two flexible electrodes with a potential difference located on separate ends of a compressible membrane are used to obtain the mechanical response from DEAs [89]. They are highly flexible materials with high energy density, strains, and the ability to emulate the behaviour of biological muscles [96]. The performance of these materials depends on their stability, breakdown voltage, and dielectric constant of them [89]. However, they generally require high voltages, usually in the kV range, to perform and leakage currents are often observed when high electric fields are applied, especially when the actuator ages [89]. Adding liquid elastomers to DEAs has been proposed as a solution for these limitations [89]. Examples of dielectric materials found in the literature include acrylic elastomers [97], which are highly deformable and possess high viscoelasticity. However, the actuator's bandwidth could be limited due to these mechanical properties [89]. Other examples include silicone-based materials [98] and PU-based elastomers [99]. The PU-based elastomers have faster reactions and can be cast into various shapes, but they perform significantly lower strains than the dielectric materials [89]. DEA solutions are constantly being researched and developed to enhance their properties in the actuation field [100].

Piezoelectric-based actuators are capable of producing voltage or electric charge in the presence of mechanical or vibrational forces (direct effect) or deformation when electrically stimulated (indirect effect) [77, 101]. These actuators can operate in room conditions for long periods and have a quick response time, typically in the milliseconds range. Also, they can hold strain under activation, inducing relatively large actuation forces [77]. However, their usability in real-world scenarios can be limited by the large AC voltages required [89]. Common piezoelectric materials for actuation and sensors include PVDF and its copolymers [102, 103], graphene [104], and zirconate titanate [105], among many others [106]. ECPs [107] are organic polymeric materials obtained by reduction or oxidation reactions [108]. They have the ability to conduct electricity with conductivities up to 10^5 S/cm, achieved through traditional sources, such as batteries or chemical reactions. Moreover, these electrically responsive materials types have been powered using biofuels, such as glucose, which shows their potential as an environmentally friendly source of energy [109]. Polypyrrole, a type of ECP obtained by the oxidative polymerization of pyrrole, is characterized by high mechanical properties and chemical stability [108] and has been shown to emulate human biological muscles due to its similar behaviour and low voltage operability [109, 110]. These characteristics make ECPs an interesting choice due to their biomimetic and biocompatible nature.

Magnetic responsive materials have potential applications as actuators since they are easily controllable through magnetic field direction and magnitude, which can penetrate most materials [89]. This feature makes them a promising solution for use in restricted or enclosed areas [111]. This actuation method is based on incorporating magnetic particles and fillers into different soft compounds such as polymers, gels, papers, or fluids [111]. This results in a magnetization profile with variable magnitude and direction [112]. In the presence of a magnetic field, the particles or fillers align to create deformation, bending, elongation, or contraction [89]. These magnetic-based actuators have a fast response time, with literature reporting speeds of up to 100 Hz [113]. However, there are some disadvantages associated with the magnetic coils used to generate magnetic fields. Their large size, high energy consumption and limited control areas where the magnetic field may not be strong enough are some handicaps to consider [89].

Thermally responsive materials, including silicone-based elastomer materials [114], liquid crystal elastomers, and synthetic hydrogels [89], can be activated by a thermal source, such as infrared (IR) radiation, thermal radiation, or Joule heating [115]. For instance, shape-memory materials (SMM) [116] can be deformed by external forces and return to their original "memorized" shape under loading or thermal cycles [89]. These materials include SMAs (typically iron-based or copper-based) [117], which return to their original shape when the temperature exceeds a certain threshold after deformation, and SMP materials (PU and thermoplastic PU) [118]. SMPs are cost-effective, have high elastic deformation, and are easy to manufacture [89]. Furthermore, they can be activated remotely, for instance, through laser incidence and are often safer than electrical fields for biomedical applications [89]. Some of these light actuators are capable of lifting objects that are up to 200 times more substantial than their own weight, to up to 5mm height [115]. However, such thermally responsive materials tend to have slower response times and are less efficient compared to other types of stimuli-based actuators [89].

Photo-responsive materials employ photochromic molecules to capture optical signals and convert them into property modifications [119, 120]. They represent an attractive wireless alternative, as they can be controlled in small sizes and consume low energy [89]. However, slow actuation speed and mechanical property degradation remain major limitations [89]. Photochromic molecules, such as spiropyran [119], may be added to various materials, such as gels, polymers, and fluids, to render them photoresponsive [89]. They respond to the light spectrum, visible or near-IR) [89].

3.3.4 Control

The majority of external skeleton or suit devices can be analyzed from two distinct perspectives: mechanical and control system, with the former including structural materials, actuators, and sources of energy, and the latter including sensors that ensure interconnection between the device and the user [121]. The control system's mission is to predict human intention, interpret signals captured by sensors, and send input to actuators, thereby allowing the skeleton to operate in parallel with the human body [122]. In passive devices that lack powered systems, a control system is unnecessary [123]. Refer to Figure 3.6 for a depiction of the control solution.

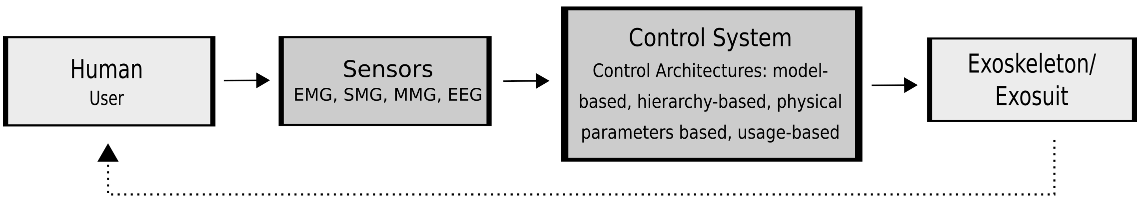


Figure 3.6: Control system overview.

Control System Architectures

The control system of external skeleton or suit devices can be categorized into four main architectures - model-based, hierarchy-based, physical parameters based, and usage-based, as shown in Figure 3.7 [121]. While none of these architectures have been used individually due to their complexity or effectiveness, they are frequently combined to achieve the desired control of a specific device [121].

In general, **model-based control systems** can be further classified into two types - dynamic and muscular models [124]. The dynamic model reflects the human motion intent by combining inertial, gravitational, Coriolis, and centrifugal effects to model the human

Table 3.2: Advantages and disadvantages of traditional and soft actuators.

Type of actuators		Energy Source			Advantages	References
Traditional actuators	Purely mechanical actuators	Unpowered			No need of an external source of energy Allow to reduce metabolic consumption	[44, 84, 86, 87]
Traditional actuators	Mechanical servomotor-based actuators	Powered electrical input			High efficiency of power conversion Quiet, clean and create no pollution Less expensive Easy of maintenance Easy to implement the remote controllable system No limitation of separation between energy source and system	[88, 89]
Traditional actuators	Pneumatic actuators	Powered compressed gas			Affordable Fast working cycle Insensitive to temperature drift No need for mechanical transmission High actuating forces	[89–91]
Traditional actuators	Hydraulic actuators	Powered compressed fluid			High stability High Stroking velocity Suitable for high loads High actuating force Stiff and incompressible source	[92, 93]
Soft actuators	Electrical responsive actuators	Powered electrical stimulus	Dielectric actuator		Soft, flexible and stretchable Scalable High power-to-weight ratio Stores and recovers kinetic energy	[89, 96–100]
Soft actuators	Electrical responsive actuators	Powered electrical stimulus	Piezoelectric actuator		Suitable for high force applications Large operation bandwidth	[77, 101–106]
Soft actuators	Electrical responsive actuators	Powered electrical stimulus	Conducting polymers		Possibility of being feed through biofuels Processability Good biological muscles emulation	[107–110]
Soft actuators	Magnetic responsive actuators	Powered magnetic stimulus			Linear effect Quick response Capacity to penetrate most materials	[89, 111–113]
Soft actuators	Thermal responsive actuators	Powered thermal stimulus	SMM	SMPs	Low cost Biodegradable Low density High elastic deformable Sustain broad range of temperature drift	[89, 115, 118]
Soft actuators	Thermal responsive actuators	Powered thermal stimulus	SMM	SMAs	Flexible in nature High energy density Low actuation temperature Provides large frequency response	[114, 116, 117]
Soft actuators	Photo responsive actuators	Powered light stimulus			Environmental-friendly Full possibility of remote controlling Ease to control the response Excelent resolution	[89, 119, 120]

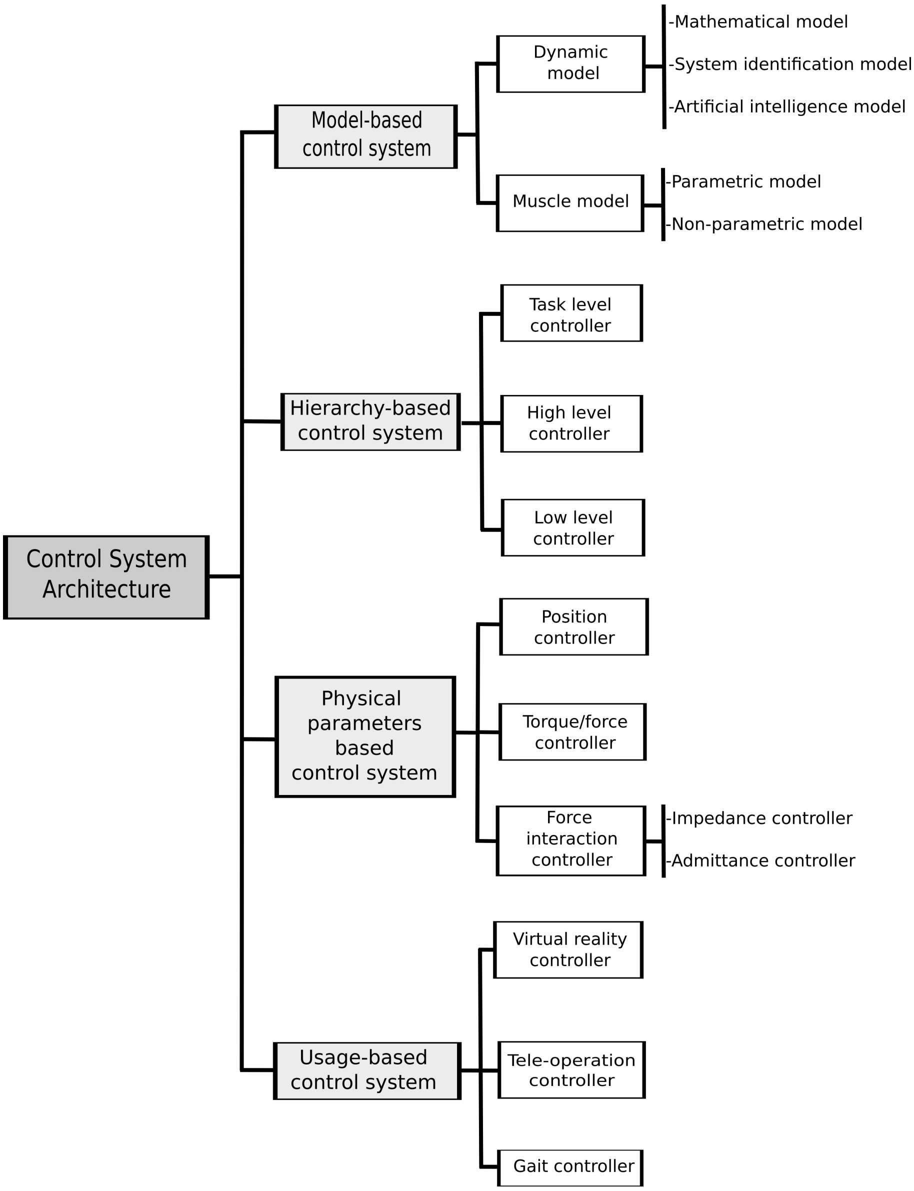


Figure 3.7: Typical control system architectures [121].

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body as a series of rigid links connected by joints (bones) [125, 126]. The control system of BLEEX is just an example of a dynamic model-based system [50]. This based-type architecture is even developed through different approaches: mathematical, system identification and artificial intelligence models. To obtain a mathematical architecture for the external device based on physical characteristics of the system, the system requires a precise dynamic model [121]. For instances in which a dynamic model cannot be adequately developed through theoretical mathematical models, the system identification model is often utilized [127]. The artificial intelligence method is the most popular approach to identifying the dynamic model due to its efficiency [127].

Muscle-based models have also been utilized in exoskeleton control systems. Unlike dynamic models, these models predict the muscle forces generated by human joints as a function of muscle neural activities and joint kinematics [126, 128]. This approach, which can be obtained by using parametric or non-parametric models, takes the electrical signal produced by muscles as input and sends force estimation as output to actuators [121]. The parametric muscle model is commonly implemented using the Hill-based model, which refers to muscle contraction and uses the estimated muscle activation level [129–131]. It is comprised of three elements: a contractile element, representing force generated by active muscle fibres; a series element, which models the mechanical response of the muscle; and a parallel element which simulates the passive resistance of muscles to stretch [131]. In addition, the output sent from this type of control model is a function of electromyographic (EMG) neural activity and muscle length [121]. In contrast, non-parametric muscle models, do not require knowledge about muscle and joint dynamics but they can be the source of control inefficiencies [132] (ex. finite impulse response model).

Shafer et al [133] developed an ankle exoskeleton controller that uses a control system based on a neuromuscular model. They conclude on the effectiveness of their model in providing a wide range of assistance torque and power. Moreover, Song et al [134] developed a novel model-based control to predict motion trajectories and amplify the forces produced by the user.

The **Hierarchy** based control system, exemplified in Huang et al [135] and Dinh et al [136], utilizes a hierarchical structure to manage inputs and outputs. The controllers are divided into three levels: task level, high level, and low level. The task level controller, which is the highest level, is responsible for performing the designated tasks [121]. The high-level controller adjusts the force of human-external device interaction based on information received from the task-level controller [121]. Finally, the low-level controller is responsible for controlling the position and/or force performed by the exoskeleton joints, therefore contacting directly to the exosuit [121].

Copaci et al [137] implemented a hierarchy-based control system in an elbow exoskeleton. Using algorithms to process EMG signals, they were capable of generating position and torque references in SMA actuators used for active rehabilitation therapies.

Control strategies such as those utilized in the ARMin [138], RUPERT IV [139], and LOPES [140] exoskeletons use **physical parameters** as a basis for their implementation. These solutions can be classified as either position, torque/force, or force interaction controllers [121]. The low level controller in the position control scheme ensures that the exoskeleton joints turn to the desired angle, while the torque/force controller regulates the desired force and/or torque [141], and is also classified as a low level controller [121].

The interaction force controller, typically functioning as a high level controller, is responsible for providing appropriate assistance to users during a task [121]. This physical parameter controller takes into consideration the force interaction between the user and the exoskeleton, which is considered in an external device [121]. The impedance controller, which accepts position and produces force, or the admittance controller, which accepts force and yields position, can be used to control this physical parameter controller [142].

The impedance controller is typically more effective for lightweight, backdrivable external devices (such as cable-driven devices) compared to other controllers [124]. It extends the position control, enabling it to not only regulate the position and force but also the relationship and interaction between the exosuit and the human body [142, 143]. This controller architecture includes an impedance module, which receives the error position of the joints and yields the force values that serve as force references for subsequent stages. The architecture also comprises a force/torque controller that attempts to ensure that the forces exerted by the exoskeleton actuators are approximately equal to force references [121].

The admittance controller is employed to regulate the force generated by the external skeleton during interaction with the user [144]. It features an admittance model, which receives forces and outputs the position, as well as a position controller that controls the joint angle based on position references from the admittance model output [121].

Wu et al [145] implemented a physical parameter-based control system in an exoskeleton for upper limb rehabilitation of disabled patients. They used a modified sliding mode control strategy incorporating a proportional integral derivative (PID) sliding surface and a fuzzy hitting control law to ensure a robust and optimal position control performance. Their approach led to best control performances in terms of tracking accuracy, response speed and robustness against external disturbances.

The **usage-based control systems**, such as those implemented in MGA [146] and L-Exos [147], can be categorized into three types: virtual reality (VR) controller, teleoperation controller [121], and gait controller, which is commonly used in lower limb solutions [140]. VR controllers are commonly employed in rehabilitation exercises for upper limb exoskeletons [148]. They allow for the guidance and assistance of patients during tasks such as moving a virtual object with their hands [139], virtually painting a wall [146], or carrying out constrained motion tasks [147]. In these applications, the exoskeleton/exosuit can be regarded as a haptic device [121].

The teleoperation controller is a form of master-slave controller, where the exoskeleton worn by the user is commonly used as the master type and a mirror robot serves as the slave [121]. In this configuration, interaction control occurs between the slave robot and the environment, as opposed to the typical interaction between the user and the exoskeleton [121]. Rahman et al. [149] implemented a teleoperation controller in an exoskeleton for rehabilitation and passive arm movement assistance (MARSE-4), constituted by an upper-limb prototype and a master exoskeleton arm (mExoArm). While mExoArm is operated by the patient, the upper-limb prototype mirrors the movement.

Liu et al [150] developed and implemented a novel systematic and algorithm of gait control based for energy efficiency. Their ultimate goal was effectively to reduce the high energy consumption of devices.

Sensors

Capturing human motion intents for external device control is a major challenge, which can be addressed through the use of sensors associated with both the control system and the device [151]. These sensors capture the user's movement intention as an input signal to the control system, which then provides output to the exosuit to perform the intended move. To ensure success, it is crucial that this input signal is precise and accurate. In addition to the intention-prediction instrumentation, other sensors such as inertial measurement units [152] (e.g., gyroscopes [153] and accelerometers [154]) or mechanical sensors [155] can be employed to measure or evaluate the output movement. However, it should be noted that these sensors are unable to predict movement beforehand [156].

Several control methods have been proposed to detect human intention through human-robot interaction dynamics, which could effectively assist able-bodied human subjects [157, 158]. While control methods using human-robot interaction dynamics are effective in assisting able-bodied humans, they may not always be suitable as the user needs to produce sufficient torque at joints to initiate movement. If this amount of torque is not generated, the device may not be effectively controlled, resulting in a problematic aspect for elderly or severely disabled individuals [159]. The ideal solution for human-robot interaction entails the prediction of movement intention, instead of a reaction to a precursor movement. This approach can improve performance in scenarios where generating sufficient torque is not possible [122].

To predict human movement, electrophysiological signals from proteins, organs, or muscles can be captured through sensors measuring voltage changes or electric current [160, 161]. **EMG sensors** (intramuscular [162], surface [163]) can measure small electrical signals [164] produced by muscle contraction, and have been successfully used in exoskeleton control [165, 166]. EMG-based methods can capture the user's intention to control the device, even if the person cannot produce sufficient joint torques or execute a particular movement [122]. However, the signal measured by EMG sensors might be biased by various factors, such as muscle crosstalk susceptibility [151], skin condition (surface sensors), muscle fatigue [156], or the inaccessibility of deep muscle fibres [167].

Besides the use of EMG sensors, there are other sensors that can be considered as alternatives for measuring muscle electrical activity. One such alternative is **mechanomyography** (MMG) sensors which are less sensitive to skin conditions compared to EMG sensors [156, 168]. These sensors measure the signal produced by muscles with respect to the gross lateral muscle movements which causes low-frequency vibration during contraction, lateral vibrations at the muscle's resonant frequency, and volumes introduced by the changes in the muscles [169]. Despite the advantages, MMG sensors have some disadvantages such as being affected by muscle fatigue as well [156]. **Sonomyography** (SMG) sets up another possibility to predict the user's movement intention by measuring muscle thickness and tracking skeletal muscle deformation from superficial to deep tissue [170, 171]. SMG sensors are also capable of classifying several motions and predicting joint kinetics during dynamic activities, such as those in the wrist [171, 172]. However, muscle fatigue is still a common issue with SMG sensors [156]. Figure 3.8 synthesizes the way these three techniques work.

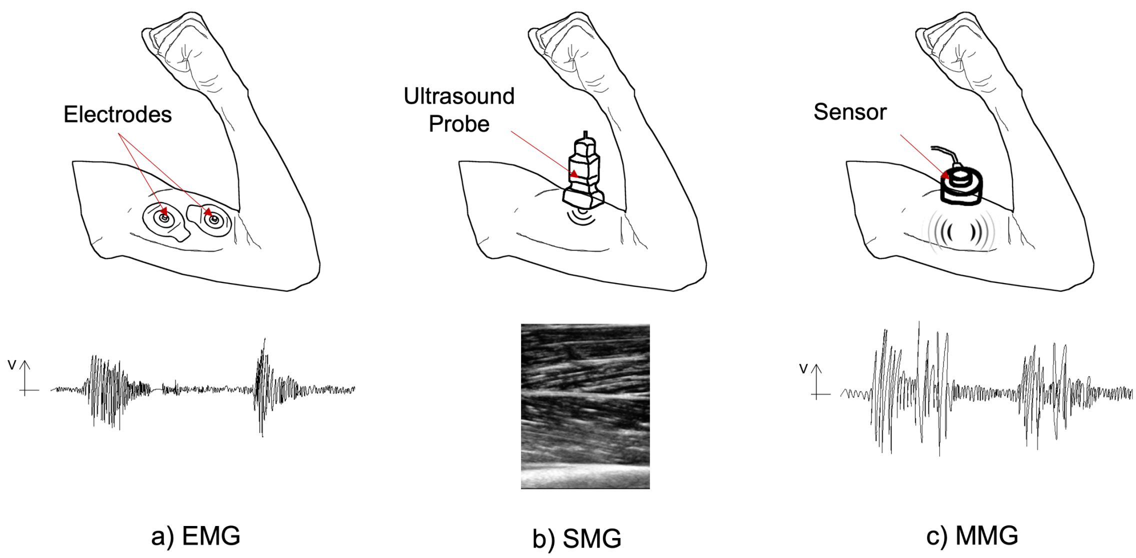

Figure 3.8: Sensors on muscles and respective outputs (SMG output [173]).

Finally, **Electroencephalogram** (EEG) sensors can capture the user's intention without using sensors that measure the signal produced directly in muscles [156, 174]. Instead, they measure the electrical activity in the brain. However, the signal captured by EEG sensors is not accurate enough and can only be used for classifying movements [156]. Table 3.3 summarizes the sensors advantages and disadvantages.

All the above-mentioned possibilities capture analogue signals, which need to be further converted into digital signals before being sent as input to actuators. This conversion can be done with affordable solutions such as an Arduino [175] and with commercial solutions already developed, such as BITalino from pluX [176, 177] or TMSi products [178, 179].

Table 3.3: Sensors, their advantages and disadvantages.

Sensors		Advantages	Disadvantages	References
EMG	Measures the electrical signals from the muscle contraction	Predict movement intention even if with any movement performed Already tested	Biasable by muscle crosstalk susceptibility, skin conditions, muscle fatigue	[162–165] [122, 166] [151, 156, 167]
MMG	Measures vibration and volume by changes in muscles	Less sensitive to skin conditions	Biasable by muscle fatigue	[156, 168, 169]
SMG	Measures thickness and deformation of muscles	Able to classify several motions and predict joint kinetics during dynamic activities	Biasable by muscle fatigue	[156, 170–172]
EEG	Measures electrical activity in the brain	No need of sensors in the muscles	Not enough accuracy	[156, 174]

3.4 Device Solutions

There are currently various exoskeleton and exosuit solutions available, not only described in the literature but also available commercially, such as Rewalk [180], Ekso [181], Cyberdyne [182], RB3D [183], and others. These solutions have been designed as powered or passive wearable devices that can assist individuals in daily living activities, including walking assistance. However, the present review will only focus on solutions discussed in literature specifically related to ankle/foot and hand/arm examples.

The structural materials used, actuation systems, control approaches, implementation, and results achieved will be described in detail.

3.4.1 Ankle/Foot Solutions

An external device known as an ankle-foot orthosis (AFO) is commonly prescribed to treat ankle impairments [184] while also helping to facilitate walking, which is essential to daily living routines. The use of an AFO has also been shown to reduce the metabolic cost of movement while rehabilitating weak ankles and feet [184]. Patients with ankle disabilities typically experience weakness in the muscles associated with plantar flexion and/or dorsiflexion movements, as illustrated in Figure 3.9. Debility in the gastrocnemius, soleus, and plantaris muscles, which are involved in plantar flexion movement, may reduce the push-off power necessary to propel the body forward during the stance phase [184]. Additionally, weakness in the tibialis anterior muscle, which is involved in dorsiflexion movement, may result in a drop-foot gait during the swing phase due to an inability to adequately lift the toes [184].

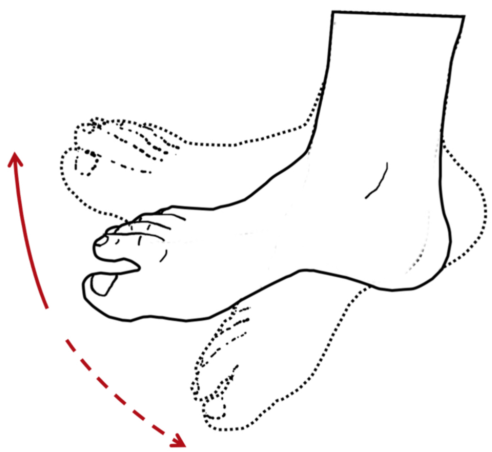

Figure 3.9: Dorsiflexion (full line) and plantar flexion (dash line) movements.

Various procedures, including surgical, therapeutic, and orthotic, can be used to treat ankle impairments. Recently, orthotic procedures have become the most commonly used [184]. In such cases, the device must be attached to the wearer and aligned with their ankle and foot to assist weak or paralyzed muscles by generating torques or forces [184]. Assistance can be provided using passive, semi-passive, or active AFO exosuits, depending on the availability of an energy source [184]. For example, Yamamoto et al. [185] and Ramsey et al. [186] have developed passive devices, while Furusho et al. [187], Mooney et al. [83], Takahashi et al. [188], and Dong et al. [189] have developed semi-passive or active ankle solutions. In addition to these authors, there are other examples of AFO devices described in the literature that provide a better understanding of what has been developed. Awad et al. [190] developed a lightweight (0.9 kg), powered, and soft wearable ankle exosuit that interfaces with the paretic limb. The exosuit is composed of functional textile anchors that are visually similar to normal clothes. The actuation method is achieved using contractile Bowden cables located in the posterior and anterior anatomic planes of the ankle joint, allowing for plantar flexion and dorsiflexion movements, respectively. A low-profile shoe insole mechanically transmits power during walking. The cables are tensioned or relaxed through a body-worn actuator and a battery attached to a waist belt. The solution proposed in this study is capable of reducing the energetic burden associated with walking movement in individuals post-stroke, which under normal conditions can cost over 60% more than usual.

The exosuit's control system used a combination of position measurements from linear potentiometers and force measurements from load cells integrated into the textiles. This instrumentation was combined with rotational velocity measurements from a gyroscope mounted in each shoe to adapt the Bowden cable position trajectories and generate the desired assistive force profile on an iterative basis. The gyroscopes enabled real-time gait segmentation, while the potentiometers and load cells enabled iterative, force-based, and position control. Together, these sensors enabled appropriately timed assistive forces with adequate magnitude. However, the control system only provides reactive help and is not suitable for individuals with severe paralysis [191]. Etenzi et al. [191] developed a lightweight unpowered passive-elastic exoskeleton made of aluminium, weighing 1.4 kg, which stores elastic energy in springs (two for each leg) that assist during walking. The energy is stored from knee extension to the end of the leg swing phase and is then released during ankle plantar flexion. The actuation control uses a ratchet and pawl system to store and return energy through compression and release phases of metal springs, which act simultaneously with the knee and ankle. This approach achieved a reduction in metabolic cost, using 11% less energy compared to disengaging the springs. However, compared to walking without the exoskeleton, the metabolic cost increased by 23%. Galle et al. [192] developed and tested a bilateral external device weighing only 0.890 kg. It consisted of an AFO at each leg, with a hinge at the ankle and actuated through pneumatic artificial muscles connected between the foot and shank segments. The actuators were contracted when inflated with compressed air and aided during plantar flexion movements, achieving a 12% reduction in metabolic consumption compared to walking without the external device. The exoskeleton's control was realized by an iterative learning algorithm that used the signal from load cells connected between the orthoses and pneumatic muscles as input, and linear displacement sensors placed between the foot and shank sections of the exoskeleton.

Bougrinat et al. [193] developed a 2.045 kg ankle-powered exoskeleton that provides at least 30 Nm of assistive plantar flexion torque using an electrical motor and Bowden cables attached from the user's waist to carbon fibre struts fixed on the boot. They implemented a hierarchical architecture control system in an off-board personal computer for controlling the device. The high-level microcontroller estimates the gait cycle percentage by dividing the time passed in each cycle by the average walking period measured over ten cycles. The force-sensitive resistors placed under the insole at the heel area provide the needed input signals. The microcontroller then communicates to the PC, which is also a high-level controller, to transmit the desired current profile to the motor driver/encoder, which is a low-level controller. This particular exoskeleton was able to reduce the metabolic cost associated with the soleus and gastrocnemius muscles by 37% and 44%, respectively [193].

The previous examples of ankle/foot external devices are summarized in Table 3.4, which shows a clear trend towards developing lightweight solutions. Figure 3.10 illustrates a generic scheme of the solutions described.

Table 3.4: Examples available in the literature for ankle/foot solutions.

	Weight	Structural materials	Actuation method	Control system	Results	References
Awad et al	0.9 kg	Textile materials	Powered Bowden cables	IMU and load cells	Reduces the metabolic cost	[190]
Etenzi et al	1.4 kg	Aluminium	Unpowered Springs	Mechanic	Increases the metabolic cost in 23%	[191]
Galle et al	0.89 kg	-	Powered Pneumatic actuators	Iterative Learning Algorithm, load cells and IMU sensors	Reduces the metabolic cost in 12%	[192]
Bougrinat et al	2.045kg (considering all components)	Carbon fiber	Powered Bowden cables	Hierarchic Control Architecture	Reduces significantly the metabolic cost of the plantar flexion muscles	[193]

3.4.2 Hand/Arm Solutions

Brown et al. [194] illustrates the initial use of hand external devices to aid people with paralysis. Subsequently, such devices were employed in rehabilitation environments [195], particularly for individuals diagnosed with neurological disorders [196]. According to Ferguson et al. [195], hand exoskeletons or exosuits can be classified into four categories: assistive, rehabilitation, augmentation, and virtual reality. Assistive hand exoskeletons, such as those developed by Lucas et al. [197] and In et al. [198], aim to reduce muscular fatigue and improve functional dexterity [199]. Due to their portable design, these devices typically have fewer and smaller actuators, resulting in a more lightweight solution.

Ferguson et al. [195] explains that rehabilitation hand devices, such as those developed by Wege et al. [200] and Kawasaki et al. [201], are not required to be portable, as they are typically intended for use in physical therapy by multiple individuals. However, this requirement and the need to accommodate multiple DOFs, impairs the conception and development of these devices. Typically, as the complexity of the solution increases, so does the weight of the device.

Ferguson et al. [195] noted that augmentation exoskeletons, such as those developed by Shields et al. [202] and Hasegawa et al. [203], aim to improve the physical abilities of able-bodied individuals. However, designing such devices entails significant challenges, such as minimizing their weight while still reproducing the DOFs of a healthy hand. Currently, there is no combination of mechanical structural materials and actuators or power supplies that can provide a meaningful augmentation force.

There is another category of hand exoskeletons [195] that differs from the other types, as their goal is not to assist or enhance hand movements. Instead, they aim to simulate interaction through VR handsets by using haptic devices [204]. Park et al. [205] prototyped a dual cable hand exoskeleton to serve as an interface for VR environments. The device just weighs 320 g and is able to feedback on the touch sensation of hard and soft objects.

Yap et al. [206] developed a soft robotic assistive glove for individuals with grasp pathologies to assist them with everyday activities. The device is capable of supporting various hand manipulation tasks, including finger and thumb movements during hand closing and grasping activities. The glove is actuated by low-profile, soft, elastomeric pneumatic actuators that require low pressure.

The manipulation control approach involves an EMG strategy associated with radio-frequency identification (RFID) to predict the user's intentions. RFID tags act as non-physical switches that enable the activation of different hand gestures. Subsequently, the Arduino microcontroller receives the input from the sensors, and the voltage regulator sends output to the pressure sensors and miniature pneumatic pumps for air pressure regulation.

Díez et al. [207, 208] developed a modular hand exoskeleton for a rehabilitation environment that was originally designed for VR environments but was later adapted for real-life scenarios. The device is made using polylactic acid (PLA) 3D printable material and actuated by electric linear actuators placed in each finger. The exoskeleton is governed by a high-level controller that relies on EMG input signals. This control approach performs successfully in 97% of the trials [207], effectively triggering the opening and closing gestures.

Agarwal et al. [128, 209] developed a unique solution that differs from previous studies by considering three closed-loop chains to manipulate the four DOFs of the thumb. Specifically, the DOFs comprise carpometacarpal (wrist) flexion-extension and abduction-adduction movements, MCP flexion-extension movements, and interphalangeal flexion-extension movements (shown in Figures 3.1 and **3.2**). This closed-loop approach also resolves issues of axis misalignment at the exoskeleton-human joints.

The actuation method of Agarwal et al. [209] employs Bowden cables connected to actuated joints, enabling the transfer of up to 0.4 Nm of torque to each exoskeleton joint, producing highly backdrivable actuators with low reflected inertia and a weight of approximately 30 g each.

Each exoskeleton joint is equipped with a pulley that has a cable attached to its circumference. The cables are pulled by a brushed DC motor, which regulates the torque of each exoskeleton joint through a PID controller. This controller tracks the desired value, ensuring that each thumb joint and movement has a root mean square error of no greater than 13%. Structurally, the device was produced using selective laser sintering, which made it lighter, with some metallic parts added for load-bearing strength and durability. According to Agarwal et al.'s findings [128, 209], the device aligns with the natural movements of all thumb joints.

The following example also involves the development of a glove exosuit for hand rehabilitation by Klug et al. [210]. The device uses structural materials, such as microfibers, elastics, and PU pleather. It weighs 0.435 kg including batteries and controllers. Wires located along the palmar and dorsal sides of the hands, resembling flexor and extensor tendons respectively, actuate the glove, allowing independent finger movements. These wires are pulled by an electrical DC servomotor capable of transmitting up to 20 N of force. The exosuit is controlled through the readings of force sensors placed at the fingertips. In some situations, this approach may limit comfort and touch sensitivity while it provides a rough force estimate. The solution depends on two distinct sensor technologies, one based on piezo-resistive bending elements mounted dorsally, and the other on electroactive-based polymers located dorsally and on the palm of the hand. Using machine learning algorithms fed by the sensor readings, the exosuit controller regulates force almost in real-time. Consequently, the hand exosuit is capable of producing a maximum force of 27.4 N, assuming both the user and the device forces, and a mean bending angle of 132°.

Table 3.5 provides a concise summary of the hand/arm exosuit examples considered. An illustrative generic representation of an exoglove is shown in figure 3.10.

Table 3.5: Hand/arm exosuit applications found in literature.

	Type	**Structural materials**	**Actuation method**	**Control system**	**Results**	**References**
Yap et al	Assistive	Elastomers textile gloves	Pneumatic Actuators	EMG RFID	Satisfactory results Maximum force achieved 1.57 N	[206]
Diíez et al	Rehabilitation	3D printable material PLA	Electric linear Actuators	EMG controller	97% success during the trials	[207, 208]
Agarwal et al	Rehabilitation	Selective laser sintering materials Metallic load bearing parts	Bowden cables with springs Brushed DC motor	-	Compatible natural motion solution Max. torque 0.4 Nm	[128, 209]
Klug et al	Rehabilitation	Glove - microfibers, elastics and PU pleather	Wires Electrical motor	Force sensors Machine learning algorithm	Max. angle motion 132° Max. force 27.4 N	[210]

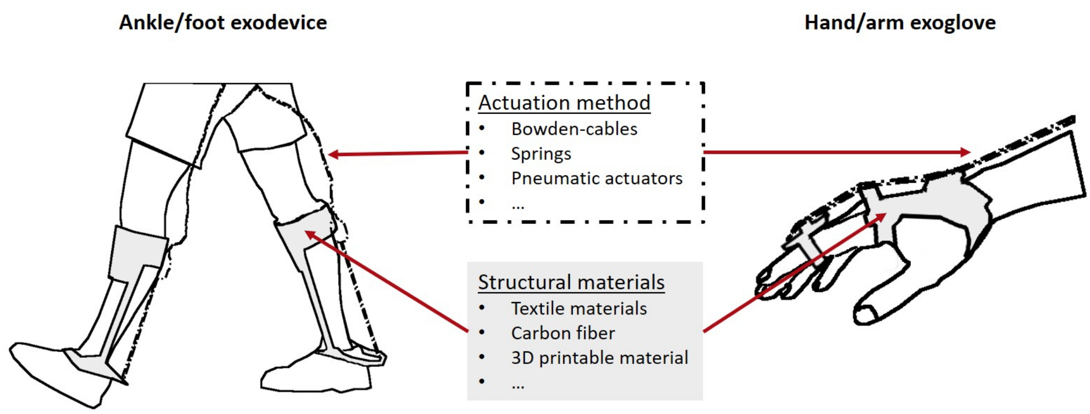

Figure 3.10: Generic illustration of an ankle/foot exodevice and a hand/arm exoglove (black dash-dot line represents the actuation method).

3.5 Ethical Issues

The adoption of external devices, such as exoskeletons or exosuits, for the purpose of enhancing physical abilities, whether for military, industrial, or rehabilitation contexts, could potentially introduce ethical, social, and legal issues to individuals and society [211, 212]. While there are undoubtedly numerous benefits associated with the use of these devices, including the ability to provide individuals with physical impairments greater freedom of movement and to increase safety conditions in the workplace, it is crucial to ensure proper regulation and monitoring to mitigate any potential negative consequences.

There's a worldwide tendency from both military forces and industrial companies to increasingly adopt the use of external devices to augment the physical capabilities of their soldiers and employees. However, there is a growing concern that these devices may result in the dehumanization of their users, as their primary aim became to achieve greater efficiency, endurance, and productivity in combat and work contexts, respectively [213].

Currently, the high cost and experimental nature of using external devices for mass rehabilitation purposes renders them inaccessible to the majority of the global population [213]. This scenario brings to light ethical concerns related to the potential of this technology to amplify existing social inequalities, and for being the source of new ones - such concerning prospects deserve an integrated societal response by all the relevant stakeholders.

3.6 Present and Future Perspectives

According to Bao et al. [214], the overall number of scientific publications about robotic exoskeletons increased exponentially since the 90s. Among the research areas (orthopaedics, computer science, automation and control) pushing this topic forward, engineering/biomedical and rehabilitation fields take the lead. Therefore, it is reasonable to expect an evolution of exoskeletons and exosuits in the near future. The review study published by Hill et al. [215], points to the potential of the technology employed in such devices, to improve the functional capabilities of individuals with neurological impairment, particularly in relation to ambulatory outcomes.

Despite their potential for mass adoption, the majority of devices found in literature are still prototypes or academic examples. Zhang et al [216] reviewed and compared several lower limb orthoses for the rehabilitation of patients with SC injuries. From the analysis, only 1 was evaluated with an A grade of recommendation. A similar investigation was made by Miguel-Fernández et al [217]. They evaluated the control strategies used in exoskeletons for gait rehabilitation in more than a thousand scientific papers. In the end, they noticed a low effectiveness of those control systems on clinical outcomes, justified by a lack of standardization in the experimental protocols which leads to high levels of heterogeneity. We see this heterogeneity as a consequence of the exploratory nature of the research in this domain. After this preliminary research stage, as resources such as artificial intelligence get a deeper integration in the control/actuation processes and help mitigating current shortcomings of existing technologies, a second generation of these devices is expected to emerge. In this phase standards and regulations both for testing and usage, are expected to emerge. The current paper aims to provide a valuable reference tool, instrumental in facilitating this evolution. Another avenue for improvement, according to Oña et al [218], depends on a better symbiosis with VR technologies required to promote a long-term recovery of motor function in daily living activities. Moreover, the recent pandemic situation caused by COVID-19 stressed the need for continuous and reliable rehabilitation therapeutics, pointing to home-based recovery solutions [219]. During home rehabilitation time, these devices might need to be worn while performing current daily tasks. As pointed out by Wolff et al [220], citing stakeholders such as healthcare professionals, such devices need to allow toileting, getting in and out of the car or even climbing stairs, among others.

The recent advances in artificial intelligence and machine learning have also improved mobile robotic exodevices used in motor rehabilitation. According to Vélez-Guerrero et al [221], there is a latent need to develop more reliable systems through clinical validation and improvement of technical characteristics.

Despite the long journey that rehabilitation devices have already taken, such as reducing hospital costs and improving the overall well-being of their users, there are still flaws and gaps that must be solved to address current and future needs.

3.7 Conclusion

A concise review of the state of the art in exoskeleton and exosuit rehabilitation solutions was presented. The materials used in the structure, the actuators (and their associated power sources), and the control systems, as described in this review, can be combined in various configurations to fabricate external devices aimed at enhancing or rehabilitating the physical capabilities of humans.

The exoskeleton and the exosuit are different concepts that can potentially be combined to address their respective limitations. For instance, leveraging the advantages of soft structural materials with the enhanced performance of traditional actuation methods can produce optimal results.

Nevertheless, it is fundamental to acknowledge that external skeletons and suits have distinct objectives dictating their design. Taking lower limb devices as an example, exoskeletons are primarily intended to bear an individual's weight, as in the case of lower limb paralysis, whereas exosuits are typically only capable of assisting with movement if there is some mobility (tough residual) in the legs.

The authors of this review are confident that they have exhaustively explored the most relevant examples pertaining to the topics discussed. With this effort, they hope to have contributed in some measure towards promoting a faster and more effective development of external devices for the benefit of humanity.

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Chapter 4

Influence of DMSO Non-Toxic Solvent on the Mechanical and Chemical Properties of a PVDF Thin Film

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Keywords: PVDF thin film; polymeric material; optical analysis; mechanical characterization; chemical modifications


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Influence of DMSO Non-Toxic Solvent on the Mechanical and Chemical Properties of a PVDF Thin Film

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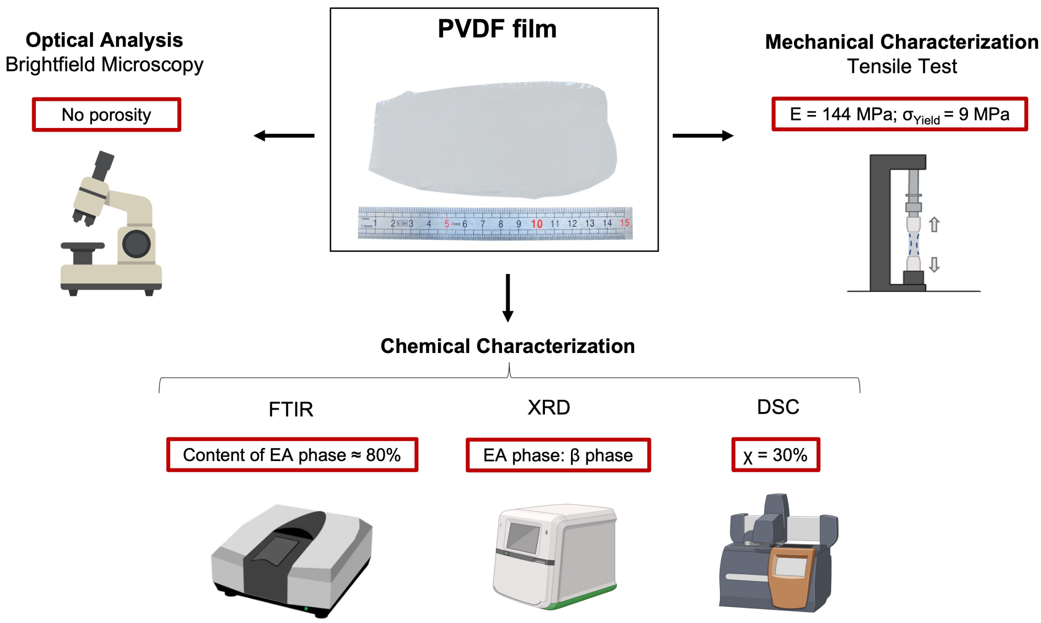
4.1 Abstract

Piezoelectric materials such as PVDF and its copolymers have been widely studied in different areas and with promising applications, such as haptic feedback actuators or deformation sensors for aided-mobility scenarios. To develop PVDF-based solutions, different protocols are reported in the literature; however, a toxic and harmful solvent is commonly used (dymethilformamide (DMF)). In the present study, a non-toxic solvent (dymethilsulfoxide (DMSO)) is used to dissolve PVDF powder, while a specific ionic liquid (IL), [PMIM][TFSI], is used to enhance piezoelectric properties. A PVDF/IL thin film is characterized. The physical material characterization is based on optical analysis (to ensure the sample's homogeneity) and on mechanical linear behaviour (Young's modulus of 144 MPa and yield stress of 9 MPa). Meanwhile, a chemical analysis focuses on the phase modifications introduced by the addition of IL (β phase increase to 80% and a degree of crystallinity, χ, of 30%). All the results obtained are in good agreement with the literature, which indicates that the proposed experimental protocol is suitable for producing PVDF-based thin films for biomedical applications.



Graphical abstract.

4.2 Introduction

Smart materials are currently a subject of high research value. They have been studied across different research fields, and there are significant applications in areas such as aerospace and aeronautics [1, 2], medicine and rehabilitation [3–5], bioengineering [6, 7], electronics [8] and architecture and design [9]. Despite their current applications, some of these materials were developed to mimic the human muscular and nervous systems [10] using non-biological materials. Given their ability to respond to external factors, these materials can be classified according to the input stimuli [11–13]. Known inputs can be thermal [14, 15], magnetic [14, 16], light [17, 18], pH [19, 20], pressure [10, 19], moisture [21] or electrical [14, 22]. Materials reacting to an electrical input, in particular the piezoelectric subclass, are well-known and are frequently used for healthcare purposes and biomedical applications [23], such as in nanomedicine and in tissue engineering scaffolds. Some examples of common materials used are zinc oxide (ZnO), cellulose nano fibril, boron nanotubes and lead zirconate titanate (PZT) [24].

Polyvinylidene fluoride (PVDF) and its copolymers have excellent properties that make them the ideal choice in several applications from electronics to bioengineering and medical applications [25, 26]. PVDF is a crystalline polymer with good chemical and weather resistance as well as good distortion and creep resistance at low and high temperatures [27]. Moreover, it is flexible, light (low density), biomimetic, biocompatible and easily processable. Such characteristics make it a good solution for muscle-like actuators [28]. Other relevant properties of PVDF are a large dielectric constant (of 10 (ε') [29]), high polarity, ionic conductivity of around 10^{-4} S/cm at 20°C [30], high piezoelectric coefficient of 49.6 pm/V [31] and high mechanical strength (Young's modulus $\approx$ 1.6 GPa and yield strength $\approx$ 45 MPa [32]). Chemically, there is evidence of at least five polymorphic modifications in its structure (α, β, γ, δ and ε), with each phase being responsible for different behaviours in the material, caused by intrinsic properties. For example, β and γ phases are the most desirable when the goal is to use PVDF as a sensor or soft actuator due to their piezoelectric, pyroelectric and ferroelectric abilities [33, 34]. These phases, mainly the β phase, can be promoted using different methodologies, including mechanical stretching of α phase [35], melting PVDF under specific and controlled conditions (i.e., high pressure) [36], applying an external electrical field [37], using ultra-fast cooling [38], solution crystallization at temperatures below 70°C [39] or by addition of nucleating fillers (e.g., ionic liquids (ILs)). The inclusion of ILs (typically composed of an organic cation and an organic/inorganic anion) brings other advantages, such as lower actuation voltage and improvement to electromechanical stability and durability, leading to higher material performance [32]. Moreover, some solvents, such as dimethyl formamide (DMF), dimethylacetamide (DMAC) and dimethylsulfoxide (DMSO) can also play a role in β phase formation [40], since higher dipole moments tend to favour the formation of phases with pieozoelectric properties [41].

Several authors have studied this polymeric material for many applications with different purposes [42]. For example, Mat Nawi et al. [43] used PVDF with polyethylene glycol dissolved in DMAC to improve the process of water treatment. In the field of medicine, Wang et al. [44] used PVDF films as the sensory component in a sensor system to monitor breathing and heartbeats during sleep. Moreover, PVDF has already been investigated as an energy harvester by Hu et al. [45]. They proposed a design that uses a PVDF film to capture energy from bending as a power source for pacemakers.

Focusing on PVDF's properties, Correia et al. [32, 46] have studied the mechanical, chemical and electrical properties of PVDF-based materials with consideration of the inclusion of different ILs and using DMF as a solvent. Despite using different process techniques, both studies focus on the influence of different ILs on the properties of the material. All ILs increased the electroactive (EA) β phase content in proportion to the IL alkali chain length [32]. The degree of crystallinity (χ) also increased [46] depending on the IL chain length, and the mechanical properties changed (plasticizing behaviour in the presence of the IL). Singh et al. [47] conducted a phase, conductivity and dielectric analysis and a morphology study of the β phase of PVDF. The objective was to use PVDF as a gel polymer electrolyte for magnesium-ion battery applications. The study reported the presence of a pure β phase PVDF membrane and good affinity with a polar organic electrolyte. Yang et al. [48] used a similar approach. They used scanning electron microscope, Fourier transform infrared spectroscopy (FTIR) and X-ray diffraction (XRD) techniques to study the morphology, structure and piezoelectric response of a composite film ($BaTiO_3$/PVDF) for human motion monitoring. As a result, a pressure sensor based on PDA@BTO/PVDF showed a fast response and a good ability to provide an energy supply with an apparent enhancement to output voltages. Despite these successful applications, neither Singh et al. [47] nor Yang et al. [48] considered the inclusion of ILs or used low-toxicity solvents as alternatives to DMF.

Regarding the solvent, DMSO evidences a large spectrum of pharmacological effects, including anti-inflammatory effects, local analgesia and weak bacteriostasis, and it is mainly used as a vehicle for other drugs [49]. It shows a high boiling point (189°C) and a coefficient of solubility of 16.4 MPa$^{1/2}$ (δ_p), which is close enough to the solute parameter (δ_p = 12.5 MPa$^{1/2}$). A relative similarity between the Hansen solubility coefficients of a solute and solvent usually indicates ease with dissolving the first into the second. However, other solvents, such as DMF, present a δ_p closer to that of PVDF, which can represent an advantage in terms of dissolution time. Still, DMSO is a good candidate to dissolve PVDF according to the literature [50, 51]. Furthermore, as a solvent, DMSO is used in situations where low toxicity is required and desirable, since conventional solvents are assumed to be severely toxic and harmful by the European Chemicals Agency [52]. The combination PVDF + DMSO has been used for different purposes: for instance, Wang et al. [53] evaluated the use of DMSO in the process of making lithium ion batteries, while Venault et al. [54] used them to make antifouling membranes. Both authors pointed out the great potential of DMSO as the ideal candidate to replace hazardous solvents.

As a result, with the present study, our research team propose a new experimental protocol to produce a PVDF-based actuator using a non-toxic solvent, which is crucial for biomedical applications. Moreover, the research team expect to better understand the mechanical properties, chemical phase content, as well as the morphology through optical analysis of a composite material based on PVDF by the addition of an IL filler. In the future, we hope the material developed and studied will be integrated into upcoming exosuit solutions, which could be used for aided mobility or rehabilitation scenarios (for instance, for muscle touch feedback for re-educational movement purposes).

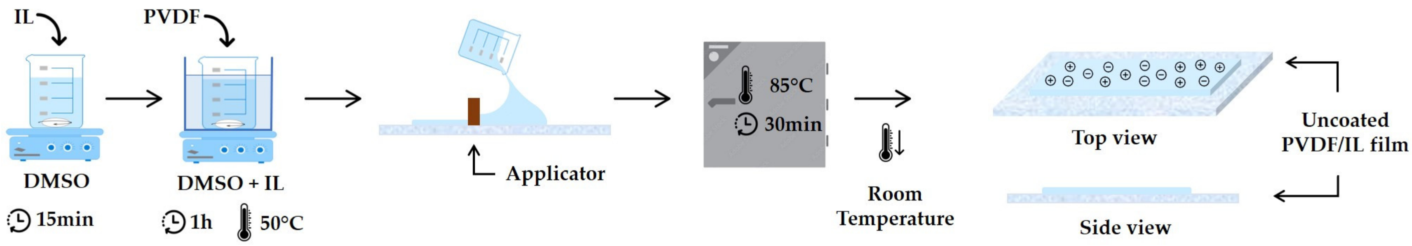
4.3 Materials and Methods

4.3.1 Materials

The samples were produced using three commercially available chemical compounds. The PVDF powder (Solef 6020, Brussels, Belgium) was offered by Solvay Lda (Brussels, Belgium). The IL (1-Methyl-3-propylimidazolium bis (trifluoromethylsulfonyl) imide, [PMIM][TFSI], $\geq$99%), which is responsible for increasing the content of the EA phase in PVDF, was purchased from IoLiTec-Ionic GmbH (Heilbronn, Germany). The polar non-toxic solvent, dimethylsulfoxide (DMSO ANH, 99.8%), was purchased from Fisher Scientific, Lda (Waltham, MA, USA).

4.3.2 Sample Preparation

The samples were produced using an experimental protocol based on Correia et al. [32] and adapted according to the objectives of this work, i.e., non-toxic for biomedical applications. In the first step, the IL was mixed with DMSO, considering a ratio of 40% w/w IL/PVDF. In the second step, the PVDF was added to the previous solution at a ratio of 12/88% w/w PVDF/DMSO. During the dissolution of the PVDF powder, the beaker was sealed and the mixture was stirred using a magnetic stirrer and heated from room temperature to 50°C in a thermal bath [55]. After achieving a transparent and homogeneous solution, the resultant mixture was cast to a glass substrate, and the wet film thickness was set to 0.6 mm using a doctor blade technique (Proceq ZUA 2000, Schwerzenbach, Switzerland). On the last step of sample preparation, the wet film was taken into an oven for total solvent evaporation for approximately 30 min. The temperature was set at 85°C in order to avoid the formation of pores [55]. Figure 4.1 is a schematic representation of the film production process, as previously described.



(a) Step-by-step production of PVDF-based samples.

(b) Homogeneous and transparent PVDF-based samples.

Figure 4.1: Schematic illustration of the production (**a**) and the final film (**b**) of PVDF-based samples.

Finally, three specimens with dimensions of 30 × 10 mm^2 (length × width) were cut from the sample and weighed. With these measures, the density (ρ) of each sample was obtained, and the mean value and standard deviation (STD) were calculated.

4.3.3 Optical Analysis

To attest the porosity level and assess if powder dissolution was completed, the samples were observed using the bright-field microscopy technique. The produced samples were cut into a square shape with dimensions of 12 × 12 mm^2 and were inspected over a grid composed of smaller squares in a Zeiss apparatus (inverted fluorescence microscope, Axiovert 200M, Oberkochen, Germany) under a 5× zoom magnification.

4.3.4 Mechanical Characterization

The mechanical tests were conducted following the standard test method for tensile properties of thin plastic sheeting, such as ASTM D882-12 [56]. The samples were cut to dimensions of 75 × 10 mm^2 (length × width) and had a thickness of 0.048 ± 0.004 mm (mean ± STD), which is the value expected after solvent evaporation according to Krebs et al. [57]. For the mechanical test, the samples were mounted with a span of 50 mm between grips. The prototype machine used is able to perform uni- and bi-axial tests and is equipped with 120 N actuators and 50 N load cells (Figure 4.2). The samples were tensile tested leading to rupture at a constant velocity of 5 mm/min (supplementary video A.1). This mechanical approach was used to obtain some of the mechanical properties in the elastic domain, such as the Young's modulus (E), the yield stress (σ_{Yield}) and the corresponding yield strain (ε_{Yield}).

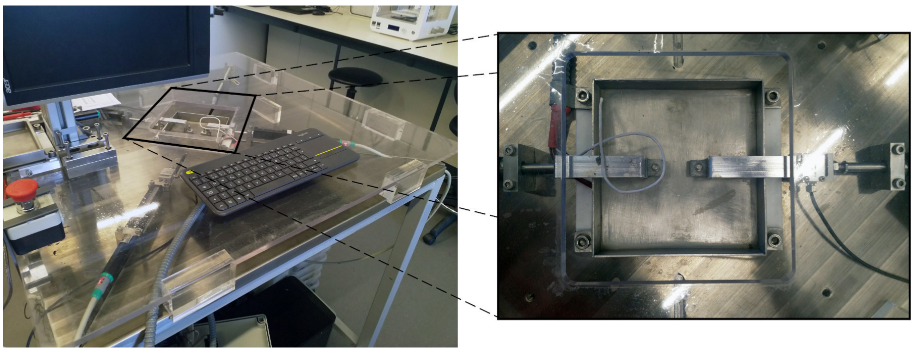


Figure 4.2: Mechanical apparatus used to mechanically tension the samples. At left, we present a general overview of the machine used, while at right, we present a detailed view of the mechanical actuators.

4.3.5 Chemical Characterization

According to Martins et al. [34], the phases of PVDF can be completely characterized by performing different chemical analysis together. FTIR, XRD and differential scanning calorimetry (DSC) were performed to distinguish and identify the prevalent electroactive phase, its content and the degree of crystallinity.

FTIR-attenuated total reflectance (ATR) measurements were conducted using a PerkinElmer FT-IR spectrometer frontier apparatus (Waltham, MA, USA). The analysis was made by considering 64 scans with a resolution of 4 cm^{-1} from 4000 to 600 cm^{-1}. From FTIR-ATR, it is possible to distinguish the non-EA phase (e.g., α phase) from the EA phases (e.g., β and γ phases) present on the material, and from Equation (4.1) [40], it is possible to calculate the total content of the EA phases (F(EA)).

$$F(EA) = \frac{A_{840,EA}}{\left(\frac{K_{840}}{K_{766}}\right) \times A_{766,\alpha} + A_{840,EA}} \tag{4.1}$$

where A_{766} and A_{840} are the absorbances at 766 cm^{-1} and 840 cm^{-1}, respectively, of the α phase and EA phases, and K_{766} and K_{840} are the corresponding absorption coefficients (6.1 $\times$ 10^4 and 7.7 $\times$ 10^4 cm^2/mol, respectively).

Besides FTIR-ATR, XRD (Bruker D8 Advance DaVinci, Billerica, MA, USA) was also used to identify the phases. This technique, in contrast to FTIR, allows clear distinction of the EA phases, such as the β phases from the γ phases. The method uses a conventional Bragg-Brentano diffractometer, and the following parameters were adopted: wavelength of the incident X-ray beam (λ) of 1.5405 °A, angle range of $5° \leq 2\theta \leq 45°$, step size of 0.02°, and 1 s per step.

The combination of both methods (FTIR-ATR and XRD) enables a robust identification of the electroactive phases presented in the material.

DSC measurement was conducted using a Hitachi DSC7020 calorimeter (Hitachi, Ibaraki, Japan) at 10 °C/min from 50 °C to 200 °C. This complementary technique allows us to obtain the degree of cristalinity, χ, of the sample through Equation (4.2) [32], as well as the melting (T_m) and the onset (T_{onset}) temperatures from the resultant curve.

$$\chi = \frac{\Delta H}{x\Delta H_\alpha + y\Delta H_{EA}} \tag{4.2}$$

where ΔH is the melting enthalpy of the material, ΔH_α and ΔH_{EA} are the melting enthalpies of the α (93.07 J/g) and EA phases (103.4 J/g), respectively, and x and y are the α and EA phase proportions, respectively, obtained from FTIR-ATR.

4.4 Results and Discussion

To evaluate the uniformity of the final sample, the density of three different specimens was calculated. The mean value of ρ was 1425.926 $\pm$ 32.075 kg/m^3. The low standard deviation (≈2.25%) indicates that all the specimens had similar densities, which means the sample was uniform.

4.4.1 Optical Analysis

Each square sample was segmented into 42 small areas. Figure 4.3 shows the overall microscopy of an analysed specimen. No porosity was observed in the analysed samples, which indicated a correct solvent evaporation temperature [39]. Moreover, the absence of air bubbles or solvent debris shows that the initially defined evaporation time was adequate for the process. However, the small dots present in the microscopic images might suggest incomplete dissolution of PVDF, indicating that more time would be needed to completely dissolve the PVDF powder and achieve a homogeneous solution at the microscopic level.

Gaihre et al. [58] prepared PVDF samples by dissolving 10% PVDF in dimethylformamide (DMF) with different additives and studied the porosity of polypyrrole (PPy)-PVDF micro-actuators. Even though they used a spin coating procedure on a glass side and dried the sample at 50 °C, they obtained samples with no porosity. Poudel et al. [59] performed digital bright-field microscopy analysis to evaluate the agglomeration of boron nitride nanotubes (BNNTs) in solvent-cast PVDF-trifluoroethylene (TrFE) films, using DMF as solvent. They chose total BNNT dispersion in the samples for lower content of nanotubes. However, the presence of porosity was reported for unpolled and annealed (at

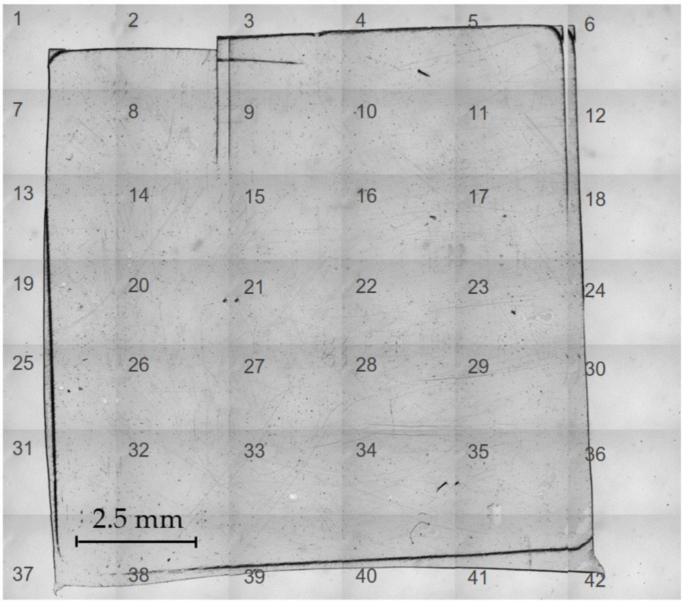


Figure 4.3: Sample viewed under microscope with 5× magnification.

25 °C) samples of PVDF-TrFE and PVDF-TrFE-BNNT 1 wt%. Other authors, such as Chen et al. [60] and Nunes-Pereira et al. [61], suggested that the samples' porosity could be controlled by the solvent choice and by evaporation temperature. However, in both studies, DMF was used to dissolve PVDF.

4.4.2 Mechanical Characterization

The mechanical properties—yield stress, strain and stiffness—were obtained through uniaxial tensile tests. Figure 4.4 shows the linear elastic behaviour of PVDF samples of 12/88% w/w (PVDF/DMSO) from 20 valid tests (gray lines), the mean curve (black full line) and standard error of the mean (SEM) (gray area). Through the mean curve, a σ_{Yield} of 9 MPa and a corresponding ε_{Yield} of 0.06 were estimated. Moreover, a Young's modulus (E) of 144 MPa could be obtained from the slope of the linear elastic region. To evaluate the influence of the concentration of PVDF on the mechanical properties, another percentage of PVDF was analysed. Maintaining the proportion of IL/PVDF but changing the ratio of PVDF/DMSO to 15/85% w/w, the result of σ_{Yield} was 9.8 MPa, and the corresponding ε_{Yield} was 0.17: both results being higher for this ratio. The value of E

was smaller at 118 MPa. This indicates that by increasing the percentage of PVDF, the yield stress and yield strain increase, but the Young's modulus decreases.

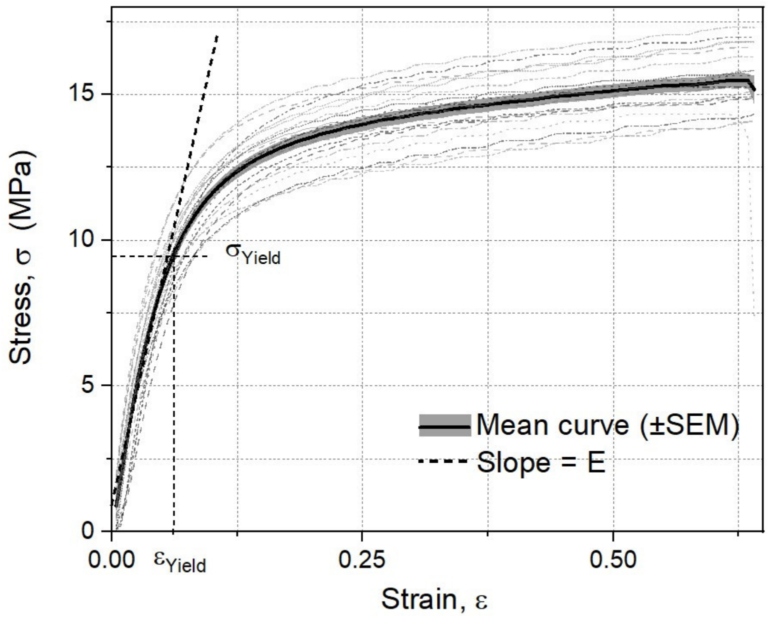


Figure 4.4: Mechanical tensile results of 12/88% w/w samples. Grey lines—20 individual specimens; black full line—mean curve; gray area—SEM.

The same relation between the increase of stiffness with the decrease in the percentage of PVDF has also been observed by other authors and has been reported in the literature [62]. Correia et al. [32, 46], using DMF as solvent, obtained an E ranging from 75 MPa to 184 MPa and a σ_{Yield} varying from 2.53 MPa to 6.4 MPa, with the range of values dependent on the IL used. Comparing the results from Correia et al. [32] with those obtained in this study for 12/88% w/w PVDF/DMSO, it is worth pointing out that E is similar and σ_{Yield} is on the same order of magnitude. However, the last parameter is higher in the present study, which might be due to different solvents and conditions used in the experimental protocols.

Bao et al. [63] studied a PVDF/IL piezo-active composite film to be used for highly sensitive pressure sensors. The samples were prepared with 1-Ethyl-3-methyl- imidazolium Chlorid ([EMIM]Cl), were dissolved in DMF and were tensile tested, and the stress–strain relation was determined. They reported that the Young's modulus decreased with the IL content. However, Kong et al. [64] studied the mechanical properties of a composite material based on PVDF and reinforced with conductive black carbon and silicon dioxide, since damage severely limits the applications of polymer membranes. The samples were prepared using DMF as solvent and were tested referring to the ASTM D 882-2012 standard [56]. They conclude that the tensile strength of PVDF increases with the content of the additive.

The results of this study show that the mechanical properties are influenced by the PVDF/DMSO ratio and decrease with the addition of fillers, as reported by Vázquez-Fernández et al. [65]. Nevertheless, the use of a different solvent, when compared to the literature, seems not to have a significant impact on the mechanical properties. Despite the property variations in comparison with PVDF, the piezoelectric material exhibits mechanical properties suitable for real-world applications. Its resistance to plastic deformation at low stress levels can be a potential advantage for applications in rehabilitation scenarios.

4.4.3 Chemical Characterization

FTIR-ATR analysis allowed us to obtain the vibrational absorption band characteristics of each phase (α and EA phases) and the transmittance values needed to calculate the phase content in the sample. Figure 4.5 shows a peak at 766 cm^{-1}, which is representative of α phase, while the peak 840 cm^{-1} validates the presence of EA phases. These results are in agreement with previous studies [32, 34] on this type of material. From Equation (4.1), the content of each phase is calculated. The samples of 12/88% PVDF/DMSO with IL contained approximately 80% EA phase, which is believed to be enough to perform a good piezoelectric response when externally stimulated by electrical inputs. Considering other ratios of PVDF/DMSO (using IL), such as 15/85% w/w, the EA content increased to approximately 86%. Without IL, those values decreased to 69% and 70%, respectively.

The samples with a PVDF/DMSO ratio of 12/88% w/w were also analysed with XRD

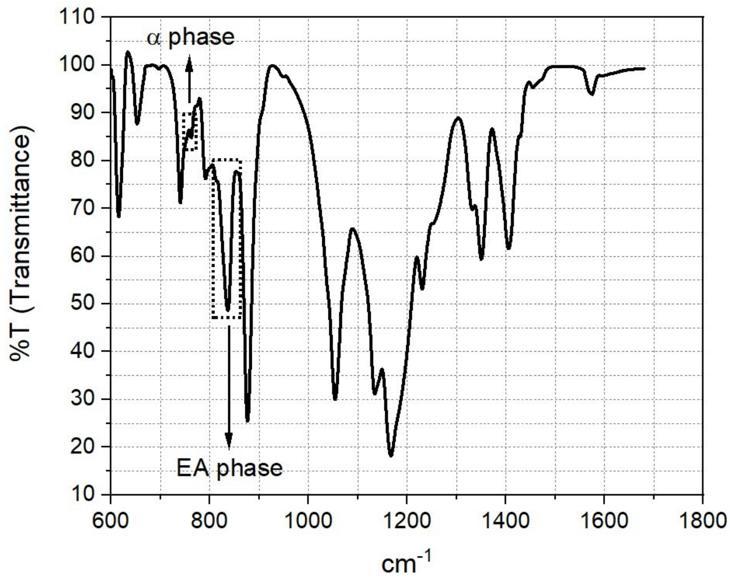


Figure 4.5: FTIR−ATR analysis of the composite material. The peaks corresponding to α and EA phases are represented in the graph and are marked with the dotted rectangles.

and DSC techniques. XRD analysis allows us to identify and distinguish the β phase from the other phases: mainly from the γ EA phase. Despite the peak around 2θ = 20° being characteristic of both EA phases of PVDF (β and γ), the absence of clear peaks before that value is an indicator of the main presence of β phase instead of γ phase (Figure 4.6 [34, 66]). This result is expected for enhanced piezoelectric characteristics of the samples, since the beta phase being the prevalent EA phase is an advantage for piezoelectric materials.

DSC analysis (Figure 4.7) quantifies the crystallinity of the sample and investigates the response of the polymer to heat. From Equation (4.2), a degree of crystallinity of approximately 30% was achieved, meaning that 30% of the polymer's mass formed crystalline regions, which directly interfere with some properties, such as the stiffness and piezoelectric behaviour [67]. Higher crystallinity means higher alignment of the polymer chains, which increases the stiffness of the material. The remaining polymer percentage, i.e., 70%, represents the amorphous regions. Moreover, it was possible to determine the melting (T_m) and the onset (T_{onset}) temperatures as 159.1 °C and 144.68 °C, respectively. At the end of this analysis, no mass loss was observed.

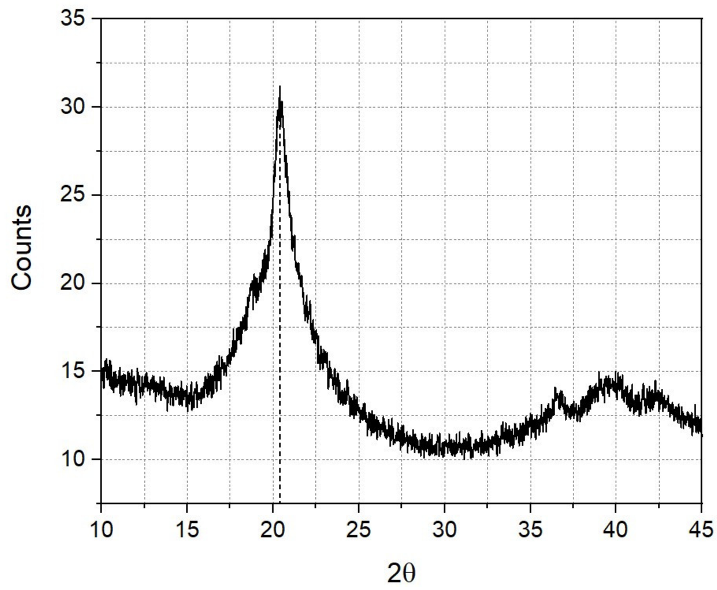


Figure 4.6: XRD analysis of the composite material. The peak at $2\theta \approx 20°$ is characteristic of EA phases.

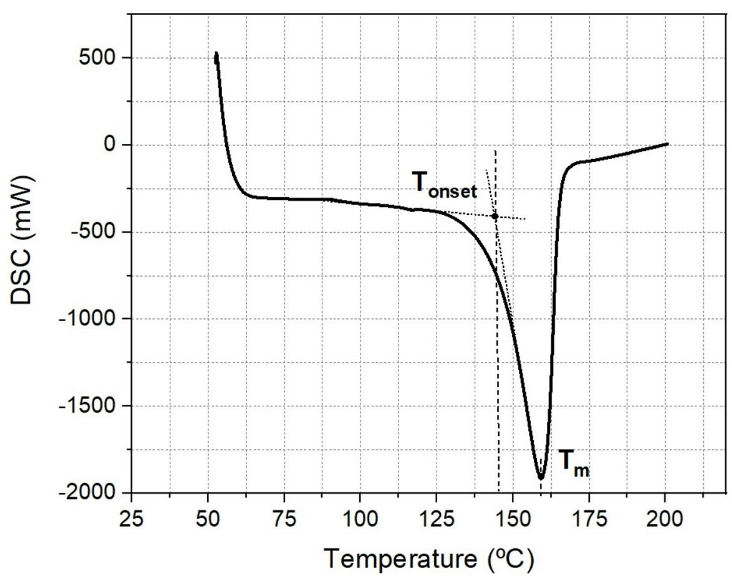


Figure 4.7: DSC analysis of the composite material. Representation of T_{onset} and T_m.

Other authors such as Correia et al. [32] and Liu et al. [68] also characterized PVDF films prepared with DMF solvent in terms of chemical properties regarding EA phases. Correia et al. [32] reported a β content from 59% to 89% using different types of IL and maintaining the IL/PVDF ratio at 40%. Compared with the results from this study, the values obtained match the range obtained by Correia et al. [32] even though different solvents were used. Liu et al. [68] obtained a β content higher than 93% by using the IL ([C2min][BF4]), which is higher than the results obtained in this study. Nevertheless, they also concluded that the polymer phases and their content were influenced by PVDF/-solvent ratios and by the presence of IL. Considering the solvent and the presence of IL, a higher concentration of PVDF increased the percentage of electroactive phases.

In addition to the β content, Correia et al. [32] also measured the crystallinity and the melting and onset temperatures. They obtained a χ result varying between 28% and 56% depending on the type of IL used. Regarding the melting temperature, the result obtained via DSC was between 154 °C and 173 °C, while the onset temperature was between 371 °C and 407 °C as measured by thermal gravimetric analyser (TGA). Begum et al. [69] studied the crystallinity and the thermal stability of PVDF nanocomposites dissolved in DMF and reinforced with epoxy functionalized multi-walled carbon nanotubes (MWCNTs). They found a T_m of 161 °C for PVDF. Moreover, although the addition of MWCNTs changed the thermal properties, the authors did not found significant variations in the temperature with an increase in the additives.

Despite the differences between experimental protocols, the results we present in this study compare favourably and are in good agreement with the results found in the literature, regardless of the solvents used.

4.5 Conclusions and Future Works

There are several experimental protocols to produce PVDF-based thin films described in the literature. However, the majority of them use toxic solvents such as DMF or methyl-ethyl-ketone (MEK), which are potentially harmful for users. Therefore, in order to produce an actuator free of toxic solvents (crucial for biomedical applications), in the present study, DMSO was chosen to create a PVDF-based film. Even though several authors have pointed to the possibility of using this solvent, only a few studies can be found in the literature. Hence, it is important to explore the properties of PVDF-based actuators produced by different protocols: especially those using non-toxic solvents. Having non-toxicity and strong piezoelectric behaviour as principal objectives in this study, our research team found an optimal ratio of 12/88% w/w PVDF/DMSO to obtain a more fluid mixture that facilitates the dissolution of the PVDF powder. Moreover, the best temperature for PVDF dissolution was also successfully achieved. A very low temperature would increase the dissolution time, while a high temperature could degrade the material. Therefore, a temperature of 50 °C was the ideal value to develop a suitable experimental protocol.

Characterization of the material revealed that the inclusion of IL changes all the properties analysed, such as the Young's modulus (E), the yield stress (σ_{Yield}) and the β phase content. Nevertheless, considering similar ratio conditions, the use of a different solvent does not evidence deep changes to the material's properties, which points to the possible use of DMSO as a replacement solvent for hazardous alternatives. In the future, we aim to study the electro–mechanical properties and the response of the PVDF-based smart material to investigate even further its use in biomedical applications. Given the promising results obtained for the properties of this polymeric material, our research team intends to develop a biomedical device using a piezoelectric PVDF thin film as the main element.

With this study, our research team hopes to have contributed significantly to the knowledge about piezoelectric PVDF-based materials and the potential use of these smart soft materials in future biomedical applications.

Author Contribution: Conceptualization, A.D.A. and P.M.; investigation, A.D.A.; methodology, A.D.A.; formal analysis, A.D.A. and A.M.T.; writing—original draft, A.D.A.; writing—review and editing, A.D.A., A.M.T. and P.M.; supervision, P.M. All authors

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Chapter 5

Piezo-Ionic Actuator for Haptic Feedback

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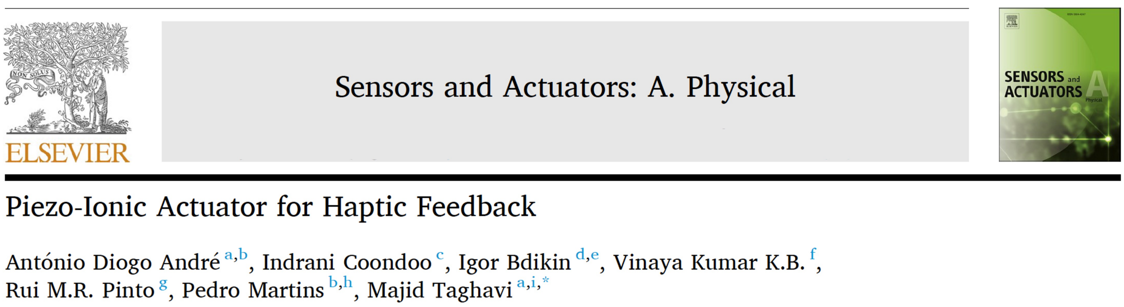
Keywords: PVDF, piezoelectric actuator, electromechanical response, haptic feedback



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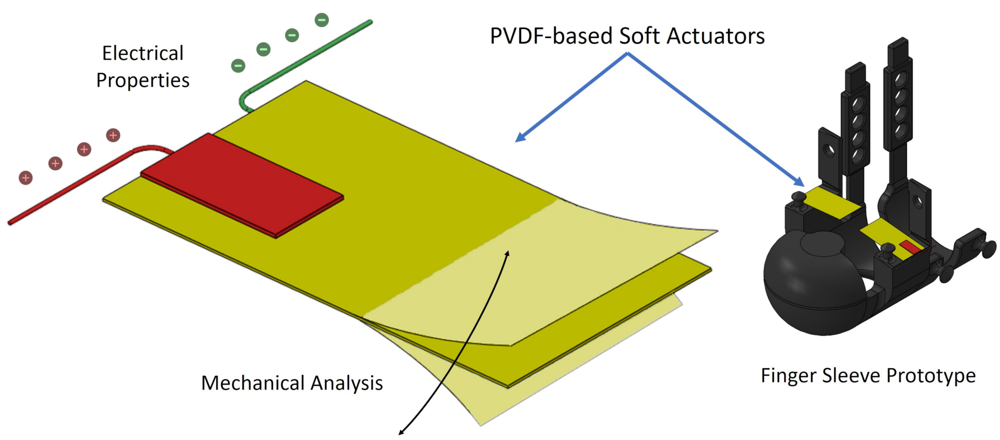
5.1 Abstract

Electroactive polymers have received substantial attention for actuation because of their muscle-like actuation behaviour. These polymers are typically studied under ionic and electric classes based on their fundamental response mechanisms. In this study, a hybrid piezo-ionic actuator is developed and characterised by its electromechanical response to analyse the piezo-ionic synergistic effect in a cantilever beam actuation design. The piezo-ionic actuator was developed using polyvinylidene fluoride (PVDF) combined with an [Pmim][TFSI] ionic liquid (IL) filler. The addition of IL into the PVDF network promotes the formation of electroactive phases (β and γ), consequently enhancing the electromechanical response of PVDF while maintaining the characteristic fast response time of piezo materials. The IL also plasticize the PVDF polymer and increases its conductivity which also causes the electrical parameters to vary with frequency. It results in higher dielectric loss, energy storage and hysteresis in PVDF/IL responses. To evaluate the actuator performance, the force generated by the hybrid actuator is measured and a finger sleeve is designed for haptic feedback analysis.



Graphical abstract.

5.2 Introduction

Electroactive polymers capable of converting electrical energy into mechanical motion and vice versa [1], have received substantial attention for their applications across different sectors, including industry [2] and healthcare [3]. The soft nature of these polymers makes them ideal for lightweight actuation while offering straightforward and flexible design options [4]; although with limited output force [5, 6]. These polymers are commonly classified into ionic and electric types based on their response mechanism [7]. In ionic actuators, the actuation response is caused by the displacement of ions within the polymer, typically requiring low voltages to trigger a mechanical response. In contrast, electric actuators respond to the electrical stimuli through the polarization of dipoles [8]. Ionic actuators are known for their slower response time and relatively lower force output, which can constrain their use in applications where minimal force is sufficient. For example, Heydt et al [9] and Ren et al [10], used ionic actuators in the development of refreshable Braille display, and Hardy et al demonstrated their application in drug delivery systems [11].

Piezoelectric materials, categorised under electric actuators, employ the converse piezoelectric effect, to alter their shape in response to electrical stimuli. Piezoelectric polymers, such as polyvinylidene fluoride (PVDF) known for its high piezoelectric constant among the polymers [12], demonstrate lower displacement when compared to ionic-based polymers [13–15]. However, their response is quicker, making the piezoelectric actuators ideal for high frequency and precision positioning tasks [16]. For example, it has been used in bimorph actuators for laser scanning actuation at high frequencies [17], ideal for high-speed manipulation as well as in haptics [18], and touch displays [19]). The actuation performance of PVDF is influenced directly by the presence of electroactive phases (β and γ), which are typically minimal in pure states [20]. These phases can be enhanced through a phase transition, solvent casting, the addition of nucleating fillers or the development of new PVDF copolymers [21]. The incorporation of ionic liquid as nucleating fillers to enhance piezoelectricity has received interest, with studies comparing different types of ionic liquids in the composition [22–24], the porosity of the polymer [25], and embedded electrodes [26]. The inclusion of IL in the PVDF network can potentially have a synergistic effect on the actuation mechanism benefiting from both ionic and piezoelectric actuation principles. First, ionic liquids act as a plasticizer in the network, reducing Young's modulus of the actuator [22, 27], thus enabling larger deformation for these actuators. Second, the inclusion of ionic liquids allows for ionic mobility and conductivity in the composite, decreasing the actuation voltage when compared to pure piezoelectric materials. The actuation behaviour of these hybrid piezo-ionic actuators, along with their hysteresis and durability, has not yet been thoroughly investigated. They are influenced by various factors, including the type of ionic liquids in the polymeric network, electrode composition, and fabrication parameters such as solvent and curing process. This work aims to address this gap by investigating the electromechanical properties and actuation performance of a PVDF-IL composite (Figure 5.1) for potential haptic applications. Our material development process involves optimizing the fabrication process with a non-toxic solvent, dimethylsulfoxide (DMSO), and leveraging the thermoplasticity of the film for a straightforward heat-transfer printing technique to embed electrodes. We have based our studies on the 1-Methyl-3-propylimidazolium bis(trifluoromethylsulfonyl)imide ([PMIM][TFSI]) as the ionic liquid, which has been reported to significantly increase the crystallization of electroactive phases of PVDF, among various cations and anions studied [22, 23]. Our mechanical characterization shows that this reduces the Young's modulus of the films by approximately tenfold compared to pure PVDF, confirming the plasticizing effect of IL [27], with the mean Young modulus of 144 MPa and a yield stress of σ_{Yield}=9 MPa.

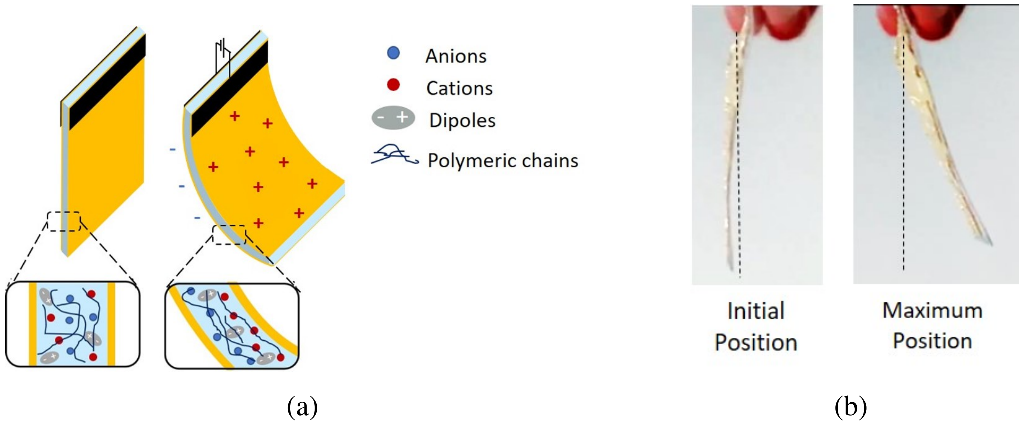


Figure 5.1: The PVDF-IL film sandwiched between electrodes: (a) Schematic representing the actuation mechanism; (b) Movement of the sample under applied voltage (10 V, 0.05 Hz).

5.3 Materials and Methods

5.3.1 Actuation development

The PVDF/IL material was developed using a PVDF powder (Solef 6020 by Solvay Lda), an ionic liquid (IL) (1-Methyl-3-propylimidazolium bis(trifluoromethylsulfonyl)imide, [PMIM][TFSI], $\geq$ 99%) from IoliTec-Ionic GmbH, and a polar non-toxic solvent to dissolve and mix PVDF with the IL, dimethylsulfoxide (DMSO, 99.9+% ACS) from Thermo Fisher Scientific.

The PVDF/IL films were prepared using the protocol outlined by André et al. [27]. First, DMSO and IL were mixed and after achieving a homogeneous mixture, PVDF was added to the solution. The ratio between the materials is 40% w/w IL/PVDF and 12/88% w/w PVDF/DMSO. The solution in a sealed beaker was stirred within a thermal bath maintained at 50°C. Once a transparent and homogeneous solution was achieved, the mixture was poured onto a glass substrate. The wet film thickness was defined as 0.6 mm through the doctor blade technique (using an applicator, Proceq ZUA 2000). For total solvent evaporation, the wet film was taken into the oven, set at 85°C to avoid the formation of pores [28], and to allow a better piezoelectric β phase crystallization [29]. Figure 5.2a provides a schematic representation summarizing this fabrication process.

For voltage application, thin gold leaf sheets were laminated on both faces of the samples using a hot-press printing technique. PVDF/IL films were sandwiched between the gold sheets on the top and the bottom faces and compressed at 180°C for 45 seconds (Figure 5.2b). Figure 5.2c shows the PVDF/IL film (with a thickness of 0.06 mm) both before and after the gold lamination step.

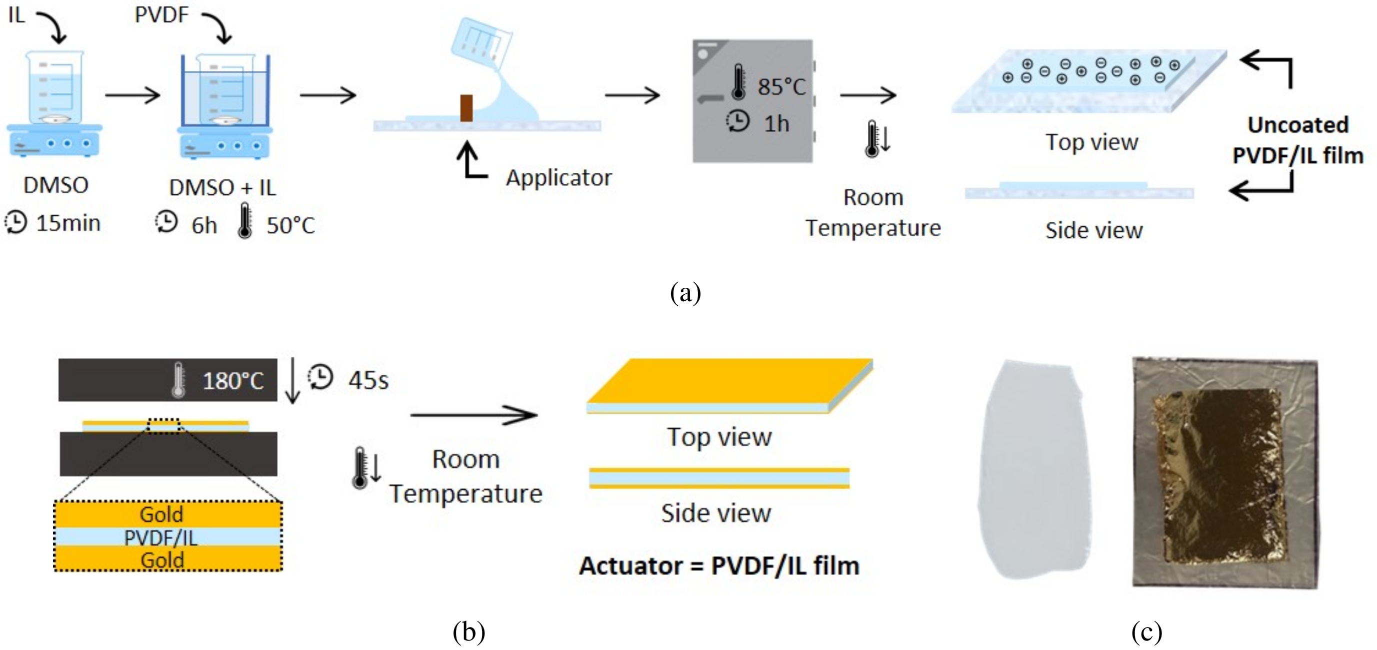


Figure 5.2: Fabrication process: (a) Schematic illustration of PVDF/IL film development; (b) Gold-layer heat printing; (c) Photograph showing the uncoated sample (left) and gold-coated sample (right).

5.3.2 Electrical Characterization

The capacitance (C) and ohmic resistance (R) of the PVDF/IL film were measured using an LCR meter (Agilent E4980A) across a frequency range of 20 Hz to 2 MHz and a voltage intensity (V_{AC}) of 1 V. Circular electrodes with a diameter of 5.3 mm were used on both faces of the samples for the measurements. Based on these results, we determined the real part of the relative permittivity (ε') using $\varepsilon' = \frac{C \cdot d}{\varepsilon_0 \cdot A}$ and AC conductivity (σ_{AC}) using $\sigma_{AC} = \frac{d}{R \cdot A}$, where d represents the thickness of the samples, ε_0 stands for the permittivity of vacuum (8.85×10^{-12} F/m) and A denotes the electrode area.

5.3.3 Electromechanical Analysis

The piezoelectric constant (d_{33}) of the PVDF/IL composite was determined using the piezoresponse force microscopy (PFM) technique. Local piezoresponse and polarization switching spectroscopy of the PVDF/IL film were conducted with a commercial scanning probe microscope (Veeco Multimode Nanoscope IV microscope, Houghton, MI, USA). A conductive probe (NSG10/Pt, NT-MDT) with a spring constant of 14 N/m was used and the piezoresponse force microscopy out-of-plane images were scanned in the single frequency mode at 7.5 V and with a frequency of 35 kHz.

We studied the electromechanical responses of the PVDF/IL film samples cut in a rectangular shape (6x30 mm^2). The samples were displaced vertically in a cantilever beam configuration, constrained at the top end and free at the bottom end, where a laser was pointed for the measurements. For sinusoidal stimulations, we used input signals with frequencies of 0.11, 0.2, 1.1 and 2 Hz and voltages of 5, 10, 15 and 20 V_{pp} for the duration of 90 seconds. Actuation signals were generated by a function generator (Multicomp Pro MP750064) and the resultant displacement was measured using a laser displacement meter (controller Keyence LK-G3001P + laser Keyence LK-G152) directed to the tip of the samples. Each PVDF/IL sample was tested once for a set of frequency and voltage. For analysis at 20 V, the mean of two sample measurements was used for each frequency due to higher movement variance in this voltage. We considered the first 15 peaks of each trial for the mean sine displacement $\pm$ standard deviation (STD) analysis, excluding the first and last peaks to ensure movement stability. The displacement at each peak (crest or trough) was regarded as the peak-to-peak amplitude.

We investigated the active displacement of the samples with a single square pulse per duration of 1, 5 and 10 seconds. Each pulse duration was applied at least 5 minutes apart to ensure complete relaxation of the samples, while the voltage amplitude was maintained at 10 V. This also includes phased displacement analysis, which reflects out-of-phase rising or relaxation of the samples. Each case was tested with 5 pulses.

The rising speed, from the time of pulse application to the moment of maximum displacement within the pulse duration, and the relaxation speed, calculated from the end of the pulse to the minimum position of the samples, were investigated at different pulse amplitudes (2.5, 5, 7.5 and 10 V). These signals were applied to 3 samples in single pulses lasting for 1 s each. Additionally, the reversal motion speed was determined by applying a bipolar square pulse 9 times, distributed across 3 samples with at least 5-minute intervals between each pulse. This measurement indicates how quickly the sample returns to its initial position after the polarity of the signal is inverted. The square waves had a period (T) of 2 s and a voltage amplitude of 20 V_{pp}.

To investigate the performance of the samples under continuous cyclic stimulation, we applied periodic sine waveforms at 0.2 Hz and input voltages of 10 V_{pp} to the actuators for 25 minutes.

The actuation force was investigated using a setup where the actuators displaced against a compression spring with a spring constant of $k = 0.0294$ N/mm (5055, Misumi) during the first half of the biphasic square pulse. A total of 9 pulses, each with a voltage of 20 V_{pp} and a period (T) of 2 were applied. The contact force between the actuator and spring is equal to the compression force of the spring, which is directly proportional to the decrease in length of the spring (x) in Hooke's law: $F = -k \cdot x$

All data was collected using a data acquisition device (National Instruments USB-6001) and subsequently processed in MATLAB.

5.4 Results and Discussion

Figure 5.3a shows the real part of the relative permittivity (ε') and conductivity σ_{AC} measured for the PVDF-IL film. The data shows that ε' decreases with the frequency, from $1.83x10^3 \pm 156.96$ to 2.91 ± 0.11, while the electrical conductivity (σ_{AC}), increases with frequency from $.86x10^{-6} \pm 2.37x10^{-7}$ S/m to $2.50x10^{-4} \pm 5.80x10^{-6}$ S/m. These trends are consistent with those reported in the literature for hybrid piezo-ionic materials

[22, 30]. This behaviour is attributed to the presence of IL in the polymeric matrix. In contrast, pure PVDF does not exhibit such frequency dependence with σ_{AC} remaining at 1.61×10^{-08} S/m and ε' at 10, considering a frequency range of 100 Hz - 1 MHz [22]. Higher values of σ_{AC} represent higher electrical losses in the piezoelectric film, while higher values of ε' indicate the material's enhanced capability to store electrical energy [31].

Figure 5.3b shows the topography of the PVDF/IL film obtained from the scanning of the surface, showing an average roughness of $\approx$120 nm. This measurement employs the standard contact mode atomic force microscopy (AFM) to reveal the topography of the samples. In comparison, the pure PVDF films show a lower average roughness of around 30 nm (see supplementary figure A.2a). The increased roughness in the PVDF/IL films can be attributed to the incorporation of the ionic liquid, which induces greater surface irregularities. Moreover, we conducted PFM measurements to evaluate the local piezoelectric response of the PVDF/IL and PVDF sample. This technique is based on the standard contact mode AFM setup, in which the cantilever and the tip are electrically conductive, and an alternating voltage is applied to the tip, evaluating the electromechanical properties in addition to the sample topography. Figure 5.3c shows the nanoscale topography of the PVDF/IL film through the PFM technique, with the corresponding topography for the pure PVDF samples shown in supplementary figure A.2b.

Moreover, figures 5.3d and 5.3e show out-of-plane amplitude and phase images, respectively for PVDF/IL samples, with the corresponding images for pure PVDF provided in the supplementary figure A.2c. These images were obtained over a scan area of 20x20 μm in the virgin state of the film. The PFM amplitude represents the piezoelectric displacement of the sample when subjected to an AC voltage, while the PFM phase specifies the orientation of polarization. The observed weak contrast regions suggest a low piezoelectric response, possibly due to the lack of the polar β phase in the scanned region. Further investigations were performed by applying a DC bias voltage of $\pm$150 V to the tip, effectively inducing microscale tip-induced poling as a means of artificially writing domains. As seen in figure 5.3f, opposite bright and dark contrasts were observed, though with some instability which can be attributed to the higher conductivity of these films. This becomes more apparent when compared to the results for pure PVDF samples, as shown in the supplementary figure A.2d.

To further study the local piezoelectric response behaviour, we investigated the hysteresis loops on the PVDF/IL and pure PVDF films (Figure 5.3g) using a cantilever tip. These loops were obtained in pulse mode by sweeping the DC bias voltage (U_{dc}) between ±150 V. The results confirm a negligible difference in the ferroelectric characteristics of the film at the nanoscale, with coercive voltages of 55 V for PVDF and 25 V for PVDF/IL, respectively. This reflects only a marginal variation in the d_{33} coefficient values, approximately 0.8 pm/V for PVDF and 0.6 pm/V for PVDF/IL.

Infrared spectroscopy also confirms that the presence of IL leads to a higher percentage of electroactive phase. Supplementary figure A.3 shows the FTIR-ATR analysis of PVDF samples with and without IL. Despite similar transmittance (%T) values at 840 cm^{-1} (corresponding to EA phases), a more subtle peak at 766 cm^{-1} (typical of non-EA phases) suggests a higher EA phase content: 80% in PVDF/IL samples compared to 69% in PVDF samples, as determined by Beer-Lambert law.

The experimental setup used to characterise the displacement of the PVDF-IL samples is shown in Figure 5.4a. Figure 5.4b shows the mean sinusoidal displacement results, investigating the effect of altering input voltage and frequency. In general, higher voltages result in greater peak displacements across all tested frequencies. This is because higher voltages induce stronger polarization, leading to increased displacement, where lower frequencies allow the samples sufficient time to respond due to their relatively slower motion. However, the peak displacement at 20 V_{pp} decreases at higher frequencies because faster frequency induces more rapid polarization changes, limiting the samples response. Supplementary videos A.4 and A.5 show typical displacements for high and low frequencies, 2 Hz and 0.02 Hz, respectively, while actuating with 15 V.

A comparison of the electromechanical response of our IL/PVDF film with other

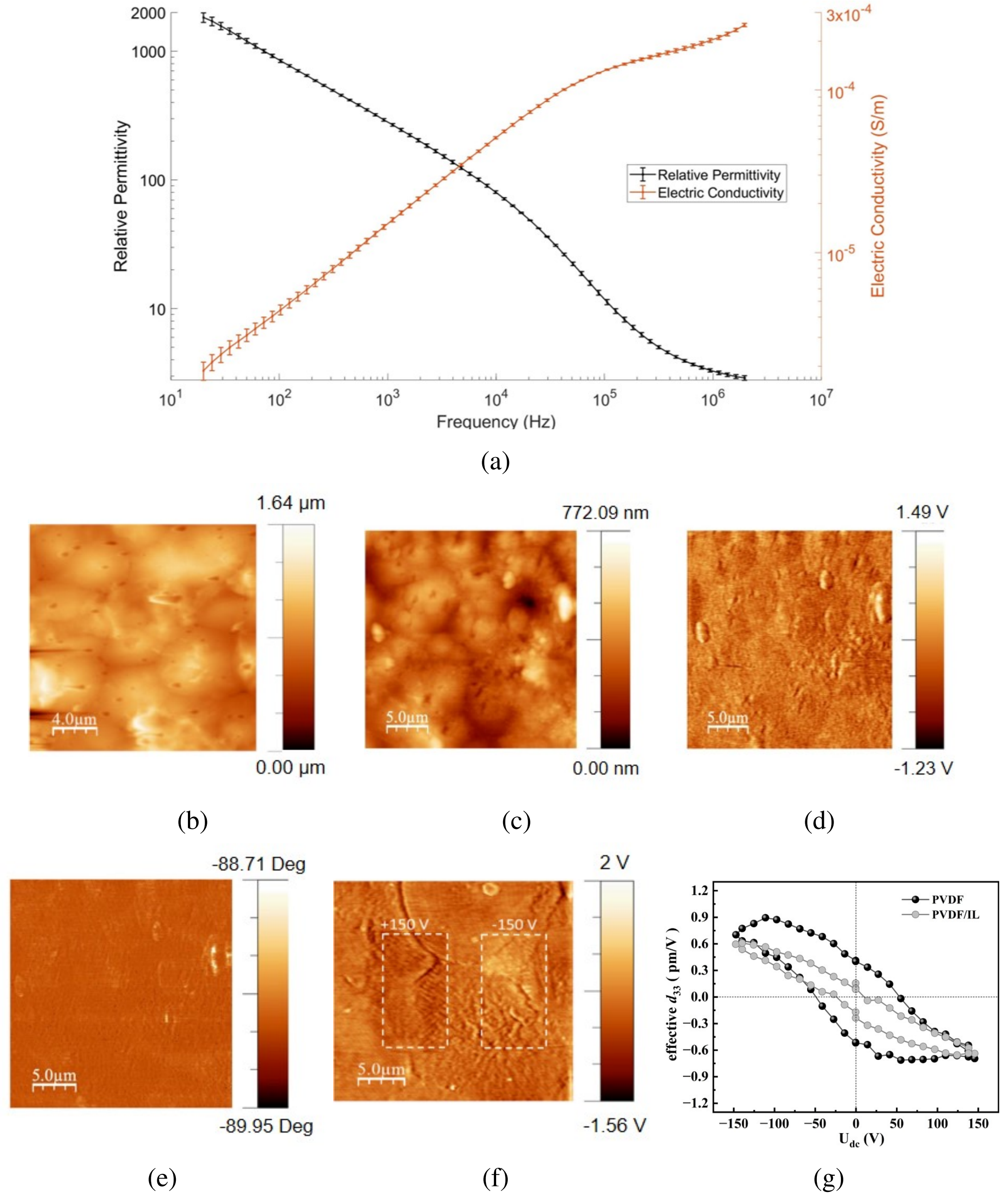


Figure 5.3: Electrical and piezoelectric characterization: (a) Relative permittivity and electric conductivity of the PVDF/IL actuators as a function of frequency; (b) AFM topography image of the PVDF/IL film; (c) PFM Topography; (d) Out-of-plane amplitude (A*cosθ); (e) Out-of-plane phase images; (f) PFM amplitude image after applying ±150 V to create artificial domains; and (g) Piezoresponse hysteresis loop obtained by PFM. Error bars in the first figure represent the standard deviation of relative permittivity (black line) and electric conductivity (orange line).

PVDF-based piezoelectric actuators with a high electroactive phase, highlights the role of IL in reducing the required input voltage for achieving greater displacement. For example, in a previously published study [18] involving P(VDF-TrFE) copolymer with similar dimensions (25x5 mm) and thickness (≈50 μm) subjected to a comparable frequency (≈1 Hz), a displacement of 0.8 mm was achieved with a sinusoidal input. In contrast, our IL/PVDF film achieved a displacement of 0.67 mm, but required a much lower electric field (0.25 MV m^{-1} compared to 50 MV m^{-1} in the P(VDF-TrFE) study) to achieve comparable displacement performance.

Figure 5.4c shows the displacement response of the film under pulse input. It shows active displacement, denoted as the highest displacement achieved by the sample during the pulse, and phased displacement, denoted as the rising displacement after the end of the pulse (as a positive displacement) or the relaxing displacement during the pulse after the maximum position (as a negative displacement). The results show greater active displacements for higher pulse duration. Exposing the soft actuators to different pulse durations leads to distinct behaviours: with shorter pulse durations (e.g. 1 s), the samples continued to rise after the end of the pulse, achieving a post-pulsed displacement higher than the one achieved during the pulse - see the top inset graph from the typical displacement result in shorter pulse duration. This could be attributed to the presence of IL in the PVDF network, partially through increasing hysteresis [32], which is associated with response delay and remnant polarization [33]. Moreover, it boosts ε' leading to increased energy storage during the pulse duration [31].

In contrast, longer pulse durations lead to a decrease of displacement immediately after reaching the maximum active displacement (figure 5.4c, bottom inset). In this study, the loss increases with the pulse duration. The observed negative phased displacement could be attributed to the dielectric loss, wherein the polarization can not be sustained. The presence of IL amplifies this dielectric loss, since the σ_{AC} increases with its addition and it is related to low IL density [22]. Moreover, compared to other phases (such as α), the β phase is associated with greater dielectric loss [34]. Figure 5.4d compares the displacement behaviour of three different samples when subjected to different pulse durations with all data filtered to minimise noise. The comparison between raw and filtered curves is detailed in the supplementary material (supplementary figure A.6).

For the 5 s and 10 s pulses, a different phenomenon is observed. After the end of the pulse, and the relaxation of the actuators, they show a secondary rise to a new peak before gradually returning to their initial position. This effect is known as the memory effect which can be attributed to the presence of IL that cross-links PVDF [35]. Moreover, the Joule effect provides a complementary explanation, as it leads to an increase in the internal temperature of the samples when current is passing through the film. Therefore, after the pulse, the temperature gradually decreases, which prevents a rapid return to the initial position [36].

Figure 5.4e shows the rising and relaxation speeds for pulse amplitudes of 5, 7.5 and 10 V. We did not observe a noticeable response for 2.5 V. Supplementary figure A.7 presents the original and filtered curves for each case. Both rising and relaxation speeds show an increase in pulse intensity, which is related to higher polarization. While 10 V pulse resulted in higher rising and relaxation speeds, it also showed higher standard deviation values. Particularly, at 7.5 V, the relaxation speed is notably lower than the rising speed. This disparity is attributed to the positive phase displacement occurring after the pulse ends, which delays the relaxation speed. In such cases, the relaxation speed was calculated by identifying the minimum decay position following this positive phased displacement (supplementary figure A.8). When employing a negative pulse to instantly reverse the actuation, as shown in figure 5.4f, the reversal motion speed showed a faster and consistent response of 9.39±2.06 mm/s. This suggests that bipolar square pulses could serve as a viable option to ensure a consistent relaxation response for practical applications such as haptic feedback devices.

Figure 5.4g shows the results of the cyclic test conducted to evaluate actuation stability under continuous sinusoidal input. This shows no significant deterioration in peak-to-peak amplitude during 300 cycles of a sine wave input at a voltage amplitude of 10 V_{pp} and a frequency of 0.2 Hz.

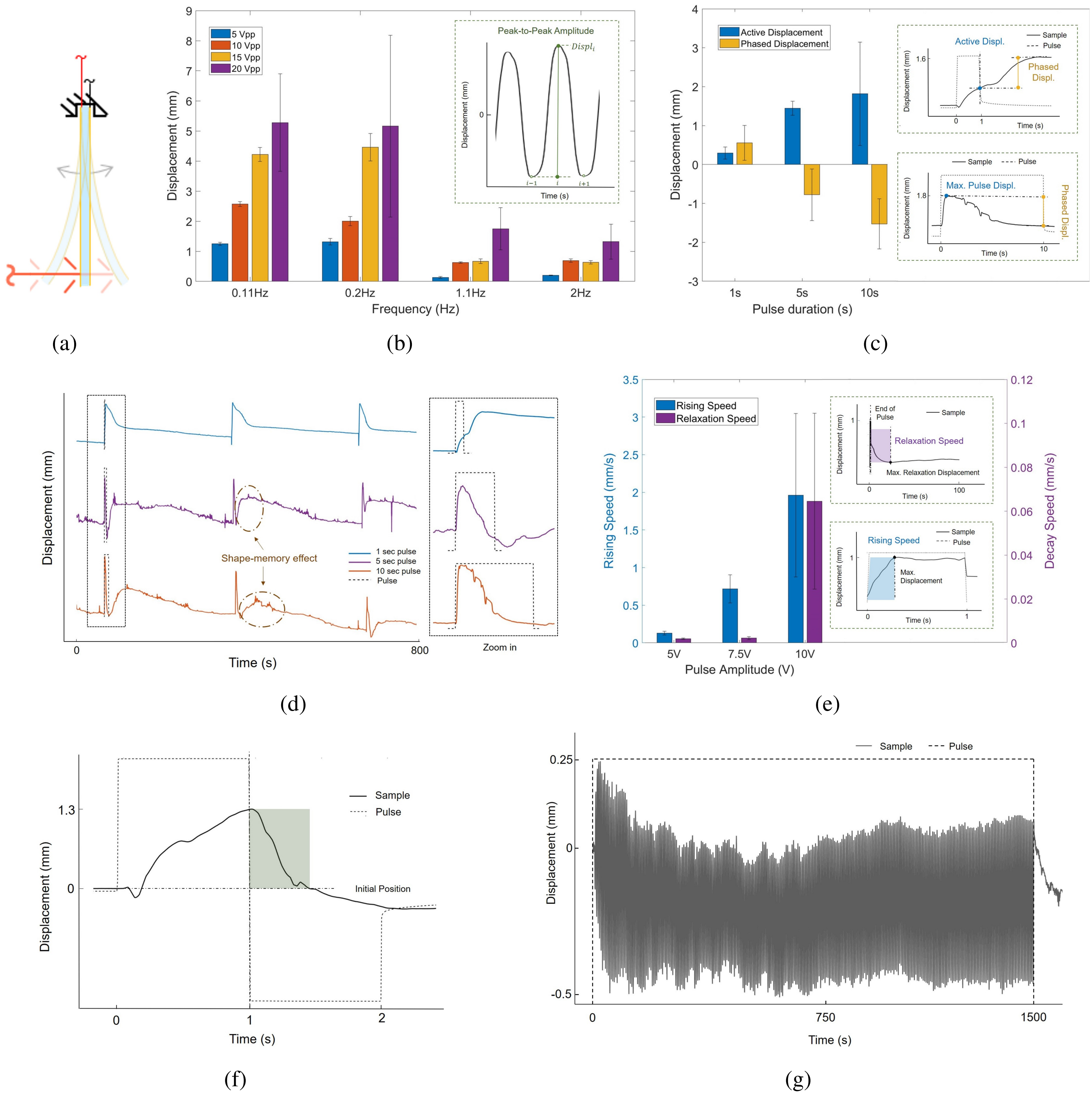


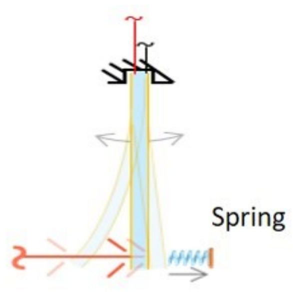
Figure 5.4: Electromechanical analysis of the PVDF/IL cantilever beam: (a) Experimental setup; (b) Displacement for sinusoidal input; (c) Displacement for different pulse durations, highlighting active and phased displacements; (d) Typical displacement behavior for square pulses; (e) Rising and relaxation speeds; (f) Reversal motion speed with bipolar pulse stimulation; (g) Cyclic test with a sine wave.

To quantify the generated force (Figures 5.5a and 5.5b) our experiments showed that the PVDF/IL can compress the spring by 0.31±0.11 mm, which corresponds to 11±3 mN according to the Hooke law. It falls within the typical threshold values for cutaneous touch perception (10-100 mN) as reported by [37], demonstrating its potential use for haptic feedback, especially considering its relatively quicker actuation response compared to the ionic actuators. Gaihre et al [38] studied the force produced by a PVDF/IL ([Li][TFSI]) actuator coated with polypyrrole (PPy) electrodes. They found a blocking force of 1.5 mN at the tip of the actuator, considering a normalized actuator dimension of 0.196 width, 0.981 length and 0.006 thickness, in comparison to the PVDF/IL developed with this study (0.196 width, 0.980 length and 0.002 thickness).[§]

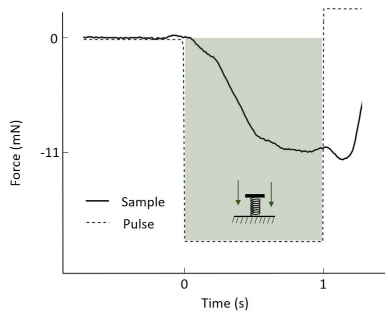
5.4.1 Finger Sleeve Design

Figure 5.5c and 5.5d show the initial design and prototype of the finger sleeve to assess haptic feedback. The sleeve is 3D printed with a flexible thermoplastic polyurethane filament, incorporating two IL/PVDF cantilever beams for force feedback. These actuators are positioned equidistantly from the finger's center, enabling simultaneous and independent feedback application on both sides. This setup offers the flexibility of applying distinct inputs to each side of the finger (left and right). The supplementary video A.9 showcases the prototype in operation, demonstrating its response to a sine input wave. A comprehensive user study will be carried out to evaluate the detectability of the applied force in different scenarios.

[§]The study of Gaihre et al was only considered for the discussion on the present dissertation as a comparison of the results achieved. It is not present in the submitted/published paper.



(a)



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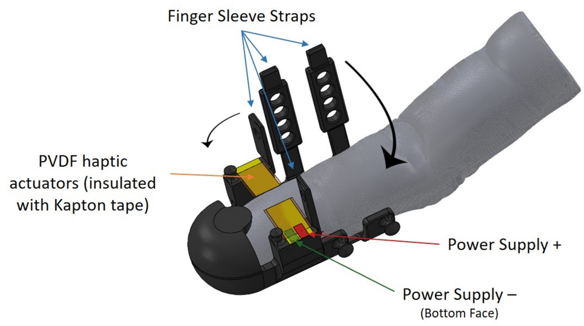

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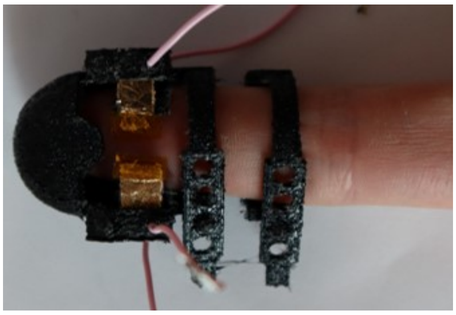

(d)

Figure 5.5: Force measurement for haptic feedback: (a) Set-up for force measurement; (b) Force result achieved from the compression of the spring. (c) Finger sleeve design; (d) Photo of the fabricated sleeve.

5.5 Conclusions and Future Works

The present study provides a critical analysis of the electromechanical response of a hybrid piezoelectric-ionic film for potential haptic feedback. The inclusion of ionic liquid into the PVDF network offers several advantages: Firstly, it enhances the electroactive phase during the polymerisation process, thereby boosting the piezoelectric effect, as observed in our sinusoidal displacement studies. Additionally, the addition of the IL reduces the required input actuation voltage, although it becomes prone to failure at voltages higher than 20 V peak to peak. The addition of IL also lowers the Young's modulus of the actuator, resulting in greater displacement. The addition of ionic liquid also causes the electrical parameters, σ_{AC} and ε', to vary significantly with frequency, which leads to higher dielectric loss and energy storage at specific frequencies. Moreover, it increases hysteresis in PVDF/IL piezoelectric responses. These effects result in phased displacement, which could make the control of these hybrid actuators more challenging. Nevertheless, using a shorter input pulse with reversed pulsing for active relaxation can offer a more stable solution.

Further studies on the type of ionic liquid and optimization of the structure and fabrication process are necessary to fully comprehend their electromechanical effect in the hybrid design for optimising actuation performance. Furthermore, user-studies are essential to assess the feasibility of using this technology for haptic feedback.

Author Contribution: Conceptualization, A.D.A., P.M. and M.T.; Methodology, A.D.A., M.T.; Formal analysis, A.D.A.; Investigation - A.D.A, P.M., M.T., I.C., I.B., K.B.V. and R.M.R.P.; Resources - P.M. and M.T.; Writing - Original Draft, A.D.A.; Writing - Review & Editing, all authors; Supervision, M.T.

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Conflict of Interest: The authors declare no competing financial interest.

5.6 Complementary Analysis*

An additional measurement was done considering Berlincourt method (BM) (quasi-static movement of the piezometer) for the piezoelectric constant, d_{33}. The samples were manufactured using the same experimental protocol described in the present chapter.

The thin films, with dimensions of 20x20 mm^2, were poled a priori. A custom-made setup was used to perform corona poling, where a needle (connected to the high voltage of 2.2 kV) was scanned across the samples placed on a grounded metal base. Scanning was along a vertical direction at a constant speed (1 mm/s) with horizontal steps of 250 μm and a needle-substrate gap of 2 mm (Figure 5.6). Then, the piezoelectric charge coefficient of the poled PVDF/IL films were measured using a piezometer (SinoCera XE2730A d_{33}-meter) under an operating frequency of 110 Hz and an applied force of 250 mN.

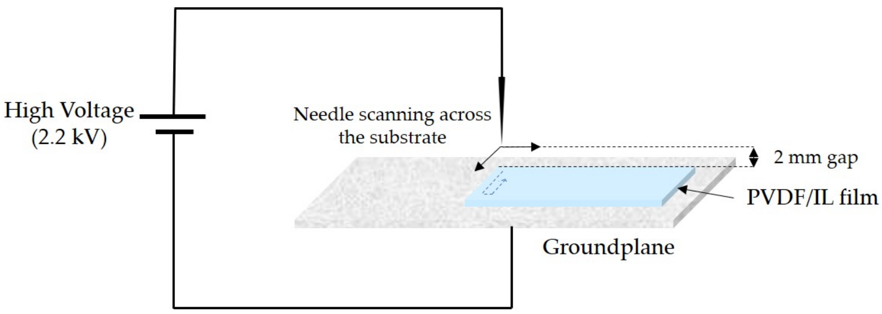

Figure 5.6: Schematic representation of corona poling experiment.

The macroscopic value measured for d_{33} was 7 pC/N. BM technique involves the analysis of many grains and measures the direct effect, in contrast to PFM measurements, which considers few domains and is based on the principle of converse piezoelectric effect [39].

In the literature, the piezoelectric constant is also reported considering bulky methods, such as BM. For example, Sukumaran et al [40] achieved values ranging from 7.2 to 11.9

*The Berlincourt Method was performed in collaboration with I.C., I.B., K.B.V. and R.M.R.P. This complementary analysis is not part of the submitted/published paper, however, it complements the electrical characterization.

pC/N in PVDF/IL samples. According to the authors, the samples have inherent potential for energy harvesting applications.

The results are very close, which shows that the properties of the material developed (PVDF/IL thin films) are in accordance with the literature.

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Chapter 6

In-Silico User Study Case: Wearable Feedback Haptic Device for Rehabilitation

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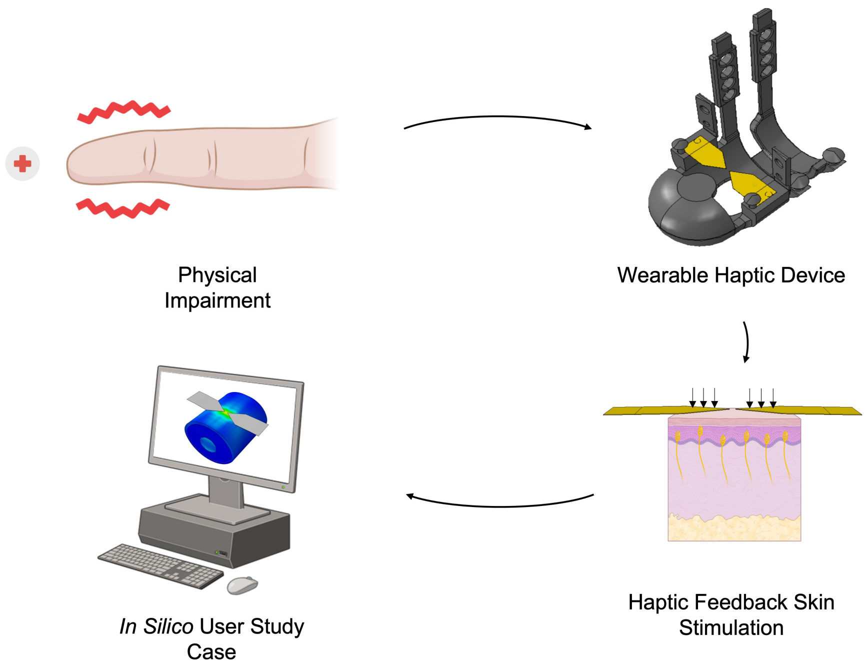
6.1 Abstract

Soft smart materials have useful properties for addressing everyday problems affecting human health and well-being, having a positive societal impact. For instance, these materials can serve as sensors for breath monitoring or as soft actuators to stimulate muscles impaired by injury or illness. A notable example of their versatility lies in piezoelectric materials, which can function both as passive elements (utilising the direct piezoelectric effect) and as active elements (employing the converse piezoelectric effect). This dual functionality showcases the broad potential of smart materials in various applications. The present study is an *in silico* simulation of a wearable piezoelectric material (polyvinylidene fluoride - PVDF), using finite element analysis (FEA) to evaluate the effectiveness of the touch sensation provided by the haptic device on human skin, using different actuators geometries and voltage input intensities. Moreover, the main active element, a PVDF-based soft actuator, was fully characterised in terms of the piezoelectric matrix, using an inverse finite element approach. In conclusion, the findings point to promising results when using this haptic technology for re-educational therapies.



Graphical abstract.

6.2 Introduction

According to the World Health Organisation (WHO) [1], in 2022, approximately 1300 million people experience some kind of physical disability. A significant part of these mobility impairments require or will need in the future some sort of therapeutic rehabilitation, motion training or motion assistance during a period of time after the onset of disorders [2].

During the last decades, the medical and research community have been trying to mitigate this problem and accelerate the recovery of those who are suffering from lack of mobility [3]. As a result, society's efforts have led to a trend of developing rehabilitative wearable devices, with the possibility to assist without tethering the patient to a specific location. However, when based on soft materials, those devices evidence limited ability to perform motion tasks [2], being essentially developed for haptic stimuli solutions on human skin.

Skin is the largest organ of the body with different functions associated. It is the first barrier against germs, helps to regulate the body temperature and, most importantly, provides the main and the first sensory channel of the body to perceive external stimuli, based on touch.

Through the skin, people can interact with the surrounding objects, feeling their properties, such as weight, temperature, textures and motion [4]. The interaction between skin and objects results in nerve stimuli, which are primarily received by different receptors in the skin and later sent to the brain. Pacinian corpuscles, Meissner corpuscles, Merkel complexes, Ruffini corpuscles, and C-fiber LTM (low threshold mechanoreceptors) are the mechano-receptors that can detect even innocuous stimuli [5], for example, responding to displacements of the skin on the order of micrometres (μm). Different stimuli, such as mechanical, electrical and thermal can feedback those receptors and transmit different sensations and information through the nerves [4].

To take advantage of the high sensitivity of skin, different technologies have been developed, using different powered-up soft materials, for rehabilitation and re-educational therapies through the use of external devices. As reviewed by André et al [6], different soft actuators can be used to provide mechanical, electrical or thermal feedback on the skin, such as electroactive materials (piezoelectric), magnetic responsive materials, thermally responsive materials or even photo-responsive materials. All these smart materials, in particular piezo polymers due to their versatility, can play an important role in actuation for tactile sensation since they are light, flexible and biomimetic.

Piezoelectric materials, in particular polymers such as polyvinylidene fluoride (PVDF), have been largely studied in rehabilitation solutions since they can be used as sensors [7, 8] or actuators [9, 10]. For example, Pan *et al.* [11] developed a mechanomyography (MMG) sensor using PVDF's piezoelectric properties for lower limb rehabilitation exoskeleton. They concluded that the approach used improved the sensitivity in driving the device. Gariya *et al.* [12], developed a pneumatic soft actuator with a PVDF membrane for sensing the bending deformation of the actuator, which was tested for the medical application of human finger rehabilitation.

Nevertheless, the use of PVDF, as a haptic feedback element, has been presented in the literature with distinct purposes. Ege and Balikci [13] introduced a haptic interface using transparent thin films of PVDF actuators to feedback touch displays. Maeda *et al.* [14] developed a wearable haptic augmentation system using a skin vibration sensor made from PVDF. In their studies, they pointed to an increase in haptic sensation of, approximately, 5% when compared to no feedback condition.

The present study aims to perform an *in-silico* user study, as proof of concept using the finite element method (FEM), of a wearable device for re-educational therapies, using PVDF/ionic liquid (IL) as haptic actuators. These feedback elements, with distinct tip geometries (rectangular, circular and triangular), will be fed with different voltage intensities, to study the influence of input voltage on the touch perception. With this approach, the authors hope to understand if the developed methodology can be effectively applied in rehabilitation scenarios and if the skin can be stimulated through gentile touch sensations provided by PVDF/IL's actuators. This work uses the index finger as a reference model, due to its high sensibility [15].

6.3 Inverse Finite Element Analysis

André et al [16, 17] characterised PVDF/IL haptic actuators experimentally, in terms of porosity and microscopic defects (high resolution scans were obtained); mechanical properties (i.e. Young modulus (E) and yield stress (σ_{Yield})); chemical properties (i.e. degree of crystallinity (χ) and electroactive phases (β phase)); and electrical, piezo and conductive properties (i.e. dielectric permittivity (ε'), AC conductivity (σ_{AC}) and piezoelectric constant (d_{33})). However, those properties are not sufficient to define a material model able to reproduce the behaviour observed experimentally, namely the electromechanical performance [17].

To better mimic the experimental phenomenon observed, an inverse finite element analysis approach was used to evaluate the piezoelectric matrix.

6.3.1 Piezoelectric Constitutive Equations

The standard form of the piezoelectric constitutive equations can be presented in four different forms by taking either two of the four field variables as independent. Considering a tensorial representation of the strain-electric displacement form and the components of stress and electric fields as the independent variables [18–20], it comes

$$S_{ij} = s^{E}_{ijkl} \cdot T_{kl} + d_{kij} \cdot E_k \tag{6.1}$$

$$D_i = d_{ikl} \cdot T_{kl} + \varepsilon^{T}_{ik} \cdot E_k \tag{6.2}$$

where the index i, j, k, l represent the different components or directions in the tensor equations, S_{ij} are the strain components, D_i are the electric displacements, s^E_{ijkl} are the elastic compliance constants, T_{kl} are the stress components, d_{kij} are the piezoelectric constants of the material, ε^T_{ik} are the permittivity constants and finally E_k is the electric field component.

In a matrix form, the equations (6.1) and (6.2) are given as [21],

$$\begin{bmatrix} S \\ D \end{bmatrix} = \begin{bmatrix} s^E & d^t \\ d & \varepsilon^T \end{bmatrix} \cdot \begin{bmatrix} T \\ E \end{bmatrix} \tag{6.3}$$

where the superscripts E and T denote that the respective constants are evaluated at constant electric field and constant stress, respectively, and t represents the transpose.

Assuming the study of a piezoelectric thin film, such as the case under analysis, matrix 6.3 can be further simplified. If the thin structure is assumed as a thin beam, based on the Euler-Bernoulli beam theory or Rayleigh beam theory [21], the stress components T_{22}, T_{33}, T_{23}, T_{13} and T_{12} are negligible ($T_{\neq 11} = 0$), since only the one-dimensional bending stress (T_{11}) has a non negligible value. Moreover, if the electrodes are placed perpendicular to the 3-direction, equation (6.3) becomes

$$\begin{bmatrix} S_{11} \\ D_{33} \end{bmatrix} = \begin{bmatrix} s^E_{11} & d_{31} \\ d_{31} & \varepsilon^T_{33} \end{bmatrix} \cdot \begin{bmatrix} T_{11} \\ E_{33} \end{bmatrix} \tag{6.4}$$

Matrix (6.4) shows that the piezoelectric constant, d_{31}, is crucial and also needed to characterise properly the piezo polymer PVDF/IL thin film.

6.3.2 Piezoelectric Matrix

Overall, the piezoelectric matrix of PVDF is given by equation (6.5), since the material evidences anisotropy [22, 23]. That fact implies that the piezoelectric properties change with direction [22].

$$d_{ij} = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{bmatrix} \tag{6.5}$$

where the subscript i refers to the direction of plane polarisation, while j is the direction of the induced strain.

However, in the literature, the piezoelectric properties of PVDF more commonly used to characterise the polymeric material are d_{31} and d_{33} [24, 25]. The d_{31} constant is the transverse coefficient, which defines the mechanical strain created in the perpendicular direction to the applied electric input; while d_{33} is the longitudinal coefficient, which defines mechanical strain in the same direction as the applied stress [26]. With a good approximation, the two coefficients are enough to define the phenomenological behaviour observed experimentally. Considering that fact, for this particular study case, matrix (6.5) could be simplified into matrix (6.6), ignoring d_{32} coefficient and the rotational and shear components (d_{24} and d_{15}).

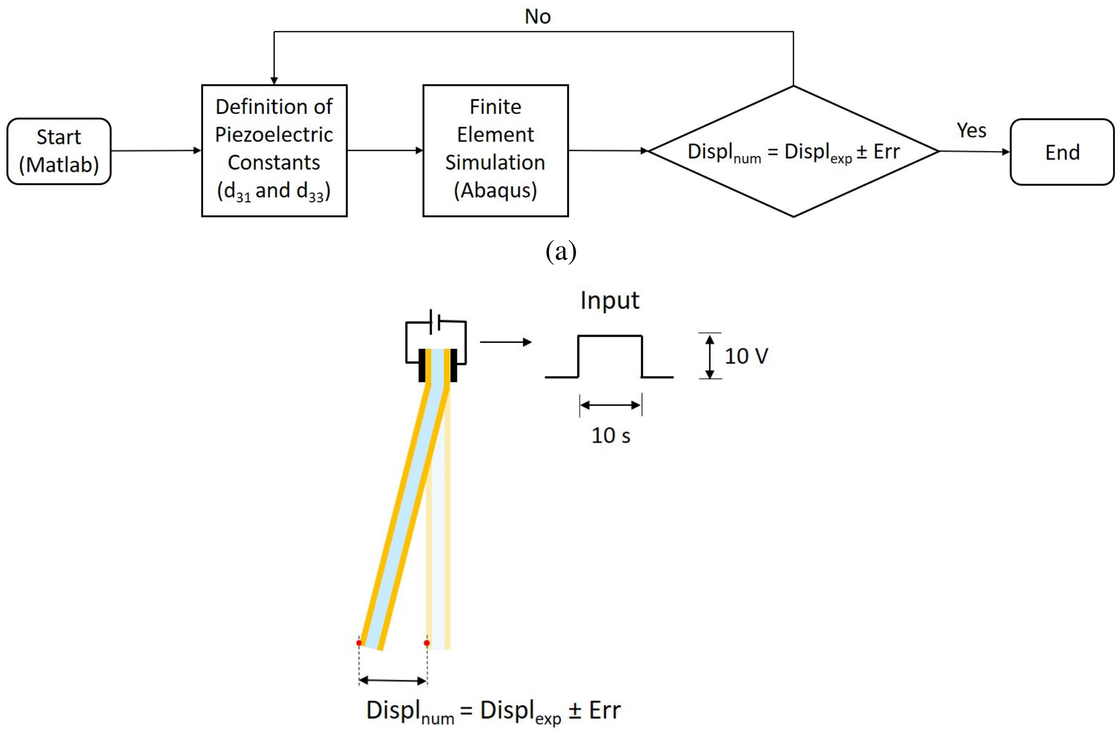
$$d_{ij} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ d_{31} & 0 & d_{33} \end{bmatrix} \tag{6.6}$$

6.3.3 Inverse-FEM Algorithm

In recent years, the identification and characterisation of the piezoelectric matrix using inverse FEM algorithms have grown and gained popularity [27]. Figure (6.1a) summarises the steps adopted to approximate numerically the piezoelectric constant d_{31}.

In general, the optimisation algorithm runs until achieving a d_{31} value that grants similar numerical behaviour when compared to the experimental behaviour. The experimental displacement (Displ_{exp}) measured at the tip of the PVDF/IL sample, when electrically stimulated with a square pulse of 10 V for 10 seconds, was 1.86 mm [17]. The optimisation algorithm optimised the initial d_{31} parameter guess (15x10^{-12} pm/V [28]) until the exit optimisation criteria is satisfied: numerical displacement (Displ$_{num}$) is equal to the Displ$_{exp}$ more or less the error (Err = 15%) (Figure 6.1b).

Applying an iterative refinement method [29], and after 8 iterations, the optimised d_{31} was 3.75x10^{-07} pm/V, with a Displ$_{num}$ of 1.87 mm (error of 0.8%).



(b)

Figure 6.1: (a) Flow chart for d_{31} optimisation; (b) Exit optimisation criteria when Displ$_{num}$ = Displ$_{exp}$ $\pm$ Err.

6.4 *In-Silico* User Study Case

A numerical model of the wearable haptic device was developed to simulate the stimuli contact and interaction between an index finger and the device, through gentile and soft haptic touch sensations.

The finger sleeve prototype used was first designed by André et al [17]. Briefly, the finger sleeve prototype was developed considering the usage of a thermoplastic polyurethane (TPU) as the main structural material. This choice grants the finger device with flexibility and high elasticity. Moreover, TPU can be used as a 3D printable material in fused filament fabrication (FFF) technique, which allows fast prototype iterations. All these properties make this material the ideal candidate for its purpose.

The prototype allows adaptability to patients' physiognomy since it has adjustable straps. As a result, the device can be adapted to different finger shapes (Figure 6.2).

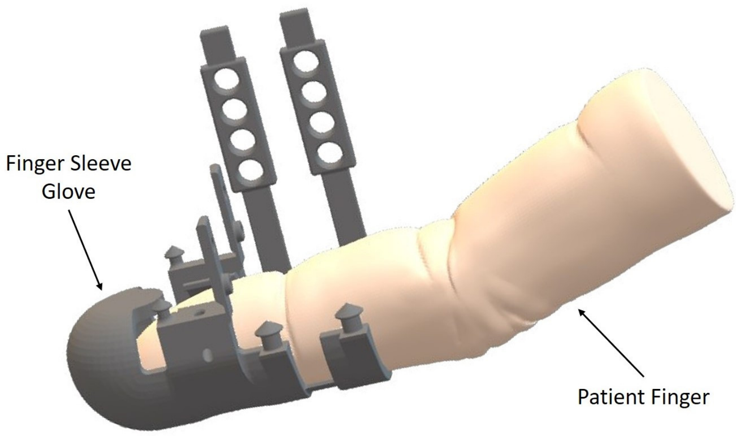


Figure 6.2: Schematic illustration of the wearable device prototype (finger sleeve) worn by an index finger.

As the PVDF/IL haptic actuators were previously characterised [16, 17, 30], as well as the finger sleeve was properly designed [17], an *in-silico* numerical model was developed on Dassault Systemès Abaqus [31] to study the device's behaviour in real scenarios. Afterwards, real interaction scenarios between the user and the haptic feedback device were simulated using a FEM approach.

6.4.1 FEM Model

The numerical model was defined in Abaqus 2022 [31]. The elements needed to perform the *in-silico* user study case were defined, i.e., the mechanical properties, interactions between parts, the boundary conditions (BCs) and the mesh.

The physiognomy of the finger model was based on literature [32–34] and drawn in Dassault Systemès SolidWorks 2020 [35]. It is composed of bone, soft tissue and skin with corresponding properties based on the literature [32, 36–38]. The wearable haptic device was modelled considering PVDF/IL soft actuators (with 6 mm width, w, 12 mm total length, l_t, 9 mm free length, l_f, and 0.06 mm thickness, t) and a TPU finger sleeve. The properties of those materials were also based on the literature [16, 17, 30, 39]. Different tip shapes for the actuation were considered, as shown in Figure 6.3a, such as rectangular, circular (with radius, R, of 4.3 mm centred at the middle of the free length) and triangular (with different tip pronunciations, started at length l=0, l=1/4, l=2/4, l=3/4 and finally l=4/4 from the free tip). Table 6.1 summarises the properties of the parts used to simulate the *in-silico* user study case.

Table 6.1: Properties of the parts used in the numerical model in Abaqus.

Parts	**Density, ρ (kg/m^3)**	**Young Modulus, E (MPa)**	**Poisson Ratio, ν (adimensional)**	**Elect. Conductivity, σ_{AC} (S/m)**	**Piezoelectric Const., d_{33} (pm/V)**	**Piezoelectric Const., d_{31} (pm/V)**
Bone [36]	1900	17x10^3	0.3	-	-	-
Soft Tissue [32]	1000	0.08	0.4	-	-	-
Skin [37, 38]	-	2.5	0.48	-	-	-
TPU finger sleeve [39]	-	2.41x10^3	0.3847	-	-	-
PVDF/IL soft actuators [16, 17, 30]	1425.9	127.0	0.18	1.98x10^{-07}	0.6	3.75x10^5

The FEM model was prepared and assembled in Abaqus, considering the wearable haptic device and the finger model (Figures 6.3b and 6.3c). Since the finger sleeve glove is not an active part of the simulation, it was not considered, resulting in a faster numerical simulation. Moreover, the contact and interaction properties, as well as the node ties were defined among all the active parts (PVDF/IL actuators ⇔ skin - contact/interaction; skin ⇔ soft tissues - tie; soft tissues ⇔ bone). The BCs were applied in the anterior and posterior sections of the finger model and on both PVDF/IL actuators. Since the finger sleeve glove is not present in the simulation, the BCs applied on the actuators satisfy the interaction with the glove. The electrical inputs were also applied in both PVDF/IL soft actuators, assuming a square pulse with different voltage intensities with a time duration of 10 seconds (Figure 6.3d). The active elements/parts of the simulation were then meshed. The finger model was meshed considering 3D stress hexagonal mesh elements (C3D8R - 8 node linear brick, reduced integration). On another hand, the PVDF/IL soft actuators were defined using piezoelectric tetrahedral mesh elements (C3D4E - 4 node linear piezoelectric brick). The model's number of elements and nodes changed according to the tip geometry (rectangular, 15303 nodes and 21728 elements; circular, 18225 nodes and 30958 elements; or triangular, 16913 nodes and 26673 elements). Figure 6.3e shows the mesh applied to a specific tip geometry situation.

6.5 Results and Discussion

The *in-silico* user study case was carried out to understand the effectiveness of the touch sensation provided by the finger sleeve prototype on the skin to be used in the context of re-educational therapy. Both PVDF/IL soft haptic actuators were input simultaneously under different initial conditions, such as the tip geometries (rectangular, circular and triangular; initiated at the middle of the free part of the actuator) and pulse intensities (2.5, 5.0, 7.5 and 10.0 V). The numerical results achieved were, then, compared against the minimum touch sensation felt by human fingers described in the literature.

6.5.1 FEM Simulation

The FEM simulation of the *in-silico* user study case, considering different voltage intensities and tip geometries, evidenced results according to our expectations. Figure 6.4 shows the results for the different tip geometries tested, considering the maximum voltage intensity (10.0 V). The best touch sensation provided by the finger sleeve prototype was achieved for the triangular tip geometry of the PVDF/IL soft actuators. According to the simulation, that value was 578.0 Pa on the skin finger model (Figure 6.4c).

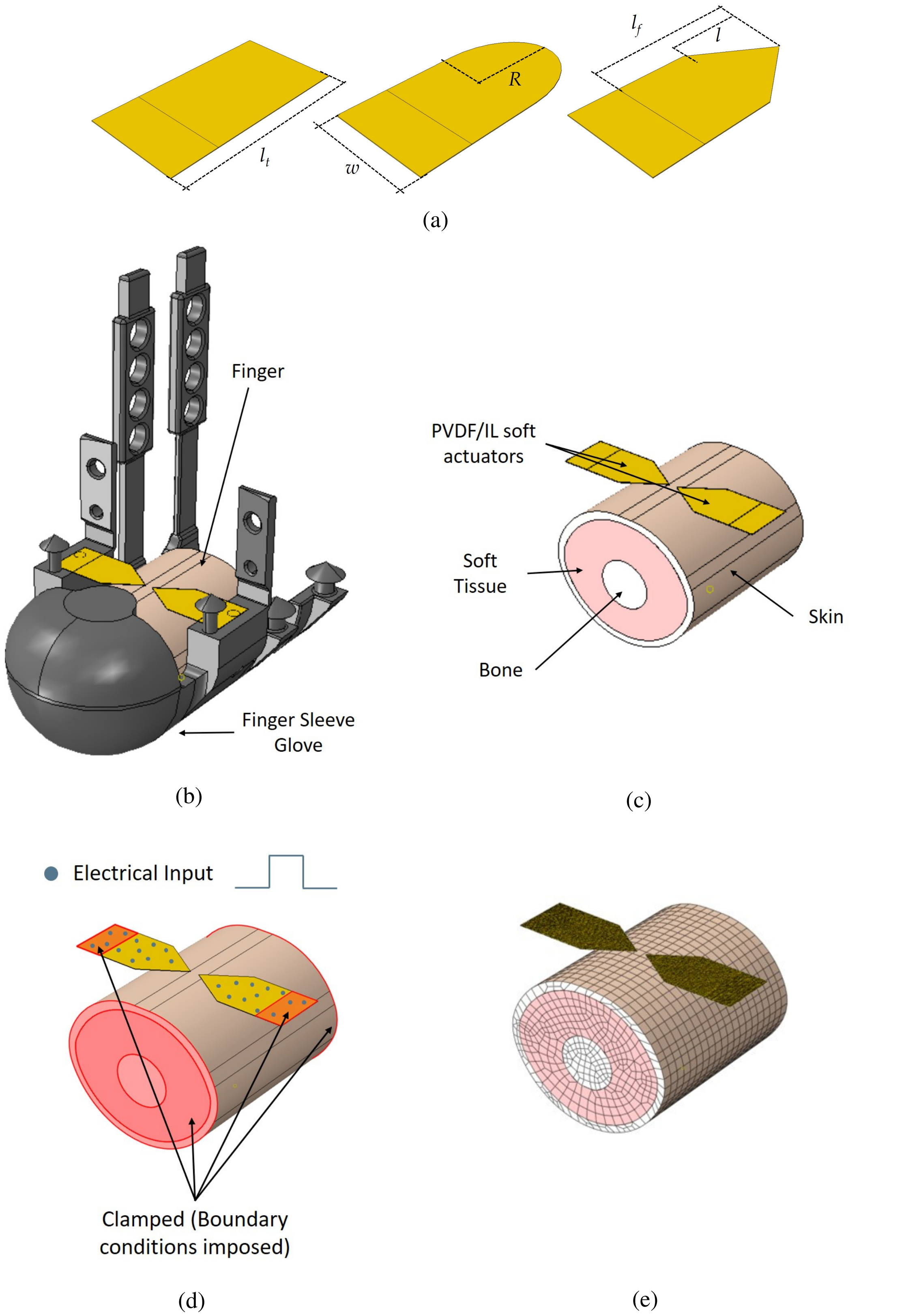


Figure 6.3: (a) Tip shapes tested: rectangular (left), circular (centre) and triangular (right). (b-e) Numerical FE models of the wearable haptic device in Abaqus: (b) schematic illustration of the finger sleeve prototype for the user study case; (c) FE model simulated in Abaqus, composed by the finger and the PVDF/IL soft actuators; (d) Electrical input and boundary conditions applied; (e) mesh applied to the numerical model.

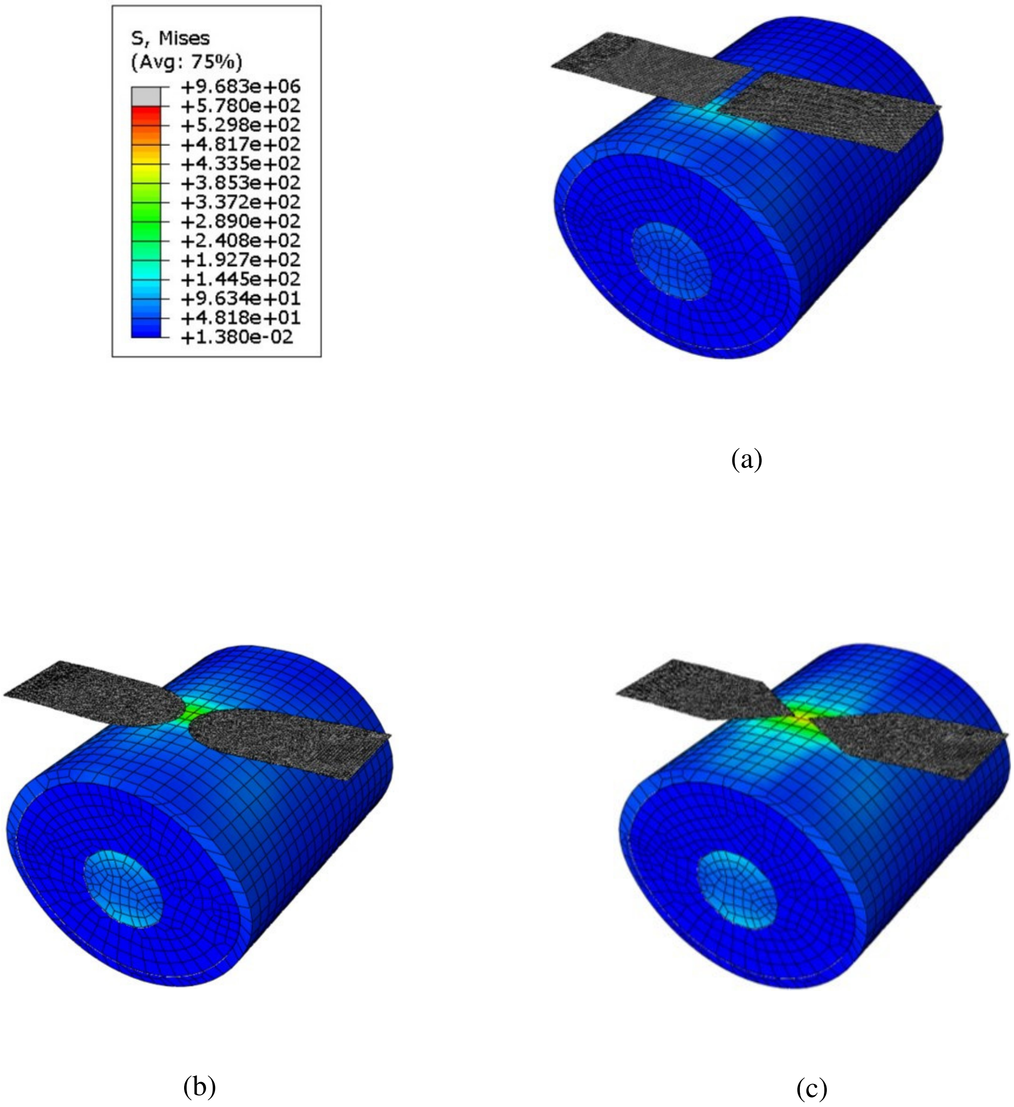


Figure 6.4: Numerical simulation considering the maximum voltage intensity (10 V) for all tip geometries tested. (a) Rectangular tip geometry; (b) Circular tip geometry; (c) Triangular tip geometry.

The results showed that the touch sensation provided by PVDF/IL soft actuators increased with the voltage intensity. For the minimum voltage intensity tested, 2.5 V, the simulation did not show any contact pressure for all the different situations. The relation between the contact pressure and the voltage intensity is justified by the proportionality of the electrical input applied and the consequent mechanical deformation induced. It means that when higher voltage intensities are applied to the samples, higher polarization is induced on them, which increases the outputs, such as the contact pressure [40] and displacement [17]. The same cause/effect is observed in other studies available in the literature. Raza et al [9] studied micro-structured porous electrolytes for highly responsive ionic soft actuators, using PVDF-co-hexafluoropropylene (PVDF-co-HFP) as the main active element. They observed a direct influence of the electrical input intensity on the mechanical deformation of the samples.

In addition, the geometry of the contact surface also had influence on the touch sensation provided by the finger sleeve prototype, in particular, by both PVDF/IL soft actuators. Independently of the input voltage applied, smaller contact areas showed higher pressure contact. From the different geometrical tips tested, the rectangular tip resulted in the smallest touch sensation. As the contact areas decreased, when the actuators were changed from rectangular to triangular (rectangular $\Rightarrow$ circular $\Rightarrow$ triangular), the contact pressure increased. This fact can be easily explained considering the mathematical equation for pressure, $p = F/A$, where p means the contact pressure, F is the force provided by the samples and A is the contact area. When similar values of force are applied to a perpendicular area or surface, the resultant pressure is inversely proportional to the contact area between the two surfaces.

The results of the tests simulated are shown in Figure 6.5 and Table 6.2.

The perception of touch can change considering different factors, such as age, gender, body region touched and health conditions [41, 42]. However, in literature, Meissners corpuscles, responsible for transmitting the sensations of fine, discriminative touch and

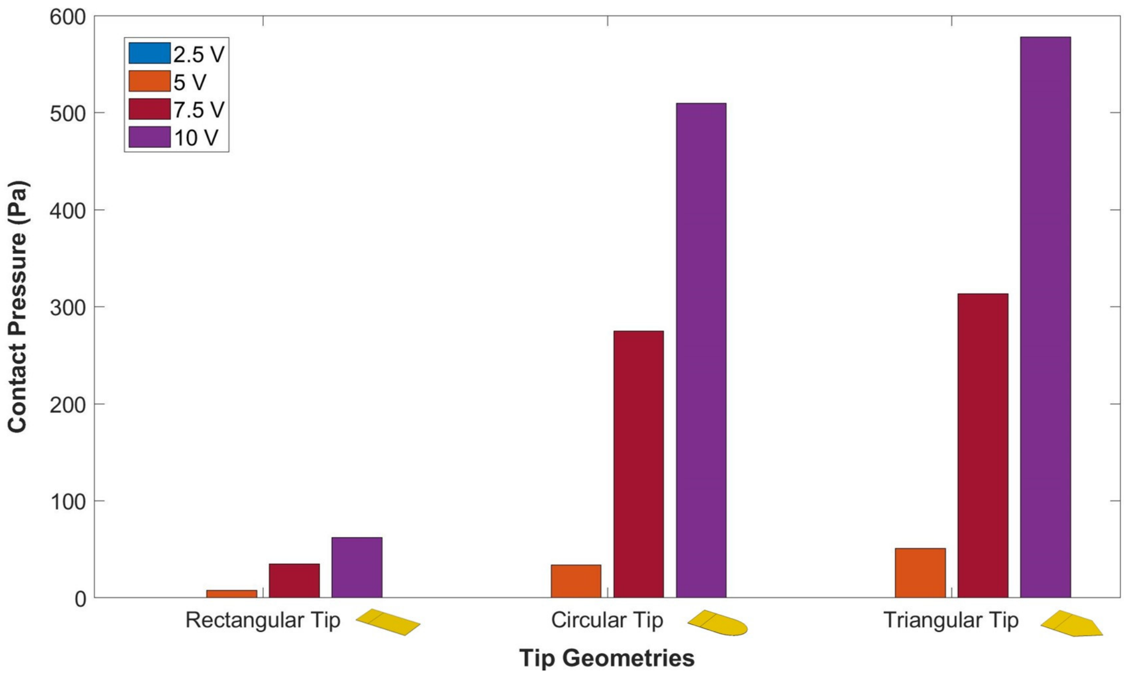


Figure 6.5: Numerical results considering the initial conditions tested in the *in-silico* user studies case, such as the tip geometry and the voltage intensity. For 2.5 V, the pressure was 0 Pa for all tip geometries.

Table 6.2: Contact pressure of each tip shape tested, considering different input voltage intensities.

	Contact Pressure (Pa)		
Input Voltage Intensity (V)	Rectangular Tip	Circular Tip	Triangular Tip
2.5	0	0	0
5.0	8.0	33.7	51.2
7.5	35.1	274.8	313.1
10.0	62.1	509.7	578

vibration, as well as allowing Braille reading in blind people, were reported as having a minimum sensitivity to skin indentation (displacement) around 10 μm [43, 44]. In the conditions of this study, the simultaneous actuation of two PVDF/IL soft actuators on the skin of an index finger resulted in different indentation results for different tip geometries, similar to what happened with the touch sensation (contact pressure) study. Considering the maximum voltage intensity, 10 V, the actuators with rectangular tip geometry indented the skin 0.80 μm, while circular and triangular tip geometries of the actuators indented 3.19 and 2.75 μm, respectively. Although quite similar, the actuators with circular tip geometry indented slightly more the finger skin than the actuators with triangular tip geometries, in contrast to the contact pressure. These values might be justified by the amount of piezoelectric material available to be actuated near the free tip, in this case, higher for the circular tip than the triangular tip.

Considering the triangular tip geometry as an example, the results for skin indentation change according to the pronunciation of the vertices. Figure 6.6 illustrates the results obtained for different triangular tips. There are minimum and maximum threshold geometries, in which the best results are achieved considering a particular tip shape.

When comparing the best result obtained considering this FEM study with the minimum value reported in the literature, it is around three times lower. This difference might be justified by the lower number of actuators used in the *in-silico* model of the device since it is expected to have higher skin indentation when more actuators are used over the same area. Finally, although a preliminary study about tip geometries was made, the optimised shape of the soft actuator might not have been achieved, which could also justify the results obtained.

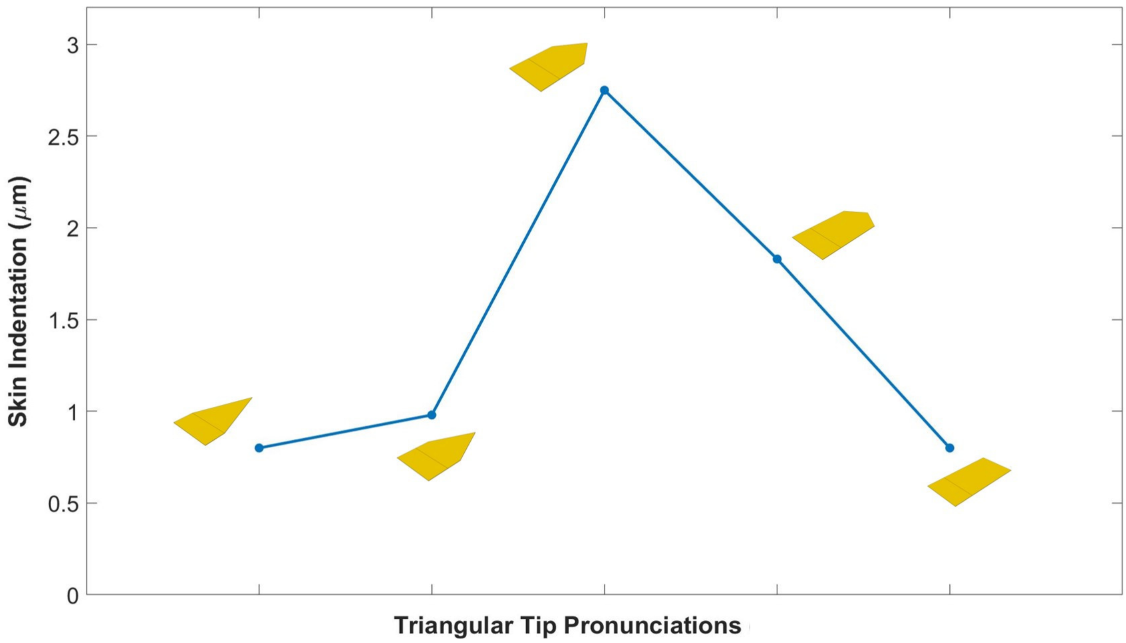


Figure 6.6: Numerical results considering different triangular tip pronunciations, from totally pronounced triangular tip geometry to rectangular tip geometry, respectively 0.80 μm, 0.98 μm, 2.75 μm, 1.83 μm and 0.80 μm of skin indentation.

6.6 Conclusions and Future Works

In conclusion, the present *in-silico* user study case allowed the authors to define some important guidelines for the development of a wearable haptic device to be used in re-educational scenarios.

The numerical study of different tip geometries highlighted the importance of the actuator's shape. As observed, the tendency for better results in terms of contact pressure and skin indentation is strongly related to the contact area between the actuator and the user. The feedback response for triangular tip geometries was the most promising, followed by circular and rectangular shapes. Nevertheless, when different triangular tip geometries were tested, the numerical model showed an optimal shape for achieving the best results, excluding the most extreme triangular shape. Moreover, the *in-silico* study also evidenced the importance of the input voltage intensity in the results. Higher input values lead to better feedback responses. However, it is important to be aware of the limitations of numerical studies such as the current one.

First of all, characterising piezoelectric materials in a proper way to be tested in *in-silico* conditions is extremely difficult, since some properties, such as the piezoelectric matrix, are not easily obtained in literature or experimentally. The piezoelectric matrix used in this study, despite limited, gives numerical displacement results similar to the experimental displacements observed on the samples' tips.

However, in the authors' opinion, the results reported are quite promising and point out in the right direction towards the future viability of haptic technologies in rehabilitative therapies, despite more developments being needed. In future works, it is important to test the wearable haptic prototype in real scenarios, through user study cases with volunteers. Only with the people's feedback is possible to have some reliable data on the effectiveness of the device, in terms of pleasantness and touch sensation.

With this study, the authors hope to have given a step forward in the analysis and characterisation of the applicability of piezo soft materials, such as PVDF-based materials, in therapeutic rehabilitative approaches.

Author Contribution: Conceptualization, A.D.A, P.M.; Formal analysis, A.D.A; Investigation, A.D.A; Methodology, A.D.A, P.M.; Software, A.D.A, M.P.; Supervision, P.M., M.P.; Writing - original draft, A.D.A.; Writing - review & editing, P.M., M.P. All authors have read and agreed to the published version of the manuscript.

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Conflict of Interest: The authors state that they have no financial, professional or other personal involvement in any product, service and/or company that would possibly affect their stance.

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Chapter 7

EMG Signals as a Way to Control Soft Actuators

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Keywords: Soft actuators, Piezo-based materials, EMG signals, Movement prediction, Exoskeleton

EMG Signals as a Way to Control Soft Actuators

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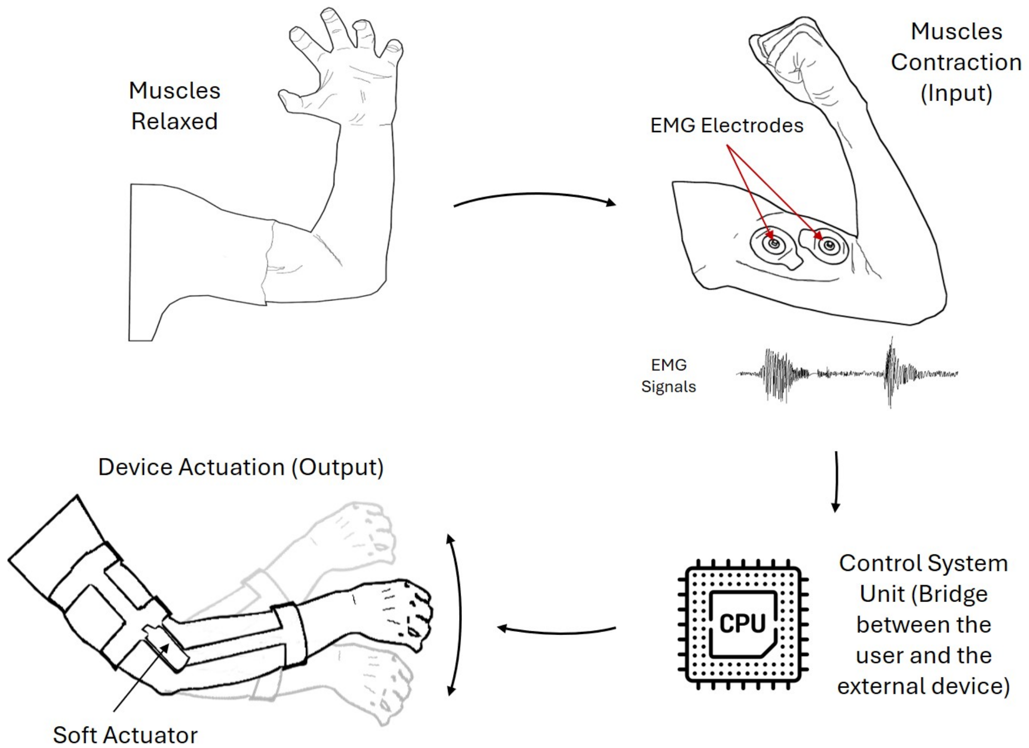
7.1 Abstract

Physical impairments have multiple causes, making them common. Locomotion disorders have been afflicting society for a long time, motivating researchers and engineers to mitigate their consequences. Nowadays, solutions such as exoskeletons and exosuits are in constant development and may become reliable options to help people in these circumstances. However, prospective solutions need a control system acting as a bridge between the external device (actuators) and the user. Among several possibilities, movement prediction is prioritized over movement reaction. This task may be done by capturing and processing biological signals from a user's body. Within this paradigm, muscle electromyographic (EMG) signals were acquired, processed and sent as input to piezoelectric soft actuators.



Graphical abstract.

7.2 Introduction

Physical impairments are very common nowadays since they could essentially be caused by three main reasons [1]: the normal ageing process, also associated with higher life expectancy [2, 3]; neurodegenerative diseases [4]; and finally, daily accidents [5].

Locomotion disorders caused by the normal ageing process are typically associated with older persons [6] and more accentuated in regions where life expectancy is higher. Some countries, such as Norway, Germany or Hong Kong (China) have a life expectancy over 80 years old, contrasting with some African nations (The Central African Republic or Nigeria have an expectancy of 54 years old). Statistics from United Nations (UN) reveal that elderly people older than 60 years old will be around 20% of the global population by 2050 [7].

Neurological pathologies are also responsible for a fragile physical state. They cause a progressive loss of structure and function of the central nervous system (CNS). The CNS is the main responsible to coordinate muscle control and command. Nowadays, it is estimated that about 50 million people worldwide could have some kind of dementia [8], with Parkinson's disease representing more than 10% of this value [9].

Injuries, particularly in the spinal cord (SC) and brain, could also cause permanent or temporary changes in muscular strength or body functions. They are the result of external traumas (such as car accidents (39.3%), falls (31.8%), gunshots (13.5%), sports-related (8%) [10]) or due to tumours in these specific locations [11]. Every year, about 40 million people suffer SC or brain injuries [12].

These locomotion issues have a significant societal impact, which motivated the effort of researchers and engineers [13] to mitigate some of their consequences and impact. Despite the solutions already in used, such as wanderers, wheeled vehicles or wheelchairs [14], the current focus is on the development of exoskeletons and exosuits equipped with different types of actuators [15].

A control system, acting as a bridge between the final user and the external device, is a fundamental requirement that does not depend on the adopted actuation system. In this work, biological signals (bioelectrical) acquired from muscle activities, will be used as inputs to control soft actuators.

Bioelectrical signals are the result of chemical activities at a cellular level, resulting in electrical potentials (typically in the mV range) [16] detected by appropriate sensors, such as EMG sensors [17, 18].

EMG sensors (intramuscular [19] or surface [20]) are able to measure small electrical signals [21] generated by muscle contraction. For this reason, they were successfully applied in the control of external devices [22, 23]. Solutions based on this method bring the advantage of reading and capturing a user's intention and control actuation, even if a person can't produce sufficient joint torques [24] to move.

However, the signal might be biased due to several factors, such as muscle crosstalk [25], a skin condition (in case of surface sensors), muscle fatigue [26] or the inability to access deep muscle fibres [27].

In the current work, a setup was developed to capture muscle bioelectrical signals, and use them as inputs to piezoelectric soft actuators - materials capable of changing their shape and size when electrically stimulated.

7.3 Control and Actuation System/Experimental Setup

An experimental setup was developed to capture bioelectrical signals and use them as inputs to induce mechanical deformation on soft materials. The solution proposed consists of the following elements (Figure 7.1): a BITalino (and EMG sensors) [28] connected to a personal computer (PC) and Matlab software [29] via Bluetooth connection; a function generator connected to a PC via USB; and finally an oscilloscope connected to the function generator for display and analysis of the output signal.

The EMG sensors connected to a BITalino board [28] acquire electrical activity at muscle level, while sending the data to software implemented in MATLAB. In parallel, an algorithm (Matlab software) processes the acquired information and if the signal fulfils pre-established criteria (such as a threshold value of signal intensity), a command is sent to the function generator to change the output signal. This signal constitutes the electrical stimulation input for the soft material, and is displayed on the oscilloscope. All data acquisition and processing happens in real-time (with maximum delays of approximately 1 second).

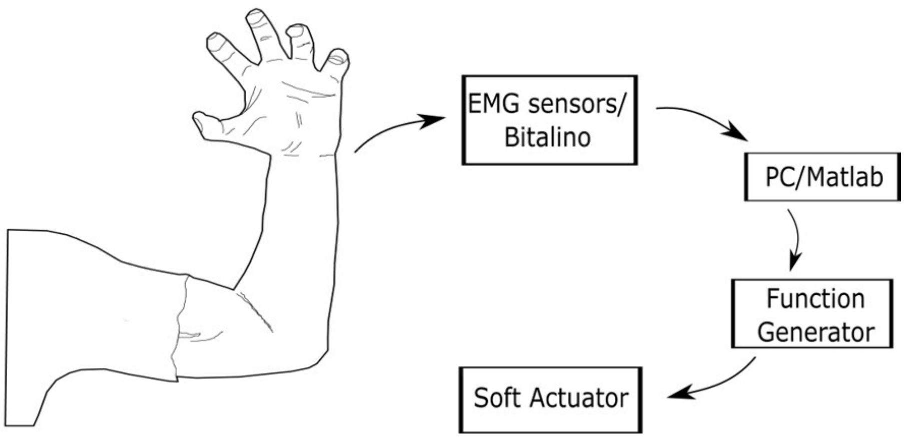


Figure 7.1: Outline of the experimental setup.

7.3.1 Hardware

The hardware setup consists of a PC (Intel Core i7-2700k and 32GB RAM) running Windows 10 Enterprise OS (OS build 19042.928), an oscilloscope (UNI-T UTD2062C) for displaying the output signal, the EMG sensors and a function generator (Joy-it JDS6600).

The EMG sensors were acquired from BITalino (Plugged Kit BT, Bluetooth version with firmware v5.1) [28]. BITalino (figure 7.2) is a microcontroller based on the AT-MEGA family, specifically developed for biometric applications with a maximum resolution of 10 bits [16].

BITalino is a modular system. The main board may be coupled with several modules. The energy module powers the device, has two energy outputs (3.3V and 1.65V) and a micro-USB connection to recharge a Li-ion battery. The central processing module hosts the microprocessor and the firmware. Finally the communication BLE module enables communication with other devices via Bluetooth. These components make the central unit of BITalino

Additionally, different sensors can be plugged into the main board, for electrocardiography (ECG) or electroencephalography (EEG) analysis [16]. In the current work, only EMG sensors were used. EMG sensor is an operational amplifier for biometric applications [28]. It has three electrodes - positive and negative placed at the muscle and a reference electrode, placed at a reference point such as an elbow.

The last component of the setup is function generator Joy-it JDS6600 [30]. The relevant characteristics of the equipment are the possibility to generate arbitrary waveforms (signals) as well as support for PC connection.

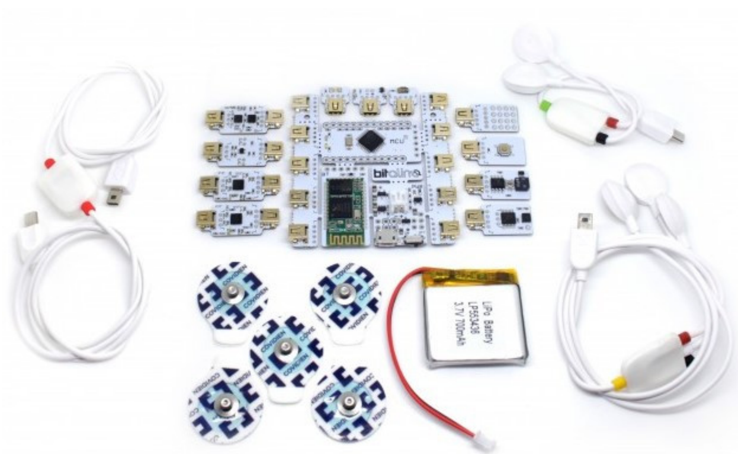


Figure 7.2: BITalino hardware.

7.3.2 Software

The control and actuation of the soft material requires software purposely built for the task. Matlab 2021a [29] and Python (v3.7) [31], are the main requirements. Additionally, the BITalino toolbox for Matlab [32] and the python module 'pyserial' [33] are needed for communication and data transfer between the BITalino and the PC.

All signal processing tasks are addressed using Matlab script EMG_signal.m, the main script which controls all hardware. While EMG_signal.m is running, two additional Python scripts are called hierarchically. The first, BITwave.py, defines the signal parameters. The second, jds6600.py, is the bridge between the PC and the function generator.

The biolectrical signal acquired with BITalino's EMG sensors, requires to be processed with a sequence of mathematical operations [28, 34, 35]. The sequence depicted in figure 7.3 gives the reader a clear understanding of the signal processing operations, applied to the raw signal. In the first step, the raw signal is converted into an EMG signal using equation 7.1 [28], rectified and displayed in the last plot of figure 7.3.

$$EMG(mV) = \frac{(\frac{ADC}{2^n} - \frac{1}{2}) \times VCC}{G_{EMG}} \times 1000 \tag{7.1}$$

wherein ADC is the value sampled from the channel, n the number of bits of the channel, VCC the operating voltage (3.3V) and, finally, G_{EMG} is the sensor gain (1009).

The last step involves an envelope peak function from Matlab libraries.

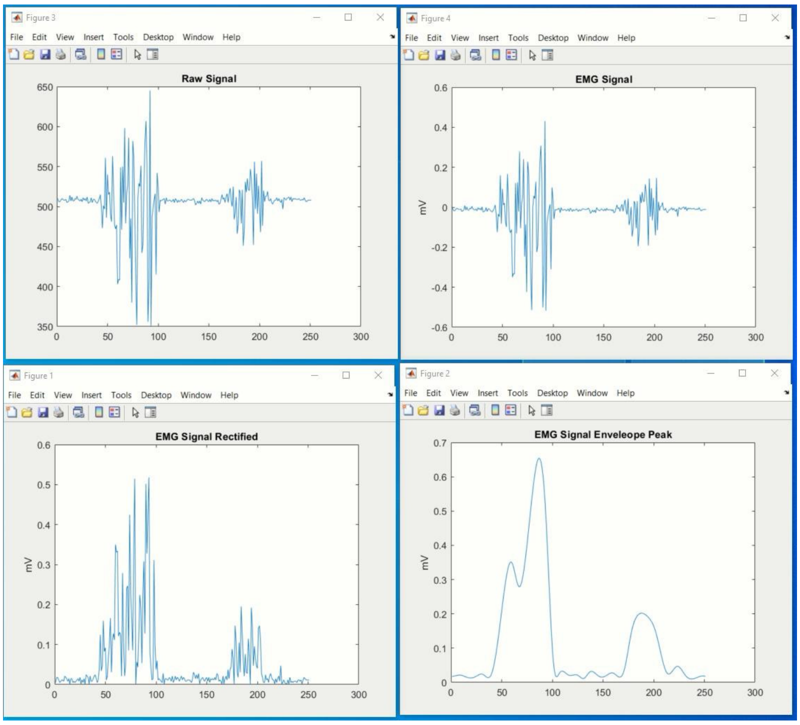


Figure 7.3: Signal acquired at different stages (example).

7.4 Preliminary Results and Discussion

Using the setup described previously, a test was carried out on a healthy male volunteer (29 years old). The results are observed in figure 7.4.

Observing the first image, when no muscle contraction was detected, the output signal (oscilloscope) is 0 V. As the muscle contraction starts, the EMG signal is acquired by EMG sensors of Bitalino and the algorithm detects the changes of the output signal from 0 V to an arbitrary positive value (second image). While the signal contraction from muscles remains constant, the output state is held (third image). Moreover, when the muscles relaxation begins and is detected by the sensors, the control system does the opposite behaviour. The output value observed returns to 0 V (fourth image).

All steps involved are processed (max delay $\approx$1 s) in real-time from data acquisition to signal output (see supplementary video A.10).

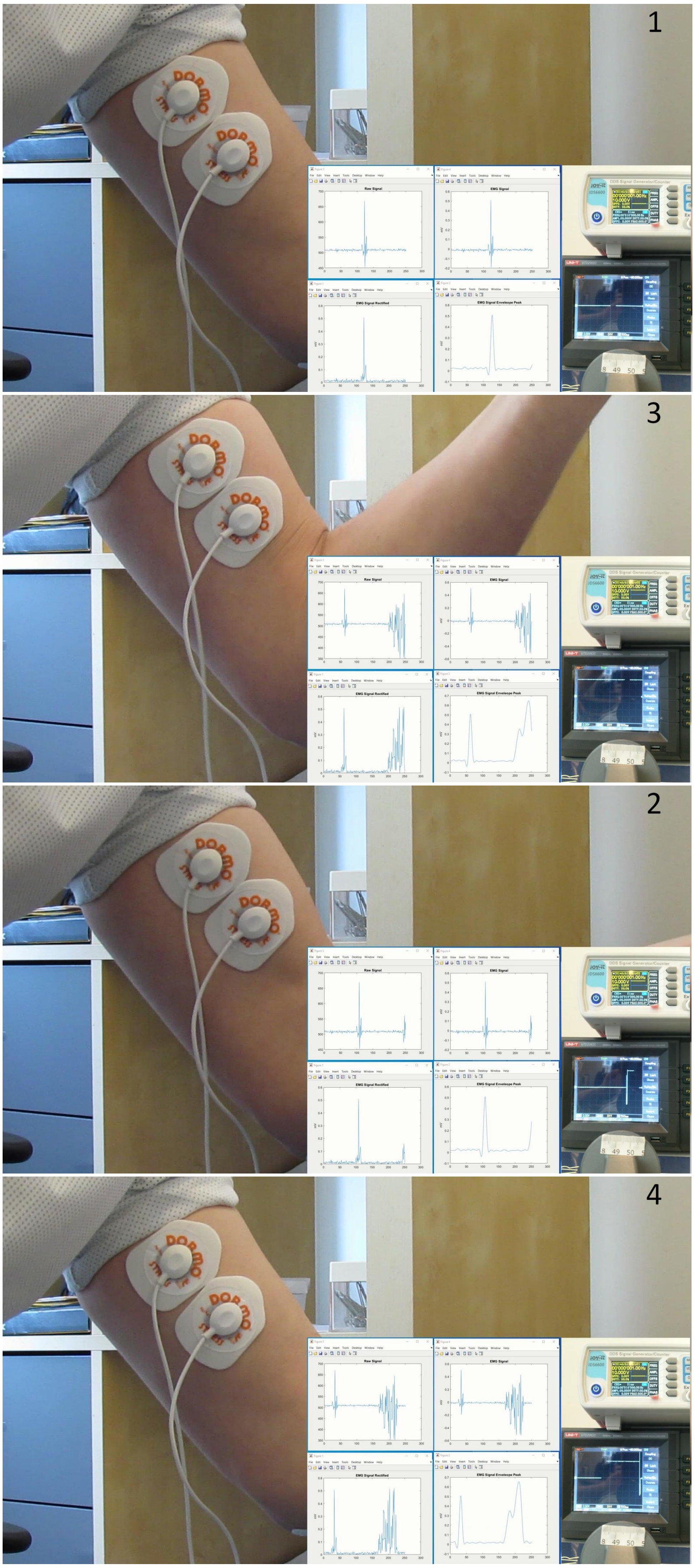


Figure 7.4: Oscilloscope output signal for four sequential instances (different stages) during muscle contraction. 1. 0 V without muscle contraction; 2. positive voltage value associated with contraction detection; 3. Stable voltage while contraction is maintained; 4. muscle relaxation induces progressive voltage drop to 0 V.

7.5 Conclusions and Future Perspectives

The procedure adopted in this work enables the use of an EMG signal as input for different use cases. The actuation of a piezoelectric soft actuator almost in real-time is a possible use of this method. Moreover, the most relevant characteristic of the proposed setup is the ability to provide adequate input to the actuator, even with slight muscle contraction. This behavior can be fully customised, since the algorithm uses sensibility parameters that can be changed according with usage requirements.

Despite its usability, the system has limitations. Since the bioelectrical signal comes from a surface EMG sensor, it could be biased due to skin conditions or muscle crosstalk signals. This fact could impair the applicability of the developed setup in real-world scenarios such as movement assistance or rehabilitation.

The research team aims to test the setup using a working soft actuator, and to address or minimize some of the technical issues already encountered. The improvement of the EMG signal via noise reduction, using filters or Fourier transforms will be one of the next iterations of the project. The creation of a portable solution will be another milestone for the project, since the team aims to combine this technology with augmented reality (AR) and virtual reality (VR) for assisted rehabilitation purposes.

Author Contribution: Conceptualization, A.D.A.; methodology, A.D.A and P.M.; software, A.D.A and A.M.T.; research, A.D.A; writing - original draft preparation, A.D.A.; writing - review and editing, A.D.A, A.M.T and P.M.; supervision, P.M. All authors have read and agreed to the published version of the manuscript.

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Conflict of Interest: The authors have nothing to declare.

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Chapter 8

Discussion

Mobility disorders tendentiously occur under specific conditions, such as the ageing process [1], accidents [2] or as a result of diseases that compromises and damage the CNS [3]. Statistics show that 16% of world population are suffering from this problem [4], whereas possibly many more are being affected indirectly, such as the case of family and closest friends. The world demography for mobility impairments is a real problem that has been fought by society, in different levels, to mitigate the consequences of lack of mobility and the suffering of patients [5].

Some political initiatives have been approved in many countries regarding the legislation of accessibility features that must be part of buildings, public spaces and public transportation. Portugal, for example, through the decree-law no. 163/2006, have implemented rules and technical requirements for facilities, such as ramps and lifts [6]. Furthermore, the scientific community has also fought against this social problem by developing exo-devices that can assist people during daily tasks and rehabilitate impaired muscles.

Despite all technical possibilities described on chapter 3 for structural materials and actuation system along with the advantages and disadvantages, the present dissertation focused on the development of an exosuit for rehabilitation purposes, with soft materials. The *finger sleeve* prototype, firstly shown in chapter 5, was developed using TPU as structural material and PVDF/IL soft actuators as main active element, following some requirements previously established, such as biomimetism, lightness and an aesthetic appearance.

Due to its soft proprieties, the device is intended to be applied for haptic feedback stimuli in re-educational movement therapies. However, before claiming its real effectiveness, the main active element (PVDF/IL soft actuators) was analysed under different perspectives. The mechanical, chemical and electric properties were studied on chapters 4 and 5, as well as the electromechanical behaviour (Chapter 5). The findings were compared to the literature to ensure the solution applicability for the goal since a unique experimental protocol was used, considering a non-toxic solvent (DMSO) and the IL [Pmim][TFSI]. The results achieved were in accordance with the literature, which validated the material developed and the tests performed.

With the PVDF/IL material fully characterized in terms of its properties and electromechanical behaviour under external stimuli, an *in silico* user study case was performed, considering the prototype developed (Chapter 6). The simulation was conducted applying FEM analysis, which allowed to have a better perception of haptic feedback interaction between the user and the device. The results achieved are in the same order of magnitude from the minimum threshold value reported on literature for skin indentation to effectively interact with the user, through haptic stimuli and touch sensation. Moreover, the study also provided strong indications of how to enhance the interaction, such as the addition of more soft actuators and different tip shapes.

A preliminary and basic control system of the soft actuation was still developed, considering as input the bioelectrical signals produced by the muscles, when they contract (Chapter 7). Considering external hardware to acquire the body signal, which will be send to the actuators for interaction with the patient, a setup was prepared. At the end, the preliminary results pointed to the real possibility of using such approach to control the actuation system.

With a light and portable device and a self-centered control actuation system, the user can play their rehabilitation exercises without place restriction or other bulky complementary devices attached.

Detailed discussions about the particular results presented in this PhD dissertation are present in each chapter, in particular on chapters 4, 5, 6 and 7.

In the literature, different authors have reported their studies about wearable haptic devices (exoskeletons or exosuits) to treat muscles impairments. Chen et al [7], for example, developed a wearable hand rehabilitation system using soft gloves for mirror and task-oriented therapies. The device was designed considering a soft flexible material. However, the actuation system was based on traditional micromotors.

Ben-Tzvi et al [8] created a portable and light haptic exoskeleton device to measure the user's hand motion and assist its movement. The actuation system was performed considering miniature DC motors through routed cables at each finger. Their experimental results demonstrated that the device was capable of assisting hand grasping motion, which show the potential of the solution. Similarly, Fujimoto et al [9] presented a force haptic feedback device for robot hand teleoperation, considering partially soft and rigid materials, such as ABS and SMAs. The authors concluded that the device developed was able to produce force, improving the operability of the five-fingered robot hand teleoperation.

Although different wearable haptic devices for rehabilitation therapies have been developed considering soft materials, only a few engineered a device totally focused on that type of materials, in terms of structure and actuation system. Skorina et al [10], for example, presented a soft robotic wearable wrist device for kinesthetic haptic device, which can apply forces directly on the human joints while maintaining flexibility. The actuation was assured by pressure driven soft actuators, which were placed on four sides of the wrist. Their results pointed to improved performance of the users following complicated paths. Chossat et al [11] presented a soft wearable device for haptic feedback using twisted and coiled polymer actuators. The results showed that the participant reaction time was comparable to those of traditional motored vibrotactile feedback system.

In comparison with literature, the wearable haptic device developed and presented in this PhD dissertation prioritizes exclusively soft and smart materials. As result, the *finger sleeve* prototype is light, adjustable to person's physiognomy, flexible and biomimetic since it is structurally made from TPU. Additionally, as it is a 3D printed prototype through FDM technology, new iterations of the device are easily and quickly achievable. Moreover, as its actuation system is based on the converse piezoelectric principle using relatively low input voltage amplitude, a small battery could be used to trigger the actuation system by input of EMG sensors strategically placed on patient. This PhD dissertation, about the development of an **Exoskeleton for Aided Mobility**, represents a complete approach since it considers the structural materials and actuation system in the same solution, while still approaches a possible control system based on EMG sensors.

Therefore, despite the intrinsic limitations of soft materials (e.g. low force actuation and low yield stress), their advantages point to the development of promising solutions to face and fight mobility impairments.

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Chapter 9

Conclusions and Future Works

Exo-devices for aided mobility are a hot topic for companies and research institutes. However, the engineering project to develop such devices is very demanding since many and crucial requirements need to be addressed, including safety, actuation and control systems. Different alternatives are viable for structural materials and actuation systems, as discussed in chapter 3, which indicated two different concepts: the exoskeletons made from rigid materials and using traditional actuation systems such as pneumatic; and the exosuits considering soft materials, such as the application of smart polymeric materials in the actuation system using, for example, piezoelectric solutions.

The present PhD dissertation focused essentially in the development of an exosuit for aided-mobility with haptic feedback purposes. PVDF/IL soft actuators are the main active element, which are responsible for the haptic interaction with the patient. As reported in this work, they evidence great biocompatibility properties along with excellent mechanical, chemical and electrical properties and electromechanical behaviour. All these features made PVDF/IL the ideal candidate to incorporate such solution, as studied and reported on chapters 4 and 5.

The *finger sleeve* prototype was engineered considering PVDF/IL actuators, as described on chapter 5. Moreover, an *in silico* user study case was performed to ensure the effectiveness of the device. Although, currently, it does not provide a considerably amount of touch sensation, it pointed to promising results. To control the actuation of the *finger sleeve* device, it was developed a simple control system able to generate and coordinate the actuation of PVDF/IL actuators based on EMG bio-signals acquired from the muscles, as referred in chapter 7.

A more detailed description about the conclusions achieved are presented in each chapter, specifically 4, 5, 6 and 7.

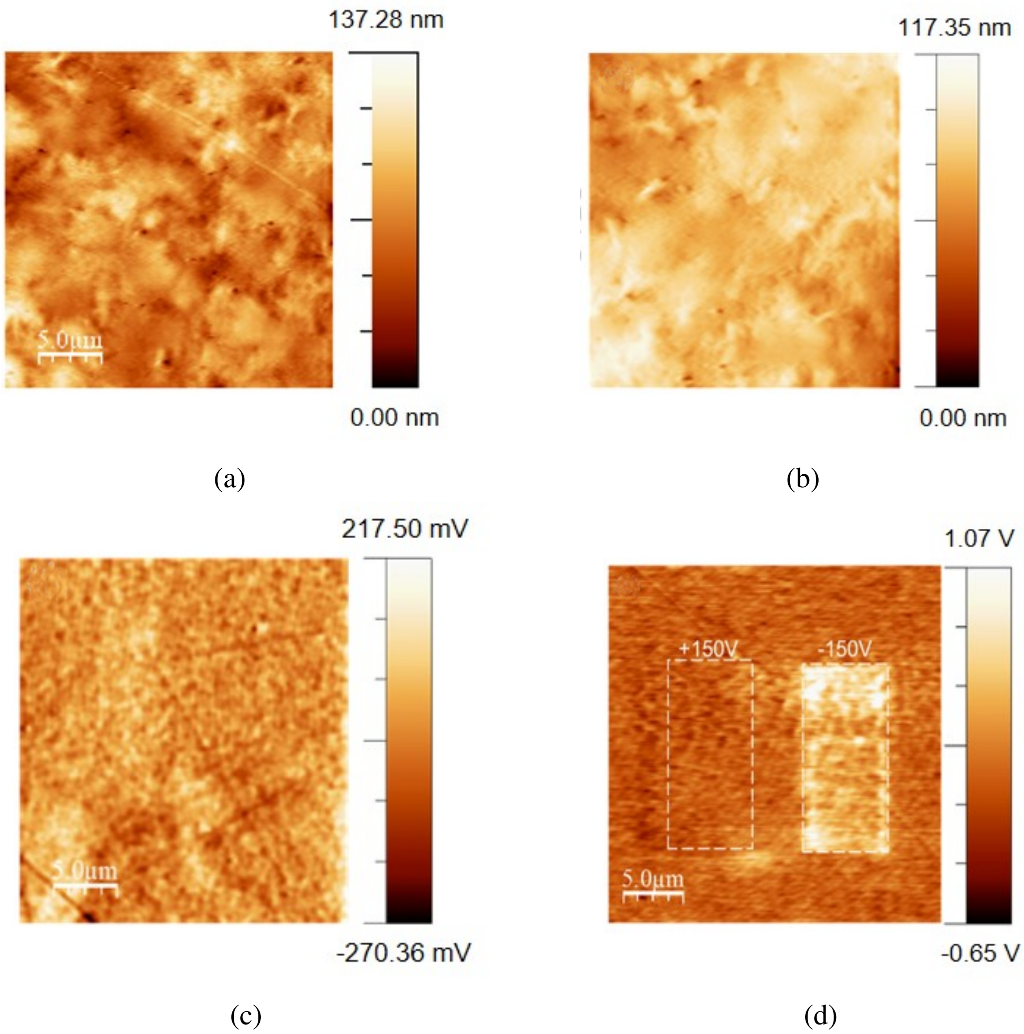
After the present study, there is room to proceed with forward studies regarding the development and iteration of the *finger sleeve* prototype. The inclusion of more soft actuators and sensors can reveal crucial for better haptic feedback stimulation and for monitoring the performance of the device. As PVDF/IL material evidences piezoelectric properties, which means besides behaving as an actuator, it can play as sensor as well. This possible dual-behaviour might allow to use the same material (PVDF/IL) for both applications in the same technical solution. Although the study of PVDF/IL as sensor has not been considered in the present dissertation, the material was already largely studied here considering its properties. Moreover, a user study case with real volunteers, instead of *in silico* studies, is fundamental to find out the real touch effectiveness and pleasantness provided by the wearable device.

Finally, the work presented in the current PhD dissertation should be faced as an early study about the use of wearable haptic devices for rehabilitation therapies, considering soft materials in their structure and actuation system. In conclusion, the exoskeleton developed points to a promising future of using such haptic feedback technologies in rehabilitation.

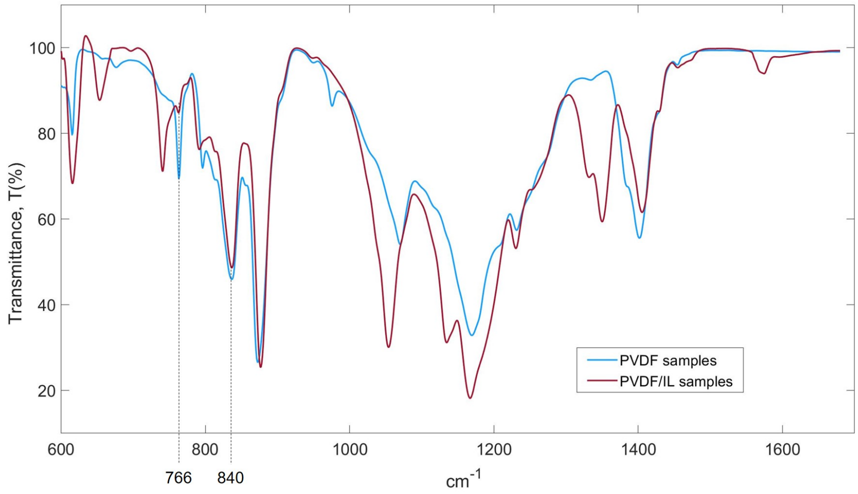
Appendix A

Supplementary Info

Supplementary Video A.1: Tensile test (sped up 15 times) (video link).



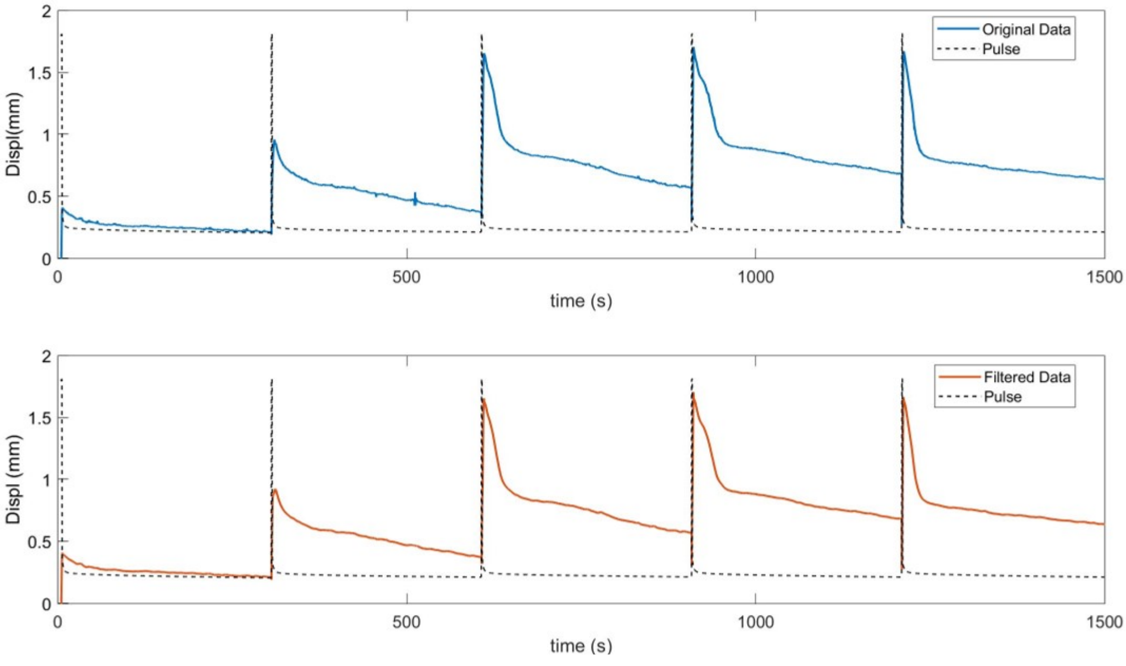
Supplementary Figure A.2: Electrical characterization of pure PVDF samples: (a) AFM topography image of the PVDF film; (b) PFM Topography; (c) Out-of-plane amplitude (A*cosθ); (e) Out-of-plane phase images; (d) PFM amplitude image after applying $\pm$150 V to create artificial domains.



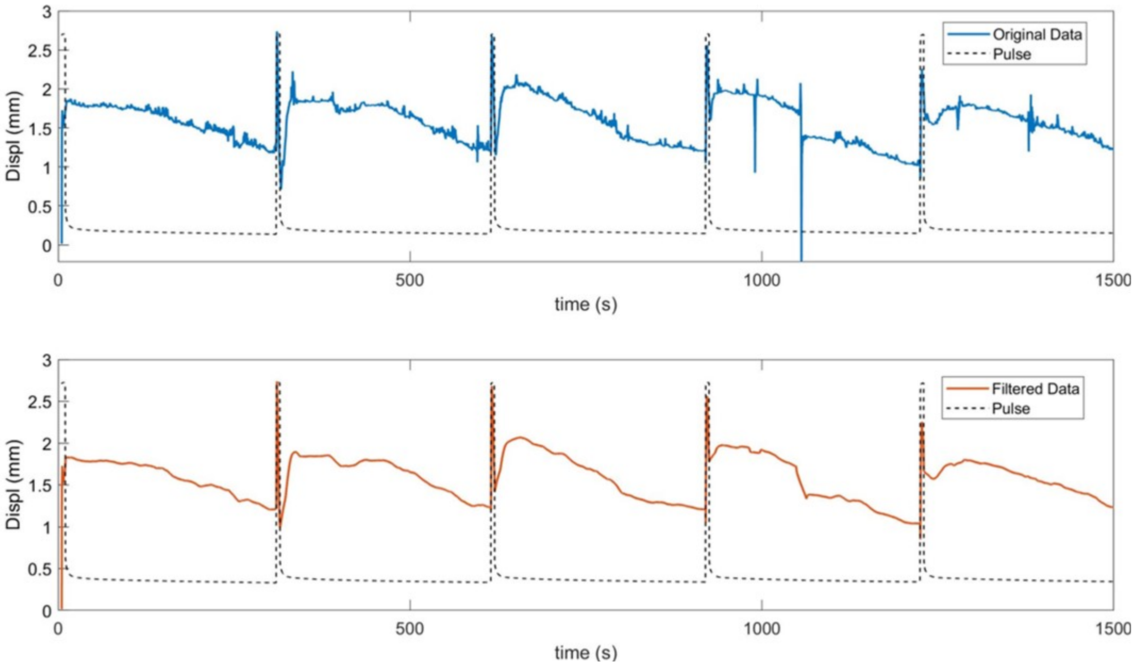
Supplementary Figure A.3: FTIR-ATR analysis. Comparison of PVDF samples with and without IL (red and blue lines, respectively). The EA phase content can be obtained using the Beer-Lambert law and the transmittance at peaks at 766 cm^{-1} and 840 cm^{-1} (marked at dashed lines), which correspond to the non-EA phase (e.g. α) and to the EA phase (e.g. β), respectively.

Supplementary Video A.4: Sine wave displacement analysis, minimum actuation displacement (video link).

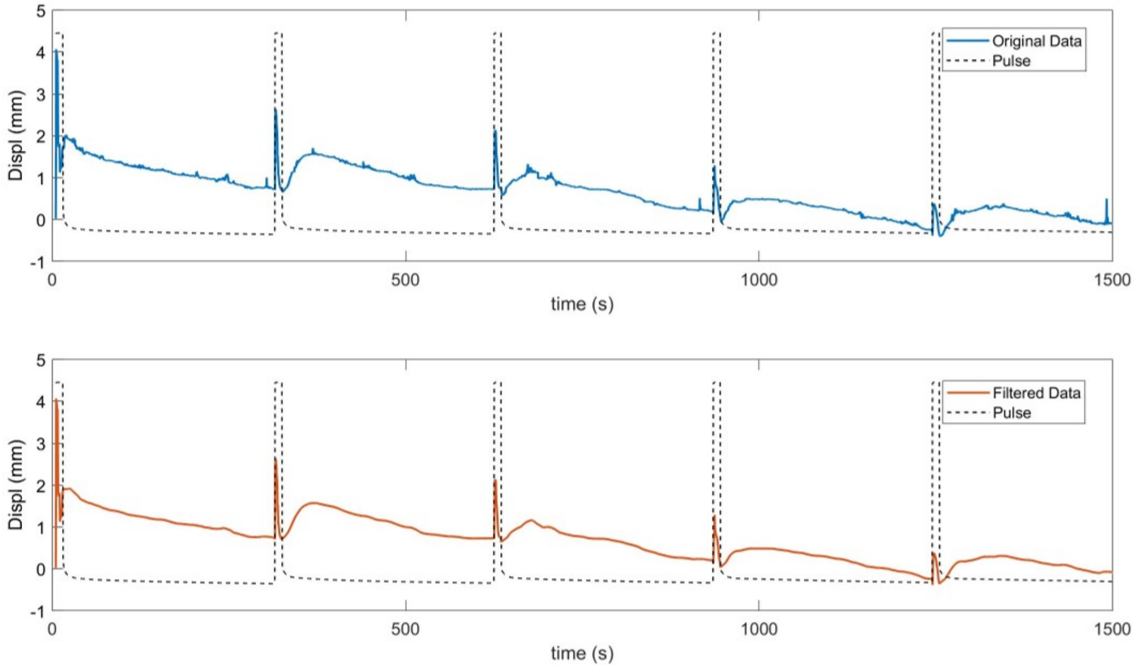
Supplementary Video A.5: Sine wave displacement analysis, maximum actuation displacement (video link).



(a) 1 second pulse time.

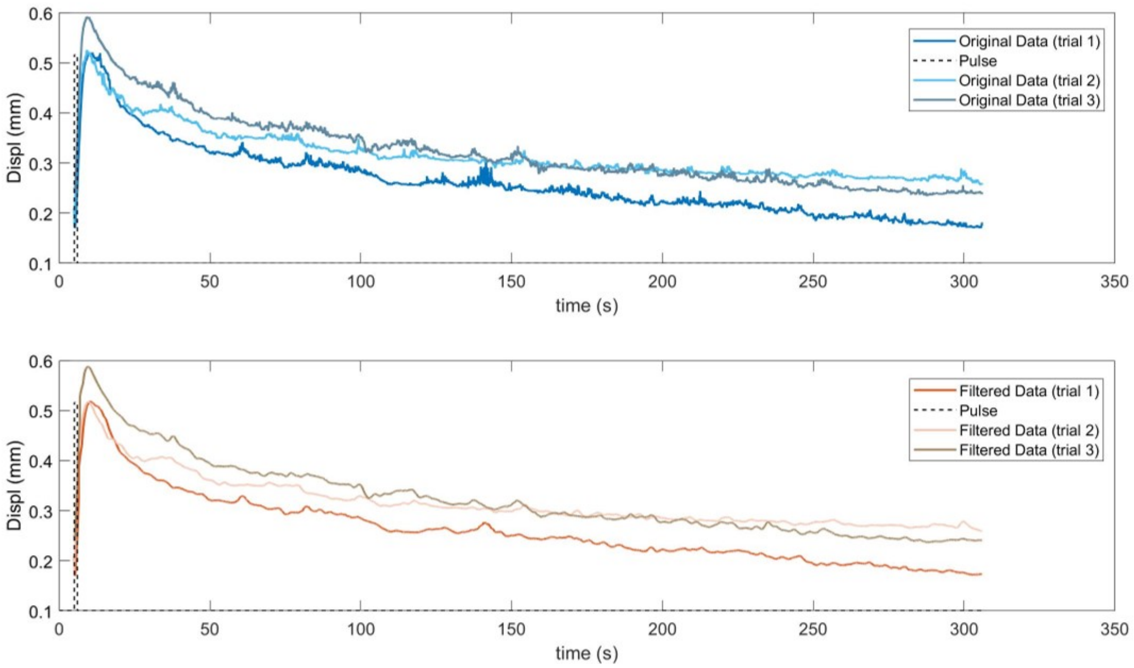


(b) 5 seconds pulse time

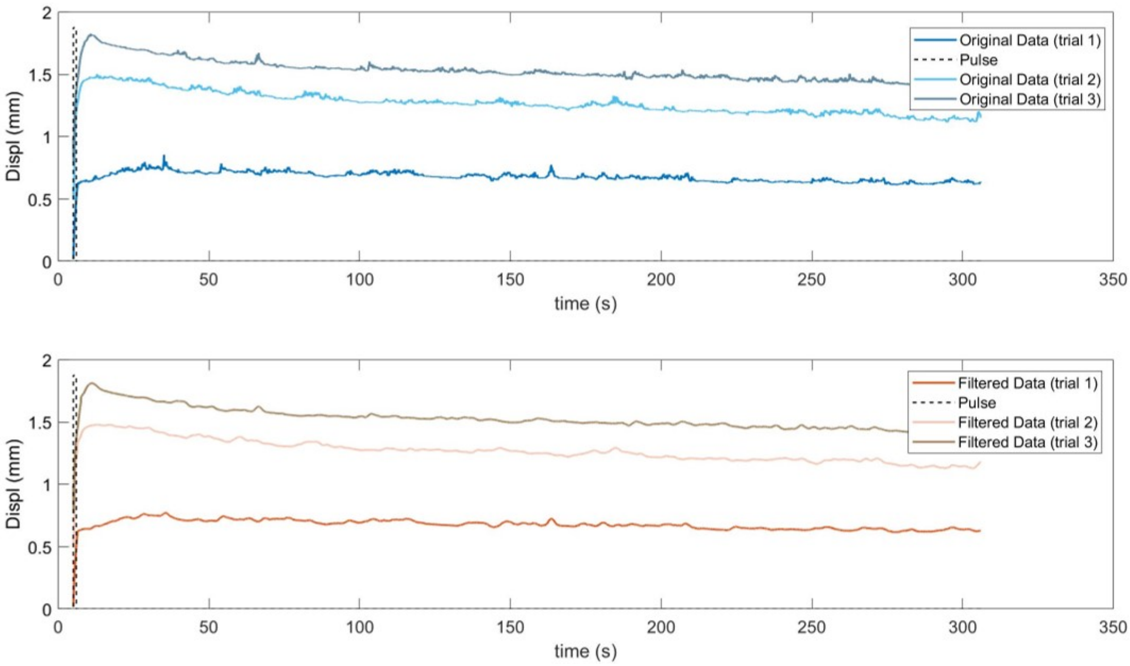


(c) 10 seconds pulse time

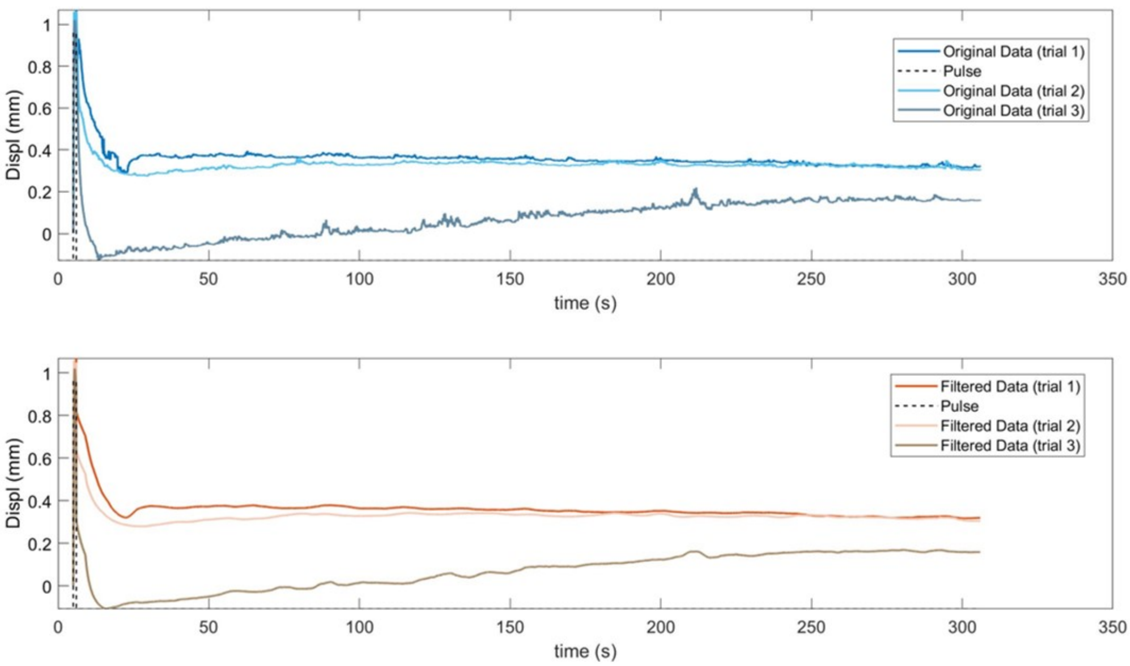
Supplementary Figure A.6: Comparison of raw data and filtered displacements for pulse time.



(a) 5 V pulse amplitude.

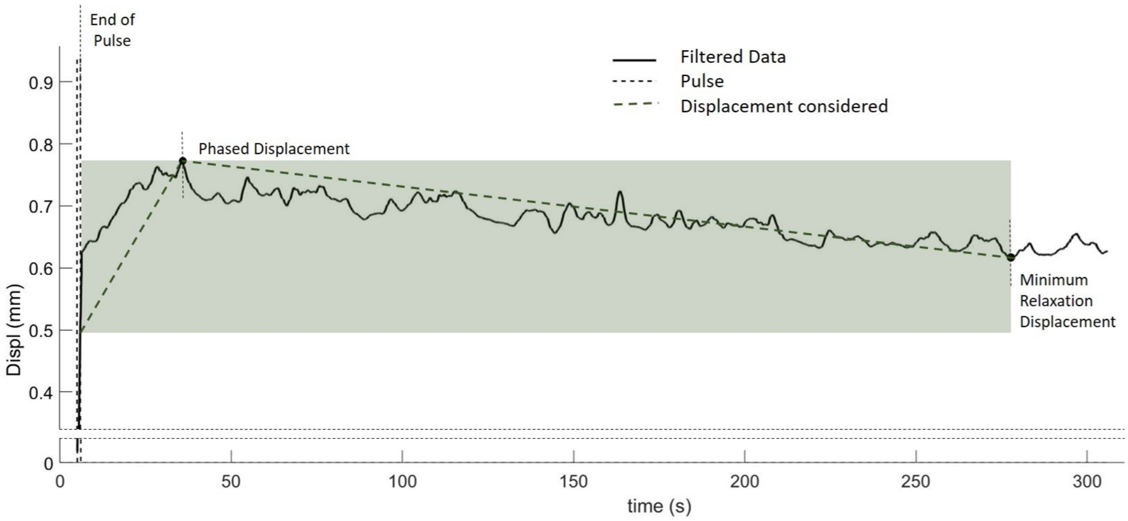


(b) 7.5 V pulse amplitude.



(c) 10 V pulse amplitude.

Supplementary Figure A.7: Original and filtered displacement data for pulse amplitude.



Supplementary Figure A.8: Relaxation speed for 7.5 V.

Supplementary Video A.9: *Finger Sleeve* Prototype - proof of concept. PVDF/IL actuators for cutaneous haptic feedback (video link).

Supplementary Video A.10: Real time data acquisition and signal output (video link).