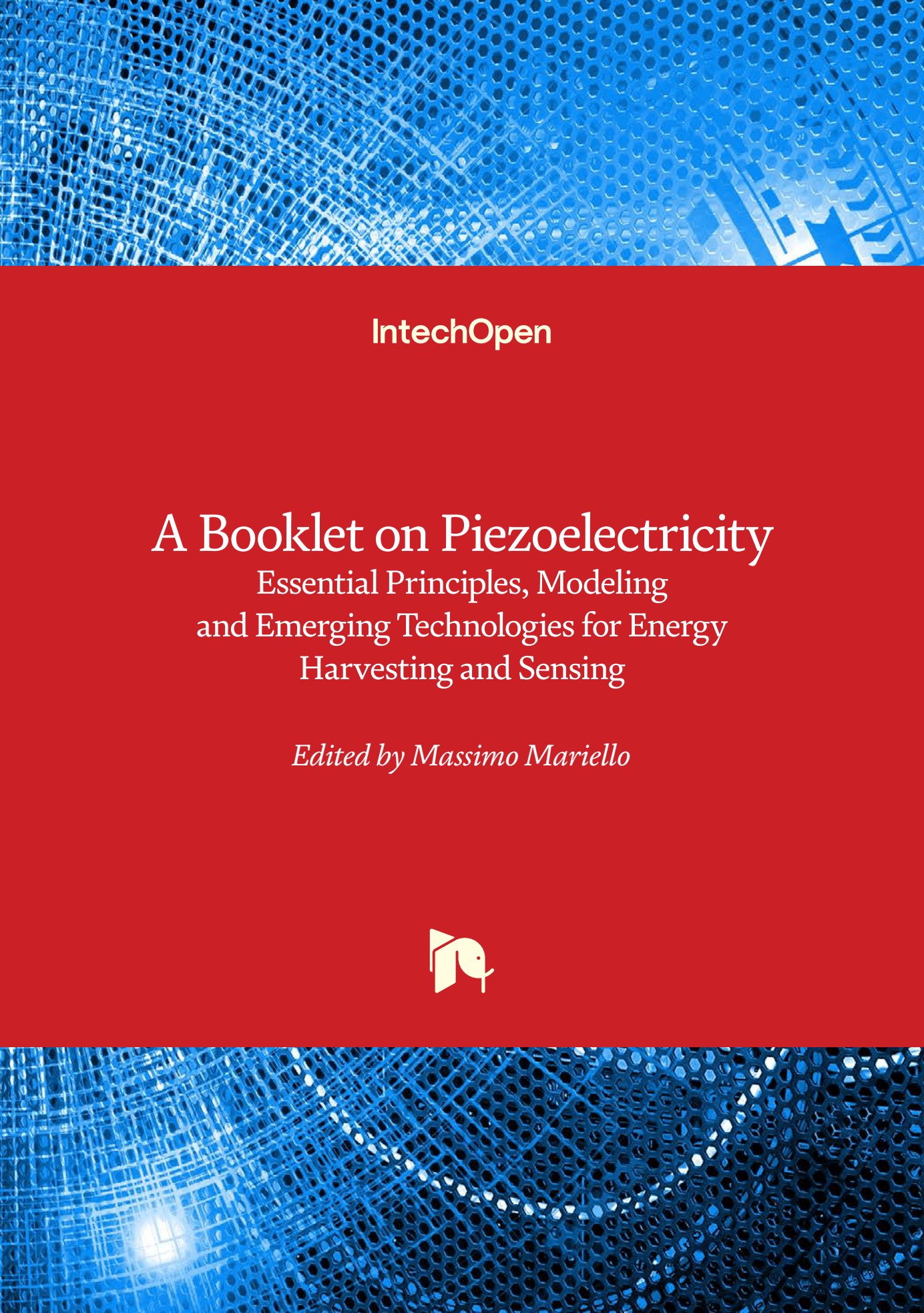




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# A Booklet on Piezoelectricity

Essential Principles, Modeling  
and Emerging Technologies for Energy  
Harvesting and Sensing

*Edited by Massimo Mariello*





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A Booklet on  
Piezoelectricity - Essential  
Principles, Modeling and  
Emerging Technologies  
for Energy Harvesting and  
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A Booklet on Piezoelectricity - Essential Principles, Modeling and Emerging Technologies for Energy Harvesting and Sensing

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Edited by Massimo Mariello

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# Meet the editor



Dr. Massimo Mariello is a senior researcher at the Institute of Biomedical Engineering in Oxford, specializing in soft bioelectronics, implantable medical devices, and electroactive materials and mechanisms. He holds a Ph.D. in Materials Engineering and Nanotechnologies from the University of Salento and the Italian Institute of Technology. He has conducted postdoctoral research at the École Polytechnique Fédérale de Lausanne (EPFL, Switzerland) and the University of Oxford (UK). His work focuses on microfabrication, electroactive materials, and flexible systems for neuromodulation, energy harvesting, biosensing, and drug delivery. Author of numerous publications in high-impact journals, he combines materials science, electronics, and biomedical engineering to advance next-generation therapeutic technologies. This book reflects his passion for making complex physical phenomena accessible through concise, interdisciplinary insight.



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# Preface

*A Booklet on Piezoelectricity – Essential Principles, Modeling and Emerging Technologies for Energy Harvesting and Sensing* was conceived as a concise introduction to one of the most elegant phenomena in physics and materials science, the conversion of mechanical stress into electrical potential, and vice versa. Known as piezoelectricity, from the Greek *piezein* meaning “to press”, this effect beautifully unites two seemingly distinct domains: mechanics and electricity.

Discovered in 1880 by Jacques and Pierre Curie, the direct piezoelectric effect revealed that certain crystals, such as quartz and Rochelle salt, generate an electric charge when mechanically deformed. Just one year later, the reverse piezoelectric effect was discovered, i.e., the ability of these same crystals to deform in response to an applied electric field. These two mirror phenomena form the cornerstone of countless technologies, from ultrasound transducers and sensors to precision actuators and microelectronic components.

This book aims to offer readers a compact, five-chapter journey through the essential aspects of piezoelectricity, combining historical perspective, material innovation, and modern applications. Each chapter touches a specific topic and represents a glimpse of what that topic can offer, based on current and previous literature. The book can be read in a few hours, yet it invites the reader to continue exploring a field that remains as dynamic today as it was revolutionary more than a century ago.

The introductory chapter, “Piezoelectricity: From Discovery to Devices”, provides an introduction to the science, history, and technological relevance of piezoelectric materials. It briefly guides the reader from the early discoveries of electromechanical coupling to the fundamental principles that govern piezoelectric behavior, explaining how mechanical stress and electrical polarization interact at the atomic level. The chapter further outlines the evolution of piezoelectric materials, from natural crystals to advanced ceramics, polymers, and composites, and their integration into modern devices. It establishes the conceptual framework for the volume, illustrating how piezoelectricity continues to bridge fundamental physics and practical innovation across sensing, actuation, and energy conversion technologies.

The second chapter, “Materials Innovation: Advances in Piezoelectric Materials”, provides an accessible yet thorough overview of how crystal symmetry and dipole alignment give rise to piezoelectric behavior. It traces the evolution from classic ceramic systems, such as PZT (lead zirconate titanate), to contemporary, lead-free, polymer-based materials, emphasizing flexibility, biocompatibility, and sustainability. This chapter lays the scientific foundation for understanding how piezoelectricity can now be engineered into soft, adaptive, and bio-integrated devices.

The third chapter, “Quartz Crystal Microbalance Modeling for Biocompatible Thin Films Studies”, focuses on the theoretical and computational modeling of

quartz crystal microbalance (QCM) systems to investigate the nanoscale behavior of thin films and coatings. By simulating the coupling between mechanical resonance, mass loading, and viscoelastic effects, this chapter demonstrates how QCM modeling provides deep insights into the structural and interfacial properties of biocompatible materials, which are crucial for the design of durable and reliable biomedical implants.

The fourth chapter, “Maximising Piezoelectric Energy Harvesting: Overcoming Challenges and Unlocking Sustainable Power Generation”, explores the expanding field of piezoelectric energy harvesting and its role in achieving self-powered technologies. It explains how environmental vibrations and mechanical motion can be converted into electrical energy through advanced material architectures, ranging from nanostructured composites to flexible thin films. These systems are designed to power sensors, wearable technologies, and implantable biomedical devices without the need for batteries. The chapter highlights energy autonomy as a central goal, emphasizing how efficient piezoelectric harvesters can contribute to a more connected, sustainable, and energy-resilient technological landscape.

The fifth and final chapter, “Efficient Interfacing Schemes to Enhance the Sensitivity and SNR Performance of Piezosensor Systems”, examines the principles and advances that underpin modern piezoelectric sensors. It discusses how optimized electronic interfaces, miniaturization, and advanced fabrication techniques enhance signal sensitivity and noise performance, enabling the precise detection of mechanical, acoustic, and physiological signals. These developments have transformed piezoelectric sensors from rigid, single-function elements into flexible, high-performance systems integrated across applications ranging from medical implants to soft robotics. The chapter highlights how innovative circuit design and material engineering are extending sensing capabilities into microscale and biological environments, driving the next generation of intelligent, adaptive sensing technologies.

Together, these chapters capture the interdisciplinary essence of piezoelectric research, where physics, chemistry, materials science, and engineering converge. Piezoelectricity’s relevance extends far beyond the laboratory: it underpins innovations in biomedical engineering, robotics, renewable energy, and communication systems. The book closes with the hope that readers, having engaged with this concise overview, will be inspired to delve deeper into the field’s rich literature and growing opportunities.

Looking ahead, the future of piezoelectricity lies in soft, biointegrated transducers, hybrid energy systems, and neuromorphic or adaptive devices capable of sensing, learning, and responding to their environments. As researchers move toward sustainability and miniaturization, piezoelectric materials will play an essential role in shaping the next generation of smart, autonomous technologies.

I want to express my gratitude to the authors who contributed to this volume and to the colleagues, reviewers, and editor assistants who offered their time and insight during its preparation. Special thanks are due to my current and previous institutions, namely the University of Salento, the Italian Institute of Technology, the École Polytechnique Fédérale de Lausanne, and the University of Oxford, for fostering the interdisciplinary environment that enables such integrative research and for shaping my cultural and scientific background.

*A Booklet on Piezoelectricity – Essential Principles, Modeling and Emerging Technologies for Energy Harvesting and Sensing* is meant to be a gateway rather than a conclusion —a small book that raises significant questions. By compressing a century of progress into four focused chapters, it invites readers to rediscover a simple truth: that even the smallest vibrations of matter can illuminate the profound unity of the physical world.

**Dr. Ing. Massimo Mariello**

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Section 1

# Introduction

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## Chapter 1

# Introductory Chapter: Piezoelectricity – From Discovery to Devices

*Massimo Mariello*

## 1. Introduction

The phenomenon of piezoelectricity, first discovered in the late nineteenth century, represents one of the most elegant and useful manifestations of electromechanical coupling in nature. It describes the mutual conversion between mechanical and electrical energy within certain crystalline or molecular structures. Though simple in definition, piezoelectricity underpins a wide array of modern technologies—from medical ultrasound scanners to precision actuators and from vibration-based energy harvesters to tactile sensors and microelectronic timing devices. This introductory chapter provides a concise overview of the science and significance of piezoelectricity, tracing its historical origins, clarifying its fundamental mechanisms, and summarizing the main material classes and technological applications that define the field today. The purpose is not to offer exhaustive technical coverage, but rather to orient the reader within the intellectual and practical landscape of piezoelectric materials and devices, establishing a coherent framework for the chapters that follow in *Libellus de Electricitate Pressoria*.

## 2. A brief history of piezoelectricity

The discovery of piezoelectricity dates to 1880, when Jacques and Pierre Curie demonstrated that certain crystals, such as quartz, tourmaline, and Rochelle salt, generate an electric charge in response to mechanical pressure—a phenomenon soon known as the direct piezoelectric effect [1]. This finding was quickly followed, in 1881, by Gabriel Lippmann's prediction and experimental verification of the converse (reverse) piezoelectric effect, where an applied electric field produces mechanical deformation [2].

Initially, a subject of academic curiosity, piezoelectricity found its first major applications in the early twentieth century. During World War I, Paul Langevin used quartz crystals to construct the first underwater sonar transducer, marking the beginning of piezoelectric technology as a tool for sound generation and detection [3]. In the subsequent decades, the development of synthetic piezoelectric ceramics—particularly barium titanate ( $\text{BaTiO}_3$ ) and lead zirconate titanate (PZT)—revolutionized the field, allowing the large-scale fabrication of stable, high-performance transducers.

By the mid-twentieth century, piezoelectricity had moved from laboratory curiosity to industrial necessity, enabling innovations in ultrasound imaging, phonograph

needles, resonators, and frequency stabilizers. The last few decades have seen the emergence of polymeric and composite piezoelectrics, alongside bio-inspired and flexible materials, expanding the field beyond rigid ceramics toward soft, wearable, and implantable systems.

### **3. Working principles of piezoelectricity**

The word piezoelectricity derives from the Greek *piezein*, meaning “to press,” and describes the coupling between mechanical strain and electrical polarization in materials lacking a center of symmetry. When mechanical stress is applied, the atomic-scale dipoles within the crystal lattice become misaligned, generating a measurable potential difference—this is the direct effect. Conversely, applying an electric field induces a strain or deformation—the converse effect [4]. Mathematically, this relationship can be expressed using piezoelectric constitutive equations and piezoelectric coefficients [4, 5]. The magnitude and sign of the piezoelectric coefficients depend on both crystal symmetry and orientation. Only non-centrosymmetric crystals exhibit piezoelectricity, with 20 of the 32 crystallographic point groups possessing this property. In practice, polycrystalline materials such as ceramics require poling—the alignment of dipoles by applying a strong electric field during fabrication—to exhibit macroscopic piezoelectric behavior.

### **4. Major piezoelectric materials**

Piezoelectric materials can be broadly categorized into single crystals, ceramics, polymers, and composites.

Single crystals, such as quartz, lithium niobate ( $\text{LiNbO}_3$ ), and gallium orthophosphate ( $\text{GaPO}_4$ ), offer high stability, linearity, and reproducibility. Their well-defined crystal axes make them ideal for resonators and frequency-control devices.

Ceramics, most notably PZT, remain the most widely used class due to their large piezoelectric coefficients, ease of processing, and tunable properties. However, the presence of lead has driven extensive research into lead-free alternatives, such as barium titanate, potassium sodium niobate (KNN), and bismuth sodium titanate (BNT) [6].

Polymers, particularly polyvinylidene fluoride (PVDF) and its copolymers, combine flexibility, low acoustic impedance, and biocompatibility, making them valuable for flexible sensors and biomedical transducers [7–10].

Composites and nanostructured materials, including piezoelectric nanowires, thin films, and polymer–ceramic blends, represent the newest frontier, allowing tailored mechanical and electrical performance for wearable, implantable, and microelectromechanical systems (MEMS).

### **5. Technological applications and categories**

The unique ability of piezoelectric materials to transduce energy between mechanical and electrical domains has led to their use across diverse technologies [5, 11–14].

## **5.1 Sensors and actuators**

From pressure sensors and accelerometers to high-precision positioning systems and inkjet printheads, piezoelectrics enable compact and precise mechanical control.

## **5.2 Ultrasound and medical imaging**

Piezoelectric transducers generate and detect ultrasonic waves in diagnostic imaging, non-destructive testing, and therapeutic ultrasound.

## **5.3 Energy harvesting**

Vibrational and biomechanical energy can be converted into electricity using piezoelectric harvesters for powering small electronic devices and IoT sensors.

## **5.4 Acoustics and communication**

Surface acoustic wave (SAW) devices and resonators, based on quartz or LiNbO<sub>3</sub>, underpin timing and filtering functions in communication electronics.

## **5.5 Emerging applications**

Flexible and bio-integrated piezoelectric devices are enabling major advances across fields such as wearable electronics, neural interfaces, and smart drug-delivery systems, where soft, adaptive materials provide mechanical compatibility with living tissues. Beyond surface-mounted sensors and actuators, implantable piezoelectric technologies are now being developed for neural stimulation, electrophysiological monitoring, and energy harvesting inside the body. For instance, miniature piezoelectric transducers can convert externally applied ultrasound or mechanical vibrations into localized electrical stimuli, enabling wireless neuromodulation of brain or peripheral nerves without the need for tethered electrodes. Similarly, piezoelectric films and nanostructures have been integrated into self-powered implants, using natural body motion or physiological pressure to generate the energy required for long-term biosensing. These developments open pathways toward closed-loop therapeutic systems, where the same piezoelectric implant can sense, stimulate, and even control drug release in response to real-time physiological cues—paving the way for a new generation of minimally invasive, adaptive, and intelligent bioelectronic medicines.

Together, these applications demonstrate the versatility of piezoelectricity as a bridge between physical forces and electronic functions—an essential technology for both classical engineering and modern bioelectronic systems.

## **6. Concluding remarks**

Piezoelectricity has evolved from a laboratory discovery to a cornerstone of modern science and technology. As we enter an era dominated by flexible, adaptive, and biointegrated systems, the relevance of piezoelectric materials continues to expand. They embody a rare synergy: a direct and reversible link between mechanics and electricity, between motion and information.

This book invites readers to approach the subject not merely as a collection of phenomena, but as a dynamic interplay between disciplines—spanning crystallography, materials engineering, physics, and biomedical design. The following chapters explore these dimensions in more detail: from materials innovation to thin-film modeling, and from energy harvesting to sensor technologies. Readers are encouraged to move through these chapters not as isolated topics, but as interconnected aspects of a single, evolving field—one that continues to shape the interface between matter and motion.

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## Chapter 2

# Materials Innovation: Advances in Piezoelectric Materials

*Huda F. Khalil and Sara Gad*

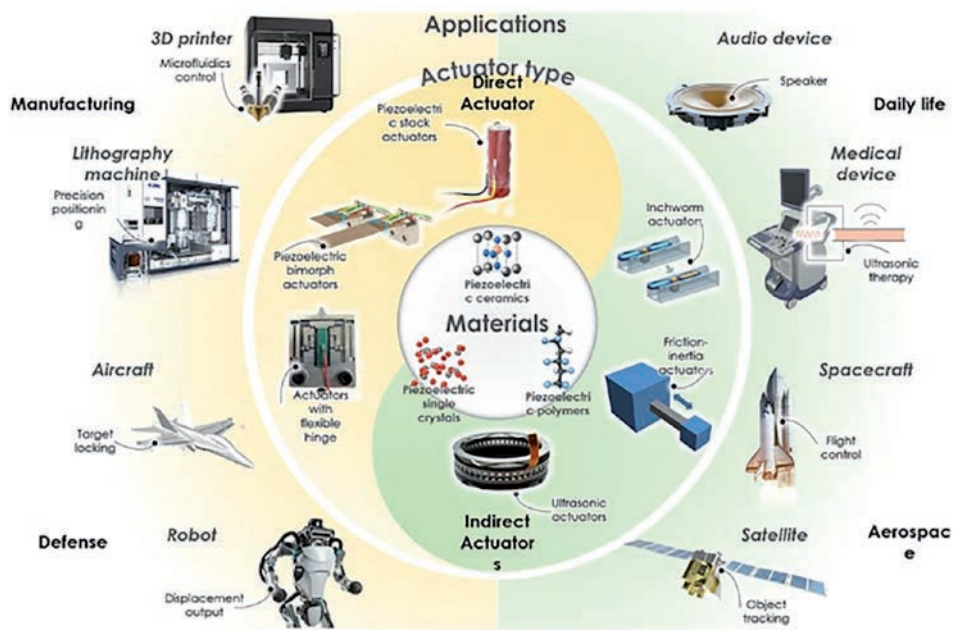
### Abstract

Piezoelectric materials are integral to modern technological innovations due to their unique ability to convert mechanical stress into electrical energy and vice versa. This chapter explores the evolution, properties, and wide-ranging applications of advanced piezoelectric materials. Beginning with a historical overview of natural materials like quartz and tourmaline, it transitions into the development of synthetic alternatives such as lead zirconate titanate (PZT) and polyvinylidene fluoride (PVDF). Fundamental concepts including the direct and inverse piezoelectric effects are discussed alongside distinctions between crystalline and amorphous structures. The chapter then delves into recent advances in organic, inorganic, and hybrid piezoelectric materials, emphasizing improvements in flexibility, sensitivity, and environmental compatibility. Key applications are highlighted across diverse fields: high-precision sensors and actuators, energy harvesting technologies, medical diagnostics and therapeutics, and consumer electronics. Innovations in manufacturing techniques such as additive manufacturing and nanostructuring are also examined for their role in enhancing material performance. Challenges related to durability, temperature sensitivity, and environmental sustainability are critically assessed, with a focus on the need for lead-free alternatives and eco-friendly fabrication. Economic factors influencing the commercialization of piezoelectric technologies are also considered. The chapter concludes with a discussion on emerging trends and future applications, underscoring the importance of interdisciplinary collaboration. Overall, this work provides a comprehensive analysis of the current landscape and future potential of piezoelectric materials in driving technological progress.

**Keywords:** piezoelectric materials, energy harvesting, smart sensors, lead-free ceramics, flexible electronics

### 1. Introduction

Piezoelectric materials are unique substances that generate an electric charge in response to applied mechanical stress. This phenomenon, known as the piezoelectric effect, results from the asymmetric arrangement of atoms within these materials, allowing them to convert mechanical energy into electrical energy and vice versa [1–3]. Common examples of piezoelectric materials include quartz crystals [4], certain ceramics



**Figure 1.**

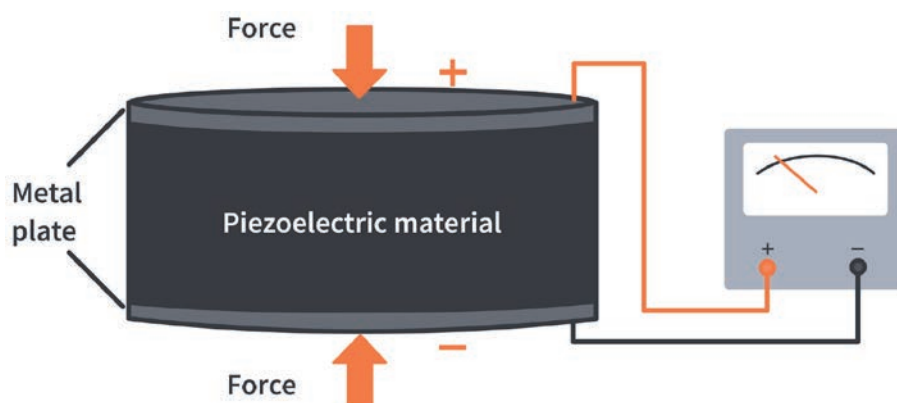
*The structure of piezoelectric materials and their wide-ranging applications, including sensors, actuators, energy harvesting devices, and advanced technologies in fields such as electronics, aerospace, and medical [12].*

(such as lead zirconate titanate [5]), and certain polymers. The importance of piezoelectric materials in modern technology cannot be overstated. They play a crucial role in a wide array of applications, from everyday devices like smartphones and microphones to advanced systems in aerospace [6], medical imaging [7], and energy harvesting [8]. Their ability to sense changes in pressure, acceleration, and force makes them indispensable in developing sensors [9], actuators [10], and transducers [11], (see **Figure 1**). As industries continue to innovate, the demand for high-performance piezoelectric materials is growing, driving research into new materials and technologies. This chapter will explore the advances in piezoelectric materials, beginning with a historical context that traces their development over time. We will delve into the fundamental principles of piezoelectricity and examine recent innovations in material composition and manufacturing techniques. The discussion will also cover the diverse applications of these materials, highlighting their significance across various sectors. Additionally, we will address the challenges faced by researchers and manufacturers in this field and provide insights into future directions for piezoelectric materials. By the end of this chapter, readers will gain a comprehensive understanding of the advancements in piezoelectric materials and their impact on technology and society.

## 2. Historical background

### 2.1 Early discoveries and applications

The phenomenon of piezoelectricity was first discovered in 1880 by the French physicists Pierre and Jacques Curie. They observed that certain crystals, such as quartz and tourmaline, produced an electric charge when subjected to mechanical



**Figure 2.**  
 Exploring piezoelectric materials [4].

stress (see **Figure 2**). This discovery laid the groundwork for future research into piezoelectric materials. Early applications included the use of quartz in phonograph pickups and other audio devices, where the material's ability to convert sound vibrations into electrical signals was harnessed [12]. As the understanding of piezoelectricity deepened, its applications expanded beyond audio technology. By the early twentieth century, piezoelectric materials were being used in various industries, including telecommunications and military technology, particularly in sonar and radar systems during World War II. Initially, the primary focus was on naturally occurring piezoelectric materials, such as quartz. However, the limitations in performance and availability prompted researchers to explore synthetic alternatives. The development of ferroelectric ceramics in the mid-twentieth century, particularly materials like lead zirconate titanate (PZT) [13], marked a significant evolution in the field. These materials exhibited enhanced piezoelectric properties, making them suitable for a broader range of applications. In the latter half of the twentieth century, the advent of polymers with piezoelectric properties, such as polyvinylidene fluoride (PVDF) [14, 15], further diversified the options available to engineers and researchers. These materials offered advantages like flexibility and lightweight characteristics, enabling their use in new sectors, including wearable technology and biomedical devices.

## 2.2 Key milestones in research and development

Several key milestones have shaped the development of piezoelectric materials:

- a. *Discovery of the piezoelectric effect (1880)* [16]: The Curie brothers' foundational work established the principles of piezoelectricity.
- b. *Introduction of ferroelectric ceramics (1950s)* [17]: The development of PZT revolutionized the industry by providing materials with superior piezoelectric response.
- c. *Advancements in polymer research (1970s)* [18]: The discovery of PVDF and other piezoelectric polymers opened new avenues for applications in flexible electronics and sensors.

- d. *Nano-piezoelectric materials (2000s)* [19]: The exploration of nanostructured piezoelectric materials, such as nanowires and thin films, has led to significant advancements in miniaturization and performance.
- e. *Recent innovations (2010s-present)* [20]: Ongoing research into composite materials and environmentally friendly alternatives has emerged, addressing concerns related to toxicity and sustainability.

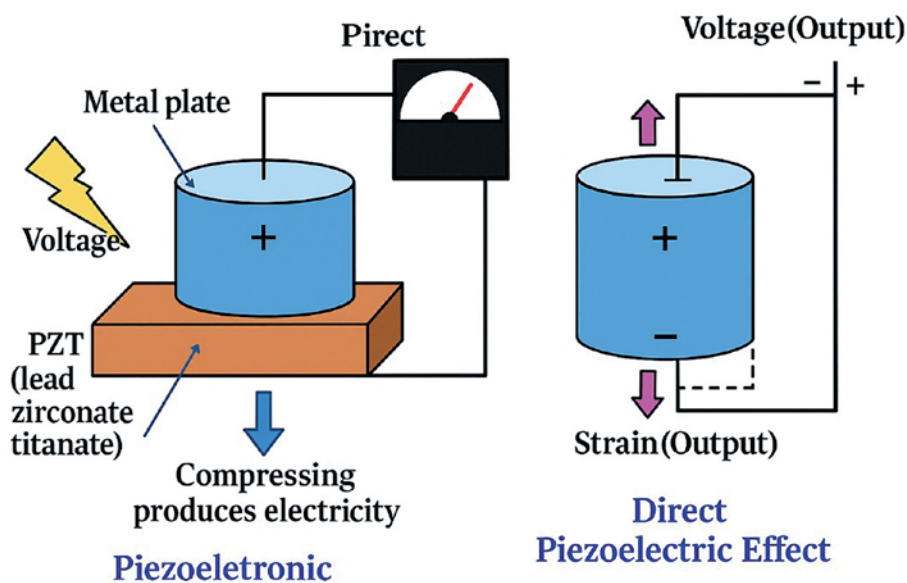
These milestones reflect a trajectory of continuous innovation and adaptation in the field of piezoelectric materials, setting the stage for the latest advancements and applications that will be discussed in subsequent sections.

### 3. Fundamentals of piezoelectricity

The piezoelectric effect is the ability of certain materials to generate an electric charge in response to applied mechanical stress. This phenomenon occurs due to the internal asymmetry in the crystal structure of piezoelectric materials [21]. When mechanical force is applied, the displacement of charged particles within the material creates a dipole moment, leading to an accumulation of charge on the surface. This effect can work in reverse as well; when an electric field is applied to a piezoelectric material, it induces mechanical deformation, allowing the material to act as an actuator. The piezoelectric effect can be classified into two categories:

- i. *Direct piezoelectric effect* [22]: The generation of electrical charge from mechanical stress.
- ii. *Conversely piezoelectric effect* [23]: The mechanical deformation caused by an applied electric field (see **Figure 3**).

These parameters help determine the suitability of a piezoelectric material for specific applications, such as sensors, actuators, energy harvesting devices, and medical ultrasound transducers. Piezoelectric materials can be natural, such as quartz, or synthetic, like lead zirconate titanate (PZT) [24], which is widely used due to its high piezoelectric response. Other notable piezoelectric materials include barium titanate ( $\text{BaTiO}_3$ ), zinc oxide (ZnO), and polyvinylidene fluoride (PVDF), a piezoelectric polymer. At the microscopic level, piezoelectricity arises from the non-centrosymmetric arrangement of atoms in the crystal lattice. In centrosymmetric materials, any applied stress results in symmetrical displacement of positive and negative charges, canceling out any net polarization. In contrast, in piezoelectric crystals [25], stress disrupts the balance of charges, resulting in a net dipole moment and observable electric polarization. Advances in material science have led to the development of piezoelectric nanomaterials, which exhibit enhanced properties due to their large surface-to-volume ratio and quantum effects. These nanostructured materials open new possibilities for miniaturized and highly sensitive piezoelectric devices, including flexible electronics and biomedical implants [26]. The efficiency and effectiveness of piezoelectric materials are quantified by parameters such as the piezoelectric coefficients, which measure the material's response to mechanical stress or electric field.



**Figure 3.**  
 Schematic diagram of the positive piezoelectric effect and the inverse piezoelectric effect (This figure generated by AI TOOL).

### 3.1 Natural vs. synthetic

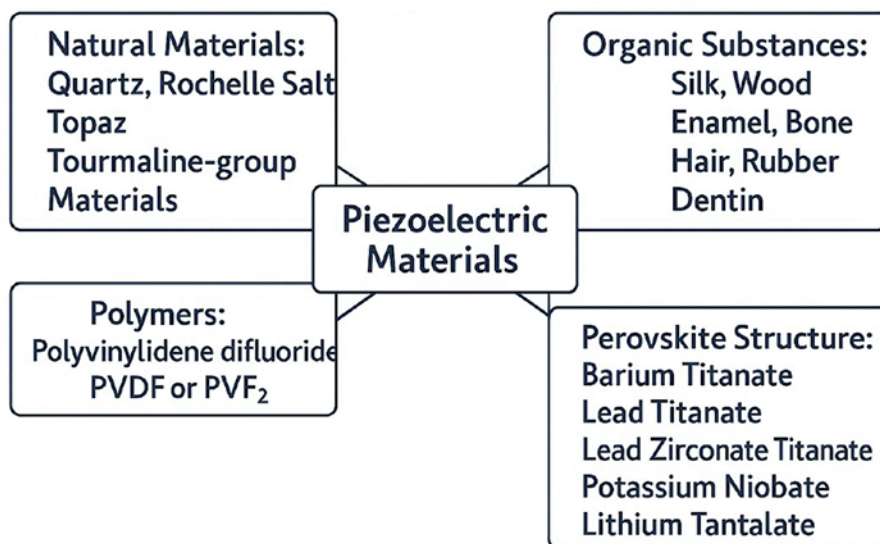
- **Natural piezoelectric materials:** These include naturally occurring minerals, such as quartz, tourmaline, and certain ceramics. While they are widely used due to their availability and ease of processing [27], their piezoelectric properties may be limited compared to synthetic alternatives.
- **Synthetic piezoelectric materials:** These materials are engineered to enhance specific properties and performance. Common examples include ferroelectric ceramics like lead zirconate titanate (PZT) [28] and polymers such as polyvinylidene fluoride (PVDF). Synthetic materials can be tailored to meet the demands of various applications, offering improved sensitivity, durability, and response times.

### 3.2 Types of piezoelectric materials

#### 3.2.1 Crystalline and non-crystalline materials

- Crystalline piezoelectric materials** [29]: These have a well-defined periodic atomic structure, which contributes to their piezoelectric properties. Common crystalline materials include quartz and various ceramics. The anisotropic nature of these materials means their piezoelectric response can vary based on the direction of applied stress.
- Non-crystalline (amorphous) piezoelectric materials** [30]: These lack a long-range ordered structure, which can lead to isotropic properties (see **Figure 4**).

## PIEZOELECTRIC MATERIALS



**Figure 4.**  
*Types of piezoelectric materials.*

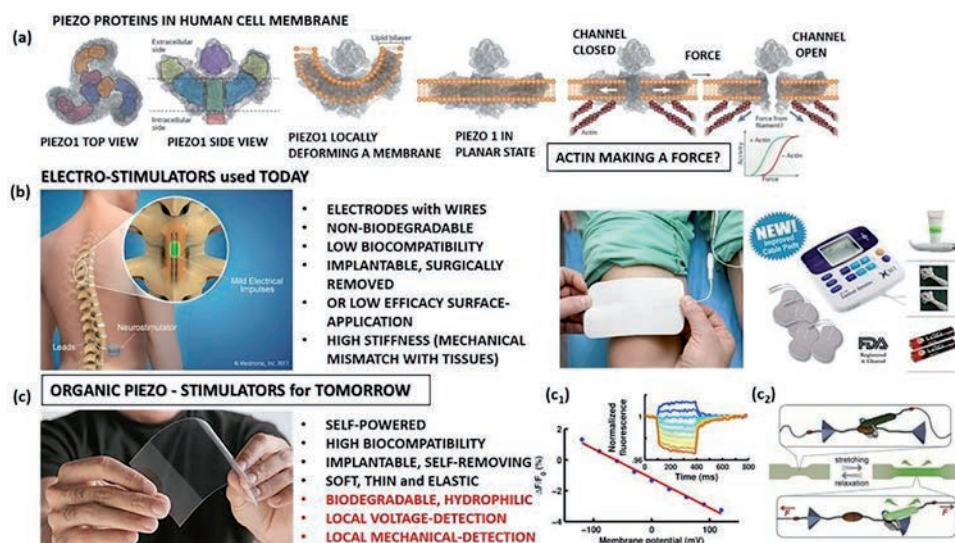
An example is certain polymers that exhibit piezoelectricity due to molecular dipoles. While they may not achieve the same levels of piezoelectric response as crystalline materials, they offer flexibility and ease of fabrication.

### 4. Recent advances in piezoelectric materials

#### 4.1 Organic piezoelectric materials

Recent years have seen significant interest in organic piezoelectric materials, particularly polymers, due to their unique properties such as flexibility, lightweight nature, and ease of processing [31]. Materials like polyvinylidene fluoride (PVDF) and its copolymers have been extensively studied and developed, resulting in enhanced piezoelectric performance (see **Figure 5**). Researchers are exploring modifications at the molecular level, such as doping with nanoparticles or blending with other polymers, to improve their piezoelectric response and thermal stability. These advancements enable organic materials to be used in applications ranging from wearable sensors to flexible electronics.

In addition to PVDF and its copolymers, other organic materials like polylactic acid (PLA) and cellulose-based materials have also exhibited piezoelectric properties, further expanding the range of options for eco-friendly [32] and biocompatible applications [33]. The inherent flexibility and processability of organic piezoelectrics make them ideal candidates for next-generation devices, including self-powered sensors, electronic skin, biomedical implants, and energy-harvesting systems. Recent innovations focus on nanostructuring techniques, such as electrospinning and 3D printing [34], to fabricate aligned nanofiber mats or patterned films that can significantly enhance the piezoelectric output.



**Figure 5.**  
 Exploring the role of piezo proteins in human cell membranes: Current applications and future innovations. This image illustrates the mechanisms of electro-stimulation used today with non-biodegradable electrodes, alongside promising organic piezo-stimulators of tomorrow that are self-powered, biodegradable, and capable of local mechanical detection [6].

Moreover, organic-inorganic hybrid composites are being developed to combine the high piezoelectric coefficient of inorganic nanoparticles with the mechanical advantages of polymers, achieving superior performance in flexible and wearable technologies.

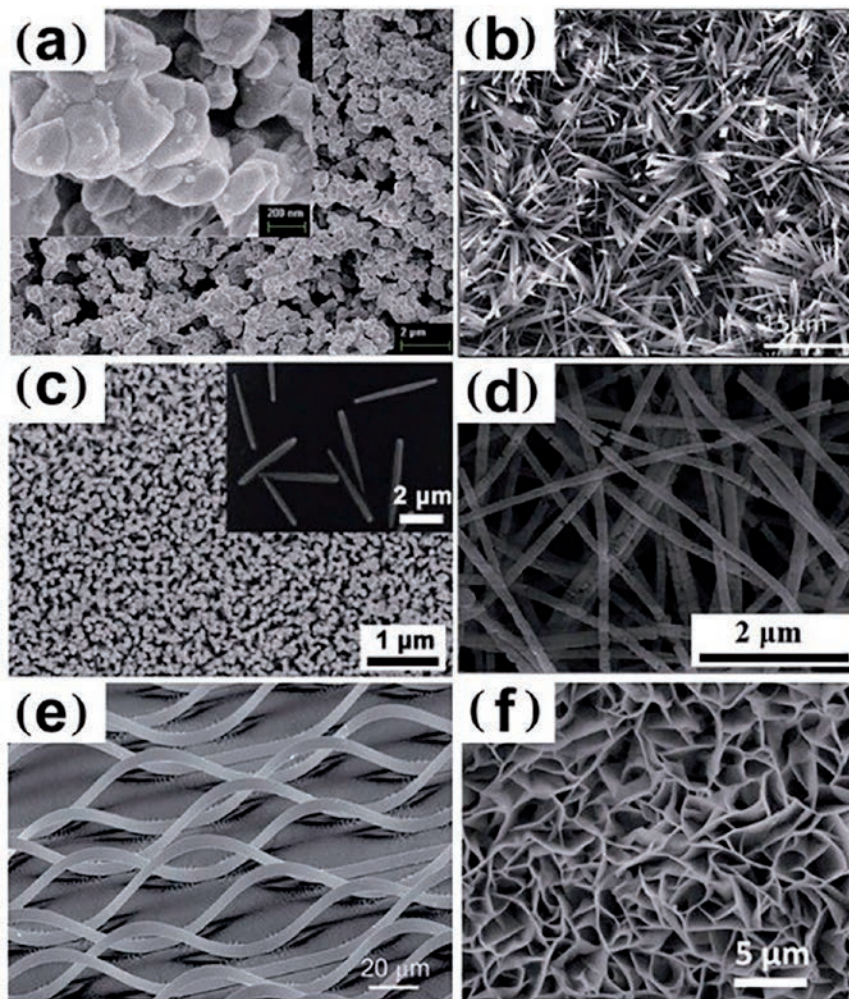
## 4.2 Inorganic piezoelectric materials

Inorganic piezoelectric materials, particularly ceramics and single crystals, continue to evolve. Lead zirconate titanate (PZT) remains a benchmark due to its high piezoelectric coefficients. However, researchers are developing lead-free alternatives, such as bismuth sodium titanate (BNT) [35] and potassium sodium niobate (KNN) [36], to address environmental concerns associated with lead-based materials, as well as Field-emission Scanning Electron Microscopy (FESEM) micrograph of BTO nanoparticle film, Scanning Electron Microscopy (SEM) image of PMN-PT nanowires, and surface morphology and longitudinal surface of ZnO nanorod arrays (see **Figure 6**). These new compositions aim to achieve similar or superior piezoelectric properties while ensuring compliance with sustainability standards.

Additionally, advancements in single-crystal piezoelectric materials, such as those based on relaxor ferroelectric compounds, are leading to enhanced performance. These materials exhibit high electromechanical coupling, making them suitable for high-precision applications like ultrasonic transducers and actuators.

## 5. Enhanced properties

Recent research has focused on enhancing the sensitivity, durability, and overall performance of piezoelectric materials. Innovations in material compositions and structures have led to:



**Figure 6.** Structures of inorganic piezoelectric materials. (a) Field-emission Scanning Electron Microscopy (FESEM) micrograph of BTO nanoparticle film. (b) Scanning Electron Microscopy (SEM) image of PMN–PT nanowires. (c) Overlooking surface morphology and longitudinal surface of ZnO nanorod arrays. (d) SEM image of  $(\text{Na}_{0.83}\text{K}_{0.17})_{0.5}\text{Bi}_{0.5}\text{TiO}_3$  nanofibers. (e) SEM image of buckled PZT ribbons under pre-strained conditions. (f) SEM image of ZnO sheets [7].

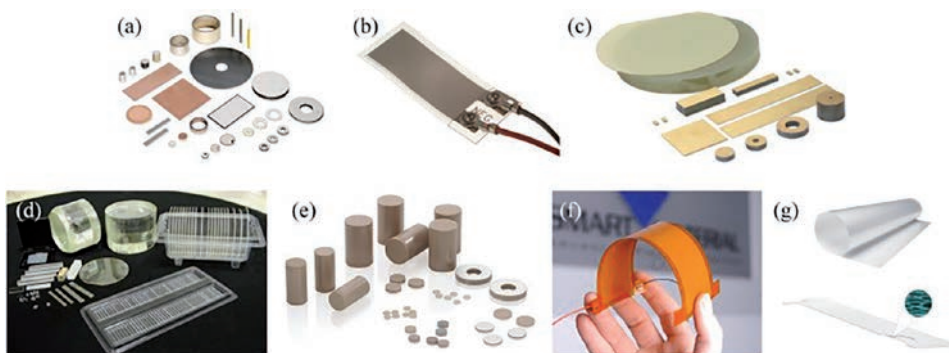
- *Increased sensitivity:* New formulations and hybrid materials combine the advantageous properties of both organic [37] and inorganic components [38], resulting in piezoelectric materials with significantly improved sensitivity. This enhancement is critical for applications in sensors and actuators where precision is essential.
- *Improved durability:* Advances in material science have led to the development of piezoelectric materials that exhibit greater resistance to fatigue and environmental factors, such as humidity and temperature fluctuations. Improved durability extends the lifespan and reliability of devices that utilize these materials.

- *Tailored mechanical properties*: Research into composite materials has enabled the creation of piezoelectric structures that can be customized for specific applications. By combining different materials, researchers can achieve tailored mechanical properties that enhance the overall performance of piezoelectric devices.

## 6. Advances in manufacturing techniques

The manufacturing techniques for piezoelectric materials have also seen significant advancements, contributing to their performance and application range (see **Figure 7**):

- *Additive manufacturing*: Techniques such as 3D printing are being employed to produce piezoelectric components with complex geometries. This allows for greater design flexibility and the creation of customized devices tailored to specific applications, such as sensors embedded in intricate structures.
- *Thin film deposition*: Methods like sol-gel processing, sputtering [39], and chemical vapor deposition [40] enable the fabrication of thin films of piezoelectric materials. These films can be used in microelectro-mechanical systems (MEMS) [41] and other miniaturized applications [42], providing high performance in compact devices.
- *Nanoengineering*: The manipulation of materials at the nanoscale has opened avenues for developing piezoelectric nanomaterials, such as nanowires and nanoparticles. These materials can exhibit enhanced piezoelectric properties and are being investigated for applications in energy harvesting and nanosensors [43].
- *Hybrid manufacturing techniques* [34]: Combining traditional methods with advanced techniques, such as integrating piezoelectric materials into existing electronic systems, is a growing trend. This approach allows for seamless integration of piezoelectric functionalities into a wide range of devices, enhancing their capabilities.



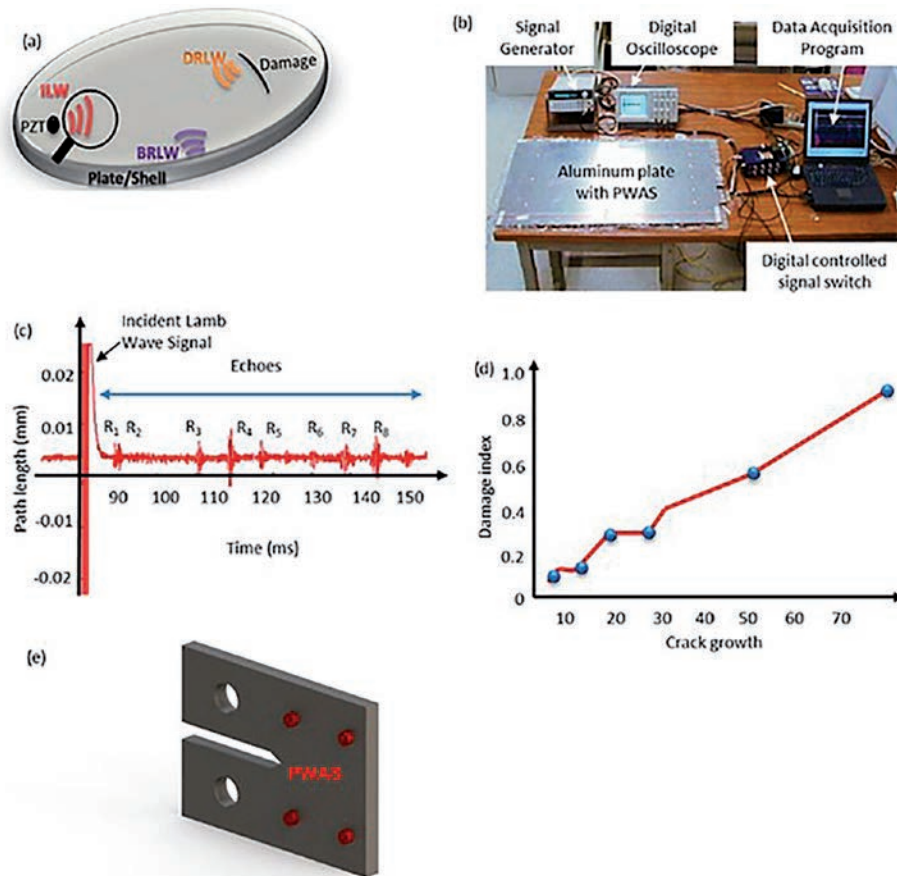
**Figure 7.** Various forms and configurations of piezoelectric materials: (a) Different shapes of ceramic piezoelectric elements; (b) Piezoelectric patch actuator; (c) Bulk piezoelectric ceramics in plates and disks; (d) Crystal-based piezoelectric materials; (e) Cylindrical and disk-shaped piezoelectric ceramics; (f) Flexible piezoelectric actuator used in wearable or adaptive devices; (g) Piezoelectric films for flexible electronics and sensor applications ) Piezoelectric films for flexible electronics and sensor applications [22].

These recent advances in novel materials, enhanced properties, and manufacturing techniques are driving the evolution of piezoelectric materials, expanding their applications and improving their performance in a variety of fields, from consumer electronics to medical devices.

## 7. Applications of advanced piezoelectric materials

### 7.1 Sensors and actuators

Advanced piezoelectric materials are integral to the development of high-performance sensors and actuators. In sensors, these materials can detect changes in pressure, force, and vibration, converting mechanical signals into electrical outputs [44, 45]. For example, piezoelectric sensors are widely used in automotive systems for crash detection and in structural health monitoring to assess the integrity of bridges and buildings (see **Figure 8**). In actuators, piezoelectric materials enable precise movement and control in



**Figure 8.** (a) A cracked aluminum plate showing incident Lamb waves (ILW), boundary reflected Lamb waves (BRLW), and damage reflected Lamb waves (DRLW) (b) Experimental setup showing the plate, active sensors, and instrumentation (c) the incident signal and the reflected (echoes) signals on sensor (d) Damage metric (e) A CT specimen with PWAS [35].

| Type of device      | Piezoelectric material        | Key functional benefits          | Additional applications                           | Important references |
|---------------------|-------------------------------|----------------------------------|---------------------------------------------------|----------------------|
| Pressure Sensor     | Quartz, PZT                   | High sensitivity, fast response  | Automotive airbag systems, touch screens          | [12]                 |
| Accelerometer       | Lead Zirconate Titanate (PZT) | Wide frequency range, durability | Seismic monitoring, industrial vibration analysis | [17]                 |
| Ultrasonic Actuator | PZT, PVDF                     | Precise displacement, low power  | Medical imaging, autofocus in cameras             | [22]                 |
| Microphone          | Quartz, PVDF                  | High fidelity, miniaturization   | Smartphones, hearing aids                         | [30]                 |
| Inkjet Printer Head | PZT                           | Accurate droplet control         | 3D printing, industrial marking                   | [44]                 |
| Force Sensor        | PVDF, PZT                     | Flexibility, high dynamic range  | Robotics, prosthetics                             | [31]                 |

**Table 1.**  
*Applications of advanced piezoelectric materials in sensors and actuators, detailing their specific types and the key functional benefits in various technological domains.*

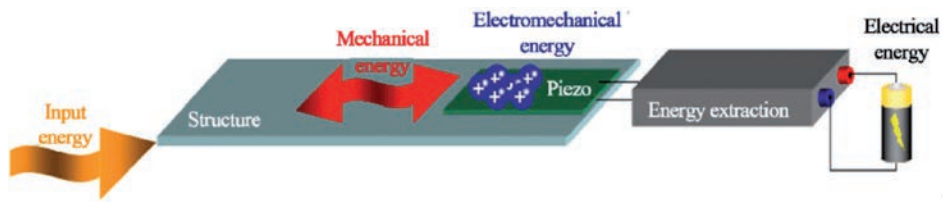
applications ranging from industrial automation to robotics (**Table 1**). They are used in devices such as piezoelectric motors and pumps, where their ability to produce rapid and controlled movements allows for high precision in tasks such as positioning and fluid delivery [46]. The combination of sensitivity and rapid response makes piezoelectric actuators essential in fields such as aerospace and telecommunications.

## 7.2 Energy harvesting technologies

Energy harvesting technologies are increasingly utilizing advanced piezoelectric materials to convert ambient mechanical energy into electrical energy. This process can capture energy from various sources, such as vibrations from machinery, foot-steps, or even wind-induced oscillations (see **Figure 9**). Piezoelectric energy harvesters are particularly valuable in powering low-energy devices [47], such as wireless sensors [48] in remote locations [49], where traditional power sources are impractical (**Table 2**). Recent innovations in material composition and design have led to more efficient energy harvesters that can generate substantial power from minimal mechanical input. These devices hold promise for applications in the Internet of Things (IoT), where they can provide sustainable power for sensor networks and smart devices.

## 7.3 Medical devices

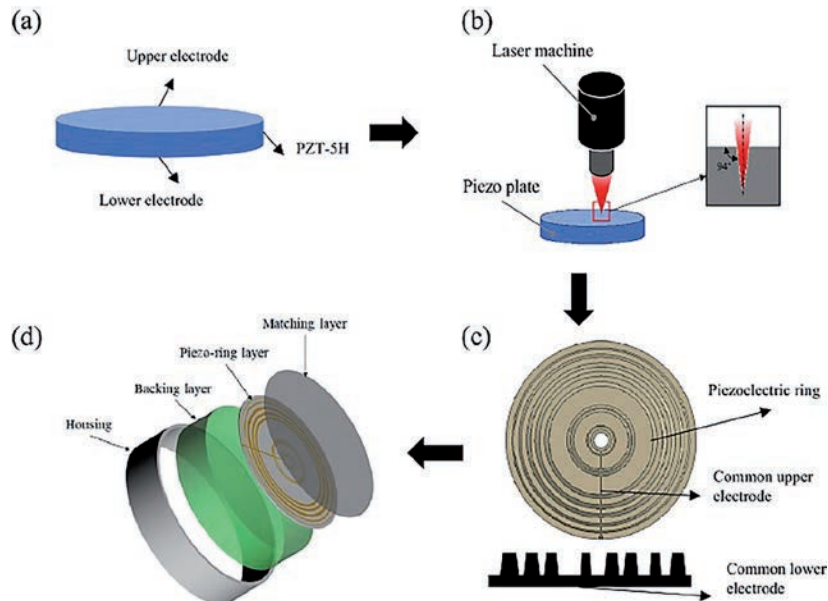
In the medical field, piezoelectric materials are crucial for several applications, most notably in ultrasound imaging. Piezoelectric transducers convert electrical signals into sound waves, which are then reflected back to create images of internal body structures [50]. The sensitivity and resolution of these transducers have significantly improved with advancements in piezoelectric materials, leading to better diagnostic capabilities (see **Figure 10**). Beyond imaging, piezoelectric materials are also employed in therapeutic ultrasound devices, which use focused ultrasound waves to treat conditions such as tumors and kidney stones (**Table 3**). Research continues to



**Figure 9.** Schematic diagram of a piezoelectric energy harvester, illustrating the conversion of ambient mechanical vibrations into electrical power. Source: [42].

| Energy source       | Harvester type             | Output power range | Application example              | Additional applications   | Important references |
|---------------------|----------------------------|--------------------|----------------------------------|---------------------------|----------------------|
| Footsteps           | Piezoelectric floor tiles  | 1–10 mW            | Smart buildings, wearable tech   | Wearable devices, bridges | [35]                 |
| Machine vibrations  | Cantilever beam harvesters | 10–100 mW          | Industrial IoT sensor nodes      | Footwear, smart watches   | [27]                 |
| Wind-induced motion | Flexible piezo             | <1 mW              | Environmental monitoring devices | Energy for micro-robots   | [19]                 |
| Acoustic Waves      | Thin Film Harvester        | 50–100 mW          | Industrial IoT sensor node       | Acoustic sensors          | [16]                 |
| Fluid Flow          | Flow-induced Harvester     | <1 mW              | Pipeline monitoring              | Marine sensors            | [32]                 |

**Table 2.** Overview of piezoelectric energy harvesting technologies, highlighting typical energy sources, corresponding harvester types, output power ranges, and representative application areas.



**Figure 10.** Illustration of a piezoelectric ultrasound transducer used in medical imaging. The transducer converts electrical signals into sound waves and vice versa, enabling internal body imaging. Copyright [26].

| Device type             | Piezoelectric component   | Functionality                     | Clinical use                      | Additional applications              | Important references |
|-------------------------|---------------------------|-----------------------------------|-----------------------------------|--------------------------------------|----------------------|
| Diagnostic ultrasound   | Piezoelectric transducer  | Imaging via sound wave reflection | Internal organ visualization      | Therapeutic ultrasound, elastography | [40]                 |
| Therapeutic ultrasound  | High-intensity transducer | Targeted heat delivery            | Tumor/kidney stone treatment      | Dental implants, spinal fusion       | [36]                 |
| Wearable health monitor | Thin piezoelectric patch  | Real-time biosignal monitoring    | Cardiac, respiratory monitoring   | Middle-ear implants                  | [42]                 |
| Drug Delivery Pump      | Piezoelectric actuator    | Precise fluid control             | Insulin pumps                     | Microfluidic lab-on-chip             | [33]                 |
| Surgical Tools          | Ultrasonic actuator       | High-precision cutting            | Cataract surgery, tissue ablation | Dental surgery                       | [11]                 |
| Bone Healing Device     | PZT                       | Electrical stimulation            | Accelerated bone repair           | Dental implants, spinal fusion       | [6]                  |

**Table 3.**  
 Key medical devices utilizing piezoelectric materials, describing their components, primary functions, and specific clinical applications in diagnostics and therapy.

explore new piezoelectric materials that enhance the performance and efficiency of medical devices, including wearable health monitors and implantable sensors [51].

## 7.4 Consumer electronics

Advanced piezoelectric materials are becoming increasingly prevalent in consumer electronics, contributing to the functionality and performance of various devices. In smartphones, piezoelectric sensors enable features such as touch sensing and haptic feedback, enhancing user interaction. Additionally, piezoelectric microphones are commonly used in audio devices, providing high-quality sound capture with minimal distortion [52]. Wearable technology is another area benefiting from piezoelectric advancements. Flexible piezoelectric materials allow for the integration of sensors into clothing and accessories, facilitating health monitoring and activity tracking. As the demand for smart and connected devices continues to grow, the integration of piezoelectric materials in consumer electronics is expected to expand further.

The applications of advanced piezoelectric materials span a wide range of fields, including sensors and actuators, energy harvesting, medical devices, and consumer electronics. The ongoing advancements in material science and manufacturing techniques are driving innovation and enabling new functionalities that enhance the performance and utility of these technologies.

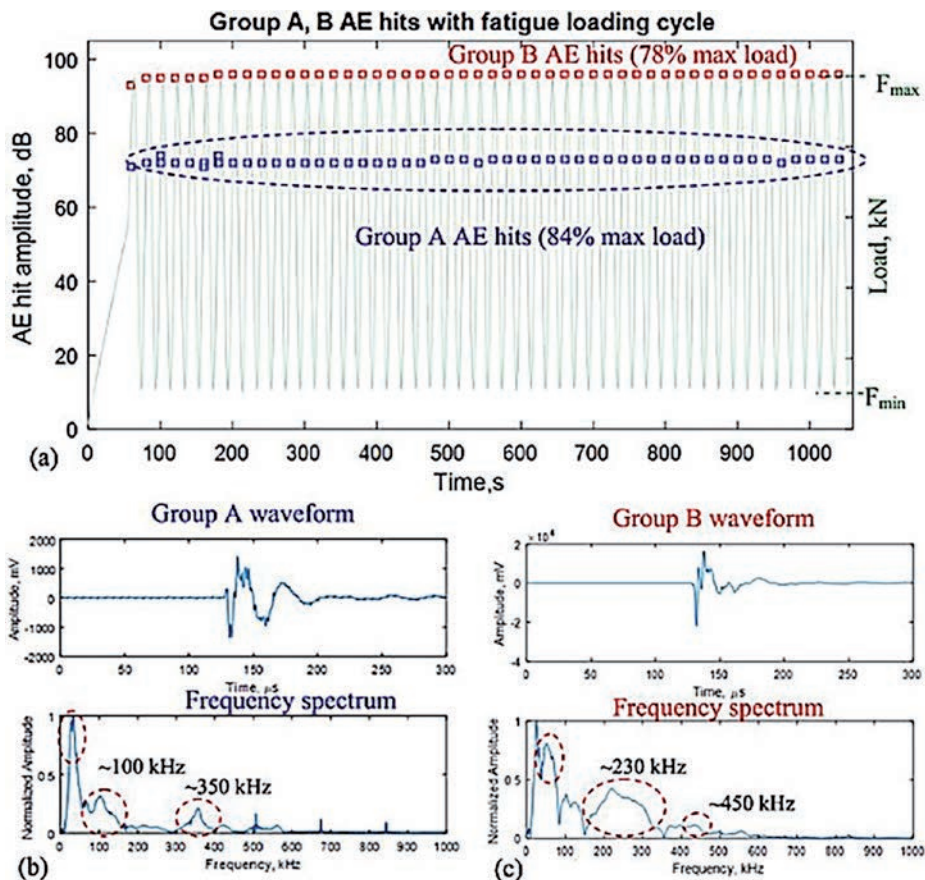
## 7.5 Material stability and performance issues

Despite the advancements in piezoelectric materials, several challenges remain regarding their stability and performance.

- i. *Fatigue and aging*: Many piezoelectric materials, particularly ceramics, can experience fatigue over time due to repeated mechanical stress. This leads to a

gradual decline in their piezoelectric properties [53], which can compromise the reliability of devices that rely on these materials (see **Figure 11**).

- ii. *Temperature sensitivity*: The performance of piezoelectric materials can be significantly affected by temperature variations (**Table 4**). Certain materials may undergo phase transitions that alter their piezoelectric properties at elevated temperatures, limiting their operating range in applications.
- iii. *Mechanical properties*: The brittleness of some inorganic piezoelectric materials, especially ceramics, presents challenges in applications where flexibility and resilience are required. Developing materials that combine high piezoelectric performance with enhanced mechanical durability remains a key focus of research.



**Figure 11.** (a) Group A, B AE hits are plotted in sync with the cyclic fatigue loading; groups A and B happened at every cycle but at different load levels; group A happened at 84% of maximum load level, while group B happened at 78% of maximum load level; (b), (c) the waveforms and frequency spectra of groups A and B, respectively; the major frequency peaks of each group are marked by dotted ellipses. Please note,  $F_{max} = 12.3$  kN,  $F_{min} = 1.23$  kN. Copyright [54].

| Composition                                                                                    | Unipolar strain at 25°C | Unipolar strain at 200°C | Temperature stability |
|------------------------------------------------------------------------------------------------|-------------------------|--------------------------|-----------------------|
| Pb(Ni <sub>0.11</sub> Hf <sub>0.33</sub> Ti <sub>0.34</sub> Nb <sub>0.22</sub> )O <sub>3</sub> | 1.63%                   | 0.24%                    | Poor                  |
| Pb(Ni <sub>0.11</sub> Hf <sub>0.21</sub> Ti <sub>0.46</sub> Nb <sub>0.22</sub> )O <sub>3</sub> | 0.78%                   | 0.79%                    | Excellent             |

**Table 4.**  
*Temperature stability of PNN–PHT piezoceramics. [37].*

## 8. Environmental considerations and sustainability

As the demand for piezoelectric materials grows, so do concerns regarding their environmental impact. So, many conventional piezoelectric materials, particularly lead-based ceramics like PZT, pose environmental and health risks due to their toxicity. As awareness of these dangers increases, there is a pressing need for alternatives that maintain performance without harmful effects (Table 5). The production processes for some piezoelectric materials can be energy-intensive and generate waste. Developing more sustainable manufacturing techniques and eco-friendly materials is crucial for minimizing the environmental footprint of piezoelectric technologies [55]. The disposal and recycling of piezoelectric devices also raise environmental concerns. Ensuring that materials can be safely recycled or disposed of is vital for long-term sustainability.

## 9. Economic factors affecting production and development

The economic landscape also plays a significant role in the development and adoption of advanced piezoelectric materials. The synthesis and processing of high-performance piezoelectric materials can be expensive, especially for cutting-edge materials like single crystals and advanced composites. These costs can hinder widespread adoption in consumer applications and limit market growth. The piezoelectric material market is competitive, with a range of alternative technologies vying for attention. Innovative solutions must not only outperform existing materials but also be cost-effective to gain market traction. Research and development in piezoelectric materials often requires significant investment. Economic downturns or shifts in funding priorities can impact the availability of resources for advancing this field, potentially slowing the pace of innovation.

Addressing these challenges requires collaborative efforts among researchers, industries, and policymakers to prioritize sustainable practices, enhance material performance, and reduce costs. By overcoming these limitations, the potential of piezoelectric materials can be fully realized across a wide range of applications.

| Sintering temperature (°C) | d <sub>33</sub> (pC/N) | k <sub>p</sub> (%) | Q <sub>m</sub> | d <sub>33</sub> * (pm/V) |
|----------------------------|------------------------|--------------------|----------------|--------------------------|
| 900                        | 650                    | 62.5               | 70             | 280                      |
| 940                        | 680                    | 65.0               | 70             | 290                      |
| 980                        | 701                    | 66.7               | 70             | 300                      |
| 1020                       | 690                    | 65.5               | 70             | 295                      |

**Table 5.**  
*Piezoelectric properties of 0.2 wt% Li<sub>2</sub>CO<sub>3</sub>-Modified PZT Ceramics.*

## **10. Future directions**

The field of piezoelectric materials is witnessing several emerging trends that promise to shape its future:

*Lead-free alternatives:* With increasing environmental concerns, research is intensifying on lead-free piezoelectric materials. Compounds such as bismuth sodium titanate (BNT) and potassium sodium niobate (KNN) are being explored for their potential to replace traditional lead-based materials while maintaining high performance.

*Nanostructured materials:* The development of nanostructured piezoelectric materials, including nanowires and nanoparticles, is gaining traction. These materials exhibit unique properties that can significantly enhance piezoelectric performance and open new avenues for applications in sensors and energy harvesting.

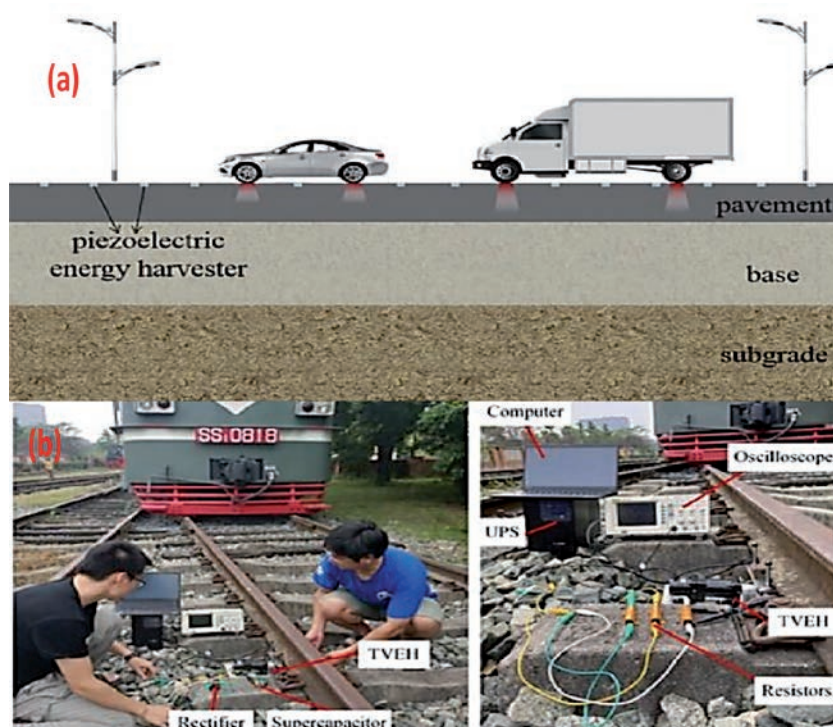
*Flexible and wearable technologies:* Research is focusing on creating flexible piezoelectric materials that can be integrated into wearable devices. Innovations in organic and hybrid materials are driving advancements in this area, offering the potential for health monitoring, fitness tracking, and smart textiles.

## **11. Potential applications in new fields**

As piezoelectric materials continue to evolve, their applications are expanding into new fields. Piezoelectric materials can play a crucial role in the development of smart city technologies, enabling energy harvesting from vibrations in urban environments, such as foot traffic or transportation systems (see **Figure 12**). This energy can power sensors and IoT devices, contributing to more sustainable urban infrastructure. The integration of advanced piezoelectric materials into biomedical devices is poised for growth. Applications may include implantable sensors for real-time health monitoring, drug delivery systems, and advanced ultrasound technologies for diagnostics and therapy. The demand for precise control and actuation in robotics is driving interest in piezoelectric materials. Their application in soft robotics, where flexibility and adaptability are essential, is an exciting frontier, enabling the development of more versatile and responsive robotic systems.

## **12. The role of interdisciplinary collaboration**

The advancement of piezoelectric materials is increasingly dependent on interdisciplinary collaboration. Collaboration between materials scientists and engineers is essential for developing new compositions and processing techniques that optimize piezoelectric properties. Understanding the fundamental mechanisms behind piezoelectric behavior requires insights from physics and chemistry, guiding the design of next-generation materials. Engaging with professionals in biomedical and environmental sciences can open up new application areas and address the sustainability



**Figure 12.**  
 (a) Example of PEH embedded inside the pavement and sustaining the vehicle load. (b) Harvester installation and system for field test. [53].

challenges associated with traditional materials. Collaborations between academia and industry are crucial for translating research findings into commercial products. Industry partners can provide insights into market needs and help streamline the development and manufacturing processes.

By fostering interdisciplinary collaboration, the field of piezoelectric materials can harness diverse expertise to drive innovation, tackle challenges, and explore new applications, ultimately enhancing their impact across various sectors. The future of piezoelectric materials is bright, with significant potential for advancements that can transform technology and improve quality of life.

### 13. Conclusion

Piezoelectric materials represent a cornerstone of modern smart materials technology, playing an increasingly vital role across various sectors including healthcare, energy, infrastructure, and robotics. This chapter has explored the evolution of these materials from natural crystals to advanced polymers and composites, highlighting the enhancements in performance, flexibility, and environmental compatibility. Emerging fabrication techniques such as additive manufacturing and nanoengineering are enabling tailored designs that meet specific application needs. Despite the challenges posed by fatigue, temperature sensitivity, and brittleness, ongoing research is steadily overcoming these barriers. The future of piezoelectric materials

lies in sustainable, lead-free alternatives; miniaturized smart systems; and interdisciplinary innovations that merge materials science, electronics, and bioengineering. As the demand for autonomous and intelligent systems grows, the continued advancement of piezoelectric technologies will be crucial to powering and sensing the next generation of devices.

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## Acronyms and abbreviations

|       |                                             |
|-------|---------------------------------------------|
| PZT   | lead zirconate titanate                     |
| PVDF  | polyvinylidene fluoride                     |
| BNT   | bismuth sodium titanate                     |
| KNN   | potassium sodium niobate                    |
| IoT   | internet of things                          |
| MEMS  | microelectromechanical systems              |
| SEM   | scanning electron microscopy                |
| FESEM | field emission scanning electron microscopy |
| AE    | acoustic emission                           |
| DRLW  | damage reflected lamb waves                 |
| BRLW  | boundary reflected lamb waves               |
| ILW   | incident lamb waves                         |
| CT    | computed tomography                         |

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## Section 2

# Modeling

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## Chapter 3

# Quartz Crystal Microbalance Modeling for Biocompatible Thin Films Studies

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### Abstract

This work presents an integrated approach combining instrumental modeling, multiscale analysis, and experimental validation to investigate water vapor interactions with thin hydrosoluble polymer films using a quartz crystal microbalance (QCM200). The advanced modeling of the QCM device, including mechanical energy trapping effects, enables accurate extraction of surface mass variations associated with water sorption. Diffusion, relaxation, and interfacial transport phenomena are first analyzed individually and then coupled within a unified framework to capture the experimentally observed sorption kinetics. The study highlights the dominant role of non-Fickian mechanisms, in particular the significant contribution of matrix mechanical relaxation. Comparative analysis of film formulations reveals the critical impact of carboxymethyl cellulose (CMC) on water uptake capacity, attributed to its strong hydrophilic affinity. Beyond its exceptional sensitivity, the QCM is shown to be a powerful investigative tool capable of resolving dynamic solvation processes within complex polymer matrices. These findings position the QCM not only as a precision measurement instrument, but also as a promising platform for the development of intelligent sensing technologies capable of real-time probing of solvent-polymer interactions.

**Keywords:** quartz crystal microbalance (QCM), biocompatible thin polymer films, water sorption, non-Fickian diffusion, interfacial mass transfer, polymer relaxation, Carboxymethyl cellulose (CMC), pharmacology, cosmetology

### 1. Introduction

Water-soluble polymer thin films play a pivotal role across a wide range of sectors, including pharmaceuticals (e.g., enteric coatings and controlled drug delivery systems) [1–3], cosmetics (e.g., transdermal patches and moisture-release facial masks) [4, 5], and the food industry (e.g., biodegradable packaging with barrier properties) [6, 7]. In all these applications, the performance of thin films critically depends on their interaction with ambient moisture, whether in the form of atmospheric water vapor or liquid water.

Understanding and controlling water sorption phenomena in these materials is essential for optimizing their mechanical properties, release functionalities, and

barrier performance. However, purely experimental approaches—though indispensable—rapidly reach their limits when it comes to providing a quantitative understanding of the underlying physicochemical mechanisms, especially in the case of complex polymer blends.

In this context, the quartz crystal microbalance (QCM) emerges as a particularly powerful and well-suited tool for detecting mass changes in the nanogram range, induced by water sorption or desorption, with exceptional sensitivity and real-time dynamic tracking capabilities. When combined with rigorous system-level modeling, QCM measurements enable the elucidation of the underlying interaction mechanisms between water molecules and the polymer matrix, providing deep insight into the coupled transport and structural relaxation processes governing sorption behavior.

This chapter focuses on modeling the interactions between water vapor in air and thin dry polymer films deposited on a quartz crystal microbalance (QCM). The dry films are obtained from aqueous solutions formulated with blends of the following water-soluble polymers: sodium alginate, sodium carboxymethylcellulose (CMC), and polyvinyl alcohol (PVA).

The present work follows a three-step integrated approach. First, an instrumental modeling of the QCM200 device is developed to rigorously relate shifts in resonance frequency to the amount of solvent absorbed or desorbed by the polymer film (see Section 2). The proposed model enables the interpretation of frequency signals as quantitative indicators of the water content evolution in the film under controlled sorption conditions. Second, a modeling framework for film-water vapor interactions is introduced (see Section 3), incorporating solvent diffusion within the material, potential structural relaxations of the polymer network, and interfacial mass transfer resistances. This framework allows the exploration of coupled regimes—diffusion-relaxation and interfacial transfer-relaxation—commonly observed in hydrophilic materials and enables the extraction of characteristic parameters from QCM-based kinetic measurements. Finally, the last part of the study presents and discusses experimental results in light of theoretical predictions (see Section 4). Sorption experiments conducted on various model films serve to assess the robustness of the proposed models and to identify the dominant underlying mechanisms.

When a dry polymer film is exposed to a humid atmosphere, it can capture water molecules both at its surface (adsorption) and within its bulk (absorption). These sorption phenomena can be modeled in various ways depending on the relative contributions of the capture mechanisms, the interfacial transfer regime, and the physical state of the film (glassy or rubbery). Water transport into the film occurs in two stages: interfacial mass transfer at the air-film boundary, followed by diffusion through the polymer matrix. Once inside the material, water molecules may be retained via chemical binding, entrapment within nanoscopic voids, or interactions with the polymer network. These interactions generate local mechanical stresses, which in turn trigger structural relaxations within the matrix. Such relaxation processes reorganize the polymer architecture and strongly influence both the sorption kinetics and the total water uptake. The overall behavior of the film thus results from a complex interplay between interfacial transfer, bulk diffusion, and mechanical relaxation of the polymer matrix.

This work aims to provide a coherent and quantitative framework for analyzing water sorption kinetics in thin water-soluble polymer films, by combining precision instrumentation, multiscale modeling, and experimental validation.

## 2. Instrumental modeling of the QCM200 quartz crystal microbalance

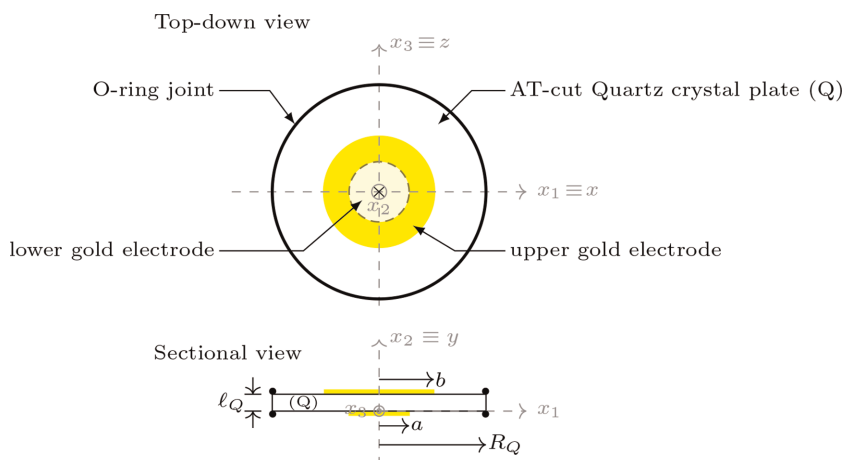
### 2.1 Operating principle of a quartz crystal microbalance (QCM)

A quartz crystal microbalance (QCM) is an ultra-sensitive mass sensor based on the measurement of the resonance frequency of an  $\alpha$ -quartz plate (see **Figure 1**), derived from a piezoelectric crystal cut along the AT orientation. This specific crystallographic cut enables pure shear-mode vibration at room temperature, with minimal thermal drift. Such a configuration is commonly employed in QCM devices due to its high frequency stability. When a thin film is deposited onto one of the electrodes of the quartz plate, any change in surface mass induces a shift in the oscillator's natural frequency. QCMs can achieve sensitivities on the order of nanograms per square centimeter, making them an ideal tool for detecting and analyzing adsorption, sorption, or deposition phenomena at the molecular scale. They are widely used in diverse fields, ranging from protein adsorption studies to the formation of lipid bilayers [8], detection of volatile organic compounds (VOCs) and air quality monitoring [9], as well as the assessment of water sorption in polymer matrices and the controlled release of active compounds in cosmetic and pharmaceutical applications.

In simplified terms, the quartz crystal microbalance (QCM) measures mass changes of a film deposited on its surface by tracking shifts in the resonance frequency of its pure shear vibrations. An increase in film mass leads to a proportional decrease in the resonance frequency.

### 2.2 Key equations and fundamental properties

Since  $\alpha$  – quartz is a piezoelectric crystal (trigonal system, class 32 [10]), the application of an electric potential difference  $V(t) = \phi(\ell_Q, t) - \phi(0, t)$  across its faces at  $y = \ell_Q$  and  $y = 0$  induces mechanical vibrations  $\mathbf{u}(\mathbf{r}, t)$  within the crystal. This



**Figure 1.** Schematic representation of an AT-cut quartz crystal (Q) used in a quartz crystal microbalance (QCM). The disk-shaped crystal (Q), with radius  $R_Q \approx 12.7$  mm and thickness  $\ell_Q \approx 330$   $\mu\text{m}$ , is equipped with two concentric metallic electrodes (gold in this case), shaped as disks with respective radii  $a \approx 3.57$  mm and  $b \approx 6.5$  mm. The thin films under investigation are typically deposited on the upper electrode of radius  $b > a$ . For clarity, the figure does not show the electrical leads connecting the electrodes to the control and measurement circuitry—responsible for exciting and tracking the crystal's resonance frequency in real time, which is not detailed in this document.

establishes an electromechanical coupling between the electric field  $\mathbf{E} = -\nabla\phi$  and the strain field  $\mathbf{u}(\mathbf{r}, t)$  inside the plate. In the ideal (lossless) case of a perfect crystal with no viscous dissipation, the system is governed by the following constitutive relations and physical properties (Einstein summation convention is assumed):

|                                                                                                                                                                                                                                                                                                                                                                                             |                                                                                                                                                                                                                                                                                                                                          |
|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| <p>Elasticity + Coupling : <math>\sigma_{jk} = C_{jklm}u_{lm} - e_{ijk}E_i</math></p> <p>Maxwell + ARQP : <math>\nabla \cdot \mathbf{D} = 0</math> and <math>\mathbf{E} = -\nabla\phi</math></p> <p>Electricity + coupling : <math>D_i = \varepsilon_{ij}E_j + e_{ijk}u_{jk}</math></p> <p>Cauchy equation : <math>\frac{\partial \sigma_{ij}}{\partial x_j} = \rho_Q \ddot{u}_i</math></p> | <p>with :</p> <p><math>\sigma_{jk}</math> : stress</p> <p><math>C_{jklm}</math> : elastic stiffness constants</p> <p><math>e_{ijk}</math> : piezoelectric coefficients</p> <p><math>\varepsilon_{ij}</math> : dielectric permittivity</p> <p><math>E_i</math> : Electric field</p> <p><math>D_i</math> : Electric displacement field</p> |
|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|

(1)

where the effect of gravity is neglected in Cauchy's equation, and the crystal is treated as a perfectly electrically insulating (ideal dielectric) medium in Maxwell's equation.

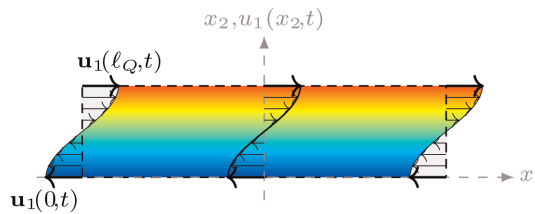
Under standard QCM operating conditions, the targeted motion is pure shear vibration. It is linearly polarized along the  $Ox_1 \equiv Ox$  direction, perpendicular to the propagation direction  $Ox_2 \equiv Oy$  (see **Figure 2**).

**Table 1** summarizes key physical properties of AT-cut quartz (adapted from Lu et al. [11] and Tiersten [12]) that are used in this work to model the operating behavior of the QCM200 quartz crystal microbalance.

## 2.3 Mathematical modeling

Different levels of modeling can be considered for the operation of a quartz crystal microbalance. In this work, we distinguish two main approaches:

- The idealized model (see Section 2.3.1), in which edge effects are neglected and the lateral dimensions of the quartz plate are assumed to be infinite compared to its thickness. Identical electrodes fully cover the plate surfaces at  $x_2 = 0$  and  $x_2 = \ell_Q$ , and a thin solid film is uniformly deposited on the top electrode at  $x_2 = \ell_Q$ .
- The edge-effect-inclusive model (see Section 2.3.3), in which the influence of the finite dimensions of both the quartz plate and its electrodes is accounted for. These effects primarily result in mechanical energy trapping between the electrodes.



**Figure 2.**

Pure shear oscillations of an ideal quartz plate with infinite lateral dimensions. The dashed rectangle represents the undeformed (rest) configuration of the crystal, while the solid line illustrates the deformation profile as a function of  $x_2$ . The amplitude of deformation has been exaggerated for visual clarity. In the ideal case, the thickness satisfies  $\ell_Q \ll \lambda_Q/2$ , where  $\lambda_Q$  denotes the wavelength of the pure shear waves within the quartz plate.

| Parameters                                                 | Values                                  | Description                                   |
|------------------------------------------------------------|-----------------------------------------|-----------------------------------------------|
| $\rho_Q$                                                   | 2648 kg/m <sup>3</sup>                  | Density of quartz                             |
| $C_{66}$                                                   | 29.47 GPa                               | Shear modulus of quartz                       |
| $\bar{C}_{66}$                                             | 29.70 GPa                               | Shear modulus with electromechanical coupling |
| $C_{55}$                                                   | 68.81 GPa                               | Shear modulus                                 |
| $C_{11}$                                                   | 86.74 GPa                               | Longitudinal shear modulus                    |
| $C_{56}$                                                   | 2.54 GPa                                | Shear modulus (cross-coupling)                |
| $e_{26}$                                                   | $-9.50 \times 10^{-2}$ C/m <sup>2</sup> | Piezoelectric constant                        |
| $\varepsilon_{22}$                                         | $39.82 \times 10^{-12}$ C/V/m           | Dielectric permittivity                       |
| $k_{26}^2 = \frac{e_{26}^2}{\bar{C}_{66}\varepsilon_{22}}$ | $7.63 \times 10^{-3}$                   | Electromechanical coupling coefficient        |
| $c_Q = \sqrt{\bar{C}_{66}/\rho_Q}$                         | 3348.8 m/s                              | Shear wave velocity in quartz                 |
| $\lambda_Q = 2\ell_Q$                                      | 660 $\mu$ m                             | Wavelength of shear waves                     |
| $\nu_{p,1} = \frac{c_Q}{2\ell_Q}$                          | 5.074004 MHz                            | Fundamental parallel resonance frequency      |

**Table 1.**  
Typical electromechanical parameters of an AT-cut quartz plate (adapted from Lu et al. [11] and Tiersten [12]).

In the present study, the analysis is restricted to the forced sinusoidal regime (FSR), which corresponds to the standard operating mode of a QCM.

### 2.3.1 Ideal model of a quartz crystal microbalance

A quartz plate with infinite dimensions in the  $(x_1, x_3)$  plane is considered first, allowing edge effects perpendicular to  $x_2$  to be neglected. Under these assumptions, the pure shear deformation field can be expressed in its idealized form as  $\mathbf{u}(\mathbf{r}, t) = u_1(x_2, t)\mathbf{e}_1$  with  $\mathbf{e}_1 \cdot \mathbf{e}_1 = 1$ , and the electric field as  $\mathbf{E}(\mathbf{r}, t) = E_2(x_2, t)\mathbf{e}_2$  with  $\mathbf{e}_2 \cdot \mathbf{e}_2 = 1$ .

With these expressions for the fields  $\mathbf{u}$  and  $\mathbf{E}$ , the constitutive relations (1) lead to the following differential equations, which describe the behavior of the idealized quartz crystal microbalance:

$$\begin{cases} \sigma_{12}(y, t) = C_{66}\partial_y u_1(y, t) + e_{26}\partial_y \phi(y, t) \\ \rho_Q \ddot{u}_1(y, t) = \bar{C}_{66}\partial_{y^2} u_1(y, t) \\ \partial_{y^2} \phi(y, t) = \frac{e_{26}}{\varepsilon_{22}} \partial_{y^2} u_1(y, t) \end{cases} \quad (2)$$

where  $\bar{C}_{66} = C_{66} + \frac{e_{26}^2}{\varepsilon_{22}}$ , and standard Voigt notation has been used. The second equation in system (2) corresponds to the propagation of a pure shear wave, whose velocity is given by  $c_Q = \sqrt{\bar{C}_{66}/\rho_Q}$ .

Quartz crystal microbalances are typically operated in the forced sinusoidal regime (FSR), using a signal generator that applies a harmonic excitation voltage  $V(t) = \phi(0, t) - \phi(\ell_Q, t) = 2\phi_0 \cos(\omega t)$  with angular frequency  $\omega = 2\pi\nu$ . In the ideal case, the shear wave solution to system (2) under FSR conditions can be expressed in the following complex form:

$$\underline{u}_1(y, t) = (\underline{A}_1 e^{ik_2 y} + \underline{A}_2 e^{-ik_2 y}) e^{i\omega t} = \hat{\underline{u}}_1(y) e^{i\omega t} \text{ and } \underline{\phi}(y, t) = \left[ \frac{e_{26}}{\epsilon_{22}} \hat{\underline{u}}_1(y) + \underline{A}_3 y + \underline{A}_4 \right] e^{i\omega t} \quad (3)$$

where  $k_2 = 2\pi\nu\sqrt{\rho_Q/\bar{C}_{66}}$  is the wavenumber, and  $\underline{A}_1, \underline{A}_2, \underline{A}_3$ , and  $\underline{A}_4$  are complex constants determined by the electrical and mechanical boundary conditions at  $y = 0$  and  $y = \ell_Q$ .

System (2) is supplemented by physical boundary conditions (BCs) on the faces  $y = 0$  and  $y = \ell_Q$ , involving both the mechanical shear stresses  $\sigma_{12}$  applied to the quartz plate (Neumann- or Robin-type conditions), and the electrical potential  $\phi$  imposed on each face (Dirichlet-type conditions). The specific nature of these boundary conditions is dictated by the operating mode of the microbalance. In this work, we focus on characterizing mass variations of a thin solid film, with uniform thickness  $\ell_F \ll \ell_Q$ , deposited on the top electrode of the QCM (see **Figure 3**).

Assuming perfect adhesion of the thin film to the top electrode, and considering that both the electrode (e) and the film (F) oscillate synchronously with the quartz surface at  $z = \ell_Q$  (since  $\ell_e, \ell_F \ll \ell_Q$ ), the application of Newton's second law to the electrode and to the film separately leads to the following ordinary differential equation:

$$\rho_e \ell_e \ddot{u}_{e,x}(\ell_Q, t) + \rho_F \ell_F \ddot{u}_{F,x}(\ell_Q, t) = -\sigma_{12}(\ell_Q, t) = -\sigma_{xy}(\ell_Q, t) \quad (4)$$

There is no slip between the film and the electrode, nor between the electrode and the quartz surface, so that  $\dot{u}_e = \dot{u}_F = \dot{u}_1$ , and the boundary condition at  $y = \ell_Q$  reads:

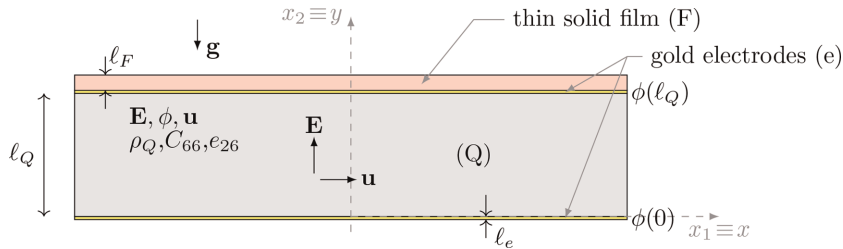
$$(\rho_e \ell_e + \rho_F \ell_F) \ddot{u}_1(\ell_Q, t) = -\sigma_{12}(\ell_Q, t) \quad (5)$$

We now define  $\sigma_e = \rho_e \ell_e$ ,  $\sigma_F = \rho_F \ell_F$ , and  $\sigma'_e = \sigma_e + \sigma_F = \sigma_e(1 + p)$ , where  $p = \sigma_F/\sigma_e$ . The boundary condition (5) thus becomes:

$$\sigma'_e \ddot{u}_1(\ell_Q, t) = -(C_{66} \partial_{x_2} u_1 + e_{26} \partial_{x_2} \phi)_{x_2=\ell_Q} \quad (6)$$

Applying the same approach to the lower electrode, which carries no film, leads to the second boundary condition:

$$\sigma_e \ddot{u}_1(0, t) = (C_{66} \partial_{x_2} u_1 + e_{26} \partial_{x_2} \phi)_{x_2=0} \quad (7)$$



**Figure 3.** Thin solid film (F) of uniform thickness  $\ell_F$  and density  $\rho_F$ , deposited on the top electrode (e) of a quartz crystal (Q) with infinite radius  $R_Q$ . The idealized case is considered here. The system is immersed in ambient air, whose effects are considered negligible.

Substituting results (3) into Eqs. (6) and (7), and using the electrical boundary conditions  $\phi(0) = \phi_0$  and  $\phi(\ell_Q) = -\phi_0$ , we obtain the following matrix system  $\mathbf{M} \cdot \mathbf{A} = (0, 0, \phi_0, -\phi_0)^T$ :

$$\begin{pmatrix} \sigma_e \omega^2 + ik_2 \bar{C}_{66} & \sigma_e \omega^2 - ik_2 \bar{C}_{66} & e_{26} & 0 \\ [ik_2 \bar{C}_{66} - \sigma'_e \omega^2] e^{ik_2 \ell_Q} & -[ik_2 \bar{C}_{66} + \sigma'_e \omega^2] e^{-ik_2 \ell_Q} & e_{26} & 0 \\ \frac{e_{26}}{\varepsilon_{22}} & \frac{e_{26}}{\varepsilon_{22}} & 0 & 1 \\ \frac{e_{26}}{\varepsilon_{22}} e^{ik_2 \ell_Q} & \frac{e_{26}}{\varepsilon_{22}} e^{-ik_2 \ell_Q} & \ell_Q & 1 \end{pmatrix} \cdot \begin{pmatrix} \underline{A}_1 \\ \underline{A}_2 \\ \underline{A}_3 \\ \underline{A}_4 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ \phi_0 \\ -\phi_0 \end{pmatrix} \quad (8)$$

which allows for the determination of the constants  $\underline{A}_1, \underline{A}_2, \underline{A}_3$ , and  $\underline{A}_4$ , provided that the determinant of matrix  $\mathbf{M}$  is nonzero for a given angular frequency  $\omega$ .

The pure shear wave  $u_1(y, t)$ , which solves system (2) under boundary conditions (6) and (7), is given from (8) by:

$$u_1(y, t) = \frac{N(y, \omega)}{D(y, \omega)} \cos(\omega t) \quad (9)$$

with:

$$\begin{cases} N(y, \omega) = 2e_{26}\varepsilon_{22}\phi_0\bar{C}_{66}k_2[\cos(k_2(\ell_Q - y)) - \cos(k_2y)] + \\ 2e_{26}\varepsilon_{22}\phi_0\omega^2[\sigma_e \sin(k_2y) - (\sigma_e + \sigma_F) \sin(k_2(\ell_Q - y))] \\ D(y, \omega) = \sin(\xi_Q) \left[ \frac{\bar{C}_{66}\xi_Q^2}{\ell_Q} - k_{26}^2\omega^2(2\sigma_e + \sigma_F) - \frac{\ell_Q\omega^4\sigma_e}{\bar{C}_{66}}(\sigma_e + \sigma_F) \right] \frac{e_{26}^2}{k_{26}^2} + \\ \frac{\bar{C}_{66}\xi_Q}{\ell_Q} \{ \cos(\xi_Q)[2e_{26}^2 + \ell_Q\omega^2\varepsilon_{22}(2\sigma_e + \sigma_F)] - 2e_{26}^2 \} \end{cases} \quad (10)$$

where  $k_{26}^2 = \frac{e_{26}^2}{\bar{C}_{66}\varepsilon_{22}}$  and  $\xi_Q = k_2\ell_Q$ .

Under standard operating conditions, the microbalance is driven by a harmonic voltage whose frequency induces resonance-referred to as series resonance and denoted here  $\nu_{s,1}^{e,F}$ -of the shear vibration amplitude  $u_1(y, t)$ . From a mathematical standpoint, the series resonance frequency corresponds to the first positive root of the denominator  $D(y, \omega)$  in the expression of  $u_1(y, t)$ . No exact analytical expression exists for the roots of  $D(y, \omega)$ , which must therefore be determined either numerically or through approximation.

Since the film (F) and the electrodes (e) have thicknesses much smaller than that of the quartz plate, these surface layers induce only a slight shift in the natural resonance frequency of the bare microbalance (i.e., without electrodes and without film), implying that the quantity  $\xi_Q$  remains close to  $\pi$ . Based on the expression (10) of  $D(y, \omega)$ , it can be readily shown that the first frequency  $\nu_{s,1}^{e,F}$  that cancels  $D(y, \omega)$  is very accurately approximated by:

$$\nu_{s,1}^{e,F} \approx \nu_{p,1} \left( 1 - \frac{4k_{26}^2}{\pi^2} \right) \left( 1 + \frac{2\sigma_e + \sigma_F}{\sigma_Q} \right)^{-1} \quad (11)$$

where  $\nu_{p,1} = \frac{c_Q}{2\ell_Q}$  is the parallel resonance frequency of the bare quartz plate, i.e., without electrodes or film (see **Table 1**). Under typical operating conditions of a quartz crystal microbalance, both  $\sigma_e$  and  $\sigma_F$  are on the order of  $10^{-3}$  kg/m<sup>2</sup>, while  $\sigma_Q$  is on the order of 1 kg/m<sup>2</sup>. It then follows that the frequency  $\nu_{s,1}^{\epsilon,F}$  can be approximated as:

$$\nu_{s,1}^{\epsilon,F} \approx \nu_{p,1} \left( 1 - R - \frac{\sigma_F}{\sigma_Q} - \frac{4k_{26}^2}{\pi^2} \right) \quad (12)$$

where  $R = 2\sigma_e/\sigma_Q \ll 1$  is the relative frequency lowering factor.

### 2.3.2 Sauerbrey relation

The standard use of a quartz crystal microbalance to determine mass variations of a thin film (F) involves measuring the changes  $\delta\nu_{s,1}^{\epsilon,F}$  in the series resonance frequency  $\nu_{s,1}^{\epsilon,F}$ , induced by changes  $\delta\sigma_F$  in the surface mass density  $\sigma_F$ , assuming constant film thickness and surface area. Based on result (12), one obtains:

$$\delta\nu_{s,1}^{\epsilon,F} = - \frac{\nu_{p,1}}{\sigma_Q} \delta\sigma_F = - C_\nu^s \delta\sigma_F \quad (13)$$

which is nothing other than the classical Sauerbrey relation [13], where  $C_\nu^s =$

$\nu_{p,1}/\rho_Q \ell_Q = 2\nu_{p,1}^2/\sqrt{\rho_Q \bar{C}_{66}}$  denotes the Sauerbrey constant of the quartz plate under consideration, in the absence of electrodes and film.

The present approach demonstrates that while the electrodes (e) can significantly affect the series resonance frequency  $\nu_{s,1}^{\epsilon,F}$  of the microbalance, they have no influence on the measurement of its variations  $\delta\nu_{s,1}^{\epsilon,F}$ . We employed relation (13) to quantify the mass changes of polymer films induced by water vapor sorption and desorption.

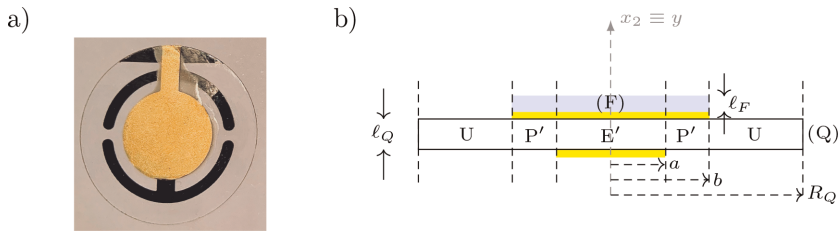
Although the Sauerbrey relation (13) is relatively simple, its practical application can be challenging when the constant  $C_\nu^s$  is not known with sufficient accuracy. The following paragraph specifies the appropriate value of  $C_\nu^s$  to be used for the QCM200 microbalance employed in this study.

### 2.3.3 Accounting for edge effects: Energy trapping

The ideal modeling presented in Section 2.3.1 predicts a uniform mass sensitivity  $S_m$ , which is not observed experimentally due to the finite dimensions of the quartz crystal (Q) and its electrodes (e). A more refined model of shear wave propagation within the crystal is therefore required, taking edge effects into account.

#### 2.3.3.1 Mathematical modeling

We now consider the more realistic configuration depicted in **Figure 4**, in which a solid film is uniformly deposited on the upper electrode (with finite radius  $b$ ) of a cylindrical quartz crystal with axis  $Oy$  and finite radius  $R_Q$  (see **Figure 4a**). The lower electrode (with finite radius  $a < b$ ), the quartz crystal, and the upper electrode are all coaxial (see **Figure 4b**).



**Figure 4.** Solid film (F) deposited on the upper electrode of a quartz crystal (Q) in a QCM200 microbalance: a) Quartz crystal and its solid coating on the upper electrode. b) Schematic representation of the microbalance and the division into regions U, P', and E'.

Three distinct regions are identified:

1. Region U ( $b \leq r \leq R_Q$ ): no electrode or film is present on either face of the quartz crystal.
2. Region P' ( $a \leq r \leq b$ ): only the upper face carries an electrode (with surface mass density  $\sigma_e$ ) and a solid film (with surface mass density  $\sigma_F$ ).
3. Region E' ( $0 \leq r \leq a$ ): both faces carry electrodes, but only the upper electrode supports the solid film, while the lower electrode remains uncovered.

To account for the influence of finite dimensions along the  $Ox_1$  and  $Ox_3$  directions, the shear wave is now expressed in the following form:

$$u_1(x_1, x_2, x_3, t) = \hat{u}_1(x_1, x_2, x_3) \cos(\omega t) \quad (14)$$

Assuming that the electric field has only one non-zero component along the  $x_2$  direction, the Cauchy and Maxwell equations simplify to the following form:

$$\begin{cases} \rho_Q \ddot{u}_1 = C_{11} \frac{\partial^2 u_1}{\partial x_1^2} + C_{66} \frac{\partial^2 u_1}{\partial x_2^2} + C_{55} \frac{\partial^2 u_1}{\partial x_3^2} + e_{26} \frac{\partial^2 \phi}{\partial x_2^2} \\ e_{26} \frac{\partial^2 u_1}{\partial x_2^2} = \epsilon_{22} \frac{\partial^2 \phi}{\partial x_2^2} \end{cases} \quad (15)$$

This system of partial differential equations (PDEs) can be reduced to a single equation:

$$\overline{C}_{66} \frac{\partial^2 u_1}{\partial x_2^2} + M_1 \overline{C}_{66} \frac{\partial^2 u_1}{\partial x_1^2} + M_3 \overline{C}_{66} \frac{\partial^2 u_1}{\partial x_3^2} = \rho_Q \ddot{u}_1 \quad (16)$$

with  $\overline{C}_{66} = C_{66} + \frac{e_{26}^2}{\epsilon_{22}}$ ,  $M_1 = \frac{C_{11}}{C_{66}} \approx 2.92$ , and  $M_3 = \frac{C_{55}}{C_{66}} \approx 2.32$ . Substituting expression (14) into Eq. (16) yields an eigenmode differential equation for the amplitude  $\hat{u}_1$ :

$$\overline{C}_{66} \frac{\partial^2 \hat{u}_1}{\partial x_2^2} + M_1 \overline{C}_{66} \frac{\partial^2 \hat{u}_1}{\partial x_1^2} + M_3 \overline{C}_{66} \frac{\partial^2 \hat{u}_1}{\partial x_3^2} = -4\pi^2 \rho_Q \nu^2 \hat{u}_1 \quad (17)$$

The eigenvalue PDE (17) does not admit an exact analytical solution over the domain  $\Omega$  formed by the cylindrical quartz plate, its coaxial electrodes, and the thin

film (see **Figure 4b**). This constitutes a fully three-dimensional (3D) boundary value problem, which can only be solved numerically-using the finite element method, for example.

Noting that the constants  $M_1$  and  $M_3$  have similar values, the dimensionality of the problem can be reduced by replacing them with  $M = (M_1 + M_3)/2$ . This leads to the following eigenvalue PDE:

$$\bar{C}_{66} \frac{\partial^2 \hat{u}_1}{\partial x_2^2} + M \bar{C}_{66} \left( \frac{\partial^2 \hat{u}_1}{\partial x_1^2} + \frac{\partial^2 \hat{u}_1}{\partial x_3^2} \right) = -4\pi^2 \rho_Q \nu^2 \hat{u}_1 \quad (18)$$

Adopting cylindrical coordinates:  $x_1 \equiv r \cos(\theta)$ ,  $x_3 \equiv r \sin(\theta)$ , and  $x_2 \equiv y$ , with  $r \in [0, R_Q]$ ,  $\theta \in [0, 2\pi]$ , and  $y \in [0, \ell_Q]$ , Eq. (18) can be rewritten as:

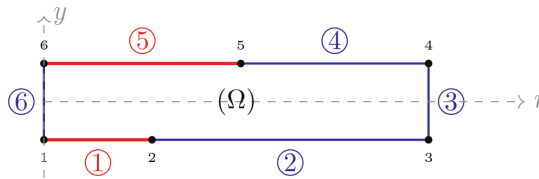
$$\bar{C}_{66} \frac{\partial^2 \hat{u}_1}{\partial y^2} + M \bar{C}_{66} \left( \frac{\partial^2 \hat{u}_1}{\partial r^2} + \frac{1}{r} \frac{\partial \hat{u}_1}{\partial r} + \frac{1}{r^2} \frac{\partial^2 \hat{u}_1}{\partial \theta^2} \right) = -4\pi^2 \rho_Q \nu^2 \hat{u}_1 \quad (19)$$

By observing the sensing surface of a quartz crystal microbalance using a scanning electrochemical microscope (SECM), Hess et al. [14] demonstrated that the amplitude  $\hat{u}_1$  of the fundamental vibration mode of the quartz crystal-equipped with its electrodes-exhibits, to a very good approximation, axial symmetry, despite the anisotropy of the quartz and its vibration mode. This observation implies that  $\hat{u}_1$  is independent of  $\theta$ , thereby allowing simplification of the PDE (19) into the following two-dimensional form:

$$\frac{\partial}{\partial y} \left( \bar{C}_{66} \frac{\partial \hat{u}_1}{\partial y} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left( M \bar{C}_{66} r \frac{\partial \hat{u}_1}{\partial r} \right) = -4\pi^2 \rho_Q \nu^2 \hat{u}_1 \quad (20)$$

We solved the partial differential Eq. (20) using the finite element method over the domain  $\Omega$  shown in **Figure 5**, where the boundary conditions (BC) on the contour  $\partial\Omega$  enclosing the domain are defined as follows:

- Segments ②, ③, ④, and ⑥: Homogeneous Neumann boundary conditions,  $(\nabla \hat{u}_1) \cdot \mathbf{n} = 0$ .
- Segment ① (lower electrode): Robin boundary condition,  $\bar{C}_{66}(\nabla \hat{u}_1) \cdot \mathbf{n} = 4\pi^2 \sigma_e \nu^2 \hat{u}_1$ .
- Segment ⑤ (upper electrode and film): Robin boundary condition,  $\bar{C}_{66}(\nabla \hat{u}_1) \cdot \mathbf{n} = 4\pi^2 (\sigma_e + \sigma_F) \nu^2 \hat{u}_1$ .



**Figure 5.** Domain  $\Omega$  and boundary  $\partial\Omega$  used for the finite element analysis of eigenmodes in the QCM200 microbalance under axial symmetry.

Surface mass sensitivity: The surface mass sensitivity  $S_m$  is a fundamental parameter of a quartz crystal microbalance (QCM). It quantifies how the series resonance frequency  $\nu_{s,1}^{\text{em}}$  responds to variations in mass distribution across the crystal surface. Assuming cylindrical symmetry of the QCM, the local surface mass sensitivity  $S_m(r, \theta)$  is defined by the following relation:

$$\Delta\nu_{s,1} = -S_m(r, \theta)\Delta m(r, \theta) \quad (21)$$

where  $\Delta\nu_{s,1}$  denotes the shift in the series resonance frequency of the quartz crystal (Q) induced by the addition of a point mass  $\Delta m$  at the polar coordinates  $(r, \theta)$ , starting from an initial configuration in which the crystal carries no additional loading other than the metallic electrode.

For a non-localized deposit characterized by a surface mass density  $\sigma_m(r, \theta)$ , Eq. (21) becomes [15]:

$$\Delta\nu_{s,1} = - \int_{\theta=0}^{2\pi} \int_{r'=0}^r S_m(r', \theta') \sigma_m(r', \theta') r' d\theta' dr' \quad (22)$$

Josse et al. proposed an analytical expression for the surface mass sensitivity  $S_m(r, \theta)$  under cylindrical symmetry, given in the following form [16]:

$$S_m(r, \theta) = \frac{|\hat{u}_1(r, \theta, \ell_Q)|^2}{\int_0^\infty \int_0^{2\pi} r' |\hat{u}_1(r', \theta, \ell_Q)|^2 d\theta dr'} C_\nu^s \quad (23)$$

The axial symmetry observed by Hess et al. [14] allows expression (23) to be simplified as follows:

$$S_m(r) = \frac{|\hat{u}_1(r, \ell_Q)|^2}{2\pi \int_0^\infty r' |\hat{u}_1(r', \ell_Q)|^2 dr'} C_\nu^s \quad (24)$$

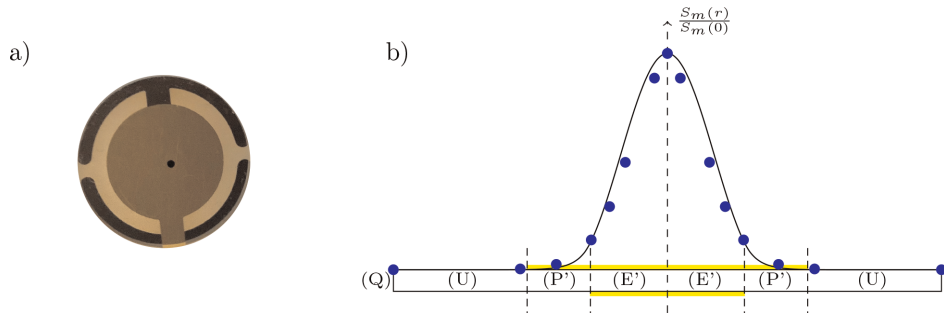
It is worth noting that the ideal case developed in Section 2.3.1 leads to a uniform mass sensitivity  $S_m^{\text{id}}$ , since in that configuration  $\hat{u}_1(r, \ell_Q)$  is constant:

$$S_m^{\text{id}} = \frac{C_\nu^s}{\pi R_Q^2} \quad (25)$$

### 2.3.3.2 Results and discussion

The experimental measurement of the mass sensitivity is carried out by depositing calibrated ink spots of equal area (one spot per measurement, see **Figure 6a**) on the upper surface of the quartz crystal. These spots are created using an Indian ink marker with a 0.7 mm tip. The frequency shift is recorded once the deposit has dried. The distance  $r$  from the spot to the center of the quartz crystal is determined through image analysis.

**Figure 6b** shows the results of the experimental characterization (symbols) of the surface mass sensitivity  $S_m(r)$  for the QCM200 microbalance used in this study, along with the prediction from the present model (solid line), which accounts for edge effects.



**Figure 6.**

Surface mass sensitivity  $S_m(r)$  of a QCM200 microbalance (without deposited film). (a) Ink spot deposited at a distance  $r \neq 0$  from the center of the upper surface of the microbalance. (b) Graphical representation of  $S_m(r)/S_m(0)$  obtained experimentally (symbols) and from axisymmetric finite element modeling (solid line).

Prior to any deposition on the upper electrode, the unloaded series resonance frequency  $\nu_{s,1}^0$  of the QCM200 is measured experimentally. For the set of quartz crystal units used in this study, the measured value was found to be  $\nu_{s,1}^0 = 5.0154335\text{MHz}$ , as shown in **Figure 7**.

To achieve the best possible agreement between the model and the experimental results, the thicknesses  $\ell_Q$  and  $\ell_e$  were adjusted in the model to  $\ell_Q = 329\text{ }\mu\text{m}$  and  $\ell_e = 428\text{ nm}$ . According to the manufacturer's datasheet, the nominal values are  $\ell_Q = 331\text{ }\mu\text{m}$  and  $\ell_e = 100 - 1000\text{ nm}$ . The adjusted values used in this study are therefore consistent with the specifications provided by the manufacturer.

From the refined model parameters, the following value of the Sauerbrey constant was obtained via finite element analysis:

$$C_\nu^s = 56.51\text{ Hz} \cdot \mu\text{g}^{-1} \cdot \text{cm}^2 \quad (26)$$

According to the manufacturer, the Sauerbrey constant is specified as  $56.6\text{ Hz} \cdot \mu\text{g}^{-1} \cdot \text{cm}^2$  at room temperature.

It is also worth noting that the curve  $S_m(r)/S_m(0)$ , shown in **Figure 6b**, demonstrates that the shear wave is primarily confined to region E'-that is, the portion of the quartz crystal (Q) where both faces are electrode-coated. As a result, the mechanical energy becomes trapped in this region (energy trapping). It follows that the active sensing area  $S_e$  of the microbalance is  $S_e = \pi a^2 \approx 0.4\text{ cm}^2$ .



**Figure 7.**

Quartz crystal microbalance QCM200 used and modeled in this study. The unloaded series resonance frequency (without any deposited material) is measured and displayed by the control electronics.

When the film spread over the upper electrode (see **Figure 4a**) is homogeneous with uniform thickness  $\ell_F$ , Eq. (22) becomes, under the assumption of axial symmetry and using expression (24):

$$\Delta\nu_{s,1} = -2\pi\sigma_F \int_{r'=0}^{R_Q} S_m(r')r'dr' = -2\pi\sigma_F \frac{\int_{r'=0}^{R_Q} r' |\hat{u}_1(r', \ell_Q)|^2 dr'}{2\pi \int_0^\infty r' |\hat{u}_1(r', \ell_Q)|^2 dr'} C_\nu^s \quad (27)$$

Since, as shown in **Figure 6b**, the mechanical wave is confined within region E', the two integrals appearing in Eq. (27) are equal, once again yielding the Sauerbrey relation, expressed as:

$$\Delta\nu_{s,1} = -\sigma_F C_\nu^s \quad (28)$$

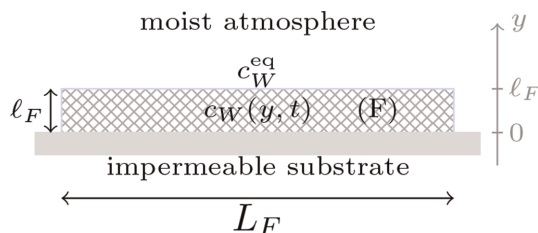
where  $\Delta\nu_{s,1} = \nu_{s,1}^F - \nu_{s,1}^0$ , with  $\nu_{s,1}^0$  and  $\nu_{s,1}^F$  being the resonance frequencies in the absence and presence of a thin film, respectively. If the film's surface mass density varies over time (due to interaction with the solvent), the result reduces to Eq. (13), previously established using the ideal model described in Section 2.3.2.

### 2.3.4 Conclusion

The study presented in this Section has demonstrated that the ideal model and the Sauerbrey relation discussed in Sections 2.3.1 and 2.3.2 can be reliably used to monitor mass variations of a thin film deposited on the upper electrode of the QCM200 quartz crystal microbalance-provided that, first, the film thickness  $\ell_F$  remains small compared to the quartz plate thickness  $\ell_Q$ , and second, the film is homogeneous and uniformly thick. Under these conditions, the Sauerbrey constant used for the polymer films examined in the sections on water vapor sorption was set to  $56.51 \text{ Hz} \cdot \mu\text{g}^{-1} \cdot \text{cm}^2$ , as determined from the advanced modeling presented in Section 2.3.3.

## 3. Modeling sorption phenomena in water-soluble polymer films

This section first reviews the most basic modeling approaches (see Section 3.1) that can be used to elucidate the modes of water solvent exchange-denoted  $W$  in the film and  $w$  in the surrounding humid atmosphere between a solid polymer film ( $F$ ) and a humid environment (see **Figure 8**). The typical lateral dimensions  $L_F$  of the film (on



**Figure 8.**  
 Interaction between water vapor ( $w$ ) from humid air and a polymer film ( $F$ ) deposited on a fixed, impermeable planar substrate.

the order of centimeters) are significantly larger than its thickness  $\ell_F$  (on the order of micrometers), allowing for the reasonable assumption that mass transport is essentially one-dimensional. We therefore denote by  $c_W(y, t)$  and  $c_w(y, t)$  the mass or molar concentration of water in the film and in the atmosphere, respectively.

The basic models presented in Section 3.1 are then combined into more advanced approaches, as discussed in Section 3.2.

### 3.1 Basic modeling

The main mechanisms involved in water transfer that are examined are pure Fickian diffusion, viscoelastic relaxation, and sorption/desorption equilibrium (-Langmuir-type model).

#### 3.1.1 Pure Fickian diffusion

Let us consider a dry, planar polymer film (F) of constant thickness  $\ell_F$ , deposited on an impermeable flat substrate (see **Figure 8**). Denote by  $D_0$  the diffusion coefficient of water (W) through the film (F), and assume that Fick's first law holds in the form  $\mathbf{j}_W^d = -D_0 \nabla c_W$ , where  $c_W(y, t)$  is the mass concentration of water in the film. The mass balance in the film is then written in its simplest form (no chemical reaction):

$$\frac{\partial c_W}{\partial t} = D_0 \frac{\partial^2 c_W}{\partial y^2} \quad (29)$$

This partial differential equation (PDE) is complemented by the following boundary and initial conditions:

$$c_W(\ell_F, t) = c_W^{\text{eq}} ; \quad \partial c_W|_{y=0} = 0 ; \quad c_W(y, 0) = c_W^0 \quad (30)$$

where  $c_W^{\text{eq}}$  and  $c_W^0$  are assumed to be constant.

The partial differential Eq. (29) can be solved analytically using various approaches. In this document, the Green's function method was employed to express  $c_W(y, t)$  in the following form:

$$c_W(y, t) = \frac{4}{\pi} \sum_{n=0}^{\infty} \frac{(-1)^{n+1}}{2n+1} \cos\left(\beta_n \frac{y}{\ell_F}\right) \left[ (c_W^0 + c_W^{\text{eq}}) e^{-t\tau_{d,n}^{-1}} - c_W^{\text{eq}} \right], \quad \forall y \in [0, \ell_F] \quad (31)$$

where:

$$\beta_n = (2n+1) \frac{\pi}{2} \quad \text{and} \quad \tau_{d,n}^{-1} = \left(n + \frac{1}{2}\right)^2 \frac{\pi^2 D_0}{\ell_F^2} = \left(n + \frac{1}{2}\right)^2 \pi^2 \tau_d^{-1} \quad (32)$$

with  $\tau_d = \ell_F^2 / D_0$  a characteristic timescale of the diffusion phenomenon. It is worth noting that, in the limit as  $t \rightarrow \infty$ , the water concentration within the film becomes uniform and equal to  $c_W^{\text{eq}}$ . Quartz crystal microbalance (QCM) measurements do not provide direct access to the water concentration but rather to the water mass  $M_t$  present in the film at time  $t$ , or to its variations  $\Delta m_W(t) = M_t - M_t^0$ , with  $M_t^0 = m_W(0)$ .

The total water mass  $M_t$  is readily obtained by integrating the result (31) over the film thickness,  $y \in [0, \ell_F]$ :

$$\frac{M_t - M_t^0}{M_t^\infty - M_t^0} = 1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{e^{-t\tau_{d,n}^{-1}}}{(2n+1)^2} \quad (33)$$

where  $M_t^\infty = m_W(\infty)$ .

**Figure 9** compares the modeling results (solid red line) based on Eq. (33) with representative experimental data (dots) for the swelling of one of the polymer films investigated in this study. A significant mismatch is observed between the pure Fickian diffusion model and the experimental results, both when plotted as a function of time  $t$  (see **Figure 9a**) and as a function of  $\sqrt{t}$  (see **Figure 9b**). The pure diffusion model yielded a characteristic diffusion time  $\tau_d = 1773$  s with a coefficient of determination  $R^2 = 0.9652$  (best-fit value).

In conclusion, the significant discrepancies between the model and the experimental data highlight that the assumption of purely Fickian diffusion alone fails to adequately capture the sorption behavior of the polymer films investigated in this study.

### 3.1.2 Pure relaxation

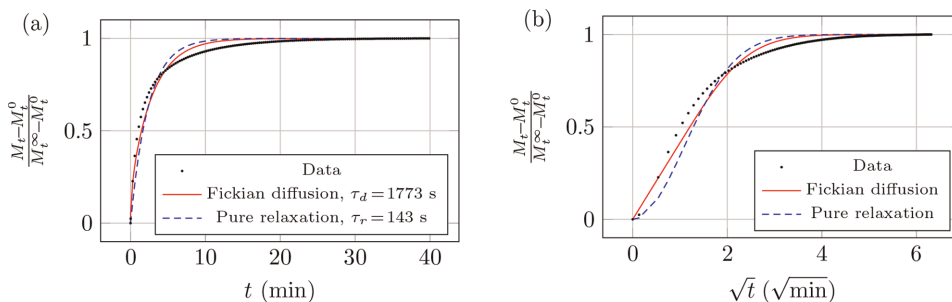
This extremely simple model assumes that the water concentration  $c_W$  is uniform throughout the film and that it follows, at each moment, an ordinary differential equation of relaxation of the following form:

$$\tau_r \dot{c}_W(t) = c_W^{\text{eq}} - c_W(t) \quad \text{when } t > 0, \quad \text{with } c_W(0) = c_W^0 \quad (34)$$

where “.” denotes the time derivative, and  $\tau_r$  is the characteristic time of the relaxation process under consideration. This first-order linear ordinary differential equation with constant coefficients has the following solution:

$$c_W(t) = c_W^0 e^{-t/\tau_r} + c_W^{\text{eq}} (1 - e^{-t/\tau_r}) \quad (35)$$

This yields the relative change in water mass resulting from the pure relaxation phenomenon, expressed as:



**Figure 9.** Typical evolutions of  $(M_t - M_t^0) / (M_t^\infty - M_t^0)$  based on data obtained during water vapor sorption experiment with one of the polymer films studied in this work. (a) Evolution as a function of time  $t$ . (b) Evolution as a function of  $\sqrt{t}$ .

$$\frac{M_t - M_t^0}{M_t^\infty - M_t^0} = 1 - e^{-t/\tau_r} \quad (36)$$

**Figure 9a** and **b** shows the fit (blue dashed line) of the experimental data (dots) using the result of Eq. (36). The pure relaxation model yields a relaxation time constant of  $\tau_r = 143$  s, with a coefficient of determination  $R^2 = 0.9341$  (best value obtained). Here again, significant discrepancies between the model and the experimental data highlight that the assumption of pure relaxation alone is not sufficient to explain the sorption behavior of the polymer films investigated in this study.

It is worth noting that at times short compared to  $\tau_r$ , a Taylor expansion of  $e^{-t/\tau_r}$  leads to a swelling kinetics of the form:

$$\frac{M_t - M_t^0}{M_t^\infty - M_t^0} \approx \frac{t}{\tau_r} \quad (37)$$

**Figure 9b** also reveals the characteristic S-shaped curve in the plot of  $(M_t - M_t^0)/(M_t^\infty - M_t^0)$  as a function of  $\sqrt{t}$ .

### 3.1.3 Sorption/desorption equilibrium (Langmuir model)

The Langmuir model has been widely used in its original form [17] and in various modified versions [18] to describe sorption phenomena in different solids. It provides a framework for describing the capture/release of water molecules (W) by trapping sites (T) within the solid film, through the following equilibrium:



whose kinetics is described by a first-order ordinary differential equation (ODE):

$$\dot{\theta}_W(t) = k_{\text{sor}} p_W^{\text{int}} (1 - \theta_W) - k_{\text{des}} \theta_W \quad (39)$$

where  $\theta_W(t) = c_W(t)/c_W^{\text{max}}$  is the fraction of water molecules trapped by the solid,  $c_W^{\text{max}}$  is the maximum trappable water concentration;  $p_W^{\text{int}}$  is the water vapor pressure at the interface between the film and the humid atmosphere;  $k_{\text{sor}}$  and  $k_{\text{des}}$  are the sorption and desorption rate constants, respectively. The ODE (39) can be rewritten in the following form:

$$\dot{\theta}_W(t) + \tau_L^{-1} \theta_W(t) = k_{\text{sor}} p_W^{\text{int}} \quad (40)$$

with  $\tau_L^{-1} = k_{\text{des}} + k_{\text{sor}} p_W^{\text{int}}$ , where  $\tau_L$  is a characteristic timescale for the sorption/desorption equilibrium. Denoting  $\theta_W^0 = \theta_W(0)$  as the initial fraction of water molecules trapped in the film, the solution  $\theta_W(t)$  of the ODE (39) is given by:

$$\frac{\theta_W(t) - \theta_W^0}{\theta_W^{\text{eq}} - \theta_W^0} = 1 - e^{-t/\tau_L} \quad (41)$$

with  $\theta_W^{\text{eq}} = \tau_L k_{\text{sor}} p_W^{\text{int}}$ . The relative variation in the mass of water trapped in the film then takes the same form as Eq. (36), where it is sufficient to replace  $\tau_r$  with  $\tau_L$ .

The conclusion of paragraph 3.1.2 also applies here: the Langmuir model used on its own does not adequately capture the sorption behavior of the polymer films studied in this work.

### 3.1.4 Linear and invariant interfacial transport

In the case of thin films with low mass Biot numbers  $Bi_m \ll 1$ , the water concentration  $c_W$  inside the film (F) can be very accurately approximated as uniform. In this regime, water transport within the film is no longer controlled by diffusion, but rather by the interfacial flux  $\Phi_w$  of water vapor (w) at the interface  $y = \ell_f$  between the film and the surrounding humid atmosphere. Within the framework of the elementary linear and invariant approach presented in this paragraph, the interfacial flux is described by a Newton-type law:

$$\Phi_w(t) = \Sigma h_W [c_W(t) - c_W^{\text{eq}}] \quad (42)$$

where  $\Sigma$  is the area of the interface between the film and the surrounding atmosphere,  $h_W$  is a constant (mass or molar) transfer coefficient, and  $c_W^{\text{eq}}$  has the same meaning as in the previous models. Since the water activity in the film depends on the water concentration  $c_W(t)$ , the Newton-type law is, strictly speaking, only valid in the linear regime, where  $c_W(t)$  and  $c_W^{\text{eq}}$  differ by only a few percent—an assumption that unfortunately rarely holds. The more general nonlinear case is also discussed in paragraph 3.2.2.

A mass balance of the water contained at time  $t$  in the film of constant thickness  $\ell_F$  leads to the following ordinary differential equation satisfied by the concentration  $c_W(t)$ :

$$\dot{c}_W(t) = - \frac{h_W}{\ell_F} [c_W(t) - c_W^{\text{eq}}] \quad (43)$$

The ordinary differential Eq. (43) has the same form as the pure relaxation Eq. (34), with a characteristic interfacial transport time  $\tau_{\text{int}}$  given here by  $\tau_{\text{int}} = \ell_F/h_W$ . The relative variation of the water mass,  $(M_t - M_t^0)/(M_t^\infty - M_t^0)$ , resulting from linear and invariant interfacial transport is therefore also given by Eq. (36), with  $\tau_r$  replaced by  $\tau_{\text{int}}$ .

The conclusion of paragraph 3.1.2 also applies here: the linear and invariant interfacial transport model, when used on its own, does not adequately capture the swelling behavior of the polymer films studied in this work.

### 3.1.5 Conclusion and discussion

When subjected to sorption phenomena, polymers can exhibit a wide range of physical behaviors. Here, we propose a non-exhaustive classification of the underlying mechanisms, aiming to organize this diversity and highlight the main characteristic regimes.

According to Masaro and Zhu [19], solvent diffusion through a solid polymer can exhibit a broad spectrum of behaviors, ranging from Fickian diffusion (Case I) to various non-Fickian regimes (Case II, anomalous diffusion, and Super Case II). Solvent transport through the polymer matrix is generally accompanied by deformation of the matrix and thus by mechanical relaxation processes, which may be more or less localized in space or time and vary in intensity depending on whether the polymer is in a glassy or rubbery state.

To classify the behavior of the solid polymer during swelling, one may use the dimensionless Deborah number  $De = \tau_r / \tau_d$ , where  $\tau_r$  and  $\tau_d$  are the characteristic timescales for relaxation of the polymer matrix and for solvent diffusion through the polymer, respectively. At short times, the relative solvent mass  $(M_t - M_t^0) / (M_t^\infty - M_t^0)$  absorbed by the polymer typically scales as  $t^n$ , where  $n$  is the characteristic exponent of the solvent transport kinetics through the polymer.

### 3.1.5.1 Fickian diffusion (or case I)

It occurs in polymers in the rubbery state, that is, at temperatures above the glass transition temperature ( $T > T_g$ ). In the rubbery state, polymer chains are sufficiently mobile to reorganize easily and rapidly as the solvent penetrates. Relaxation times are short in this case compared to the diffusion timescale:  $\tau_r \ll \tau_d$ , and  $De \ll 1$ . In this regime, solvent transport is primarily governed by diffusion, with a constant diffusion coefficient  $D_0$ , characterizing the so-called linear case. The relative mass of solvent absorbed by the solid polymer at short times scales as:

$$\frac{M_t - M_t^0}{M_t^\infty - M_t^0} \propto t^{1/2} \quad (44)$$

This  $n = 1/2$  behavior is clearly observed in **Figure 9b** for the solid red curve (corresponding to the purely Fickian model), which appears linear at short times when plotted as a function of  $\sqrt{t}$ .

### 3.1.5.2 Case II

This corresponds to a case of non-Fickian diffusion, where transport no longer follows Fick's law. It is typically encountered in glassy polymers ( $T < T_g$ ), for which the solvent penetrates with difficulty and induces only limited deformation of the polymer matrix. In this situation, the characteristic relaxation times of the matrix become much longer than the diffusion timescale:  $\tau_r \gg \tau_d$ , and  $De \gg 1$ . Solvent transport is then strongly decoupled from the mechanical relaxation of the polymer matrix; these two processes do not occur simultaneously at the same location.

It is often observed in this regime that a sharp solvent penetration front forms, behind which the polymer undergoes substantial swelling, accompanied by localized mechanical relaxation in the swollen zone. In this case, the relative mass of solvent absorbed by the solid polymer at short times typically scales as:

$$\frac{M_t - M_t^0}{M_t^\infty - M_t^0} \propto t \quad (45)$$

This  $n = 1$  kinetics is also obtained in the case of pure relaxation discussed in paragraph 3.1.2, Eq. (37). This correspondence highlights the key role of relaxation in Case II, which can no longer be described as purely Fickian.

### 3.1.5.3 Anomalous case

This situation arises when the characteristic timescales of diffusion  $\tau_d$  and relaxation  $\tau_r$  are comparable:  $\tau_r \approx \tau_d$ , or equivalently  $De \approx 1$ . In this case, there is no clear

separation between diffusion and relaxation phenomena, which occur simultaneously at the same location. This represents an intermediate regime between Case I and Case II, in which the short-time solvent uptake kinetics follows:

$$\frac{M_t - M_t^0}{M_t^\infty - M_t^0} \propto t^n, \quad \text{with} \quad \frac{1}{2} < n < 1 \quad (46)$$

#### 3.1.5.4 Super case II

This situation also corresponds to  $\tau_r \gg \tau_d$  and  $De \gg 1$ , but in this case the swelling kinetics is faster than in Case II, as it follows  $n > 1$ :

$$\frac{M_t - M_t^0}{M_t^\infty - M_t^0} \propto t^n, \quad \text{with} \quad n > 1 \quad (47)$$

#### 3.1.5.5 Interfacial transport

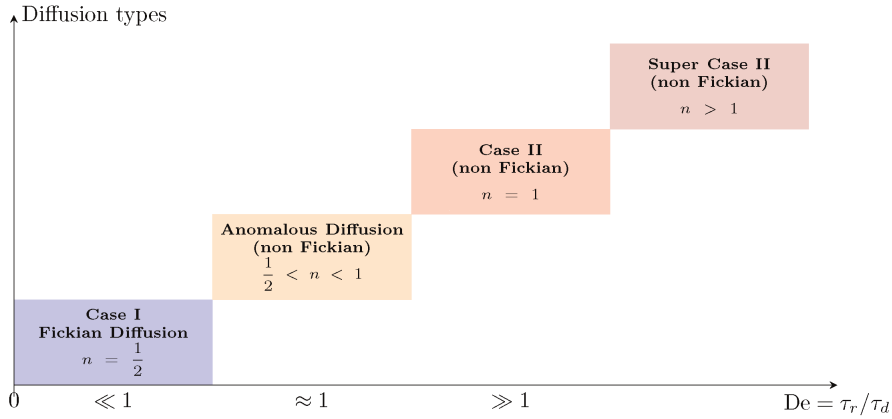
When the polymer takes the form of a thin solid film of thickness  $\ell_F$ , solvent transport through the material may also be governed by interfacial transfer (rather than diffusion) and mechanical relaxations. Under these conditions, the results presented in paragraph 3.1.2 show that the swelling kinetics also follows a first-order behavior with exponent  $n = 1$ :

$$\frac{M_t - M_t^0}{M_t^\infty - M_t^0} \propto t \quad (48)$$

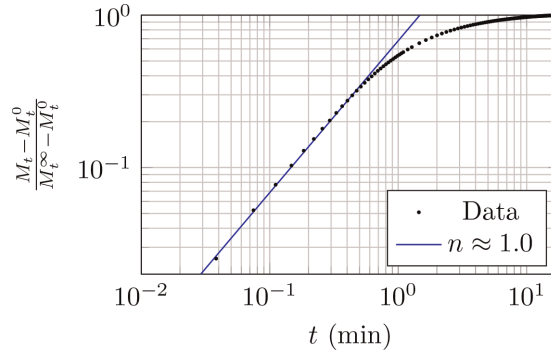
Let  $h_W$  be the interfacial mass transfer coefficient for the solvent. The mass Biot number, defined as  $Bi_m = h_W \ell_F / D_0$ , compares the resistance to solvent diffusion within the film to the resistance to solvent transport at the interface between the film and the surrounding atmosphere. When  $Bi_m \ll 1$ , the solvent distribution inside the film remains approximately uniform. In this case, the product  $Bi_m De = h_W \tau_r / \ell_F = \tau_r / \tau_{\text{int}}$  helps identify whether interfacial transport or polymer relaxation is the dominant phenomenon [20]:

- If  $Bi_m De \ll 1$ , then  $\tau_r \ll \tau_{\text{int}}$ , meaning that the polymer relaxes much faster than the interfacial solvent flux. In this case, interfacial transport is the limiting phenomenon, and the sorption process is controlled by interfacial transport.
- If  $Bi_m De \gg 1$ , then  $\tau_r \gg \tau_{\text{int}}$ , meaning that the polymer relaxes much more slowly than the interfacial solvent flux. In this case, interfacial transport is no longer the limiting phenomenon, and the swelling process is controlled by the mechanical relaxations of the polymer matrix.
- If  $Bi_m De \approx 1$ , then  $\tau_r \approx \tau_{\text{int}}$ , and the mechanical relaxations of the polymer and the interfacial transport occur on comparable timescales, leading to a strong coupling between mechanical and transport phenomena.

**Figure 10** provides a schematic summary outlining the classification of the different types of diffusion phenomena that may arise during solvent sorption in solid polymers. It should be noted that, in practice, the boundaries between these modes are rarely as clear-cut as suggested by this schematic overview.



**Figure 10.**  
Typical solvent transport modes that may occur during the sorption process in a solid polymer.



**Figure 11.**  
Log-log representation of the typical evolution of  $(M_t - M_t^0) / (M_t^\infty - M_t^0)$ , based on experimental measurements (data) obtained during the solvent sorption process of one of the polymer films studied in this work.

The graphical representation of experimental data on log-log scales at short times can help determine the kinetic order of sorption and thus provide valuable insight into the mechanisms governing solvent transport through the film. **Figure 11** shows the relative variations in the water mass sorbed by the film, plotted on logarithmic scales.

It can be observed in **Figure 11** that the sorption kinetics follows an order of approximately  $n \approx 1$  at short times, which corresponds to a regime at the boundary between anomalous diffusion and Case II, both of which are non-Fickian. Fickian diffusion alone is therefore insufficient to describe solvent transport within the film, and it is necessary in this case to also account for the relaxation of the polymer matrix.

### 3.2 Couplings

The discussions in paragraph 3.1 have shown that the swelling behavior of the polymer films considered in this study cannot be explained by a single physical mechanism. We now consider the combination of some of the elementary mechanisms previously introduced.

Only the approaches that successfully reproduced the experimental sorption data are presented and discussed in this document.

### 3.2.1 Diffusion/relaxation modeling

To explain the sorption curves obtained experimentally with glassy polymer powders, Berens and Hopfenberg proposed a phenomenological model of non-Fickian diffusion, involving two processes: the relaxation of the polymer matrix and the Fickian diffusion of the solvent through that matrix [21]. They suggested describing the complex sorption behavior in such polymers by expressing the water mass  $M_t$  absorbed by the solid as follows:

$$M_t = M_t^F + M_t^R \quad (49)$$

This formulation independently accounts for both Fickian diffusion, through the term  $M_t^F$ , and relaxation phenomena, through the term  $M_t^R$ . Using results (33) and (36),  $M_t$  can be expressed in the following form:

$$\frac{M_t - M_t^0}{M_t^\infty} = \frac{M_t^{F,\infty} - M_t^{F,0}}{M_t^\infty} \left[ 1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{e^{-t\tau_{d,n}^{-1}}}{(2n+1)^2} \right] + \frac{M_t^{R,\infty} - M_t^{R,0}}{M_t^\infty} (1 - e^{-t/\tau_r}) \quad (50)$$

where  $M_t^0 = M_t^{F,0} + M_t^{R,0}$  is the initial amount of water present in the film. It is customary to denote  $\phi = (M_t^{F,\infty} - M_t^{F,0})/M_t^\infty$  as the fraction of solvent taken up by the film via diffusion between the initial time  $t = 0$  and equilibrium. With this notation, the fraction of water sorbed by the film at time  $t$  can be written as:

$$\frac{M_t - M_t^0}{M_t^\infty} = \phi \left[ 1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{e^{-t\tau_{d,n}^{-1}}}{(2n+1)^2} \right] + \left( 1 - \phi - \frac{M_t^0}{M_t^\infty} \right) (1 - e^{-t/\tau_r}) \quad (51)$$

This relation will be used in paragraph 4 to interpret the swelling measurements of the polymer films studied in this work.

### 3.2.2 Discussion

- Treating diffusion and relaxation phenomena as independent is generally an approximation, which makes the Berens-Hopfenberg model a phenomenological approach. However, this approach can be considered exact when the two phenomena are decoupled, that is, when the characteristic times  $\tau_d$  and  $\tau_r$  differ significantly. According to Sun, only the case where  $De \gg 1$  allows for a rigorous decoupling of the two phenomena [22].
- The experimental situation is generally considered where the film is initially almost dry, that is, such that  $M_t^0 \ll M_t^\infty$ .
- From Eq. (32), one can write  $\tau_{d,n}^{-1} = (n + \frac{1}{2})^2 \pi^2 \tau_d^{-1}$ , with  $\tau_d = \ell_F^2/D_0$ . By introducing the dimensionless time  $t_r = t/\tau_d$ , relation (51) can be further rewritten as:

$$\frac{M_{t_r} - M_t^0}{M_t^\infty} = \phi \left[ 1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{e^{-(n+\frac{1}{2})^2 \pi^2 t_r}}{(2n+1)^2} \right] + \left( 1 - \phi - \frac{M_t^0}{M_t^\infty} \right) (1 - e^{-\psi t_r}) \quad (52)$$

where  $\psi = \tau_d/\tau_r = 1/De$ .

### 3.2.3 Interfacial transport/relaxation modeling

In certain situations—particularly with thin solid films—diffusion may no longer be the dominant mode of solvent transport. Instead, interfacial transport may control the overall flux of solvent through the film during sorption. This paragraph therefore introduces a simple model that accounts for both interfacial transport and mechanical relaxation in describing the sorption behavior of a polymer film.

#### 3.2.3.1 Presentation—Assumptions

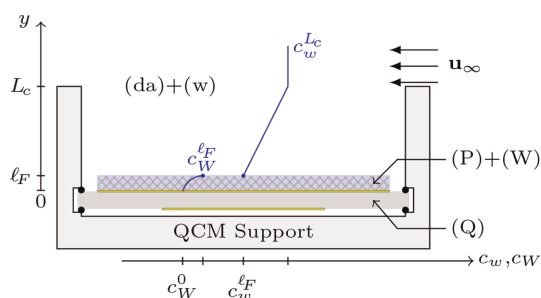
**Figure 12** schematically illustrates the situation considered in this study. A thin film composed of a mixture of water-soluble polymers is deposited on the gold electrode of a quartz crystal (Q) used in a QCM200 quartz crystal microbalance. The initially dry film ( $M_t^0 \ll M_t^\infty$ ) is exposed to a humid atmosphere (da) + (w), where (w) denotes water vapor and (da) dry air, both treated as ideal gases. The notation (W) refers to the water present inside the film, and it is assumed that dry air (da) does not penetrate the film. Given the very small film thickness  $\ell_F$  (on the order of micrometers) compared to the diameter  $d = 2b = 1.3$  cm of the electrode on which it is deposited, water exchange processes are assumed to be one-dimensional. The mass concentrations of water in the film and in the air are therefore denoted  $c_w(y, t)$  and  $c_w(y, t)$ , respectively.

Variations in the film thickness  $\ell_F$  during the sorption process are usually neglected.

Because the microbalance support and the measurement rod prevent the development of a boundary layer near the film, it can be shown [23] that the water vapor profile  $c_w(y, t)$  in the humid atmosphere above the film is, to a very good approximation, linear and takes the following form:

$$c_w(y, t) = \frac{c_w^{L_c} - c_w^{\ell_F}(t)}{L_c - \ell_F} (y - \ell_F) + c_w^{\ell_F}(t) \quad (53)$$

where  $c_w^{\ell_F}(t)$  and  $c_w^{L_c} = \text{const}$  are the water vapor concentrations at the interface between the film and the humid atmosphere and far from the film, respectively.



**Figure 12.**

Polymer film (F) = (P) + (W), of thickness  $\ell_F \ll L_c$ , deposited on a quartz crystal (Q) of a QCM200 microbalance, positioned at the end of the Teflon measurement rod (see also **Figure 7**). The film undergoes isothermal and isobaric sorption of water. For clarity of illustration, the spatial and concentration scales for water,  $c_w$  and  $c_W$  (blue line), are not represented to scale.

Under these conditions, the water mass flux  $\Phi_w$  between the film and the atmosphere can be written in the following form:

$$\Phi_w(t) = \Sigma h_w(\mathbf{u}_\infty) [c_w^{\ell_F}(t) - c_w^{L_c}(t)] \quad (54)$$

with:  $\Phi_w < 0$  corresponding to water uptake (and vice versa);  $\Sigma = \pi b^2$ ;  $h_w(\mathbf{u}_\infty)$  the interfacial mass transfer coefficient (in m/s); and  $\mathbf{u}_\infty$  is the flow velocity of humid air far from the film (F). As water is exchanged between the film and the humid atmosphere, the vapor concentration  $c_w^{\ell_F}(t) = c_w(\ell_F, t)$  in the atmosphere and the water concentration  $c_W^{\ell_F}(t) = c_W(\ell_F, t)$  in the film evolve over time, until they reach their equilibrium values, at which point the sorption process comes to an end.

A direct relationship exists between the water concentration  $c_w^{\ell_F}$  in the humid air and  $c_W^{\ell_F}$  in the polymer film, at the interface  $y = \ell_F$ . This relationship is governed by the water chemical activity  $a_W$  or the water fugacity  $f_W$  in the film (F). Assuming that local thermodynamic equilibrium is maintained at all times at the interface  $y = \ell_F$  between the film and the humid atmosphere, one can write the equality of chemical fugacities  $f_w(T, p) = f_W(T, p)$ . Since water vapor (w) is treated as an ideal gas, its fugacity in the humid air is given by  $f_w(T, p) = p_w = c_w r_w T$ , where  $r_w = R/M_w$ ,  $R$  is the universal gas constant, and  $M_w = 18$  g/mol is the molar mass of water. On the other hand, the fugacity of water (W) in the solid polymer film can be written as  $f_W(T, p) = a_W(c_W, T, p)p_w^{\text{sat}}(T)$ , where  $a_W$  is the chemical activity of water in the film, and  $p_w^{\text{sat}}(T)$  is the saturation vapor pressure of water at the film absolute temperature  $T$ . The local thermodynamic equilibrium at the interface  $y = \ell_F$  therefore results in the following relation:

$$c_w^{\ell_F}(t) = \frac{M_w}{RT} a_W(c_W^{\ell_F}(t), T, p) p_w^{\text{sat}}(T) \quad (55)$$

The mass flux  $\Phi_w$  then becomes, at constant  $T$  and  $p$  (which will no longer be explicitly mentioned hereafter):

$$\Phi_w(t) = \Sigma h_w \frac{M_w}{RT} p_w^{\text{sat}}(T) [a_W(c_W^{\ell_F}(t)) - a_W^\infty(t)] \quad (56)$$

To fully express the mass flux  $\Phi_w$ , it is necessary to specify the chemical activity of water  $a_W^{\ell_F}(t) \equiv a_W(c_W^{\ell_F}(t))$  in the polymer film as a function of the mass concentration  $c_W^{\ell_F}$  at the interface between the film and the humid atmosphere. Several approaches can be considered.

### 3.2.3.2 Use of a model for $a_W$

Several models exist to determine the chemical activity of water  $a_W$  in a polymer or polymer blend, generally expressed as a function of temperature  $T$  and water content  $\omega = m_W/m_P$ , where  $m_P$  is the mass of dry polymer. Among these models, only the most common ones are recalled here.

The BET model (Brunauer, Emmett, and Teller [24]) is one of the earliest models proposed to describe gas adsorption on solid surfaces. This model has been applied to numerous polymers of different types, including biopolymers, food polymers, and other hygroscopic solid materials [25, 26]. The GAB model (Guggenheim, Anderson,

and de Boer) covers a generally wider range of chemical activities  $a_W$  than the BET model [27]. In this case, the chemical activity is implicitly expressed as a function of water content  $\omega$  in the following form:

$$\omega = \frac{\omega_m CKa_W}{(1 - Ka_W)(1 - Ka_W + CKa_W)} \quad (57)$$

where  $\omega_m$ ,  $C$ , and  $K$  are temperature-dependent parameters (following an Arrhenius-type law), whose values are obtained for a given solid by fitting the model to experimental data [28]. Recalling that the mass concentration  $c_W$  is related to the water content  $\omega$  by the relation  $c_W = \omega/(\omega/\rho_W + 1/\rho_S)$ , where  $\rho_W$  and  $\rho_S$  are the densities of pure water and dry solid, respectively, and the chemical activity  $a_W$  can consequently also be expressed as a function of the mass concentration  $c_W$ .

### 3.2.3.3 Linear approximation

This approximation is strictly valid when the difference between  $a_W^{\ell_F}(t)$  and  $a_W^\infty(t)$  remains small during the sorption process. In this case, it can be well approximated that  $a_W(c_W^{\ell_F}(t)) - a_W^\infty(t) \propto c_W^{\ell_F}(t) - c_W^\infty(t)$ . The mass flux  $\Phi_w$  can then be expressed in the following form:

$$\Phi_w(t) = \Sigma h_W [c_W^{\ell_F}(t) - c_W^\infty(t)] \quad (58)$$

where  $c_W^\infty$  is the mass concentration of water in the film when it is in equilibrium with the surrounding humid atmosphere, and  $h_W$  is a mass transfer coefficient that can be expressed as a function of  $h_w$ . This approach was implemented by Satterfield et al. to describe the water sorption dynamics of Nafion membranes [20].

In the absence of reliable experimental data available in the literature for the parameters  $\omega_m$ ,  $C$ , and  $K$  for the polymer blends considered, the linear approximation is used in the present study.

### 3.2.3.4 Mathematical model

In the present case of a polymer film of thickness  $\ell_F$  deposited on a fixed, impermeable flat substrate at  $y = 0$ , the linear approximation equations governing the transport of water molecules within the polymer film are as follows, where  $y = \ell_F$  denotes the film/humid atmosphere interface:

$$\frac{\partial}{\partial t}(\delta c_W) = D_0 \frac{\partial^2}{\partial y^2}(\delta c_W) ; \quad \frac{\partial}{\partial y}(\delta c_W) \Big|_{y=0} = 0 ; \quad \frac{\partial}{\partial y}(\delta c_W) \Big|_{y=\ell_F} = -h_W(\delta c_W \Big|_{y=\ell_F} - \delta c_W^\infty) \quad (59)$$

where  $\delta c_W = c_W - c_W^0$ , with  $c_W^0$  the initial mass concentration of water in the film;  $h_W$  the interfacial mass transfer coefficient (in m/s); and  $\delta c_W^\infty = c_W^\infty - c_W^0 \neq 0$ , where  $c_W^\infty$  is the water concentration in the film when it has reached equilibrium with the surrounding humid atmosphere.

The Eq. (59) can be nondimensionalized using the following variable changes:

$$Y = \frac{y}{\ell_F} \in [0, 1]; \quad \tau_d = \frac{\ell_F^2}{D_0}; \quad t_r = \frac{t}{\tau_d}; \quad \text{and} \quad \text{Bi}_m = \frac{h_W \ell_F}{D_0} \quad (60)$$

Eq. (59) is solved in Appendix A for any value of  $\delta c_W^\infty$  (constant or time-dependent) using the Laplace transform. The resulting expression for the water mass  $\delta m_W(t_r)$  absorbed via interfacial transport is:

$$\delta m_W(t_r) = \ell_F \Sigma \sum_{n=1}^{\infty} \frac{2\text{Bi}_m^2}{\text{Bi}_m(\text{Bi}_m + 1) + \beta_n^2} \int_0^{t_r} \delta c_W^\infty(\tau) e^{-\beta_n^2(t_r - \tau)} d\tau \quad (61)$$

where it is recalled that  $\beta_n$  is the nonzero solution of the transcendental equation  $\beta_n \tan \beta_n = \text{Bi}_m$ . Experimentally, we imposed abrupt humidity changes on the polymer film (F), which corresponds to subjecting (F) to a humidity step

$$\delta c_W^\infty(\tau) = (c_W^\infty - c_W^0) \Theta(\tau) \quad (62)$$

where  $\Theta(\tau)$  is the Heaviside step function. In this case, result (61) leads to the following expression for  $\delta m_W(t_r)$ :

$$\delta m_W(t_r) = \ell_F \Sigma (c_W^\infty - c_W^0) \sum_{n=1}^{\infty} \frac{2\text{Bi}_m^2}{\beta_n^2 [\text{Bi}_m(\text{Bi}_m + 1) + \beta_n^2]} (1 - e^{-\beta_n^2 t_r}) \quad (63)$$

The mass variation  $\delta m_W(t_r)$  approaching at equilibrium  $\delta m_W^\infty = \ell_F \Sigma (c_W^\infty - c_W^0)$ ; therefore, we have in the present case:

$$\frac{\delta m_W(t_r)}{\delta m_W^\infty} = \sum_{n=1}^{\infty} \frac{2\text{Bi}_m^2}{\beta_n^2 [\text{Bi}_m(\text{Bi}_m + 1) + \beta_n^2]} (1 - e^{-\beta_n^2 t_r}) \quad (64)$$

### 3.2.3.5 Coupling between interfacial transport and relaxation

As in the case of paragraph 3.2.1, we now write  $M_t = M_t^{\text{TI}} + M_t^R$  to account for the two modes of solvent transport through the polymer film, with TI standing for interfacial transport and R for relaxation. According to results (36) and (64), we obtain, using dimensionless time  $t_r = t/\tau_d$ :

$$\frac{M_{t_r} - M_t^0}{M_t^\infty} = \phi \sum_{n=1}^{\infty} \frac{2\text{Bi}_m^2}{\beta_n^2 [\text{Bi}_m(\text{Bi}_m + 1) + \beta_n^2]} (1 - e^{-\beta_n^2 t_r}) + \left(1 - \phi - \frac{M_t^0}{M_t^\infty}\right) (1 - e^{-\psi t_r}) \quad (65)$$

where  $\psi = \tau_d/\tau_r$ ,  $M_t^0 = M_t^{\text{TI},0} + M_t^{R,0}$ ,  $M_t^\infty = M_t^{\text{TI},\infty} + M_t^{R,\infty}$ , and  $\phi = (M_t^{\text{TI},\infty} - M_t^{\text{TI},0})/M_t^\infty$  is the fraction of solvent sorbed by the film via interfacial transport between the initial time  $t = 0$  and equilibrium.

### 3.2.4 Conclusion

Expressions (51) and (65) were successively used to fit the experimental results obtained with a QCM200 quartz crystal microbalance during the sorption study of various initially dry polymer films. The experimental results, as well as those obtained from the two modeling approaches (diffusion/relaxation and interfacial transport/relaxation) introduced in this paragraph, are discussed in the following section.

## 4. Experimental results and discussion

### 4.1 Methods and materials

The films studied were prepared from powders containing varying proportions of three biocompatible polymers:

- *Sodium alginate (ANa, hereafter referred to as A)*: commonly used as a binder and viscosity control agent in cosmetic formulations;
- *Sodium carboxymethyl cellulose (CMC, hereafter C)*: commonly employed as an emulsion stabilizer, film-forming agent, and thickener;
- *Polyvinyl alcohol (PVA, hereafter P)*: commonly used as a film-forming agent and viscosity regulator.

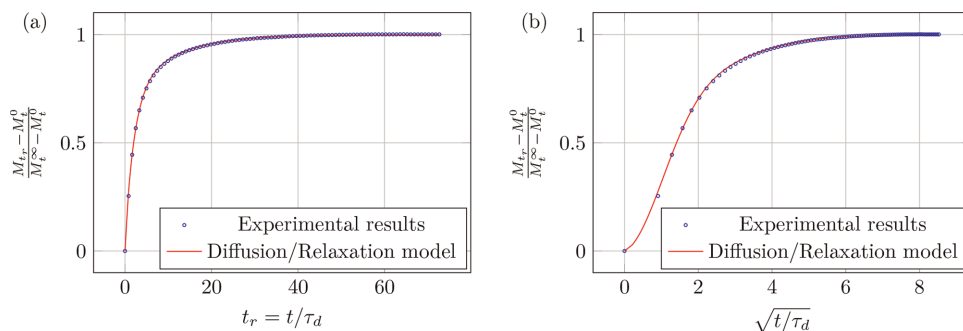
Three polymer powder blends  $P_xA_yC_z$  were studied, where  $x, y$ , and  $z = 1 - x - y$  represent the mass fractions of polyvinyl alcohol, sodium alginate, and sodium carboxymethyl cellulose, respectively. The polymer properties, preparation methods of the formulations, film casting, and drying procedures are detailed in [29] and will not be reiterated here. The masses of water absorbed and released were measured using the QCM200 quartz crystal microbalance described and modeled in Section 2.

### 4.2 Results and discussion

**Figure 13** presents the experimental results (circles) obtained during the sorption of an initially dry  $P_{3/4}A_{1/8}C_{1/8}$  when exposed at  $t = 0$  to a humid atmosphere at temperature  $T = 298$  K and controlled relative humidity  $RH = 95\%$ . The solid curve represents the fit of the diffusion/relaxation model (51) to the experimental data.

The curves in **Figure 13** demonstrate excellent agreement between the model and the experimental results, as confirmed by the coefficient of determination  $R^2 = 0.9992$ , which is very close to 1 (see **Table 2**).

The graphical representation in **Figure 13b** confirms that diffusion alone is insufficient to describe the water uptake of the film, as  $(M_{tr} - M_t^0) / (M_t^\infty - M_t^0)$  is not proportional to  $\sqrt{t_r}$  at the onset of sorption (for  $t_r \rightarrow 0$ ). In the present case, the



**Figure 13.** Experimental results and diffusion/relaxation modeling for the sorption of the dry  $P_{3/4}A_{1/8}C_{1/8}$  film. Evolution of water uptake  $(M_{tr} - M_t^0) / (M_t^\infty - M_t^0)$  as a function of: (a) dimensionless time  $t_r = t / \tau_d$ ; (b)  $\sqrt{t} / \sqrt{\tau_d}$ .

| Composition                                        | $\rho$ | $\frac{M_t^\infty - M_t^0}{m_F^0}$ | $D_0$ (m <sup>2</sup> /s) | $Bi_m \times De$ | $\phi_{D/R}$ | $\phi_{T/R}$ | $\tau_d$ (s) | $\tau_r$ (s) | $R_{T/R}^2$ | $R_{D/R}^2$ |
|----------------------------------------------------|--------|------------------------------------|---------------------------|------------------|--------------|--------------|--------------|--------------|-------------|-------------|
| P <sub>3/4</sub> A <sub>1/8</sub> C <sub>1/8</sub> | 1028   | 404                                | $6.69 \cdot 10^{-14}$     | 0.0083           | 0.343        | 0.312        | 26.44        | 2.17         | 0.9998      | 0.9992      |
| P <sub>1/8</sub> A <sub>3/4</sub> C <sub>1/8</sub> | 1025   | 407                                | $5.34 \cdot 10^{-14}$     | 0.0062           | 0.525        | 0.457        | 33.14        | 2.47         | 0.9970      | 0.9966      |
| P <sub>1/8</sub> A <sub>1/8</sub> C <sub>3/4</sub> | 992    | 522                                | $1.09 \cdot 10^{-13}$     | 0.0703           | 0.597        | 0.462        | 16.16        | 29.51        | 0.9993      | 0.9973      |

**Table 2.**

Compositions of the three films studied, along with the parameter values of the diffusion-relaxation and interfacial transport-relaxation models for water uptake performed under the same conditions: initial thickness

$\ell_F^0 = 1.33$   $\mu$ m, surface area  $\Sigma = 1.327$  cm<sup>2</sup>, temperature  $T = 298$  K, and relative humidity RH = 95%. The dry film mass is  $m_F^0 = \rho \ell_F^0 \Sigma$ . Densities  $\rho$  are given in kg/m<sup>3</sup>, and water uptake values  $(M_t^\infty - M_t^0)/m_F^0$  are given in mg/g.

kinetic exponent  $n$  of the sorption process is approximately 1, which is also observed for the two other polymer blends studied.

For the polymer blend P<sub>3/4</sub>A<sub>1/8</sub>C<sub>1/8</sub>, the low value of the product  $Bi_m \times De = 0.0083$  (see **Table 2**) indicates that relaxation phenomena (mechanical relaxation and interfacial transport) predominate over diffusion during the water uptake process by the dry film [30]. It is also noted that the water mass uptake rate  $(M_t^\infty - M_t^0)/m_F^0 = 404$  mg/g (with  $m_F^0$  the dry film mass) is the lowest among the three blends studied here, which can be partially explained by the limited contribution of relaxation to water uptake for the P<sub>3/4</sub>A<sub>1/8</sub>C<sub>1/8</sub> blend. These analyses apply similarly to the blend P<sub>1/8</sub>A<sub>3/4</sub>C<sub>1/8</sub> for which the proportions of PVA and ANa are reversed, while the CMC proportion remains constant. As shown in **Table 2**, the film containing the highest proportion of CMC (P<sub>1/8</sub>A<sub>1/8</sub>C<sub>3/4</sub>) is the one that absorbs the most water during sorption:  $(M_t^\infty - M_t^0)/m_F^0 = 522$  mg/g, all other factors being equal. These observations highlight the fundamental role of CMC as a facilitator of water uptake for the three polymer blends considered in this study.

The results of fitting models (51) and (65) to the sorption measurements of the three dry films exposed to water vapor are also summarized in **Table 2** (the series were truncated at  $N = 20$  terms for each model). The combined models of interfacial transport/relaxation and diffusion/relaxation are denoted as  $T/R$  and  $D/R$ , respectively. For each film studied, it is observed that the coefficients of determination satisfy the inequality  $R_{T/R}^2 > R_{D/R}^2$ ; thus, the interfacial transport/relaxation model yielded the best results, which is reasonable given the thinness of the films examined. Moreover, the low values of the products  $Bi_m \times De$  also reflect the predominance of interfacial transport over diffusion in the solvent transfer through the film (see discussion in Section 3.1.5). It is also worth noting that the low diffusion coefficients  $D_0$  indicate the glassy state of the dry films.

It is also observed in **Table 2** that the two blends with low CMC content correspond to the lowest values of the diffusion coefficient  $D_0$ . From these it can be inferred that, in the present case, the proportions of PVA and ANa polymers have little influence on both the diffusion coefficient of water within the solid matrix and the uptake rate of water by the dry film.

The results gathered in **Table 2** also reflect the influence of CMC content on both the water mass uptake rates, which increase with higher CMC proportions, and the coupling between the solid matrix relaxation and water diffusion. Indeed, it is observed that the characteristic diffusion time  $\tau_d$  decreases noticeably for the highest

| Formulation                                | Water content (mg/g) | References              |
|--------------------------------------------|----------------------|-------------------------|
| CMC/PVP/Acrylamide                         | 4000                 | Firoz et al. [31]       |
| CMC/MCC + 5% citric acid                   | 3227                 | Susi et al. [32]        |
| CMC/PVA crosslinked +6% citric acid        | 725                  | Pratinthong et al. [33] |
| CMC/5% ammonium carbonate + lyophilization | 610                  | Lavlinskaya et al. [34] |

**Table 3.**

*Water uptake capacities of films and gels based on CMC, according to various recent formulations. PVP: polyvinylpyrrolidone; MCC: microcrystalline cellulose.*

CMC content, while simultaneously the characteristic relaxation time  $\tau_r$  increases significantly. Thus, increasing the CMC fraction in the blend promotes a slowdown of mechanical relaxation processes, which appears to favor water uptake by the dry films, as evidenced by the significantly increased value of  $(M_t^\infty - M_t^0)/m_F^0$  with higher CMC mass fraction.

The tendency of the studied films to absorb increasingly more water with higher CMC mass fraction is linked to the intrinsic property of sodium carboxymethyl cellulose to capture water vapor. As a highly hydrophilic polysaccharide, CMC is widely used in polymer formulations for its excellent water retention capabilities. These strong water uptake properties have also been observed in other CMC-containing systems, as evidenced by the studies compiled in **Table 3**.

## 5. Conclusion

This work demonstrated the value of an integrated approach combining rigorous instrumental modeling, multiscale modeling of water/film transport, and experimental validation using a quartz crystal microbalance to unravel the complex sorption mechanisms in water-soluble polymer films. The advanced use of the QCM200, coupled with detailed modeling of edge effects (energy trapping) and sorption dynamics, enabled surpassing the limitations of classical approaches (pure Fickian diffusion, isolated relaxation), revealing coupled regimes such as diffusion-relaxation and interfacial transport-relaxation.

The results obtained demonstrate that the sorption kinetics observed in the studied films fall within a strongly coupled non-Fickian regime, characterized by a kinetic exponent close to 1 at short times, reflecting the significance of mechanical relaxation and interfacial uptake phenomena. The numerically fitted Sauerbrey constant, the spatial distribution of mass sensitivity, and Berens-Hopfenberg-type models enabled faithful reconstruction of the experimental data while providing physically interpretable physicochemical parameters.

In particular, the comparative analysis of films formulated from different blends of alginate, polyvinyl alcohol (PVA), and carboxymethyl cellulose (CMC) highlighted the central role of the latter in water uptake, notably through its ability to promote the formation of highly hydrophilic structures and stabilize the sorbed states. The presence of CMC in these water-soluble polymer blends significantly contributes to the increase in water mass sorbed, thereby confirming its structuring effect within the polymer network and its capacity to interact effectively with water vapor.

Beyond the static characterization of adsorption properties, the presented approach highlights the potential of the quartz crystal microbalance as a dynamic

investigative tool for solvent-polymer matrix interactions. Thanks to its exceptional mass sensitivity and its capability to monitor sorption/desorption phenomena in real time, the QCM stands out as a premier platform to finely explore interaction kinetics, detect mechano-structural transitions, and reveal formulation-specific signatures.

This work opens several high-value research perspectives, such as:

- the development of intelligent QCM-based sensors capable of identifying or quantifying targeted vapors or solvents via the mechanical response of tailored polymer films;
- the exploration of coupling with external stimuli (temperature, electric field, or light), paving the way for adaptive materials and multi-parameter sensing platforms.

By combining advanced physical modeling with high-precision instrumentation, this study contributes to establishing a predictive science of thin polymer film behavior, while outlining the foundations for future detection technologies based on controlled interactions between solvents and polymer matrices.

In addition, recent advances in the literature have demonstrated the breadth of QCM applications across diverse materials and contexts. For instance, molecularly imprinted polymer (MIP)-based QCM sensors have reached  $\text{sub-ng}\cdot\text{mL}^{-1}$  detection limits for antibiotics such as oxytetracycline ( $\text{LOD} \approx 0.019 \text{ ng}\cdot\text{mL}^{-1}$ ) [35] and  $\text{ng}\cdot\text{mL}^{-1}$ -level quantification for sparfloxacin in food matrices, with recoveries of 96–105% and relative standard deviation (RSD) below 5% [36]. Other studies have shown ppb-level gas detection, such as CO at  $\approx 500$  ppb using  $\text{CeO}_2$  nanorod coatings [37]. These achievements, obtained by different groups, illustrate the capacity of QCM to be extended well beyond the biopolymer films studied here, providing useful benchmarks for future developments.

The use of QCM, combined with the modeling approach presented in this work, has proven to be perfectly adequate for describing the interaction of the studied biocompatible polymer films with water vapor. Nevertheless, other techniques such as QCM-D, which additionally monitors energy dissipation within the film, could also be employed to provide complementary information on the viscoelastic properties of thin films [38, 39].

Future work will benefit from expanding into such directions: coupling QCM with other surface-sensitive methods (SPR, ellipsometry, AFM, and electrochemistry) to disentangle solvent mass uptake and mechanical contributions [40–42]; advancing surface functionalization strategies (molecularly imprinted polymers, nanocarbons, metal-organic frameworks, oxides) to enhance selectivity and robustness [36, 39]; and developing portable readout electronics: phase locked-loop (PLL) circuits and MEMS-based devices [43, 44]. Beyond PLL-based solutions, dedicated gain and phase detector integrated circuits such as the AD8302 from analog devices can also be employed for QCM readout, providing accurate frequency and dissipation tracking with relatively simple and compact electronics (openQCM Q-1, Novaetech, Italy).

Finally, we plan to investigate the drying of thin films containing solvents more volatile than water (e.g., alcohols or water-alcohol mixtures), as well as systems incorporating nanoparticles (e.g., silica). These configurations exhibit much faster drying kinetics than those considered in this chapter. The underlying physics is therefore different, essentially involving coupled phenomena of diffusion, sedimentation, and surface tension in a liquid film. In this context, the modeling must be revised, and the signal processing techniques applied to the electrical response of the QCM must be

rethought in order to track the rapid variations of the quartz crystal series resonance frequency during the fast solvent evaporation [45, 46].

## **Appendix A: Impulse response of a polymer film subjected to interfacial transport**

The impulse response of a polymer film subjected to interfacial transport is calculated by applying the Laplace transform (LT) to the boundary value problem (59). Denoting  $\underline{u}(Y, s) = LT[\delta c_w(Y, t_r)]$ , we obtain:

$$\frac{d^2 \underline{u}}{dY^2} = s\underline{u} ; \quad \left. \frac{d\underline{u}}{dY} \right|_{Y=0} = 0 ; \quad \left. \frac{d\underline{u}}{dY} \right|_{Y=1} = -\text{Bi}_m(\underline{u}(1, s) - \underline{u}^\infty(s)) \quad (66)$$

where  $\underline{u}^\infty(s) = TL[\delta c_w^\infty(t_r)]$ .

To solve the boundary value problem (66), we seek the impulse response  $h_F(Y, t_r)$  of the film. By introducing the parameter  $\beta^2 = -s$ , the ordinary differential equation satisfied by  $\underline{u}(Y, s)$  can be rewritten as

$$\frac{d^2 \underline{u}}{dY^2} = i^2 \beta^2(s) \underline{u} \quad (67)$$

whose general solution is

$$\underline{u}(Y, s) = A e^{i\beta(s)Y} + B e^{-i\beta(s)Y} \quad (68)$$

where  $i^2 = -1$ , and the constants  $A$  and  $B$  are determined from the boundary conditions (66). After calculations, the closed-form solution for  $\underline{u}(Y, s)$  in Laplace space is obtained as follows:

$$\underline{u}(Y, s) = \frac{\text{Bi}_m \cos(Y\beta(s))}{\text{Bi}_m \cos(\beta(s)) - \beta(s) \sin(\beta(s))} \underline{u}^\infty(s) = H_F(Y, s) \underline{u}^\infty(s) \quad (69)$$

where  $H_F(Y, s)$  is the Laplace transfer function of the polymer film. The impulse response  $h_F(Y, t_r)$  of the film can be deduced from  $H_F(Y, s)$  using the following result (see [47], p. 27):

$$H_F(Y, s) = \frac{\text{Bi}_m \cos(Y\beta(s))}{P(s)} = \sum_{s_n} \frac{\text{Bi}_m \cos(Y\beta(s_n))}{(s - s_n) P'[\beta(s)]_{s=s_n}} \quad (70)$$

where the  $s_n$  are the roots of the equation  $P(s) = 0$ , with  $P(s) = \text{Bi}_m \cos(\beta(s)) - \beta(s) \sin(\beta(s))$ . The result (70) is applicable for a finite domain, as is the case here since  $Y \in (0, 1)$ . Solving the nonlinear algebraic equation  $P(s) = 0$  leads to the following transcendental equation:

$$\beta_n \tan \beta_n = \text{Bi}_m, \quad \text{with} \quad \beta_n = \beta(s_n) \quad (71)$$

which was solved numerically as a function of  $\text{Bi}_m$  using the Newton-Raphson method. The calculation of  $P'[\beta(s)]$  uses the result  $\beta^2(s) = -s \Rightarrow \beta\beta' = -\frac{1}{2}$ , from which follows:

$$P'[\beta(s)] = \frac{1}{2} \left[ (\text{Bi} + 1) \frac{\sin \beta}{\beta} + \cos \beta \right] \quad (72)$$

which allows expressing the transfer function  $H_F(Y, s)$  in the form of the following infinite series expansion:

$$H_F(Y, s) = \sum_n \frac{2\text{Bi}\beta_n \cos(Y\beta_n)}{(\text{Bi} + 1) \sin \beta_n + \beta_n \cos \beta_n} \frac{1}{s + \beta_n^2} \quad (73)$$

which can be expressed as an infinite linear combination of first-order low-pass filters, making it straightforward to invert. The inversion of this expression leads to the following impulse response  $h_F(Y, t_r)$  of the film:

$$h_F(Y, t_r) = \text{TL}^{-1}[H_F(Y, s)] = \sum_n \frac{2\text{Bi}\beta_n \cos(Y\beta_n)}{(\text{Bi} + 1) \sin \beta_n + \beta_n \cos \beta_n} e^{-\beta_n^2 t_r} \quad (74)$$

Integrating the transfer function (73) and the impulse response (74) over the dimensionless film thickness ( $Y \in (0, 1)$ ) yields the average transfer function  $\bar{H}_F(s)$  and the average impulse response of the film  $\bar{h}_F(t_r)$ , respectively, expressed as follows:

$$\bar{H}_F(s) = \sum_{n=1}^{\infty} \frac{2\text{Bi}_m^2}{\text{Bi}_m(\text{Bi}_m + 1) + \beta_n^2} \frac{1}{s + \beta_n^2} \quad (75)$$

and

$$\bar{h}_F(t_r) = \sum_{n=1}^{\infty} \frac{2\text{Bi}_m^2}{\text{Bi}_m(\text{Bi}_m + 1) + \beta_n^2} e^{-\beta_n^2 t_r} \quad (76)$$

Finally, the temporal variation  $\delta m_W(t_r)$  of the film's mass can be calculated for any variation of concentration  $\delta c_W^\infty(t_r)$  using  $\bar{h}_F(t_r)$  and the convolution theorem as follows:

$$\delta m_W(t_r) = \ell_F \Sigma \int_0^{t_r} \bar{h}_F(t_r - \tau) \delta c_W^\infty(\tau) d\tau \quad (77)$$

which can be expressed as:

$$\delta m_W(t_r) = \ell_F \Sigma \sum_{n=1}^{\infty} \frac{2\text{Bi}_m^2}{\text{Bi}_m(\text{Bi}_m + 1) + \beta_n^2} \int_0^{t_r} \delta c_W^\infty(\tau) e^{-\beta_n^2(t_r - \tau)} d\tau \quad (78)$$

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## Section 3

# Enhancing Energy Generation

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# Maximising Piezoelectric Energy Harvesting: Overcoming Challenges and Unlocking Sustainable Power Generation

*Hwa San Sim, Alain Cheong, Samson Chang, Lian Hock Lim and Pradeepkumar Krishnamoorthy*

## Abstract

Piezoelectric generation presents a promising path towards sustainable energy. Transforming mechanical pressure into electrical power supports our pursuit of environmentally friendly solutions, contributing to a cleaner and more efficient energy landscape while reducing our reliance on traditional sources. While piezoelectric generation holds immense potential, a notable challenge is its relatively low power output. Due to the limited mechanical stress available from ambient sources like walking or vibrations, the generated energy often falls short for many applications. Innovative strategies are essential to overcome this limitation and unlock their full energy harvesting capabilities. This research focuses on optimising the power output of piezoelectric energy harvesting systems by enhancing both mechanical stress application and electrical conversion efficiency. We propose the integration of rubber stoppers and mechanical spacers onto the piezoelectric transducers to induce greater mechanical deformation, thereby improving energy conversion performance. Furthermore, we investigate the strategy of vertically stacking multiple piezoelectric transducers mounted on PCB assemblies. This stacking approach demonstrates a significant boost in cumulative energy output, with power generation increasing proportionally to the number of layered piezoelectric modules. Importantly, these advancements go beyond theoretical concepts. The improved piezoelectric system offers practical applications, such as powering LED walkways and electrical appliances. This translates into a viable and sustainable energy source that holds promise across a spectrum of real-world scenarios. By overcoming the challenges of limited power output, we unlock the full potential of piezoelectric energy harvesting, contributing to a greener future.

**Keywords:** energy harvesting, piezoelectric energy, smart infrastructure, power generation, sustainable technology, renewable energy systems, IoT integration, footstep-powered applications, system optimisation

## **1. Introduction**

As the world shifts towards cleaner energy systems, the need for innovative and decentralised power solutions has become increasingly urgent. According to the International Energy Agency, global electricity demand is projected to grow by nearly 80% between 2012 and 2040, highlighting the challenge of meeting future needs without increasing carbon emissions [1]. To address this growing demand sustainably, researchers have intensified their efforts to harness ambient energy sources, including solar radiation, thermal gradients, and mechanical vibrations.

Energy harvesting technologies are generally classified into three categories: solar, thermal, and mechanical [2]. While solar and thermal systems are well-established, they are often constrained by environmental dependencies such as sunlight availability, surface orientation, or temperature differences. Mechanical energy, on the other hand, is readily available in urban settings—from foot traffic, machinery, and vehicle motion—yet much of it remains untapped.

Piezoelectric energy harvesting (PEH) offers a promising route to capture this mechanical energy and convert it into electrical power using piezoelectric transducers (PTs). These systems are increasingly considered for low-power applications such as structural health monitoring, embedded Internet of Things (IoT) devices, wearable electronics, and smart infrastructure [3, 4]. Their appeal lies in their compact size, passive operation, and maintenance-free nature, making them ideal for decentralised deployment in built environments.

Despite these advantages, PEH systems face a fundamental limitation: low power output, particularly when subjected to ambient, low-intensity mechanical forces such as footsteps or low-frequency vibrations [5]. This limitation significantly reduces their effectiveness in real-world applications and presents a key challenge for widespread adoption.

To address this, prior research has proposed several strategies, including impedance-matching circuits, i.e., Maximum Power Point Tracking (MPPT) [6], and advanced AC-DC rectification methods [7]. While effective in controlled environments, these techniques often introduce complexity, are costly to implement, or perform poorly under irregular, human-induced mechanical input.

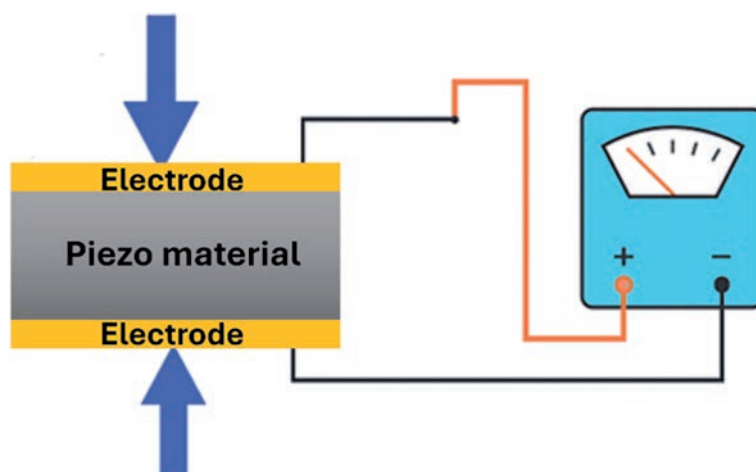
In contrast, this work explores practical mechanical and structural enhancements to maximise energy output under low-force, footstep-based conditions. Focusing on lead zirconate titanate (PZT) transducers, we investigate how simple yet effective design modifications—such as rubber stoppers and mechanical spacers—can significantly enhance deformation and thus energy conversion. We further propose a stacked configuration of PCB-mounted PTs to boost energy density without increasing system footprint. These strategies are experimentally validated and offer actionable, scalable insights for real-world applications of piezoelectric harvesting that support sustainable development goals.

## **2. Methodology**

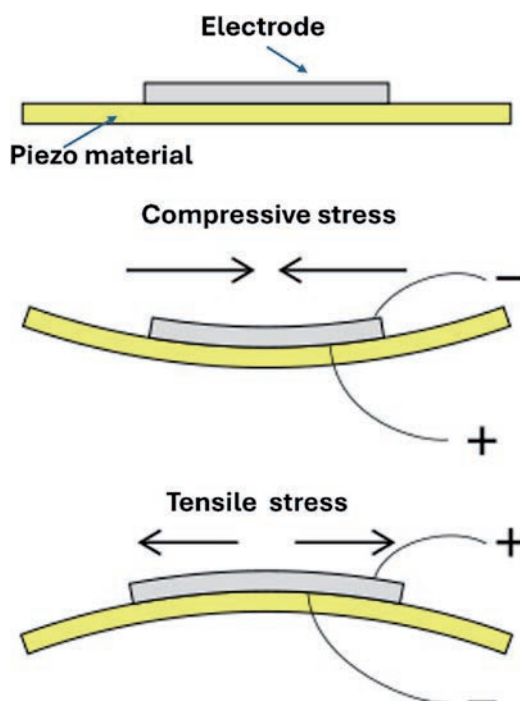
This study explores structural and electrical enhancements to optimise the energy output of PTs, with a focus on practical replication and scalability. The methodology is structured to deepen understanding of piezoelectric working principles, justify material and design choices, and describe the experimental configuration used to evaluate performance.

## 2.1 Working principle of piezoelectricity

The term *piezoelectricity* derives from the Greek word *piezein*, meaning “to press” or “squeeze.” Piezoelectric materials produce an electric field when subjected to mechanical stress due to shifts in internal electric dipoles. When at rest, these materials are electrically neutral; however, applied force disturbs this balance, generating charge separation and surface voltage (Refer to **Figures 1** and **2**). The amount of



**Figure 1.**  
 A piezo material generating an electric voltage upon compression.



**Figure 2.**  
 Piezo material upon compression.

voltage generated is directly proportional to the degree of deformation, a key design consideration in energy harvesting systems.

This electromechanical interaction makes piezoelectric materials ideal for harvesting low-frequency mechanical energy, such as that from human footsteps. Maximising deformation under load directly increases the electrical output, guiding the structural modifications explored in this study. Furthermore, understanding the relationship between mechanical input and electrical response is essential for optimising both material selection and transducer design in real-world applications.

## 2.2 Material and transducer selection

More than 200 types of piezoelectric materials exist [7], each with unique mechanical, electrical, and cost profiles. Quartz, for example, offers high precision but low sensitivity and is costly to fabricate. In contrast, piezoceramics such as PZT offer high piezoelectric coefficients, cost-effectiveness, and mechanical robustness, making them suitable for energy harvesting under pedestrian-induced forces.

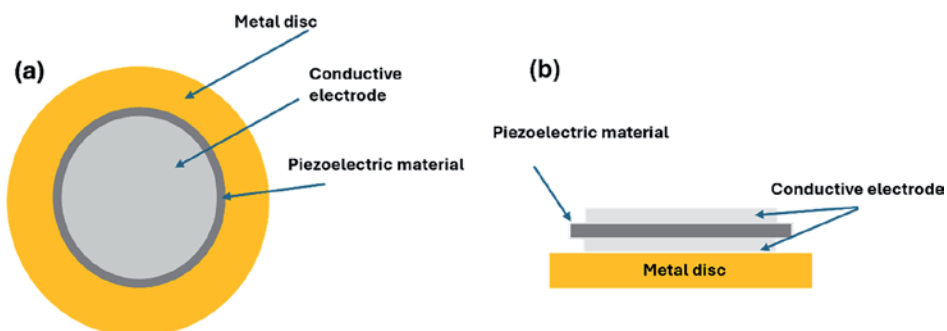
This study employs PZT circular diaphragm transducers (See **Figure 3**). Their shape enables even force distribution, and their commercial availability makes them practical for scalable deployment in educational and urban contexts.

## 2.3 Mechanical modes of operation

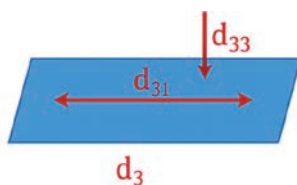
The mechanical–electrical response of a PT depends on how force is applied. Two principal operating modes are relevant:

- d33 mode (transverse): Compression is applied perpendicular to the surface (along thickness), producing charge in the same axis.
- d31 mode (longitudinal): Force is applied parallel to the surface, causing lateral deformation and generating charge across the thickness.

These modes are visualised in **Figure 4**. To activate both modes efficiently, this study introduces mechanical enhancements—namely rubber stoppers and wire mechanical spacers—that amplify deflection and direct strain appropriately across the transducer surface.



**Figure 3.**  
*Circular diaphragm transducer: (a) Front view; (b) Side view.*



**Figure 4.**  
 $d_{33}$  and  $d_{31}$  mode and the directions of the force.

## 2.4 Energy harvesting circuit

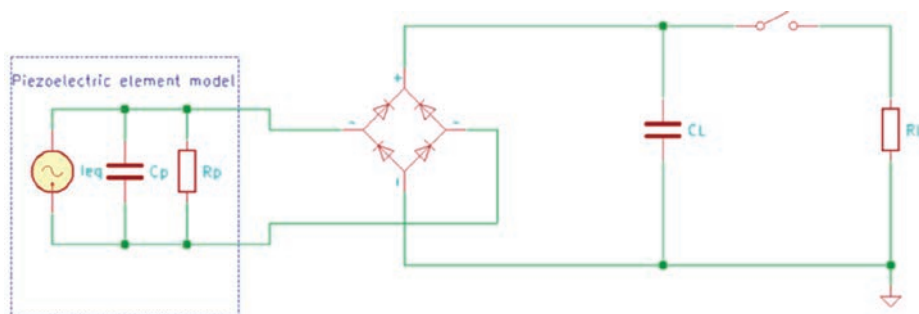
The energy produced by PTs is in alternating current (AC) form. To power devices or store the energy, conversion to direct current (DC) is required [6, 7]. A full-wave bridge rectifier was selected for its simplicity and high efficiency in converting bipolar signals to unidirectional output.

The circuit, illustrated in **Figure 5**, models the PT as an equivalent current source ( $I_{eq}$ ) in parallel with a capacitance ( $C_p$ ) and leakage resistance ( $R_p$ ). The rectified voltage is stored in a capacitor ( $C_L$ ), which can later be discharged across a load ( $R_L$ ) for energy calculation or practical use. This configuration reflects real-world application scenarios such as powering LEDs or sensors.

## 2.5 Experimental design for replication and scalability

This research prioritises not only performance metrics but also real-world usability. All setups were built using standard lab tools and commercially available components, ensuring they can be replicated in educational institutions and scaled for field applications. The approach of vertically stacking multiple PCBAs, each embedded with modified PTs, reflects a deliberate strategy to increase energy density without expanding the system's surface footprint.

Additionally, the inclusion of rubber stoppers and wire spacers introduces controlled mechanical deformation, allowing comparisons between unmodified and modified transducer setups. These design modifications aim to enhance energy capture efficiency under footstep-like loading, typical of smart infrastructure use cases.



**Figure 5.**  
 Standard AC-DC energy harvesting circuit with electromechanical piezoelectric model as described by Covaci et al. [7].

This methodology enables a robust evaluation of both the electrical performance and deployment potential of the system, setting the stage for experimental results that are practical, replicable, and aligned with global sustainability goals.

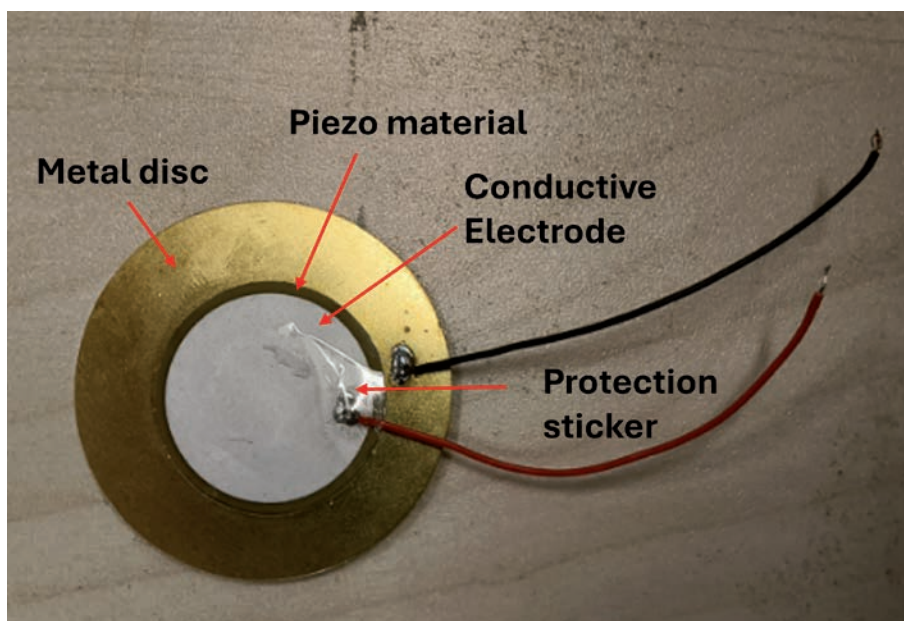
### 3. Experimental setup

Circular diaphragm PTs were used, each consisting of a 41 mm-diameter metal shim with a 24.5 mm inner electrode coated in conductive paste. These were paired with DF005SA-E3/77 bridge rectifier integrated circuits sourced from Digikey Electronics. The experimental setup included the PTs, full-wave bridge rectifiers, a 10  $\mu\text{F}$  capacitor (C1), a calibrated force gauge with a 1.5 cm diameter flat-surfaced tip, and an oscilloscope to capture voltage responses under varying mechanical force conditions.

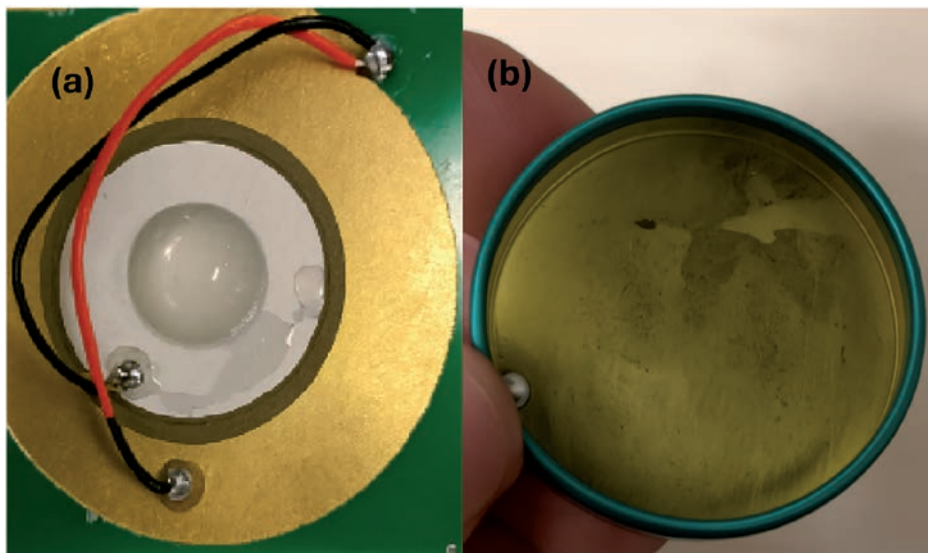
The unmodified PT, shown in **Figure 6**, served as the baseline device. Its conductive electrode and piezo material film are essential for power generation, and any surface damage can impair performance. To protect the piezo material and the conductive electrode during handling and testing, a sticker was applied over it, as illustrated in **Figure 6**.

To investigate the influence of different working modes on energy harvesting, the study focused on two primary piezoelectric modes: d33 and d31. The d33 mode is activated by a vertical force applied perpendicular to the PT surface, inducing compression along the thickness direction. In contrast, the d31 mode involves in-plane strain, triggered when the deformation is redirected laterally across the disc surface due to structural constraints that promote bending.

To enable controlled activation of both modes, the PT was structurally modified as shown in **Figure 7**. A dome-shaped rubber stopper (10 mm diameter, 5 mm height) was added to the top surface (**Figure 7(a)**) to facilitate vertical loading, primarily



**Figure 6.**  
*Unmodified PT with protective sticker on the unmodified PT.*



**Figure 7.**  
 (a) Rubber dome stopper on the modified PT and (b) A wire loop is positioned around the back of the piezo disc to act as a mechanical spacer, enabling depression to a defined depth corresponding to the wire's thickness.

activating the d33 mode while also contributing to lateral strain. A 1.2 mm-thick wire loop was positioned beneath the disc (**Figure 7(b)**) to elevate the disc edges and encourage bending during compression, thereby enhancing d31 mode activation. While both modes are simultaneously excited under applied stress, these structural enhancements influence their relative contributions, enabling a more complete assessment of piezoelectric behaviour under varying mechanical inputs.

To assess energy harvesting from multiple transducers, a 200 mm × 200 mm printed circuit board assembly (PCBA) was developed, incorporating sixteen modified PTs arranged from PT1 to PT16 and connected in parallel (**Figure 8(a)**).

The decision to integrate 16 PTs onto a single 200 mm × 200 mm PCBA was driven by the need to balance energy output with spatial efficiency while accommodating real-world use cases. Each circular PT, measuring approximately 41 mm in diameter, was arranged to maximise surface coverage within the footprint of a typical human foot. This layout ensures that the applied pressure from a single footstep can effectively activate multiple PTs simultaneously. The parallel configuration was chosen due to its superior voltage and energy output compared to the series connection [8]. Human-centred testing—such as simulating the force of a person standing on one foot—validated the harvester's functionality under realistic conditions, making it suitable for dynamic environments such as corridors, event spaces, or pedestrian walkways.

A key challenge was ensuring uniform force distribution across all PTs, which was addressed by employing a rigid overlay plate. Standard PCB layout practices were followed to ensure clean signal routing and reliable convergence of rectified outputs at capacitor C1, supporting accurate energy measurement with minimal loss. This experimental setup reflects practical deployment scenarios such as building floors, corridors, and access points where uniform activation and robust data capture are essential.

As shown in **Figure 8(b)**, each PT is individually connected to its own bridge rectifier, which converts the AC signal generated by the PT into DC. These rectified

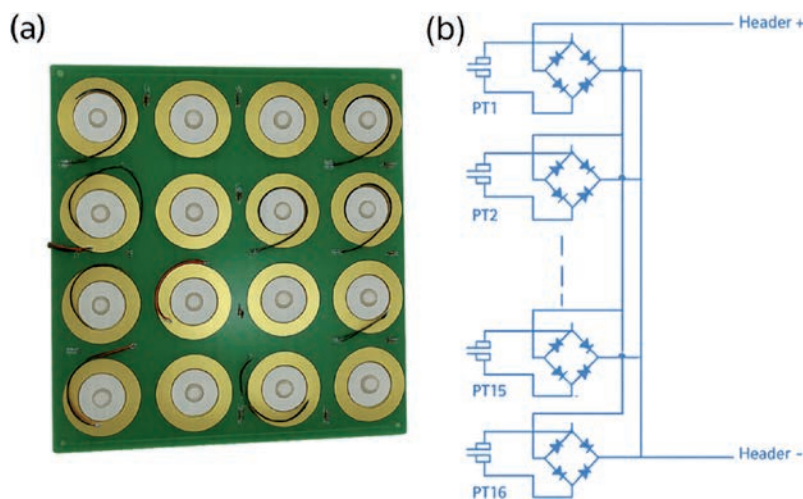
outputs are then combined in parallel to form a unified energy output, ensuring consistent performance and efficient energy harvesting across the entire array.

To ensure uniform mechanical loading, the PCBA was placed on a rigid, flat surface to prevent deformation during force application. A single compressive force was applied vertically to the centre of the board, enabling simultaneous activation of all 16 PTs under consistent loading conditions. This setup is shown in **Figure 8**, while **Figures 9** and **10** illustrate the configurations for the single-layer and stacked PCBAs, respectively.

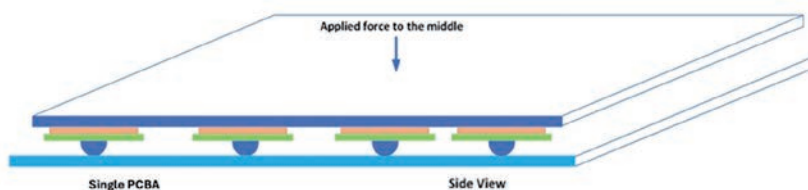
To estimate energy generation, the PTs were connected to a measurement circuit as shown in **Figure 11**, where the output voltage was rectified and stored in capacitor C1. The energy harvested was calculated using:

$$E = \frac{1}{2} \times C \times V^2 \quad (1)$$

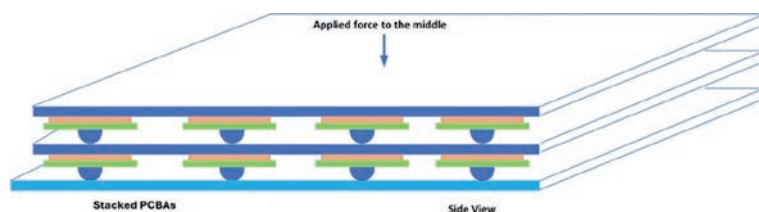
where E is the energy in joules, C is the capacitance of C1 (10  $\mu$ F), and V is the peak voltage across the capacitor. Prior to each measurement, capacitor C1 was fully discharged by shorting its terminals with a wire. The oscilloscope probe was then connected across the capacitor to record the voltage solely generated by an applied force.



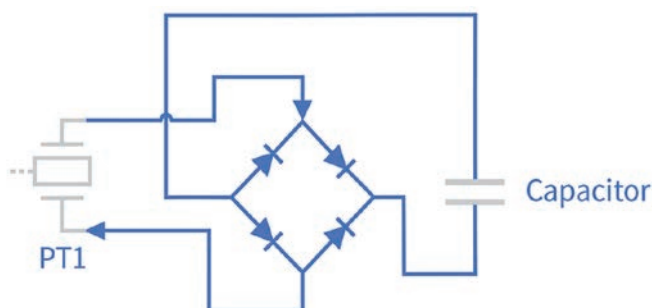
**Figure 8.** (a) PCBA of 16 modified PTs with rubber dome stopper facing downward, (b) Corresponding schematic diagram illustrating the circuit configuration of 16 PTs.



**Figure 9.** Side view of the experimental setup showing applied force on a single PCBA populated with 16 modified PTs.



**Figure 10.**  
 Side view of the experimental setup showing applied force on a stacked configuration of two PCBAs, each populated with 16 modified PTs (PTs).



**Figure 11.**  
 Setup for estimating power generated from the PT [6, 7].

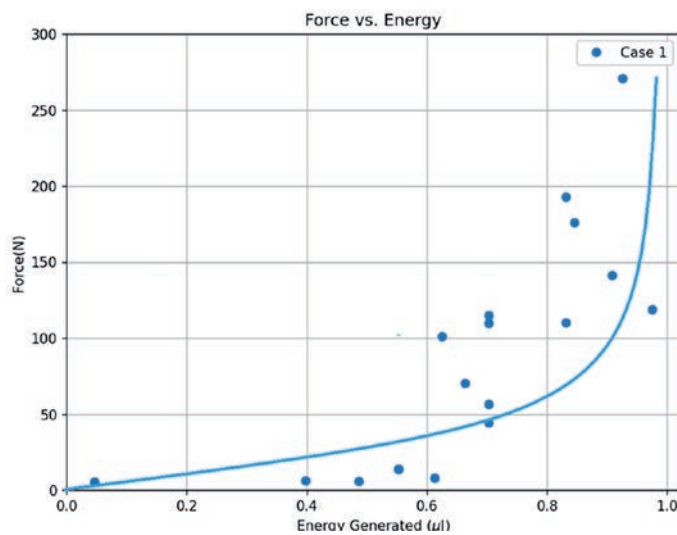
The experimental cases evaluated were as follows:

- Case 1: A single unmodified PT was tested with the applied force gradually increased from 0 to 1000 N.
- Case 2: A single modified PT, incorporating a dome-shaped stopper and a wire spacer (**Figure 7**), was tested under the same force range.
- Case 3: Sixteen modified PTs were integrated onto a single PCBA and subjected to increasing force up to 1000 N.
- Case 4: Two identical PCBAs from Case 3 were stacked vertically to form a dual-layer configuration and tested under the same force conditions.

A compressive force ranging from 0 to 1000 N was applied to simulate the weight of a typical human using a calibrated force gauge with a flat circular tip. The force was applied perpendicularly at the centre of the PT (Cases 1–2) or the centre of a PCBA (Cases 3–4), ensuring consistent distribution of the excitation force. The peak force and resulting voltage were recorded for each trial. For all cases, measurements were taken until energy output saturation was observed.

## 4. Results and discussion

**Figure 12** illustrates that for a single unmodified PT (Case 1), the energy output reaches a saturation point, as evidenced by identical energy values recorded at both 175 N and 200 N of applied force. This plateau indicates that the transducer has



**Figure 12.**  
Case 1 plot (Unmodified PT as per Figure 6 was used).

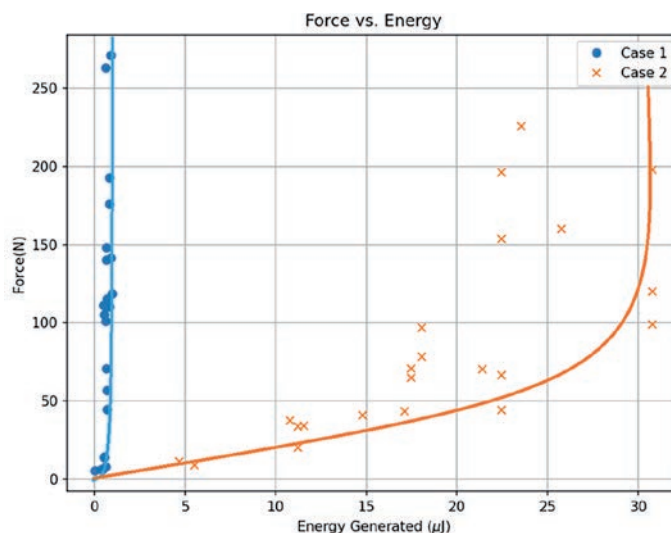
reached its mechanical compression limit, beyond which further deformation—and thus additional energy conversion—is not possible. The peak energy output recorded is relatively low, at only 0.95  $\mu\text{J}$ .

This result aligns with findings reported by Godwin Chukwunonyelum et al. [9], where a 50 kg weight applied over 100 footfalls produced 55.5 mV and 6.3 mA from twelve piezoelectric discs. Based on the assumption of one footfall per second, the estimated energy generated per transducer per footfall was approximately 0.29  $\mu\text{J}$ . This value furthermore falls within the microjoule range, consistent with the findings of the current study. The low energy generated is likely as a result of limitations in the applied mechanical stress and electric field intensity relative to the material's polarisation direction—specifically, the restricted mechanical deformation in the d33 and d31 modes caused by the fixed mechanical flex or stress of the unmodified PT.

As previously described, the modified PT used in Case 2 incorporates a centrally placed dome-shaped stopper and a wire ring measuring 1.2 mm in thickness acting as a mechanical spacer. These structural enhancements enabled greater deformation under applied force, improving the activation of both d33 and d31 modes, and thereby significantly increasing energy output.

**Figure 13** illustrates the enhanced deformation behaviour and improved energy performance of the modified PT. The addition of the wire spacer beneath the disc raises its edges slightly, allowing vertical force to induce lateral strain across the disc surface, activating the d31 mode in conjunction with the d33 mode. This dual-mode activation significantly improves the PT's energy conversion efficiency. The dome-shaped stopper also contributes to controlled and repeatable force application at the disc's centre. Together, these modifications result in greater overall deformation under the same applied force, accounting for the superior energy output observed in the modified configuration compared to the unmodified PT.

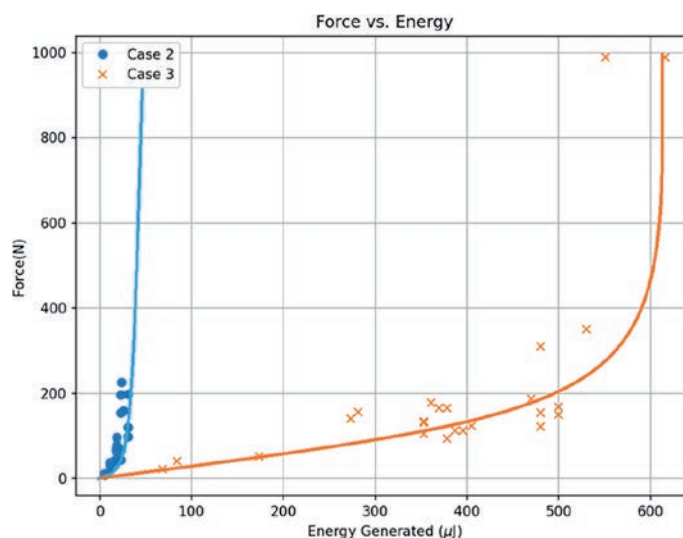
Experimental results reinforce the impact of design improvements on power generation. Case 1 (unmodified PT) exhibited limited energy output due to constrained mechanical deformation. In contrast, Case 2 (modified PT) achieved a notable increase in energy output, attributed to improved deflection made possible by the



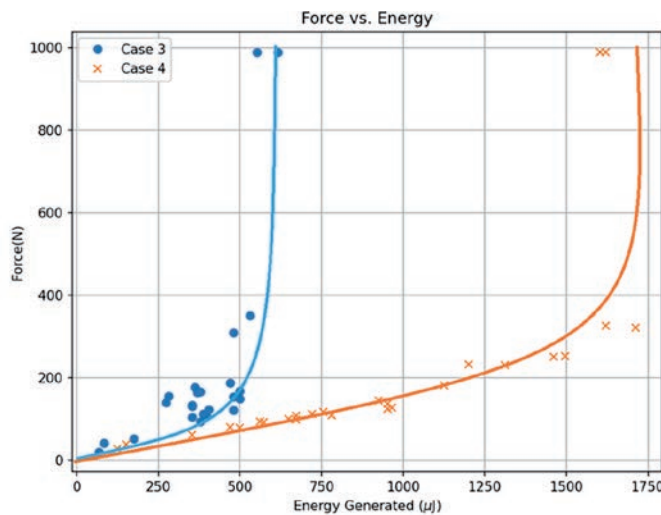
**Figure 13.** Comparing the energy generated by Case 1 (single unmodified PT, refer to **Figure 6**) and Case 2 (Single modified PT, refer to **Figure 7**).

structural enhancements. These modifications activated multidirectional piezoelectric modes and significantly improved energy harvesting efficiency.

**Figure 14** demonstrates the performance comparison between Case 2 (a single modified PT) and Case 3 (a configuration of 16 modified PTs connected in parallel). The results exhibit that Case 3 produces a substantially greater energy output, many times greater than Case 2, demonstrating the effectiveness of parallel integration. This substantial improvement arises from the combined output of 16 identical modified PTs operating simultaneously.



**Figure 14.** Illustration of Case 2 (single modified PT) and Case 3 (16 modified PTs), demonstrating that the use of 16 modified PTs results in a significantly higher power output than that of a single modified PT.



**Figure 15.** Illustration of Case 3 (single-layer of 16 PT PCBA) and Case 4 (two-layer of 16 PT PCBA stack one on top of the other).

Case 4 explores the vertical stacking of PT units to further enhance energy output within a limited footprint. Each unit consists of 16 modified PTs assembled on a single PCB. In this configuration, two such units are physically stacked, with their electrical outputs connected in parallel to enable simultaneous energy harvesting from both layers.

**Figure 15** compares the energy output of Case 3 (a single-layer configuration) and Case 4 (a stacked two-layer configuration). The results show that Case 4 consistently generates significantly higher energy across a range of applied forces, including at lower force levels, demonstrating the effectiveness of vertical stacking in enhancing energy harvesting performance. Tests involving a human subject applying approximately 1000 N of force, by standing on one foot on the PCB assembly, confirmed that both configurations eventually reach an output saturation point, highlighting a physical limit to energy conversion under such loading. Achieving higher energy yields would likely require additional stacking, though this introduces trade-offs in size, weight, and system complexity. Importantly, the rate of energy increase with applied force differed notably between the configurations. Case 4 produced nearly 2.5 times more energy than Case 3 under identical force conditions, affirming the potential of stacking to improve energy density where floor space is constrained. This finding aligns with studies by Shenck and Paradiso [10], which showed that improvements in electrode configuration and force distribution can lead to exponential efficiency gains. Additionally, the incorporation of structural stoppers and spacers in this study enabled activation of both d32 and d33 modes, promoting multidirectional deformation. This form of mechanical coupling—often overlooked in conventional designs—proved crucial in enhancing energy output.

## 5. Applications

Piezoelectric energy harvesting presents a highly adaptable solution for generating electrical energy in environments rich in mechanical motion, particularly where

vibrations or repeated human activity occur. By converting kinetic energy into usable electrical power, piezoelectric systems enable decentralised, low-maintenance energy sources that are especially valuable for powering small-scale devices and infrastructure.

In public transportation systems, for instance, piezoelectric tiles strategically embedded at station entrances, ticketing zones, or turnstiles can capture the mechanical energy of pedestrian footfalls. The harvested energy can be used to operate nearby systems such as ambient lighting, digital display panels, surveillance cameras, or environmental sensors. These deployments can improve energy efficiency while reducing reliance on traditional power grids. Pilot projects in Japan and Israel have demonstrated that footstep energy from thousands of commuters can meaningfully power nearby devices [11–13].

In the broader context of smart cities, piezoelectric systems offer a sustainable means of powering urban services. Applications include powering air quality monitoring sensors, traffic counters, street lighting in low-traffic walkways, and interactive information kiosks. Their passive and self-sustaining nature reduces both operating and maintenance costs over time and contributes to carbon emissions reduction.

Consumer technology is another promising domain for piezoelectric integration. Wearable electronics—such as smart footwear, backpacks, or fitness gear—can incorporate miniature piezoelectric elements to convert human motion into energy. This energy can support embedded devices like health monitors, fitness trackers, GPS units, and emergency alert systems. In scenarios where battery life is a constraint, such as remote outdoor activities or disaster zones, self-powered systems provide meaningful advantages.

In high-footfall indoor environments, including hospitals, airports, shopping centres, and educational campuses, piezoelectric flooring offers a practical means of passive energy generation. These installations can support illuminated emergency signage, motion-triggered lighting, occupancy detection, and real-time footfall analytics for building management. Such systems contribute to operational efficiency, safety, and sustainability without requiring extensive infrastructure changes.

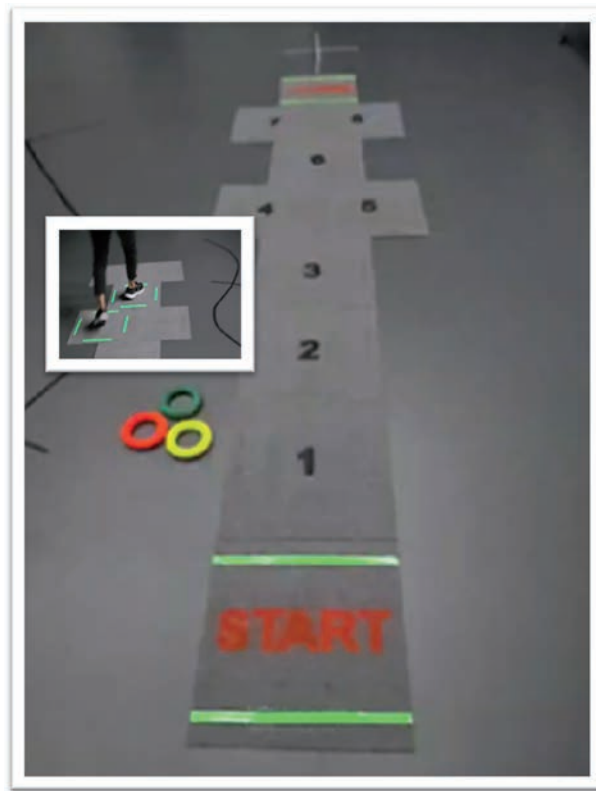
To demonstrate the feasibility and educational value of these applications, three working prototypes were designed and implemented within the campus environment of ITE College West, one of three colleges under the Institute of Technical Education (ITE). These installations not only provided functional demonstrations but also served as tools to promote sustainability awareness among students and visitors.

### **5.1 Kinetic Hopscotch**

This interactive installation is designed to make energy harvesting both visible and engaging. It lights up embedded LED strips in response to foot pressure from users, showcasing real-time mechanical-to-electrical energy conversion. Installed at the Drone and Robot Hub at ITE College West, the system was showcased during a course promotion activity, where students participated in an experiential learning session on renewable energy and electronics (**Figure 16**). The installation served both as an educational tool and a practical example of user-triggered energy harvesting.

### **5.2 Kinetic emergency lighting**

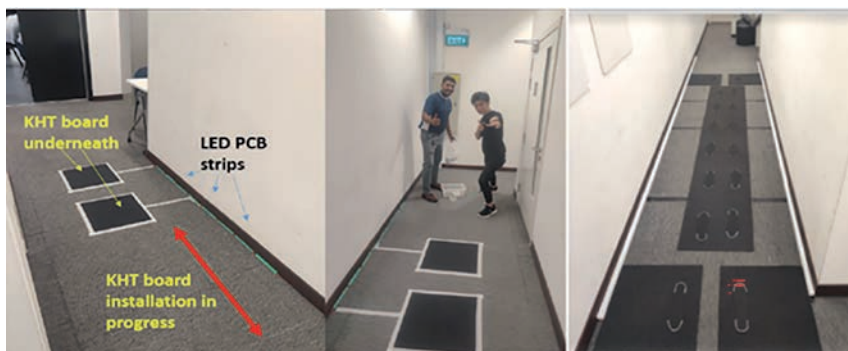
This corridor-based system uses piezoelectric tiles to power LED strips that illuminate only when stepped on. Installed near emergency exits, it provides a responsive



**Figure 16.**

*Implementation of the Kinetic Hopscotch at the Drone and Robot Hub ITE College West. (See attached video: <https://bit.ly/4dGLQhZ>).*

and energy-efficient safety solution. Unlike conventional systems that remain on continuously, this lighting activates only in the presence of foot pressure, reducing energy consumption while ensuring real-time visibility when needed (**Figure 17**). Such a system is particularly valuable in facilities where emergency lighting is seldom used but must remain functional.



**Figure 17.**

*Kinetic emergency lighting for fire exits. (See Video: <https://bit.ly/4dGLQhZ>).*



**Figure 18.**  
*Deployment and implementation of Kinetic Game Station. (See Video: <https://bit.ly/4dGLQhZ>).*

### 5.3 Kinetic game station

This platform combines a piezoelectric flooring system with a rechargeable battery and a ticket dispenser. As users step on the tiles, energy is stored until a set threshold is reached. Once enough energy is accumulated, the stored charge powers a ticket dispenser that releases a reward, gamifying the energy harvesting experience. Used during school engagement events, the system not only demonstrated the viability of kinetic energy capture but also served as a playful and memorable sustainability outreach tool (**Figure 18**). By integrating real-world energy generation with a reward mechanism, it made abstract sustainability concepts tangible and fun.

## 6. Conclusion

In summary, this chapter has examined the principles, structural configurations, and practical potential of piezoelectric energy harvesting systems. By comparing the performance of modified and unmodified PTs, the study showed that structural enhancements, such as the integration of rubber stoppers and mechanical spacers, significantly improve mechanical deformation, thereby boosting energy output. These enhancements increase the efficiency with which PTs convert mechanical pressure into electrical energy, particularly under low-force conditions typical of pedestrian activity.

The findings highlight the critical importance of how PTs are physically arranged and supported, referred to here as structural configurations. Our experimental results confirm that the layout and mechanical reinforcement of PTs directly influence both the quantity and consistency of harvested energy. In particular, activating multiple piezoelectric modes through well-designed mechanical interfaces emerged as a key strategy for performance enhancement.

A particularly significant innovation explored in this work is the vertical stacking of multiple PCBAs, each embedded with modified PTs. This approach enables greater energy output without increasing the system's surface footprint. By stacking PT-equipped PCBAs, the same floor area can produce a higher energy yield, making the method ideal for use in space-limited environments such as pedestrian walkways, staircases, transport hubs, and building entrances. The modular design of these stacked assemblies also allows energy output to be scaled to suit specific application needs, ranging from small sensors and LED indicators to more complex, self-powered Internet of Things (IoT) systems.

Practical implementations—such as the Kinetic Hopscotch, Emergency Lighting, and Game Station—demonstrate how this technology can transition from controlled laboratory conditions to real-world deployment in educational and public settings. These installations not only validate the technical feasibility of piezoelectric systems but also promote sustainability awareness through interactive, user-driven engagement. Their integration into high-traffic environments illustrates the viability of kinetic energy harvesting as a clean, embedded power source.

Looking ahead, continued advancements in piezoelectric material science, circuit optimisation, and energy storage technologies will further improve the performance and applicability of piezoelectric energy harvesting. Integration with smart city infrastructure, predictive analytics, and wireless communication technologies could extend the role of these systems into responsive, decentralised energy networks. There is also strong potential for cross-sector adoption, spanning transportation, healthcare, retail, and education, where footfall and vibration can be converted into useful power. As the demand for sustainable, low-carbon solutions intensifies globally, piezoelectric harvesting systems—especially those built with optimised and stackable configurations—are well-positioned to become vital components in building greener, more resilient infrastructure.

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## Section 4

# Glimpse on Piezoelectric Sensors

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## Chapter 5

# Efficient Interfacing Schemes to Enhance the Sensitivity and SNR Performance of Piezosensor Systems

*Byju Chamblon and Sreenath Vijayakumar*

### Abstract

This chapter explores innovative interfacing techniques designed to improve the sensitivity and signal-to-noise ratio (SNR) of piezoelectric sensor systems. It begins with an overview of conventional interfacing approaches, namely the standard voltage amplifier and charge amplifier configurations. The chapter then introduces a novel enhanced voltage amplifier scheme specifically designed to mitigate the detrimental effects of cable parasitic capacitance, which often degrades the performance of piezosensor systems deployed in harsh environments requiring long cable connections between the sensor and signal-conditioning electronics. In such applications, long cables introduce signal degradation due to capacitive loading, triboelectric noise, and external electromagnetic interference—factors that significantly impair sensitivity and SNR. Notably, piezopolymer-based sensors such as polyvinylidene fluoride (PVDF) exhibit high voltage sensitivity relative to charge sensitivity, making voltage amplification inherently advantageous. However, in practice, charge amplifiers are often used to combat cable-induced loading, resulting in reduced overall sensitivity due to the poor charge sensitivity of piezopolymer sensors. To address this limitation, the chapter details the design, simulation, and implementation of an enhanced voltage amplifier architecture that retains the benefits of voltage amplification while remaining insensitive to cable parasitic effects. Simulations of the proposed scheme were performed using LTspice and compared experimentally with standard voltage and charge amplifier configurations. Prototypes of each scheme were evaluated under controlled conditions with varying cable parasitic capacitance and common-mode interference. Comprehensive experimental results are presented, demonstrating the superior performance of the enhanced voltage amplifier in maintaining high sensitivity and SNR in challenging deployment scenarios.

**Keywords:** cable parasitic, charge amplifier, piezoelectric sensor, piezopolymer, PVDF sensor, sensitivity, voltage amplifier, piezoelectric charge constant, piezoelectric voltage constant

## 1. Introduction

Piezoelectric sensors are integral to a broad spectrum of modern applications, ranging from process instrumentation and structural health monitoring to automotive systems, medical diagnostics, consumer electronics, and non-destructive testing [1]. These sensors operate on the principle of the piezoelectric effect, whereby mechanical stress or strain is converted into an electrical signal in the form of voltage or charge [2]. Their widespread adoption is attributed to several compelling characteristics, including high reliability, wide dynamic range, excellent linearity, mechanical robustness, insensitivity to electromagnetic interference, and stable performance across a broad temperature range [1, 2].

Different piezoelectric materials play a crucial role in determining the performance and suitability of piezoelectric sensors for various applications. Commonly used materials include ceramics like lead zirconate titanate (PZT), polymers such as polyvinylidene fluoride (PVDF), and single crystals like quartz and lithium niobate. PZT offers high piezoelectric coefficients and is widely used in industrial and medical sensors, while PVDF provides flexibility, low acoustic impedance, and is ideal for wearable and biomedical applications [3]. Quartz, known for its thermal stability and precision, is often used in timing and frequency control devices. The choice of material significantly influences the sensitivity, frequency response, temperature tolerance, and mechanical robustness of the sensor. PVDF, in particular, has emerged as a material of choice in numerous applications due to its favorable properties such as high piezoelectric voltage constant, low acoustic impedance, mechanical flexibility, chemical stability, and biocompatibility. These characteristics make PVDF well suited for a diverse set of applications including hydrophones and sonar transducers [4], biosensors [5, 6], energy harvesting systems [3], medical ultrasound imaging [5], pressure sensors [6], acoustic detection (e.g., microphones), and structural health monitoring [7, 8].

A critical consideration in piezoelectric sensor systems is the choice of interfacing circuitry, which directly affects system sensitivity and signal fidelity. Piezoelectric sensors require specialized interfacing circuits to effectively convert their high-impedance output into measurable voltage signals. Common approaches include charge amplifiers, which minimize signal distortion by directly amplifying the charge output, and voltage amplifiers, which are simpler but sensitive to cable capacitance. High-input-impedance JFET or CMOS-based amplifiers are often used to reduce loading effects, while resonant circuits enhance sensitivity at specific frequencies. Additionally, transimpedance amplifiers (TIAs) can be employed for current-mode readout, improving noise performance. Some designs incorporate buffered voltage followers to isolate the sensor from external noise. Recent advancements also explore negative capacitance and integrated electronics to enhance sensitivity and bandwidth. However, challenges such as drift, cable interference, and frequency response limitations persist, driving ongoing research into optimized conditioning circuits for piezo sensors.

The two most widely used interfacing schemes are the charge amplifier and the voltage amplifier [5, 9]. The selection between these configurations depends largely on the inherent properties of the piezoelectric material. For materials with high charge sensitivity—such as piezoceramics—charge amplifiers are typically preferred. In contrast, piezopolymer sensors like PVDF, which exhibit significantly higher voltage sensitivity ( $g_{13} \approx 0.216 \text{ V}\cdot\text{m}/\text{N}$ ) compared to their charge sensitivity ( $d_{33} \approx 33 \text{ pC}/\text{N}$ ) [10], perform optimally when interfaced using voltage amplifiers [10].

However, in many real-world PVDF-based sensor applications—such as structural health monitoring, biosensing, medical imaging, and underwater sonar—the sensor and electronics are spatially separated and connected *via* long cables. These cables introduce parasitic capacitance, triboelectric noise, and external interference, which degrade sensitivity and signal-to-noise ratio (SNR). To address capacitive loading, charge amplifiers are often employed, albeit at the cost of reduced sensitivity due to the low charge constant of PVDF sensors. This trade-off creates a significant performance bottleneck in high-fidelity sensing applications.

To overcome these limitations, this chapter presents an enhanced voltage amplifier scheme specifically designed to be insensitive to cable parasitic capacitance. The proposed interfacing approach enables the use of voltage amplifiers even in long-cable scenarios, thereby leveraging the high voltage sensitivity of piezopolymer sensors without suffering from signal degradation. Enhanced and negative capacitance-based voltage amplifier architectures are discussed as alternatives to conventional charge amplifier-based designs for such scenarios.

The remainder of this chapter is organized as follows:

Section II outlines the key criteria for selecting amplifier configurations and provides an overview of the operation principles of standard voltage and charge amplifiers. Section III details the design and functionality of the enhanced voltage amplifier scheme. Section IV discusses how various nonidealities influence the functionality and performance of the proposed scheme. Section V presents simulation studies (performed using LTspice), experimental setup, and results comparing the proposed scheme with conventional approaches in terms of sensitivity and SNR under various conditions, including the presence of cable parasitic and common-mode interference.

This chapter aims to bridge the gap between theoretical design and practical deployment, offering robust interfacing strategies that enhance the performance of piezoelectric sensor systems in demanding environments.

## **2. Criteria for amplifier selection and overview of interfacing schemes**

The performance of a piezoelectric sensor system is significantly influenced by the characteristics of the interfacing circuitry used to process its output signal. Since piezoelectric sensors act as high-impedance charge or voltage sources, the design of the front-end amplifier must be carefully tailored to preserve signal fidelity while minimizing noise and loading effects. Selecting the appropriate amplifier configuration, either a voltage amplifier or a charge amplifier requires a clear understanding of the sensor material properties, application environment, signal levels, and the nature of the signal transmission path.

### **2.1 Criteria for amplifier selection**

Selecting an appropriate amplifier configuration is a critical design consideration in piezoelectric sensor systems, as it directly influences the sensitivity, signal integrity, and overall system performance. The two most widely adopted interfacing schemes are charge-mode and voltage-mode amplifiers, each with distinct operational characteristics, benefits, and limitations depending on the application context [9, 11–13].

Several factors govern the choice between charge and voltage amplification, among which the most significant are the intrinsic material properties of the

piezoelectric sensor, the electrical environment of deployment (including cable length and parasitic), and application-specific requirements related to bandwidth, mechanical resonance, and noise performance.

### *2.1.1 Piezoelectric constants and material properties*

One of the primary criteria for amplifier selection lies in the intrinsic piezoelectric constants—specifically, the piezoelectric charge constant ( $d_{ij}$ ) and voltage constant ( $g_{ij}$ ). These constants influence how the sensor converts mechanical input into electrical output and thereby dictate the optimal interfacing strategy.

Piezoceramic materials, such as lead zirconate titanate (PZT), exhibit a high charge constant ( $d_{ij}$ ) and a high dielectric constant, resulting in large capacitance per unit area. Due to these properties, piezoceramic sensors are best interfaced using charge amplifiers, which offer improved sensitivity and SNR in such configurations.

In contrast, piezopolymer materials such as polyvinylidene fluoride (PVDF) possess a high voltage constant ( $g_{ij}$ ) but low dielectric constant, typically yielding capacitance values in the range of 10 pF to 100 pF. In these cases, voltage amplifiers are more suitable as they leverage the high voltage sensitivity of PVDF to provide superior output signal levels and improved SNR performance compared to charge amplifiers [10].

### *2.1.2 Sensor electrical loading and cable effects*

The type and length of the cable linking the piezoelectric sensor to the front-end electronics plays a significant role in determining the appropriate amplifier selection. When using voltage amplifiers, the sensor operates in an open circuit configuration and is thus highly susceptible to capacitive loading from the connecting cable. This results in signal attenuation, especially when the cable length increases, since typical conductor-to-conductor capacitance can be in the order of 100 pF/m.

To mitigate this loading effect, charge amplifiers are traditionally used in systems where the sensor is located far from the signal-conditioning electronics. Operating the sensor in a virtual short-circuit configuration, the charge amplifier becomes largely insensitive to cable parasitic capacitance, preserving signal integrity even with extended cable runs [12, 14, 15]. However, this benefit comes at the cost of reduced sensitivity when applied to piezopolymer sensors due to their inherently low charge output.

Voltage amplifiers, on the other hand, are generally employed in applications where the sensor and amplifier are co-located or separated by very short cable lengths. In such scenarios, voltage-mode preamplifiers prevent signal degradation due to cable capacitance and help maintain a high SNR.

### *2.1.3 Impact on resonant frequency and bandwidth*

Another important but often overlooked consideration is the effect of the interfacing configuration on the sensor's mechanical stiffness and resonant behavior. A piezoelectric sensor presents different stiffness characteristics under open-circuit (voltage mode) and short-circuit (charge mode) conditions. These changes influence the sensor's resonant frequency and effective bandwidth [16]. As a result, voltage amplifiers are preferred in applications where consistency in resonant behavior is critical, such as in ultrasonic imaging or high-frequency dynamic pressure measurements, where even slight shifts in frequency response can affect measurement accuracy.

#### 2.1.4 Environmental noise and signal fidelity

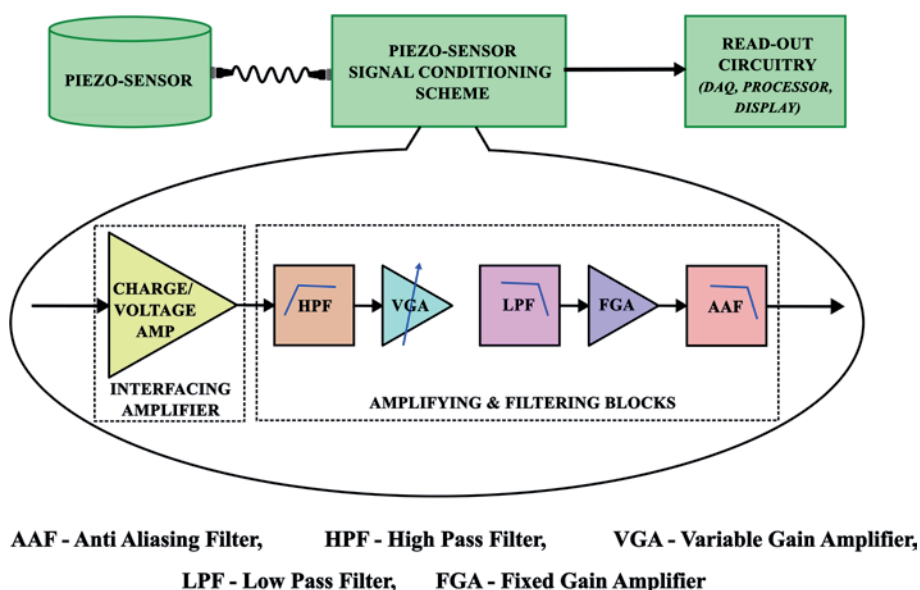
In electrically noisy environments, long cable runs not only introduce capacitive loading but also increase susceptibility to triboelectric noise and external electromagnetic interference. Charge amplifiers inherently reject cable-induced noise due to their low input impedance and feedback operation. However, modern advancements—such as the proposed enhanced voltage amplifier architecture with cable-capacitance compensation—offer new alternatives that maintain high sensitivity and SNR while remaining robust against environmental interference, especially for PVDF-based sensors.

## 2.2 Overview of interfacing schemes

**Figure 1** illustrates a typical signal conditioning architecture for piezoelectric sensors. This configuration comprises interfacing amplifier stages, both fixed and variable gain amplifiers, and filtering stages. Piezoelectric sensors require specialized interfacing circuits to convert the electrical signal generated in response to mechanical stimuli into a usable output. Due to their high output impedance and susceptibility to noise, the signal conditioning stage must be carefully designed to preserve signal fidelity, ensure adequate sensitivity, and reject interference. The two primary circuit topologies used for interfacing piezoelectric sensors are voltage amplifiers and charge amplifiers, each suited to specific types of piezoelectric materials and application conditions.

### 2.2.1 Voltage amplifier configuration

A voltage amplifier is the simplest and most direct method for interfacing a piezoelectric sensor. In this configuration, the sensor is connected to the



**Figure 1.**  
 Block diagram of a standard signal conditioning architecture for piezoelectric sensors.

high-input-impedance terminal of an operational amplifier. The amplifier senses the voltage generated across the sensor and outputs an amplified version of the signal. This approach is particularly well-suited for piezopolymer sensors such as PVDF, which have a high piezoelectric voltage constant ( $g_{ij}$ ) and inherently generate relatively large voltage signals even at low mechanical excitation.

Voltage amplifiers offer several advantages that make them particularly suitable for interfacing piezopolymer sensors such as PVDF. They effectively exploit the inherently high voltage sensitivity of PVDF, enabling strong signal generation even under low mechanical excitation. When the cable capacitance is minimal—as in short-distance or integrated setups—voltage amplifiers provide excellent signal-to-noise ratio (SNR), preserving the fidelity of the sensor output. Their compact design, ease of implementation, and low power consumption make them well-suited for embedded and portable sensor systems.

**Figure 2** illustrates the schematic of a voltage amplifier configuration used for interfacing a piezoelectric sensor *via* long cables. In voltage-mode operation, the interface cable introduces a parasitic capacitance  $C_c$  that, when combined with the sensor capacitance  $C_p$ , forms a capacitive voltage divider. This results in signal attenuation, as the output voltage becomes a function of the total capacitive loading. Furthermore, any mechanical movement or vibration of the cable can dynamically alter  $C_c$ , leading to fluctuations in the output signal and reduced measurement stability [17, 18].

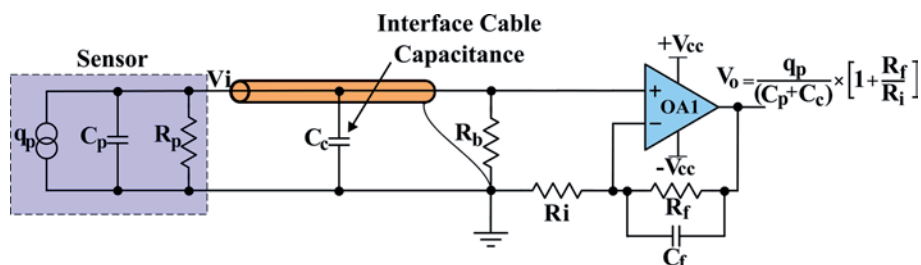
In this configuration, the resistor  $R_b$  provides a DC bias path to the non-inverting input of the operational amplifier  $OA_1$ . The feedback network comprising  $R_f$  and  $C_f$  defines the high-pass characteristics and sets the upper cutoff frequency of the amplifier. The combination of  $R_p$ ,  $R_b$ ,  $C_p$ , and  $C_c$  determines the lower cutoff frequency of the system [11, 12]. The output voltage  $V_o$  is given by Eq. (1):

$$V_o = \frac{q_p}{C_p + C_c} \times \left(1 + \frac{R_f}{R_i}\right) \quad (1)$$

where  $q_p$  is the charge generated by the piezoelectric sensor and  $R_i$  is the input resistor. This expression highlights the dependence of the output on both the sensor and cable capacitance, emphasizing the sensitivity of voltage-mode configurations to interface cable characteristics.

### 2.2.2 Charge amplifier configuration

Charge amplifiers are designed to convert the electric charge generated by the sensor into a voltage signal using a high-gain transimpedance amplifier with a feedback



**Figure 2.** Schematic of a voltage amplifier circuit for piezoelectric sensor interfacing.

capacitor. This topology essentially holds the input node at virtual ground, thereby making the system immune to the effects of cable parasitic capacitance and triboelectric interference [14, 15]. Charge amplifiers are highly effective in a variety of scenarios where signal integrity and noise immunity are critical. They are particularly well-suited for interfacing piezoceramic sensors, such as those based on PZT, which generate high charge outputs. Their ability to maintain performance regardless of cable length makes them ideal for applications where the sensor must be connected to the electronics *via* long cables. Additionally, in electrically noisy environments, charge amplifiers offer superior common-mode rejection and reduced susceptibility to interference, making them a robust choice for high-fidelity signal acquisition in harsh conditions.

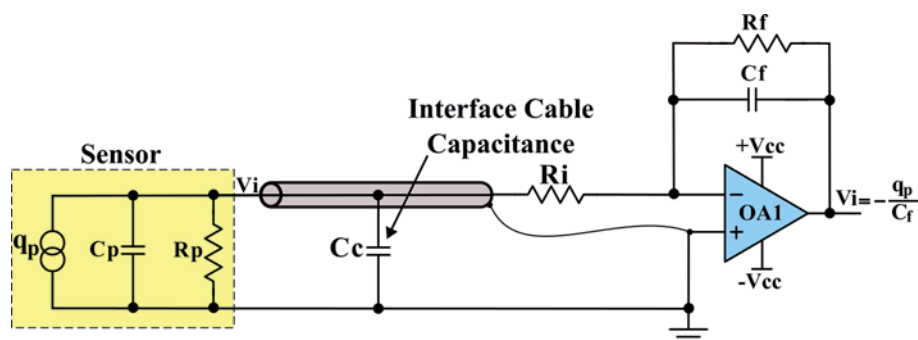
**Figure 3** illustrates the schematic diagram of a charge amplifier configuration commonly used for interfacing piezoelectric sensors, particularly in applications requiring long cable connections. In this setup, the piezoelectric sensor is modeled by a charge source  $q_p$  in parallel with its intrinsic capacitance  $C_p$  and leakage resistance  $R_p$ . The interface cable contributes an additional parasitic capacitance  $C_c$ , which is effectively rendered negligible by the virtual ground at the inverting input of the operational amplifier  $OA_1$ .

The core of the charge amplifier is the operational amplifier configured with a feedback capacitor  $C_f$  and resistor  $R_f$ . The feedback capacitor determines the charge-to-voltage gain of the amplifier, while the resistor  $R_f$  provides a discharge path for the capacitor, defining the low-frequency response and ensuring DC stability. The resistor  $R_i$ , connected between the cable and the amplifier input, may serve to limit inrush current or provide additional filtering in some designs.

One of the principal advantages of this configuration is its insensitivity to variations in sensor and cable capacitance. Since the input node is maintained at virtual ground, the total input impedance is extremely low, allowing the amplifier to integrate the total charge generated by the sensor without being affected by the parasitic capacitance of the cable. As a result, the output voltage  $V_o$  is governed solely by the relationship:

$$V_o = -\frac{q_p}{C_f} \quad (2)$$

This topology ensures that the amplifier delivers a consistent and reliable output irrespective of cable length or environmental noise sources, making it highly suitable for remote sensing applications, high impedance piezoceramic sensors, and electrically noisy operating conditions [5, 11, 12, 19, 20].



**Figure 3.**  
 Schematic of a charge amplifier circuit for piezoelectric sensor interfacing.

### 3. Enhanced voltage amplifier interfacing scheme

To overcome the trade-offs inherent in traditional voltage amplifier designs—particularly their susceptibility to cable parasitic—enhanced voltage amplifier schemes have been developed [21]. The novel circuit allows the system to retain the benefits of voltage-mode operation (i.e., high sensitivity for PVDF sensors) while mitigating the drawbacks associated with long cables. Enhanced voltage amplifiers have shown significant promise in extending the applicability of voltage-mode interfacing to real-world scenarios that were traditionally the domain of charge amplifiers. Applications such as distributed structural health monitoring, biomedical sensing, and underwater acoustics, where PVDF sensors and long cable runs are commonly used, stand to benefit greatly from this approach. The subsequent sections provide a detailed explanation of the design and functioning of the enhanced voltage amplifier scheme.

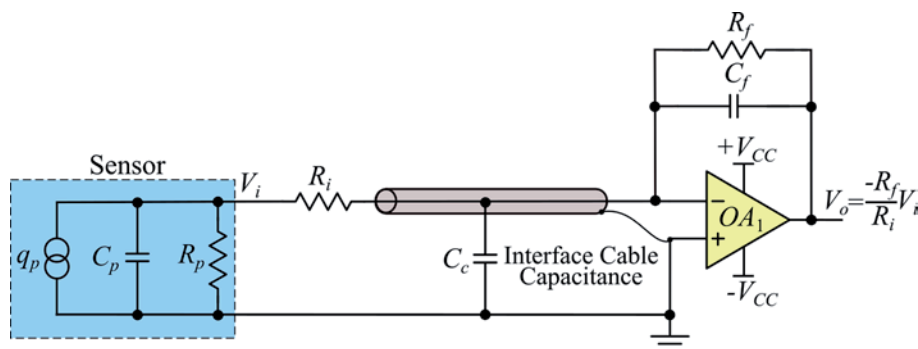
#### 3.1 Description and operation of circuit

**Figure 4** shows the circuit schematic of the enhanced voltage amplifier configuration for interfacing piezoelectric sensors through extended cable connections. The primary objective of this configuration is to mitigate the loading effect of the cable parasitic capacitance  $C_c$ , which typically degrades signal amplitude and fidelity in conventional voltage amplifier configurations.

In conventional voltage-mode amplifiers, the high output impedance of the piezoelectric sensor is influenced by the voltage division between the sensor capacitance  $C_p$  and the cable capacitance  $C_c$ , leading to a reduction in the amplitude of the measured signal. In contrast, charge amplifiers eliminate this loading by maintaining a virtual ground at the inverting input of the operational amplifier, effectively shorting the cable capacitance and preventing signal loss.

The enhanced voltage amplifier replicates this beneficial characteristic of the charge amplifier. By placing the interface cable between the two input terminals of the operational amplifier—specifically, between the far end of the input resistor  $R_i$  and the inverting terminal of the op-amp—the cable experiences a near-zero voltage differential during operation. This arrangement effectively presents a short circuit across the cable, nullifying the impact of  $C_c$  on signal transmission.

Moreover, the input resistor  $R_i$  is relocated to the sensor side, ensuring that the sensor sees only the high input impedance of the amplifier without the capacitive



**Figure 4.** Circuit diagram of enhanced voltage amplifier scheme (single-ended configuration) [20].

loading of the cable. The feedback components  $R_f$  and  $C_f$  define the gain and bandwidth of the amplifier, while the signal voltage  $V_o$  is governed by the expression:

$$V_o = -\frac{R_f}{R_i} \times V_i \quad (3)$$

This configuration enables voltage-mode amplification while maintaining immunity to cable-induced signal degradation, making it particularly advantageous for high-voltage-sensitivity sensors such as PVDF used in remote or distributed sensing applications.

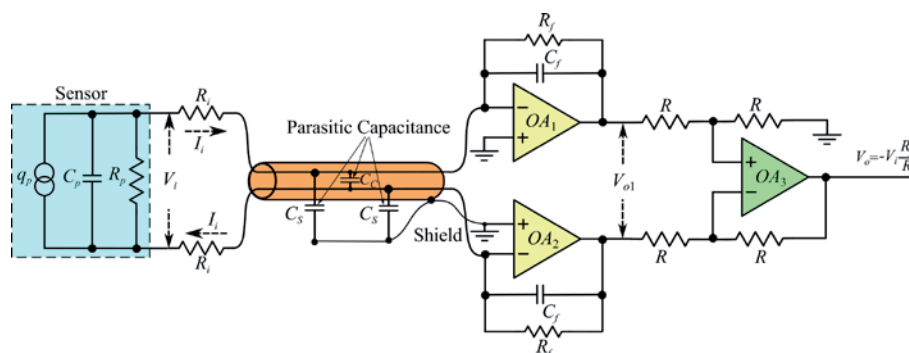
### 3.2 Differential scheme for the enhanced voltage amplifier

**Figure 5** illustrates the schematic of a differential enhanced voltage amplifier configuration designed to interface piezoelectric sensors through long cables while preserving high sensitivity and signal fidelity [22]. This scheme effectively addresses the limitations posed by cable parasitic capacitance and common-mode interference, both of which are critical challenges in remote sensing applications.

In this differential setup, the individual cores of a shielded dual-core cable are connected to the inverting inputs of two matched operational amplifiers, labeled  $OA_1$  and  $OA_2$ . The non-inverting inputs of both amplifiers are grounded and also connected to the cable shield, establishing a common reference. The sensor is modeled as a charge source  $q_p$  in parallel with its intrinsic capacitance  $C_p$  and leakage resistance  $R_p$ , and is interfaced to the input resistors  $R_i$ , which are relocated to the sensor end. The cable links the opposite ends of each  $R_i$  to the respective inverting inputs of operational amplifiers  $OA_1$  and  $OA_2$ .

During operation, the virtual ground established at the inverting inputs of  $OA_1$  and  $OA_2$  ensures that the parasitic capacitances associated with the cable—namely the conductor-to-ground capacitance  $C_s$  and the inter-conductor capacitance  $C_c$ —are effectively short-circuited. As a result, these parasitic do not draw any input current or distort the signal, thereby preventing sensor output loading. The entire current  $I_i$  generated by the sensor flows through the respective input resistors  $R_i$  and feedback resistors  $R_f$ , without being diverted into the cable parasitic. The voltage gain  $G$  of each inverting amplifier is given by Eq. (4):

$$G = \frac{R_f}{R_i} \quad (4)$$



**Figure 5.**  
 Schematic of the enhanced differential voltage amplifier used for piezoelectric sensor interfacing.

This gain remains unaffected by cable length or its associated capacitance, preserving the intended sensitivity performance of the system. The differential outputs of  $OA_1$  and  $OA_2$  are subsequently combined using a differential-to-single-ended converter implemented with operational amplifier  $OA_3$ . This final stage, configured for unity gain using matched resistors  $R$ , provides the single-ended output  $V_o$  corresponding to the differential signal  $V_{o1}$  generated across the inverting stages.

The differential structure of this enhanced amplifier not only suppresses the effects of cable capacitance but also significantly attenuates common-mode noise. As both conductors of the cable are maintained at virtual ground relative to the amplifier inputs, any common-mode noise acquired along the cable is efficiently directed to ground, resulting in an improved overall signal-to-noise ratio (SNR).

In summary, the proposed enhanced differential voltage amplifier enables voltage-mode interfacing to be effectively used with high voltage sensitivity piezoelectric sensors like PVDF, even when long cable connections are involved. It achieves this without compromising sensitivity or SNR, enabling reliable and high-performance signal acquisition in challenging environments traditionally served by charge amplifier configurations.

The influence of nonideal factors on the functionality and performance of the proposed scheme is examined. The enhanced differential voltage amplifier was simulated using LTspice to validate its resilience to parasitic cable capacitance and its robustness against common-mode interference (CMI). A prototype of the amplifier was developed, and experimental tests were carried out to assess its performance under conditions involving cable capacitance and CMI. The following sections present a detailed discussion of the error analysis, along with the results from both simulations and experimental evaluations.

#### **4. Error analysis of the enhanced differential voltage amplifier**

While the enhanced differential voltage amplifier configuration shown in **Figure 5** has been conceptually discussed under ideal conditions, real-world circuit implementations are invariably influenced by several nonidealities [22]. These include both active component limitations and passive parasitic such as input bias characteristics of operational amplifiers, resistor mismatches, thermal effects, and the influence of parasitic capacitances. This section examines the qualitative effects of these nonidealities on the performance of the proposed scheme.

##### **4.1 Effect of DC characteristics of operational amplifiers**

The output of piezoelectric sensors is typically AC in nature. As a result, the DC characteristics of operational amplifiers  $OA_1$ ,  $OA_2$ , and  $OA_3$ —such as input bias current, input offset voltage, input offset current, and thermal drift—have a negligible effect on the overall performance of the proposed scheme. Any DC offset that may arise at the output can be efficiently removed through AC coupling, either between amplifier stages or at the output, thereby preserving signal integrity.

##### **4.2 Effect of AC characteristics of operational amplifiers**

The AC performance of the amplifier is influenced by the internal parasitic capacitances of the operational amplifiers as well as the external parasitic cable capacitances  $C_c$  and  $C_s$ . These parasitic elements influence the amplifier's frequency response, slew

rate, and overall stability. The parasitic cable capacitance appearing at the inverting inputs of OA<sub>1</sub> and OA<sub>2</sub> introduces an extra pole in the system. When combined with the feedback resistance R<sub>f</sub>, this additional pole leads to a reduction in both the gain margin and phase margin.

The modified feedback factor  $\beta$  for the inverting amplifier, considering the effect of cable capacitance, is given by Eq. (5):

$$\beta = \frac{1}{\left(1 + \frac{R_f}{R_i}\right) + s R_f C_c} \quad (5)$$

This additional pole also shifts the lower cutoff frequency of the amplifier, thereby altering its bandwidth and affecting the overall frequency response [23].

#### 4.3 Effect of gain-setting resistor mismatches

The ideal differential gain of the proposed scheme is defined as  $G = R_f / R_i$ , assuming perfectly matched gain-setting resistors for OA<sub>1</sub> and OA<sub>2</sub>. In practice, resistor mismatches result in unequal gains for the two differential paths. Let  $G_1 = G(1 + \delta)$  and  $G_2 = G(1 - \delta)$ , where  $\delta$  is the mismatch factor. This imbalance introduces a finite common-mode gain, approximately given by Eq. (6):

$$\text{Common mode gain} \approx 2\delta \frac{G}{1 + G} \quad (6)$$

However, this contribution is typically much smaller than the differential gain and does not significantly affect overall output accuracy, provided resistor tolerances are kept within acceptable limits.

#### 4.4 Effect of common-mode interference

A key benefit of the differential architecture depicted in **Figure 5** is its inherent immunity to common-mode interference. During normal operation, the inverting inputs of OA<sub>1</sub> and OA<sub>2</sub> are maintained at virtual ground, which effectively suppresses any common-mode voltage induced along the shielded dual-core cable. As a result, electromagnetic interference coupled equally into both conductors is rejected. Furthermore, mismatches in the gain-setting resistors do not significantly degrade this immunity, ensuring high common-mode rejection ratio (CMRR) and improved signal-to-noise ratio (SNR).

#### 4.5 Effect of parasitic cable capacitance

Although the cable parasitic capacitances C<sub>c</sub> and C<sub>s</sub> do not load the sensor directly—due to the virtual ground condition at the amplifier inputs—they still influence the stability and dynamic behavior of the circuit. These parasitic introduce additional poles into the feedback network, shifting the lower cutoff frequency and potentially degrading phase margin and system stability. While the sensitivity performance remains unaffected, careful consideration of parasitic capacitances is essential in high-frequency applications to prevent oscillations and ensure consistent bandwidth performance.

## 5. Experimental setup and results

The performance of the enhanced differential voltage amplifier scheme was evaluated through both simulation and experimental validation [22]. LTspice simulations were performed to evaluate the amplifier's robustness against parasitic cable capacitance and its ability to reject common-mode interference (CMI). Prototype implementation was subsequently developed to verify these characteristics under practical conditions. The results of these studies are presented below.

### 5.1 Simulation and results

Two amplifier configurations were simulated using LTspice: a conventional differential voltage amplifier and the enhanced differential voltage amplifier. Both setups incorporated cable parasitic capacitance models to assess their impact. The goal was to validate the proposed design's ability to maintain high sensitivity and reject CMI even in the presence of long interconnects.

The piezoelectric sensor was modeled using a voltage source  $V_s$ , a series resistance  $R_s = 100\ \Omega$ , a sensor capacitance  $C_p = 1000\ \text{pF}$ , and a parallel leakage resistance  $R_p = 10\ \text{M}\Omega$ . The dual-core cable was represented by a series resistance  $R_c = 2.5\ \Omega$ , a parasitic inter-core capacitance  $C_c = 800\ \text{pF}$ , and a shield-to-core capacitance  $C_s = 1000\ \text{pF}$ . The amplifier stages ( $OA_1$ ,  $OA_2$ , and  $OA_3$ ) were implemented using LT6234 op-amp models—chosen for their low noise, low bias current, and rail-to-rail performance. The amplifier was designed for a nominal gain of  $-10$ .

#### 5.1.1 Effect of parasitic cable capacitance

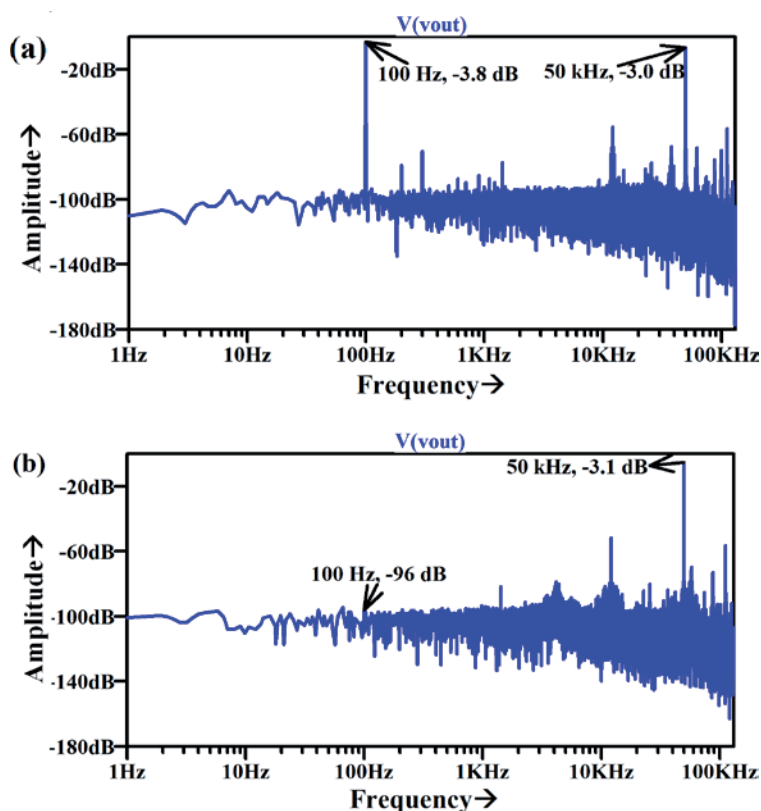
The source signal  $V_s$  was configured as a 50 kHz sine wave with a 0.2 V peak-to-peak amplitude. In the case of the conventional differential amplifier, a significant voltage drop was observed at the amplifier input: from 0.2 V pp. at the source to 0.1 V pp. ( $-6\ \text{dB}$ ) at the amplifier input, indicating signal attenuation due to cable loading. Consequently, the output signal was also attenuated, resulting in reduced sensitivity.

In contrast, the enhanced differential voltage amplifier maintained the full input signal amplitude, with no observed attenuation due to the cable. The measured gain was  $-10.02$ , closely matching the theoretical value. These results confirm that the proposed scheme effectively mitigates the impact of parasitic cable capacitance, preserving sensitivity even with long cable lengths.

#### 5.1.2 Effect of common-mode interference

To evaluate the immunity of the proposed amplifier to common-mode interference, a 1 kV, 100 Hz AC source was applied equally to both conductors of the modeled cable through high-value resistors ( $100\ \text{M}\Omega$ ), simulating a strong CMI condition. The fast Fourier transform (FFT) of the amplifier output was analyzed using the LTspice scope function. The output spectrum is shown in **Figure 6(a)**.

For the proposed differential scheme, the FFT revealed that the amplitude of the 100 Hz common-mode component was  $-96\ \text{dB}$ , demonstrating exceptional rejection of the interference signal. For comparison, the same CMI test was repeated using the single-ended voltage amplifier configuration (as shown in **Figure 4**). In this case, the 100 Hz component was only  $-3.8\ \text{dB}$  relative to the signal amplitude, as shown



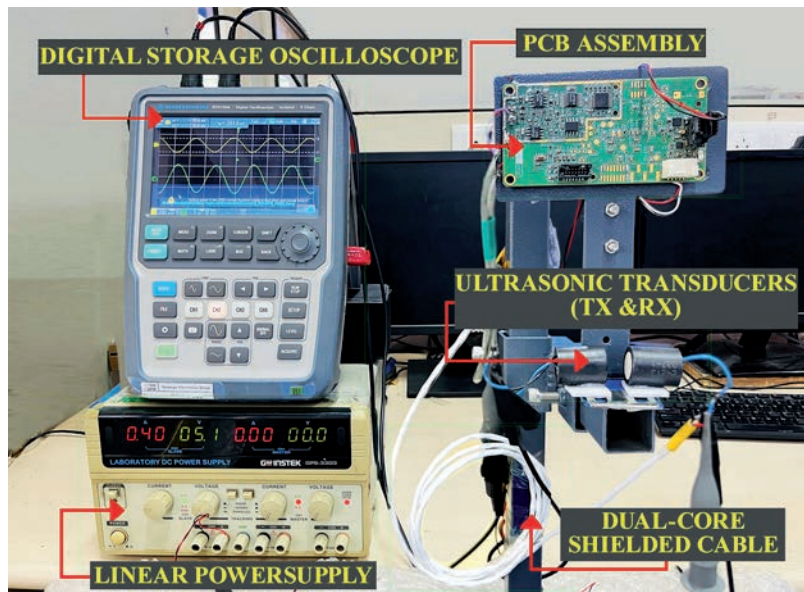
**Figure 6.**  
 Simulated fast Fourier transform (FFT) output plot for (a) the single-ended configuration of the proposed voltage amplifier, and (b) the proposed differential voltage amplifier in the presence of common-mode interference (CMI).

in **Figure 6(b)**. This corresponds to a CMI rejection improvement of approximately 92.2 dB in the differential configuration.

These findings clearly demonstrate the superior common-mode rejection capability of the enhanced differential voltage amplifier. The significant reduction in the output spectral content of the interference confirms that the virtual ground maintained at the inverting inputs of  $OA_1$  and  $OA_2$  effectively suppresses common-mode signals. This improved noise immunity directly contributes to enhanced signal-to-noise ratio (SNR), making the configuration highly suitable for applications involving long cables and electrically noisy environments.

## 5.2 Hardware implementation and experimental validation

A hardware prototype of the enhanced differential voltage amplifier was developed and experimentally validated to evaluate its effectiveness in real-world conditions. The experimental setup and the PCB implementation are shown in **Figures 7** and **8**, respectively. The evaluation focused on two primary aspects: (i) sensitivity degradation due to parasitic cable capacitance and (ii) immunity to common-mode interference (CMI). A comparative analysis was also carried out between the proposed differential scheme and a conventional single-ended voltage amplifier configuration.

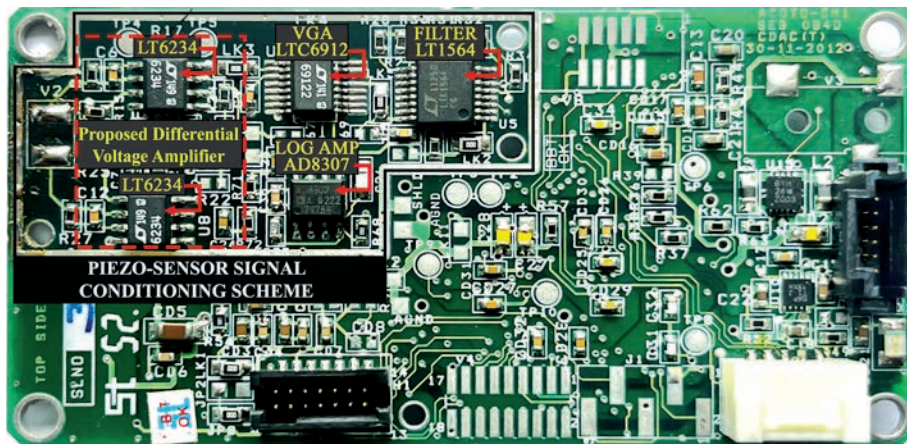


**Figure 7.**  
Experimental setup used to evaluate the performance of the proposed differential amplifier configurations.

### 5.2.1 Experimental setup

The experimental setup comprises ultrasonic air-coupled transducers (Model: AT200, AIRMAR), a dual  $\pm 5$  V benchtop linear power supply, a digital storage oscilloscope (DSO), and a custom-designed signal conditioning PCB (**Figure 8**).

The LT6234, a low-noise, low-bias current, wide-bandwidth (60 MHz) operational amplifier capable of single-supply operation, is employed in the differential amplifier configuration. A differential amplifier gain of  $-10$  was configured using matched resistors ( $5.1$  k $\Omega$  input,  $51$  k $\Omega$  feedback). The input resistors were strategically placed at the sensor end, and a 5-meter dual-core shielded cable was routed between the



**Figure 8.**  
Prototype PCB implementation of the enhanced differential amplifier design.

resistors and the inverting inputs of OA<sub>1</sub> and OA<sub>2</sub>. The sensor capacitance (510 pF) was measured using an LCR meter.

The DDS-based transmitter circuit (using AD9833) on the PCB generates an 18 V peak-to-peak continuous sine wave at 200 kHz to excite the transmitting transducer. The received signal from the closely positioned receiver transducer is then amplified by the differential voltage amplifier. A photograph of the piezosensor signal conditioning PCB highlighting key functional blocks—such as the proposed amplifier, VGA, filter, and log amp—is provided in **Figure 8**.

The PCB assembly operates using a dual  $\pm 5$  V DC linear power supply. A digital storage oscilloscope (DSO) is used to monitor the input and output voltages of both the standard and enhanced differential voltage amplifiers, with and without the presence of the connecting cable.

### 5.3 Experimental results

The impact of cable parasitic capacitance and common-mode interference on the performance of both the standard and enhanced voltage amplifiers has been assessed, and a comparative evaluation of their performance has been carried out.

#### 5.3.1 Effect of parasitic cable capacitance

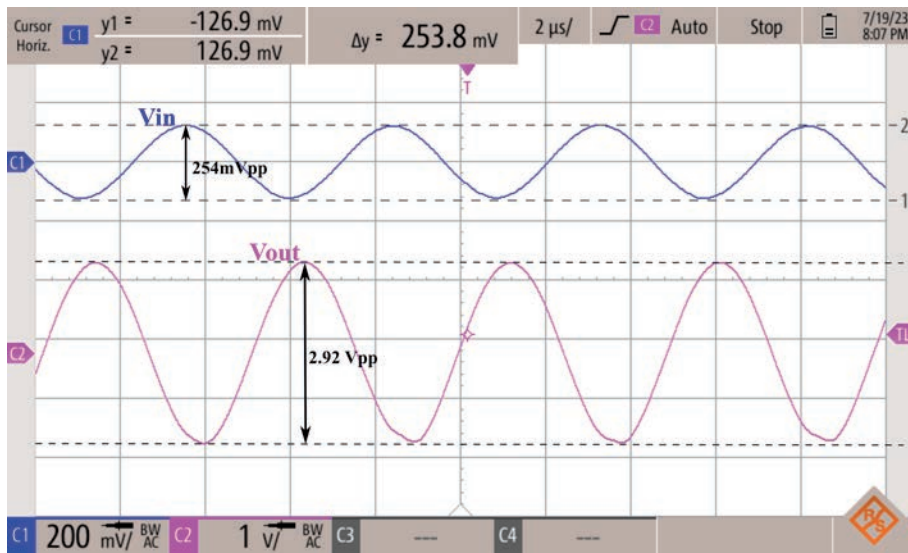
The impact of cable parasitic capacitance was first evaluated using a conventional differential voltage amplifier. When no cable was connected, the measured input and output voltages were 261 mV peak-to-peak and 2.558 V peak-to-peak, respectively. When a 5-meter dual-core shielded cable was inserted between the transducer and the amplifier input, the input voltage reduced to 177 mV pp. and the output voltage dropped to 1.73 V pp., corresponding to an approximate attenuation of  $-3.4$  dB. This attenuation is attributed to the capacitive loading effect, which degrades sensitivity in the standard configuration.

To counter this, the input resistors were moved to the sensor side, and the cable was placed between the resistors and the inverting terminals of OA<sub>1</sub> and OA<sub>2</sub>, realizing the proposed differential scheme. As shown in **Figure 9**, the input voltage experienced a slight drop from 261 mV pp. (without the cable) to 254 mV pp. (with the cable), while the output voltage showed a minor increase from 2.558 V pp. to 2.92 V pp. This increase is a result of gain peaking caused by the interaction of cable parasitic with the amplifier bandwidth, indicating a potential for instability. Despite these variations, the observed gain remained close to the intended design value (approximately  $-11.5$ ), indicating that the proposed scheme maintains its sensitivity and is largely unaffected by cable parasitic capacitance.

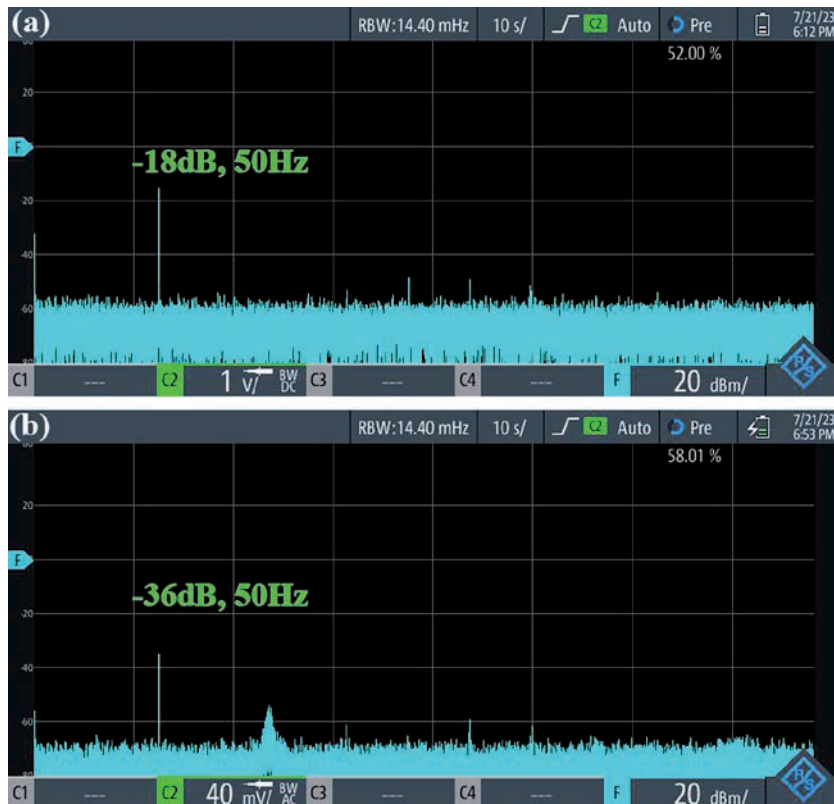
#### 5.3.2 Effect of common-mode interference

Experimental validation was carried out to demonstrate the differential amplifier's resilience to common-mode interference. The experiment was conducted in an environment with significant 50 Hz electromagnetic interference, primarily caused by heavy machinery operating in proximity to the test setup. FFT plots of the amplifier output for both configurations—single-ended and differential—were recorded using the DSO and are shown in **Figure 10**.

In the single-ended amplifier setup (**Figure 10a**), the 50 Hz interference component appeared at  $-18$  dB, significantly affecting the output signal. In contrast, the proposed



**Figure 9.**  
DSO waveform capture shows input and output signals of the enhanced differential voltage amplifier.



**Figure 10.**  
Frequency spectra of output voltage for (a) single-ended amplifier, and (b) proposed differential amplifier under common-mode interference.

differential amplifier configuration (**Figure 10b**) demonstrated a marked improvement, with the 50 Hz interference reduced to  $-36$  dB—a suppression of 18 dB. This confirms the superior CMI rejection capability of the differential scheme, owing to the virtual ground maintained at the inverting terminals and the symmetric signal path architecture.

## 6. Conclusions

This chapter presented an enhanced differential voltage amplifier scheme for interfacing piezoelectric sensors. The proposed design effectively addresses two major challenges in piezosensor signal conditioning: the degradation of sensitivity due to parasitic cable capacitance and the impact of common-mode interference (CMI), both of which are critical in applications involving long cable interconnects between sensors and front-end electronics.

The proposed amplifier architecture repositions the input resistors at the sensor end and places the dual-core shielded cable between the sensor and the inverting inputs of the amplifier. This configuration ensures a virtual ground across the cable parasitic during operation, thereby eliminating signal loading effects and significantly suppressing common-mode noise. The system retains the high voltage sensitivity advantages of piezopolymer sensors, such as PVDF, which are otherwise compromised when using charge amplifiers in long-cable applications.

The functionality of the proposed differential voltage amplifier was validated through comprehensive simulations using LTspice, followed by hardware implementation and experimental testing. The experimental results confirmed that the design is insensitive to cable parasitic capacitance and exhibits excellent immunity to common-mode interference. Notably, the sensitivity of the proposed differential voltage amplifier improved by 4.55 dB compared to a standard differential amplifier when a 5-meter dual-core cable was used to interface an AT200 piezoelectric air transducer. Furthermore, the proposed scheme demonstrated an 18 dB improvement in CMI rejection over a single-ended configuration, thereby significantly enhancing the signal-to-noise ratio (SNR) performance.

Currently, there are limited interfacing techniques that allow the use of voltage amplifiers—rather than charge amplifiers—for piezopolymer sensor applications involving long cables. The proposed solution fills this gap, enabling the use of high-voltage-sensitivity piezopolymer sensors without compromising performance, even in harsh and noise-prone environments.

The simplicity of the proposed scheme, combined with its proven performance, makes it an attractive alternative to traditional charge amplifiers, particularly for applications such as

- Underwater sonar and acoustic sensing.
- Structural health monitoring in infrastructure and aerospace.
- Biomedical instrumentation and biosensor systems.
- Medical ultrasound imaging.

The comparative analysis of the proposed amplifier against conventional configurations is summarized in **Table 1**, demonstrating its superior performance in sensitivity, SNR, and noise immunity. In conclusion, the enhanced differential voltage

| <b>Amplifier configuration</b>                            | <b>Pros</b>                                                                                                                                                                                                                                                                                            | <b>Cons</b>                                                                                                                                                                                                                                                                                  |
|-----------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Voltage amplifier [9] (Preamplifier)                      | <ul style="list-style-type: none"> <li>• High sensitivity and SNR</li> <li>• Insensitive to cable parasitic capacitance</li> <li>• Drives long cables</li> <li>• Immune to external interference</li> </ul>                                                                                            | <ul style="list-style-type: none"> <li>• Not ideal for high-temperature environments</li> <li>• Reliability concerns from embedded electronics</li> <li>• Bulky sensor design due to internal electronics</li> <li>• Additional power cable needed for amplifier operation</li> </ul>        |
| Voltage amplifier [12] (With cable)                       | <ul style="list-style-type: none"> <li>• Remote electronics for high-temp applications</li> <li>• Adjustable gain</li> <li>• Minimal impact on stability and bandwidth</li> </ul>                                                                                                                      | <ul style="list-style-type: none"> <li>• Sensitive to cable parasitic capacitance</li> <li>• Sensitive to triboelectric effect</li> <li>• Poor sensitivity</li> <li>• Susceptible to interference</li> </ul>                                                                                 |
| Charge amplifier [12, 20, 24] (With cable)                | <ul style="list-style-type: none"> <li>• High sensitivity</li> <li>• Insensitive to cable parasitic capacitance</li> <li>• Insensitive to triboelectric effect</li> <li>• Simple configuration</li> </ul>                                                                                              | <ul style="list-style-type: none"> <li>• Incompatible with low-capacitance piezo sensors</li> <li>• Alters resonant frequency and bandwidth</li> <li>• Bandwidth and stability impacted by cable capacitance</li> <li>• Requires bleeder resistor, increasing noise gain [18, 24]</li> </ul> |
| Enhanced voltage amplifier [21] (With cable)              | <ul style="list-style-type: none"> <li>• Insensitive to cable parasitic capacitance</li> <li>• Insensitive to triboelectric effect</li> <li>• High sensitivity</li> <li>• Variable gain amplification possible</li> <li>• Compatible for low capacitance piezo sensors</li> </ul>                      | <ul style="list-style-type: none"> <li>• Cable capacitance impacts signal integrity</li> <li>• Higher noise gain due to cable capacitance [18]</li> <li>• Input resistance influences bandwidth</li> <li>• Susceptibility to common-mode interference</li> </ul>                             |
| Differential voltage amplifier [23, 24] (With cable)      | <ul style="list-style-type: none"> <li>• Common mode signal rejection</li> <li>• Improved SNR performance</li> <li>• High linearity</li> <li>• High signal integrity</li> <li>• Increased signal swing</li> </ul>                                                                                      | <ul style="list-style-type: none"> <li>• Sensitive to cable parasitic capacitance</li> <li>• Sensitive to triboelectric effect</li> <li>• Poor sensitivity due to cable loading</li> <li>• Matching of components required</li> </ul>                                                        |
| Enhanced differential voltage amplifier [22] (With cable) | <ul style="list-style-type: none"> <li>• High immunity to common mode signals</li> <li>• Insensitive to cable parasitic capacitance</li> <li>• Insensitive to triboelectric effect</li> <li>• High sensitivity and SNR</li> <li>• Compatible for high voltage constant piezopolymer sensors</li> </ul> | <ul style="list-style-type: none"> <li>• The cable capacitance affects the bandwidth and stability</li> <li>• Increased noise gains due to cable capacitance [18]</li> <li>• Input resistance affects the bandwidth</li> <li>• Matching of components required</li> </ul>                    |

**Table 1.**  
*Comparison between different amplifier configurations.*

amplifier provides a robust and efficient solution for interfacing piezopolymer-based piezoelectric sensors in long-cable scenarios, expanding the applicability of voltage-mode signal conditioning in advanced sensor systems.

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*Edited by Massimo Mariello*

*A Booklet on Piezoelectricity - Essential Principles, Modeling and Emerging Technologies for Energy Harvesting and Sensing* offers a concise journey into one of the most fascinating properties of matter: the conversion between mechanical force and electrical charge. In compact chapters, this book introduces readers to the essential principles, materials, and applications of piezoelectricity — from modeling crystal lattices to the device engineering of sensors and energy-harvesting systems. Written in a clear, accessible style, it bridges physics, materials science, and engineering, illustrating how tiny atomic-level deformations generate signals that power technologies shaping modern life. The book's concise structure reflects its essence: to distill the beauty and utility of piezoelectric phenomena without overwhelming the reader, while instilling curiosity to explore the world of piezoelectricity and its many applications, intersections, and developments. Each chapter offers a logical glimpse, from the historical discovery of the effect by the Curie brothers to today's flexible, soft piezoelectric materials driving advances in robotics, medicine, and human-machine interfaces. Edited by Dr. Massimo Mariello, this book draws on years of interdisciplinary research in bioelectronics, materials engineering, and biomedical science, combining straightforward explanations with an appreciation for scientific creativity. Whether read by a student, a scientist, or a curious mind, this small book opens a gateway to the powerful simplicity of piezoelectricity, where pressure becomes potential, and the smallest vibrations reveal the profound connection between mechanics and electricity. Designed to be read in just a few hours, it leaves the reader with lasting curiosity and a sense of the vast scientific landscape beyond these concise chapters.

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