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## **DESIGN AND DEVELOPMENT OF 3D-PRINTED PIEZOELECTRIC SENSORS**

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### **ABSTRACT :**

Additive Manufacturing (AM), or 3D printing, is gaining attention for creating complex, personalized parts from polymers, ceramics, and metals without molds or machining. Future advancements will focus on integrating smart materials like sensors and actuators. This thesis explores 3D printing a fully dense, biphasic, lead-free piezoelectric sensor with low-volume fillers, analyzing how nozzle size and temperature affect the final product. It also investigates how various printing settings impact sensor performance. Additionally, the thesis develops a design for in-situ fabrication of 1-3 connectivity piezoelectric composites, examining how printing speed, electrode spacing, and temperature affect performance. Recommendations for optimizing settings and setup adjustments are provided.

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**Keywords:** Additive Manufacturing (AM), Three Dimensional (3D), Piezoelectric Effect (PE), Piezoelectric Sensor (PS)

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### **Introduction :**

Recent advancements in Additive Manufacturing (AM) technologies have opened new avenues for creating complex structures with high precision, enabling the fabrication of piezoelectric sensors with enhanced design flexibility and customization. The growing need for innovative sensing solutions and cost-effective, scalable production methods highlights the importance of utilizing 3D printing in sensor manufacturing. 3D printing's capability for rapid prototyping and iterative design allows for quick customization and optimization of sensors for specific applications. Additionally, exploring new materials and composites further boosts the potential for developing high-performance sensors. This thesis aims to leverage 3D printing technology to meet industry needs, drive sensor innovation, and advance additive manufacturing in sensing applications.

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### **Nomenclature**

**3D Printing (3DP):** An additive manufacturing process that creates three-dimensional objects by successively layering material.

**Piezoelectric Sensor (PS):** A device that converts mechanical energy (such as pressure, force, or vibration) into electrical energy using the piezoelectric effect.

**Sensitivity:** The ratio of a sensor's output (voltage or current) to the applied input (force or pressure).

**Stability:** The capacity of a sensor to maintain its characteristics over time and under varying environmental conditions.

**Repeatability:** The ability of a sensor to produce consistent output for repeated measurements under the same conditions.

**Frequency Response (FR):** The range of frequencies over which the sensor can accurately measure dynamic signals.

**Linearity:** The degree to which the sensor's output is directly proportional to the input.

**Hysteresis:** The difference in sensor output for the same input when the input is increasing compared to when it is decreasing.

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### **Requirement Specification**

3D Printed Piezoelectric Sensor is a revolutionary approach in sensor technology. This innovative method enables the creation of sensors with enhanced customization, scalability, and efficiency compared to traditional manufacturing techniques. By leveraging the capabilities of additive manufacturing, engineers can produce intricate sensor designs, optimize material properties, and tailor sensors for specific applications. This shortens development cycles, reduces production costs, and opens doors to new possibilities in sensor design. In this presentation, we'll delve into the principles, advantages, and applications of 3D printed piezoelectric sensors, exploring their role in shaping the future of sensing technology.

What Are Piezoelectric Sensors?

Piezoelectric sensors are devices that change mechanical energy into electrical signals and vice versa. They work using the piezoelectric effect, which happens in materials like quartz, ceramics, and certain crystals. When these materials are stressed mechanically, they produce an electric charge. Conversely, applying an electric field to them causes them to change shape.

This ability to switch between mechanical and electrical forms allows piezoelectric sensors to measure things like pressure, force, acceleration, and vibration. These sensors are used in many industries, including automotive, aerospace, healthcare, and consumer electronics, for tasks such as detecting impacts, measuring pressure, and harvesting energy.

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## **Development of Piezoelectric Sensor :**

### ***3.1 Introduction***

Materials science has always been important for technological progress. It has improved existing methods and explored new ways to create and use materials. By understanding how materials work, we have been able to build complex devices that meet our needs and broaden our knowledge.

One key group of materials is piezoelectric-ferroelectrics, studied since the 19th century. These materials can change mechanical energy into electrical energy and vice versa. This is called the piezoelectric effect, and it occurs in various materials like single crystals, piezoelectric ceramics, polymers, and composites.

Because of their strong performance and versatility, piezoelectric materials are used in many areas. They are found in sensors, actuators, microphones, sonar devices, beepers, buzzers, gas lighters, and energy harvesters. These materials are used across many industries, including aerospace, automotive, biomedical, and electronics.

In this section, we will look at printed piezoelectric sensors. We will first examine them using Thick-Film technology (TF) and then explore Additive Manufacturing (AM), Solid Freeform Fabrication (SFF), and Rapid Prototyping (RP) technologies. These new manufacturing methods have opened up more possibilities for using piezoelectric sensors.

### ***3.2 Components and Classification***

#### **i. THICK FILMS**

Thick-Film technology (TF) is one of the most cited divisions of microelectronics that despite the fact that, it was introduced during the late 60s, it continues to have a significant role in today's technologic advancement. By definition TF regards films of thickness between 1 to 100  $\mu\text{m}$  printed onto a ceramic, insulating or semiconducting substrate, consisting of a ink/paste mixture, which contains a number of different ingredients such as filler (e.g. conductive, resistive, dielectric etc.), a binder that plays the role of bonding agent (e.g. glass, metal-oxide etc.), dispersants and solvent. Moreover, due to their high level of automation, low-cost and good performance, thick-films present a vast variety of applications from everyday use devices, such as televisions and telephones, to more technically advanced ones as missile guidance systems and automotive/aerospace electronics.

#### **ii. SCREEN PRINTING**

One method of categorizing the various printing techniques utilized in the TF field is by distinguishing between those that utilize a mask during the printing process and those that do not. Screen printing falls under the category of mask-using techniques, as it relies on a screen with a stencil (composed of a screen-frame and ink/paste blocking layer) that features the negative image of the printing surface (see Fig.2.1.). The ink/paste, typically exhibiting viscous and thixotropic behavior, is applied to the stencil and pressed through the mesh apertures to be printed onto the substrate in specific patterns.

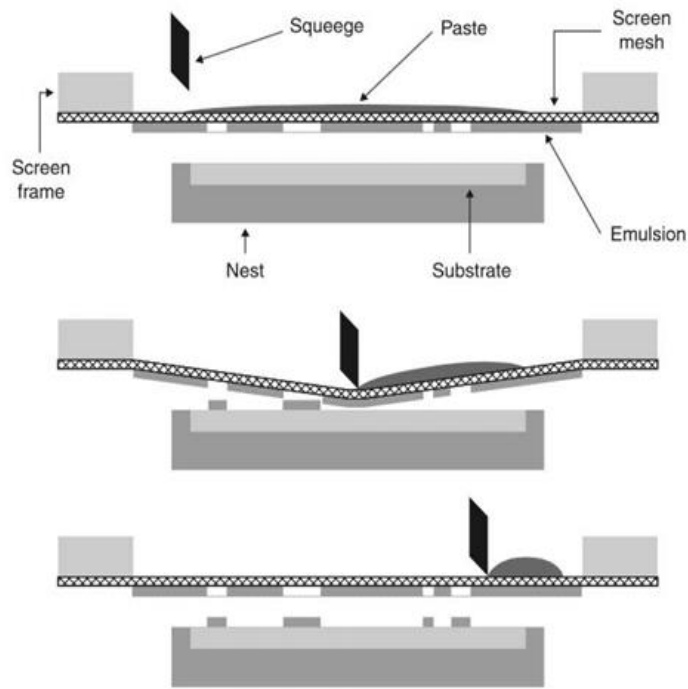


Figure 2.1: Schematic representation of the screen printing process

iii. INK-JET PRINTING

The initial emergence of this specific printing technology occurred around the mid-1970s. As illustrated in Figure 2.2, it stands out as one of the most advanced techniques in the Direct-Writing field. Ink-jet printing distinguishes itself from other printing methods by its capability to print on various substrate materials without the need for a guided mask. This results in significant cost reductions, either through minimal material consumption or reduced production waste. Additionally, the division of ink-jet printing into Continuous and Drop-on-Demand operational types offers further advantages in terms of processing ease and the range of features that can be printed. The Continuous ink-jet printing method, depicted in Figure 2.3 a, relies on the unique behavior of a continuous liquid jet to break up and flow under surface tension when excited in a specific manner, resulting in a continuous stream of ink. On the other hand, the Drop-on-Demand ink-jet printing method (shown in Figure 2.3 b) involves applying pulsed pressure behind the ink vessel to generate ink droplets that are then released through the nozzle onto the substrate.

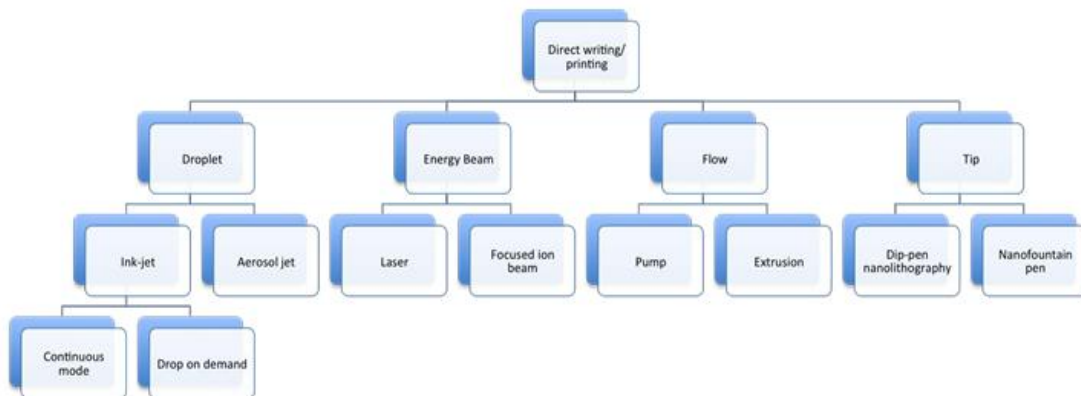
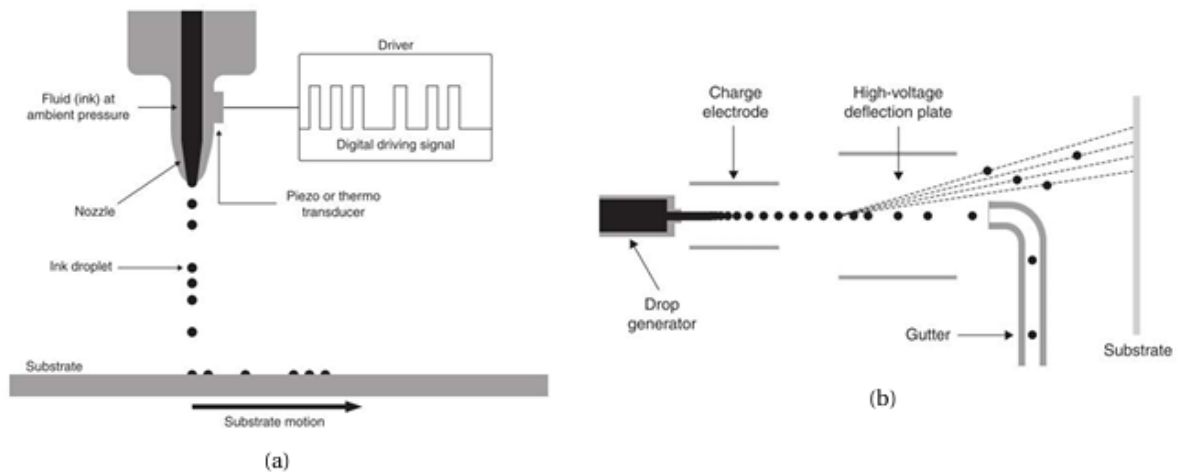


Figure2.2: A schematic representation of the classification of the various Direct Writing techniques

Two different methods are currently being explored for the production of printed ceramic parts using ink-jet printing. The first method involves a combination of ceramic particles and wax, which solidifies when it comes into contact with a substrate that is at a lower temperature than the hot paste. The second method involves the use of a solvent that evaporates after deposition. Both of these techniques are being studied for their potential in creating various piezoelectric products, including thick- and thin-films, as well as different types of transducers, such as piezoelectric micro-machined ultrasonic transducers (pMUT), in various shapes and sizes.

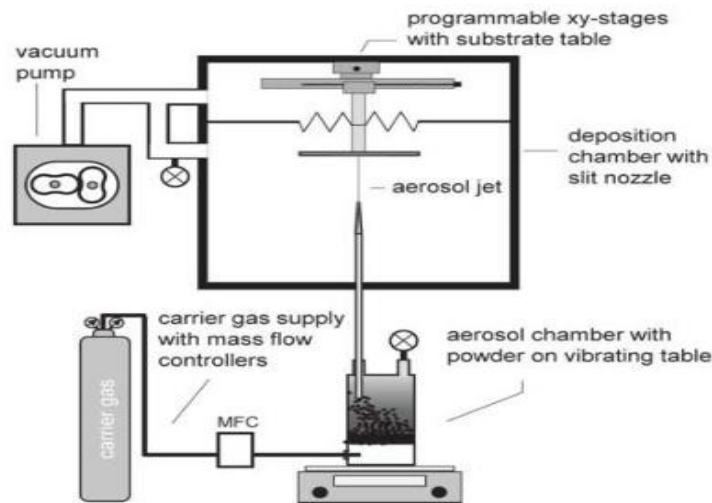


**Figure 2.3: Schematic representation of the two types of ink-jet printing: (a) Drop-on-Demand approach, (b) Continuous approach**

#### iv. AEROSOL JET DEPOSITION

In the mid-1980s, researchers began exploring gas deposition methods, which were later refined in the 1990s to create thick films from metallic or ceramic particles. More recently, aerosol deposition (AD) has gained interest because it can produce dense, pore-free coatings and films on various surfaces without needing additional sintering.

The AD process works by using a gas carrier to move through a vibrating chamber filled with liquid or powder. This creates an aerosol. The aerosol is then pushed by pressure into a deposition chamber, where it is sprayed onto the target surface.



**Figure 2.4: A schematic representation of the aerosol deposition technique**

Numerous materials have been investigated as pastes for the AD method, as indicated by research studies. In the late 90s, Akedo et al. conducted the first investigation on piezoelectric applications using PZT filler. Their study proposed that the post-annealing process directly affects the dielectric properties of the film due to grain growth mechanism occurring in the system. This research by Akedo paved the way for further studies on the AD of piezoelectric ceramics, focusing on post-annealing and grain size analysis.

### Development of Printed 0-3 Piezoelectric sensors

Based on the research guidelines that were set in section 1.3 for the development of a fully dense, two-phase random piezoelectric sensor through FDM technology, in this chapter we are going to commence by briefly exhibiting all the processing and characterization techniques that were utilized in the experimental phase. Moreover, a section of the produced results with an elaborate discussion on them will be exhibited. Finally, conclusions and recommendations based on the outcomes of the project will be presented.

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## Proposed System Development of Printed 1-3 Aligned Piezoelectric sensors

In this we explore the development of a printed aligned piezoelectric sensor. In the first section the experimental steps that were carried out will be presented, along with the setup that was utilized for this purpose. Subsequently, an analysis of the obtained DEP results and of the parameters the influence the in-situ printing/DEP process will be discussed. Finally, our conclusions and recommendations for future work on the in-situ process will be presented.

### 5.1 EXPERIMENTAL

All the experimental setups that were used, along with our concept design for the implementation of an in situ printing/DEP process will be addressed in this section. The first part considers a brief presentation of the dielectrophoretic alignment setup for high temperatures, which was utilised in this project. Then, in the second part an introduction of our concept design for the development of an in-situ printing/DEP process will be provided.

#### 5.2.1. HIGH TEMPERATURE DIELECTROPHORESIS SETUP

In the initial experiments a high temperature dielectrophoresis setup, which is a patented design process by NovAM at TU Delft, was utilized. The setup which is kept in a plexiglass box during operation time is accompanied by a heater device (equipped with a thermocouple), a high voltage amplifier, a wave generator and an oscilloscope. The setup consists of two alumina plates that are clamped during operation, a heating element placed on the bottom alumina plate, two kapton foils, top/bottom electrodes and a Teflon mould of circular cut shape.

#### 5.2.2. IN-SITU PRINTING/DEP SETUP

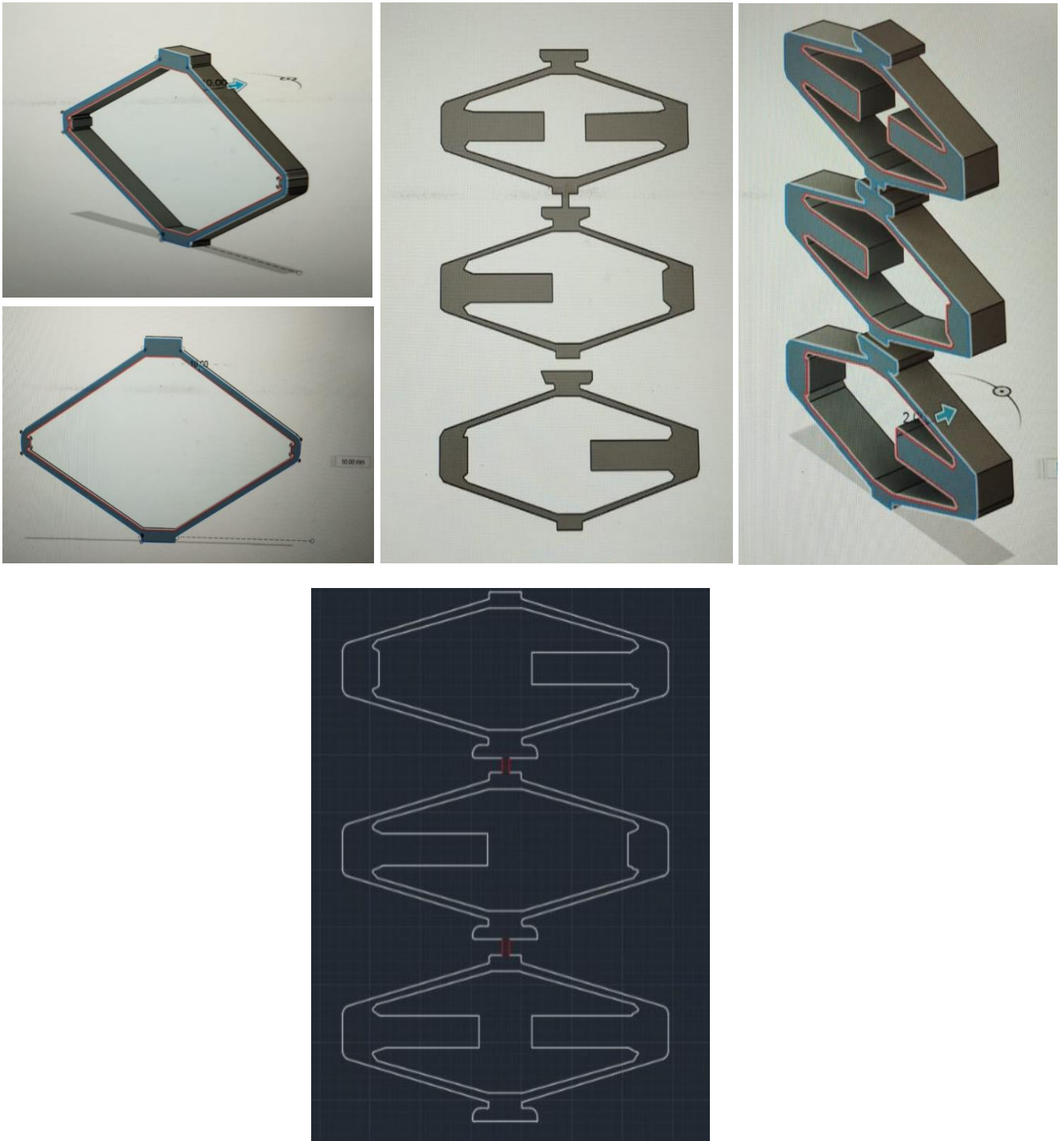
The in-situ printing/DEP setup is a concept design developed by NovAM at TU Delft for the in-situ alignment of the ceramic filler while 3D printing is under process. The setup is composed of the full nozzle-head configuration of the Ultimaker's 2+, which includes the Olsson block, the thermocouple and the heating element, along with a newly designed nozzle of 0.6mm size, that is attached to the rest of the printer by a 8mm long boron nitride threaded tube that acts as an electrical insulator. In Fig.4.1 a schematic representation of the new nozzle concept design is displayed. The process follows the standard FDM printing technique with the composite filament entering the Olsson block from the top. While moving forward, the filament begins to melt due to high temperature and flows down through a boron nitride threaded tube (that provides electrical insulation between the nozzle and the Olsson block). Subsequently, the extruded material comes out from the nozzle and is instantly quenched at the orifice. The dielectrophoretic alignment of the ceramic filler takes place by applying a high voltage electric field between the Olsson block and the nozzle simultaneously.

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## Results and Discussion :

This work demonstrates that structured piezoelectric composites of (quasi)1-3 connectivity, of multiple PLA based compositions, were developed at a certain electric field and frequency of 1kV/mm and 1 kHz, respectively. Through the investigation on the major parameters that influence the performance of the in-situ printing/DEP concept design, it was shown that by incorporating an electrical insulating threaded tube between the heating element (ground electrode) and the nozzle (hot electrode), a temperature difference of 15°C was established inside our setup. Therefore the possibility of performing dielectrophoretic alignment at a range of temperatures between 180°C and 210°C, was explored. From the obtained results, it was shown that particle chains were formed for both BT- PLA and KNLN- PLA composites at the whole temperature range. In addition to the above, research on the influence of the length between top/bottom electrodes and that of printing speed, was carried out. In a brief parametric analysis on their linear relationship, it was indicated that maximum available exposure time frame was 20s for the minimum printing speed value and for 20mm length. The range of applied voltage that is required based on the configuration of our setup was to be 13 20kV for an electric field of 1kV/mm. Finally, it was presented that except from the secondary setting of exposure time, the parameter of viscosity affects the particle alignment performance of the new set up and that it is controlled by the setting of printing speed and temperature.

In order to fully understand the behavior of PLA under the conceptual setup, rheological measurements utilizing a capillary rheometer are recommended for a PLA-based piezoelectric composite filament. From these results we can infer important information regarding the rheological properties of the filament as it is extruded through the printer's nozzle, as well as, as it behaves along the length between the two electrodes. Furthermore, it is recommended, along with the optimization of the new setup process, the development of the cascade concept design through a collaboration between TU Delft and Ultimaker.



### Conclusion :

In this thesis the prospects of fabricating a randomly oriented, printed piezoelectric sensor through the Additive Manufacturing (AM) technique of Fused Deposition Modeling (FDM) were explored, as well as, the development of a concept design process for in-situ printing/DEP. The investigations into the development of a randomly oriented, lead-free piezoelectric sensor of multiple filler content and low volume fraction filler, presented that a nozzle size higher than 0.25mm is required in order achieve a fully dense biphasic composite. In addition, a link between the dielectric constant of the printed sample disk and the printing settings of both layer height and infill density was observed, indicating that due to a decrease in void content, dielectric constant increases as the layer height decreases and infill density increases respectively. Moreover, the optimum printing nozzle temperature was found to be a function of filament composition, which is probably due to particle size distribution of the filler. From characterization of the piezoelectric properties of the printed KNLN- PLA samples of KNLN- PLA, a proportional relationship between charge piezoelectric charge constant ( $d_{33}$ ) and poling temperature, was noticed. Piezoelectric performance tends to increase as poling temperature increases, up until the point that

polymeric matrix loses its solid state to a liquid one. Through characterization analysis, the obtained  $d_{33}$  values were  $0.17 \pm 0.09 \text{ pC/N}$  and  $1.35 \pm 0.21 \text{ pC/N}$  for BT- PLA and KNLN- PLA, respectively. Moreover, the piezoelectric charge constant ( $g_{33}$ ) which stands as a figure-of-merit indicator, presented to be  $2.83 \pm 0.9 \text{ mV m/N}$  for BT-based composites and  $28.62 \pm 2.1 \text{ mV m/N}$  for KNLN-based ones.

Through our work on structured piezoelectric composites, it was found that particle chain alignment can be developed for multiple PLA-based compositions at a certain electric field and frequency. By separating the heating element and the nozzle for our concept design of in-situ printing/DEP process, a temperature difference was established in the system. By varying the DEP temperature for both piezoelectric compositions, dielectrophoretic alignment was achieved at arrange of temperatures. Furthermore, by taking into consideration all the major parameters (i.e. such as the length between the top/bottom electrodes, temperature and printing speed) that are involved during operation, a thorough investigation on their influence on the net particle alignment performance of our setup was carried out. It was shown that except from the setting of exposure time, which is influenced from the length of the electrodes and the printing speed, the parameter of viscosity affects the particle alignment performance of the new setup and that it is controlled by the setting of printing speed and temperature.

Unfortunately not all optimizations were possible in the time frame of our project. Therefore certain recommendations for future work are advised. A logical next step for the development of flexible randomly piezoelectric electronics through a FDM process, can be the selection of a polymeric matrix that fits the processing window of the printing technique used, has good chemical compatibility with the chosen ceramic filler, and shows suitable poling parameters ( $\epsilon$ ,  $\sigma$ ,  $T_g$ ). Additionally for enhanced ease of processing, a filler of a well-defined particle size is recommended. Moreover, in order to achieve higher piezoelectric properties with a flexible polymer matrix, higher filler's volume fraction is required by adjusting the feeding system of the FDM printer to handle filaments of higher brittleness.

The increasing interest in in-situ printing/DEP setup, due to its potential to produce structured piezoelectric composites in a both time and cost efficient manner, indicates that future work should be carried out for the optimization of the process, either experimentally or via modeling its major parameters. To fully develop the process and being able to control all the major parameters that influence its performance, a series of rheological measurements utilizing a capillary rheometer are recommended in order to understand the behavior of the PLA-based piezoelectric composite filament inside the system. Finally, a possible optimization of the setup's configuration to a more cascade structure is considered a possible route of improvement and potential collaboration between TU Delft and industry.

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