A Comprehensive Review of Nanocomposite PVDF as a Piezoelectric Material: Evaluating Manufacturing Methods, Energy Efficiency and Performance

Article history:
Received: 12-09-2023
Revised: 26-11-2023
Accepted: 12-12-2023

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Abstract: Given the escalating concerns surrounding high energy consumption during manufacturing and the environmental impact of piezoelectric materials, the pursuit of sustainable alternatives has emerged as a critical challenge in shaping our technological future. In light of this imperative, this review paper investigates the domain of polymeric piezoelectric materials, with a specific focus on Polyvinylidene fluoride (PVDF) as a promising avenue for sustainable piezoelectric materials with a low-energy production process. The primary objective of this investigation is to conduct a comprehensive assessment of the existing research on the manufacturing processes of polymeric piezoelectric materials to enhance piezoelectric properties while minimizing energy-intensive production techniques. Through rigorous evaluation, the effectiveness of each manufacturing method is scrutinized, enabling the identification of the most energy-efficient approaches. This review paper paves the way for sustainable development and advancement of piezoelectric technologies.

Keywords: Review paper; Piezoelectric materials; PVDF; Manufacturing methods.

1. INTRODUCTION

The rising rates of energy consumption and environmental pollution have become significant obstacles to our future livelihood and technological advancement. Moreover, the rapid progress of mobile electronic devices, their increasing demand for higher battery energy density, and the limitations of traditional batteries have underscored the necessity for alternative energy sources. Hence, it is imperative to seriously contemplate the advancement of sustainable, economically viable, and eco-friendly energy sources capable of meeting the forthcoming energy requirements (Brown *et al.*, 2018; S. Fotouhi *et al.*, 2019; Mahapatra *et al.*, 2021). There are various types of renewable clean energies with different energy conversion systems for harvesting energy, including wind energy (S. Li & Yuan), solar energy (Yoon & Yu, 2016), hydropower (Vu, Le, & Ahn, 2022), biomass (Ko *et al.*, 2017) and mechanical vibration (Zuo & Tang, 2013).

Using green energies in micromechanical devices are somehow challenging due to their significant power supply and hard usage condition (Megdich, Habibi, & Laperrière, 2023). The preferred energy in microelectronic systems with power supply in the scale of (mW and μW) is mechanical energy. The mechanical energy generated

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through motion or vibration can be transformed into electrical energy through three distinct approaches: Piezoelectric (Liang, Hao, & Olszewski, 2021; Hanbo Shao, Chen, & He, 2021), Electromagnetic(Shen & Zhu, 2015; X. Zhao *et al.*, 2018), and electrostatic (Naito & Uenishi, 2019; Vysotskyi *et al.*, 2018) methods.

These techniques have been extensively examined in various academic works. However, the majority of research has centered on piezoelectric materials, primarily because of their remarkable piezoelectric sensitivity and significant energy conversion capabilities (A. Rahman, Farrok, Islam, & Xu, 2020). Piezoelectric materials demonstrate an extraordinary capability to transform mechanical stress into electric charges through the direct piezoelectric effect. Additionally, it has been observed that applying a voltage across two electrodes of a piezoelectric material induces mechanical strain or deformation through the converse piezoelectric effect (Kapat, Shubhra, Zhou, & Leeuwenburgh, 2020).

This inherent potential of piezoelectric materials makes them suitable for energy-harvesting and actuator applicants (Elnabawy *et al.*, 2021; W. Feng, Chen, Wang, & Yu, 2022; Sakineh Fotouhi, Fotouhi, Pavlovic, & Djordjevic, 2018; X. Li *et al.*, 2021; Smith & Kar-Narayan, 2022; Y. Wang *et al.*, 2021). The piezoelectric phenomenon is evident in both naturally occurring and synthetic substances. Among the most effective piezoelectric materials, known for their elevated dielectric and piezoelectric coefficients, are piezoceramics. Different ceramics such as lead zirconate titanate (PZT) (Jain, KJ, Sharma, Jain, & PN, 2015; Jung, Do, Kang, & Kang, 2013), Sodium and Potassium niobate (Na/$KNbO_3$) (Mohammadpourfazeli *et al.*, 2023), barium titanate (BaTiO) (Chang *et al.*, 2016; Sood *et al.*, 2023), Zinc Oxide (ZnO) (Kumar & Kim, 2012; D Ponnamma & Cabibihan, 2019), lithium niobate ($LiNbO_3$) (Yun *et al.*, 2014), lead magnesium niobate-lead titanate (PMN-PT) (Hwang *et al.*, 2014; Paralı, Koç, & Akça, 2023) are used widely as a piezoelectric material. While Piezoceramics exhibit exceptional piezoelectric properties, they are not without limitations. These drawbacks include brittleness, non-biocompatibility, challenging processability, and toxicity, which impose constraints on their applications. In contrast, Piezopolymers, despite having lower piezoelectric coefficients, offer advantageous features such as flexibility and biocompatibility. Consequently, Piezopolymers hold promise to overcome several limitations associated with Piezoceramics. The remarkable flexibility of piezopolymers renders them particularly suitable for applications that demand high flexibility, such as biomedical applications(Z. Feng, Wang, Liu, Han, & Yu, 2023).

PVDF stands out as one of the main promising electroactive materials, offering exceptional piezoelectric characteristics. PVDF and its copolymers, such as trifluoro ethylene (PVDF-TrFE) and hexa-fluor propylene (PVDF-HFP), find extensive use as piezoelectric materials owing to their distinctive properties: lightweight, corrosion resistance, flexibility, biocompatibility, adequate mechanical behavior, and eco-friendliness (Correia *et al.*, 2016; Damaraju *et al.*, 2017; L. Wu, Jin, Liu, Ning, Liu, Alamusi, *et al.*, 2022; L. Wu, Jin, Liu, Ning, Liu, & Hu, 2022). So, these polymers can be a suitable candidate to replace piezoceramics.

The piezoelectric nanogenerator (PENG) has the potential to become a promising.

Given that the piezoelectric coefficient of PVDF is typically in the range of 20-40 C/N, significantly lower than that of piezoceramics such as PZT (which can reach up to 500 pC/N), it becomes imperative to enhance the piezoelectricity of PVDF. This enhancement is critical for advancing the performance and efficacy of PVDF-based piezoelectric devices (Z. Yang, Zhou, Zu, & Inman, 2018). Moreover, PVDF-based sensors are presently employed to provide power and monitor sensors within Internet of Things (IoT) networks using wireless communication (Chung *et al.*, 2019).The piezoelectric properties are directly related to the amount of β-phase in the PVDF, so the improvement of β-phase content is essential for enhancing the piezoelectric output of PVDF products (Z. Liu, Li, Zhu, Mi, & Zheng, 2022). The piezoelectric properties of PVDF can be enhanced by incorporating piezoelectric ceramics, single crystals, or certain nanoparticles into its composition. This blending approach leads to improved piezoelectric capabilities in PVDF-based materials (Bodkhe, Turcot, Gosselin, & Therriault, 2017; Y. Chen *et al.*, 2022; Pei, Shi, Chen, Xiong, & Lv, 2022; X. Wan *et al.*, 2023; M. Yuan *et al.*, 2023). The topic of piezoelectric polymers has been reviewed numerously from different points of view (Ju *et al.*, 2023; Kalimuldina *et al.*, 2020a; L. Lu, Ding, Liu, & Yang, 2020; Mahapatra *et al.*, 2021; Malini, Indumathy, Gunasekhar, & Prabu, 2022; Megdich *et al.*, 2023; Mohammadpourfazeli *et al.*, 2023; Fatemeh Mokhtari, Azimi, Salehi, Hashemikia, & Danti, 2021; Smith & Kar-Narayan, 2022; Surmenev *et al.*, 2019; Tuloup *et al.*, 2019; X. Wan *et al.*, 2023; L. Wu, Jin, Liu, Ning, Liu, & Hu, 2022). This review research provides a comprehensive summary of the latest advancements in the manufacturing processes of piezoelectric materials, with a particular emphasis on their economic efficiency. The primary objective of this study is to categorize and analyze existing research on piezoelectric materials, with a specific focus on the manufacturing methods employed. Recognizing the significant influence of manufacturing processes on the properties and performance of these materials, the research aims to conduct a thorough investigation and evaluation of each method using a comprehensive categorization approach.

2. PRINCIPLE OF PIEZOELECTRIC EFFECTS AND THEORY

The concept of "Piezoelectricity" originates from the Greek term "piezein," which translates to "to press." (Vassiliadis & Matsouka, 2018). The Curie brothers made a significant breakthrough in 1880 by discovering the piezoelectric effect while researching quartz crystals (Curie & Curie, 1880). More than 30% of the materials show piezoelectric effect. While this property is present in a diverse range of materials, only a select few of them can be practically utilized (Sappati & Bhadra, 2018). The direct piezoelectric effect is the term used to describe the ability to convert mechanical energy into electrical energy. Soon it was found that mechanical strain (deformation) is produced when these materials are subjected to an electrical field (Ramadan, Sameoto, & Evoy, 2014). It means that a piezoelectric material is polarized and generates a voltage when mechanical stress is applied to it (C. Wan & Bowen, 2017). This phenomenon is known as the indirect effect. A schematic diagram illustrating both the direct and inverse piezoelectric effects can be observed in Figure 1. The direct piezoelectric effect is particularly suitable for energy harvesting, while the converse piezoelectric effect finds applications in acoustic emitters, dampers, and actuators.

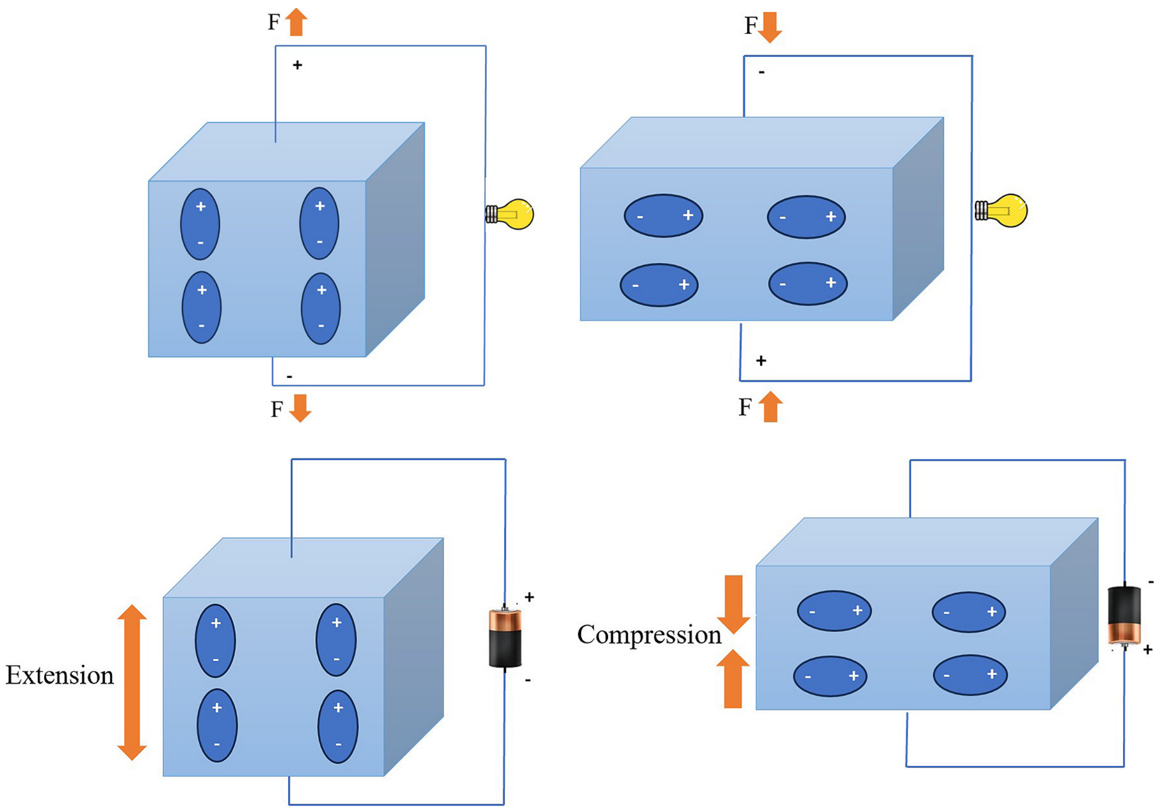

Figure 1. Diagram depicting the direct and Indirect piezoelectric effects.

Piezoelectric effects can be mathematically described by the constitutive strain-charge equations (1) and (2), which offer a comprehensive portrayal of these phenomena (Jean-Mistral, Basrour, & Chaillout, 2010; Kochervinskii, 2003; Mohammadpourfazeli *et al.*, 2023; Smith & Kar-Narayan, 2022):

$$Dk = d_{jk}\, T_j + \varepsilon^{T}_{km}\, E_m \quad (1)$$

$$Si = s^{E}_{ij}\, T_j + d_{im}\, E_m \quad (2)$$

The direct piezoelectric formula is represented by Eq (1), which relates the electric displacement (D) to stress (T). Also, Eq (2) describes the converse piezoelectric effect, depicting the connection between the electric field (E) and the resulting strain (S). These equations incorporate various parameters such as the piezoelectric coefficients (d), elastic compliance (s), and dielectric permittivity (ε).

Piezoelectric characteristics are frequently evaluated through the utilization of coefficients represented as dij. These coefficients are arranged within a matrix, with the first subscript (i) signifying the direction of the electric field, while the second subscript (j) signifies the direction strain. The transverse coefficient, d31, describes a situation in which the resulting electrical polarization is perpendicular to the applied stress. Conversely, the d33 coefficient, illustrates a scenario where both the electrical polarization and the stress align in the same direction (refer to Fig. 2). These coefficients provide valuable insights into the polarization behavior of piezoelectric materials (Ramadan *et al.*, 2014).

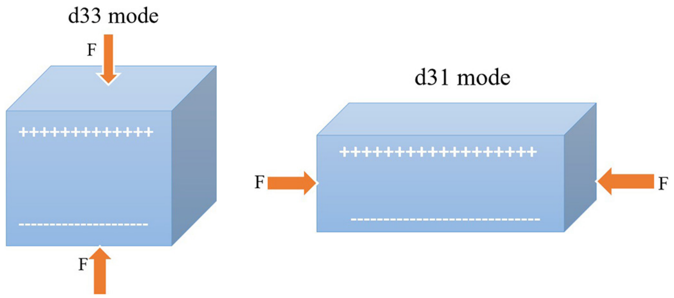

Figure 2. The d_{33} and d_{31} modes.

To evaluate the energy efficiency of the PVDF-based system, It utilizes various key measuring parameters, including voltage output, current, and power generation. These parameters allow us to conduct a quantitative assessment of the system's ability to efficiently convert mechanical vibrations or other energy forms into usable electrical power. Furthermore, elaborating on the precise methodologies employed for these measurements serves to enhance the reader's comprehension of the robustness and precision of research outcomes.

3. PIEZOELECTRIC MATERIAL

Piezoelectric materials refer to substances that exhibit the piezoelectric effect, enabling them to convert mechanical energy into electrical energy. These materials find diverse applications in fields such as sensors, actuators, energy harvesting, nanogenerators, health monitoring, and more. Their ability to transform energy between mechanical and electrical forms makes them highly suitable for a wide range of technological applications (H. Yuan, Lei, Qin, & Yang, 2019).

Piezoelectric materials can be categorized into four main groups based on their composition: inorganic piezoelectric materials, organic piezoelectric materials, piezoelectric composites, and natural piezoelectric materials. This classification provides a framework for understanding the different types of materials that exhibit piezoelectric properties.

3.1. Inorganic piezoelectric

The category of inorganic piezoelectric materials encompasses piezoelectric single crystals and ceramics, which were the first types of piezoelectric materials to be discovered. Due to the inherent asymmetry of their crystal structures, these materials exhibit remarkable piezoelectric properties without requiring a polarization process. Inorganic piezoelectric materials possess distinctive characteristics, including excellent temperature stability, high dielectric constant, high piezoelectric coefficient, and the ability to achieve high orientation. However, their processing costs tend to be high, and their fabrication process can be complex (X. Wan *et al.*, 2023). Among the commonly used piezoelectric crystals are Quartz, SiO_2, $LiGeO_3$, $LiNbO_3$, and others. These crystals exhibit reliable piezoelectric properties. Piezoelectric ceramics, on the other hand, are characterized by high piezoelectric and dielectric constants, cost-effectiveness, and advanced preparation techniques. They find wide applications in fields such as ultrasonic imaging and underwater acoustic transducers. However, due to their low toughness and inherent brittleness, they are not suitable for flexible sensor applications. The most common piezoelectric ceramics are PZT (Lead Zirconate Titanate), ZnO, $BaTiO_3$, $BiSrTiO_3$, $CaTiO_3$, $PbTiO_3$, $SrTiO_3$, and so on (Gavrilyatchenko, Semenchev, & Fresenko, 1994; Kang, Jung, Kang, & Yoon, 2016; K.-I. Park *et al.*, 2010; D.-J. Shin, Jeong, Seo, Cho, & Koh, 2015; Sundarakannan, Kakimoto, & Ohsato, 2003).

The PZT is recognized as a piezoelectric material renowned for its exceptional piezoelectric performance. However, it is important to note that PZT possesses certain drawbacks. Firstly, it is considered toxic and harmful to both human health and the environment. Additionally, PZT exhibits weak mechanical properties, which can limit its applications in certain scenarios. These considerations highlight the need to carefully address the environmental and health concerns associated with PZT and explore alternative materials with improved mechanical characteristics. (Sun *et al.*, 2020). On the other hand, BTO due to its environmental friendliness along with comprehensive piezoelectric properties, has attracted much attention (Yaqoob & Kim, 2018).

3.2. Organic piezoelectric material

Piezoelectric polymers, also referred to as organic piezoelectric materials, play a significant role in the field. Several commonly used piezoelectric polymers include poly (vinylidene fluoride) (PVDF), Nylon, epoxy, silicon, poly (L-lactide) (PLLA), polyimides (PI), polyacrylonitrile (PAN), and more. These polymers exhibit inherent piezoelectric properties, making them valuable for various applications that require flexibility, biocompatibility, or lightweight design (Ali & Khan, 2021; Anwar *et al.*, 2021; Curry *et al.*, 2020; Datta, Choi, Chalmers, Ou, & Kar-Narayan, 2017; Q. Lu, Liu, Lan, Liu, & Leng, 2016; Hao Shao *et al.*, 2021; Toroń, Szperlich, & Kozioł, 2020; X. Wan *et al.*, 2023; Zabek, Pullins, Pearson, Grzebielec, & Skoczkowski, 2021).

Type of materials	Piezoelectric coefficient d_{ij} (*pC/N*)			Ref
	d_{33}	d_{31}	d_{14}	
PVDF	-10 to -33	8 to 23	—	(Gomes, Nunes, Sencadas, & Lanceros-Méndez, 2010; Z. He, Rault, Lewandowski, Mohsenzadeh, & Salaün, 2021; Fatemeh Mokhtari, Cheng, Raad, Xi, & Foroughi, 2020; Zaarour, Zhu, Huang, & Jin, 2018)
PVDF-TrFE	-25 to -40	12 to 25	—	(Calavalle *et al.,* 2020; Lutkenhaus, McEnnis, Serghei, & Russell, 2010; P Martins, Lopes, & Lanceros-Mendez, 2014; Soulestin, Ladmiral, Dos Santos, & Ameduri, 2017; Jiang Yang *et al.,* 2021; You, Zhang, Gui, Cui, & Guo, 2019; Zhou *et al.,* 2019)
PVDF-CTFE	140	—	—	(Z. Li, Wang, & Cheng, 2006)
PVDF-HFP	-18 to -24	30 to 43	—	(Badatya *et al.,* 2021; Huan, Liu, Yang, & Wu, 2007; Huang *et al.,* 2021; Deepalekshmi Ponnamma, Aljarod, Parangusan, & Al-Maadeed, 2020; Y. Wu, Qu, Daoud, Wang, & Qi, 2019)
polylactic acid (PLA)	12 to 19	1.58	9.82	(Baheti, Militky, & Marsalkova, 2013; Bernard, Gimeno, Viala, Gusarov, & Cugat, 2017; Smith & Kar-Narayan, 2022; Tai *et al.,* 2021)
Cellulose	5.7 and 14.5	1.88 to 30.6	—	(E. S. Choi *et al.,* 2021; Jaehwan Kim *et al.,* 2010; Rajala *et al.,* 2016)
Polyacrylonitrile (PAN)	3	2	—	(Fang & Lin, 2019; Peng *et al.,* 2021; Hao Shao *et al.,* 2020; Street, Minagawa, Vengrenyuk, & Schauer, 2019)
Polyimide	2.5 to 16.5	—	—	(Ramadan *et al.,* 2014)

Table 1. Overview of piezoelectric characteristics of polymers.

Piezoelectric polymers have gained significant attention due to their flexible nature, environmental friendliness, chemical resistance, lightweight composition, low acoustic impedance, low electrical permittivity, affordability, and ease of processing. These characteristics make them highly desirable in current times. The flexibility of piezoelectric polymers grants them an advantage over piezoceramics, making them suitable for intricate designs of piezoelectric sensors. Table 1 summarizes the piezoelectric properties of various polymers. Among these, PVDF and its copolymers stand out as semi-crystalline piezoelectric polymers with unique properties. The initial observation of the piezoelectric effect in PVDF was made by Kawai (Kawai, 1969) in the year 1969. This breakthrough finding has contributed significantly to the development and understanding of piezoelectric polymers. Through Kawai's observations, it was noted that the piezoelectric coefficients of PVDF exceeded those of other known synthetic polymers by an order of magnitude (refer to Fig. 3). Although PVDF exhibits lower piezoelectric properties compared to piezoelectric ceramics, significant efforts have done to improve it (F Mokhtari, Latifi, & Shamshirsaz, 2016). Practically, it is observed that the incorporation of nanoparticles can improve the piezoelectric properties of PVDF (Motamedi, Mirzadeh, Hajiesmaeilbaigi, Bagheri-Khoulenjani, & Shokrgozar, 2017).

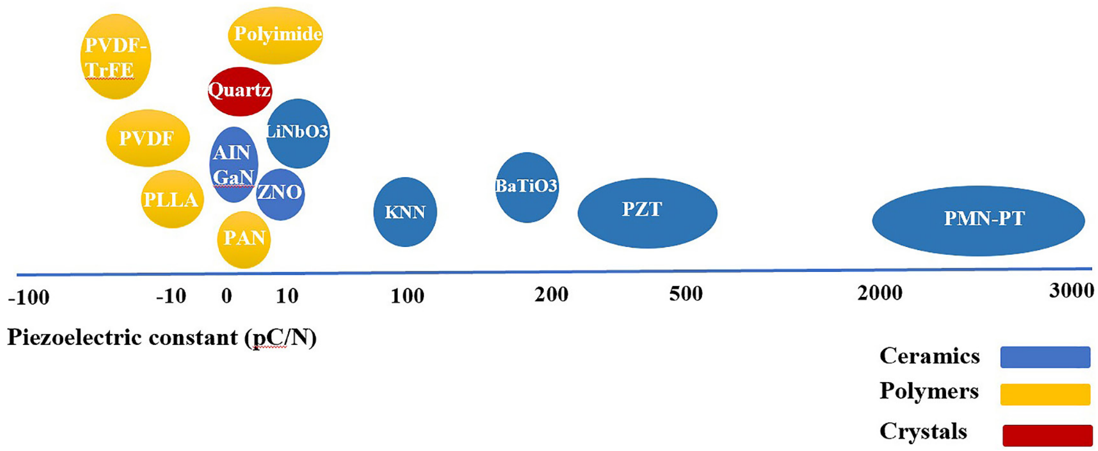

Figure 3. The charge constants in three types of piezoelectric materials (ceramics, polymers, crystals).

PVDF displays five distinct crystalline phases, which comprise: α (non-polar), β (polar), γ (polar), σ (polar), and ε phases. However, PVDF primarily consists of α and β crystals (Fig. 4). Presence of these distinct phases is determined by the conformation of the polymeric chains (T. He *et al.*, 2019; Jayoung Kim, Campbell, de Ávila, & Wang, 2019).

Certainly, addressing the challenges and limitations of nanocomposite PVDF as a piezoelectric material is crucial for a comprehensive review. Here are some insights into the scalability, durability, and environmental concerns associated with this material:

- Scalability: Nanocomposite PVDF materials have demonstrated potential in laboratory settings; however, scaling up their production to an industrial level presents formidable hurdles. The industrial-scale synthesis and processing of these nanocomposites may necessitate substantial investments in specialized equipment and rigorous process optimization. These challenges can have repercussions on the cost-effectiveness and broader acceptance of nanocomposite PVDF in real-world applications(Havelka *et al.*, 2023).
- Durability: While nanocomposite PVDF materials offer improved piezoelectric properties, their long-term durability in real-world conditions is a concern. Environmental factors, such as temperature fluctuations, humidity, and mechanical stress, can impact the stability of the nanocomposite structure over time. Investigating the material's resilience and lifespan under various operating conditions is essential for assessing its suitability for long-term applications(J. Fu, Hou, Gao, Zheng, & Zhu, 2018).

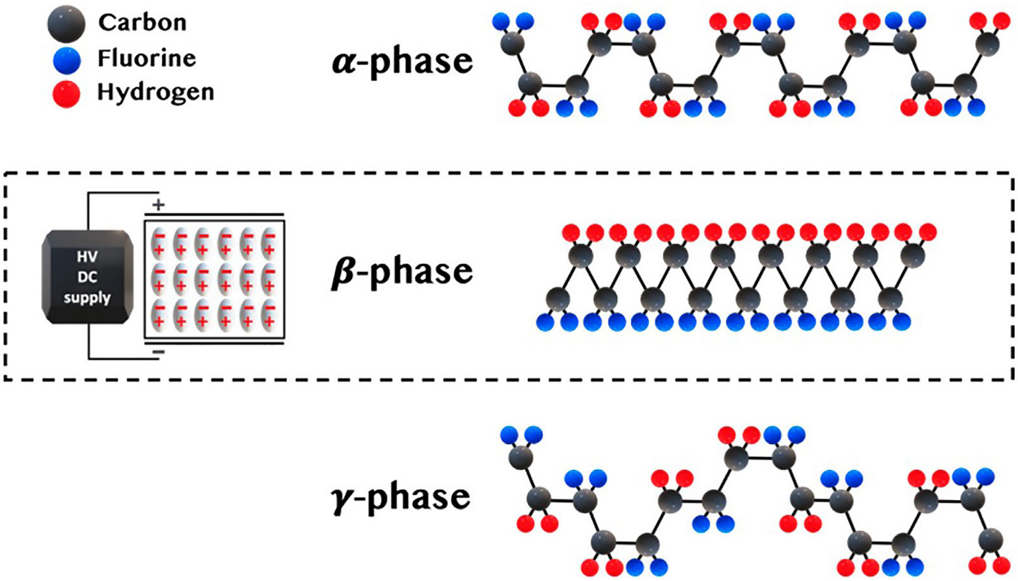

Figure 4. Primary phases of PVDF and the formation of β phase through high-voltage application(Kalimuldina *et al.*, 2020a).

- Environmental Concerns: The incorporation of nanoparticle additives like carbon nanotubes or graphene into PVDF nanocomposites can give rise to environmental and safety considerations. The entire life cycle of nanomaterials, from production to utilization and disposal, carries potential risks to both human well-being and the natural environment. Consequently, it is imperative to confront these issues and delve into strategies aimed at the secure management, recycling, or disposal of nanocomposite PVDF materials (B. Ouyang *et al.*, 2021).

3.3. The piezoelectric composites

As previously noted, piezopolymers have gained recent attention from researchers due to their unique attributes, including flexibility. However, their applications are limited because of their low piezoelectric coefficients. As a result, a new group of materials is made by the combination of organic and inorganic piezoelectric materials, which are named piezoelectric composites. Some typical inorganic materials such as PZT, ZnO, BTO, and KNN are widely used to fabricate piezoelectric composites (J. Li *et al.*, 2019; Niu *et al.*, 2018). The combination of PVDF with piezoelectric ceramic fillers such as ZnO, BTO, PZT, $BaTiO_3$ and KNN, made a composite with good flexibility and piezoelectric properties (Y. Chen *et al.*, 2022; C. Liu *et al.*, 2015; Pei *et al.*, 2022; C. Yang, Chen, Sun, & Chen, 2021; M. Yuan *et al.*, 2023). Various investigations have shown that the incorporation of nanofillers can enhance the formation of β-crystals in PVDF, leading to substantial improvements in the piezoelectric characteristics of polymer composites. These polymer composites can be produced with common solvent-assisted techniques such as electrospinning (Dai *et al.*, 2021; L. Xue *et al.*, 2021) and 3D printing (Cui *et al.*, 2019; X. Yuan *et al.*, 2020).

4. FABRICATION METHODS OF PIEZOELECTRIC PVDF

This article discusses the different fabrication methods for preparing piezoelectric PVDF-based materials. Table 3 explores and contrasts various techniques, providing a comparative analysis of the piezoelectric properties exhibited by these polymers. The methods mentioned earlier were thoroughly examined, and a comprehensive evaluation of the piezoelectric properties and performance of each fabrication method is provided in Table 2.

4.1. Electrospinning

Among the various techniques available, electrospinning stands out as a highly efficient and cost-effective method for producing PVDF nanofiber films. This method is also employed for the production of nanofibers for various applications, including enhancing the toughness of composite laminates(Gholizadeh, Najafabadi, Saghafi, & Mohammadi, 2018; Mohammadi, Ahmadi Najafabadi, Saghafi, Saeedifar, & Zarouchas, 2021; Mohammadi *et al.*, 2023; Mohammadi, Najafabadi, Saghafi, & Zarouchas, 2020a, 2020b; Saeedifar, Saghafi, Mohammadi, & Zarouchas, 2021; Saghafi, Nikbakht, Mohammadi, & Zarouchas, 2021). This approach offers several advantages, including its affordability and ease of operation. The resulting nanofibers possess a small diameter, providing a large specific surface area. Moreover, their remarkable flexibility enables their widespread application in various fields such as sensors, tissue engineering, and wearable devices, among others (Batra, Sampson, Davis, Currie, & Vaseashta, 2023; Y. Zhao *et al.*, 2015). The process of electrospinning occurs within a high-voltage electric field, wherein the applied voltage aids in the polarization of nanofibers and the formation of the polar β-phase. As a result, there is no requirement for additional polarization. Furthermore, the mechanical stretching caused by the elongation of the polymer jet during fiber formation also contributes to the in-situ generation of piezoelectricity within the nanofibers (Kitsara *et al.*, 2019). Consequently, the PVDF film fabricated by this method, shows enhanced piezoelectric properties without any further polling post-process. The constructed PVDF fibrous film should be used in the form of a sandwich in piezoelectric sensors. Nanofiber membranes are inherently flexible, so for having a flexible sensor, flexible electrodes and packaging are necessary (Fig. 5).

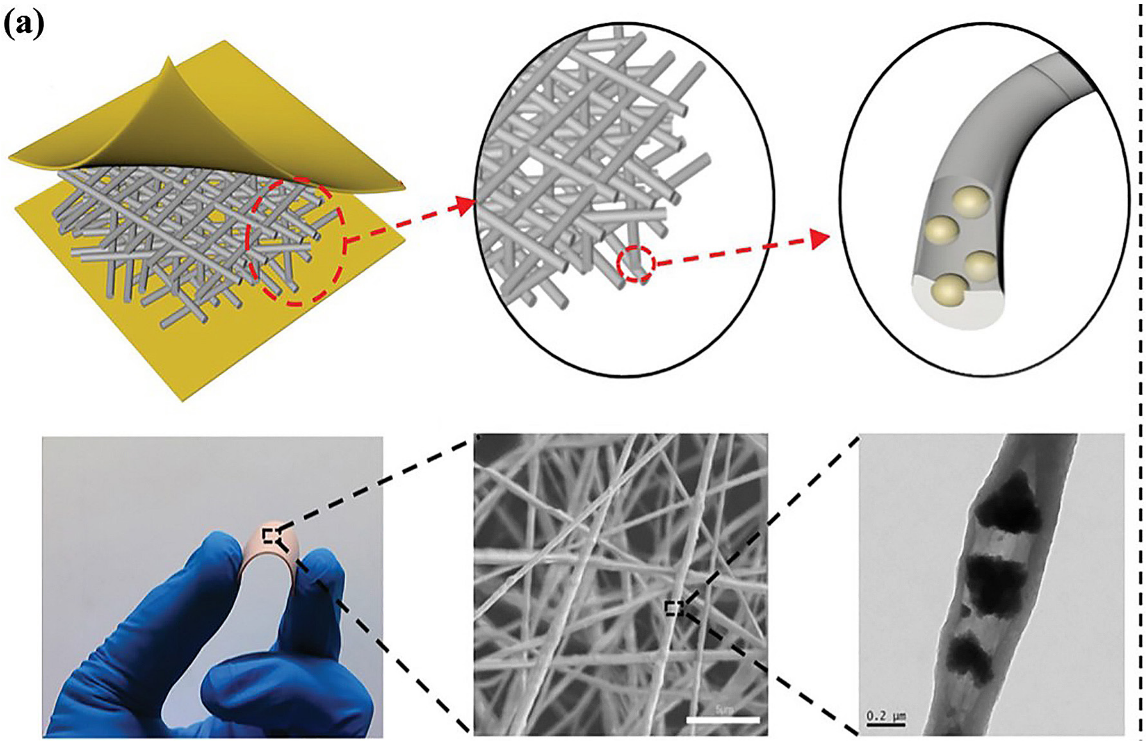

Figure 5. The electrospun PVDF/ZnO nanofiber (Deng *et al.*, 2019).

Fu et al. (G. Fu, Shi, He, Xie, & Liang, 2022) made multifunctional nanofiber films with an electrospinning method which can be used as an energy storage device and pressure sensor. They used a double filler system in order to investigate the synergic effect on mechanical, fluorescence, and piezoelectric properties of polyvinylidene fluoride-hexafluoropropylene (P(VDF-HFP)). The sandwich-structured piezoelectric composite, consisting of polyurethane (PU/P(VDF-HFP)-Eu $(TTA)_3(TPPO)_2$-$BaTiO_3$/PU) as depicted in Figure 6, exhibits a notable energy density of 30.45 mJ/cm^3 and demonstrates low-pressure sensitivity at 0.49 kPa^{-1}.

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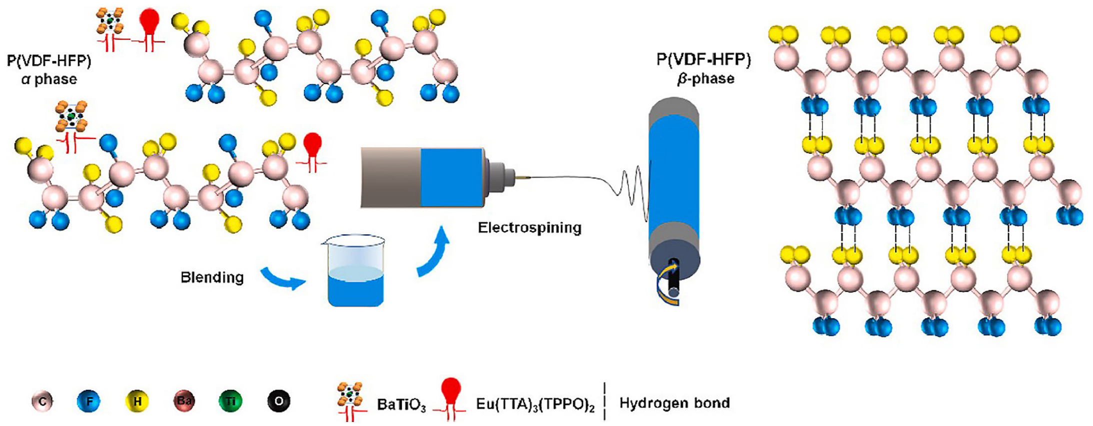

Figure 6. Electrospinning process for the production of P(VDF-HFP) nanofibers with Eu^{3+} complex and $BaTiO_3$ fillers (G. Fu, Shi, He, *et al.*, 2022).

To enhance the piezo PVDF output achieved through the electrospinning process, it is essential to gain a comprehensive understanding of the electrospinning process parameters and precisely fine-tune these parameters (Kalimuldina *et al.*, 2020b). Kumar et al. (R. K. Singh, Lye, & Miao, 2021) conducted research to investigate the impact of various electrospinning input parameters on the proportion of β-phase within electrospun nanofibers. Their findings indicated that the collector distance had the most significant influence on the percentage of β-phase. Other influential parameters included the amount of electric field, drum speed, and the feed rate.

In another study, Fu et al. (G. Fu, Shi, Liang, *et al.*, 2022) fabricated a piezoelectric sensor using electrospun nanofibers. They used graphene oxide as a 2D conductive filler. It could considerably improve the mechanical and piezoelectrical properties of the piezoelectric composite. The high β-phase content (96.3%) leads to good piezoelectricity of nanofibers. The pressure sensor can attain a maximum voltage of 4.5 volts. Abdolrasouli et al. (Haji Abdolrasouli, Abdollahi, & Samadi, 2022) developed PVDF-based electrospinning nanofibers reinforced with graphene oxide and TiO_2-Fe_3O_4 nanofillers. The findings indicated that the inclusion of 2 wt% TiO_2-Fe_3O_4-GO led to an enhancement in β-phase, reaching 79%, and resulted in a maximum voltage of 4.63 V.

Certain flexible conductive materials have found application as electrodes in PVDF devices, with graphene being a prominent choice. The utilization of graphene in this context has garnered significant attention due to its exceptional physical characteristics, encompassing superior electrical conductivity, remarkable thermal conductivity, and optical transparency, making it a promising candidate for transparent electrode material exploration(S. Park *et al.*, 2017).

4.2. Solution casting

Solvent casting stands out as one of the most convenient techniques for producing piezoelectric polymers. This method is highly favored due to its cost-effectiveness, quick preparation time, and ease of processing (Wahid, Khan, Hussain, & Ullah, 2018) . The process involves dissolving the polymer and various nanoparticles in a special solvent which can be volatile. Following a thorough mixing and blending process, the solution is molded into the proper shape. However, solvent casting has its drawbacks, such as inadequate dispersion and the potential for nanoparticle agglomeration. Additionally, it can result in high porosity, brittleness, and a translucent appearance in the final structure. These factors directly impact the strength and piezoelectric characteristics of the nano-composites, presenting challenges in their performance (Mohammadpourfazeli *et al.*, 2023).

Fig. 7 schematically shows this method. Kulkarni and Kumari (Kulkarni & Kumari, 2023) created a PVDF-TiO_2 nanocomposite intended for use in nanogenerator applications using a solvent casting method. The TiO_2-reinforced nanocomposite exhibited a substantial enhancement in tensile strength, elasticity, and storage modulus when compared to pure PVDF. In addition, the conductivity and dielectric constant increased by 214% and 143% respectively and the PVDF-TiO_2 nanocomposite, produced higher piezoelectric output voltage.

Pereira et al. (Nunes-Pereira *et al.*, 2015) produced P(VDF-TrFE)/$BaTiO_3$ nanocomposites through a casting method. Within the P(VDF-TrFE) matrix, $BaTiO_3$ fillers of diameters 10 nm, 100 nm, and 500 nm were incorporated at loadings ranging from 0% to 20% by weight. Notably, the sample containing 10 nm $BaTiO_3$ particles at a 20% loading exhibited the maximum output of 0.28 µW. Additionally, samples containing 100 nm and 500 nm particles at 5% weight loadings displayed identical outputs.

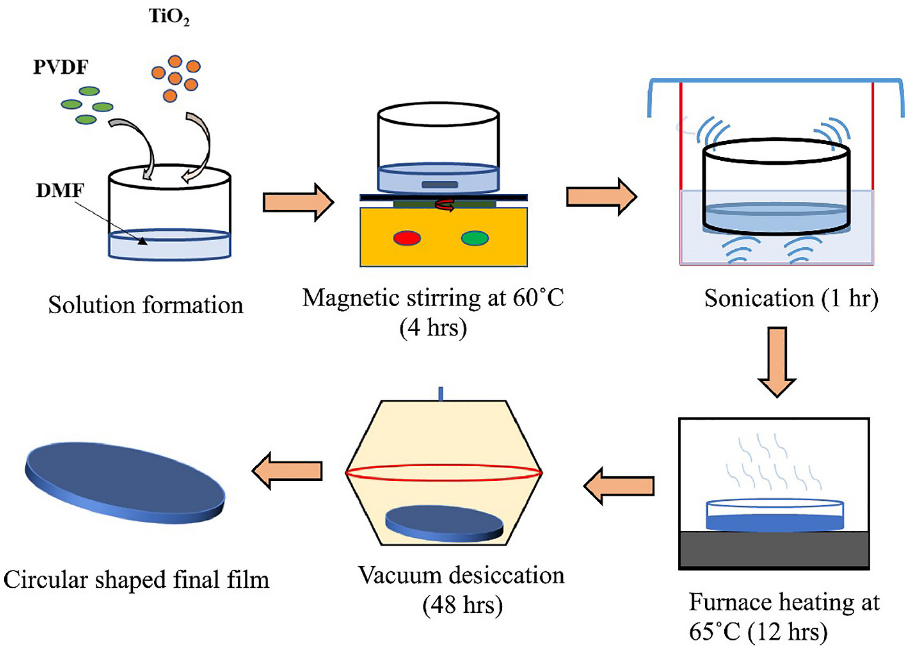

Figure 7. Fabrication of piezoelectric nanocomposite by using solution casting method (Kulkarni & Kumari, 2023).

Previous research showed that by incorporation of TrFE (20 to 50 mol%) into PVDF, the β phase content considerably increases (Yagi, Tatemoto, & Sako, 1980). Xia et al. (Xia, Che, Ren, Zhang, & Cao, 2023) produced a (P(VDF-TrFE) film using the solution casting method. The corona polarization under a 20 KV DC power generator electrical field at 80 ◦C was used for the polarization of P(VDF-TrFE) films. This film exhibited great piezoelectric coefficient (d_{33}) of −24 pC/N. Also, the piezoelectric sensor manufactured by this film could reach the large output of 8.9 V by applying a 1.5 MPa stress.

Kar et al. (Kar *et al.*, 2019) developed a self-polarized piezoelectric nanogenerator, including SnO_2 nanosheet and PVDF, with a solution casting method. This organic/inorganic nanogenerator shows outstanding output power enhancement. When subjected to gentle tapping by a human finger, the SnO_2/PVDF composite yielded an output voltage of 42 V and a current density of 6.25 $\mu A \cdot cm^{-2}$. This resulted in an impressive output power density of 4900 $W \cdot m^{-3}$, accompanied by an efficiency of 16.3%, as illustrated in Figure 8.

4.3. 3D PRINTING METHOD

In comparison with other traditional manufacturing methods for 3D devices, 3D printing is the most favorable technology with considerable advantages. Low-cost manufacturing technology, capability of production of multi-material parts, and extremely complex structures (Ngo, Kashani, Imbalzano, Nguyen, & Hui, 2018; Zolfagharian *et al.*, 2016). There are 7 different classifications for 3D printing techniques: Inkjet Printing (IJP); Direct Wiring (DIW); Fused Deposition Modelling (FDM); Stereolithography (SLA); Selective Laser Sintering (SLS); Laminated Object Manufacturing (LOM); and direct energy deposition (Megdich *et al.*, 2023).

Inkjet printing (IJP) is widely used for printing electronic devices due to its ability to print complex structures and use a wide range of materials (Andò *et al.*, 2017; Godard *et al.*, 2020; Machida *et al.*, 2012). Two key processes of IJP are polymer jetting and binder jetting.

In the direct ink writing (DIW) method, a paste-like ink is extruded (C. Chen *et al.*, 2019; Nick A

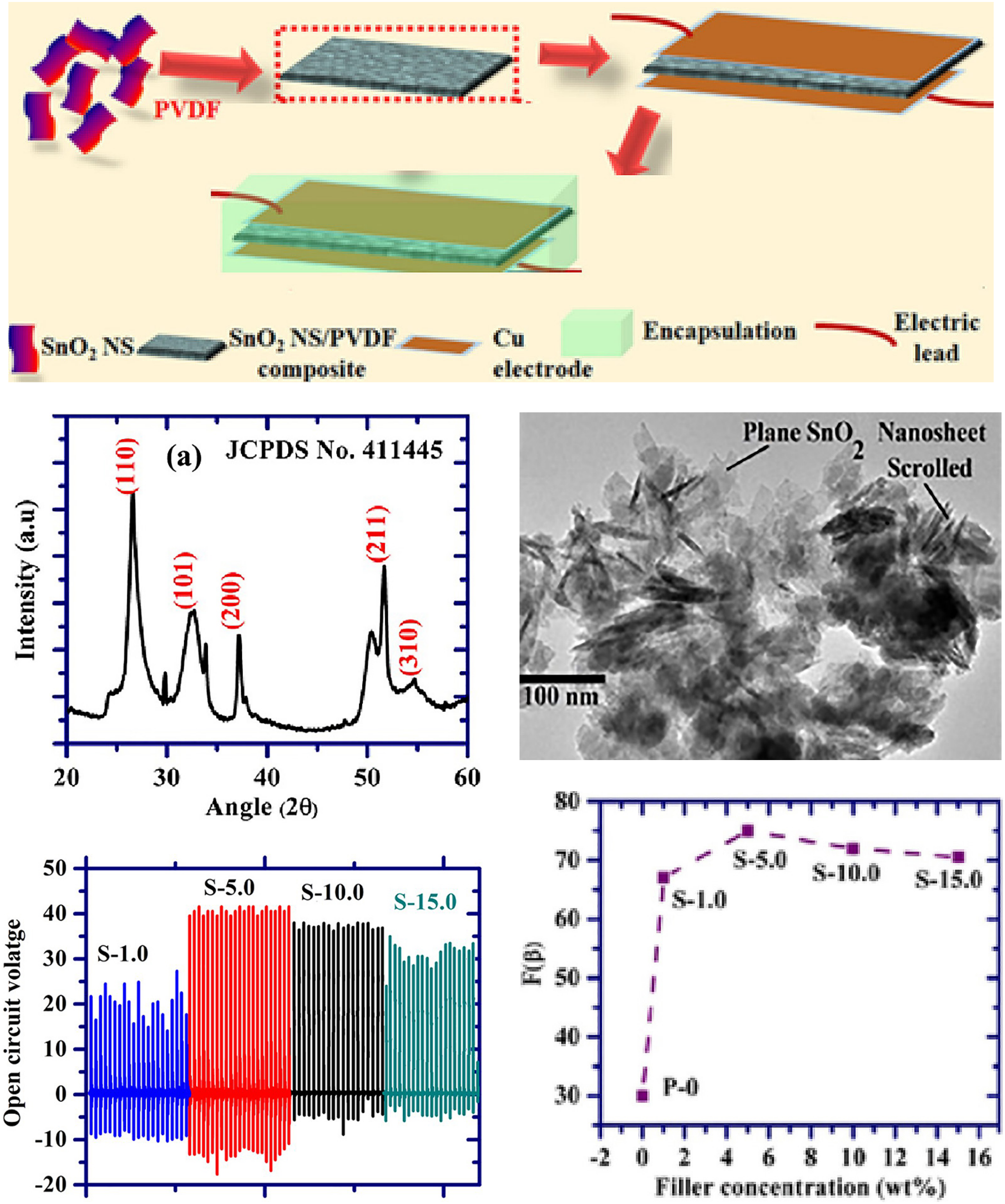

Figure 8. (a) The fabrication procedure of PSNG (b) The X-ray graph of SnO_2 nanocomposite. (c) TEM image of SnO_2 nanocomposite (d) Variation of the output voltage of the NGs (e) Variation of electroactive beta phase fraction (Kar *et al.*, 2019)

Shepelin *et al.*, 2019). The ink with high yield stress and storage modulus should be used, in order to allow distortion-free bridging of spanning filaments and protection of extruded lines (Megdich *et al.*, 2023). In the selective laser sintering (SLS) method, a high-power laser is used to fuse small particles into the last layer of 3D structure. In this method laser fuses powdered material on the surface of the bed according to the 3D digital description of the component (Qi *et al.*, 2021; Shuai *et al.*, 2020; S. Song, Li, Wang, & Zhang, 2021; C. Yang, F. Chen, *et al.*, 2021).

Stereolithography (SLA) is another technology for the fabrication of 3D structures (Z. Chen *et al.*, 2016; Seol *et al.*, 2018). In this method, light is used for the solidification of photocurable resin. The laser spots over photocurable resin cause photopolymerization and solidification. By the focus on using a laser source, the SLA technique can be used for the fabrication of incredibly tiny structures.

The most widely used technique is fused deposition modeling (FDM) due to its advantages over the other methods (Bodaghi *et al.*, 2020; L. He *et al.*, 2022; H. Kim, Fernando, Li, Lin, & Tseng, 2018; X. Liu, Liu, He, Shang, & Zhang, 2022; X. Liu, Shang, Liu, Shao, & Zhang, 2022; Pei, Xie, Xiong, Lv, & Chen, 2021; S. Tiwari, Gaur, Kumar, & Maiti, 2021). Easy operation, environmental friendliness, low cost, wide range of molding materials, and high efficiency (X. Liu, Shang, Zhang, & Zhang, 2021). In this method filament-shaped thermoplastic materials are used to build 3D structures layer by layer. The extruded material solidifies after cooling and forms a cross-section of the object.

Since in the molten state α- crystals are more stable, the production of PVDF devices with good piezoelectric performance via FDM printing is somehow challenging. So, the following treatment (electrical polling) is necessary to enhance the content of β-crystals to reach satisfactory piezoelectric performance. Researchers have proposed new methods such as subsequent polarization, synchronous electric polling, and adding nucleating agents (H. Kim *et al.*, 2018; Lee & Tarbutton, 2019; Porter, Hoang, & Berfield, 2017; Tarbuttona, Leb, Helfrichb, & Kirkpatrickb, 2017). However, according to previous literature, the content of β-phase can reach 56%, which is considerably lower than the accepted range in PVDF piezoelectric devices (more than 80%) (H. Kim *et al.*, 2018). Due to these limitations, it is highly required to develop a convenient method for FDM processing of PVDF devices with high β-phase. Liu et al. for the first time introduce ionic liquid (IL)-assisted FDM for the printing of PVDF-based piezoelectric devices with desired β-phase (X. Liu *et al.*, 2021). According to the result IL can maintain the β-phase content in melted PVDF, and this amount may reach 98.3% directly without any further process Fig. 9. schematically shows the process of FDM printing of IL-assisted PVDF.

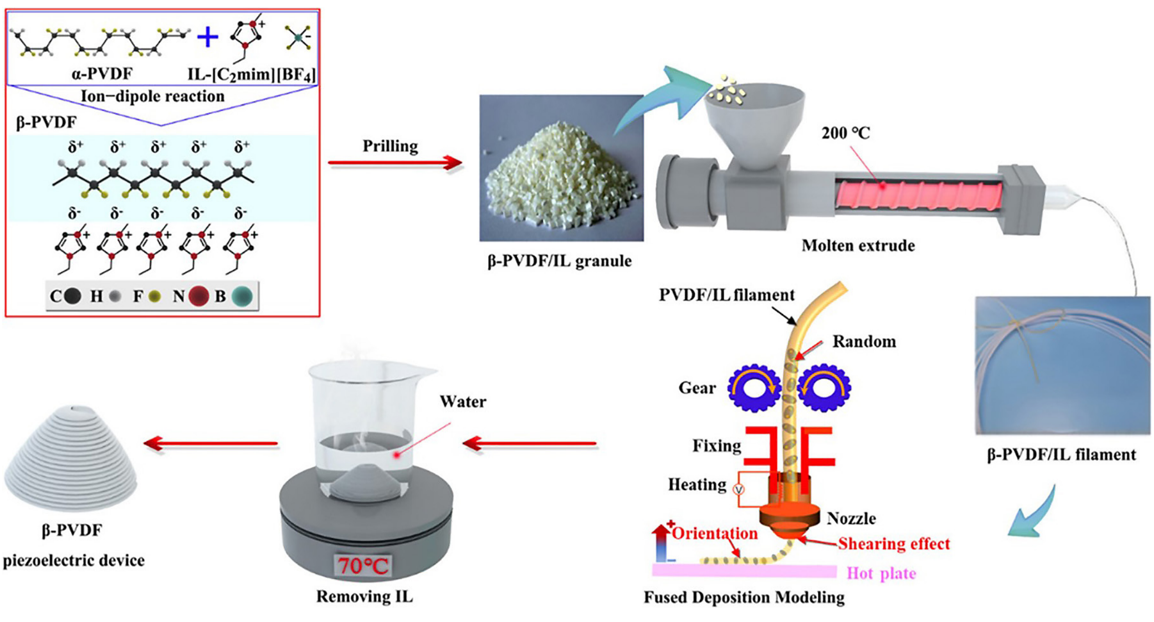

Figure 9. The process of FDM printing of IL-assisted PVDF (X. Liu *et al.*, 2021).

Bodkhe *et al.* produced a PVDF/$BaTiO_3$ nanosheet through 3D printing method (Bodkhe *et al.*, 2017). The 3D contact sensor can generate 4 V with a gentle finger tapping. The piezoelectric coefficient d_{31} of this nanocomposite was about 18 $^{pC}/_{N}$. This amount is comparable with poled and stretched PVDF films. Figure 10 depicts the emergence of the β-phase throughout the 3D printing procedure. Ball milling of $BaTiO_3$, activates these nanoparticles and increases the adhesion between $BaTiO_3$ and PVDF. During the extrusion process, the polymer chains undergo crystallization, forming an aligned β-phase. The $BaTiO_3$ nanoparticles limit the PVDF chains and force them to solidify in the β-phase.

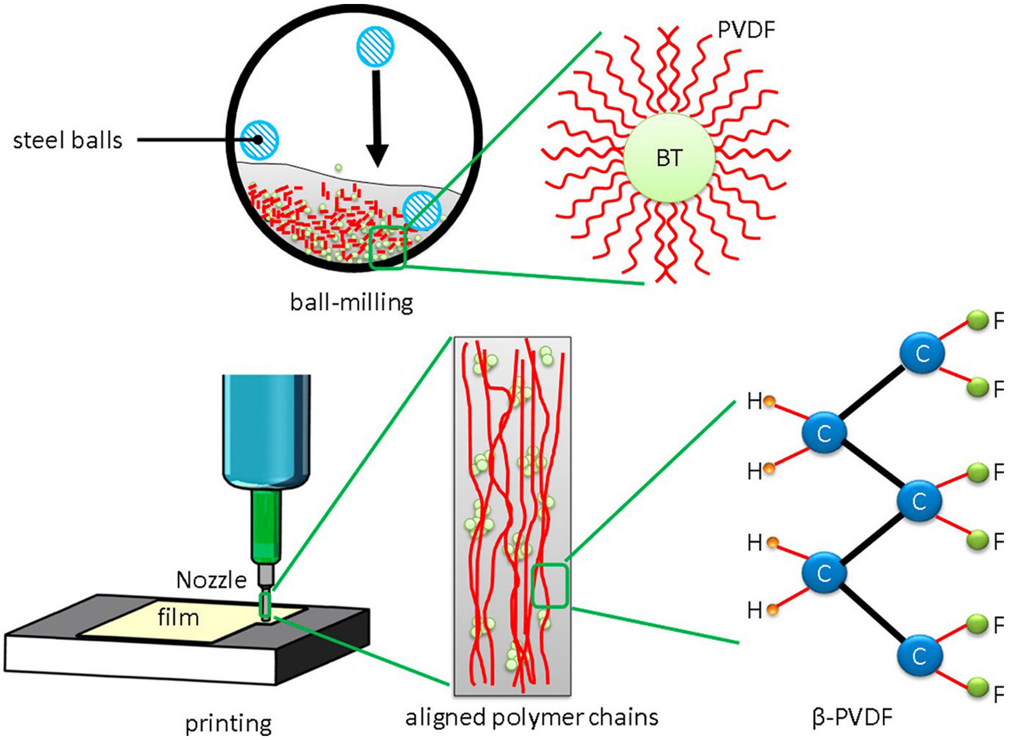

Figure 10. Increasing of the β-phase throughout the 3D printing procedure by adding the $BaTiO_3$ nanoparticles (Bodkhe *et al.,* 2017)

In another work, Kim et al. used the FDM method for the preparation of PVDF-based nanocomposite by incorporation of Multi-Walled Carbon Nanotubes (MWCNTs) and $BaTiO_3$ nanoparticles (H. Kim, Torres, Islam, *et al.*, 2017). The inclusion of MWCNTs improved the formation of a stress-reinforcing network and provided an electron conduction pathway within the PVDF matrix. By incorporation of 0.3% MWCNTs and 18% $BaTiO_3$, the piezoelectric constant was 0.13 $^{pC}/_{N}$. The output voltage and under bending and finger tap was 120 and 435 mV respectively.

4.4. Spin coating

Spin coating is one of the cost-effective methods that is widely used for obtaining thin and uniform films. In this method, a droplet of filler/polymer mixed solution is transferred on a horizontal, smooth rotating substrate. The centrifugal force causes evaporation of most of the solvent, so the layer of functional material remains, and a thin solid film is formed. This method has some advantages compared to other methods such as thickness control, energy saving, cost-effective, easy process, and low pollution. The fine-tuning of film properties can be accomplished by controlling input parameters such as rotation speed, droplet quantity, and solution ratio (L. Lu *et al.*, 2020; X. Zhang *et al.*, 2022). The creation of β-phase in the spin coating process was also investigated by Shaik et al (Shaik *et al.*, 2017). In this work effects of spin rotational speed, annealing temperature and solution concentration were studied. The optimized conditions for achieving The β-phase content increased significantly, reaching 93%, under the following conditions:

- Solution concentration: 15%
- Rotational frequency: 9000 rpm
- Annealing temperature: 100 °C

Within the spin-coating process, the nanocomposite thin layer experiences shear forces induced by the rotational speed. This orientation process encourages the alignment of polymer molecular chains, fostering the creation of the β-phase and amplifying the piezoelectric properties. Consequently, spin-coating exhibits considerable promise across various applications, including nanogenerators, nanophononics, and beyond. Figure 11 depicts a visual representation of the thin PVDF film preparation.

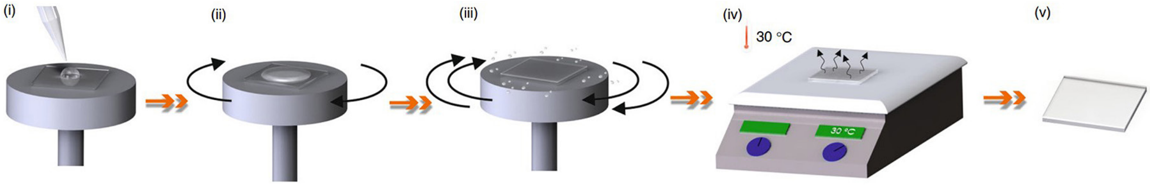

Figure 11. A thin PVDF layer preparation by spin coating method (Ribeiro *et al.*, 2018)

Also, some attention was paid to the synergistic effects of different composite layers on piezoelectric properties. Cardoso et al. (Cardoso, Minas, & Lanceros-Méndez, 2013) showed that one layer of spin-coated PVDF has more β-phase content in comparison to 3 layers of film. Furthermore, Hu et al. (Hu, Yan, Zhao, Zhang, & Niu, 2018) fabricated a bilayer piezoelectric nanogenerator using the spin coating method. This nanocomposite film is composed of two different layers, $BaTiO_3$/PVDF and pure PVDF layer. The findings indicate that a bilayer film comprising 20% $BaTiO_3$ exhibits exceptional output characteristics, yielding an output voltage of 6.7 V and a current of 2.4 µA.

4.5. Hot press

The hot-press method involves heating a polymer or polymer composite material to near its melting point and pressing it to obtain a piezoelectric composite. The electrical and piezoelectric properties of the composite completely depend on the temperature and pressure applied during the molding process (L. Lu *et al.*, 2020). Hot-press process conventionally contains a high-temperature press followed by a high-pressure cooling. With temperature and pressure increments, the porosity decreases, and a compact crystal structure is created. These phenomena make the polarization process more easily and improve piezoelectric properties. It should be mentioned that if the temperature is too high, the shrinkage of PVDF causes internal defects and leads to loss of piezoelectric performance (J. Fu *et al.*, 2018; J. Fu, Hou, Zheng, & Zhu, 2020). The temperature most of the time should be higher than 150 °C to ensure the complete melting of PVDF matrix (L. Wu, Jin, Liu, Ning, Liu, & Hu, 2022).

Despite many advantages of the hot-pressing method, controlling the distribution of the filler phase during powder mixing is still a big challenge, which often results in a serious agglomeration of fillers into the matrix. This leads to a poor interface between the fillers and polymer and can negatively impact piezoelectric properties.

Fu et al. (J. Fu *et al.*, 2018) developed a new flexible piezoelectric energy harvester using the hot-pressing method (Fig. 12). In this study, PVDF polymer matrix was filled by oriented $BaTi_2O_5$ (BT2) nanorods. The horizontal direction of BT_2 nanorods can significantly increase the power generation in the cantilever beam mode. The fabricated composite with the incorporation of 5% BT_2 exhibited a high power density of 27.4 µW/cm^3.

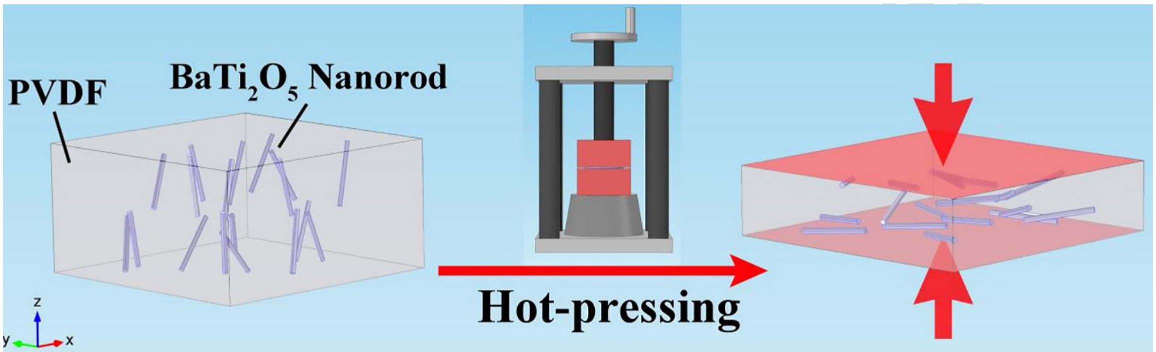

Figure 12. Schematic picture of the formation of BT_2/PVDF nanocomposite by using a hot-pressing process (J. Fu *et al.*, 2018)

4.6. Polling and Self-polling

The piezoelectric properties of PVDF are strongly impacted by the relative proportion of its crystalline phases, particularly the β-phase. While the α-phase can be obtained directly from a solution or in a molten state, the conversion of the nonpolar α-phase to the polar β-phase is of utmost importance in enhancing PVDF's piezoelectric capabilities. This transformation, known as "poling" or "polarization," is a critical step in the manufacturing process and significantly contributes to the overall piezoelectric performance of PVDF. Various techniques are employed to increase the piezoelectricity of PVDF. These include annealing and cooling at high pressure (Jiang *et al.*, 2007), drawing (cold stretching with high ratio) (L. Li, Zhang, Rong, & Ruan, 2014), electric polling (Parangusan, Ponnamma, & Al-Maadeed, 2018), and melting under a high-voltage electric field (Tian *et al.*, 2019). All of these methods are commonly used to improve the piezoelectric properties of PVDF (Scheffler & Poulin, 2022), as depicted in Figure 13. Furthermore, the introduction of nanofillers like metal oxides, carbon nanotubes, and graphene also influences the formation of polar molecular chains and leads to an increase in the content of the β-phase within the PVDF structure. This additional enhancement further contributes to its piezoelectric behavior (Parangusan, Ponnamma, & Al-Maadeed, 2018).

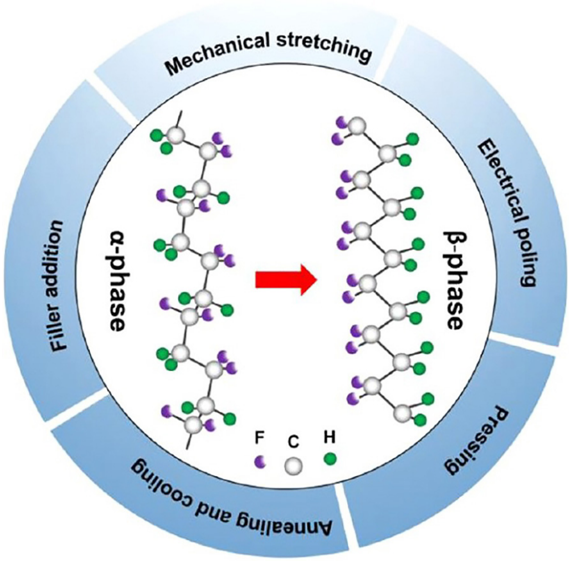

Figure 13. The effect of different manufacturing techniques on the PVDF molecular chain and formation of different microstructures (X. Wan *et al.*, 2023)

The process of poling involves the following sequential steps:

- Sample Preparation: PVDF is typically available as a film or a sheet. Before poling, the material is cut or shaped into the desired dimensions suitable for its intended application.
- Electrodes: Two electrodes are positioned on opposite sides of the PVDF material. These electrodes serve as the positive and negative terminals during the poling procedure.
- Applying Electric Field: To initiate poling, a strong electric field is introduced to the PVDF material. This application of the electrical field helps to align the polar molecules within the material. The magnitude and duration of the electric field are determined based on specific requirements and the desired properties of the piezoelectric device.
- Temperature Control: Poling is often carried out at an elevated temperature to facilitate the alignment of molecules within the material. Precise temperature control is maintained throughout the poling procedure.
- Cooling: After the poling process concludes, the PVDF material undergoes a gradual cooling phase while remaining subjected to the electric field's influence. This step helps in retaining the desired molecular alignment achieved during the poling procedure.

The piezoelectric characteristics of PVDF can undergo substantial alteration based on the specific parameters governing the applied voltage, temperature, and duration during the poling process. Achieving optimal results requires careful determination of suitable parameters based on the specific materials, fillers, and dimensions of the sensor. However, this polling process and parameter optimization can be both time-consuming and costly. The traditional manufacturing process, which includes a post-poling procedure, is a two-step methodology involving the necessity of applying an electric field of no less than 120 MV/m at a temperature of 70 °C for an extended duration. (Ghosh *et al.*, 2016). As a result, it is advantageous to explore more efficient preparation methods that eliminate the need for post-poling steps.

In response to this challenge, numerous researches have been made to develop self-poling nanocomposites, where aligned dipoles are achieved without the need for subsequent processing steps. Several techniques, including spin coating, electrospinning, and the Langmuir-Blodgett method, have been explored. These methodologies leverage potent mechanisms such as shearing forces and electric fields to orient PVDF molecular chains in a specific direction. By employing these approaches, it is possible to create self-poling fabricate with aligned dipoles, thereby streamlining the fabrication process and enhancing the overall efficiency of the preparation.

Guo et al. (Guo, Nie, & Wang, 2021), developed a self-poling PVDF piezoelectric featuring a highly aligned β-phase through the application of a standard melting technique. They employed microinjection to generate a strong flow field, which can significantly influence the molecular chains and their orientations. This innovative approach has enabled the fabrication of a flexible PVDF generator with enhanced piezoelectric properties and improved crystalline structure orientation. Xiang et al. (X. Yuan *et al.*, 2022), employed mechanical stress to produce a self-poling nanocomposite. Their study revealed that the application of dual mechanically directional stress fields induced the transformation of the molecular chain as a dipole in PVDF. This transformation resulted in a significantly enhanced poling-free piezoelectric coefficient. Building on their research, the team utilized 3D-printing method to fabricate a self-powered circular pressure sensor which demonstrated exceptional performance (235 mV/kPa). Remarkably, the power density achieved was nearly 8 times higher compared to conventional, poled single-layer PVDF sensors. This novel method for self-polling has opened new avenues for the advancement of exceptionally efficient and responsive self-owered PVDF piezoelectric fabrications. In Figure 14, the diagram illustrates the process of polling through a directional stress field, as employed by the researchers.

In a separate study, Nick et al. (Nick A. Shepelin *et al.*, 2020) presented a novel and energy-efficient method to produce a PVDF-TrFE transparent piezoelectric film. They utilized the extrusion printing method to create dipole. This method yielded a striking increase of up to 500% in the piezoelectric charge coefficient (d33) when compared to pristine PVDF-TrFE printed through extrusion. Furthermore, they also noted corresponding improvements in energy harvesting capabilities, exemplified by a power of up to 20 μWcm^{-3} with the inclusion of 0.02 wt% SWCNTs. This advancement in manufacturing methods represents a significant enhancement in the piezoelectric generator's performance, positioning it as a highly effective and promising solution for transparent and flexible energy harvesting applications. In conclusion, Table 2 offers a comprehensive summary of the manufacturing techniques and performance metrics associated with PVDF piezoelectric materials developed by different researchers. The table provides valuable information regarding the types of fillers used, the manufacturing procedures employed, the polling conditions applied, and the resulting output voltage and power. This information serves as a valuable resource for understanding the diverse approaches and outcomes achieved in the field of PVDF piezoelectric research.

Certainly, exploring potential avenues for future research in the field of PVDF nanocomposite piezoelectric materials can lead to exciting developments. Here are some areas where current gaps in knowledge exist, along with suggestions on how researchers can address them:

- Self-Poled Nanocomposites: Investigate methods to design PVDF nanocomposites that inherently possess or develop piezoelectric polarization during their synthesis or processing stages. This would eliminate the need for external poling and enhance the convenience of using these materials. Explore how self-polling techniques can improve the long-term stability of piezoelectric properties in PVDF nanocomposites, especially under mechanical stress or

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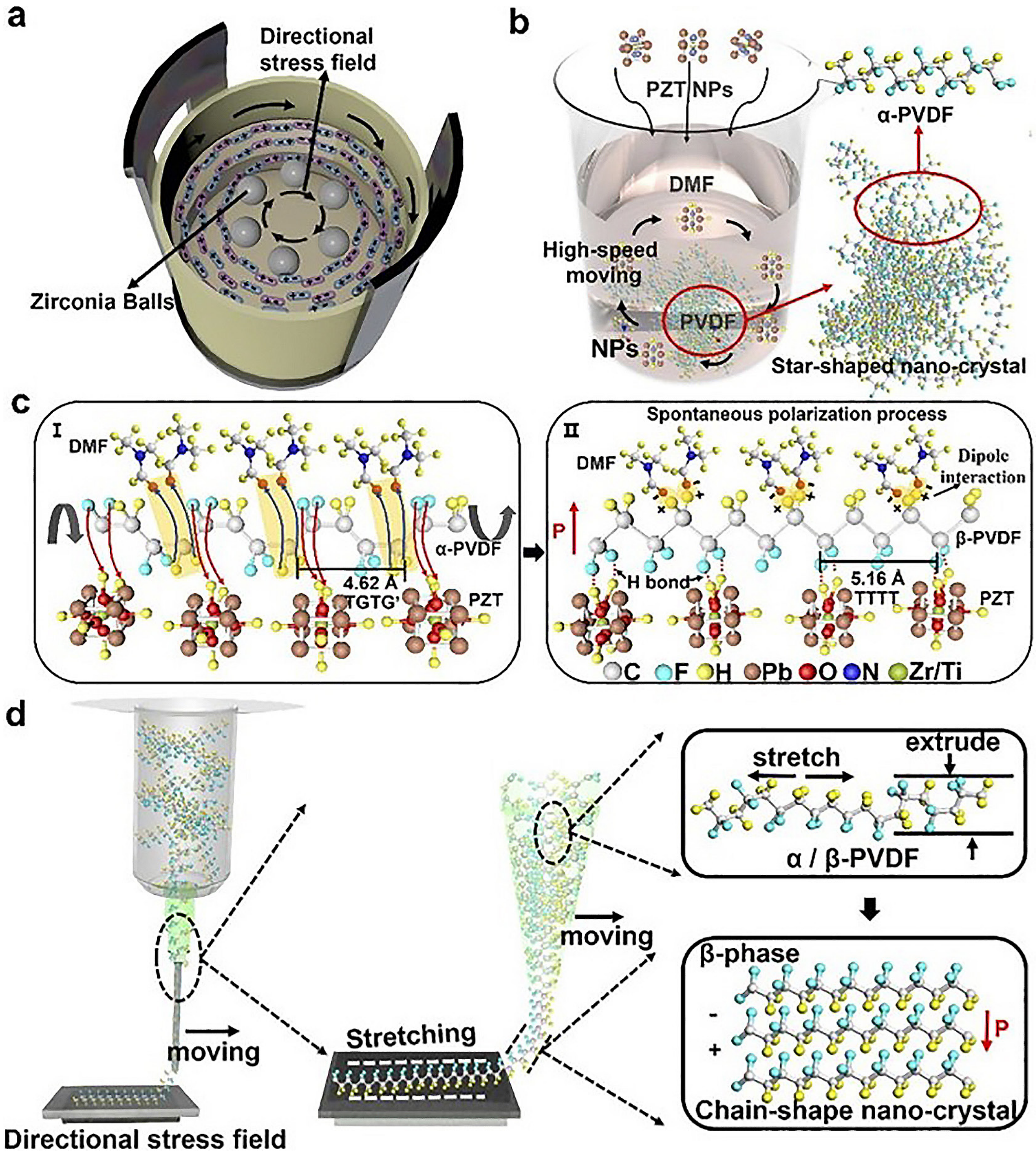

Figure 14. Schematic of self-poling process by mechanical stress field[114].

environmental conditions. This could involve the development of self-repair mechanisms or self-polarization maintenance strategies.

- Adaptive Materials: Research materials that can adapt their piezoelectric response based on external stimuli or changes in the environment. For example, materials that can self-adjust their properties in response to temperature variations, strain levels, or humidity changes.
- Characterization Techniques: Develop advanced characterization techniques to better understand the nanoscale behavior of PVDF nanocomposites. This includes studying the dispersion of nanoparticles, crystalline structures, and interface interactions to optimize material properties.
- Multifunctionality: Investigate the integration of multifunctionality into PVDF nanocomposites. Research could focus on designing materials that not only exhibit superior piezoelectric properties but also offer other functionalities, such as self-healing, energy storage, or sensing capabilities. This approach can broaden the scope of applications for these materials.

Materials	Nanoparticles	Manufacturing	Piezoelectric constant (d_{31}/d_{33}) pC/N	Polling Condition	Applied Pressure / load / Strain%	Output Voltage	Current density / Current	Power density / Power	Ref
PVDF	—	Electrospinning at 1.2 KV/cm	—	No poling	—	330 mV	—	—	(R. K. Singh *et al.*, 2021)
P(VDF-HFP)	$Eu(TTA)_3(TPPO)_2$ and GO	Electrospinning	$d_{33} = 4$	No poling	20 N, Frequency 3	4.5 V	35 nA	—	(G. Fu, Shi, Liang, *et al.*, 2022)
P(VDF-HFP)	Co-doped ZnO nanorods	Electrospinning at 12 KV	—	No poling	2.5 N	2.8 V	—	—	(Parangusan, Ponnamma, & Al-Maadeed, 2018)
PVDF	ZnO nanoparticle	Electrospinning at 16 KV	—	No poling	—	1.1 V	—	—	(Sorayani Bafqi, Bagherzadeh, & Latifi, 2015)
P(VDF-HFP)	Cellulose NC and Fe-doped ZnO	Electrospinning	—	No poling	2.5 N	12 V	1.9 $\mu A\ cm^{-2}$	490 $\mu W cm^{-3}$	(Deepalekshmi Ponnamma, Parangusan, Tanvir, & AlMa'adeed, 2019)
PVDF	Cellulose nanocrystals	Electrospinning at 15 KV	—	No poling	Forced by hammer	6.3	2 μA	—	(Fashandi *et al.*, 2016)
PVDF	SiO_2 nanoparticles	Electrospinning at 13 KV	—	No poling	13.9 N	24.6 V	—	—	(Haddadi, Ramazani SA, Talebi, Fattahpour, & Hasany, 2017)
PVDF	Ce-doped Fe_2O_3 nanoparticles	Electrospinning at 12 KV	—	No poling	2.5 N	20 V	0.01 $\mu A\ cm^{-2}$	700.64 $\mu W cm^{-3}$	(Parangusan, Ponnamma, & AlMaadeed, 2019)
PVDF	Ce-doped Co_3O_4	Electrospinning at 12 KV	—	No poling	2.5 N	15V	0.005 $\mu A\ cm^{-2}$	334.39 $\mu W cm^{-3}$	(Parangusan *et al.*, 2019)
PVDF	Nano-clay	Electrospinning at 12.5 KV	—	No poling	Finger tapping	70 V	20 nA	68 $mW\ cm^{-2}$	(S. Tiwari, Gaur, Kumar, & Maiti, 2019)
PVDF	Talc nanoparticles	Electrospinning at 18 KV	—	No poling	3.8 N	9.1 V	16.5 nA	1.12 $\mu W cm^{-2}$	(Shetty, Mahendran, & Anandhan, 2020)
PVDF	MoS_2 nanosheet	Electrospinning	—	No poling	Finger touch	14 V	8 nA	—	(Maity *et al.*, 2017)
PVDF	Ag nanoparticles	Electrospinning	—	No poling	1MΩ load resistance	2V	2μA	—	(Issa, Al-Maadeed, Luyt, Ponnamma, & Hassan, 2017)
PVDF	Ag nanowires	Electrospinning at 12 KV	30	No poling	—	—	—	—	(B. Li, Xu, Zheng, & Xu, 2014)
PVDF	Pt nanoparticles	Electrospinning at150 kVm^{-1}	44	No poling	0.3 MPa	30 V	6 $mA\ cm^{-2}$	22 $\mu W cm^{-2}$	(Ghosh & Mandal, 2018)
PVDF	MWCNTs	Electrospinning at 18 KV	—	No poling	4 MPa	6 V	—	81.8 nW	(Yu *et al.*, 2013)

Materials	Nanoparticles	Manufacturing	Piezoelectric constant (d_{31}/d_{33}) pC/N	Polling Condition	Applied Pressure / load / Strain%	Output Voltage	Current density / Current	Power density / Power	Ref
PVDF	CNT	Electrospinning	31.3	No poling	350N	1.89 V	11 nA	—	(C.-M. Wu, Chou, & Zeng, 2018)
PVDF	COOH functio-nalized CNTs & Ag-CNTs	Electrospinning at 15 KV	54pm V^{-1} for Ag-CNTs	No poling	—	—	—	—	(Sharma, Srinivas, Madras, & Bose, 2016)
PVDF	Graphene nanoplatelets	Electrospinning at 20 KV	—	No poling	—	7.9 V	4.5 μA	—	(Abolhasani, Shirvanimoghaddam, & Naebe, 2017)
PVDF	Ce^{3+} doped Graphene	Electrospinning at 12 KV	—	No poling	6.6 KPa	11 V	0.07 μA	0.56 μWcm^{-2}	(Garain, Jana, Sinha, & Mandal, 2016)
PVDF	Graphene and Poly benzoxazole	Electrospinning at 16 KV	—	No poling	—	60 V	—	—	(Barstugan, Barstugan, & Ozaytekin, 2019)
PVDF	CdS doped rGO	Electrospinning	—	No poling	Finger imparting	4 V	—	—	(Pusty, Sharma, Sinha, Chaudhary, & Shirage, 2017)
PVDF	Carboxylate and fluorinated graphene oxide	Electrospinning at 16 KV	63 forfluori-nated GO	No poling	—	—	—	—	(Gebrekrstos, Madras, & Bose, 2018)
PVDF	TiO_2-F_e3O_4-MWCNT	Electrospinning at 12 KV	51.42	No poling	1.32 N	0.68 V	—	—	(Samadi, Ahmadi, & Hosseini, 2019)
PVDF	halloysite nanotubes	Electrospinning	—	No poling	0.49 N	0.1 V	0.1 μA	—	(Abbasipour, Khajavi, Yousefi, Yazdanshenas, & Razaghian, 2017)
PVDF	Fe_3O_4-Graphene oxide	Electrospinning at 12 KV	—	No poling	1.32 N	0.23 V	—	—	(Samadi, Hosseini, & Mohseni, 2018)
PVDF	Hexagonal boron nitride	Electrospinning	—	No poling	—	68 V	100 nA	53.2 μWcm^{-2}	(Yadav, Raju, & Badhulika, 2020)
PVDF	Ionic Liquid	Electrospinning	—	No poling	—	48 V	—	47 μWcm^{-2}	(S. Tiwari *et al.*, 2021)
PVDF	Graphene oxide	Electrospinning	—	No poling	0.0075 KPa	23 V	3 mA	55 μWcm^{-2}	(Ramasamy, Rahaman, & Kim, 2021)
PVDF	ZnO nanorods	Electrospinning	—	No poling	13.3 KPa	0.356	0.456 mA	0.054 μWcm^{-2}	(Fakhri *et al.*, 2019)
PVDF	Halloysite nanotubes	Electrospinning	—	No poling	0.714 KPa	1.5 V	50 nA	15 $mWcm^{-2}$	(Abbasipour *et al.*, 2019)
PVDF	Tetra-n-butyl ammonium chloride	Electrospinning	—	No poling	—	17.2	70 nA	1.4 μWcm^{-2}	(Ekbote, Khalifa, Mahendran, & Anandhan, 2021)

Materials	Nanoparticles	Manufacturing	Piezoelectric constant (d_{31}/d_{33}) pC/N	Polling Condition	Applied Pressure / load / Strain%	Output Voltage	Current density / Current	Power density / Power	Ref
PVDF	Halloysite nano-tube and PANi	Electrospinning	—	No poling	—	7.2 v	0.75 mA	0.25 μWcm^{-2}	(Khalifa, Mahendran, & Anandhan, 2019)
PVDF	PDA@$BaTiO_3$	Electrospinning	—	No poling	—	8.2	0.4 mA	0.365 μWcm^{-2}	(Su, Li, *et al.*, 2021)
PVDF	PDA@BTO	Electrospinning	—	—	5.55 KPa	11 V	2.5 mA	7.639 μWcm^{-2}	(Su, Chen, *et al.*, 2021)
PVDF	Graphene oxide	Electrospinning	—	—	—	16 V	0.7 mA	2.8 μWcm^{-2}	(Jie Yang *et al.*, 2021)
PVDF-TrFE	AlN	Electrospinning	—	—	26.66 KPa	0.106	1.1 nA	0.003 μWcm^{-2}	(Jiang Yang *et al.*, 2021)
PVDF-TrFE	Ce/$BaTiO_3$	Electrospinning	—	—	0.0028 KPa	0.58 V	2.5 nA	0.00096 μWcm^{-2}	(Zhuang *et al.*, 2020)
PVDF-TrFE	ZnO NRs	Electrospinning	—	—	—	61 V	22 mA	25. 62 μWcm^{-2}	(Ye *et al.*, 2021)
PVDF-TrFE	1% ZnO+0.01% EGO	Electrospinning	—	—	—	0.4 V	0.23 nA	—	(Cherumannil Karumuthil, Prabha Rajeev, Valiyaneerilakkal, Athiyanathil, & Varghese, 2019)
PVDF-TrFE	MWCNTs	Electrospinning	—	—	—	18.23 V	2.1 mA	6.53 μWcm^{-2}	(C. Zhao, Niu, Zhang, Li, & Hu, 2019)
PVDF-TrFE	Polydopamine modified $BaTiO_3$	Electrospinning	—	—	—	6 V	1.5 mA	8780 μWcm^{-2}	(Guan, Xu, & Gong, 2020)
PVDF-TrFE	BZT-BCTNWs	Electrospinning	—	—	60 KPa	13.01 V	-	1.44 μWcm^{-2}	(Jie Liu *et al.*, 2020)
PVDF-TrFE	$BaTiO_3$ NWs	Electrospinning	—	—	—	12.6 V	1.3 mA	4.25 μWcm^{-2}	(Shi *et al.*, 2021)
PVDF-TrFE	Mxene	Electrospinning	—	—	200 KPa	1.58 V	—	3.64 μWcm^{-2}	(S. Wang *et al.*, 2021)
PVDF	TiO_2-Fe_3O_4-GO	Electrospinning at 12 kV	—	No poling	—	4.63 v	—	—	(Haji Abdolrasouli *et al.*, 2022)
PVDF-TrFE	Ag-decorated BCZT	Electrospinning at 10 KV and sol gel	—	50 kV/mm at 60 °C in an oil bath for 6 h.	—	3.5 V	—	4.5 mWm^{-1}	(Yan *et al.*, 2022)

Materials	Nanoparticles	Manufacturing	Piezoelectric constant (d_{31}/d_{33}) pC/N	Polling Condition	Applied Pressure / load / Strain%	Output Voltage	Current density / Current	Power density / Power	Ref
PVDF-HFP	$BaTiO_3$ nanoparticles	Solvent evaporation	—	100 $kVcm^{-1}$ at 100 °C for 20 h	0.23 MPa	75 V	15μA	—	(S.-H. Shin, Kim, Lee, Jung, & Nah, 2014)
PVDF-HFP	$BaTiO_3$ nanoparticles	Solvent evaporation	—	100 $kVcm^{-1}$ at 100 °C for 20 h	0.23 MPa	110 V	22 μA	0.48 Wcm^{-3}	(S.-H. Shin, Kim, Jung, Lee, & Nah, 2014)
PVDF	$BaTiO_3$ nanoparticles	Solvent evaporation	—	2 $KVcm^{-1}$ at for 8h	10 MPa	150 V	1.5μA	—	(Y. Zhao *et al.*, 2015)
PVDF	$BaTiO_3$ nanoparticles	Solvent casting	—	1 KV for 30 min at RT	100 MΩ	7.2	38 nA	0.8 $\mu W\ cm^{-2}$	(Sahu *et al.*, 2021)
PVDF	$BaTiO_3$ nanoparticles	Solvent evaporation	—	15 $MVcm^{-1}$ at 100 °C for 1 h	2 N	10 V	2.5 μA	—	(Yaqoob, Uddin, & Chung, 2017)
PVDF	ZnO nanoparticles	solution mixing	—	50 $KVcm^{-1}$ at 60 °C	—	—	—	—	(Indolia & Gaur, 2013)
PVDF	ZnO nanoparticles	Solution casting sol gel	–6.4	No poling	8.43 KPa	28 V	450 nA	0.4 μW	(Jana, Garain, Ghosh, Sen, & Mandal, 2016)
P(VDF-HFP)	Ni-dopped ZnO nanoparticles	Solution casting	20	No poling	2.5 N	1V	19-21 nA	—	(Parangusan, Ponnamma, & AlMaadeed, 2017)
PVDF	Fe- dopped ZnO nanoparticles	Solution casting	9.44	No poling	2.5 N	2.4 V	25 nA	1.17 μWcm^{-2}	(Parangusan, Ponnamma, & AlMaadeed, 2018)
PVDF	Zirconate titanate nanoparticles	Solution casting	—	No poling	16.5 KPa	25.7 V	1.2μA	8.22 μWcm^{-2}	(Si *et al.*, 2018)
PVDF	$GaFeO_3$ nanoparticles	Solvent casting	—	No poling	—	4 V	4 nA	—	(Mishra, Roy, Dash, & Mukherjee, 2018)
PVDF	PZT powder	Solution casting	84	10 $KVmm^{-1}$ at 80°C in silicone oil bath	—	—	—	—	(V. Tiwari & Srivastava, 2015)

Materials	Nanoparticles	Manufacturing	Piezoelectric constant (d_{31}/d_{33}) pC/N	Polling Condition	Applied Pressure / load / Strain%	Output Voltage	Current density / Current	Power density / Power	Ref
PVDF	NiO nanoparticles	Solution casting	—	No poling	—	—	—	—	(Dutta, Bose, Kar, Das, & Mukherjee, 2017)
PVDF	SiO_2 coated NiO nanoparticles	Solution casting	—	No poling	0.3 MPa	53 V	0.3 $\mu A\ cm^{-2}$	685 $W\ m^{-3}$	(Dutta, Kar, Bose, & Mukherjee, 2018)
PVDF-TrFE	MgO nanoparticles	Solution casting	–65	No poling	Finger tapping	2 V	—	—	(D. Singh, Choudhary, & Garg, 2018)
PVDF	$CoFe_2O_4$ nanoparticles	Solvent evaporation	33	Corona poling at 80°C for 0.5 h	—	—	—	—	(Pedro Martins *et al.*, 2012)
PVDF	Fe_3O_4 nanoparticles	Solution mixing	37	35 MVm^{-1} at 60°C for 1 h	—	—	—	—	(Z.-W. Ouyang, Chen, & Wu, 2015)
PVDF-HFP	Li dopped montmorillonite (Mt)	Solution casting	45	No poling	Pressing by finger	5V	50 nA	—	(Ma, Tong, Wang, An, & Zhang, 2018)
PVDF	Laponite nano-clay	Solvent evaporation	—	No poling	300 N	6 V	70 nA	—	(W. Rahman, Ghosh, Middya, & Mandal, 2017)
PVDF	SnO_2 nanosheets	Solution casting	36.52	No poling	0.3 MPa	42 V	6.25 $\mu A\ cm^{-2}$	4900 Wm^{-3}	(Kar *et al.*, 2019)
PVDF	MWCNTs	Solution casting	—	60 MVm^{-1}	—	3.7 V	—	—	(Ning *et al.*, 2013)
PVDF	MWCNTs coated with TiO_2	Solution casting	41	120 $V\mu m^{-1}$ at 70 °C for 1.2 h	—	—	—	—	(L. Yang, Ji, Zhu, Wang, & Qiu, 2016)
PVDF-TrFE	Graphene	Solution casting	34.3 ± 7.2	stepwise from 10 to 60 MVm^{-1}	20.37 MΩ	12.43	0.6 μA	148.06 Wm^{-3}	(L. Wu *et al.*, 2019)
PVDF	Graphite nanosheets	Solution casting	6.7	50 $kVmm^{-1}$ for 30 min at 130 °C	—	—	—	—	(Zhang *et al.*, 2013)
PVDF	Graphene-Ag doped nanosheets	Solution casting	—	No poling	5.2 KPa	0.1 V	0.1 nA	—	(Sinha *et al.*, 2016)

https://doi.org/10.37819/nanofab.008.1775

Materials	Nanoparticles	Manufacturing	Piezoelectric constant (d_{31}/d_{33}) pC/N	Polling Condition	Applied Pressure / load / Strain%	Output Voltage	Current density / Current	Power density / Power	Ref
PVDF	Reduced graphene oxide (rGo)	Solution casting	—	Stepwise poling at 60MVm^{-1} at 8 min intervals	Vibration test at 30 Hz	3.28 V	—	—	(J. Xue *et al.*, 2012)
PVDF	ZnO dopped rGO	Solution casting	—	no poling	—	—	—	—	(Jaleh & Jabbari, 2014)
PVDF	AlO dopped rGO	Solution casting	45	No poling	31.19 KPa	36 V	0.8 μA	27.97 μWcm^{-3}	(Karan *et al.*, 2016)
PVDF	Graphene oxide nanosheets	Solution casting	—	No poling	—	—	—	—	(El Achaby, Arrakhiz, Vaudreuil, Essassi, & Qaiss, 2012)
PVDF-HFP	Carbon black nanoparticles	Solution casting	—	polling at 90 MVm^{-1}	—	3.68 V	—	13 Wm^{-3}	(L. Wu *et al.*, 2014)
PVDF-HFP	$BaTiO_3$ NPs and hexagonal boron nitride nanolayers	Solution mixing	—	—	—	2.4 V	—	—	(Deepalekshmi Ponnamma & Al-Maadeed, 2019)
PVDF	TiO_2 nanolayers and rGO	Solution mixing	—	No poling	—	—	—	—	(Al-Saygh *et al.*, 2017)
PVDF	TiO_2 nanotubes and CNT	Solution casting	—	Corona poling at 8 KV for 7 s	2.5 N	1.3 V	—	—	(Deepalekshmi Ponnamma, Sharma, Saharan, & Al-Maadeed, 2020)
PVDF	MnO_2/graphene/ MWCNT hybrid	Solution casting and rolling	17-33	50-80 MV m^{-1}	—	—	—	—	(L. Yang *et al.*, 2018)
PVDF	—	Solution casting	–5	20 kV cm^{-1}	—	—	—	—	(Panigrahi, Sitikantha, Bhuyan, Panda, & Mohanta, 2021)
PVDF-TrFE	$BaTiO_3$ nanoparticles	Solution casting	—	—	—	—	—	0.28 μW	(Nunes-Pereira *et al.*, 2015)
PVDF	TiO_2	Solvent casting	—	40 KV cm^{-1}	—	—	—	—	(Kulkarni & Kumari, 2023)
PVDF-TrFE	—	Solution casting	d33 = –24	Corona poling at 20 KV at 80 degrees	1.5 MPa	8.9 V	—	—	(Xia *et al.*, 2023)

Materials	Nanoparticles	Manufacturing	Piezoelectric constant (d_{31}/d_{33}) pC/N	β-phase	Polling Condition	Applied load/ Pressure/ Strain%	Output Voltage	Current density / Current	Power density / Power	Ref
PVDF	Ionic Liquid	FDM	-	98.3	No poling	—	4.2 V	17.5 nAcm-2	—	(X. Liu et al., 2021)
PVDF	$BaTiO_3$	DIW	18	78	No poling	Finger tapping	4V	—	—	(Bodkhe et al., 2017)
PVDF	Graphene	DIW	–8.7	61.52	No poling	—	0.35 V	—	—	(C. Chen et al., 2019)
PVDF-TrFE	—	DIW	—	75-80	No poling	50 N	298.3 mV	—	—	(Nick A Shepelin et al., 2019)
PVDF	$BaTiO_3$/CNT	SLS+Sc–CO_2	2.6	79.5	corona polarization at RT for 60 min	—	19.3 V	415 nA	—	(C. Yang, Song, Chen, & Chen, 2021)
PVDF	Graphene	SLS	—	—	0.142 $KVcm^{-1}$	—	16.97 V	274 nA	—	(S. Song et al., 2021)
PVDF	$BaTiO_3$/Ag	SLS	8.1	—	40 $KVcm^{-1}$ for 30 min at 70 degrees	—	10 V	142 nA	—	(Shuai et al., 2020)
PVDF	$BaTiO_3$/Carbon	SLS	—	92.2	corona polarization with a voltage of 10 kV	—	5.7 V	79.8 nA	—	(Qi et al., 2021)
PVDF	Ionic Liquid	FDM	—	93.3	No poling	—	8.69 V	90.8 nA	—	(Jingfeng Liu, Shang, Shao, Liu, & Zhang, 2021)
PVDF	$BaTi_2O_5$	FDM	—	95.9	Corona polarization with 10 kV/mm for 30 min	—	13.4 V	142.5 nA	—	(X. Liu, Y. Shang, et al., 2022)
PVDF	tetraphenylphosphonium chloride (TPPC)/$BaTiO_3$	shear milling	4.2	85.2	10 kV/mm at 100 °C in a silicone oil bath for 6 h	—	11.5 V	220 nA	—	(Pei et al., 2022)
PVDF	Ionic Liquid	FDM	—	86.72	No poling	—	13 V	0.27 μAcm^{-2}	—	(L. Song, Dai, Li, Wang, & Zhang, 2021)

Materials	Nanoparticles	Manufacturing	Piezoelectric constant (d_{31}/d_{33}) pC/N	β-phase	Polling Condition	Applied load/ Pressure/ Strain%	Output Voltage	Current density / Current	Power density / Power	Ref
PVDF	$BaTiO_3$	SLS+Sc–CO_2	—	80.6	8 kV/mm polarization in an 80 °C silicone oil bath for 30 min	—	20.9 V	0.371 μAcm^{-2}	—	(C. Yang, F. Chen, et al., 2021)
PVDF	Ionic Liquid	FDM	—	97.4	No poling	—	8.2 V	300 nA	—	(X. Liu, J. Liu, et al., 2022)
PVDF	—	FDM	0.048	56.83	Corona poling	—	—	0.106 nA	—	(H. Kim, Torres, Wu, et al., 2017)
PVDF	Ionic Liquid	FDM	—	90	No poling	—	6 V	83 nA	—	(L. He et al., 2022)
PVDF	tetraphenyl-phosphonium chloride (TPPC)	FDM	–1.85	83.8	No poling	—	6.62 V	108.15 $nAcm^{-2}$	—	(Pei et al., 2021)
PVDF	—	DIW	—	—	Corona poling at 80 degrees	—	0.2 V	—	—	(Porter et al., 2017)
PVDF	—	FDM + electric poling assisted additive manufacturing (EPAM)	—	—	2 MV m^{-1}	—	—	037 nA	—	(Lee & Tarbutton, 2014)
PVDF	—	FDM + EPAM	0.66 pC/N	—	30MV/m	—	—	—	—	(Tarbuttona et al., 2017)
PVDF	1% MWCNT + 1.5% Ionic Liquid	FDM	—	76.8	No Poling	—	—	—	—	(H. Zhang et al., 2022)
PVDF	—	FFF, 3D-printing	0.72	—	electrode polling 16.5 MV/m	—	—	—	—	(Košir & Slavič, 2022)
PVDF/TrFE	SWCNT	Inkjet 3D printing	–12 pm/V	72.7	No poling	—	—	—	20 μWcm^{-3}	(Nick A. Shepelin et al., 2020)
PVDF	$BaTiO_3$	solvent evaporation	–18 pC/N	—	No poling	—	—	—	—	(Bodkhe, Noonan, Gosselin, & Therriault, 2018)
PVDF	Mxene	microinjection molding process	—	93.4	No poling	—	15.2 V	497.3 nA	18.9 μWcm^{-2}	(R. Han et al., 2021)

Materials	Nanoparticles	Manufacturing	Piezoelectric constant (d_{31}/d_{33}) pC/N	β-phase	Polling Condition	Applied load/ Pressure/ Strain%	Output Voltage	Current density / Current	Power density / Power	Ref
PVDF	PZT	Inkjet 3D printing	—	97.8	No poling	255 Kpa	60 V	—	0.9 mW cm^{-2}	(X. Yuan et al., 2022)
PVDF	$BaTiO_3$ and graphene quantum dots	Spin-coating	—	—	No polling	265 mN	4.6 V	4.13 pA cm^{-2}	11.2 μWcm^{-3}	(Bakar et al., 2018)
PVDF	Gd_5Si_4 nanoparticles	Spin-coating	—	—	No polling	2-3 N	1.2 V	—	—	(Harstad et al., 2017)
PVDF	ZnO nanowires	Spin-coating	—	—	1.2 MV cm^{-1}	3.2% strain	0.4 V	30 nA	—	(M. Choi et al., 2017)
PVDF-TrFE	ZnO nanoparticles	Spin-coating	32.2	—	no polling	65 g load	7.5 V	-	-	(J. Han et al., 2019)
PVDF-TrFE	$BaTiO_3$ nanoparticles	Spin-coating	—	—	100 MV m^{-1} at RT for 6 h	0.5 N mm-2	9.8 V	0.69 μA, 1.4 μA cm^{-2}	13.5 μWcm^{-2}	(Siddiqui et al., 2015)
PVDF	$BaTiO_3$	Spin-coating	—	—	—	—	6.7 V	2.4 μA	—	(Hu et al., 2018)
PVDF	$BaTi_2O_5$ nanorods	Hot pressing	—	—	20 kV mm^{-1} at 80 °C for 6h	22 MΩ	27.5 V	1.7 μA	27.4 μWcm^{-3}	(J. Fu et al., 2018)
PVDF	$BaTiO_3$ nanoparticles	Hot pressing	25	—	5kVmm^{-1} at 120 °C for 30 min	—	—	—	—	(R. Li, Zhao, Chen, & Pei, 2017)
PVDF	$FeTiNbO_6$ (FTN)	Hot pressing	28	—	poling in a silicone oil bath	—	—	—	110 μWcm^{-3}	(J. Fu et al., 2020)

Table 2. The performance of PVDF-based piezoelectric material using different manufacturing methods.

5. CONCLUSION

This review paper underscores the escalating significance of confronting the environmental and energy-related hurdles presented by conventional piezoelectric materials employed in manufacturing processes. Heightened concerns regarding excessive energy consumption and ecological repercussions have triggered an exploration of viable sustainable alternatives. Among these alternatives, PVDF has surfaced as a particularly promising avenue, offering an environmentally sound solution characterized by a resource-efficient production procedure. This paper meticulously examined the various manufacturing techniques for PVDF piezoelectric materials. As per the authors' viewpoint, a recent trend among researchers involves a shift towards self-poling techniques. This shift is driven by the understanding that the piezoelectric characteristics of PVDF can be markedly impacted by variables such as applied voltage, temperature, and duration during the poling phase. To achieve optimal outcomes, a judicious selection of these parameters is imperative, contingent upon the distinct properties of the materials, additives, and dimensions of the sensor. However, the traditional poling process and the intricate parameter fine-tuning it entails can be both time-intensive and financially burdensome. In light of this challenge, substantial endeavors have been directed towards the development of self-poling PVDF, which attains uniformly aligned dipoles devoid of supplementary post-processing steps. In fact, the upcoming research direction concerning PVDF piezoelectric materials revolves around devising manufacturing methods that prioritize cost-effectiveness, reliability, and environmental sustainability.

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