International Journal of Science and Research Archive

eISSN: 2582-8185

Cross Ref DOI: 10.30574/ijsra

Journal homepage: https://ijsra.net/

(REVIEW ARTICLE)

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Synthesis of Polyvinylidene Difluoride (PVDF)-Based Metal Oxide Nanoparticle Nanocomposite Thin Films: A Comprehensive Review

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International Journal of Science and Research Archive, 2025, 17(01), 813-818

Publication history: Received on 12September 2025; revised on 19 October 2025; accepted on 21 October 2025

Article DOI: https://doi.org/10.30574/ijsra.2025.17.1.2846

Abstract

Polyvinylidene fluoride (PVDF) is a multifaceted polymer known for its excellent piezoelectric, ferroelectric, and dielectric properties. The incorporation of metal oxide nanoparticles into PVDF matrices has led to the advancement of nanocomposite thin films with enhanced multifunctional characteristics. This review explores the synthesis techniques, material combinations, structural and functional properties, and emerging applications of PVDF-based metal oxide nanocomposite thin films. Emphasis is placed on the role of nanoparticle dispersion, phase transformation, and interfacial interactions in determining the performance of these advanced materials.

Keywords: PVDF - Metal oxide synthesis; Methods of synthesis; Characterization; Applications

1. Introduction

PVDF has emerged as a leading polymer for implementation in sensors [10], actuators, energy harvesting, and biomedical devices due to its unique electroactive properties. However, its performance can be significantly enhanced by incorporating inorganic fillers, particularly metal oxide nanoparticles. These nanocomposites exhibit synergistic effects [9], combining the flexibility and processability of PVDF with the functional attributes of metal oxides such as ZnO, TiO_2, $BaTiO_3$ and Fe_2O_3. The development of batteries with higher capacities [15] will also favor the use of the fastest and most powerful charging modes.

- PVDF is a semicrystalline polymer with a repeat unit of $CH_2 – CF_2$[**Source: Royal Society of Chemistry**]
- Polyvinylidene difluoride is a semicrystalline thermoplastic fluoropolymer. It is readily melted – processible (has the lowest melting point of 178 °C).
- PVDF possesses high permittivity, low dissipation factor [7][10]
- It can be fabricated by injection and compression molding. It integrates high mechanical strength with good processability.
- Strongest piezoelectric properties (ability to generate an electric charge in response to applied mechanical stress).
- It has a high purity with service temperature up to 150 °C.
- It is a flexible crystal.

About Zinc Oxide (ZnO):

- ZnO is a piezoelectric compound with a high piezoelectric coefficient.

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- Current research has shown that hybrid fibers consisting of ZnO nanofillers in PVDF polymer have demonstrated more efficient conversion of mechanical energy into electrical energy.
- Furthermore, good chemical resistance, higher mechanical strength [24], low cost and easy processability make them suitable for such device applications. Moreover, good crystalline nature with high mechanical flexibility makes PVDF/ZnO composite fiber a proper element for flexible electronic, piezo-tronic and piezo-phototronic devices [25] [5][7].
- In ZnO, Zn^{2+} and O^{2-} ions are bonded in a tetrahedral shape, making one bond slightly longer than the others. This distorted nature results in a permanent electrostatic dipole moment, which exhibits piezoelectric behavior [5][7][20].

2. Synthesis Techniques

2.1. Solution Casting

A widely used method involves dissolving PVDF in solvents such as DMF or acetone, followed by the scattering of nanoparticles and film formation through solvent evaporation. It is simple but may suffer from nanoparticle agglomeration.

2.2. Electrospinning

It produces nanofibrous films with high surface area and porosity. Electrospinning promotes the formation of the β-phase in PVDF and is suitable for sensor and filtration applications.

2.3. Spin Coating

It provides homogeneous thin films with controlled thickness. And is suitable for microelectronic applications where precision is critical.

2.4. Sol–Gel Processing

Used primarily for synthesizing metal oxide nano-particles before incorporation into PVDF. Offers sway over particle size and morphology.

2.5. In Situ Polymerization

This allows for better nanoparticle dispersion and interfacial bonding. Often used for functionalizing nanoparticles to improve compatibility with PVDF. Material and methods

Table 1 Metal Oxide Nanoparticles in PVDF

Nanoparticle	Key Properties	Impact on PVDF
ZnO	Piezoelectric, UV-sensitive	Enhances piezoelectric output and UV sensing
TiO_2	Photocatalytic, dielectric	Improves dielectric constant and photocatalytic activity
$BaTiO_3$	Ferroelectric, high permittivity	Boosts energy storage and ferroelectric behaviour
Fe_2O_3	Magnetic, catalytic	Adds magnetic functionality and catalytic potential

Table 2 Literature Review

Polymer Base element	Nanofillers	Synthesis	Results	Applications
Polyvinylidene fluoride (β-phase and	Fe_2O_3	Solvent casting method	Improve the optical and dielectric properties of polymer nanocomposites.	Batteries, Sensors, devices storing chemical energy and converting it

γ-phase)	Cu_2O	Solvothermal technique (Casting technique)	An improvement of the electroactive property and an increase of the thermal stability and the crystallinity of the PVDF.	into electricity(like powering smartphones, laptops, aerospace technology, electric vehicles (EVs), medical tools, and more. PVDF-ZnO nano-composites can be used as a UV shielding material because of their excellent absorption capability. [3],[4],[5],[6],[7],[8],[9],[10],[11],[12].
	ZnO (Combustion method)	Solvent casting method	improves the piezoelectric performance with a maximum observed sensitivity of 103 mV/N for zinc oxide incorporated devices.	
	ZnO/FeO hybrid nanomaterial	Electrospinning	Enhance the charge separation and influence the charge transfer reactions at the interface.	
	NiO particles with SiO_2		Highest Output Voltage – 53V	
Polyvinylidene fluoride (β – phase and γ – phase)	TiO_2	Polymer blending	Nanomaterials improve the fouling and the hydrophilicity of membranes. Amplify the mass transfer during the pre-evaporation technique. Enhance selectivity and solute rejection efficiency of the membrane. Improve the mechanical and thermal properties	Waste water treatment
	Ag – CuO/rGO nano-particles	Ag – CuO nano-particles by the co-precipitation method	The PVDF/Ag-CuO/rGO nano-composites demonstrate promising potential for polymer battery applications, offering high energy density,capacity retention, and mechanical flexibility.	Battery

3. Structural and Functional Characterisation

3.1. Crystallinity and Phase Analysis

- **XRD** reveals phase composition and crystallinity.
- **FTIR** identifies α and β phases of PVDF, with β-phase being crucial for piezoelectricity.

3.2. Morphology

- **SEM/TEM** used to observe nanoparticle dispersion and film uniformity.
- Agglomeration can reduce performance and must be minimized.

3.3. Thermal Properties

- **DSC/TGA** assess thermal stability and phase transitions.
- Nanoparticles often increase decomposition temperature and thermal resistance.

3.4. Electrical and Piezoelectric Properties

- Impedance spectroscopy and piezoelectric coefficient (d_{33}) measurements quantify enhancements.
- Nanocomposites show improved dielectric constant and piezoelectric output.

3.5. Mechanisms of Enhancement

- **β-Phase Promotion**: Nanoparticles act as nucleating agents, promoting the electroactive β-phase in PVDF.
- **Interfacial Polarization**: Interfaces between PVDF and nanoparticles contribute to increased dielectric properties.
- **Charge Transfer**: Metal oxides facilitate charge mobility, enhancing conductivity and sensor response.

3.6. Applications

3.6.1. Energy Harvesting

- Piezoelectric nano-generators using ZnO/PVDF and $BaTiO_3$/PVDF films.[12]

3.6.2. Sensors

- Pressure, UV, and gas sensors leveraging the multifunctionality of metal oxides.[11]

3.6.3. Biomedical Devices

- Electro-spun PVDF/TiO_2 scaffolds for tissue engineering and drug delivery.[10]

3.6.4. Flexible Electronics

- Thin film capacitors and transducers integrated into wearable devices.[8],[9]

3.7. Challenges and Future Directions[13],[14],[15]

- **Dispersion Control**: Preventing nanoparticle agglomeration remains a key challenge.
- **Scalability**: Transitioning lab-scale synthesis to industrial production.
- **Environmental Impact**: Developing green synthesis routes for nanoparticles.
- **Multifunctionality**: Designing composites with modified properties for specific applications.

4. Conclusion

PVDF-based metal oxide nanocomposite thin filmsrepresent a promising class of materials with wide-ranging applications. Advances in synthesis techniques and nanoparticle engineering have enabled significant improvements in their structural and functional properties. Continued research into interfacial phenomena, phase behaviour, and scalable fabrication will drive the next generation of smart materials.

Compliance with ethical standards

Disclosure of conflict of interest

No conflict of interest to be disclosed.

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