Advances in Nanostructured Functional Hydrogels for Energy Harvesting

Maulida Zakia[1], *Merreta Noorenza* Biutty[2], *Hermawan Dwi* Ariyanto[3], *Indrasukma* Permanadewi[1], *Ramavi* Akbar[1], *Irene Nindita* Pradnya[1] and *Marcelinus* Christwardana[4,5**]

[1]Department of Chemical Engineering, Faculty of Engineering, Universitas Negeri Semarang, Semarang, 50229, Indonesia
[2]Department of Chemical Engineering, Faculty of Industrial Engineering, Universitas Pembangunan Nasional "Veteran" Yogyakarta, Yogyakarta, 55283, Indonesia
[3]Department of Industrial Technology, Vocational College of Diponegoro University, Semarang, 50275, Indonesia
[4]Master Program of Energy, School of Postgraduate Studies, Diponegoro University, Semarang, 50241, Indonesia
[5]Research Collaboration Center for Electrochemistry, BRIN - Diponegoro University, Indonesia 50275

Abstract. Nanostructured functional hydrogels have recently emerged as highly versatile and promising materials for sustainable energy harvesting because of their tunable chemical composition, inherent mechanical flexibility, and outstanding ionic conductivity. Their unique three-dimensional polymer networks provide abundant space for the integration of nanostructures, which can significantly improve charge transport pathways, mechanical stability, and responsiveness to a wide range of external stimuli such as pressure, temperature, or humidity. By tailoring molecular design and incorporating functional nanomaterials, these hydrogels can be engineered to achieve superior electrochemical and mechanical performance. This review highlights recent progress in the rational design, fabrication strategies, and advanced applications of nanostructured functional hydrogels in energy harvesting technologies, with particular focus on triboelectric nanogenerators (TENGs), piezoelectric devices, and bioenergy systems. Finally, current challenges, emerging trends, and opportunities for developing next-generation hydrogel-based energy harvesters are critically discussed.

1 Introduction

The extensive consumption of non-renewable fossil fuels has raised global concerns regarding the risk of energy depletion and the associated environmental impacts. Hence, the transition toward clean and renewable energy sources has become essential. Renewable resources such as solar, wind, and tidal energy provide sustainable alternatives to fossil

[1] Corresponding author: maulida.zakia@mail.unnes.ac.id
** Co-corresponding author: marcelinus@live.undip.ac.id

© The Authors, published by EDP Sciences. This is an open access article distributed under the terms of the Creative Commons Attribution License 4.0 (https://creativecommons.org/licenses/by/4.0/).

fuels. However, their reliance on weather, season, and location restricts their ability to meet continuous energy demand, driving the development of technologies to effectively harvest and store energy from ambient sources. Energy-harvesting devices, including triboelectric nanogenerators (TENGs), piezoelectric nanogenerators, and biofuel cells have emerged as promising solutions to bridge this gap. These systems are capable of scavenging mechanical, thermal, and biochemical energy from the surrounding environment and converting it into usable electrical energy. Despite their promise, the utilization of energy-harvesting technologies still faces several challenges, including low energy conversion efficiency, limited mechanical durability under cyclic loading, and integration difficulties with flexible and biocompatible platforms.

Nanostructured functional hydrogels have recently emerged as an attractive class of materials to address these limitations [1-3]. Hydrogels can closely mimic biological tissues and accommodate large deformations without mechanical failure, owing to their three-dimensional polymer networks, high water content, mechanical softness, and ionic conductivity [1]. More importantly, their highly tunable chemical structure allows precise control over crosslinking density, porosity, and surface functionality. Additionally, the incorporation of various nanomaterials further enhances their electrochemical and energy conversion performance [4-6]. These unique characteristics make nanostructured hydrogels promising candidates for next-generation energy-harvesting devices.

This review summarizes recent progress in the development and application of nanostructured functional hydrogels for energy-harvesting technologies. We first discuss the design strategies and functionalization approaches that govern their mechanical, electrical, and electrochemical performance. Subsequently, their integration into representative devices, including TENGs, piezoelectric nanogenerators, and biofuel cells also highlighted. Finally, we outline the challenges and future directions for advancing hydrogel materials toward practical, scalable, and multifunctional energy-harvesting systems. This review aims to provide researchers with a comprehensive understanding of the potential of nanostructured hydrogels as a platform for self-powered technologies.

2 Design Strategies for Nanostructured Functional Hydrogels

The performance of hydrogel-based energy-harvesting devices are primarily controlled by the structural architecture and chemical functionalization of the hydrogel matrix. These approaches include polymer network engineering, nanostructure incorporation, and stimuli-responsive functionalization, as depicted in Fig 1.

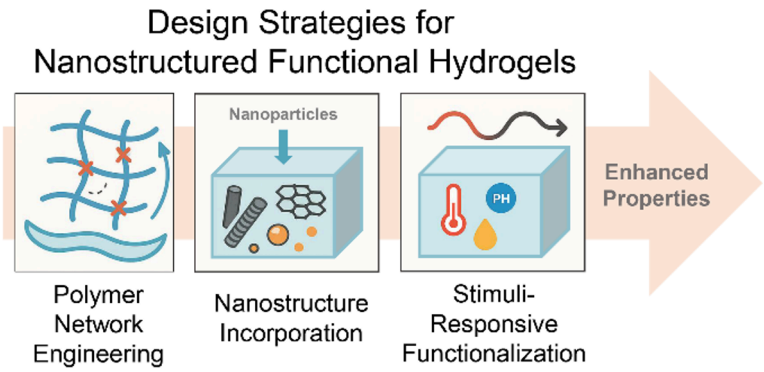

Fig. 1. Nanostructured Hydrogel

2.1 Polymer Network Engineering

Controlling hydrogel crosslinking, porosity, and hydrophilicity tunes their mechanics and ion transport, directly impacting energy-harvesting performance [1]. Tuning the crosslinking density allows a balance between mechanical robustness and flexibility, preventing brittle fracture while maintaining sufficient elasticity for cyclic deformation. Similarly, modulating porosity enhances ion diffusion and facilitates rapid charge transport within the three-dimensional network, thereby improving the overall electrochemical response.

Recent studies have highlighted double-network hydrogels, in which a rigid primary network and a ductile secondary network work synergistically to enhance mechanical resilience and dissipative capacity under applied stress [2]. In parallel, supramolecular hydrogels, which rely on reversible non-covalent interactions have been developed to deliver self-healing capabilities and dynamic responsiveness [3]. These characteristics are valuable for wearable and flexible energy-harvesting devices, where repeated mechanical loading and potential micro-damage can compromise long-term stability. By integrating these structural design principles, researchers can create hydrogel systems that combine high stretchability, rapid recovery, and efficient ion transport. Thus, ensuring consistent performance in dynamic and mechanically demanding environments.

2.2 Nanostructure Incorporation

The incorporation of nanomaterials into hydrogel matrix has emerged as a powerful strategy to enhance their physicochemical and electrochemical properties. Nanostructures such as carbon nanotubes (CNTs), graphene, and metal nanoparticles (i.e. Au, Ag, Pt) have been widely employed as functional additives to significantly improve the electrical conductivity, dielectric constant, and specific surface area of hydrogels [4-6]. These nanomaterials create interconnected conductive pathways within the hydrogel, enabling fast charge transport and improving energy-conversion efficiency. Furthermore, the high surface-to-volume ratio of nanomaterials provides abundant electroactive sites, promoting efficient charge separation and transfer at the electrode–electrolyte interface [5]. For instance, graphene-based hydrogels demonstrate enhanced mechanical robustness and electrochemical stability, attributed to the synergistic interactions between graphene sheets and the polymer network [4]. Metal nanoparticles having catalytic activity can be used in biofuel cells and electrocatalytic energy-conversion systems [5]. Overall, the integration of nanomaterials not only improves the electrical and mechanical properties of hydrogels but also allows for the design of multifunctional, stimuli-responsive hybrid systems that can adapt to dynamic operational environments in advanced energy-harvesting devices.

2.3 Stimuli-Responsive Functionalization

The functionalization of hydrogels with stimuli-responsive moieties has opened new opportunities for creating adaptive and intelligent energy-harvesting systems. The incorporation of functional groups or supramolecular structures into hydrogel can dynamically modulate its swelling behavior, ionic conductivity, and mechanical properties [7,8]. For example, thermo-responsive hydrogels composed of poly(N-isopropylacrylamide) (PNIPAM) display a reversible volume phase transition near physiological temperature,

thereby facilitating controlled ion transport and improved energy conversion efficiency under fluctuating thermal conditions [7]. Additionally, pH-sensitive hydrogels functionalized with weak acidic or basic groups can alter their ionization state and swelling ratio in response to local pH changes leading to enhance performance of biofuel cells and implantable energy devices operating in physiological environments [8]. Such stimuli-adaptive functionality enables the design of smart hydrogel-based energy harvesters capable of self-regulation, enhanced durability, and real-time adaptability to dynamic environmental or biological conditions. These advances pave the way for the development of next-generation self-powered systems for wearable electronics, implantable medical devices, and autonomous sensing platforms.

3 Applications in Energy Harvesting

Nanostructured functional hydrogels exhibit considerable promise for various energy-harvesting applications, owing to their tunable mechanical, electrical, and electrochemical properties, as illustrated in Fig 2.

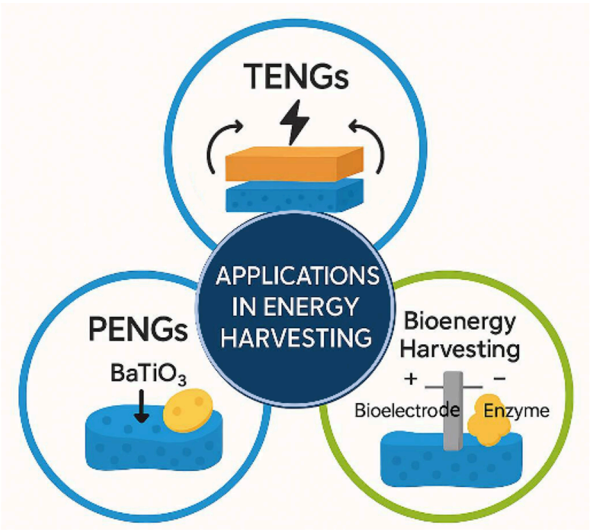

Fig. 2. Nanostructured functional hydrogels applications

3.1 Triboelectric Nanogenerators (TENGs)

Hydrogel-based triboelectric nanogenerators (TENGs) are a promising class of energy-harvesting devices that convert mechanical energy into electricity through the triboelectric effect [9]. The high-water content of hydrogels allows conformal contact with irregular surfaces, enhancing effective contact area and triboelectric charge generation. In addition, the high ionic conductivity of hydrogels allows them to serve as effective ionic conductors or electrodes to promote charge transport during mechanical activation in TENGs. Recent studies have reported hydrogel-based TENGs capable of harvesting biomechanical energy from human motion [10], respiration [11], or joint movement [12]. Such systems demonstrate excellent mechanical compliance, durability over thousands of

operation cycles, and compatibility with flexible and skin-mounted electronics, underscoring their potential for wearable and self-powered health-monitoring devices.

3.2 Piezoelectric Nanogenerators

The incorporation of piezoelectric nanostructures such as barium titanate ($BaTiO_3$), zinc oxide (ZnO), or poly(vinylidene fluoride) (PVDF)-based nanofillers into hydrogel matrix has been recognized as an effective strategy for developing flexible piezoelectric energy-harvesting systems [13-15]. These hybrid materials take advantage of the piezoelectric effect to generate electrical potential. Integration of piezoelectric nanostructures also significantly improves the sensitivity and energy conversion efficiency of the system. The porous and stretchable network of the hydrogel facilitates uniform dispersion of piezoelectric particles, maximizing their effective surface area and enabling efficient stress transfer from the matrix to the active phase [13]. Recent studies have shown that $BaTiO_3$–hydrogel composites can achieve higher output voltages and currents compared to pristine hydrogels, making them attractive for biomechanical energy harvesting from motions such as bending, pressing, and stretching [14]. Similarly, ZnO–hydrogel hybrids have been used to create flexible piezoelectric nanogenerators capable of real-time physiological monitoring [15]. In addition to improved electromechanical performance, the combination of piezoelectric nanostructures with conductive or ionic hydrogels offers multifunctionality, including self-healing, biocompatibility, and stretchability, which are highly desirable for wearable and implantable energy devices. These advances highlight the potential of piezoelectric hydrogel hybrids as next-generation power sources for soft electronics, medical implants, and autonomous sensing platforms.

3.3 Bioenergy Harvesting

Nanostructured hydrogels have emerged as promising bioelectrode materials for enzymatic biofuel cells (EBFCs), owing to their unique ability to provide a hydrated, three-dimensional, and biocompatible microenvironment that preserves enzyme activity and facilitates efficient mass and charge transport [16]. The high-water content and interconnected porous network of hydrogels allow rapid diffusion of substrates (e.g., glucose, lactate) and oxygen to the immobilized biocatalysts, thereby enhancing the overall reaction kinetics. Moreover, the soft and flexible nature of hydrogels enables intimate contact with biological tissues, making them highly attractive for implantable and wearable biofuel cell systems. Incorporation of nanostructures such as carbon nanotubes, graphene, and metal nanoparticles within the hydrogel matrix has been shown to significantly improve the electrical conductivity and electron transfer rate between the redox centers of enzymes and the electrode surface. For example, CNT- or graphene-functionalized hydrogels have achieved higher power densities by lowering charge-transfer resistance and promoting direct electron transfer (DET) pathways. Additionally, functionalization with redox mediators or conductive polymers (e.g., polypyrrole, PEDOT:PSS) further boosts current output and operational stability. Recent studies have demonstrated that nanostructured hydrogel-based EBFCs can deliver long-term stable power generation under physiological conditions, enabling applications in self-powered biosensors, continuous glucose monitoring systems, and even low-power implantable medical devices. These advances

underscore the potential of nanostructured hydrogels as a versatile platform for next-generation bioenergy harvesting and biomedical power supply technologies.

4 Conclusion and Future Perspectives

Despite the remarkable advances achieved in recent years, hydrogel-based energy harvesting technologies still face several critical challenges that must be addressed before they can be widely adopted in practical applications. One of the most pressing issues is the limited long-term stability of hydrogels, as their high-water content often leads to dehydration under ambient conditions, resulting in performance degradation over time. In addition, the scalability of current manufacturing processes remains a bottleneck, as many fabrication methods are either costly, time-consuming, or difficult to reproduce consistently at industrial levels. Addressing these limitations will require the design of advanced hydrogels with antifreeze, anti-drying, and self-healing properties, enabling them to maintain structural integrity and functional performance even under harsh environmental conditions. Future research directions should also emphasize the integration of hydrogels with wireless data transmission systems to create self-powered wearable and implantable electronic devices that can operate continuously without external power sources. Furthermore, the use of machine learning-guided material design, coupled with advanced nanofabrication techniques, offers exciting opportunities to accelerate the discovery and optimization of hydrogel systems. By combining predictive modelling with high-throughput experimental approaches, researchers can rapidly identify next-generation nanostructured hydrogels with superior efficiency, multifunctionality, and adaptability for diverse energy harvesting applications.

Acknowledgements

This work was supported by DPPM Kemdiktisaintek 2025, grant number 94.2.6/UN37/PPK.11/2025 date 2 June 2025.

References

[1]. Z. Wang, N. Li, Z. Zhang, X. Cui, H. Zhang, Nanoenergy Adv. **3**, 315 (2023)

[2]. K. Ou, M. Wang, C. Meng, K. Guo, N.S. Emon, J. Li, K. Qi, Y. Dai, B. Wang, Compos. Sci. Technol. Composites Science and Technology **255**, 110732 (2024)

[3]. Y. Liu, M. Zhou, Y. Liu, X. Han, X. Zhang, S. Liu, Supramol. Chem., **32**, 445 (2020)

[4]. P. Manchi, M.V. Paranjape, A. Kurakula, V.S. Kavarthapu, C. Kim, J.S. Yu, Nano Energy, **142,** 111184 (2025)

[5]. B. Bagchi, P. Datta, C.S. Fernandez, L. Xu, P. Gupta, W. Hung, A.L. David, D. Siassakos, S. Homer-Vaniasinkam, M.K. Tiwari, Nano Energy, **107,** 108127 (2023)

[6]. G. Tan, N. Xu, D. Gao, X. Zhu, Energy, **258,** 124843 (2022)

[7]. B. Zhao, K. Lu, W. Zhang, C. Jin, Q. Xuan, G. Pei, Solar Energy, **288,** 113306 (2025)

[8]. R. García-Sobrino, I. Casado-Losada, L. Bruno-Pérez, C.García, H. Reinecke, C. Elvira, J. Rodríguez-Hernández, A. Gallardo, E. Martínez-Campos, Eur. Polym. J., **169**, 111131 (2022)

[9]. F.G. Torres, O.P. Troncoso, G.E. De-la-Torre, Int. J. Energy Res., **46,** 5603 (2022)

[10]. B. Xie, Y. Ma, N. Luo, Y. Chen, Y. Liu, K. Nie, Y. Jia, R. Yin, Y. Liu, Nano Energy, **130,** 110095 (2024)

[11]. Q. Yang, M. Yu, H. Zhang, N. Li, J. Du, L. Xu, J. Xu, Nano Energy, **134,** 110530 (2025)

[12]. G. Feng, Y. Wang, D. Liu, Z. Cheng, Q. Feng, H. Wang, W. Han, C. Jia, Materials, **18,** 33 (2025)

[13]. Y. Khazani, E. Rafiee, A. Samadi, M. Mahmoodi, Coll. Surf. A. Physicochem. Eng. Asp., **687,** 133537 (2024)

[14]. Z. Wang, Z. Liu, G. Zhao, Z. Zhang, X. Zhao, X. Wan, Y. Zhang, Z.L. Wang, L. Li, ACS Nano, **16,** 1661 (2022)

[15]. X. Song, H. Zou, S. Cao, B. Jiang, M. Li, L. Huang, Y. Zhang, Q. Yuan, Chem. Eng. J., **475,** 146184 (2023)