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Waste-derived hydroxyapatite reinforced piezoelectric geopolymer hybrid composite for sustainable energy-harvesting applications

Arun Murugesan[1], Nidhya Rathinavel[1], Karthick Jaisankar[1], Balaji Ravi[1], and Natrayan Lakshmaiya[2,*]

[1] *Department of Civil Engineering, PSG Institute of Technology and Applied Research, Neelambur, Coimbatore 641062, India*
[2] *Department of Research and Innovation, Saveetha School of Engineering, SIMATS, Chennai, Tamil Nadu 602105, India*

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ABSTRACT

Traditional piezoelectric materials are expensive and environmentally unsustainable, limiting their adoption in energy-harvesting building applications. This study proposes an eco-friendly approach for developing a waste-derived hydroxyapatite (HAP)-reinforced piezoelectric geopolymer hybrid composite using eggshell waste and fly ash. Eggshell-derived calcium carbonate was converted into hydroxyapatite through a controlled precipitation process, and the synthesized HAP was characterized for structural purity, morphology, and dielectric properties. Geopolymer composite specimens of 50 mm cubic geometry were fabricated by incorporating 15 wt% HAP, 25 wt% ZnO, and 10 wt% TiO_2 into a Class F fly ash-based geopolymer matrix and cured at 70 °C for 48 h. Under uniaxial compressive loading, the hybrid composite generated a maximum piezoelectric voltage output of 0.149 V at a compressive stress of 22.12 MPa, while maintaining a compressive strength suitable for non-structural construction applications. Microstructural analysis confirmed uniform dispersion of HAP within the geopolymer matrix, contributing to enhanced interfacial polarization and dielectric response. The results demonstrate that waste-derived HAP can function as an effective, lead-free piezoelectric phase, enabling the development of sustainable, energy-responsive geopolymer materials for self-sensing and energy-harvesting applications.

1 Introduction

The global construction industry faces increasing pressure to mitigate its environmental footprint while maintaining structural performance. Ordinary Portland cement (OPC), the dominant binder in construction, is responsible for nearly 8–10% of global CO_2 emissions due to its energy-intensive clinkerization process and limestone calcination [1–3]. To achieve carbon neutrality, the development of sustainable, multifunctional alternatives to OPC has become imperative [4–6]. Geopolymer binders, synthesized through the alkaline activation of aluminosilicate rich by-products such as fly ash, slag, or metakaolin, have emerged as a

Address correspondence to E-mail: natrayanphd@gmail.com

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viable substitute. These binders exhibit high compressive strength, chemical stability, and up to 80% lower CO_2 emissions than OPC. Beyond sustainability, geopolymers serve as adaptable matrices for incorporating functional materials, enabling smart features such as self-sensing and energy harvesting [7–10].

Among these functionalities, piezoelectricity, the ability to generate electrical energy from mechanical stress, offers unique opportunities for self-powered sensing and energy-harvesting infrastructure [11, 12]. Embedding piezoelectric phases within geopolymer matrices can transform passive structural components into active systems capable of monitoring stress, vibration, or damage in real time. However, conventional piezoelectric ceramics such as lead zirconate titanate (PZT) contain toxic lead and require high sintering temperatures, conflicting with environmental and sustainability objectives [13–15].

Hydroxyapatite (HAP), a calcium phosphate compound ($Ca_{10}(PO_4)_6(OH)_2$), has emerged as a promising, lead-free alternative. HAP possesses a non-centrosymmetric hexagonal crystal structure that enables moderate piezoelectric response, along with excellent biocompatibility and chemical stability [16, 17]. While traditionally used in biomedical applications, recent studies suggest its potential as a functional piezoelectric phase for composite materials. Importantly, HAP can be synthesized from low-cost, renewable precursors, making it attractive for sustainable engineering applications [17].

Specifically, eco-efficient method for HAP synthesis involves the valorization of eggshell waste, a calcium rich by-product of the food industry. Globally, millions of tons of eggshells are discarded annually, creating disposal and environmental challenges [6, 18–22]. Since eggshells consist primarily of calcium carbonate ($CaCO_3$), they can be thermally converted to calcium oxide (CaO) and subsequently reacted with phosphoric acid to produce high-purity HAP. This conversion not only reduces waste and landfill burden but also aligns with circular-economy and low-energy manufacturing principles [23, 24].

Despite its sustainability advantages, the integration of waste-derived HAP into geopolymer matrices remains underexplored, particularly regarding its impact on piezoelectric behaviour. The hydroxyl rich geopolymer network offers favourable interfacial bonding with HAP, potentially enhancing dielectric and electromechanical performance. Additionally, incorporating semiconducting oxides such as zinc oxide (ZnO) and titanium dioxide (TiO_2) can create synergistic effects that improve charge mobility and polarization within the hybrid composite.

This study aims to synthesize hydroxyapatite from eggshell waste and evaluate its performance as a piezoelectric additive in fly ash, based geopolymer composites. The work investigates structural, dielectric, and electromechanical properties of hybrid systems containing HAP, ZnO, and TiO_2 nanoparticles. Comprehensive characterization using X-ray diffraction (XRD), scanning electron microscopy (SEM), and energy-dispersive spectroscopy (EDS) was conducted to confirm phase formation, morphology, and interfacial bonding. By integrating waste valorization, sustainable geopolymer chemistry, and functional material design, this research contributes to developing eco-efficient, energy-responsive construction materials. The outcomes demonstrate a pathway toward self-sensing and energy-harvesting infrastructure that advances both environmental sustainability and material innovation.

2 Research gap and novelty

Previous research has examined piezoelectric fillers in cement and geopolymer matrices, yet there has been minimal research on using waste-derived hydroxyapatite as a multifunctional additive. The research still lacks understanding about how eggshell-derived HAP and ZnO and TiO_2 nanoparticles interact to produce electromechanical coupling effects in geopolymer systems.

The present study addresses these gaps by developing a low-temperature, lead-free, waste-derived hybrid composite and systematically investigating its structural, dielectric, and piezoelectric performance. The research introduces an innovative method that combines biogenic calcium materials with geopolymer binders to create sustainable energy-harvesting capabilities.

3 Materials and methods

The experimental program for the geopolymer-piezoelectric composite consists of the principles of piezoelectric generation, raw material selection, HAP characterisation, and geopolymer-piezoelectric composite performance evaluation under mechanical stress.

consistent piezoelectric performance and durability over a long period of time [44, 45].

The hybrid composite's compressive strength was found to be 22.12 MPa, which indicates that the addition of functional filler did not affect the mechanical performance of the composite as much as the case of conventional geopolymers with similar composition. The strength can be used for non-structural and light-load-bearing applications. The small reduction in compressive strength that can be observed in the hybrid composite with respect to pure geopolymer reference samples can be explained by the presence of rigid particles which hinder gel formation. Nevertheless, the trade-off is compensated by the additional electrical functionality.

5 Potential applications and limitations

The developed hybrid geopolymer, hydroxyapatite composite offers significant potential for smart and sustainable construction applications. Its ability to convert mechanical stress into electrical output enables use in self-sensing components, energy-harvesting pavements, and façade or flooring systems where electromechanical responsiveness enhances performance. The low-temperature synthesis and reliance on waste-derived resources make the material particularly suitable for environmentally conscious infrastructure projects. However, certain limitations must be addressed before large-scale implementation. The generated voltage, while measurable, remains relatively low compared with conventional piezoelectric ceramics, necessitating optimization of phase dispersion, poling, and interfacial bonding. Additionally, long-term durability under cyclic loading and variable environmental conditions requires further evaluation. Despite these challenges, the material's scalability, low embodied energy, and compatibility with standard cementitious processing position it as a promising candidate for next-generation, self-powered sensing elements in sustainable construction systems.

6 Conclusions

The research ensures the sustainable and operational way for developing piezoelectric geopolymer composites from the hydroxyapatite prepared from waste eggshell powder. The key features of the study includes eggshell waste has been turned into high-purity hydroxyapatite using a controlled wet-chemical precipitation method at room temperature. XRD confirmed the presence of crystalline HAP with hexagonal symmetry and an average crystallite size of about 45 nm. EDS analysis gave a Ca/P ratio of 1.66, which is in line with stoichiometric hydroxyapatite. The addition of 15 wt % HAP, along with ZnO (25%) and TiO_2 (10%) nanoparticles, to the fly ash geopolymer matrix resulted in a composite that was dense and homogeneous. Further, the hybrid composite generated a voltage output of 0.149 V under 22.12 MPa compressive stress, demonstrating effective electromechanical coupling. The synergistic interaction between HAP, ZnO, and TiO_2 contributed to improved charge polarization and dielectric stability. The compressive strength of the modified geopolymer stayed at approximately 22 MPa, adequate for non-heavily structural uses, hence electricity function was gained without substantial trade-off in the load-bearing ability.

The research ensures that development of eco-friendly piezoelectric geopolymer composites is feasible by utilizing hydroxyapatite from eggshells in combination with fly ash. The method paves the way for environmentally friendly and low-temperature thermal smart building materials. Composites are primarily recommended for non-structural applications like façade panels, tiles, and pavements, where electromechanical responsiveness can be a significant functional added value. Overall, the present framework, is technically sound and sustainable for the use of waste-derived, energy-responsive geopolymer composites in smart infrastructure.

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Author Contributions

Arun Murugesan: Conceptualization; Methodology; Experimental work; Data curation; Formal analysis; Writing – original draft preparation. Nidhya Rathinavel: Material preparation; Laboratory investigations; Critical review and editing; Visualization; Data interpretation; Writing – review and editing. Karthick Jaisankar: Hydroxyapatite synthesis; Characterization studies (XRD, SEM, EDS); Validation; Draft editing. Balaji Ravi: Geopolymer formulation; Mechanical testing; Resources; Supervision of laboratory procedures. Natrayan Lakshmaiya: Conceptualization; Supervision; Project administration; Interpretation of results; Final approval of the manuscript. All authors have read and approved the final manuscript.

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Data availability

All data supporting the findings of this study are available from the corresponding author upon reasonable request.

Declarations

Competing Interest The authors declare no competing interests.

Ethical approval This study did not involve human participants or animal testing. All experimental procedures complied with institutional environmental and safety regulations.

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