Received: 11 August 2025 | Revised: 22 September 2025 | Accepted: 13 October 2025 | Published online: 28 October 2025

RESEARCH ARTICLE

Smart Wearable Technology
2025, Vol. 00(00) 1–7
DOI: 10.47852/bonviewSWT52027158

Influence of Nano Zinc Oxide Reinforcement on the Diffusion Coefficient of Unsaturated Polyester and Nitrile Butadiene Rubber Composites

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Abstract: This research investigates the influence of zinc oxide (ZnO) nanoparticle reinforcement on the diffusion coefficient of epoxy/nitrile butadiene rubber (EP/NBR) composites. The study evaluates various ZnO concentrations (1%, 2%, and 3% by weight) incorporated into the EP/NBR matrix. The dispersion of ZnO nanoparticles (ZnONP) was analyzed using X-ray diffraction and scanning electron microscopy, revealing improved particle dispersion with increased ZnO content, contributing to better structural stability. Diffusion coefficient measurements in water, nitric acid, and sodium hydroxide (NaOH) solutions indicated that higher ZnO concentrations reduce the diffusion coefficient for water absorption, enhancing moisture resistance. However, a slight increase in diffusion coefficients was observed in NaOH solutions, demonstrating the modified composite's variable interaction with different environmental conditions. The findings emphasize the role of ZnONPs in enhancing the mechanical and physical properties of EP/NBR composites, particularly in improving their resistance to moisture and alkaline environments, thereby broadening their potential applications in moisture-sensitive and chemically aggressive environments.

Keywords: zinc oxide (ZnO), polymer nanocomposites, nanoparticle dispersion, diffusion coefficient, moisture absorption

1. Introduction

Composites represent solid materials consisting of two or more distinct phases that are deliberately combined to produce enhanced properties, setting them apart from traditional homogeneous mixtures like alloys or blends [1]. Unlike alloys, which are typically made from metal combinations exhibiting similar physical characteristics, or homogeneous mixtures in which components are evenly distributed, composites are designed to bring together materials that possess varied and complementary properties [2]. These materials can range from metals and polymers to ceramics and glass, strategically integrated to create a material that surpasses the individual properties of its constituents. The choice of these materials depends on the specific application requirements, aiming to achieve optimal mechanical, thermal, and electrical properties [3, 4]. In recent years, the development of polymer–nanoclay composites has significantly advanced the field of material science. With the improvement of fabrication techniques, researchers have been able to design advanced composites with superior performance characteristics [5, 6].

These innovations have opened up new possibilities for various industries, particularly those that require materials with high strength, durability, and reduced weight. Applications in aerospace, automotive, construction, petrochemical industries, biomedicine, and wastewater treatment have become increasingly reliant on these advanced composites [7]. Polymer nanocomposites exhibit remarkable attributes, such as increased density, enhanced mechanical strength, large surface areas, and elevated elastic moduli, in addition to flame resistance, and superior thermomechanical, optoelectronic, and magnetic properties [8, 9].

These qualities are largely due to the inclusion of nanoparticles, such as nanoclays, which interact at the molecular level with the polymer matrix. This interaction significantly alters the composite's overall performance by improving strength, durability, and other crucial properties. The high surface area and the ability of nanoparticles to disperse uniformly within the polymer matrix contribute to the composite's enhanced mechanical properties [10]. Among the variety of nanoparticles used in composites, zinc oxide (ZnO) has garnered significant attention due to its semiconducting nature, as well as its optical, electronic, and photonic characteristics. ZnO's properties make it an ideal candidate for enhancing the functionality of polymer nanocomposites [11].

Building upon these foundational studies, the present research aims to evaluate the influence of ZnO nanoparticle (ZnONP) incorporation on the physical properties of epoxy/nitrile butadiene rubber (EP/NBR) composites. The study particularly focuses on nanoparticle dispersion behavior within the polymer matrix and assesses the water diffusion coefficient under various environmental conditions, including aqueous, acidic, and alkaline media, through systematic experimental investigations.

2. Literature Review

In the field of composite materials, extensive research has been conducted on the effects of moisture absorption and diffusion in particle-reinforced composites. For example, Siedlaczek et al. [12] investigated the moisture uptake and diffusion behavior in epoxy

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and vinyl ester matrices that contained varying filler levels. Their results showed that the rate of moisture absorption was primarily influenced by relative humidity, while temperature had a lesser effect, particularly in epoxy-based systems. This highlights the importance of environmental factors, especially in high-humidity conditions. They employed a hybrid sorption isotherm model to describe the moisture behavior and confirmed that moisture diffusion followed Fickian diffusion, a behavior commonly observed in polymeric materials. Moreover, the study identified three key moisture transport pathways within the composite material: the polymer matrix, the matrix-filler interface, and the matrix-filler continuum. These insights are essential for understanding how moisture penetrates the composite and affects its long-term performance, as moisture absorption can reduce mechanical strength, which could compromise the material's structural integrity over time.

Similarly, a review by Korkees focused on the moisture and environmental exposure effects on carbon fiber-reinforced epoxy composites. This research highlighted how the interaction between moisture and composite structure, particularly with epoxy resins and carbon fibers, significantly impacts the diffusion behavior and overall material durability. These findings are especially relevant when designing polymer composites for harsh environments where exposure to moisture can accelerate material degradation [13].

In a different study, Mertgenç (2025) examined the degradation effects of seawater exposure on glass fiber-reinforced polymer (GFRP) and carbon fiber-reinforced polymer (CFRP) composites. His research showed a substantial decrease in mechanical performance due to water absorption, with GFRP samples exhibiting a more significant reduction in Young's modulus compared to CFRP samples after prolonged exposure to seawater. These findings underline the importance of selecting suitable fillers and reinforcing agents when designing composites that must endure exposure to challenging environments, such as seawater [14].

3. Materials

Epoxy resin and nitrile butadiene rubber (NBR) were supplied by Saudi Industrial Resins Limited Company (SIR)™. ZnONPs were supplied by Hongwu International Group Ltd-China (purity: 99.8%; melting point: 1975 °C; density at 20 °C: 5.06 g/cm^3).

3.1. Preparation of nanocomposites

The epoxy resin and NBR were mixed in a 70:30 ratio to form a polymeric blend. ZnONPs were added at 1%, 2%, and 3% by weight, and the mixture was stirred using a magnetic stirrer for 30 minutes to ensure uniform dispersion. A 1% hardener was then added, and the mixture was manually stirred for an additional 2 minutes before being poured into an oily glass mold for easy removal of the sample.

The mixture was left to solidify in the mold for two days, allowing the formation of a cross-linked polymer network. This process ensured that the final composite exhibited enhanced mechanical properties, including improved strength, durability, and resistance to moisture and environmental degradation, making it suitable for various industrial applications requiring high performance under harsh conditions.

3.2. Diffusion coefficient

The relation between weight gain% and the root square for the time of immersion in water for all specimens according to Fick's law to evaluate the value of the diffusion coefficient, which is given by Equation [4]:

$$D = \pi\left(\frac{kb}{4M\infty}\right) \tag{1}$$

where D is the diffusion coefficient, k is the slope of weight gain vs. square root of time of immersion diagram, b is the thickness of the sample, and $M\infty$ is the highest value of weight gain% for the specimens. To calculate the percentage of weight gain for the specimens, we use the following equation [15]:

$$Weight\ Gain\% = \frac{M_2 - M_1}{M_2} \tag{2}$$

where M_1 is the weight of the specimen before immersion in liquid (g) and M_2 is the weight of the specimen after immersion in liquid (g) [16].

3.3. Characterization

X-ray diffraction (XRD) patterns of nano-ZnO were obtained using a Miniflex XRD machine (Rigaku, Japan). The crystalline domain dimension (D) was calculated using Scherrer's equation [17]:

$$D = {}^{K\lambda}\!/_{\beta cos\theta} \tag{3}$$

where λ represents the wavelength of the incident X-ray beam, θ is Bragg's diffraction angle, β is the width of the X-ray pattern line at half-peak height in radians, and the dimensionless shape factor (K) has a normal value of 0.89 but varies depending on the actual crystalline structure. Scanning electron microscopy (SEM) (FEI Company, USA) was used to study the morphology of the materials.

4. Results and Discussion

4.1. XRD of ZnONPs

Figure 1 presents the representative XRD pattern of ZnONPs, which was employed to analyze their crystal structure and estimate particle size. The diffraction peaks provide clear evidence of the crystalline nature of the ZnONPs, with pronounced features indicative of high crystallinity. Notably, sharp diffraction peaks appeared at 2θ values of 31.79°, 34.45°, 36.26°, and 47.53°, corresponding to the (100), (002), (101), and (102) crystal planes. These reflections are characteristic of ZnO's hexagonal wurtzite structure and fall within the standard diffraction range of 20°–80°. The pattern aligns closely with the findings reported by Nassif et al. [18], confirming the phase purity of the synthesized nanoparticles. Furthermore, the average crystallite size of the ZnO particles was calculated to be approximately 18.36 nm using the Scherrer equation, based on the broadening of the major diffraction peaks as described in Equation (3).

4.2. The X-ray diffraction (XRD) of nanocomposites

Analysis of the samples is presented in Figure 2. The XRD intensity of the pure blend exhibited the highest peak, while the intensities of the composites were either similar or closely matched. These results contrast with findings reported by Balogun et al. [1]. The observed increase in intensity indicates a higher level of crystallinity. The XRD pattern for the pure blend shows prominent peaks at 2θ values of 18.9°, 22.77°, and 29.32°, which are consistent with prior studies by Muthukumar and Thilagavathi [19]. The impact of the rubber component appears minimal due to its low concentration. The NBR itself showed no distinct peaks,

Figure 1
Typical XRD pattern of ZnONPs

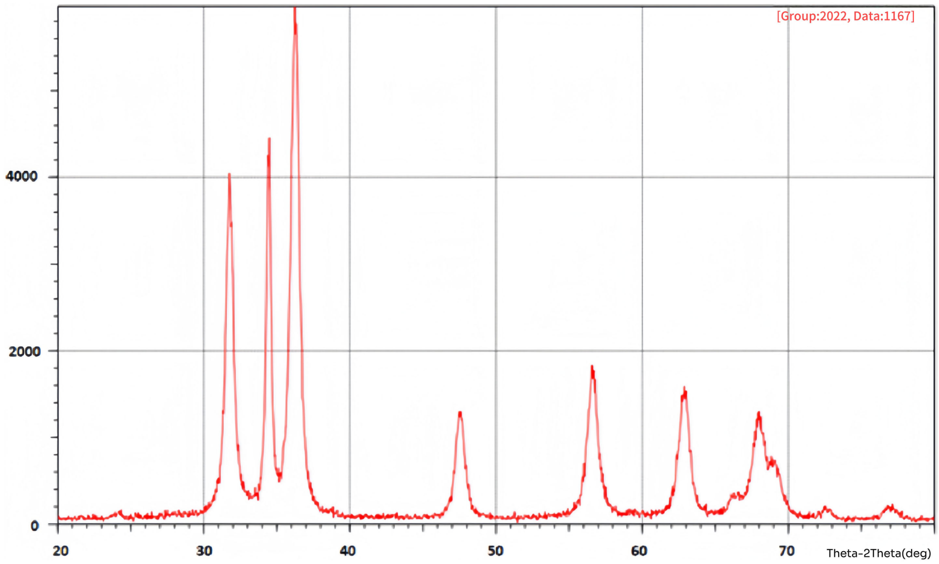
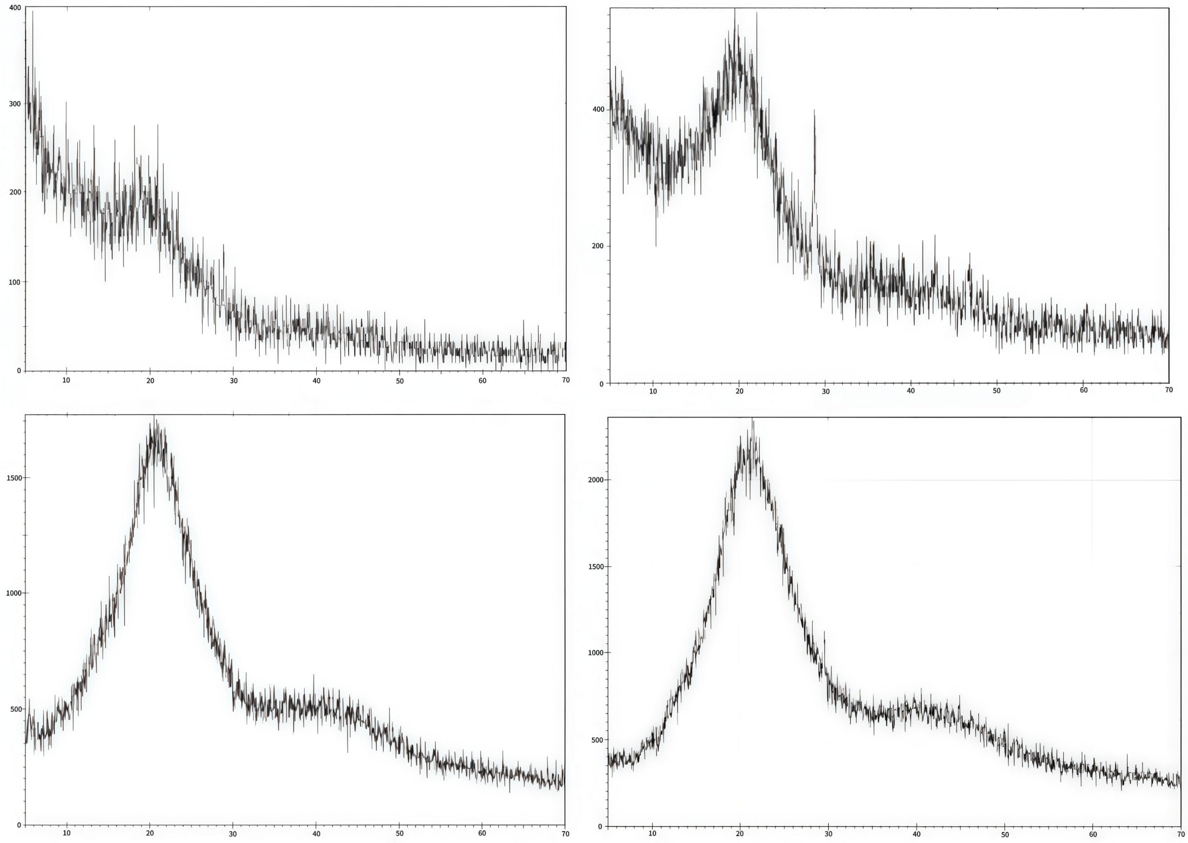
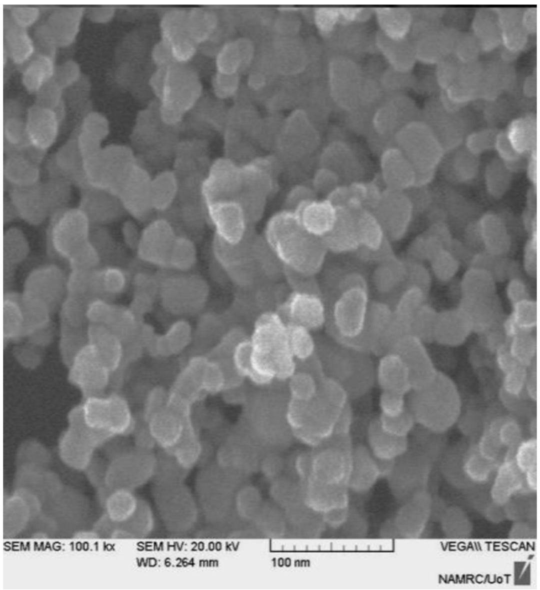


Figure 2
(a) XRD of an unreinforced blend; (b) a blend reinforced by 1% ZnONPs; (c) a blend reinforced by 2% ZnONPs; (d) a blend reinforced by 3% ZnONPs



which aligns with its expected amorphous nature, as noted in earlier studies by Thomas et al. [20]. Increasing the concentration of ZnONPs led to a shift in the 2θ values of the composites. The composite containing 1% ZnONPs exhibited the lowest 2θ values when compared to the other composites, suggesting an interaction between the ZnONPs and the polymer matrix. As the ZnO concentration increased, the 2θ values gradually increased, which can be attributed to particle agglomeration or de-intercalation

Figure 3
SEM of ZnONPs



occurring with the addition of a higher amount of filler. Such trends have been previously documented in the literature by Hwang et al. [21].

4.3. SEM

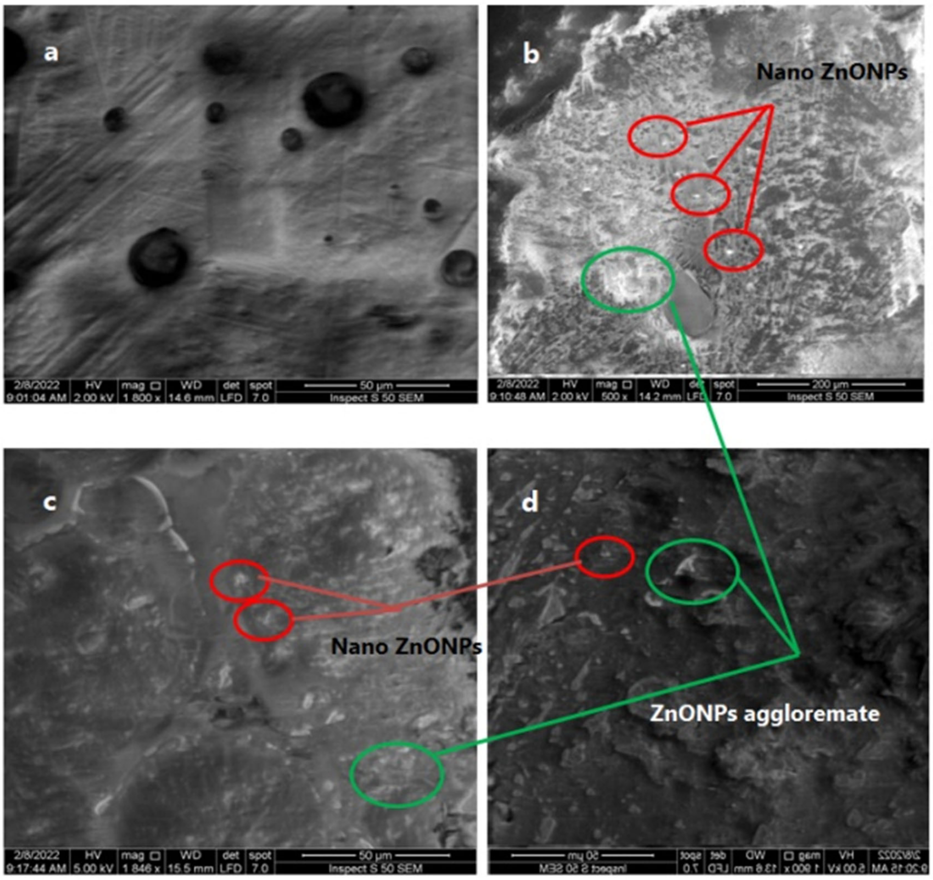
Figure 3 illustrates the morphological characteristics of the ZnONPs. The micrograph clearly shows that the particles possess a smooth, nearly spherical shape, indicating uniform morphology across the entire sample. These observations are consistent with the morphological characteristics reported by Lin et al. [22], further validating the synthesis approach employed in this study. The nearly spherical and smooth shape of the nanoparticles plays a key role in their dispersion within the polymer matrix. Spherical nanoparticles typically disperse more evenly than other shapes, like cylindrical or needlelike particles, due to their lower interaction with the matrix.

This uniform dispersion enhances the physical and mechanical properties of the composite. ZnONPs, when incorporated into the polymer, strengthen the material, improving its durability and flexibility. Thus, these morphological features confirm that the synthesis method effectively produced nanoparticles with a consistent shape, critical for enhancing the composite's performance.

4.4. SEM of the nanocomposites

Figure 4 illustrates the surface morphology of the samples under investigation, as observed using SEM. In Figure 4(a), the SEM image of the pure polymer blend, composed of 70% epoxy resin and 30% NBR, reveals significant surface irregularities, including prominent protrusions and uneven regions. These characteristics suggest poor miscibility between the two polymer components, likely due to

Figure 4
(a) SEM micrographs of an unreinforced blend; (b) a blend reinforced by 1% ZnONPs; (c) a blend reinforced by 2% ZnONPs; and (d) a blend reinforced by 3% ZnONPs at 1900× magnification



incomplete phase integration [23]. At a magnification of 1800×, the microstructure indicates limited molecular interaction between the epoxy resin (EP) and NBR, which could negatively affect the mechanical strength and thermal stability of the material.

Upon adding 1% nano-sized ZnO, as shown in Figure 4(b) (magnified at 4000×), a notable improvement in the dispersion of the nanoparticles throughout the polymer matrix is observed. The ZnONPs are more uniformly distributed in comparison to the unreinforced sample, resulting in a smoother surface with fewer large agglomerates. Although some surface roughness remains, the improved dispersion suggests better phase compatibility and may lead to enhanced mechanical properties.

Further enhancement is evident with the add fraction of 2% ZnONPs, as seen in Figure 4(c) (magnified at 3325×). At this concentration, the nanoparticles appear more uniformly integrated within the polymer matrix, and the surface exhibits reduced heterogeneity. This suggests a greater degree of compatibility between the ZnONPs and the polymer blend, promoting a more cohesive and stable matrix. When compared to the 1% ZnO sample, the 2% composition shows improved structural continuity and potentially superior performance characteristics.

Figure 4(d) shows the SEM image of the blend reinforced with 3% nano-ZnO, captured at a magnification of 3808×. This sample exhibits the most homogeneous dispersion of nanoparticles among all the compositions tested. The surface is smoother, with minimal roughness and negligible aggregation. The even distribution of ZnONPs throughout the polymer network at this concentration suggests that 3% ZnO provides optimal dispersion, which is likely to result in improved mechanical integrity and enhanced thermal resistance [11, 24]. Thus, this concentration appears to offer the most balanced reinforcement effect within the composite system.

4.5. Chemical solutions uptake

Figures 5 and 6 illustrate that the polymeric materials demonstrate nearly identical behavior when submerged in both water and nitric acid over an 8-week period. Figure 4 depicts the relationship between the percentage of weight gain and time ($Time^{0.5}$) for samples containing varying concentrations of ZnONPs (0%, 1%, 2%, and 3%). The data show that the 0% ZnO sample experiences a rapid initial weight gain, followed by stabilization, indicating significant water absorption that eventually reaches equilibrium. This behavior can be attributed to the interaction of water, a low molecular weight liquid, with the resin. The water molecules quickly penetrate the polymer matrix, filling microcavities and gaps between the structural components [25]. This process involves the sequential movement of polymer chains [26]. In contrast, the other composite samples exhibited a slower weight gain. As the ZnO concentration increases, the weight gain progressively decreases, with the 2% ZnO sample exhibiting the smallest weight gain. This trend suggests that ZnONPs enhance the material's resistance to nitric acid (HNO_3) absorption, with higher ZnO concentrations providing improved protection by reinforcing the material's resistance to acidic environments.

Figure 5
Weight gain vs. time square root of all samples immersed in water

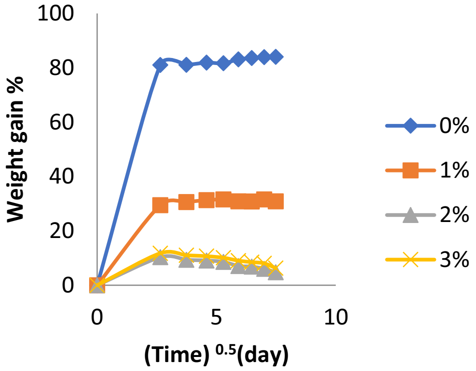


Figure 6
Weight gain vs. time square root of all samples immersed in $HN0_3$

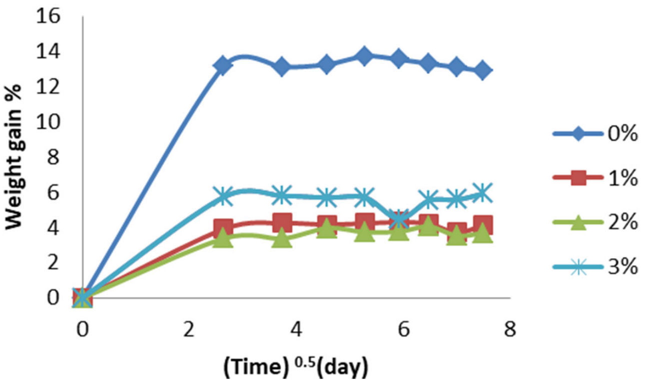


Figure 7 shows the effect of different concentrations of ZnONPs (0%, 1%, 2%, and 3%) on the weight gain percentage (% weight gain) of samples immersed in NaOH solution over 8 weeks. The 0% ZnO sample shows the lowest weight gain, while an increase in the ZnO concentration results in a higher weight gain, with the 3% ZnO sample exhibiting the highest weight gain, indicating that higher ZnO concentrations provide the best protection against absorption in alkaline environments.

Figure 8 illustrates the diffusion coefficients of water, HNO_3, and NaOH solutions as a function of the ZnONP filler concentration, ranging from 0% to 3%. For water, the diffusion coefficient decreases as the concentration of ZnO increases, suggesting that higher ZnO concentrations decrease the material's permeability to water. In contrast, for NaOH, an increase in ZnO concentration leads to a rise in the diffusion coefficient, implying that ZnO may enhance the material's permeability to alkaline solutions. Similarly, for HNO_3, the diffusion coefficient increases with ZnO content, reaching its peak at 3%, indicating that the material becomes more permeable to acidic solutions with higher ZnO concentrations. These observations emphasize that ZnONPs

Figure 7
Weight gain vs. time square root of all samples immersed in NaOH

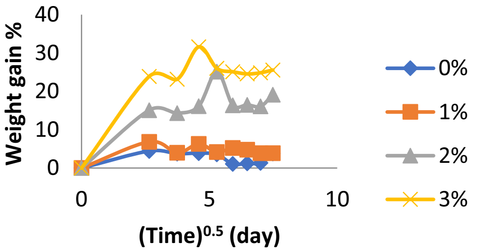
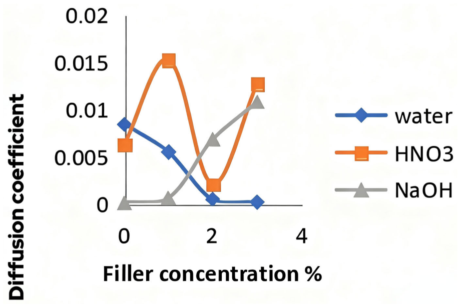


Figure 8
Diffusion coefficients as a function of filler content of all samples



influence the material's interaction with different solutions, such as water, HNO_3, and NaOH, implying that the ZnO concentration can be adjusted to optimize the material's chemical resistance and permeability [15]. Generally, the inclusion of nanofillers in epoxy tends to reduce the absorption of water and other liquids, resulting in a significantly smaller weight change upon immersion, as these nanoparticles hinder the movement of liquid molecules into the inner structure of the material [27].

5. Conclusion

The research demonstrated that integrating ZnONPs into EP/NBR composites significantly enhanced the dispersion of nanoparticles within the polymer matrix. As the concentration of ZnO increased, the nanoparticle dispersion improved, leading to better composite properties overall. Diffusion tests revealed a reduction in the water diffusion coefficient with higher ZnO concentrations, suggesting that ZnO enhances the composite's resistance to moisture absorption. However, fluctuations were noted in the diffusion coefficients when the composites were exposed to acidic and alkaline solutions. The sample with 1% ZnO showed the best performance in acidic conditions, indicating its superior behavior under such environments. The results highlight the dual nature of ZnO's impact on the composite's performance in various environments. These findings underscore the potential of ZnO-enhanced EP/NBR composites for applications that require superior moisture and chemical resistance. With improved mechanical and physical properties, these composites are well-suited for demanding industrial applications where durability and long-term performance under challenging conditions are essential.

Acknowledgment

The authors are grateful to the College of Applied Science – University of Technology – Iraq for its contribution to this work.

Ethical Statement

This study does not contain any studies with human or animal subjects performed by any of the authors.

Conflicts of Interest

The authors declare that they have no conflicts of interest to this work.

Data Availability Statement

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

Author Contribution Statement

Rafah Alwan Nassif: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Resources, Data curation, Writing – original draft, Writing – review & editing, Visualization, Supervision, Project administration. **Raghad Hamid Hilal:** Software, Validation, Formal analysis, Investigation, Resources, Data curation, Writing – original draft, Visualization, Project administration.

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How to Cite: Nassif, R. A., & Hilal, R. H. (2025). Influence of Nano Zinc Oxide Reinforcement on the Diffusion Coefficient of Unsaturated Polyester and Nitrile Butadiene Rubber Composites. *Smart Wearable Technology*. https://doi.org/10.47852/bonviewSWT52027158