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Study of the Structural Properties of Recycling Polyethylene Terephthalate as a Matrix to Prepare Polymer Nano Composites with Nano Nickel Oxide Synthesized via Green Method

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Abstract

This work employs the green manufacturing of nickel oxide (NiO) nanoparticles using rosemary (Rosmarinus officinalis) extract as a bioreductant. The NiO nanoparticles were combined with recycled polyethylene terephthalate (rPET) to generate a composite material. Atomic Force Microscopy (AFM), scanning electron microscopy (SEM), Energy Dispersive X-ray (EDS), X-ray diffraction (XRD), Thermogravimetric analysis (TGA), and differential scanning calorimetry (DSC) were used to thoroughly examine the NiO/PET nanocomposite. AFM and SEM investigations confirmed NiO nanoparticles' homogeneous distribution and surface shape inside the rPET matrix. EDX validated the elemental composition, whereas XRD revealed information about the crystalline structure of the produced nanoparticles and the nanocomposite. TGA and DSC were used to examine thermal stability and breakdown behavior, demonstrating the NiO/rPET composite's improved thermal characteristics. The composite's anti-corrosion ability was also investigated, and it showed a considerable increase in corrosive resistance when compared to pure rPET. This work emphasizes the feasibility of employing green synthesis methods to produce metal oxide nanoparticles and their application in improving the characteristics of polymer composites, notably their anti-corrosion capabilities.

Article Info.

Keywords:
Polyethylene Terephthalate, NPs, Green Method, Nanocomposites, Anti-Corrosion.

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1. Introduction

In recent years, extensive research has been conducted worldwide on nanotechnology due to its unique properties, such as its ability to manipulate matter at the atomic and molecular scale with enhanced surface area-to-volume ratio and its potential for creating materials with novel mechanical, electrical, and optical properties. [1, 2]. Nanomaterials are advantageous compared to bulk materials when considering the properties they possess. These properties enable the creation of advanced materials with superior strength, lightness, and conductivity, as well as enhanced chemical reactivity. The benefits of these nano-properties include more efficient energy storage and conversion, targeted drug delivery systems in medicine, improved sensors, and the development of stronger yet lighter materials for aerospace and automotive industries [3, 4]. In the past decade, metal oxides, especially transition metal nanostructures, have played a major role due to their unique and distinctive properties, such as electronic, optical, sensing, chemical, magnetic, and mechanical properties, and their application in various fields in medicine, engineering, industries, and anti-corrosion coatings due to their size, surface effect, and electrical properties [5]. Nickel oxide (NiO) is an appealing semiconductor as its properties are beneficial for various applications in optical and chemical sensing, batteries, nanowires [6], fibers, coatings, alloys, and constrained catalysts [7-11]. The environmentally friendly green synthesis method that adheres to the green chemistry approach involving the use of plant exudates and extracts is a popular

approach for the synthesis of nanoparticles due to its speed, simplicity, and low synthesis costs. Nano oxides have been combined with synthetic polymer waste to generate new materials with new properties to produce nanoparticles of desired metal oxides [12-14]. Plant extracts such as rosemary have been used as reducing and stabilizing agents for the synthesis of nickel oxide nanoparticles. Synthetic polymers are designed and developed to be durable, strong [15-17], and long-lasting, but failure to dispose of them leads to their continued presence in the environment.

The persistence of this artificial material in the environment has become a known environmental pollution problem, causing known concerns and challenges for waste management [18, 19]. Recycled plastics modified with other materials have been improved and converted to obtain recycled products with desirable properties, such as recycled polyethylene terephthalate (PET) polymer reinforced with nano-nickel oxide, which has been recycled to enhance mechanical strength and thermal stability, which meets wide industrial needs and is also used as anti-corrosion coatings [20, 21].

This work examines the green production of nickel oxide nanoparticles utilizing rosemary extract as a reducing and stabilizing agent. Additionally, the aim of the research is to evaluate the possible improvement in mechanical strength and thermal stability of recycled PET polymers when reinforced with NiO nanoparticles, with an emphasis on generating materials suited for industrial applications, such as anti-corrosion coatings. This research follows green chemistry principles, aiming to reduce environmental waste and develop long-lasting nanocomposite materials.

2. Materials and Method

The materials used in this work are: Nickel nitrate ($Ni(NO_3)_2 \cdot 6H_2O$), deionized water, rosemary leaves, and polyethylene terephthalate (PET) plastic bottles collected from waste. Dimethyl sulfoxide (DMSO) was used as a solvent.

2. 1. Extraction of Rosemary

Rosemary leaves were washed, dried in the shade, and ground. A quantity of the powder was mixed with 50 mL of distilled water and heated to boiling point for 20 minutes. It was left to cool at room temperature to obtain a concentrated extract. After that, it was filtered with filter paper. The extract was later used to prepare the nano oxide.

2. 2. Green Synthesis of the Metal Oxide NPs

Nickel nitrate salt was dissolved in distilled water and heated for a few minutes; then, rosemary extract was added to it gradually using a dropper while heating at a temperature of 80 °C with stirring until the biosynthesis was completed. The precipitate was then collected by filtration or centrifugation, washed several times with distilled water, dried at a temperature of 100 °C, and then calcined at 500 °C to improve the crystallinity of the nanoparticles.

2. 3. Prepared Nanocomposites

Plastic bottles were collected from waste, washed with distilled water, cut into small pieces, and dissolved in 20 ml of dimethyl sulfoxide solvent in a round flask in a sublimation device with stirring and heating to 80°C for 20 minutes until the polymer dissolved. Then, nano nickel oxide was added to the mixture with stirring and heating for 3-4 hours. Then, it was placed in plates and dried at 100°C to remove the solvent and obtain the PET/NiO nanocomposite.

3. Results and Discussion

3. 1. Atomic Force Microscopy (AFM)

AFM was used to investigate the synthesized NiO nanoparticles' surface morphology, particle size, and roughness. AFM gave a 3D surface profile of NiO nanoparticles, as shown in Fig. 1(a), providing information about their shape and distribution. Generally, green-manufactured NiO nanoparticles are spherical or quasi-spherical. The average particle size was 30.5 nm, as shown in Table 1.

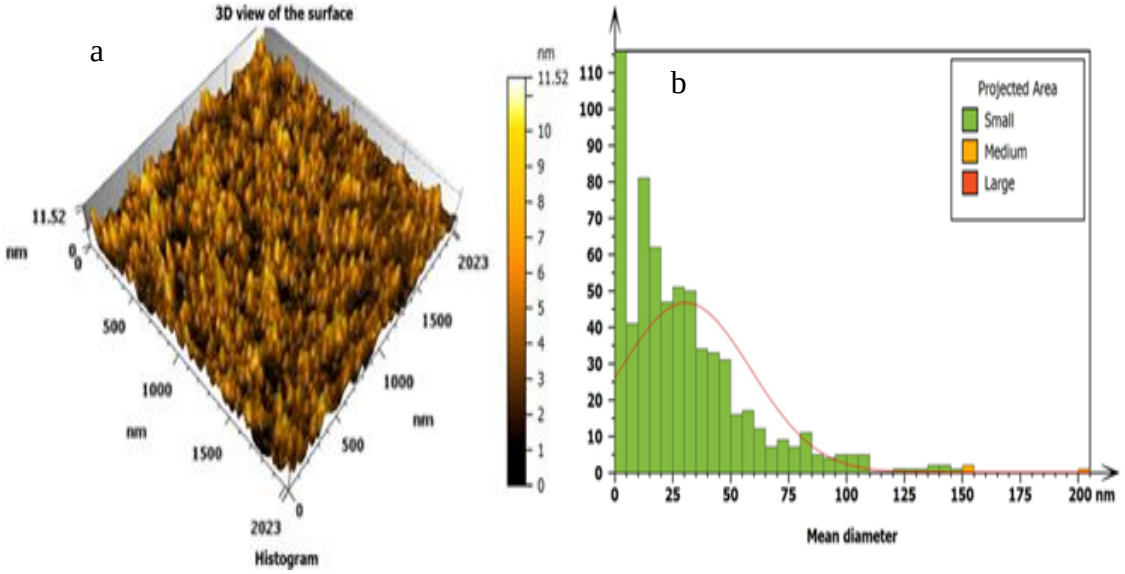

Figure 1: AFM (a)3D image of NiO nanoparticles, (b) histogram of distribution.

Table 1: Dimensions of the synthesized NiO nanoparticle.

Average Value (nm)	Minimum (nm)	Maximum (nm)
30.52	2.9	155

3. 2. Scanning Electron Microscopy and Energy Dispersive X-ray (SEM/EDS)

SEM images revealed details regarding the shape and size of the PET/NiO composite nanostructures. Green synthesis with rosemary extracts frequently produces spherical, flower-like nanostructures or have various complicated morphologies, depending on the synthesis conditions. Fig. 2 shows the spherical shapes nanostructures SEM images of PET/NiO Nanocomposite (a) 500µm and (b) 100µm.

EDX performs a qualitative and quantitative examination of the constituents in the sample. The primary elements detected in NiO nanostructures synthesized with rosemary extract were nickel (Ni) and oxygen (O), confirming the formation of NiO. Peaks corresponding to carbon (C) and possibly other elements, such as nitrogen (N) or sulfur (S) could be attributed to the organic residues from the rosemary extract. These residues might be present as a capping layer on the nanoparticles or as part of the overall structure. Different percentages of other elements may be attributed to impurities in the recycled polymer, as shown in Fig. 3.

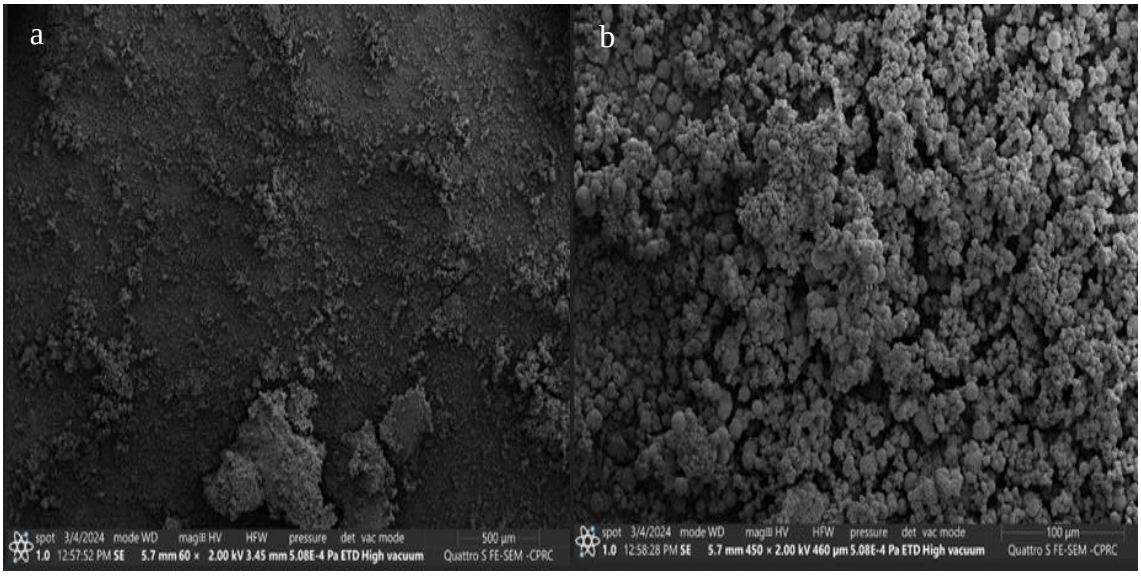

Figure 2: (a) SEM image of PET/NiO Nanocomposite 500µm, (b) SEM image of PET/NiO Nanocomposite 100µm.

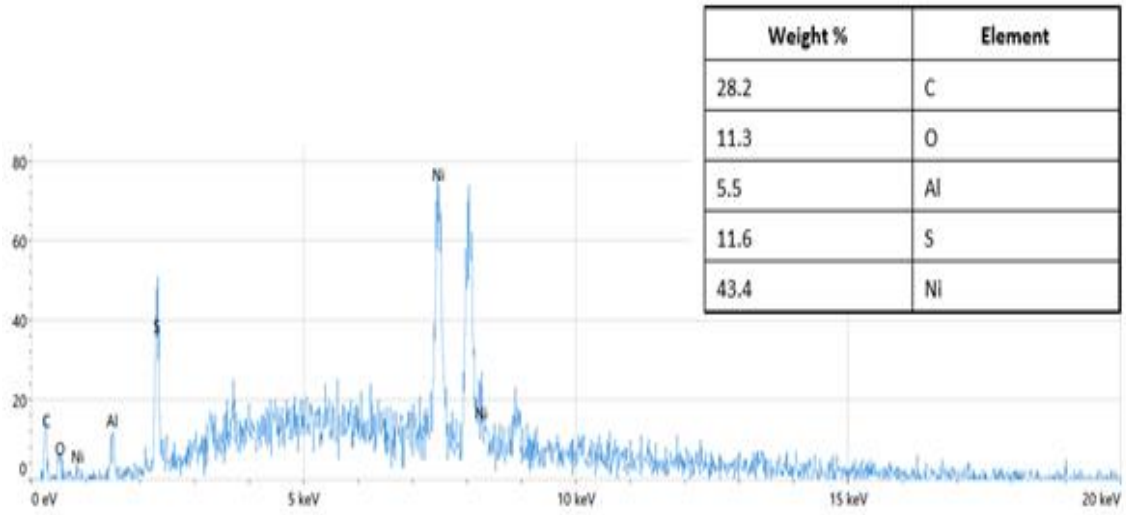

Figure 3: EDS of PET/NiO nanocomposite.

3. 3. X-Ray Diffraction of PET /NiO Nanocomposite

XRD patterns are used to detect the crystalline phases that exist in the produced material. XRD pattern of NiO exhibited diffraction peaks at certain angles (2θ) that correspond to (111), (200), and (220) planes of its cubic crystal structure.

The sharp, well-defined peaks at these angles indicate the production of crystalline NiO. The strength and sharpness of the XRD peaks reveal the crystallinity of the composite. Sharp peaks suggest high crystallinity, as shown in Fig. 4, but broader peaks might represent amorphous phases or small crystallites. Other phases in the composite (such as nickel hydroxide or rosemary organic remnants) will yield distinctive peaks that may be recognized and examined. Peaks representing additional compounds, such as unreacted nickel precursors or organic components from the rosemary extract, may appear in the XRD pattern, indicating the presence of impurities or partial reactions.

Scherrer equation was applied to the XRD peak broadening to estimate the average crystallite size (D) of the NiO nanoparticles, which was 15.38 nm.

Green synthesis methods often produce smaller crystallites due to the mild reaction conditions. This is typical in green synthesis due to the presence of organic molecules.

Rosemary extract can help reduce crystal formation. Peaks in the PET/NiO nanocomposite were seen at 21.3°, 31.8°, 39.1°, 51.5°, and 53.9°, corresponding to the same Miller indices 200, 002, 102, 112, and 422 as the reference card (JCPDS card No. 65-6920), as shown in Table 2 [22, 23].

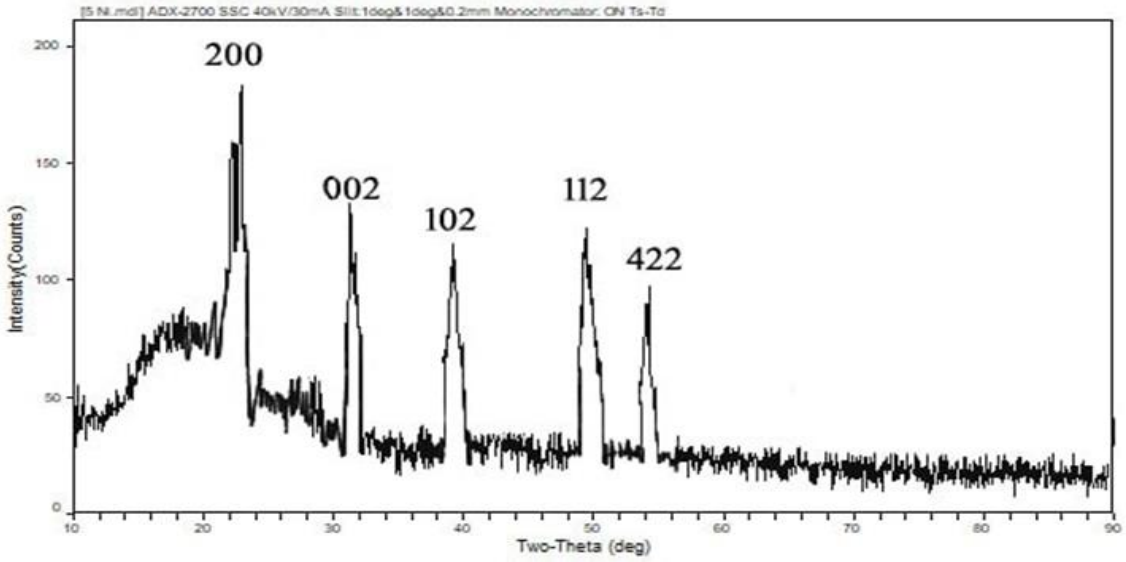

Figure 4: X-Ray Diffraction Pattern of PET/NiO nanocomposite.

Table 2: The structure parameter of PET/NiO nanocomposite obtained from XRD pattern.

2-Theta	d(nm)	(hkl)	FWHM	Phase	D(nm)
21.3	0.41681	(200)	0.35	PET/NiO	23.6
31.804	0.28114	(002)	0.488	PET/NiO	17.2
39.105	0.23016	(102)	0.31	PET/NiO	27.9
51.585	0.17703	(112)	0.546	PET/NiO	16.4
53.943	0.16983	(422)	0.257	PET/NiO	35.8

3. 4. Thermogravimetric Analysis (TGA) and Differential Scanning Calorimetry (DSC) of PET/NiO Nanocomposite

Thermogravimetric analysis (TG) is a technique that can provide information about the amount of organic material present in composite interlayers. TGA can also show how too much surface modification results in a lower thermal degradation temperature and an assortment of negative effects on the composite properties. The sample starts at full weight, indicating its pristine condition before heating. The nanocomposite PET/NiO synthesized by the green method was decomposed in three stages as follows:

1. First Decomposition Stage (~100°C–200°C):

Weight loss is likely due to the evaporation of moisture or volatile organic compounds from the PET/NiO composite. Cause: Adsorbed water or solvents from the synthesis process.

2. Second Decomposition Stage (~300°C–500°C):

Significant weight of (~73.62%) is lost in this stage, representing the thermal degradation of the polyethylene terephthalate (PET) matrix. The breakdown of polymer chains in PET releases volatiles, contributing to weight reduction. Cause: Thermal degradation of the organic polymer matrix.

3. Final Stabilization (~600°C–1000°C):

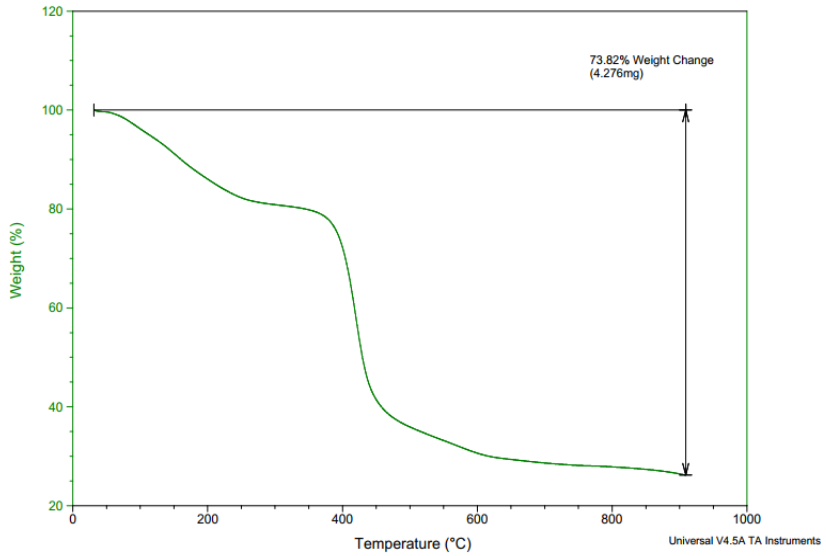

Figure 5: TGA curve of PET/NiO.

The residual weight corresponds to the inorganic component (NiO nanoparticles) that remain thermally stable up to 1000°C. The presence of NiO stabilizes the composite and resists further degradation. Weight Change Annotation (73.62%), which indicates the percentage of mass lost during heating, attributed to the decomposition of the PET component.

Incorporating NiO nanoparticles can affect the glass transition temperature (T_g) of the rPET matrix, leading to an increase in T_g due to restricted polymer chain mobility. Additionally, DSC analysis of the heat flow during decomposition indicates the composite's thermal stability, with a shift in the thermal degradation peak to higher temperatures indicating enhanced stability due to NiO. It considers the glass transition compared with standard reference at 101.7 °C, as shown in Fig. 6. The glass transition temperature of PET is reported in the literature to be between 69°C and 85°C [24] .

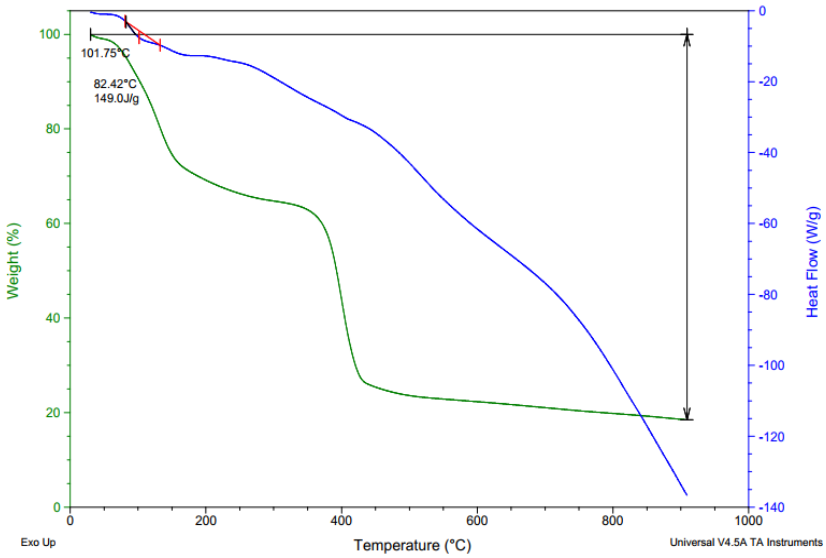

Figure 6: DSC curve of PET/NiO.

4. Corrosion Protectiveness Studies

The sample under test was placed inside a corrosion cell with a diameter of 16.55 cm^2, with the surface area exposed to the solution (1M HCl).

The corrosion parameters were calculated using the data given in Table3 and Fig. 7. The corrosion current density (i_{corr}) and corrosion potential (E_{corr}) were calculated by extrapolating the cathodic and anodic Tafel's in the absence and presence of inhibitor molecules in 1M HCl solution. The anodic (ba) and cathodic (bc) Tafel slopes were also calculated using Fig. 7. Table 3 shows the values for the corrosion potential E_{corr} (mV), corrosion current density i_{corr} (A/cm^2), cathodic and anodic Tafel slopes (mV/Dec), and protection efficiency PE% [25].

$$\%PE = \frac{(icorr)o - (icorr)}{(icorr)o} \quad (1)$$

where %PE represents the protection efficiency, $(i_{corr})_O$ represents the corrosion current density in the absence of inhibitors and (i_{corr}) represents the corrosion current density without the inhibitors [26]. Table 3 shows the corrosion parameters or various substances and blanks in HCl solutions.

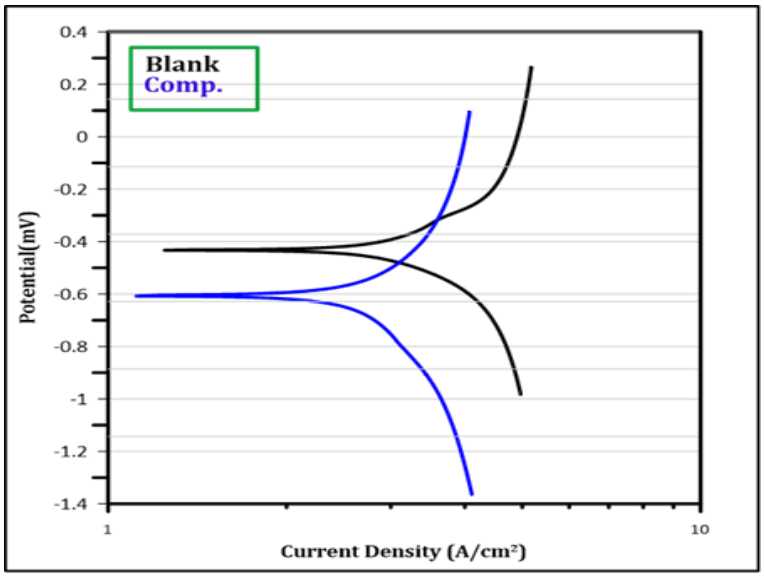

Figure 7: Polarization curves for corrosion of PET/NiO.

Table 3: Corrosion parameters for blank and PET /NiO in HCl solutions and different.

Comp.	Blank	PET/Ni
-Ecorr (mV)	-0.427	-0.723
Icorr (µA/cm^2)	493.9	39.37
I corr./ r (A/cm^2)	9.87E-4	7.873E-5
Resis. (Ω)	70.31	1519
-Bc (mV/Dec)	0.156	0.230
Ba (mV/Dec)	0.164	0.343
Corr. rate, (mm/y)	4.848	0.386
IE%	-	92

5. Conclusions

Nanocomposites with nickel oxide synthesized via green technologies offer a sustainable, eco-friendly approach to materials science. This method combines recycled PET with NiO nanostructures, produced using natural extracts like rosemary, enhancing

the environmental benefits by addressing plastic waste and eliminating toxic chemicals during synthesis. The nanocomposites were characterized using AFM, which revealed NiO particle sizes ranging from 2.9 nm to 155 nm. SEM) and Energy Dispersive X-ray (EDX confirmed the uniform distribution of NiO within the PET matrix. XRD patterns showed distinct crystalline peaks, verifying the crystalline structure of NiO. Thermal properties were assessed using TGA and DSC, indicating enhanced thermal stability, with a degradation temperature of 101.7°C. This technique not only solves environmental concerns about PET disposal but also improves the creation of high-performance materials for a wide range of applications, including electronics, catalysis, and anticorrosion coatings.

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Conflicts of Interest

The authors declare they have no competing interests.

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إعادة تدوير متعدد إيثيلين تيرفثالات واستخدامه كمصفوفة لإعداد متراكبات نانوية بوليمرية مع أكسيد النيكل المحضر بطريقة الخضراء

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الخلاصة

يتناول هذا العمل التصنيع الأخضر لجسيمات أكسيد النيكل النانوية (NiO) باستخدام مستخلص إكليل الجبل (Rosmarinus officinalis) كمختزل حيوي. ثم تم دمج جسيمات أكسيد النيكل النانوية مع بولي إيثيلين تيريفثالات المعاد تدويره (rPET) لتوليد مادة مركبة. تم استخدام AFM و SEM و EDX و XRD و TGA و DSC لفحص مركب NiO / rPET بدقة. أكدت تحقيقات AFM و SEM التوزيع المتجانس لجسيمات أكسيد النيكل النانوية وشكل السطح داخل مصفوفة rPET. أثبت EDX تركيب العناصر، بينما كشف XRD عن معلومات حول البنية البلورية للجسيمات النانوية المنتجة والمواد المركبة. تم استخدام TGA و DSC لفحص الاستقرار الحراري وسلوك الانهيار، مما يدل على الخصائص الحرارية المحسنة لمركب NiO / rPET. كما تم التحقيق في قدرة المركب على مقاومة التآكل، وأظهر زيادة كبيرة في مقاومة التآكل عند مقارنته بـ rPET النقي. يؤكد هذا العمل على جدوى استخدام طرق التركيب الأخضر لإنتاج جسيمات نانوية من أكسيد المعادن وتطبيقها في تحسين خصائص المركبات البوليمرية، ولا سيما قدراتها المضادة للتآكل.

الكلمات المفتاحية: بولي اثلين تير فثالات، NPs، طريقة خضراء، المركبات النانوية، مضاد التآكل.