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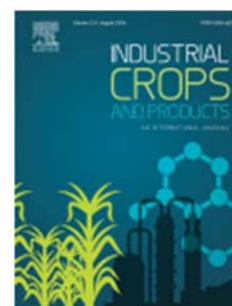
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Development of functional graphene oxide-urethane coating systems from *Ricinus communis* seed oil

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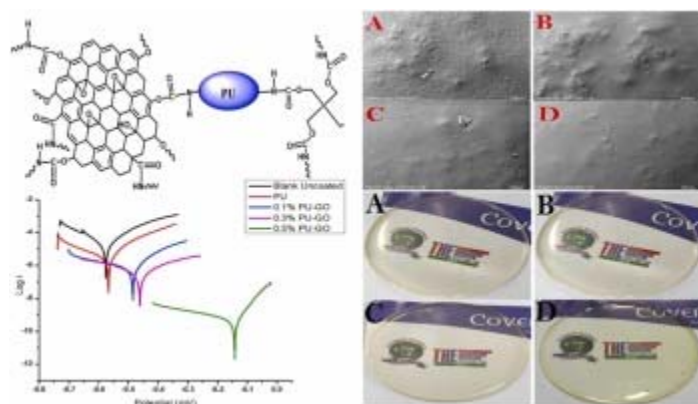
Highlights

- • Graphene nanoparticles were successfully modified and characterized.
- • Modified materials were dispersed within the polymer matrix uniformly.
- • Hydrophobicity of the synthesized urethane films is confirmed.
- • Composite films inhibit microbes.
- • Composite-coated mild steels show improved salt spray and Tafel test evaluations.

Abstract

The surface-modified graphene oxide (GO) nanoparticles and their blending with a fixed percentage of trimethylpropane (TMP) in *Ricinus communis* seed oil were successfully prepared in a one-pot urethane reaction using 4,4'-diisocyanato dicyclohexylmethane (H_{12} MDI) and methyl isobutyl ketone (MIBK) as the reaction solvent. The structural elucidation and surface morphology of pristine and hybrid composites of the polyurethane coating films were investigated with the aid of Fourier transform infrared spectroscopy (FT-IR), Energy-dispersive X-ray spectroscopy (EDX), Proton nuclear magnetic resonance (1H NMR), X-ray diffraction (XRD), and Scanning electron microscope (SEM). The presence of FT-IR absorption peaks at 790 cm^{-1} to 870 cm^{-1} , 990 cm^{-1} , and 1017 cm^{-1} confirms the following functional groups phenyl -CH bend, stretching phenolic -CO, and epoxy -C-O-C, respectively in modified graphene oxide. An evaluation of the thermal stability of the coating films that were synthesised was carried out with the use of a thermogravimetric analyzer (TGA). It was seen that as the amount of modified graphene oxide in the urethane films increased, so did the water contact angle from 0% to 0.5% . Antimicrobial and anticorrosive properties of the materials were also evaluated.

Graphical Abstract



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Introduction

The usefulness of polyurethane (PU) in various fields, especially in corrosion prevention, which is a global phenomenon, cannot be over-emphasised. However, the synthesis of PU from fossil fuels, which is nonrenewable, has been of great concern to environmentalists. Polyurethanes, a very versatile and extensively utilised industrial material, have been successfully employed in various applications. The utilisation of renewable resources for the synthesis of polyurethane polymeric materials holds substantial economic and ecological importance. Acceptability of these polyurethanes derived from renewable resources is increasing due to the appealing qualities associated with the unique structures of the triglycerides that are found in vegetable oils, along with the environmental and industrial sustainability benefits they offer (Adeboye et al., 2023, Paraskar et al., 2021, Pradhan and Mohanty, 2015). Corrosion is

commonly defined as the process by which a material deforms or deteriorates due to its contact with the surrounding environment, such as gas, moisture, and ultraviolet (UV) radiation (Ibrahimi et al., 2021). Mitigation or prevention of corrosion has been a multimillion-dollar global industry (Chauhan et al., 2020). Corrosion is a prominent factor contributing to the failure of process equipment. This can lead to an untimely deterioration of metallic elements, which can cause adverse financial consequences, environmental pollution, and potential harm or fatality (Fu et al., 2020, Ibrahimi et al., 2021). The corrosion phenomenon has also been responsible for a significant loss of human life within the engineering community, leading to substantial financial burdens of billions of dollars (Sunday and Abimbola, 2019). There are many types of corrosion that stakeholders combat daily (Chauhan et al., 2020). Thus, developing durable, affordable, and eco-friendly corrosion inhibition technologies to safeguard infrastructure is an urgent and strategic priority for global economies (Ibrahimi et al., 2021, Kawsihan et al., 2023, Sunday and Abimbola, 2019). Among the most prevalent approaches employed for corrosion prevention are anti-corrosion coatings and corrosion inhibitors. Corrosion inhibitors (CI) are widely employed in mitigating general and pitting corrosion (Chauhan et al., 2020). Hydrophobic coatings are a surface treatment that repels water extraordinarily, thus inhibiting corrosion (Ahmad et al., 2018). These coatings are designed to create a highly water-repellent surface by reducing the contact area between water droplets and the coated surface, resulting in the droplets rolling off or bouncing off the surface (Bai et al., 2021). There are numerous natural superhydrophobic surfaces, including plant and insect surfaces, like the lotus plant and the chitinous cuticle of crustaceans. Paint is inherently hydrophilic and susceptible to contamination by waterborne pollutants, and due to this hydrophilic nature, paint attracts water and dirt. Consequently, the paint's visible appearance deteriorates, giving it an aged and dirty look (Ahmad et al., 2018). However, applying polyurethane coatings in virtually every industry has generated research interest in introducing nanoparticles (especially their modified forms) into polymeric matrices, thereby improving the hydrophobicity of the composites (Das and Mahanwar, 2020). Modified nanoparticles that have shown such hydrophobic properties in urethane systems are graphene oxide (GO), Zinc oxide (ZnO) (Siyabola et al., 2013), Titanium (iv) oxide (TiO_2) (Siyabola et al., 2021), etc. Graphene is a flat, single-layer arrangement of carbon atoms connected by covalent bonds in an SP^2 hybridization pattern. These carbon atoms are arranged in a hexagonal lattice, forming a honeycomb-like network (Sun, 2019, Yu et al., 2020). As a monolayer allotrope of carbon, graphene functions as the fundamental building block for several other carbon-based allotropes (Adetayo and Runsewe, 2019); thus, graphite flake, which is an intrinsic mineral that occurs naturally, undergoes a purification process for the removal of heteroatomic impurities. Graphite is an inorganic element commonly regarded as a mineral that can be used to synthesise graphene oxide through the oxidation process. This method facilitates the addition of functional groups that contain oxygen, which has the beneficial characteristic of being easily dispersed in water (Han et al., 2020). These functional groups boost the hydrophilicity of GO and facilitate its dispersion in solvents, hence facilitating its integration into composite materials (Jena and Philip, 2022). In addition, the inherent hydrophilic nature of graphene oxide enables its homogeneous deposition onto many surfaces, forming thin films (Marcano et al., 2010). Graphene oxide has properties that are different from

graphene, such as reduced electrical conductivity and increased solubility (Adetayo and Runsewe, 2019). Other distinctive characteristics of GO are good mechanical strength, a high aspect ratio, and a large surface area (Duong et al., 2020). These inherent features of GO render it a promising material for application in coatings to enhance its hydrophobicity (Crawford and Escarsega, 2000).

Furthermore, one crucial consideration is that hydrophobic surfaces must possess a textured structure to maintain their hydrophobic properties for an extended period, enabling them to exhibit longevity and durability over time (Adetayo and Runsewe, 2019). Previous works showed that incorporating modified graphene oxide (GO) into the polyurethane (PU) coating can greatly improve the mechanical properties and the thermal stability of the polyurethane/graphene oxide polymer (Alrashed et al., 2019), but no work has been done with the unique combination of castor seed oil (CSO), trimethylolpropane (TMP) and GO. In this study, the GO is incorporated into functional polymer nanocomposite coatings as a reinforcing nanofiller to enhance mechanical, thermal, barrier, and electrical properties at low dosages of 0.1 wt%, 0.3 wt%, and 0.5 wt%. For the first time, this research details the successful development of a graphene oxide-reinforced polyurethane hybrid coating with TMP and castor seed oil for superior anticorrosive performance combined with enhanced mechanical durability and thermal stability. The synergistic integration of the passive barrier protection afforded by the graphene oxide nanoplatelets, along with the hard segments and crosslinked network structure of the polyurethane polymer matrix, resulted in a structurally robust yet flexible surface treatment.

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Section snippets

Material and methods

Castor seed oil (CSO) obtained from Ado-Ekiti, Southwestern Nigeria, was extracted as previously reported (Siyanbola et al., 2023). 4,4'-Diisocyanato dicyclohexylmethane (H_{12} MDI) was obtained from Sigma-Aldrich, USA. Trimethylolpropane (TMP), sulphuric acid (H_2SO_4), and phosphoric acid (H_3PO_4) were also products of Sigma-Aldrich, USA. Graphite powder was acquired from Swift Services, India. Potassium permanganate ($KMnO_4$), hydrogen peroxide (H_2O_2), and 4-methyl pentan-2-one (MIBK) were purchased

Physico-chemical characteristics

Table 1 shows the comparison of the physicochemical analyses of CSO, *Thevetia peruviana* seed oil (TPSO), and *Jatropha curcas* oil (JCO) [28], CSO and TPSO are both non-drying seed oils (owing to their iodine values less than 115 g/100 g (Siyanbola

et al., 2023), the contrary is that of JCO having the highest of iodine value of the three oils under investigation. However, the lower acid value of CSO (3.8831 mg KOH/g) indicates that the oil is less prone to rancidity when exposed to air,

Conclusion

The inorganic primary status of graphene was successfully modified into graphene oxide (hybrid) with carboxylic groups at the surface of the material, which was conveniently incorporated into the polymer matrix of the prepared composites. The drying time, chemical resistance, thermal stability, water contact angle, XRD, corrosion studies, and antimicrobial evaluations of synthesised composites were evaluated. It is important to note that the hybrid nanoparticles (GO) enhanced the coating

CRedit authorship contribution statement

Adesola Ajayi: Writing – review & editing, Resources, Project administration, Methodology, Investigation, Formal analysis. **Hesdh Irevebo:** Resources, Methodology, Formal analysis. **Olayemi Arigbede:** Resources, Methodology, Data curation. **Oluwafayokunmi Adebamiro:** Methodology, Investigation, Data curation. **Samuel Adeboye:** Writing – original draft, Resources, Methodology, Investigation, Formal analysis, Data curation. **Tolutope Siyanbola:** Writing – review & editing, Writing – original draft,

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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