

DEGRADATION STUDY OF POLYLACTIC  
ACID/GRAPHENE NANOCOMPOSITES BY  
USING ACCELERATED WEATHERING

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We hereby declare that we have checked this thesis, and in my opinion, this thesis is adequate in terms of scope and quality for the award of the degree of Master of Science.



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I hereby declare that the work in this thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institutions.

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## ABSTRAK

Salah satu polimer terbiodegradasi dan berasaskan bio yang banyak ialah polilaktik asid (PLA), yang boleh didapati secara komersil pada harga yang berpatutan dan mempunyai sifat bahan yang unik. Poliester alifatik, PLA mempunyai pelbagai kegunaan penting dalam industri pembungkusan, automobil, bioperubatan dan pertanian. Walau bagaimanapun, ia mempunyai beberapa kelemahan, seperti rapuh dalam kekuatan tegangan dan sifat terma yang lemah. Pengenalan pengisi nano meningkatkan sifat mekanikal dan terma matriks polimer. Menyiasat tingkah laku penguraian komposit PLA/graphene adalah tujuan utama kajian ini. Peringkat pertama kajian mengkaji kandungan nanoplatelet graphene (GNP) optimum bagi nanokomposit PLA/GNP berdasarkan sifat mekanikal, haba dan morfologi. Peringkat kedua ialah kajian degradasi PLA dan nanokomposit (iaitu, GNP dan nanoplatelet carboxyl graphene (GNP-COOH)) dengan teknik luluhawa dipercepat, termasuk mengkaji sifat terma, mekanikal dan morfologi nanokomposit PLA. Nanokomposit PLA/GNP telah disediakan dengan pelbagai kandungan GNP (0, 0.1, 0.3, 0.5, dan 1.0 wt%) dengan kaedah pengadunan cair menggunakan penyemperit skru berkembar. Luluhawa dipercepat telah dijalankan dengan lima kitaran berbeza (8, 16, 32, 48, dan 64 kitaran). Ciri-ciri PLA dan nanokomposit sebelum dan selepas ujian luluhawa dipercepat dianalisis dengan menggunakan Mikroskopi Elektron Pengimbasan Pancaran Lapangan (FESEM), pembelauan sinar-X (XRD), ujian tegangan, kalorimetri pengimbasan pembezaan (DSC), analisis termogravimetrik (TGA), dan analisis Spektroskopi inframerah transformasi Fourier (FTIR). Pada level GNP yang rendah, nanokomposit yang disediakan telah meningkatkan ciri tegangan dengan ketara. Kekuatan tegangan nanokomposit meningkat dengan ketara apabila kandungan GNP dalam PLA dinaikkan, mencapai nilai maksimum 50.3 MPa dalam PLA dengan 0.3 wt% GNP berbanding dengan komposisi lain. GNP terdispersi secara seragam dalam matriks polimer PLA/0.3GNP, sesuai dengan hasil FESEM. Menurut penemuan kajian ini, nanokomposit PLA/GNP dengan 0.3 wt% GNP mempunyai sifat mekanikal terbaik daripada semua formulasi yang diuji. Kehabluran PLA diperhatikan meningkat pada 0.3 wt% daripada GNP daripada 27.0% kepada 32.7% berbanding dengan komposisi lain. Sementara itu, kandungan optimum pengisi pada 0.3 wt% digunakan untuk mengkaji degradasi PLA dan nanokomposit. Akibatnya, kehabluran PLA didapati meningkat sehingga 50.6% selepas luluhawa dipercepat. Sebaliknya, kehabluran PLA/0.3GNP, dan PLA/0.3GNP-COOH didapati meningkat masing-masing pada 58.1% dan 66.4% selepas luluhawa dipercepat. Tambahan pula, sifat mekanikal keseluruhan (iaitu, kekuatan tegangan, modulus Young's dan pemanjangan semasa putus) nanokomposit PLA, dan PLA menurun selepas luluhawa dipercepat. Punca utama pengurangan sifat mekanikal nanokomposit PLA didapati ialah penyerapan air, pemusnahan integriti nanofiller, degradasi matriks PLA, dan pembentukan retakan dan liang. Lekatan antara muka antara matriks PLA dan GNP dan GNP-COOH dilemahkan oleh cahaya UV, lembapan dan haba, yang menyebabkan nanokomposit merendahkan dan mengurangkan kestabilan mekanikal dan terma nanokomposit.

## ABSTRACT

One of the most researched biodegradable and biobased polymers is polylactic acid (PLA), which is readily available commercially for a reasonable price and has unique material properties. An aliphatic polyester, PLA has a wide range of essential uses in the packaging, automobile, biomedical, and agricultural industries. Nevertheless, it has some drawbacks, such as brittle in tensile behaviour and poor thermal properties. The introduction of nanofiller improves the polymer matrix's mechanical and thermal properties. Investigating the degrading behaviour of PLA/graphene nanocomposites is the main purpose of this study. The first stage of the study examines the optimum graphene nanoplatelets (GNP) contents of PLA/GNP nanocomposites based on mechanical, thermal, and morphological properties. The second stage is the degradation study of PLA and nanocomposites (i.e., GNP and carboxyl graphene nanoplatelets (GNP-COOH)) by accelerated weathering technique, including studying the thermal, mechanical and morphological properties of PLA nanocomposites. The PLA/GNP nanocomposite was prepared with various contents of GNP (0, 0.1, 0.3, 0.5, and 1.0 wt%) by melt blending method using a twin-screw extruder. The accelerated weathering was conducted with five different cycles (8, 16, 32, 48, and 64 cycles). The characteristics of the PLA and nanocomposites before and after the accelerated weathering test were analysed by using Field Emission Scanning Electron Microscopy (FESEM), X-ray diffraction (XRD), tensile test, differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), and Fourier Transform Infrared Spectroscopy (FTIR) analysis. At low GNP levels, the prepared nanocomposites significantly improved tensile characteristics. The tensile strength of nanocomposites increased considerably when the GNP content in PLA was raised, reaching a maximum value of 50.3 MPa in PLA with 0.3 wt% GNP compared to other compositions. The GNP dispersed uniformly in the PLA/0.3GNP polymer matrix, according to the FESEM results. According to the findings of this study, PLA/GNP nanocomposites with 0.3 wt% GNP had the best mechanical properties of all the formulations tested. The crystallinity of PLA was observed to increase at 0.3 wt% of GNP from 27.0% to 32.7% compared to other compositions. The optimum content of filler at 0.3 wt% was used to study the degradation of PLA and nanocomposites. As a result, the crystallinity of PLA was found to increase up to 50.6% after accelerated weathering. In contrast, the crystallinity of PLA/0.3GNP and PLA/0.3GNP-COOH was found to increase at 58.1% and 66.4% after accelerated weathering, respectively. Furthermore, the overall mechanical properties (i.e., tensile strength, Young's modulus and elongation at break) of the PLA and PLA nanocomposites decreased after accelerated weathering. The leading causes of reduction in the mechanical properties of PLA nanocomposites were found to be water absorption, destruction of nanofiller integrity, degradation of PLA matrix, and formation of cracks and pores. The interface adhesion between the PLA matrix and the GNP and GNP-COOH is weakened by UV light, moisture, and heat, which causes the nanocomposites to degrade and reduce their mechanical and thermal stability of nanocomposites.

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