POTENTIAL OF JATROPHA OIL AND DERIVATIVES AS AN ULTRAVIOLET CURABLE SELF-HEALING COATING

By

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Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Doctor of Philosophy

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Chair : Professor Luqman Chuah Abdullah, PhD
Faculty : Engineering

Coatings are applied at various substrates with the aim of protecting the surface and give aesthetic value. However, the coating degrades and loses its performance due to ageing and environmental factors. Therefore, self-healing coating is invented to extend the shelf-life of the protected material and hence requires less maintenance and monitoring service. This smart coating can repair damage or recover the functional performance of the coating with minimal or no intervention.

In coating preparation, the strategy of using vegetable oil to replace petrochemical resin in the coating industry has been started for a few decades. However, most of the oils that have been used such as soy, linseed and sunflower are edible oils. In Malaysia, jatropha oil is a natural source with high content of the unsaturated part. This feature reflects its reactivity during chemical modification and can potentially be applied in the coating industry. Moreover, it is a nonedible oil and does not compete with the food supply. As far as it is known, no study on an ultraviolet (UV) curable self-healing coating based on jatropha oil and its derivatives has been reported. This study investigates the feasibility of jatropha oil and its derivatives as a self-healing coating using UV curing to produce the film. Following that, the microcapsule embedment technique was chosen for the preparation of the self-healing coating.

The coating primer was prepared from jatropha oil-based polyurethane acrylate (JPUA), whilst the healing agent was pure jatropha oil (JO) and its derivatives were encapsulated into a microcapsule. JPUA-TDI had a higher viscosity and molar mass with a broader polydispersity index (PDI) value than JPUA-IPDI. With the addition of monomer and photoinitiator, coating formulations based on JPUA-TDI and JPUA-IPDI were developed and irradiated under UV subjected to the curing process. JPUA-TDI- and JPUA-IPDI-based films showed the best

mechanical properties based on coating formulation, with a monomer to JPUA ratio of 35:65. The JPUA-TDI-based coating formulation had a lower hardness but better adhesion than the JPUA-IPDI-based coating formulation. Pure JO and JPUA-IPDI coating mixtures behaved Newtonian at 25°C, whereas JPUA-TDI coating mixtures behaved as shear thickening fluids.

JO and JPUA-IPDI coating mixture were encapsulated into microcapsules by *in-situ* polymerisation, where polyurea formaldehyde was chosen as the shell material of the microcapsule. The variation in the agitation rate affected the morphology and the core content of the microcapsules. At the speed of 400 rpm, JO 400 (71.6 ± 2.7%) and IPDI 400 (40.0 ± 4.1%) had the highest core content. Therefore, JO 400 and IPDI 400 were chosen for self-healing coating preparation to be incorporated into the JPUA-TDI coating primer.

The microcapsules were loaded into the JPUA-TDI coating primer and exposed to UV light for the curing process for film thickness variations and the percentage of the microcapsule. At a film thickness of 50 µm with 5% loading of both JO and JPUA-IPDI-based microcapsules, respectively, the prepared self-healing coating had hardness and adhesion properties that were comparable with the control sample. During the scratch test, 5% JO did not show a self-healing property with response to air. The 5% IPDI showed self-repairing action on the scratch area after exposure to UV light. Proof-of-concept of self-healing coating is offered by jatropha oil derivatives where JPUA-TDI was used as the coating primer and JPUA-IPDI coating mixture as the healing agent in response to UV light.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

POTENSI MINYAK JATROPHA DAN TERBITAN SEBAGAI SALUTAN PEMBAIKAN KENDIRI BAGI SINARAN ULTRALEMBAYUNG TERMATANG

Oleh

NURUL HUDA BINTI MUDRI

Januari 2023

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Salutan telah digunakan pada pelbagai substrat dengan tujuan untuk melindungi permukaan dan memberikan nilai estatika. Walaubagaimanapun, salutan terurai dan hilang keupayaannya disebabkan penuaan dan faktor alam sekitar. Oleh itu, salutan pembaikan kendiri telah dicipta untuk menambah jangka hayat bahan yang dilindungi dan seterusnya mengurangkan kerja-kerja penyelenggaraan dan pemantauan. Salutan pintar ini mampu membaiki kerosakan atau mengembalikan fungsi keupayaan salutan secara minimum atau tiada tindakbalas luaran.

Dalam penghasilan salutan, strategi dengan menggunakan minyak tumbuhan untuk menggantikan resin petrokimia dalam bidang salutan telah dimulakan sejak beberapa dekad yang lalu. Walaubagaimanapun, kebanyakan minyak yang digunakan seperti soya, biji rami dan bunga matahari adalah minyak yang boleh dimakan. Di Malaysia, minyak jatropha merupakan minyak dari sumber semulajadi yang memiliki kandungan bahagian tidak tepu yang tinggi. Sifat ini menggambarkan kereaktifannya semasa pengubahsuaian kimia dan berpotensi untuk digunakan dalam industri salutan. Tambahan pula, ianya merupakan minyak yang tidak boleh dimakan dan tidak bersaing dengan rantaian makanan. Setakat yang diketahui, tiada kajian terhadap salutan pembaikan kendiri bagi sinaran ultralembayung termatang berasaskan minyak jatropha dan terbitannya telah dilaporkan. Kajian ini mengenalpasti kemampuan minyak jatropha (JO) dan terbitannya sebagai salutan pembaikan kendiri menggunakan pematangan ultralembayung untuk menghasilkan filem, manakala teknik mikrokapsul digunakan untuk menghasilkan salutan pembaikan kendiri.

Salutan asas disediakan daripada poliuritena akrilat berasakan minyak jatropha (JPUA), manakala agen pembaikannya terdiri daripada minyak jatropha dan terbitannya. JPUA-TDI mempunyai kelikatan dan berat molekut yang tinggi serta

nilai indeks poliserakan (PDI) yang lebih lebar berbanding JPUA-IPDI. Dengan penambahan monomer dan pemula foto, formulasi salutan berasaskan JPUA-TDI dan JPUA-IPDI telah dibangunkan dan disinarkan di bawah sinaran ultralembayung bagi proses pematangan. Filem berasaskan JPUA-TDI dan JPUA-IPDI menunjukkan sifat mekanikal terbaik pada nisbah monomer ke JPUA pada 35:65. Filem berasaskan formulasi JPUA-TDI mempunyai nilai kekerasan yang rendah tetapi kelekatan yang baik berbanding salutan formulasi berasaskan JPUA-IPDI. JO asli dan campuran salutan JPUA-IPDI bersifat Newtonian pada 25°C, manakala campuran salutan JPUA-TDI bersifat cecair penebalan ricih.

JO dan campuran salutan JPUA-IPDI dikapsulkan ke dalam mikrokapsul menggunakan pempolimeran *in-situ* manakala poliurea formaldehida dipilih sebagai bahan bagi penghasilan kelongsong mikrokapsul. Kepelbagaian kadar pengacauan memberi kesan terhadap morfoloji dan kandungan isirong mikrokapsul. Pada kelajuan 400 rpm, JO 400 (71.6 ± 2.7%) dan IPDI 400 (40.0 ± 4.%) mempunyai nilai isirong tertinggi. Oleh itu, JO 400 dan IPDI 400 telah dipilih bagi penghasilan salutan pembaikan kendiri serta digabungkan ke dalam salutan asas JPUA-TDI.

Mikrokapsul dimasukkan ke dalam salutan asas JPUA-TDI dan didedahkan kepada sinaran ultralembayung bagi proses pematangan bagi pelbagai ketebalan filem dan peratusan mikrokapsul. Bagi ketebalan filem 50 µm dengan 5% kandungan mikrokapsul berasaskan JO and JPUA-IPDI, salutan pembaikan kendiri yang dihasilkan mengekalkan sifat kekerasan dan kelekatan yang setara dengan sampel kawalan. Semasa ujian kecalaran, 5% JO tidak menunjukkan tindakbalas pembaikan kendiri terhadap dedahan udara. 5% IPDI menunjukkan pembaikan kendiri pada kawasan calaran setelah didedahkan dengan sinaran ultralembayung. Pembuktian konsep salutan pembaikan kendiri berjaya ditunjukkan bagi terbitan minyak jatropha di mana JPUA-TDI digunakan sebagai salutan asas dan campuran salutan JPUA-IPDI sebagai agen pembaikan semasa tindakbalas terhadap sinaran ultralembayung.

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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LIST OF ABBREVIATIONS

<	Less than
2,4-TDI	2,4-Toluene diisocyanate
AOCS	American Oil Chemists' Society
ASTM	American Standard for Testing Material
ATR	Attenuated total reflectance
d_{50}	Average particle size
DBTDL	Dibutyltin dilaurate
DMPA	Dimethylol propionic acid
EB	Electron beam
EC	Ethyl cellulose
EJO	Epoxidised jatropha oil
EW	Equivalent weight
FESEM	Field Emission Scanning Electron Microscopy
FTIR	Fourier Transform Infrared Spectroscopy
GPC	Gel Permeation Chromatography
h	Hour
ha	Hectare
HBr	Hydrogen bromide
HDDA	1,6-hexanediol diacrylate
HDI	1,6-hexamethylene diisocyanate
HEA	Hydroxyethyl acrylate
HEMA	2-hydroxyethyl methacrylate
IPDI	Isophorone diisocyanate
IV	Iodin Value
JO	Jatropha oil
JOL	Jatropha oil-based polyol
JPUA	Jatropha oil-based polyurethane acrylate
KHPt	Potassium hydrogen phtalate
MDI	4,4′-methylenediphenyl diisocyanate
Mn	Average molecular number

Mw	Molar mass
N	Normality
NMR	Nuclear Magnetic Resonance
OHV	Hydroxyl value
OOC	Oxygen oxirane content
Pa	Pascal
PAP	Pyridine phtalic anhydride
PDI	Polydispersity Index
PU	Polyurethane
PUA	Polyurethane acrylate
PUF	Poly-urea-formaldehyde
PVA	Polyvinyl alcohol
ROMP	Ring opening metathesis polymerisation
rpm	Rotation per minute
SDBS	Sodium dodecylbenzene sulfonic acid
SEM	Scanning Electron Microscopy
TCDDA	Tricyclodecane dimethanol diacrylate
TEA	Triethanolamine
T_g	Glass transition temperature
TGA	Thermal Gravimetric Analysis
THF	Tetrahydrofuran
TMPTA	Trimethylol propane triacrylate
TMPTMA	Trimethylolpropane trimethacrylate
UV	Ultraviolet
VO	Vegetable oil
VOCs	Volatile organic compounds
w/w	Weight by weight
wt	Weight
x	Times
ZrO_2	Zirconium oxide

CHAPTER 1

INTRODUCTION

1.1 Research Background

A coating is a mixture of film-forming elements that may contain a pigment, solvent, and additives. The purpose of a coating is to protect and beautify various surfaces, including metal, glass, wood, paper, and concrete. Coatings have been shown to protect surfaces from corrosion, keep them clean, and provide safety, particularly on slippery surfaces. In daily life, paints, drying oils, varnishes, and clear synthetic coatings are a subclass of the coatings that are commonly used for surface treatments (Javadi et al., 2020).

However, ageing and environmental factors can cause the coating to degrade. For example, during storage and transit of a structure, the protective coating for high-performance materials may develop microcracks. The microcrack would then spread and cause the coatings to fail if no effort was taken to correct the situation (Liu et al., 2012). Corrosion can occur on a metal substrate if corrosive agents such as air and moisture are present on the exposed surface. In some cases, corrosion can result in catastrophic events such as the collapse of buildings and bridges (Petrovic, 2016).

As a result, a natural-inspired self-healing coating has been developed. In plants and animals, healing action can occur through regeneration, repair, or replacement of damaged tissue (Cremaldi & Bhushan, 2018). Self-healing coatings are a smart material that can detect damage and subsequently initiate the repair response. The fascinating potential of self-healing coatings to self-restore the structural integrity when microcracks occur explains the growing interest in self-healing coatings. This functionality results in fewer maintenance and cost savings, which is particularly motivating for offshore constructions such as oil rigs and wind turbines (Nesterova et al., 2012). Such a coating can increase the working life and save money on active monitoring and external repair because of its auto-repairing response, especially when the plant needs to be shut down and stopped during preventive and maintenance services (Pannamma et al., 2017).

The first generation of self-healing materials was created in 2001 by the lab of Scott R. White at the University of Illinois (White et al., 2001). The self-healing process began when the implanted microcapsules ruptured due to a crack. Dicyclopentadiene, which was released as the core material from the burst capsules, reacted with Grubbs' catalyst, which was dispersed throughout the coating matrix. The polymerisation process occurred based on ring opening metathesis polymerisation (ROMP) to restore the crack (Wei et al., 2015).

Currently, self-healing coatings are applied for high-end applications such as wind turbine rotor blades, turbine blades, aviation, and the automobile industry (Lutz et al., 2016; Urdl et al., 2017).

1.2 Problem Statement

Protective coatings have a finite shelf life and can degrade due to external forces such as scratches and cracks. As a result, a self-healing coating has been invented to self-repair after damage and extend the working life of the coating while needing fewer inspections. The mechanism of the damage restoration to the initial state depends on the types of polymers employed. For thermoset polymers, a microcapsule-based system is a potential technique for an autonomous extrinsic system such as coating applications (Pannamma et al., 2017).

Vinyl, acrylic, epoxy, and polyester are examples of traditional petroleum-based chemicals used to make coatings. However, due to a global lack of fossil fuels, the price of these compounds fluctuates frequently in the market. Furthermore, they require hazardous solvents during processing and coating application, which may result in the production of volatile organic compounds (VOCs) that affect human health and the environment. Vegetable oil has been developed as a viable alternative, with economic effectiveness, biodegradability, and reduced solvent consumption. Aside from that, vegetable oils are typically derived from agricultural products, making them a renewable resource with abundant supply (Sharmin et al., 2015). Therefore, vegetable oil resin is expected to reduce the dependency on petrochemical resin in the coating industry. Soy (Sahoo et al., 2016), palm (Ling et al., 2014), olive, canola and castor (Zhang et al., 2015) are examples of vegetable oils that have been researched for surface coating application.

Palm oil has been the most important commodity in Malaysia since the 1970s. In 2020, Malaysia was responsible for 25.8% of global palm oil output and 34.3% of global palm oil exports (MPOC, 2022). However, edible oil as a feedstock has been criticised due to food supplies being used for non-food purposes (Taib et al., 2017). Jatropha oil (JO) has appeared as one of the most promising options as it is classified as non-edible oil. It contains a toxic phobic ester compound that should not be consumed (Ahmed & Salimon, 2009; Pandey et al., 2012). Furthermore, jatropha can be grown on marginal and non-agricultural sites without interfering with the production of existing food crops (Kalam et al., 2012). This will avoid the deforestation issue as highlighted by European countries concerning palm oil products (Amri et al., 2021). JO is extracted from the seed of the fruit of *Jatropha curcas* and contains about 79% of unsaturated fatty acid, which is mainly contributed by oleic acid (43.5%) and linoleic acid (35.2%) (Oskoueian et al., 2011). Apart from that, the iodine value (IV) of JO is around 94 to 120 mg/g (Abdullah et al., 2013; Khalil et al., 2013), which is higher than palm oil (44 to 58 mg/g). This reflects the sensitivity of JO during chemical modification compared to palm oil.

Radiation curing is an effective process for turning a liquid into a solid material. UV (ultraviolet) and electron beam (EB) curing systems are two types of radiation curing systems (Abliz et al., 2013). UV curing is preferred over EB curing because the UV curing equipment is simple and cost-effective. UV curing currently accounts for over 90% of the radiation curing business in Europe. UV curing requires using a photoinitiator to initiate the curing process, which results in a thin film (Sangermano et al., 2014). Moreover, no solvent has been released to the atmosphere during the UV curing process as all components, such as oligomer, monomer and photoinitiator, are crosslinked efficiently within a few seconds (Glöckner et al., 2008). Furthermore, compared to thermally cured coatings, this process has advantages such as a more comprehensive formulation range, lower energy usage, and is suitable for coating heat-sensitive substrates (Habib & Bajpai, 2011). As a result, the sophisticated self-healing coating technology, which combines biodegradable jatropha oil with an efficient and safe UV curing procedure, provides an effective solution to lengthen the shelf life of the coating while also achieving future sustainable development.

1.3 Objectives of Study

The main aim of this study is to investigate the feasibility of jatropha oil and its derivatives as a self-healing coating using the microencapsulation method. To achieve this goal, the following specific objectives have been established:

a. To examine the effect of diisocyanate correlating with the physical, chemical and thermal properties of jatropha oil-based polyurethane acrylate (JPUA);
b. To optimise the coating formulation containing pure jatropha oil and mixture of JPUA with the variation ratio of the monomer using mechanical tests and rheological study;
c. To investigate the influence of stirring rate during the preparation of microcapsules containing the jatropha oil and its derivatives in connection with the physical, chemical and thermal properties of the microcapsules; and
d. To demonstrate the concept of a self-healing coating when exposed to air and UV light and evaluate the mechanical properties.

1.4 Scope of Study

The scope of the present research is explained as follows:

a. To produce the jatropha oil-based polyurethane acrylate (JPUA), three stepwise chemical reactions known as epoxidation, hydroxylation, and isocyanation were performed. During the synthesis of jatropha oil-based polyol (JOL), the chemical reaction of epoxidation and hydroxylation was performed according to (Saalah et al., 2017) with minor modification. Two series of diisocyanates were selected during isocyanation; 2,4-toluene diisocyanate (2,4-TDI) and isophorone diisocyanate (IPDI) to be reacted with the synthesised polyol with hydroxyethyl methacrylate (HEMA) as a chain terminator. The molar ratio between the polyol:diisocyanate:chain terminator was calculated to be 1:1:1. The influence of the different diisocyanates on the physical, chemical and thermal properties of the JPUA is to be investigated.

b. The preparation of the UV curable coating involves the various ratios between the monomer and the synthesised JPUA. This study chose trimethylolpropane triacrylate (TMPTA) as the monomer in the coating formulation. The monomer:JPUA ratio was set at 25:75, 30:70, 35:65, 40:60, 45:55 and 50:50. The benzophenone was selected as the photoinitiator and fixed at 4% (*w*/*w*) of total amount of monomer and JPUA. After the coating and UV curing process, the effect of JPUA types at different monomer ratios was examined for hardness, adhesion, haze, and transmission tests. Rheological study was conducted to determine the feasibility of the healing agent during the encapsulation process. The pure jatropha oil and the best JPUA coating formulation (based on mechanical properties) were examined for the rheological study at a shear rate of 0 to 100 s^{-1} with a temperature of 25°C, 40°C, 60°C, and 80°C respectively to predict the fluid behaviour during the microcapsule preparation.

c. During the synthesis of a microcapsule containing healing agent, the stirring speed parameter was chosen to be 200 rpm, 400 rpm and 800 rpm. The relationships between stirring speed and particle size, morphology and core content will be discussed.

d. For the self-healing coating formulation, the percentage of microcapsule loading was examined for 5% and 10% with a thickness of 50 µm, 100 µm, 150 µm and 200 µm respectively. The self-healing coating without microcapsule loading was used as a control. The effect of microcapsule loading on hardness, adhesive, haze and transmission properties will be investigated. The proof-of-concept of the self-healing coating is to be evaluated by a scratch test after exposure to air and UV light for pure JO and JPUA based healing agents, respectively.

1.5 Significant Contribution and Limitations

Mechanical damage and weathering causes a cracking problem that may cause a deterioration of both coating and substrate (Wang & Zhou, 2018). In a microcrack problem, detecting and repairing the damaged area is difficult, especially for bulk materials (Ataei et al., 2019). Therefore, self-healing coatings were invented with the idea of automatically repairing features (Zhang et al., 2018). The addition of microcapsule that containing a reservoir of healing agent give extra protection and may increase the lifespan of the coating and substrate. At the same time, the need for frequent surface treatment can be avoided and thus save cost on preventive and maintenance service. Moreover, the microcapsule-based self-healing coating allow a controllable releasing of the healing agent only in crack or scratch occurs (Shisode et al., 2018). These features become a motivating factor for technologists to continuously develop self-healing coating.

The prepared microcapsules are typically in the submicron range of up to 1 mm with healing agents inside them. As cracks propagate, the healing agent flows into the crack lines. The healing agents come into contact with the active materials, inducing polymerisation and finally sealing the crack (Mirabedini et al., 2015). However, this method is limited to one cycle repairing mode. Once the healing agent has been used, the microcapsule-based self-healing coating cannot restore the crack if it occurs in the same area (Hia et al., 2016). Apart from that, the processing and application of microcapsule-based self-healing coating require temperature conditions of less than 200 ℃. This is due to the limitation of the thermal stability of the shell materials (polyurea formaldehyde) and the healing agent (vegetable oil-based resin), as reported in previous studies (H. Li et al., 2017; Saman et al., 2018; Shisode et al., 2018).

1.6 Structure of Thesis

The thesis is divided into seven chapters. The background of the current research is explained in Chapter 1, which includes an introduction to the concept of a self-healing coating, as well as the problem statement and the scope of the study.

In Chapter 2, the literature concerning polyurethane, vegetable oil, radiation curing, jatropha oil and self-healing coating using the microencapsulation method was reviewed. Chapter 3 demonstrates the experimental procedure for synthesising jatropha oil-based polyurethane acrylate, the UV curing method, followed by mechanical tests and rheological studies. For the self-healing part, the process of encapsulating the jatropha oil-based healing agent and the evaluation of a self-healing coating will be explained.

Chapter 4 discusses the results of the synthesised JPUA. This includes the effect of different diisocyanates on the structural, chemical, and thermal properties of the resin. The results of the mechanical and rheological properties of the coatings based on JPUA are demonstrated in Chapter 5.

For the microencapsulation parts, the results and discussions are presented in Chapter 6. This chapter also covers the proof of concept of jatropha oil and its derivatives as a self-healing agent. Finally, the conclusion and the recommendation are summarised in Chapter 7.

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