THERMO-CHEMICAL AND MECHANICAL PROPERTIES OF TEA TREE *(MELALEUCA ALTERNIFOLIA)* FIBRE REINFORCED TAPIOCA STARCH COMPOSITES

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FACULTY OF SCIENCE AND NATURAL RESOURCES
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ABSTRACT

Melaleuca alternifolia or commonly known as tea tree is a tall shrub or small tree in the plant genus Melaleuca. It is popular for its oil, which is tea tree oil where it has been employed largely in various industries of its antimicrobial properties. Tea tree fibres as the underutilized fibres were investigated physically, chemically and mechanically. From this study, it was found out that the tea tree leaf (TTL) had the highest density 0.42 g/cm^3, and had the highest percentage of water absorption, 69.9%. From the tensile strength, tea tree trunk (TTT) gave the highest value, 65.44MPa, followed by tea tree branch (TTB), 48.43MPa and tea tree leaf (TTL), 47.47MPa. The chemical composition of tea tree fibres showed TTT had the highest cellulose content, which is 33.9%, followed by TTB, 27.2% and TTL, 13.5%. Meanwhile, TTL had the highest extractive value, 16.4%, almost 3 times higher than TTB and TTT due to the existence of tea tree oil in TTL. The fabrication of tea tree fibres reinforced tapioca starch (TS) composite successfully developed using casting method. The physical, thermo-chemical, and mechanical properties are undergone in order to get the characterization of the composite. The addition of tea tree fibres do not affect the chemical properties of composite as shown in Fourier transform infrared (FTIR) results, where all of the tea tree fibres and its reinforced composites show same pattern. From the mechanical test, the addition of 5% v/v of tea tree fibre as filler, improved the tensile strength of TS composite up to 34.39% in TTL/TS, 82.80% in TTB/TS and 203.18% in TTT/TS. Thermogravimetric analysis (TGA) proves that tea tree fibres increase thermal stability of composites. Scanning electron microscopy (SEM) shows a good dispersion of the tea tree fibre in the TS matrix. The water absorption and swelling thickness test of all tea tree fibres reinforced composites decreased compared to TS composite. Above all, all parts of the tea tree waste, namely TTL, TTB and TTT have the potential novel fibres which can act as reinforcement in developing a green biocomposite.

ABSTRAK

SIFAT-SIFAT TERMO- KIMIA DAN MEKANIKAL BAGI SERAT TEA TREE (MELALEUCA ALTERNIFOLIA) BERTETULANG KANJI UBI KAYU

Melaleuca alternifolia atau lebih dikenali sebagai tea tree merupakan pokok renek tinggi atau pokok kecil dalam genus tumbuhan Melaleuca. Pokok ini sangat popular dengan minyaknya, di mana ia telah digunakan dalam pelbagai industri kerana sifat anti-mikrob yang terkandung dalam minyaknya. Serat tea tree yang jarang diguna dan diberi perhatian secara umumnya, telah dikaji dari segi fizikal, kimia dan mekanikal. Daripada kajian ini, didapati bahawa daun tea tree (TTL) mempunyai ketumpatan paling tinggi 0.42 g/cm^3 dan mempunyai peratusan tertinggi penyerapan air, iaitu sebanyak 69.9%. Dari segi kekuatan mekanikal, batang tea tree (TTT) memberi nilai yang paling tinggi, 65.44MPa, diikuti ranting pokok tea tree (TTB), 48.43MPa dan daun tea tree (TTL), 47.47MPa. Komposisi kimia serat tea tree menunjukkan TTT mempunyai kandungan selulosa tertinggi, iaitu 33.9%, diikuti TTB, 27.2% dan TTL, 13.5%. Sementara itu, TTL mempunyai nilai ekstraktif tertinggi, iaitu sebanyak 16.4%, hampir 3 kali lebih tinggi daripada TTB dan TTT kerana kewujudan minyak tea tree dalam TTL. Fabrikasi serat tea tree bertetulang kanji ubi kayu (TS) komposit berjaya dihasilkan menggunakan kaedah casting. Sifat-sifat fizikal, termo-kimia, dan mekanikal bagi komposit telah dikaji. Penambahan serat tea tree tidak mempengaruhi sifat kimia komposit seperti yang ditunjukkan dalam keputusan Fourier transform infrared (FTIR) di mana kesemua serat tea tree dan komposit bertetulangnya menunjukkan paten yang sama. Melalui ujian mekanikal, penambahan 5% v/v serat tea tree sebagai tetulang, meningkatkan kekuatan tegangan TS komposit sehingga 34.39% dalam TTL/TS, 82.80% dalam TTB/TS dan 203.18% dalam TTT/TS. Thermogravimetric analysis (TGA) membuktikan bahawa serat tea tree meningkatkan kestabilan haba komposit. Scanning electron microscopy (SEM) menunjukkan penyebaran serat tea tree yang sekata di dalam matrik TS. Ujian penyerapan air dan pengembangan ketebalan semua serat tea tree komposit bertetulang menurun berbanding TS komposit. Secara tuntasnya, semua bahagian sisa serat tea tree, iaitu TTL, TTB dan TTT memiliki sumber serat novel yang berpotensi dan boleh berfungsi sebagai tetulang dalam menghasilkan biokomposit mesra alam.

LIST OF CONTENTS

CHAPTER 4: RESULTS AND DISCUSSION

CHAPTER 5: CONCLUSIONS AND RECOMMENDATIONS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

ASTM	American Society for Testing and Material
AUD	Australin Dollar
A.D	Air dry
FTIR	Fourier transform infrared
ISO	International Standards Organisation
KS	Potassium sorbate
NaAlg	Sodium alginate
NaOH	Sodium Hydroxide
NFM	Non-fibrous material
O.D	Oven dry
OPEFB	Oil palm empty fruit bunch
PALF	Pineapple leaf fiber
PE	Polyethylene
PCL	Poly-3-caprolactone
PHA	Polyhydroxy-alkanoate
PLA	Polylactide
PP	Polypropylene
PVC	Polyvinyl chloride
SEDIA	Sabah Economic Development & Investment Authority
SEM	Scanning electron microscope
SPI	Soy protein isolate
TAPPI	Technical Association of the Pulp and Paper Industry
TGA	Thermogravimetric analysis
TS	Tapioca starch
TTB	Tea tree branch
TTL	Tea tree leaf
TTO	Tea tree oil
TTT	Tea tree trunk
WGL	Whole ground pineapple leaf
TTB/TS	Tea tree branch reinforced tapioca starch

TTL/TS	Tea tree leaf reinforced tapioca starch
TTT/TS	Tea tree trunk reinforced tapioca starch

LIST OF EQUATIONS

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 Background of the Study

Malaysia is one of the tropical countries blessed with large reserved rainforest areas. The land which are fertile, offer so much plants to grow. One of the potential plants that is famous for its essential oil is tea tree *(Melaleuca alternifolia)* (Indu and Ng, 2000). Tea tree oil is widely used in many kinds of value-added products as a preservative, antiseptic, antibacterial, antifungal and even anti-pest agent (Southwell and Lowe, 1999; Colton *et al.*, 2000). Tea tree oil was one of the products with success stories in Australian essential oil production during the early 90's with prices topping AUD 65/kg. This attracted large numbers of new growers entering the industry until production exceeded demands and prices tumbled to less than AUD 50/kg, especially in the late 90's (Davis, 1999). This has shaken out the industry and all the largest producers ceased production, leaving small and medium sized producers to supply the market.

Few years after that, prices have been on the increase again and are breaching the AUD 50/kg mark and are still increasing. However, many large customers including some of the major European and USA retail chains who carried the products have discontinued their demand, due to lack of confidence in future supply. Likewise, many major personal care companies have also switched to other natural additives in their product range, leaving the task ahead for tea tree producers to convince the cosmetic and retail industries to support the product. This unstable period has opened the door for producers in countries like China and other Asian countries to step up production and compete with the Australian producers.

We have witness a few decades back, the depletion of petroleum resources coupled with awareness of global environmental problem, create the alternatives for a new green material, compatible with environment and independent to fossil fuels. These natural fibre products based on renewable plant and agricultural residues are eco-efficient and sustainable, and can be used in various applications such as automotive, building, furniture and packaging industries which currently dominated by products based on petroleum feedstock. However, products made from natural fibre composites are yet to be seen in magnificent numbers.

Customers these days are looking forward for products that can satisfy their needs in terms of safety, quality and cost. As a result, scientists, technologists and researchers work hard to produce a new product that can fulfill those needs while maintaining the technical standards at a reasonable cost. The balance between those three factors will ensure the success of the product development and satisfaction of the customers. Economic as well as environmental factors are recently driving the trend towards utilization of material based on natural sources.

Increasing awareness among the world population to protect our environment has promoted research in agriculture residues. This is due to the abundant sources of agriculture crop wastes that cause problem to handle them. Agricultural residues such as in oil palm, pineapple leaf, banana, sugar palm and many more produced in billions of tons around the world. They can be obtained in abundance, low cost, and they are also renewable sources of biomass. Among these large amounts of residues, only small quantities of these residues are converted as household fuel or fertilizer. The major portion of the residues is burned which results in negative impact to our environment due to its contribution to air pollution (Abdul Khalil *et al.*, 2008).

One of the vital alternatives to solve this problem is to use the agriculture residues as reinforcement material in the development of composites. It is not necessary to produce 100% biobased material as subtitutes for petroleum-based material immediately. A viable solution is to combine petroleum and bioresources to produce a useful product for our daily applications (Mohanty *et al.*, 2005). These new green resources have many advantages such as low cost, low density, abundance, sustainability, recyclability and biodegradable which makes natural fibre more interesting to the applied research community in order to tap the full potential of the materials.

This research is to utilize by-products from the tea tree. Its purpose is to convert all the by-products into value-added product such as biocomposites. In order to completely drive the potential of tea tree fibres for new applications, a comprehensive and detailed investigation of fundamental information such as chemical composition and mechanical properties are necessary (Abdul Khalil *et al.*, 2008). Hence economic utilization of these fibres can be beneficial to our nation.

1.2 Problem Statements

Petroleum has contributed many things to mankind since ancient times. The most important part is the invention of the internal combustion engine for transportation. Today, about 90% of vehicles are moving around the world with fuel from petroleum (Sahari, 2013b). Besides, our qualities of lives are improved due to availability of plastics such as plastic bags, bottles, plates, cups and others, which are derived from petroleum. Plastics have already substituted many traditional materials, such as stone, leather, wood, metal and ceramic due to its good performances. Petroleum resources are the only major mineral commodities where many parties fear that their depletion will cause significant scarcities over the decades to come.

Environmental problem is also one of the issues why industries turn to natural fibre in producing new type of products. The impact of agriculture on air, land and water are issues that must be considered for the benefit of our next generation. Even though synthetic fibre such as polyester, acrylic, nylon, rayon, asbestos, acetate, kevlar and more is very useful in composite materials, it would not be degraded when disposed and causes negative effect to the environment (Bachtiar *et al.*, 2008; Leman *et al.*, 2008). The issues of climate change and global warming due to deforestation also become a major reason why agriculture residues are used, despite of limited space to handle all of these residues. The use of natural fibres as reinforcement in biocomposites can contribute in reducing the dependency on timbers.

In term of cost, synthetic fibres are more expensive compared to natural fibre. The limited source of timber also makes the price of wood higher. This phenomenon is to protect our forest from deforestation. For these reasons, enormous efforts, time and money have been spent on research to discover a new green material that can replace synthetic fibres and timber. In addition, synthetic fibres have serious effects to human health especially to the skin and eye. More seriously, inhalation of fibrous synthetic fibres such as asbestos can cause long terms or deadly effects such as lung cancer (Ishak, 2009).

Investigation on natural fibre including banana pseudo-stem, kenaf, pineapple leaf, rice husk and bamboo has gained popularity among the researchers recently because of its abundantly available, renewable as well as degradable materials (Awang *et al.*, 2009). However, tea tree have not been widely studied among the researchers all over the world. The importance of tea tree in our daily life is the tea tree oil, where it has been employed largely in various industries of its antimicrobial properties, which lead to areas of preservative, antiseptic, antibacterial, antifungal and even anti-pest agent (Southwell and Lowe, 1999; Colton *et al.*, 2000). Various research works are still ongoing mainly focusing on tea tree oil, however none of them investigating on the leaf residue which contain the fibre. Apparently, the new potential green materials which are produced by the combination of tea tree fibre from different parts of tea tree reinforced polymer composites such as tapioca starch can reduce the usage of petroleum and synthetic fibre.

1.3 Research Objectives

The general aim of this study is to determine the thermo-chemical properties and mechanical properties of tea tree *(Melaleuca alternifolia)* fibre (leaves, branch and trunk) reinforced tapioca starch composites.

The specific objectives of this research are:

a. To study the physical, chemical and mechanical properties of tea tree fibre (leaves, branch and trunk)

b. To develop new biocomposites from tea tree fibre (leaves, branch and trunk) reinforced tapioca starch

c. To analyse the physical, thermo-chemical and mechanical properties of tea tree fibre (leaves, branch and trunk) reinforced tapioca starch composites.

1.4 Significant of study

The significance of this study is to utilize the new resources of natural fibre which can be found in abundance from tea tree. The natural fibres become the most important material in developing the biocomposite products. These biocomposites are low in cost production, renewable, partially biodegradable and environmentally friendly. Besides, the use of natural fibre composite for the applications of furniture and building is expected to reduce the demand of timber which is now facing the problem of deforestations. This idea can also help to solve the problem of handling residue from tea tree. It also plays an important role in upgrading the economic life to the poor rural people through establishment of employment (Mogea *et al.*, 1991; Mahmud, 1991). Moreover, this study is significant as it would generate new ideas and could evaluate new potential use of natural resources tea tree waste and turn them into new valuable green products.

1.5 Scope and Limitation of Study

This study focuses on the measurement of chemical and tensile properties of single fibre from different parts of tea tree such as tea tree leaf, tea tree branch, and tea tree trunk. The chemical properties include the measurement of moisture content, extractive (TAPPI standard TS os-73), lignin (TAPPI standard T 222os-74), holocellulose (Wise *et al.*, 1946), cellulose (TAPPI standard T203 os) and ash (TAPPI standard T15 os-58). The Fourier transform infrared (FTIR) spectroscopy was used to detect the presence of the functional groups that exist in tea tree fibre. The tensile properties of single fibres were conducted using the ASTM D3379-75 (1989) standard. The mechanical testings such as tensile test was performed in order to determine the mechanical properties of the tea tree fibre reinforced tapioca starch composites. These testings were carried out in accordance to ASTM

D5083 (1996). Thermogravimetric analysis (TGA) was carried out to measure the mass change, thermal decomposition and thermal stability of material. Finally, the observation on the surface morphology of the fibres and fractured surface of composites failure test specimen completed using scanning electron microscope (SEM).

1.6 Structure of Thesis

Chapter 1 presents the background of study, problem statements, objectives, significance of the study, scope of the study and structure of thesis. A literature review on previous research work in various areas which is relevant to this research is presented in Chapter 2. The chapter started with a comprehensive literature survey on the natural fibre and tea tree. Review of the chemical and mechanical properties of fibres and its composites are also included in this chapter. The methodology of the study is described in Chapter 3. This chapter presents the determination of tensile and chemical properties of single fibre from different parts of tea tree. This chapter also includes the techniques for preparation of composites and the determination of the mechanical properties of tea tree fibre reinforced tapioca starch composite. Chapter 4 presents the results and discussion of the tensile and chemical properties for single fibre. It also includes the discussion on mechanical properties of the tea tree fibre reinforced tapioca starch composite. Surface morphology of the fibre and fractured specimen is also evaluated in this chapter using scanning electron microscope (SEM). Thermal decomposition and thermal stability of material also evaluated through thermogravimetric analysis (TGA). Finally, Chapter 5 presents the conclusions and recommendations for future works.