POTENTIAL OF USING FRUIT AS NATURAL DYE

SITI MARIAM BINTI AZMI

FACULTY OF EARTH SCIENCE

UNIVERSITI MALAYSIA KELANTAN

2017

POTENTIAL OF USING FRUIT AS NATURAL DYE

by

SITI MARIAM BINTI AZMI

A report submitted in fulfilment of the requirements for the degree of Bachelor of Applied Science (Sustainable Science) with Honours

FACULTY OF EARTH SCIENCE

UNIVERSITI MALAYSIA KELANTAN

2017

DECLARATION

I declare that this thesis entitled “Potential of Using Fruit as Natural Dye” is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

Signature: _____

Name: SITI MARIAM BINTI AZMI

Date: _____

ACKNOWLEDGEMENT

First of all, my praise is to Allah, the Almighty, the most Gracious and the most Compassionate which we depend for guidance. Then my appreciation goes to my supervisor, Dr Nurul Akmar Che Zaudin, who always giving me supports, guidance, extra information, also careful reading and constructive comments that were valuable for me. Her timely and efficient contribution helped me to shape this into its final form.

My sincere thanks go to my friends who helped me a lot during my laboratory sessions. And also, thanks, tribute and appreciation to Puan Faridah Mat Nor, the staff of Kraftangan Malaysia Cawangan Kelantan for her cooperation and guidance in completing my laboratory sessions there. An appreciation is also given to all my examiners who have contributed to the successful completion of this thesis.

I would like to thank my family for their support. I am deeply and forever indebted to my parents, Azmi Ibrahim and Che Pah Ismail, for their love, support and encouragement throughout my entire life.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

BSLA	Brine Shrimp Lethal Assay
$CuSO_4$	Copper Sulphate
G	Gram
ISO	International Organization for Standardization
LC_{50}	Lethal Concentration of 50%
mL	Millilitre
o.w.f	On Weight Fabric
UV	Ultraviolet
µg/mL	Microgram per Millilitre

LIST OF SYMBOLS

%	Percentage
>	Greater than
<	Less than
°C	Temperature (degree Celcious)
µ	Micro (Mu)
×	Multiplication

Potential of Using Fruit as Natural Dye

ABSTRACT

Synthetic dyes in textile industry are associated to be toxic and could harm the environment and human health. Following these issues, dyes derived from natural sources have emerged as an important alternative to synthetic dyes. Therefore, this study aimed at production of natural dye extract from mangosteen peel using distilled water. The content of the dye extract was manipulated with and without addition of copper sulphate mordant using simultaneous mordanting method where the presence of the mordant gave various hues on cotton and silk fabrics. The dye extract produced was aimed to be tested for toxicity test and colour fastness test to wash and light exposure. Toxicity of the dye extracts with and without mordant was investigated by using Brine Shrimps Lethality Assay (BSLA). 7 different concentrations of the dye extracts was used for toxicity test containing 10 matured brine shrimps; 15.625, 31.25, 62.5, 125, 250, 500 and 1000 µg/mL, for triplicate. The dye extracts with and without mordant were found to be non-toxic with the LC_{50} value of the extracts were more than 1000 µg/mL, according to Meyer's toxicity index. The resulting wash and light fastness of the fabrics were moderate to good and the mordanting did not result in any significant improvement in fastness properties compared to dye extract without mordant. The indication of accepted fastness properties were according to the ISO 105-C01 and ISO 105-B02 for wash fastness and light fastness, respectively. Based on the studies on toxicity and fastness properties, it was inferred that natural dye from mangosteen peel has good potential to be commercialized in textile industry.

Potensi Menggunakan Buah Sebagai Pewarna Semula Jadi

ABSTRAK

Pewarna sintetik dalam industri tekstil adalah bertoksik dan boleh memberi kesan buruk kepada alam sekitar dan kesihatan manusia. Berikutan isu ini, pewarna yang diperbuat daripada sumber semula jadi telah muncul sebagai alternatif penting untuk menggantikan pewarna sintetik. Oleh itu, kajian ini bertujuan untuk pengeluaran ekstrak pewarna semula jadi daripada kulit manggis dengan menggunakan air suling. Kandungan ekstrak pewarna telah dimanipulasikan dengan dan tanpa penambahan mordant sulfat tembaga menggunakan kaedah mordant serentak, di mana kehadiran mordant itu memberikan kepelbagaian warna pada kain kapas dan sutera. Ekstrak pewarna dihasilkan bertujuan untuk menguji ketoksikan dan mutu ketahanan warna kepada basuhan dan pendedahan cahaya. Ujian ketoksikan ekstrak pewarna dengan dan tanpa mordant dijalankan dengan menggunakan asai kematian udang air garam (BSLA). 7 kepekatan yang berbeza daripada ekstrak pewarna telah digunakan untuk ujian ketoksikan yang mengandungi 10 udang air garam yang matang; 15.625, 31.25, 62.5, 125, 250, 500 dan 1000 μg/mL, untuk tiga kali ulangan. Ekstrak pewarna dengan dan tanpa mordant didapati tidak toksik menurut indeks ketoksikan Meyer, dengan nilai LC_{50} daripada ekstrak lebih daripada 1000 μg/mL. Yang terhasil daripada ujian mutu ketahanan warna kepada basuhan dan cahaya pada fabrik kapas dan sutera adalah sederhana kepada baik, dan penambahan mordant tidak menyebabkan apa-apa peningkatan yang ketara dalam sifat-sifat mutu warna berbanding pewarna ekstrak tanpa mordant. Piawaian bagi mutu ketahanan warna yang diterima adalah mengikut ISO 105-C01 dan ISO 105-B02, masing-masing untuk mutu ketahana basuhan dan mutu ketahanan cahaya. Berdasarkan kajian mengenai ketoksikan dan mutu ketahan warna, ia disimpulkan bahawa pewarna semula jadi daripada kulit manggis mempunyai potensi yang baik untuk dikomersialkan dalam industri tekstil.

CHAPTER 1

INTRODUCTION

1.1 Background of Study

Natural dyes provide rare, soothing and light hues compared to artificial dyes (Alemayehu and Teklemariam, 2014). The used of synthetic dye could lead to adverse impacts to the environment and human health although its use would help to divulge strong colour (Adeel *et al.*, 2009).

This initiative is not only beneficial for the health but also create awareness for the environment as a sustainable source of dyes for the local food and textile industries (Summoogum-Utchanah and Joyram, 2015). The alternative of using natural sources in dye industry is one of the efforts to protect and conserve our environment and its ecological system from the consequences of abundant synthetic dyes (Alemahayu and Teklemariam, 2014).

Besides, abundance of wastes is contributed by industries such as food and beverages which generated huge amount of bio-wastes such as vegetables and fruit peels. Hence, these wasted parts can be fully exploited for extraction of dyes and their synthetic counterparts. Consequently, production of natural dyes needs to be boosted to fulfil the demand of the market and reduce the waste disposal issues. This effort of exploiting wasted parts for extracting natural dyes was first taken by Germany, followed by Netherlands, India and some other countries (Patel, 2011; Alemahayu and Teklemariam, 2014; Summoogum-Utchanah and Joyram, 2015).

However, there have been some problems in textile dyeing using natural dyes such as the colour produced, complexity of the processes, reproducibility

consequences, inadequate shades yield, blending problems and poor fastness properties (Sachan and Kapoor, 2007; Siva, 2007). Using mordants, the metal salts that strengthen the bond of the fabric with the dye molecules, can be initiatively solved these issues in textile natural dyeing process (Vankar *et al.*, 2009; Samanta and Agarwal, 2009).

In this study, mangosteen peel was used as the source of natural dye. Mangosteen or *Garcinia mangostana* L. is a tropical fruit origin in Southeast Asia. It is typically well-known as "the queen of fruit" in Thailand. The peel instantly become unwanted and discarded once the fruit being used (Maiaugree *et al*., 2015). It is able to produce staining juice that can leave marks on hand and clothes. This noticeable feature of mangosteen peel shows the possibility of the peel to be produced as a natural dye (Batisah, 2015).

There are two types of sample fabrics that were used in this study. The first sample was a natural cellulose fiber or commonly known as cotton material. It is mostly used in the textile industry because of its soft and comfortable while touch. It has high absorption ability, easy in handling and has good sewing qualities. Other than cotton, silk was also used in this study. Silk has the strongest natural protein fiber and has its own luster. It absorbs moisture, that cooling in the summer and warming in the winter, contrarily. The good absorption ability of silk makes it definitely stain in many deep colours (Deshpande and Chaturvedi, 2012).

According to Samanta and Agarwal (2009), the dyed materials are tested for light fastness and wash fastness where the colour fastness tested on both cotton and silk are generally graded by the changes of colour depth in original sample or could also be rated by discolouration scales.

1.2 Problem Statement

The commonly used synthetic dyes in textile dyeing could give adverse impacts to the environment and human health as they are known to be toxic (Sivakumar *et al*., 2000; Ozturk and Abdullah, 2006). Toxicant contained from the synthetic dyes effect the natural water sources through the wastewater emitted. As an alternative to synthetic dyes, natural dyes in fabric dyeing has emerged to possibly reduce the potential risk caused by the synthetic dyes. However, the natural dyes applied on fabrics generally have poor fastness properties compared to the synthetic dyes. Therefore, the fastness properties of the peel of mangosteen are important to be investigated as they have never been studied before. The findings will be useful for future study on this topic.

1.3 Objectives

1. To produce natural dyes from mangosteen peel by extraction method.
2. To determine the toxicity level of natural dyes produced.
3. To determine the light and wash fastness properties of natural dyes produced on cotton and silk fabrics.

1.4 Significance of Study

The findings of this study proved that the mangosteen peel discarded can be extracted as natural dye. This study is beneficial for environment and human health. By mitigating the use of synthetic dye in textile industry, the environment became healthier as the wastewater emitted can be less affected by the flow of chemical contained in synthetic dyes used.

The extract can also be harmless and safe to be used because of the less toxicity of natural dye. This study also beneficial the textile industry as the sources to extract the natural dye are discarded as wastes.

CHAPTER 2

LITERATURE REVIEW

2.1 Natural Dye For Textile Coloration

The use of colour extraction from fruits as a natural dye for various purposes, including textiles and food colouring has been studied by researchers as an effort to protect the ecosystem from the potential hazards caused by the use of some synthetic dyes.

According to Samanta and Agarwal (2009), as compared to artificial dyes, natural dyes give very rare, gentle and light hues. Synthetic or artificial dyes, on the other hand could be more commercial as it yields colours variability. These dyes however cause much harmfulness to human body and environment. They are produced by the process of isolation of chemicals from the by-products of petroleum at a particular condition with high temperature and pressure (Barhanpurkar *et al*., 2015).

The dyestuffs from artificial chemicals production contribute in economic sector and have become profitable in textile industry. However, there are many cancer-causing chemicals used during the industrial process of the dyes that would lead to the production of contaminated by-products. These by-products are released into the main water flow such as rivers or left in open, hence caused undecorated water and air pollution (Barhanpurkar *et al*., 2015).

Today, the global awareness prefers the application of natural obtainable resources for the purposes of conserving the environment and ecosystem from contamination and other environmental problems. This current phenomenon is

concerned more towards the exploitation of the infinite variety of colour pigments obtained from natural resources for their consumption in many purposes including foodstuff provisions, medications and fabrics, corresponding to their artificial complements (Jothi, 2008). The emergent of natural dyes that are non-hazardous and environmental friendly on textiles has an importance to improve the concerns toward nature. Accordingly, this is to evade certain harmfulness from synthetic dyes (Agarwal, 2009).

Devi and Karuppan (2015) reported that the high potential and their wide ranges of variability caused the use of natural dyes on textile fabrics became more probable to be used as colorant than synthetic dyes. The potential of natural dyes to be used on textile fabrics has been attracting many associated studies recently.

In textile industry, there are three main continuous processes of applying natural dyes on fabric materials, in which all the steps are related to each other. The steps are first started by extracting dye from natural source, mordanting, and dyeing, where the mordanting method is the most significant process. This is because the nature did not improve colours with textiles by itself unless with the help of metallic salt, mordant. Natural dye could not bond naturally with textile fibers. Thus, the use of mordant as a medium is essential to bond the dye with textiles. This proved that the combination of both natural dyes with a specific amount of metallic salts helps to serve different results for textile coloration (Freeman, 2015).

Natural dyes are frequently applied on cotton and silk. The concentration of the dye extraction applied on cotton and silk fabrics, and the fiber structure of those fabrics would influent the colour strength. As the concentration of the extraction increased, the colour seen on the fabrics will be more strengthen. The different structures of the fiber in cotton and silk would also vary the depth of shades between

the fabrics even though the same concentration is used. The depth of shades formed on silk is higher than on cotton fabric without manipulating the dyeing condition and the concentration of the extraction (Hasan *et al.*, 2015; Freeman, 2015).

The identical and suitable procedures are needed in order to apply a correct dyeing method on sample materials using natural dye. It is significant to regard on the colour production obtained and the fastness properties by conducting the correct scientific process of producing the selective natural dyes in order to attain various colours from natural dyes (Samanta and Agarwal, 2009)

2.2 The Use of Mangosteen Peel as Natural Dye

Natural dyes are colorants resulting from naturally obtained sources that can be found in flora, fauna, or minerals. Mostly, natural dyes are dyes that derived from specific parts of plant such as roots, leaves, flower and fruits, and other organic sources such as fungi and lichens (Barhanpurkar *et al*., 2015).

Mangosteen peel is part of the fruits that are discarded as a waste. The reddish-purple of mangosteen peel have an attractive colour that leaves stains when touch and might produce good natural dyes for fabrics dyeing (Batisah, 2015).

Peel is the only part of the mangosteen being extracted in the process of obtaining natural dye from the fruit. There is a red pigment that highly contained in the unwanted mangosteen peel. The pigments extracted from mangosteen peel have a high potential to be used as a natural dye. The *anthocyanin* pigments give the red-purple colour range to the different parts of plants such as fruits and leaves, which is then can be processed as a natural dye, an alternative of synthetic dyes (Chang and Lo, 2010; Sudarmi, *et al*., 2015). This pigment that gives striking red colour extraction would beneficially help to decrease potential health problems, lowering

the toxicity levels and evading reactions of allergens (Kimler *et al.*, 1971; Chairat *et al*., 2007).

Temperature is one of the significance factors to be considered in extracting this pigment. Higher temperature during the extracting process would lose the stability even though this pigment is potentially extracted after drying and being exposed to elevated heat. An optimal temperature is needed in order to give the best and effective spread of *anthocyanin* pigments in the extract. The lower or higher temperature than the optimal one during heat exposure would reduce the content of *anthocyanin* extraction (Strack *et al*., 2003; Stintzing and Carle, 2004; Cai *et al.*, 2005; Herbach *et al*., 2004; Harivaindaran *et al*., 2008).

Many studies have discovered that the extract of mangosteen peel could stop and prevent some bacterial activities from causing inflammatory, diseases and infections. It could also avoid cancer cell spreading and antioxidant possessions (Gopalakrishnan *et al*., 1980; Farnsworth and Bunyapraphatsara, 1992; Williams *et al*., 1995).

Those unwanted peel from discarded fruit wastes can be reprocessed and use again as to earn its significant ranking in our fabric industry (Basitah, 2015). Other than that, it has been initiatively used as a natural source for cotton and silk dyeing in certain part of Thailand because of this non-toxic characteristic and it is also proved to be safely applied on the fabric materials (Chairat *et al*., 2007).

However, the wash and light fastness properties of dye extract from the discarded parts of fruits are needed to be enhanced since they contribute poor fastness properties on fabrics (Chairat *et al*., 2007).

2.3 Mordanting Methods In Natural Dye

Mordanting is a treatment process for dyed textile fabric. This treatment is applied by adding metallic salts or other complex forming agents with mordantable natural dyes onto the textile fabrics. The added metallic salts or complex forming agents are known as mordant. Primarily, the additional of mordant in the treatment process is to bind the natural dyes with textile fabrics (Samanta and Agarwal, 2009).

According to Siva (2007), Jothi (2008) and Samanta and Agarwal (2009), most natural dyes required a mordant to be stable onto the fabric. The commonly used mordant such as copper sulphate, provide an attraction for the dye pigment and the fabric as the precipitation formed insolubly with the dye pigment in the fabric. There were studies about the effect of copper sulphate on silk and cotton. It was found that its application could influent the colour fastness properties as it showed excellent fastness properties on those fabrics (Mahale *et al*., 2003; Dayal *et al*., 2006).

Copper sulphate or also recognized as blue vitriol has high water solubility, thus allow it to be easily applied as a mordant in natural dyeing process to improve the colour fastness. Its use in natural dyeing method helps in giving unique properties for the hues obtained, which could not be attained before. The depth of the shades appeared on silk fabric is also better with copper sulphate as the mordant. The metallic salt applied with natural dye would produce high depth of colour on the same type of silk material (Maulik and Pal, 2005; Samanta and Konar, 2011).

Various kinds of mordants applied would yield various colours, even though the same source of natural dye is used. As for that reason, the attained colour on the fabric is dependent on the types of mordants used, apart from the colour produced by the natural sources itself (Samanta and Konar, 2011).

Devi *et al.*, (2006), Shrivastava *et al.*, (2006) and Radhika *et al.*, (2007), reported that the mordanting method enhanced the emergent of various colour hues and implement an association to the dyeing substances to stay permanent on fiber. Mordanting in natural dyes could influent the colour fastness behaviour by enhancing the resistance to the exposure of light, temperature and washing (Deshpande and Chaturvedi, 2012).

Different methods of mordant or different mordanting methods combinations can improve the colour shades of natural dyes on textile. The mordanting method with natural dye on fabric can be either applied by pre-mordanting, simultaneously mordanting or post mordanting. The selective methods in mordanting would give different result on the shades of colours. It helps to enhance the fastness properties of any natural dye once applied on fabric (Samanta and Agarwal, 2009).

According to Deshpande and Chaturvedi (2012), a good shades will form on silk and cotton cloth with the application of simultaneous mordanting method. The simultaneous mordanting and dyeing method are progressed by immersing the prepared bath solution of dye with the fabric sample material accordingly with certain quantity of mordant to be used. This is because the simultaneous mordanting helps to maximize the production of colour stained on the fabric sample. Thus, the presence of a mordant in natural dye is more important than the dye itself (Samanta and Konar, 2011).

2.4 Dyeing of Fabric

For fabric dyeing, the cloth is generally boiled with an aqueous extracted solution of specific natural dye until all the colouring matter absorbed by the cloth. The dye required minimum time of 1 hour to dye cloths with effective colour. As

natures fibres are different from each other, different extract formula gives two different shades on both fibres (Despande and Chaturvedi, 2012).

In the dyeing process, the tinctorial strength or the dyeing, colouring, or staining properties also needs to be considered. The tinctorial strength is including the weight of fiber materials and the depth of fiber needed to produce a colour. Importantly, the property of colour of a solvent varies associated with the application for which it is intended and the amount of colour that can be tolerated being dependent on the colour characteristics of the material in which it is used. (Jothi, 2008, Samanta and Kumar, 2011; Kechi *et al*., 2013).

The optimum dyeing technique for dyeing fiber by determining dye material concentration, extraction time, dyeing time, concentration of mordant, as well as many other factors (Jothi, 2008).

The standardization and optimization of dyeing conditions are essential for effectively colouring any textile in a particular shade in techno-economic way to produce maximum colour yield (Samanta and Kumar, 2011).

2.5 Fastness Of Natural Dye

The colour fastness in textile dyeing refers to the resistance of a sample to transfer any of its colour appearances or degree of changes of its stains to adjacent white samples. The behaviour of the colour fastness of a textile is generally could be graded by changes of colour depth in original sample. It could also be rated by the scales of discolouration tested the original material. Specifically, it is associated when white sample get coloured from its original fabric colour. Within every kinds of colour fastness of dyed fabrics, the light fastness and wash fastness are significant for any type of textile (Samanta and Agarwal, 2009).

According to Crews (1982) and Balankina *et al.*, (2006), both natural dyes' wash and light fastness rates were strongly influenced by the factor of nature, type and the concentration of mordant used. The colour fastness behaviour of natural dye affected via complex with mordant, as it makes colour fast by insolubilizing the dye. An application of natural dyes on textiles could be faded or paled with the additional of mordant because it is significantly important in fading colours on fabrics. These two fastness test of dye on textile are tested accordingly to International Organization for Standardization (ISO) standard method as shown in Table 2.1.

Table 2.1: ISO standard method for colour fastness test

ISO method	Colour fastness test
MS ISO 105-B02 (1988)	Colour fastness to light
MS ISO 105-C01 (1989)	Colour fastness to wash

It has been considered that the colour fastness properties of natural dyes are not simply be determined by chemical nature with just the sort of colorants. It is also be influenced by chemical nature with the sort of mordant applied on the sample fiber. Thus, it is prominent for the dye handler to concern on using the best combinations of fibre-mordant to obtain less colour change at the end of the fastness test done (Agarwal, 2009).

2.5.1 Wash Fastness

The degree of distribution of dye and its state inside the textile material would strongly influent the natural dyes wash fastness. Particularly, the wash fastness is dependent on the affinity to aggregate the dye molecule inside the fabric (Jothi, 2008). It is foreseeable that the method of applying mordant is purposely to

enhance not only the depth of colour but also the wash fastness of the dyed samples (Kechi *et al*., 2013).

In this study, the wash fastness of the fabric samples dyed without and with mordant was tested accordingly to the MS ISO 105-C01 (1989) methods. This standard method of wash fastness on fabric indicates the staining of adjacent fabric sample and the change in colour by standard grey scale grading (Adeel *et al*., 2009; Samanta and Agarwal, 2009).

The standard grey scale is apportioned into two parts of scaling the wash fastness of a dyed sample. The sample will be graded into the grey scale for colour change and the grey scale for colour staining for different indications of the wash fastness. However, the minimum grade obtained from the grey scale colour change is the same minimum grade colour staining of the grey scale for the commercial scale of the dye extract on textile (Kothari, 1999; Adeel *et al*., 2009; Samanta and Agarwal, 2009).

Grey scale for colour change as shown in Figure 2.1 shows the scale used as the fading or alteration of colour occur on a dyed fabric. It relates to the amount of fading or colour alteration with environmental exposure or washing. In grey scale for colour change, the amount of contrast between the treated and untreated fabric is related to one of the standard pairs to yield the grey scale rating. On this grey scale, 5 indicate that almost no colour was lost, and 1 indicates that most of the colour was lost (Trotman, 1984).

Meanwhile, the amount of staining of adjacent materials that occurs with washing of a specimen as shown in Figure 2.2 is indicated by the grey scale for staining. In grey scale for colour staining, the transference of colour from the tested sample fabric to an adjacent fabric is evaluated in a manner very similar to that of

grey scale for colour change. Similarly, the standard is according to 5 standards but one half of each standard is white, and the second half ranges from white or no staining to a grey scale with the Chroma value of the test specimen that great staining. A value of 5 corresponds to virtually no staining occurred by the washing test, whereas 1 indicates poor colour fastness of washing (Trotman, 1984).

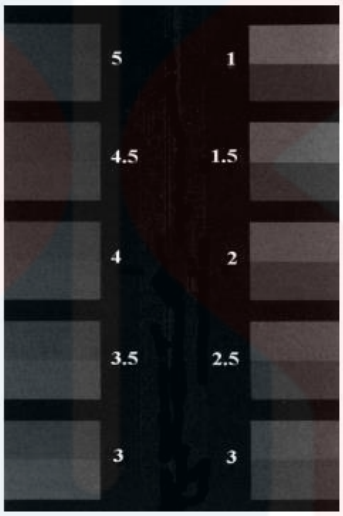

Figure 2.1: Grey Scale for colour change

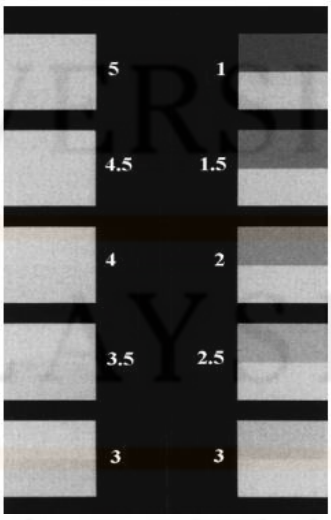

Figure 2.2: Grey scale for colour staining

Both scales for colour change and staining are based on relative small differences between a product standard and lot of samples of any colour in comparison to these two grey scales. The dye extracts with fastness grade of more than 3 can be used as a source of natural dye for dyeing of fabric on commercial scale from the ranging scale of 1 to 5 (Kothari, 1999).

2.5.2 Light Fastness

Agarwal (2009) reported in her study on the 'Application of natural dyes on textiles' that the light fastness of natural dyes were majorly poor than that of artificial dyes. Thus, the colours of artificial dye appeared in the structure of textile were regularly dissimilar from the nature colours as the artificial dyes represented the full range of light fastness properties from poor to excellent.

Table 2.2: Grade and degree of fading of light fastness test

Grade	Degree of Fading	Light Fastness Type
8	No fading	Outstanding
7	Very slight fading	Excellent
6	Slight fading	Very good
5	Moderate fading	Good
4	Appreciable fading	Moderate
3	Significant fading	Fair
2	Extensive fading	Poor
1	Very extensive fading	Very poor

(Kiron, 2014)

In day, sunlight falls on the fabric surface. So it needs to know how much protection ability a fabric was to sun light. It is determined by an experiment called colour fastness to light. To measure the colour fastness, a blue scale is used. After

completing the test, sample is compared with the blue scale and the grades of the colour fastness to light for every fabric materials are scaled as shown in Table 2.2 (Kiron, 2014).

The performance of light fastness of a dye extract on textile could be influenced by external factors. The external factors such as the source and the intensity of light, temperature and humidity as well as the atmospheric pollution can affect the light fastness presentation. Within the optimal conditions of light exposure with optimal temperature and humidity, the rate of fading of dyed fabric materials was found as a drop in relative from 65 to 45% of humidity had very little effect. However, there is a significant reduction of fading as caused by a further decrease up to 25% humidity (Egerton and Morgan, 1970).

On the other hand, the presence of substance such as starch and gums might accelerate the fading process (Gupta, 1999). Cumming *et al*. (1956) in 'A Study of the Photochemistry of Dyes on Proteins and other Substrates' attributed that the fading on cellulose to an oxidative process, whereas on protein fibers the process had a reductive nature.

Besides, several internal factors may influence the light fastness performance of a dye extract on textile. The internal factors that could be manipulated in this test are the chemical and the physical state of dye, the dye concentration, the nature of the fibers, and the mordant type and concentration (Cristae and Vilarem, 2006). Light fastness of dyed textiles is related to the chemical structure and physical characteristics of the fiber itself. Mainly, the light fastness of a dyed fiber usually increases with increasing dye concentration. However, Sahoo *et al*. (2012) proved that the copper sulphate of 1% to 3% concentration as a mordant to silk fabric did not

affect the colour fastness to light as the grades were remained the same for all concentration tested.

According to Gantz and Summer (1957) and Sahoo *et al*. (2012), the fabric sample of silk showed poor fastness without mordant application. It showed that the silk material gives fairly good colour fastness after being exposed to light exposure. The amount of fading is then measured by comparison to the original colour and a rating between 0 and 8 is awarded. Zero denotes extremely poor colour fastness whilst a rating of 8 is deemed not to have altered from the original. Most apparel will have a light fastness of 4. The higher the rating, the better the fabric will perform to daylight. The textile yarns must have a minimum of 3/4 light fastness according to the standard blue scale as shown in Figure 2.3.

Figure 2.3: Standard blue scale for light fastness

2.6 Toxicity In Dye

By the term of toxicity, there are probabilities of any specific compound or molecules to cause negative consequences to the society. Toxicity of dye is

associated with the provision of evidence in the toxicity statistics of the dye (Zippel, 2004; Joshi and Purwar, 2004).

The assessment of the toxicity of textile dyes is very important. The toxicity test is prominent due to the different effects that they caused to the environment and the organisms exposed to them. The biological activities also differed greatly between the dyes (Marechal *et al*., 1997).

The test of toxicity focused on the effects of irritation to skin and eye, as well as the potential of sensitization, which is affected within a short period of time. Apart from that, toxicity test would also be potentially tested for the long term consequences, including the toxicity effects of carcinogenic, mutagenic or reproductive, for natural dye or any other materials before they are being used (Zippel, 2004; Joshi and Purwar, 2004).

The textile sector is liable for the large volumes of residues with high organic load and strong colouration. This appears as the main environmental issues caused by the textile industry (Ekici *et al.*, 2001). The inefficiencies in the colouring during textile dyeing process using synthetic dyes could cause an effluent of enormous amount of residues, which then directly released into the water bodies. The release of this residue will contaminate the environment, especially aquatic lives. This consequence occurred because of the impurities of the removal of the crude material and residual chemical reagents used in such processes (Correia *et al*., 1994).

Natural dyes are not causing bad consequences to human health and ecological system (Onal, 1996; Siva, 2007; Adeel, *et al.*, 2009). The application of mordant onto fiber sample in natural dyeing process in order to advance the colour fastness properties is fortunately proved as non-allergic which will not cause

infections to human skin as it is scientifically evidenced by Deshpande and Chaturvedi (2012).

The dyed fabrics with mordant are friendly to ecological and naturally biodegradable. Hence, the presence of metallic salts in natural mordantable dyes could evade potential risk of having skin infections as compared to the used of purely synthetic dyes in industry. It is generally known that the lethal concentration of 50% is the best toxicity assessment (Khan *et al*., 2003).

2.6.1 Lethal Concentration 50%

Lethal concentration of 50% or LC_{50} emphasizes the lethal concentration of the animal tested for 50% where it is considered the quantity of substances that executes half of the tested animals in the form of concentration. The LC_{50} is a way to measure the short-term poisoning potential or the acute toxicity of a material (Khan *et al*., 2003).

LC_{50} gives measure of the immediate or acute toxicity of a chemical. Acute toxicity describes the adverse effects of a substance that result either from a single exposure or from multiple exposures in a short period of time that usually less than 24 hours (Goldstein *et al*., 1968).

LC_{50} determination is a simple preliminary assessment of the toxicity of a compound by determination of the median lethal concentration which is capable of killing 50% animals under stated conditions (Goldstein *et al*., 1968). In terms of signs or symptoms, LC_{50} is redefined as the concentration effective for producing a certain sign in 50% of the experimental animals (William *et al*., 1976).

The lethality was calculated from the mean survival of animal tested in the extracts and that of control. Mean percentage mortality are plotted against the

logarithm of concentrations. The concentration that killing 50% of the animal tested is calculated from the linear equation by taking the antilogarithm (Naidu, *et al*., 2014)

The assessment of exposure can be performed by measuring the concentration of a substance administered to a particular organism (Oga, 2003). The study of concentration-response or concentration-effect in toxicology is essential and is used to determine the LC_{50} of drugs and other chemicals (Goodman and Gilman, 2007).

LC_{50} tests measure the susceptibility and survival potential of organisms during exposure to particular toxic substances. Pollutants with higher LC_{50} values are less toxic for organisms because greater concentrations are required to result in mortality (Sparling *et al*., 1997).

The toxicity of plant extracts expressed as LC_{50} values is commonly classified either by comparison to Meyer's toxicity index as listed in Table 2.3 and 2.4 (Meyer *et al*., 1982; Clarkson *et al*. 2004).

Table 2.3: Clarkson's toxicity criterion for the toxicity assessment of plant extracts

LC_{50} (μg/mL)	Toxicity level
Above 1000	Non-toxic
500-1000	Low toxic
100-500	Medium toxic
0-100	Highly toxic

(Clarkson *et al*. 2004)

Table 2.4: Meyer's toxicity index

LC_{50} (μg/mL)	Toxicity level
$LC_{50} < 1000$	Toxic
$LC_{50} > 1000$	Non-toxic

(Meyer *et al*., 1982)

2.6.2 Brine Shrimp Lethality Assay

Brine shrimp lethality bioassay (BSLA) is considered as a useful tool for preliminary assessment of toxicity. It is a convenient system used for monitoring the biological activities of various plant species. The method is attractive as it is very simple, inexpensive and sensitive (Krishnaraj *et al*., 2005; Gadir, 2012; Naidu *et al.,* 2014). It is used as an indicator for general toxicity and also as a guide for detection of antitumor and pesticidal compounds (Meyer *et al*., 1982). This assay has been noted as a very useful tool for the isolation of bioactive compounds from plants extracts (Sam, 1993).

The ability of the *Artemia salina* to produce dormant eggs, known as cysts, has led to extensive use of *Artemia salina* in aquaculture. The cysts may be stored for long periods and hatched on demand. Nauplii of the brine shrimp *Artemia salina* constitute the most widely used food item, and over 2000 tonnes of dry *Artemia salina* cysts are marketed worldwide annually. In addition, the resilience of *Artemia salina* makes them ideal animals for running biological toxicity assays and it has become a model organism used to test the toxicity of chemicals. It is normally found in brackish water and is a very hearty little organism able to tolerate high salt concentrations (Sorgeloos *et al*., 2001).

Artemia salina found favour as a model organism for use in toxicological assays, despite the recognition that it is too robust an organism to be a sensitive indicator species. In pollution research, the brine shrimp has had extensive use as a test organism and in some circumstances is an acceptable alternative to the toxicity testing of mammals in the laboratory. The fact that millions of brine shrimp are so easily reared has been an important help in assessing the effects of a

large number of environmental pollutants on the shrimps under well controlled experimental conditions (Lewan *et al*., 1992).

The process of toxicity test could be extended to maximum of 60 hours of monitoring the brine shrimp amount. However, most data of the tests done are relevantly for the LC_{50} calculations obtained after 24 hours of exposure to the samples (Vanhaecke *et al*., 1981; Meyer *et al*., 1982; Vanhaecke and Persoone, 1984; McLaughlin *et al*., 1998).

CHAPTER 3

MATERIALS AND METHODS

3.1 Materials

600g of mangosteen peel was used with; distilled water, 1 g of copper sulphate, 450 of living shrimps, 2 pieces of 21 cm x 27.9 cm of each white cotton and silk materials, muslin cloth, a 500 mL round bottom flask, a 50 mL measuring cylinder, a 5 mL measuring cylinder, a micropipette, rotary evaporator, hot plate, weighing scale, beaker, 10 petri dishes, a Xenotest220 machine, a Linitest machine, a stopwatch.

3.2 Methods

3.2.1 Extraction

The sample of mangosteen peel was collected and washed thoroughly with water to remove the impurities. This sample was tossed to prevent the excessive moisture in the extract and was ready to be extracted. 150 g of mangosteen peel was weighed and taken into a 500 mL round bottom flask. The same round bottom flask was added with 200 mL of distilled water and connected to a rotary evaporator. The round bottom flask was immersed into the water bath as the temperature for water bath was set to 80°C. The rotary evaporator was started to rotate the round bottom flask for an hour. The colour from mangosteen peel was started to extracted. After 1 hour of heating, the round bottom flask was taken out from the water bath and disconnected from the rotary evaporator. Both mangosteen peel residues and the extracted juice were allowed for cooling process at room temperature. The residues

and dye extract were then separated by filtering them using muslin cloth into a 500 mL beaker. The volume of the extract obtained was measured by using a 250 mL measuring cylinder. The dye extract was kept cold at 4°C for further use. The extraction was repeated until all the 600 g of mangosteen peel extracted. The dye extract was ready to be mordanted using simultaneous mordanting method (Deshpande and Chaturdevi, 2012; Kannadasan, *et al*., 2013).

3.2.2 Mordanting

Cotton and silk sample fabrics were obtained. Two samples size of cotton materials were cut into 21 cm x 29.7 cm. Each of the cotton samples was accurately weighed using digital weighing scale. The dye extract solution was mixed with 5% of copper sulphate mordant to prepare a mixture of dye with mordant.

A 5% copper sulphate mordant solution was prepared by adding 1 g of copper sulphate powder into 20 mL of distilled water. The 2% based on weight fabric (o.w.f) prepared mordant was mixed into dye extract solution to make a liquor ratio 1:40. The mixture was heated at 100°C for 10 minutes and was ready for toxicity test and dyeing process (Jothi, 2008; Pruthi *et al*., 2008).

3.2.3 Toxicity Test

The hatched brine shrimp (*Artemia salina*) were put in 33.4 g/L artificial sea water prepared using commercial sea salt. The 33.4 g/L artificial sea water was prepared by mixing 33.4 g of commercial sea salt with 1 L of distilled water. A lamp was placed above the open side of the tank to attract the hatched shrimps closed to the tank wall. After 24 hours, the shrimps matured as nauplii (*Artemia salina*) and were ready for the assay (Sasidharan *et al*., 2008).

Lethal Concentration 50% (LC_{50}) for toxicity test of prepared mangosteen peel dye extract without mordant and the dye extract with copper sulphate mordant was conducted by using matured living shrimps. 450 of matured living shrimps were obtained. 10 matured living shrimps were placed into a control 2mL of artificial sea water. The other 50 matured living shrimps were equally placed into 5 petri dishes. Each of five petri dish were filled with 2mL of total volume dye extract and artificial sea water with 7 different concentrations of 1000, 500, 250, 125, 62.5, 31.25 and 15.625 µg/mL dye extract without mordant by serial dilution method. The same steps for LC_{50} were repeated by replacing the dye sample without mordant with the dye sample with copper sulphate mordant. The number of matured living shrimp was observed after 24 hours of exposure and the number of died brine shrimps are recorded into table of data. This step was triplicated. The value of LC_{50} was calculated by using the equation obtained from the graph of log concentration versus percentage of mortality of brine shrimps (Meyer *et al*., 1982; Klemola *et al*., 2006; Mshelia *et al*., 2016).

3.2.4 Dyeing

A weighed cotton sample was immersed into the dye bath solution without copper sulphate mordant with material to liquor ratio 1:40 and heated at 100°C for an hour. Then, the other weighed cotton materials was immersed in a dye bath solution with the presence of mordant copper sulphate with the same material to liquor ratio and let for an hour at 100°C. After 1 hour, both samples were taken out from the dye extract solution. The samples were dried in the laboratory by air circulation. The same procedures were applied on the sample silk material (Samanta and Kumar, 2011; Sahoo *et al*., 2012; Hecker, 2014).

3.2.5 Fastness Tests

(a) Wash Fastness

The wash fastness test was carried out by soaking all of the dyed fiber of cotton and silk using 100 mL of washing detergent. The colour fastness to washing of the samples was determined by using washing fastness machine (Linitest). The wash fastness rating was assessed using grey scale as per ISO-105-C01 to test the loss of colour shade or depth. 4 cm × 10 cm of cotton samples were stitched between a same size of white wool and cotton fabrics. The same size as cotton samples of silk fibers were stitched between a same size of wool and silk fabrics. All the four samples of dyed fabrics were soaked inside the washing pots and were inserted inside the pot holes. The machine was operated and run for 30 minutes. The washed samples were removed and rinsed under running distilled water and squeeze to remove excess water on it and will be shade dried, before ready for grading by using standard grey scale (Adeel *et al*., 2009; Samanta and Agarwal, 2009; Umar, 2013).

(b) Light Fastness

The colour fastness to light of the samples was determined as per MS ISO 105-B02 method. 2 cm × 1 cm of sample materials were stapled to a prepared card plate each and were assembled into the light fastness machine plates. Light fastness was analysed by exposing the dyed materials to Ultraviolet light (UVL) in a Xenotest220 machine, for 24 hours. The light fastness ratings were assessed using standard blue scale to grading the colour change (Adeel *et al*., 2009).

CHAPTER 4

RESULTS AND DISCUSSIONS

4.1 Extraction of Mangosteen Peel

The extracted mangosteen peel was controlled under a fixed temperature of 80°C of water bath for 60 minutes. According to Hasan *et al*. (2015), these controlled variables were an optimum temperature and time in order to extract the best yield of natural dye produced by mangosteen peel. The extraction of mangosteen peel produced was 656.0 mL of dye extract from 150 g of discarded, weighed and cleaned mangosteen peel as shown in Table 4.1. The peel was extracted for four times separately.

Mangosteen peel dye extract was produced from the extraction method with 200 mL of distilled water using rotary evaporator for 60 minutes each process. The extraction process produced reddish-purple colour of dye extract liquid and this extract was later applied onto cotton and silk fabrics for further experimentation.

Table 4.1: The production of mangoesteen peel dye by using extraction method

	Colour observed	Volume (mL)
Extraction	Reddish-purple	656.0

4.2 Effect of Copper Sulphate ($CuSO_4$) Mordant in Mangosteen Peel Dye Extract on Cotton and Silk Fabric

Copper sulphate was used as a mordant for this study. The mordant was mixed with mangosteen peel dye extract and the varieties of the shades performance

of the dye extract were observed. It was found that the addition of the mordant gave a different appearance functions on cotton and silk fibers as shown in Table 4.2.

Table 4.2: The shades appeared on cotton and silk fabric dyed with mangosteen peel dye extract

	Cotton	Silk
Dye extract without mordant		
Dye extract with $CuSO_4$ mordant		

From the shades observed in Table 4.2, the appearance of hue on cotton fabric with the addition of copper sulphate is darker than without mordant. Through this visual observation, the copper sulphate mordant used has varied the shades performance of mangosteen peel dye extract on cotton fabric by giving the fabric a darker hue.

The images of the silk fabrics in Table 4.2 were also found to have different hues after being dyed with the dye extract with and without mordant. The hue of silk fabric dyed with mangosteen peel dye extract and copper sulphate mordant gave a paler hue than the silk fabric dyed with mangosteen peel dye extract without mordant.

The application of mordant in natural dyes could produce unique and various hues that could not be attained in natural dyes without any addition of mordant

(Maulik and Pal, 2005). According to Devi *et al.*, (2006), Shrivastava *et al.*, (2006) and Radhika *et al.*, (2007), the emergence of various colour hues have been enhanced with the presence of mordant in natural dye extracts and implemented an association to the dyeing substances to stay permanent on fiber.

Based on the visual observation, it could be seen that the present of copper sulphate as mordant in mangosteen peel dye extracts had given various shades on cotton and silk fabrics.

4.3 Toxicity of Mangosteen Peel Dye Extract

In Clarkson's toxicity index, the lower the LC_{50} concentration, the higher the level of toxicity in dye extracts. This is because the LC_{50} values calculated are indicating the concentration to cause 50% of living brine shrimps killed. Meyer's toxicity index stated that the plant extracts with the LC_{50} value of more than 1000 µg/mL are non-toxic, whilst the lower values indicate that the plant extracts are toxic (Meyer *et al*., 1982; Clarkson *et al*., 2004).

Data in Table 4.3 shows that there are no brine shrimps killed in 15.625µg/mL and 31.25 µg/mL of mangosteen dye extract without mordant. 6.67% of brine shrimps were started to die in 62.5 µg/mL of the dye extract without mordant after 24 hours. Whilst, 3.33% of died brine shrimps are counted at the 31.25 µg/mL of dye extract with mordant within the same exposure time interval. The data also shows that the percentage of brine shrimps mortality in their nature of sea water increases accordingly to the increase of dye extract concentrations in both dye extracts with and without mordant.

Table 4.3: Toxicity of mangosteen peel dye extract without and with $CuSO_4$ mordant

	Concentration dye extract (µg/mL)	Mortality (%)	LC50 (µg/mL)	Toxic/ non-toxic
Control	0	0	-	-
Dye without mordant	15.625	0	2512.71	Non-toxic
	31.25	0		
	62.5	6.67		
	125	16.67		
	250	23.33		
	500	30.00		
	1000	40.00		
Dye with mordant ($CuSO_4$)	15.625	0	1807.96	Non-toxic
	31.25	3.33		
	62.5	13.33		
	125	26.67		
	250	30.00		
	500	36.67		
	1000	43.33		

The concentrations of dye extract that could cause half of the matured brine shrimps killed were calculated in terms of LC_{50}. Based on the different concentrations of mangosteen peel dye extract manipulated in the artificial sea water, the numbers of remaining brine shrimps were counted in order to plot a graph of log concentration against the percentage of mortality (Mshelia *et al.*, 2016). A linear equation was formed in the graph plotted using Microsoft Excel. The equation formed was used to calculate the specific value of the LC_{50}. The graphs of log concentration versus percentage of mortality for dye extract with and without mordant were plotted as shown in Figure 4.1 and 4.2. The value of LC_{50} calculated for the concentration that caused 50% of the brine shrimps killed after 24 hours of

exposure were 2512.71µg/mL for dye extract without mordant and 1807.96 µg/mL for dye extract with mordant.

The LC_{50} of the mangosteen peel dye extract without mordant was higher than the dye extract with mordant. This difference was most probably caused by the presence of $CuSO_4$, the chemical used as the mordant (Dalziel, 2012). This result was expected as the addition of chemical substances could influence the mortality of brine shrimp after 24 hours. However, the LC_{50} for mangosteen peel dye extract with mordant was still more than 1000 µg/mL, which according to Meyer's and Clarkson's toxicity index, these tested extracts were classified as non-toxic, harmless and safe to be used commercially as natural dye in textile industry.

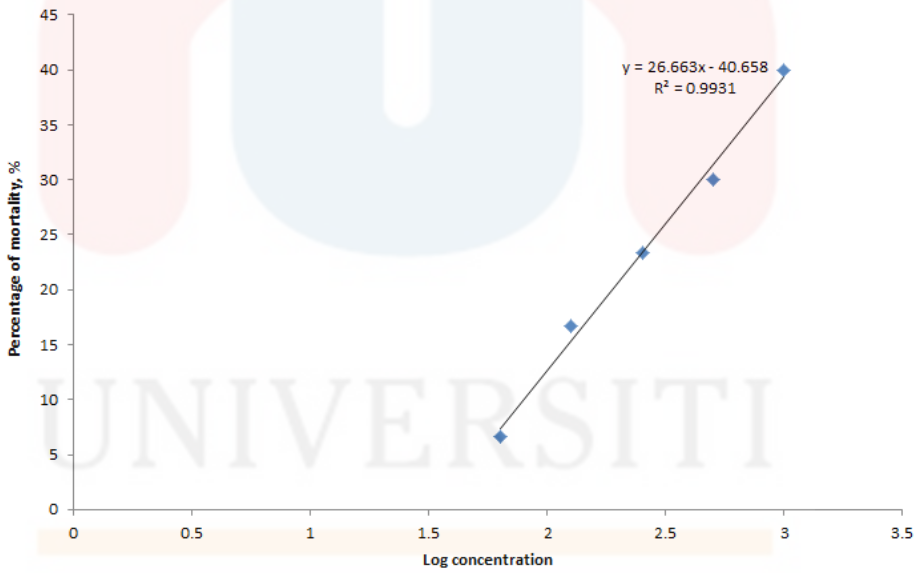

Figure 4.1: Log concentration against percentage of mortality for dye extract without mordant

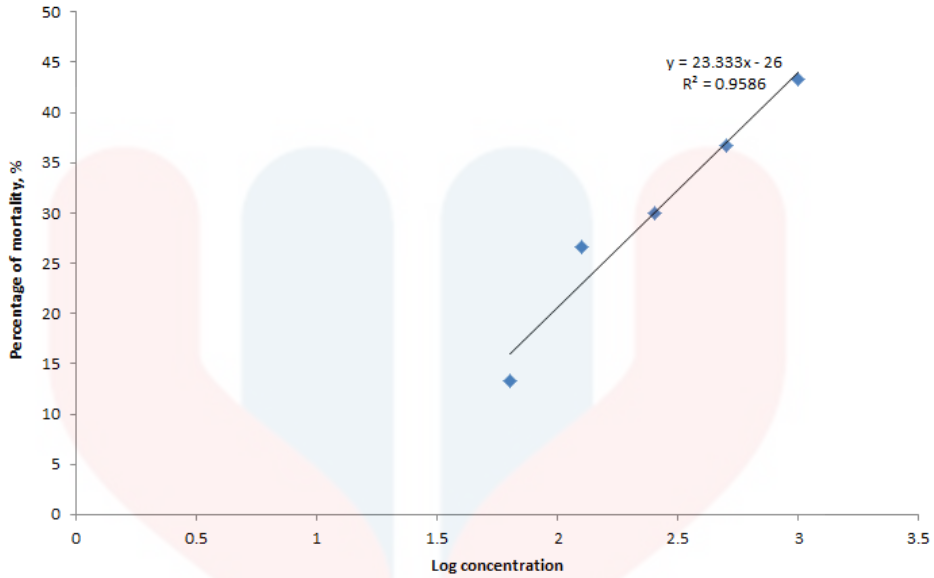

Figure 4.2: Log concentration against percentage of mortality for dye extract with mordant

4.4 Wash Fastness of Mangosteen Peel Dye Extract on Cotton and Silk

According to Kothari (1999) and Kumaresan *et al*. (2011), a dye extract with fastness grade of more than 3 for washing properties had a good potential to be used as a source of natural dye for dyeing of fabric. This commercial grade scales ranged from 1 to 5, as referred to the standard of grey scale as per ISO-05-C01.

Table 4.4 shows the results for the colour change and colour staining of mangosteen peel dye extract with and without mordant according to the stated ISO standard. Regarding to the wash fastness test, the addition or the absence of copper sulphate mordant in mangosteen peel dye extract on cotton and silk fiber shows that the grades of colour change and colour staining of both cotton and silk fabrics dyed were more than 3 which were higher than the minimum requirement scale for the wash fastness.

Table 4.4: Wash fastness of mangosteen peel dye extract on cotton and silk fabric

	Cotton		Silk	
	Colour Change	Colour Staining	Colour Change	Colour Staining
Dye without mordant	4/5	4/5	4	4/5
Dye with mordant	4/5	4/5	3/4	4/5

Simultaneous mordanting method was used in this study where the copper sulphate as the mordant was mixed together with the natural dye prior to the application of the mixture on fabrics. Table 4.4 shows that the same results were obtained for colour change and colour staining on cotton fabric. The grades recorded for dye extract with and without copper sulphate mordant was 4/5 for both colour change and colour staining on cotton fabric. This was expected as the grade for wash fastness properties on cotton fabric dyed with natural dye containing mordant to be higher that without mordant as the role of mordant is to effectively bind the mangosteen peel dye extract to cotton fabric. This result indicates that the use of copper sulphate as a mordant to bind the mangosteen peel dye extract to cotton fabric was less effective than expected.

Differently, the dyeing process with the presence of copper sulphate effected the wash fastness on the silk fabric as the grade for the colour change recorded on mangosteen peel dye extract with and without mordant were 4 and 3/4, respectively. The higher grade of the colour change by using dye extract without mordant on silk fabric compared to the one with mordant might be caused by the type of mordant used, as well as the fabric structure of silk fiber (Mahale *et al*., 2003; Dayal *et al*., 2006; Samanta and Agarwal, 2009). Thus, the chemical structure of mordant used and type of fabrics should be considered in natural dyeing process.

Therefore, based on the results obtained in Table 4.4, the mangosteen peel dye extract has a good potential to be used as a natural dye for cotton and silk.

4.5 Light Fastness of Mangosteen Peel Dye Extract on Cotton and Silk

The ISO 105-B02 standard for blue wool scale stated that the minimum rate for light fastness for a dye sample to be potentially considered as a commercial dye in textile industry is 5. The range for blue wool scale is 1 to 8 (Kumaresan *et al*., 2011).

Table 4.5 shows that mangosteen peel dye extracts with and without mordant applied on cotton fabric give the same grade of light fastness as both were recorded as 4/5. The same scale grades obtained on both dye extracts indicates that the properties or the ratio concentration of copper sulphate used was ineffective to strongly bind the mangosteen peel dye extract on cotton fabric. Similarly, the applications of both mangosteen dye extracts with and without copper sulphate as mordant did not produced a good light fastness grade on silk, as the grade obtained was 3 for dye extract without mordant and 2/3 for dye extract with mordant. This test results also showed that the mangosteen peel dye extract without copper sulphate mordant was better than the light fastness of mangosteen peel dye with copper sulphate mordant when applied on the silk fabric.

Comparing to the ISO 105-B02 standard, the light fastness of both mangosteen peel dye extracts with and without copper sulphate mordant applied onto cotton and silk fabric were lower than the minimum requirement rate for the colour fastness to light.

Table 4.5: Light fastness of mangosteen peel dye extract on cotton and silk

	Cotton	Silk
Dye without mordant	4/5	3
Dye with mordant	4/5	2/3

Different chemical fixations of mordants in natural dye contribute to different functions of fabrics appearance as the mordants bind the natural dye with textile fabrics. Most natural dyes require a mordant to be stable onto the fabric (Siva, 2007; Jothi, 2008; Samanta and Agarwal, 2009). The mordant fixed are aimed either to form dull, different shades, or mainly to improve the light fastness properties. These appearance characteristics are however may change after the dyed fabric being exposed to the sunlight or the UV light in artificial Xenotest220 machine for 24 hours. The changes of appearance gradually change the light fastness properties graded along the blue wool scale. The unsuitability mordant used together with mangosteen peel dye extract could cause the light fastness to be unacceptable to be applied onto cotton and silk fabric. The mordanted fabrics may experience the destruction of dye molecules during the light exposure.

These moderate and poor performances of the mangosteen peel dye extracts dyed on silk fabric material are changed more deeply than to the light fastness properties on cotton fabric. Cotton and silk textile materials contain different fiber structures that would vary the depth of shades of the natural dye extract applied without considering the type and concentration of dye extract used, as well as the method for dyeing used (Hasan *et al.*, 2015; Freeman, 2015). From the data in Table 4.5, the light fastness properties performed on cotton is better than on silk fabric for both mangosteen peel dye extract without mordant and that of with presence of mordant applications.

The problem with the grey scale and the blue wool scale is that both of the scales are subjective test. It depends on an experienced operator in comparing the changes in test swatches versus the reference swatches. Thus, this factor can lead to some variability in grading the light fastness scores. Besides, the significant changes on the blue wool scale are not particularly onerous with respect to outdoor performance of most moulded or fabricated products.

CHAPTER 5

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

Mangosteen peel as the discarded part of the fruit contains deep reddish purple pigment when ripe. When touch, the colour do stains on hands and cloths.

The mangosteen peel extracted with distilled water is non-toxic according to Meyer's and Clarkson's toxicity index. The presence of copper sulphate in the mangosteen peel dye extract as a mordant is also found to be non-toxic according to the same toxicity index mentioned earlier.

The colour fastness to wash test of the mangosteen peel dye extract with and without copper sulphate mordant has a potential to be accepted as a commercial dye extract for cotton and silk fabrics as the grade obtained for this test was comply with the ISO 105-C01 standard.

The colour fastness to light test of the mangosteen peel dye extract with and without copper sulphate mordant on cotton gave a better grade than on silk fabric. The grade obtained for the natural dye application on cotton was very close to the grade accepted for commercial use based on the ISO 105-B02 standard scale.

Mangosteen peel dye extract with mordant is expected to improve the natural dye extract performance when being applied on cotton and silk fabrics. However, the copper sulphate mordant performance as a binder between natural dye and fabrics was found to be moderate based on the results obtained from the tests.

It could be concluded that the mangosteen peel dye extract has a potential to be used as natural dye in textile industry with some improvement on the type of mordant used and other enhancement that will be further discussed in 5.2.

5.2 Recommendations

A further experiment could be conducted regarding the type of mordant used in natural dyeing process. There are many other types of mordant that could be easily found in the market such as alum, potassium dichromate and ferrous sulphate. A test should be run to find the most suitable and most effective mordant to be used with mangosteen peel dye extract.

Other than that, all three different mordanting methods; pre-mordanting, simultaneous mordanting and post mordanting, could also be run to find the best method that give a better result for colour fastness test. This approach is importance to find the most effective mordanting method for copper sulphate mordant in mangosteen dye extract to be used on cotton and silk.

Furthermore, the fiber structures of cotton and silk fabric materials should be identified and studied in order to improve the dyeing process of those fabric materials. Different chemical structures of fiber require different process to bind the fabric with the dye extracts prepared.

REFERENCES

Adeel, S., Ali, S., Bhatti, I. A. & Zsila, F., 2009. Dyeing Of Cotton Fabric Using Pomegranate Punica Granatum) Aqueous Extract. *Asian Journal of Chemistry*, Vol.21 (5): 3493-3499.

Agarwal, K., 2009. Application of natural dyes on textiles, *Indian Journal of Fibre & Textile Research*, Vol.34: 384-399.

Alemaheyu, T. & Teklemariam, Z., 2014. Application of Natural Dyes on Textile: A Review. *International Journal of Research – Granthaalayah*, Vol.2 (2): 61-68.

Balankina, G. G., Vasiliev, V. G., Karpova, E. V. & V. I. Mamatyuk, 2006. *Dyes and Pigments*, Vol.71: 54-60.

Barhanpurkar, S., Bhat, P., Kumar, A. & Purwar, R., 2015. Studies of Banana SAP used as mordant for natural dye, *International Journal on Textile Engineering and Processes*, Vol.1 (4): 56-62.

Basitah, T., 2015. Extraction, Characterization and Application of Natural Dyes from the Fresh Rind of Index Colour 5 Mangosteen (*Garcinia mangostana L.*). *International Journal of Chemical, Molecular, Nuclear, Materials and Metallurgical Engineering*, Vol.9 (7): 871-874.

Cai, Y., Sun, M. & Corke, H., 2005. Characterization and application of betalain pigments from plants of the *Amaranthaceae. Trends in Food Science and Technology*, Vol.16: 370-376.

Chairat, M., Bremner, J. B. & Chantrapromma, K., 2007. Dyeing of cotton and silk yarn with the extracted dye from the fruit hulls of mangosteen, *Garcinia mangostana* Linn. *Journal of Fibers and Polymers*, Vol.8 (6): 613-619.

Chang H. and Lo Y. J. 2010. Pomegranate leaves and mulberry fruit as natural sensitizers for dye-sensitized solar cells. *Solar Energy*, Vol.84:.1833-1837.

Clarkson, C., Maharaj, V. J., Crouch, N. R., Grace, O. M., Pillay, P., Matsabisa, M. G., Bhagwandin, N., Smith, P. J. & Folb, P.I., 2004. In Vitro Antiplasmodial Activity of Medicinal Plants Native to or Naturalized in South Africa. *Journal of Ethnopharmaceutical*, Vol.92: 177-191.

Crews, P. C., 1982. The Influence of Mordant on the Light Fastness of Yellow Natural Dyes. *Journal of American Institute Conservation*, Vol.21: 43-58.

Cristae, D. & Vilarem, G., 2006. Improving Light Fastness of Natural Dyes on Cotton Yarn. *Dyes and Pigments*, Vol.71 : 39-46.

Correia, V. M., Stephenson, T. & Judd, S. J., 1994. Characterization of textiles wastewaters – A review. *Environmental Technology*, Vol.15: 917-929.

Cumming, J. W., Giles, C. H. & McEachran, A. E., 1956. A Study of the Photochemistry of Dyes on Proteins and other Substrates. *Journal of the Society of Dyers and Colourists*, Vol.72: 373-453.

Dalziel, C. J., 2012. Joybilee Farm: Are natural dyes safe? Retrieved from http://joybileefarm.com/are-natural-dyes-safe-clearing-up-the-confusion-about-chemical-dyes-and-metal-mordants/

Dayal, R., Dhobal, P. C., Kumar, R., Onial, P. & Rawat, R. D., 2006. Natural Dye from *Parthenium hysterophorus*. *Colourage*, Vol.52 (12): 53.

Deshpande, R. & Chaturvedi, A., 2012. Colourant Potential Of *Ricinus Communis* L. On Silk And Cotton Cloth. *Science Research Reporter,* Vol.2 (3): 302-306.

Devi, A. S., Sumathy, B. S. & Katyayini, V. K. L. T., 2006. Trailing *Eclipta* – A Natural Green Colour for Textiles, *Natural Dyes Scope and Challenges* (ed. M. Daniel) Scientific Publication (India) Jodhpur, 103-112.

Devi, S. & Karuppan, P., 2015, Reddish brown pigments from *Alternaria alternata* for textile dyeing and printing. *Indian Journal of Fibre & Textile Research*, Vol.40: 315-319.

Egerton, G. S. & Morgan, A. G., 1970. The Photochemistry of Dyes. II. Some Aspects of the Fading Process. *Journal of the Society of Dyers and Colourists*, Vol.86: 242-251.

Ekici, P., Leupol, G. & Parlar, H., 2001. Degradability of selected azo dye metabolites in activated sludge systems. *Chemosphere*, Vol.44: 721-728.

Farnsworth, N. R. & Bunyapraphatsara, N., 1992. Thai Medicinal Plants: Recommended for Primary Health Care System. *Prancheon Company, Bangkok.*

Freeman, H. S., 2015. Textile Chemistry Development. *Textile Engineering, Chemistry and Science.*

Gadir, S. A., 2012. Assessment of Bioactivity of some Sundanese Medicinal Plants Using Brine Shrimp (Artemia salina) Lethality Assay. *Journal of Chemical and Pharmaceutical Research*, Vol.4: 5145-5148.

Gantz, G. M. & Summer, W. G., 1957. Stable Ultraviolet light Absorbers. *Textile Research Journal*, Vol.27: 244-295.

Goldstein, A., Anonov, I. & Kalman, S., 1968. *Principle of Drug Action: The Basis of Pharmacology*. New York: John Wiley and Sons.

Goodman, L. and Gilman, J. S., 2007. *As bases farmacologicas da terapeutica. 11th ed.* Rio de Janeiro: McGraw-Hill, 607-629.

Gopalakrishnan, C., Shankaranarayanan, D., Kameswara, L. & Nazimudern, S. K., 1980. *Indian Journal of Experimental Biology*. Vol.18: 843.

Gupta, D., 1999. Fastness Properties of Natural Dyes Part 1: Introduction and Review of Literature. *Colourage*, Vol.46 (7): 35-43.

Harivaindaran, K. V., Rebecca, O. P. S. & Chandran, S., 2008. Study of Optimal Temperature, pH and Stability of Dragon Fruit (*Hylocereus polyrhizus*) Peel for Use as Potential Natural Colorant. *Pakistan Journal of Biological Sciences*, Vol.11 (18): 2259-2263.

Hasan, M. M., Nayem, K. A., Azim, A. Y. M. A. & Ghosh, N. C., 2015. Application of Purified Lawsone as Natural Dye on Cotton and Silk Fabric. *Journal of Textiles*, 1-7.

Hecker, S., 2014. Extraction of B-carotene from orange peel and carrot waste for cotton dyeing. *Thesis for the Degree of Master in Science with a major in Textile Engineering The Swedish School of Textiles*, Vol.18.

Herbach, K. M., Stintzing, F. C. & Carle, R., 2004. Thermal degradation of betacyanins in juices from purple pitaya (Hylocereus polyrhizus [Weber] Britton and Rose) monitored by high-performance liquid chromatography-tandem mass spectrometric analyses. *European Food Research and Technol*ogy, Vol.219: 377-385.

Jothi, D., 2008. Extraction of Natural Dyes From African Marigold Flower (*Tagetes Ereecta* L) For Textile Coloration, *AUTEX Research Journal*, Vol.8: 49.

Joshi, M. & Purwar, R., 2004. A review of developments in new processes for colour removal from textile effluents. *Review of Progress in Colouration*, Vol.34: 57-58.

Kannadasan, T., Gnanaprakasam, A., Harikrishnan, R., Nganyeikonyak & Rajkumar, A., 2013. Extraction of Natural Dye (*Xanthopyll*) from Marigold Flower. *Journal of Advanced Scientific Research*. Vol.4 (2): 48-50.

Kechi, A., Chavan, R. B. & Moeckel, R., 2013. Dye Yield, Colour Strength and Dyeing Properties of Natural Dyes Extracted from Ethiopian Dye Plants. *Textiles and Light Industrial Science and Technology*, Vol.2 (3): 137-145.

Khan, M. R., Omoloso, A. D. & Kihara, M., 2002. Antimicrobial activity of *Michelia champaca*. *Fitoterapia*, Vol.74 (5): 501.

Kimler, L., Larson, R. A., Messenger, L., Moore, J. B. & Mabry, T. J., 1971. Betalanic acid , a new naturally occurring pigment. *Journal of Chemical Society*, Vol.21: 1329-1330.

Kiron, M. I., 2014. Textile Learner: Colour Fastness Test to Light, Light Fastness Grades. Retrieved from http://textilelearner.blogspot.my/2012/05/color-fastness-test-to-light-light.html

Klemola, K., Honkalampi-Hamalainen, U., Liesivuori, J., Pearson, J. & Lindstrom-Seppa, P., 2006. Evaluating the Toxicity of Reactive Dyes and Fabrics with the Spermatozoa Motility Inhibition Test. *Association of Universities for Textiles Research Journal*. Vol. 6 (3): 182-190.

Kothari, V. K., 1999. *Progress in Textiles: Science and Technology: Testing and Quality Management, 1st ed*. New Delhi: IAFL, 413-423.

Krishnaraj, A. V., Rao, T. V. N., Sundaraju, D., Vanisree, M., Tsay, H. S. & Subbaraju, G. V., 2005. Assessment of Bioactivity of Indian Medicinal Plants Using Brine Shrimp (*Artemia salina*) Lethality Assay. *International Journal of Applied Science and Engineering Research*, Vol.3: 125-134.

Kumaresan, M., Alanisamy, P. N. & Kumar, P. E., 2011. Dyeing of Cotton and Silk using flower of *Spathodea campanulata*: Effects of mordanting and colour fastness properties. *Journal of Basic and Applied Scientific Research*, Vol.1 (8): 933-941.

Lewan, L., Anderrson, M. & Morales-Gomez, P., 1992. The Use of *Artemia Salina* in Toxicity Testing. *Alternative to Laboratory Animals*, Vol.20: 297-301.

Maiaugree, W., Lowpa, S., Towannang, M., Rutphonsan, P., Tangtrakarn, A., Pimanpang, S., Maiaugree, P., Ratchapolthavisin, N., Sang-aroon, W., Jarernboon, W. & Amornkitbamrung, V., 2015. A dye sensitized solar cell using natural counter electrode and natural dye derived from mangosteen peel waste. *Scientific Report*, Vol.5: 1-12.

Mahale, G., Sakshi & Sunanda, R. K., 2003. *Indian Journal Fibre and Textile Research*, Vol.28 3): 86.

Marechal, A. M, Slokar, Y. M. & Taufer, T., 1997. Decolouration of Chlorotriazine Reactive Azo Dyes with H_2O_2. *Dyes Pigments*, Vol.33: 281-298.

Maulik, S. R. & Pal, P., 2005. *Man-made Text India*, Vol.48 (1): 19.

McLaughlin, J. L., Rogers, L. L. & Anderson, J. E., 1998. The use of Biological assays to evaluate botanicals. *Drug Information Journal*, Vol.32: 513-524.

Meyer, B. N., Ferrigni, N. R., Putnam, J. E., Jacobsen, L. B., Nichols, D. E. & McLaughin, J. L., 1982. Brine Shrimp: A convenient general bioassay for active plant constituents. *Planta Medica*, Vol.45: 31-34.

Mshelia, E. H., Adamu, H. M., Abayeh, O. J., Maigari, A. U. & Watirahyel, E. M., 2016. The Quantitative Phytochemicals Content of the Root Bark of Grewia Mollis, The Antioxidant and Cytotoxicity Activities of Its Extracts. *European Journal of Pure and Applied Chemistry*, Vol.3 (2): 1-11.

Naidu, J. R.., Ismail, R. & Sasidharan, S., 2014. Acute Oral Toxicity and Brine Shrimp Lethality of Methanol Extract of *Mentha spicata* L. (Lamiaceae). *Tropical Journal of Pharmaceutical Research*, Vol.13: 139-144.

Oga, S., 2003. *Fundamentos de Toxicologia 2nd ed*. Sao Paolo: Atheneu, 696.

Onal, A., 1996. Extraction of Dyestuff from Onion (*Allium cepa* L.) and its Application in Dyeing of Wool, Feathered-Leather and Cotton. *Turkish Journal of Chem*istry, Vol.20: 194-203.

Ozturk, A. W. & Abdullah, A. D., 2006. Toxicological effect of indole and its azo dye derivatives on some microorganisms under aerobic conditions. *Science of the Total Environ*ment, Vol.358 (1): 137-142.

Patel, N. K., 2011. Natural Dye Based Sindoor. *Life sciences Leaflets*, Vol.11: 355-361.

Pruthi, N., Chawla, G. & Yadav, S., 2008. Dyeing of Silk with Barberry Bark Dye Using Mordant Combination. *Natural Product Radiance*, Vol.7 (1): 40-44.

Radhika, A., Neelam, P. & Jeet, S.S., 2007 Effect of Mordants on Printing with Marigold Flowers dye. *Natural Product Radiance.*, Vol.6 (4): 306-309.

Sachan, K. & Kapoor, V. P., 2007. Optimization of Extraction and Dyeing Conditions for Traditional Turmeric Dye. *Indian Journal of Textile Knowledge*, Vol.6 (2): 270-278.

Sahoo, T., Bhattacharya, G., Das, P. & Dash, S. K., 2012. Colour Intensity, Fastness and Antimicrobial Characteristics of Silk Fabric Dyed with Mahua Bark. *Universal Journal of Environment Research and Technology*, Vol.2 (6): 591-600.

Samanta, A. K. & Agarwal, P., 2009. Review Article: Application of natural dyes on textiles, *Indian Journal of Fibre & Textile Research*, Vol. 34: 384- 399.

Samanta, A. K. & Konar, A., 2011. Dyeing Textiles with Natural Dyes. *Natural Dyes.* 29-56.

Samanta, A. K. & Kumar, P. E., 2011. Dyeing of Cotton Using Flower of *Cordia sebestina*: Effects of mordanting and fastness properties. *Journal of Asian Scientific Research*, Vol.1 (4): 137-142.

Sam, T. W., 1993. *Toxicity Testing Using the Brine Shrimp Artemia salina in: Bioactive natural products detection, isolation, and structural determination,* CRC Press, Boca Raton: FL, 442-456.

Sasidharan, S., Darah, I. & Jain, K., 2008. In Vivo and In Vitro Toxicity Study of *Gracilaria changii*. *Pharmaceutical Biolology*, Vol.46 (7): 413.

Shrivastava, A. & Dedhia, E.M., 2006. Extraction and Dyeing Methods of Natural dyes, *Natural Dyes Scope and Challenges* (ed. M. Daniel) Scientific Publication (India) Jodhpur, 67-79.

Siva, R., 2007. Status of Natural Dyes and Dye-Yielding Plants in India. *Current Science*, Vol.92 (7).

Sivakumar, V. S., Ravi, Q. T., Verma, P. G., Rao, U. D. & Swaminathan, G. F., 2000. Studies on the use of power ultrasound in solid-liquid myrobalan extraction process. *Journal of Cleaner Production*, Vol.15: 1815-1820.

Sorgeloos, P., Dhert, P & Candreva, P., 2001. Use of the brine shrimp, *Artemia* spp., in marine fish larviculture. *Aquaculture*, Vol.200: 147-159.

Sparling, D. W., Lowe, T. P. & Campbell, P. G. C., 1997. *Ecotoxicology of aluminum fish and wildlife. In: Research issues in aluminum toxicity*. USA: CRC Press, 47-68.

Stintzing, F. C. & Carle, R., 2004. Functional properties of *anthocyanins* and *betalains* in plants, food and in human nutrition. *Trends in Food Science and Technol*ogy, Vol.15: 19-38.

Strack, D., Vogt, T. & Schliemann, W., 2003. Recent advances in *betalain* research. *Phytochemistry*, Vol.62: 247-269.

Sudarmi, S., Subagyo, P., Susanti, A. & Wahyuningsih, A. S., 2015. Simple Extraction of Dragon Fruit (*Hylocereus polyrhizus*) Peel as Natural Dye Colorant. *Jurnal Prodi Teknik Kimia UPN "Veteran" Yokyakarta*, Vol.12 (1): 5-7.

Summoogum-Utchanah, S. L. & Joyram, H., 2015. An Investigation on the Potential of Extracting Natural Dyes from Beetroot and Turmeric. *International Journal of Research in Engineering and Technology*, Vol.4 (2): 401-416.

Trotman, E.R., 1984. *Dyeing and Chemical Technology of Textile Fibres*, Charles Griffin and Co. Ltd.

Umar, I. A., 2013. Fastness Properties of Colorant Extracted from Tamarind Fruits Pod to Dye Cotton and Silk Fabrics. *Journal of Natural Sciences Research*, Vol.3 (4): 60-66.

Vanhaecke, P., Persoone, G., Claus, C. & Sorgeloos, P., 1981. Proposal for a Short-term Toxicity Test with Artemia nauplii. *Ecotoxicology and Environmental Safety*, Vol.5: 382-385.

Vanhaecke, P. & Persoone, G., 1984. The ARC-test: a standardized short-term routine toxicity test with Artemia nauplii. Methodology and evaluation. *Ecotoxicology Test of Marine Environment*, Vol. 2: 588.

Vankar, P. S., Shankar, R. & Wijayapala, S., 2009. Dyeing Cotton, Silk and Wool Yarn with Extract of *Garnicia mangostana* Pericarp. *Journal of Textile and Apparel, Technology and Management,* Vol.6 (1): 1-10.

William, B. B., Gary, D. O. & Gray, A. V., 1976. *Clinical and Diagnostic Veterinary Toxicology*. Denmark: Gary, D. O.

Williams, P., Ongsakul, M., Proudfoot, J., Croft, K. & Bellin, L., 1995. Mangostin inhibits the oxidative modification of human low density lipoprotein. *Free Radical Resreach,* Vol.23: 175-184.

Zippel, E., 2004. Analysis of Textile Colorants and Coloured Textile. *Review of Progress in Colouration and Related Topics*, Vol.34(1): 1-11.

APPENDIX A

Table A.1: Raw data for the production of mangoesteen peel dye by using extraction method

Extraction	Colour observed	Volume (mL)
1	Reddish-purple	164.5
2	Reddish-purple	164.0
3	Reddish-purple	164.5
4	Reddish-purple	163.0
Total		656.0

Table A.2: Raw data for toxicity test of mangosteen peel dye extract on matured brine shrimp

	Concentration dye extract (μg/mL)	Log concentration	Number of killed brine shrimp			Mortality (%)	LC_{50} (μg/mL)
			1st	2nd	3rd		
Control	0	-	0	0	0	0	-
Dye without mordant	15.625	1.19	0	0	0	0	2512.71
	31.25	1.49	0	0	0	0	
	62.5	1.80	0	1	1	6.67	
	125	2.10	2	2	1	16.67	
	250	2.40	3	2	2	23.33	
	500	2.70	3	3	3	30.00	
	1000	3.00	4	4	4	40.00	
Dye with mordant ($CuSO_4$)	15.625	1.19	0	0	0	0	1807.96
	31.25	1.49	0	0	1	3.33	
	62.5	1.80	1	2	1	13.33	
	125	2.10	3	3	2	26.67	
	250	2.40	3	3	3	30.00	
	500	2.70	4	4	3	36.67	
	1000	3.00	5	4	4	43.33	

Calculation for LC_{50}

To find the concentration for causing 50% of brine shrimp mortality in mangosteen peel dye extract without mordant:

$$y = mx + c$$
$$y = 26.663x - 40.658$$
$$50 = 26.663x - 40.658$$
$$x = 3.40$$
$$\text{Anti-log concentration} = 2512.71\ \mu g/mL$$

Thus, the concentration of mangosteen peel dye extract without mordant to cause 50% of brine shrimp is 2512.71 µg/mL.

To find the concentration for causing 50% of brine shrimp mortality in mangosteen peel dye extract with $CuSO_4$ mordant:

$$y = mx + c$$
$$y = 23.333x - 26$$
$$50 = 23.333x - 26$$
$$x = 3.26$$
$$\text{Anti-log concentration} = 1807.96\ \mu g/mL$$

Thus, the concentration of mangosteen peel dye extract with $CuSO_4$ mordant to cause 50% of brine shrimp is 1807.96 µg/mL.

Where y is the percentage of mortality, m is slope of the graph plot, x is the log concentration of dye extract in sea salt water, and c is the y-intercept.

APPENDIX B

Wash fastness test of cotton and silk fabric dye with mangosteen dye extract

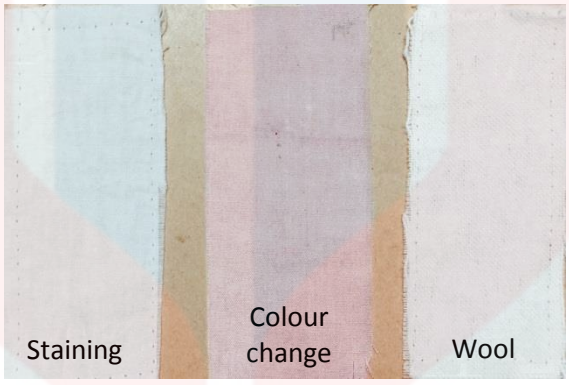

Figure B.1: The colour staining and colour change observed on cotton fabric tested dyed with mangosteen peel dye extract without mordant

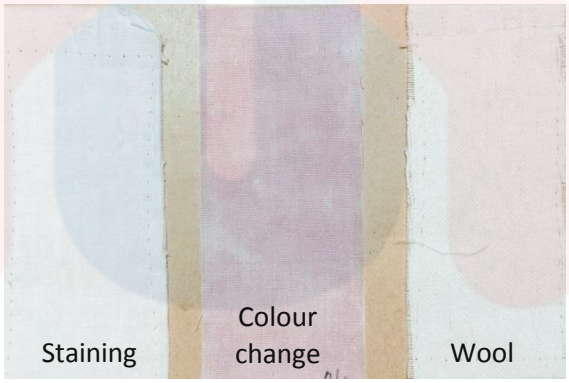

Figure B.2: The colour staining and colour change observed on cotton fabric tested dyed with mangosteen peel dye extract with $CuSO_4$ mordant

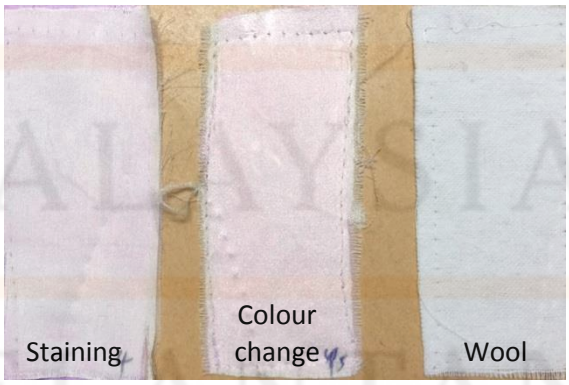

Figure B.3: The colour staining and colour change observed on silk fabric tested dyed with mangosteen peel dye extract without mordant

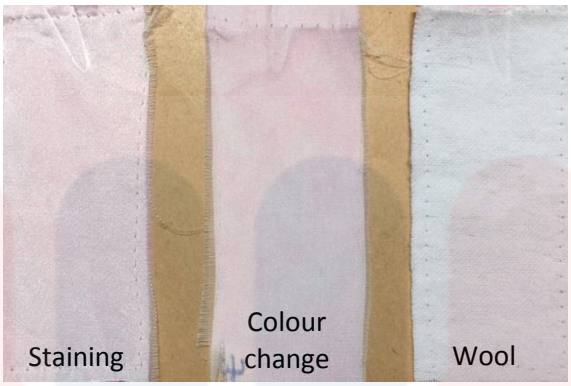

Figure B.4: The colour staining and colour change observed on silk fabric tested dyed with mangosteen peel dye extract with $CuSO_4$ mordant

Light fastness test of cotton and silk fabric dye with mangosteen dye extract

(a) (b)

Figure B.5: Cotton (a) and silk (b) fabrics dyed with mangosteen peel dye extract without mordant after 24 hours being exposed to UV light in Xenotest220 machine

(a) (b)

Figure B.6: Cotton (a) and silk (b) fabrics dyed with mangosteen peel dye extract with $CuSO_4$ mordant after 24 hours being exposed to UV light in Xenotest220 machine