OPTIMIZATION OF CONGO RED DYE REMOVAL USING JACKFRUIT PEEL AS LOW COST ADSORBENT.

by

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DECLARATION

I Nur Syuhada Binti Zulkifli declare that this thesis entitled Optimization of dye removal (Congo red) using jackfruit peel as low cost adsorbent 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 : ______________________________

Date : ______________________________

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LIST OF ABBREVIATIONS

pH	Potential of hydrogen
UV-vis	Ultraviolet-visible spectroscopy
nm	Nanometer
CO_2	Carbon dioxide
SO_2	Sulphur dioxide
O_2	Oxygen
$AlCl_3$	Aluminium hydroxide
$ZnCl_2$	Zinc chloride
HCl	Hydrochloric acid
KOH	Potassium hydroxide
NaOH	Sodium hydroxide
HNO_3	Nitric acid
$FeCl_3$	Iron (iii) chloride
$CaCl_2$	Calcium chloride
K_2CO_3	Potassium carbonate
H_3PO_4	Phosphoric acid
H_2SO_4	Sulphuric acid
$ZnOCl_2$	Zincconyl chloride
min	Minutes
ml	milligrams
m/L	Milligrams per litre
ppm	Parts per million
rpm	Revolutions per minute
SPSS	Statistical packages for Social Sciences
ANOVA	Analysis of variances
BET	Brunauer-Emmett-Teller
λmax	Maximum lambda

LIST OF SYMBOLS

%	Percentages
g	Gram
°C	Degree celcius
×	Multiply
≤	Less than and equal
≥	Greater than and equal
α	Significance level
=	Equal

ABSTRACT

Congo red dye is one of the carcinogenic of dye which can be affecting the human health and also the ecosystem of fresh water. This type of dye normally used in the textiles industry in the dye and rinsing process. The previous study had shown that adsorption is one of technology that most efficient in removing the pollutants from waste water. Commercial activated carbon had been used in this adsorption process but the production is high cost. So, there are many researcher try to find another alternatives such as produced the activated carbon from low cost of agricultural wastes. In this study, jackfruit peel was used as raw material because it has high carbocenous material. The activated carbon produced from the jackfruit peel was investigated under several parameters such as carbonization time, effect of contact time and initial concentration of dye. An amount of 0.5 g of best activated carbon removes 98.84% of 10 ppm dye with constant time 1 hour at 100 rpm. The best contact time was found to be at 1 hour because the removal percentage was decreasing as the contact of time increase. It also can be said that at 1 hour it has reached the equilibrium state which the active site becomes saturated. Other than that, the percentage removal also decreasing as the initial concentration of congo red dye solution increase. The initial concentration used start from 10 ppm until 100 ppm and the percentage removal was decreasing from 98.84% until 75.67%. The outcomes indicated that jackfruit peel can be used as a good low cost alternative for treatment of effluents containing congo red in synthesis waste water.

ABSTRAK

Pewarna Congo merah adalah salah satu pewarna yang boleh menjejaskan kesihatan manusia dan juga ekosistem air tawar. Pewarna jenis ini biasanya digunakan dalam industri tekstil seperti dalam proses mewarna dan membilas. Kajian sebelumnya telah menunjukkan yang proses penjerapan adalah satu teknologi yang paling berkesan dalam menghapuskan bahan pencemar daripada air sisa. Komersial carbon aktif digunakan dalam proses penjerapan ini tetapi kos terlau tinggi. Jadi, terdapat banyak penyelidik cuba untuk mencari alternatif lain seperti menghasilkan karbon aktif yang ber kos rendah daripada sisa pertanian. Dalam kajian ini, kulit nangka telah digunakan sebagai bahan mentah kerana ia mempunyai bahan carbocenous tinggi. Karbon aktif yang dihasilkan daripada kulit nangka telah diuji kaji mengikut beberapa parameter seperti masa karbonisasi, kesan interaksi masa dan kepekatan awal pewarna. Sejumlah 0.5 g karbon aktif yang terbaik membuang 98.84% daripada 10 ppm pewarna dengan pemalar masa 1 jam pada 100 rpm. Interaksi masa terbaik didapati 1 jam kerana peratusan penyingkiran itu berkurangan jika interaksi masa semakin bertambah. Ia juga boleh dikatakan bahawa pada 1 jam ia telah mencapai keadaan keseimbangan yang di mana ,tapak aktif menjadi tepu. Selain daripada itu, peratusan penyingkiran juga turut berkurangan seiring dengan peningkatan kepekatan awal pewarna. Kepekatan awal digunakan bermula dari 10 ppm hingga 100 ppm dan penyingkiran peratusan itu berkurangan daripada 98.84% sehingga 75,67%. Hasil menunjukkan bahawa kulit nangka boleh digunakan sebagai alternatif yang baik kerana kos rendah untuk rawatan air yang mengandungi congo merah di dalam air sisa .

CHAPTER 1

INTRODUCTION

1.1 Background

Nowadays, the pollution has become big concern issue especially in developed and developing country. Developed country has faced this pollution problem as they have the big industrial of agriculture; technology and industry that include textile industry and each country have their own problem because of different environment management. Water play an important role in our life as it was the need that every human must have in their life and human cannot survive without water. That's why many researchers have shown a high interest about how to reduce the water pollution in order to increase the sources and water quality for the next generation.

Water can be said contaminated if there are some substances or situation that can cause the water cannot be used for an important purpose (Owa, 2014). The sources of the contaminants are highly comes from the industrial and chemical industry that become a main environmental problems facing by the people around the world. The major sources of water pollution are comes from heavy metals, dyes, pharmaceuticals, pesticides, fluoride, phenols, insecticides, pesticides and detergents (Reddy & Lee, 2012).

Textiles industry is one of the most essential and popular sector in the world. Dyeing is a very synonym with the textile industry for the coloring purpose and this process have consumes large amounts of water and produces many wastewaters in the process of dyeing of the fabric (Patel & Vashi, 2012). The wastewater that contained dyes is very dangerous because it was hard to degrade in the water due to their resistant to fading when it was exposed to the sunlight and this is because of their chemicals structures.

The discharges of dyes direct into the water can affect the ecosystem and food chain of the aquatic life. Wastewater with higher organic pollutant usually have higher suspended material which will reduce the light from penetrates into the water and this process also can affect the photosynthesis process of aquatic life. The dyes also can be very dangerous when exposed to the human, since it can cause an allergy reaction and carcinogenic to the human (Sharma & Janveja, 2008)

1.2 Problem Statement

According to the issue of water pollution problem over the world, many researchers had shown an interest and do a research on how to remove the highly toxic organic compound (Congo red) from the water. Many advanced technologies have been discovered to remove the dye from the waste water in order to minimize the environmental pollution and impact. There are three type of the technologies have been use which are physical, chemical and biological method. Example of these methods includes the membrane filtration process, sorption techniques, coagulation and flocculation, aerobic and anaerobic microbial degradation. All the technique that were mentioned are very efficient in removing dyes from waste water however all the techniques are very highly cost and not too commercial for the use in the industry but all the technique have their own advantages and disadvantages (Pereira & Alves, 2012).

The abundance of agriculture waste also becomes a concern towards the environment because of the development of the development of many agriculture sectors and the food industry. This industry has produced many wastes and no proper way to disposed of this waste without produced the pollution. This study will help to utilize the amount of agriculture waste and how it can be used as a low cost adsorbent for removing the dye.

1.3 Purpose/Objective of Research

1. To explore the feasibility of jackfruit peel for the removal of Congo-red dye from aqueous solutions

2. To identify the optimum parameters which affecting removal of the dye process by using activated carbon from jackfruit peel.

1.4 Significance of study

This study will produce activated carbon from jackfruit peel for the removal of the congo-red dye from the aqueous solution. It also will help to utilize the amount of agricultural waste in the environment. Besides that, this study can help to sustain and recover the resources of water.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction of Dye Pollution

Many colors have been used in the fabric industry because it was the main attraction of human in choosing the clothes. Before the synthetic dyes have been discovered, the natural dyes have been used in colour the fabric. The natural dyes is not dangerous to the environment since it was extracted from the vegetables, fruits, flowers, certain insects and fish but this natural dyes have their own disadvantages and not suitable for the using in the textile industry because it’s have limitation of the colour and the colour that produced from the natural dyes is not too strong and dull. Other than that, the colour also easy to fade when exposed to the sunlight and washing (Kant, 2012)

Textiles industry is one of the largest industries in this world that contributed the most of the water pollution because large quantity of water was used in fabric processing (Pereira & Alves, 2012). Water pollution by dyes becomes the one of the most concerning environmental problem because these pollutants are toxic, carcinogenic and some of it can cause allergic reaction to the human. In Malaysia, homemade textile industry which is known as Batik industries is very famous especially in the east and coast of peninsular Malaysia. This industry has contribute large income for our country economic growth since it have a high demand from locally and abroad and this industry use many types of dye in processing the Batik (Ahmad *et al*., 2012).

Dyes are very dangerous because it can affect the chemical and physical properties of water and the aquatic flora and fauna. Besides that, dyes also resistant to the light exposure and difficult to fade away because of their stable chemical structure which can give problems to the aquatic life and the food chain (Sharma & Janveja, 2008). It has been estimated about 7×10^5 tons of dyes were discharged annually from all around the world by certain industrial activities (Chequer *et al.*, 2013).

Normally, the dyes that treated with the classical treatment do not effective and the adsorption process is an effective treatment method for their removal (Demirbas *et al.*, 2009). In this study, activated carbon was produced to study the efficiency of dyes removal by adsorption process.

2.2 Agriculture Waste

By products of agricultural waste activities are usually referred to as "agricultural waste" because they are not the primary product (Sabiiti, 2011). These agriculture waste is usually comes from the crop residue such as corn cobs, jackfruit peel, pineapple peel, orange peel and mango peel. Agriculture waste is usually come with large amount and can be used as resource since it can be renewable. In developed country, the production of agriculture waste is higher but they do not have the proper management on how to dispose this waste. Sometimes these agriculture waste just left until it rot or eliminate them by burning process, but the burning of agriculture waste can lead to the air pollution problem. The use of agriculture waste as a resources in process to produce the activated carbon can help in reduces the amount of waste and it can prevent the environmental problem from occur.

In this study, jackfruit peel was used as the low cost of agricultural waste to produced activated carbon. The jackfruit peel was treated with the chemical and put in the furnace at a certain time and activated carbon will be formed. Jackfruit peel is very common fruit for the Asian people but this jackfruit is originally come from the India country. In the jackfruit peel it contains lignocellulose and it also contain 4% ash, 10% moisture, 50% of volatile matter and 36% fixed carbon (Prahas *et al*., 2008).

2.3 Dyes

Application of dyes actually has been started over 4000 years ago in the textiles industry. The type of dye that was used during this time is natural dyes which produced from the extract of natural ingredients such as flower, vegetables and insects. Synthetic dyes does not exist during that time and not all people can wear the colored fabric because only certain people such as kings can wear colorful fabrics.

Dyes can be defined as substances that, when applied to a substrate it will provide color by a process that alters, at least temporarily, any crystal structure of the colored substances. Dyes widely used in the industrial activity such as cosmetics, plastics, paper, textiles and rubber to colour their product (Sharma & Janveja, 2008). Dyes can be categorized by their application, chemical structure and the group of atoms that composed by dye which is known as chromophores that will be responsible for the dye colour (Chequer *et al.*, 2009). Other than that, dye is colored because it absorbs the spectrum in the visible range at a certain wavelength and normally strong colour can be produced even it involve small amount of dye in aqueous solution because it related with the high molar extinction coefficient (Pereira & Alves, 2012).

2.4 Types of dyes

There are two types of dyes that were used in colouring process which are natural dyes and synthetic dyes. Natural and synthetic dyes have their own advantages and disadvantages.

2.4.1 Natural dyes

Natural dyes was discovered and use from a long time ago, before the synthetic dyes was discovered and widely used all over the textiles industry. Natural dyes same with its name comes from the natural resources such as plant leaves, root, bark, flower, vegetables and animal. Refer to the Table 2.1, there are some examples of natural dyes and this dye normally will be extracting from the natural resources and will be used in the colouring process of fabrics or other things. Natural dyes were very popular before the synthetic dyes were discovered and now only 1% of the industry used the natural dyes because it cannot fulfill or meet the huge demands from the people. Advantages of these natural dyes are it does not cause environmental problem since it comes from the natural resources but the colour that produces from the natural dyes will be fade easily when exposed to the sunlight or washing process (Saxena & Raja, 2014)

Table 2.1: Some example of natural dyes

Common name of the plant	Botanical plant	Part used	Colour obtained
1. Siam Weeds	*Eupatorium odoratum*	Whole plant	yellow
2. Gulmohar	*Deloxonic regia*	Flower	Olive green
3. Water lilly	*Nymphae alba*	Rhizomes	Blue
4. Dahlia	*Dahlia variabilis*	Flowers	Orange
5. Indian Jujube	*Berziziphus mauritiana*	Leaf	Pink
6. Golden dock	*Rumex maritmus*	Seeds	Brown
7. Red sandalwood	*Pterocarpus santalinus*	Wood	Red

2.4.2 Synthetic dyes

The first synthetic dye was discovered by William Henry Perkin and after that about 10,000 of new synthetic dye have been discovered and widely used all around the world. Eventhough, these dyes have contributes many pollution problems on the environment but it still win over the natural dyes. This is because synthetic dyes produced strong colour and the colour does not easily fade away when exposed to the sunlight or washing process because of their stable chemical structure which is stable. Nowadays, mostly of the industries preferred synthetic dyes more than the natural dyes.

2.5 Congo red dye

Congo red dye is the adsorbate which its colour is red and this red colour will appeared when the aqueous phase at pH>5 and turn to blue colour when at more acidic pH. Congo red dye can be categorized as dangerous because it is carcinogenic which comes from the effluents of textile industries during rinsing and dyeing process. Congo red also one of the most important dyes found in the waste water which has higher solubility in the water about 1g/30ml (Salman *et al.*, 2015). The chemical formula of Congo red dye is $C_{23}H_{22}N_6O_6S_2Na_2$ and its formulae weight is equal to 696.65 g/mol. Maximum wavelength for the Congo red dye adsorbance in the UV-vis spectrophotometer is 497 nm. Molecular structure for Congo red in two dimensions was shown in the Figure 2.1 below:

(Rong *et al*., 2015)

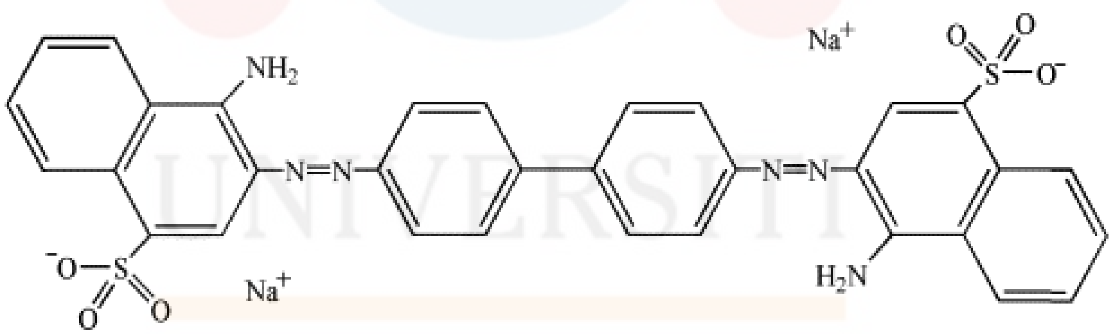

Figure 2.1: Congo red dye structure

2.6 Water pollution

Water is the most important thing in this earth because without water human and other living things cannot survive. According to this statement, clean water resources are needed but today, the resources of freshwater was depleted because of the rapid development in the urban and non-urban country. Nowadays, there is too much pollution of water caused by human activities such as industrial activity.

Water can be defined as polluted if there are some substances or condition that cause the water cannot be used for a specific purpose (Owa, 2014). Increasing number of contaminants that entering the water are normally comes from the industrial activity such as dyes, pharmaceutical, agriculture, pesticides, detergent and heavy metal (Reddy & Lee, 2012). The consequences from these contaminants will affect the quantity of clean water and lead to the depletion of water resources.

2.7 Harmful Effect of Dyes

There are many water bodies in this world that was polluted by dyes. Dyes largely consumed in textile industry and it can be considered as the largest contribution polluter in this world because this industry consumed large quantity of water in the dyeing process. The waste water of dye that discharged directly into the river or stream without any treatment can contaminated the water and give adverse effect to the aquatic life. More than that, it also will affect clearness and aesthetic of water. Some of textiles dyes can give bad impact to the human such as allergies, dermatitis and tumor and it can lead to a serious problem if the pollution of dyes increasing for every year.

Congo red is dangerous dye which is not safe to use because of it is extremely toxicity and have the carcinogenic properties which can affecting the cells of mammals and also can cause anaphylactic shock in human (Bekçi *et al.,* 2009). Direct discharge of dyes in the water can cause decreasing solubility of gases in the water and exhibits the effect to the organisms and causes adverse impact to the human being. Dyes are persistent to exposure of light and very difficult to fade away because of their stable chemical structure. This situation will lead to the reduction of biosynthesis process since the sunlight cannot penetrate into the water and it also will affect the food chain and aquatic life. The decreasing of dissolved oxygen in the water can disturb the photosynthesis process and automatically will affect the marine life which will lead to the fish death.

India is the most country that consumed large quantity of dyes in the textiles industry which is at 80%, including every type of dyes and pigment produced; this amount is close to 80,000 tonnes. Eventhough, India is the country that consumed large quantity of dyes; China is the number one in exported the dyestuff and the India is the second after China. There are about 10^6 tons of dyes produced annually and 1-1.5×10^5 tons are released into the environment in wastewater (Pereira & Alves, 2012). Dyes are normally resistant to the light and the colour is not easy to diminished or wash away. Desorption process is one of the effective treatment method in removal of dyes (Demirbas et al., 2009).

2.8 Methods Used For Dyes Removal

There are many technology have been developed from the ancient time until today but not all the technologies is suitable for the removal of dyes because some of the treatment are not commercial to be used and the cost is too expensive. Table 2.2 show that every treatment has their own advantages and disadvantages but the adsorption treatment is more preferable because of the cost is low and efficient in removing the dyes. There are many technologies that have invented such as:

2.8.1 Coagulation-Flocculation

Coagulation-flocculation process is where the coagulant was added in order to remove the suspended material from the wastewater. The coagulants can divide in two types which is organic coagulant and inorganic coagulant compound such as Aluminium sulphate, Aluminium Hydroxide chloride or high molecular weight cationic polymer. There are few factors that can accelerator effect the coagulation-flocculation process which is retention time, temperature, tank design, and condition of the equipment. Without coagulation-flocculation process sedimentation can only remove the coarse suspended material (Awaleh & Soubaneh, 2014).

2.8.2 Adsorption

The adsorption treatment is usually was said to be similar with absorption process because the concept is when the substances in a gas or liquid is attached to the solid. In our case we can said that the pollutant as adsorbate which it attached to the solid substance. Other than that, the adsorption also was said as a process of purification technique for removing the suspended material or compounds that contains in the wastewater that comes from the industrial effluents. Adsorption also the most popular and widely use technique for the removal or low concentration of non- degradable organic that comes from the drinking water preparation, groundwater, process water or as tertiary cleansing after (Ozhukil Kollath *et al*., 2015).

Usually the activated carbon was used in the adsorption study because activated carbon can be produced from low cost agricultural waste. The factors that affecting the adsorption study is surface area adsorbent, particle size, contact time or residence time, degree ionization of the adsorbance, ph, temperature, effect of adsorbent dosage and effect of initial concentration of metal (Regina *et al*., 2015). Table below shows advantages and disadvantages of treatment of dyes that was available.

Table 2.2: Type of treatment with their advantages and disadvantages

Treatment methodology	**Stage of treatment**	**Type of industry**	**Advantages**	**Disadvantages**
Physical methods				
1. Adsorption a. Activated carbon b. Bagasse c. Peat d. Wood chips	Pre/post treatment Pre treatment Pre treatment Pre treatment	Brewery/tannery/textile Sugar/Brewery Any industry Any industry	-Economical, good removal efficiency -Good adsorption for colourants	-Regeneration is costly -Post treatment disposal -Huge quantity required
2. Irradiation	Post treatment	Kraft mill, paper and pulp	Effective removal at low volumes	Requirement of dissolved oxygen

3. Ion exchange	Main treatment	Any industry	Low cost regeneration	Limited applications
Chemical methods				
1. Oxidation a. Fenton's regeneration b. Ozonation C.Electrochemical oxidation.	Pre treatment Main treatment Post treatment	Textile Brewery Textile	-wide range of decolourization. -Effective for both soluble and insoluble dyes.	-Expensive -Unsuitable for dispersed dyes -Cost intensive
2. Coagulation	Pre treatment	Sugar/pulp and paper	Low capital cost	Dewatering and sludge handling problems.
Biological methods				
1. Aerobic process	Main treatment	Kraftmill/Tannery	Colour as well as COD removal	Longer detention time
2. Anaerobic process	Main treatment	Pulp and paper/sugar/distillery	Biogas produced for steam generation.	Longer acclimatization phase
Single Cell (Fungal, Algae/Bacterial)	Post treatment	Any industry	Good removal efficiency for low concentrations and volumes	Cost intensive, unable to treat large volumes.

2.9 Activated carbon

Activated carbon was known as the oldest treatment and widely used in treat the wastewater that was polluted by organic and inorganic pollutants (Prahas *et al.*, 2008). Activated carbon widely used because of their extreme high surface area and micropores volume. Normally activated carbon was produced from materials that have carbocenous material and rich in carbon in it. Besides that, activated carbon always used in the adsorption process because of their surface chemistry and pore structure of porous carbon. Activated carbon also can be produced from agriculture waste such as jackfruit peel, orange peel, apple peel, sawdust and coconut husk (Bhatnagar *et al.*, 2013).

2.10 Modification of adsorbents

Modification of adsorbents is the process to increase the adsorption capacity of adsorbents by using different activation methods and the researchers usually used Langmuir isotherms in order to test the efficiency of the activation process. Activation process is usually can be divided into two types which are the physical activation and chemical activation (Bharathi & Ramesh, 2013). Example of the physical activation of the adsorbent is such as carbonization of the material and chemical activation is usually using chemicals as an activating agent. In this experiment, jackfruit peel will be treating by using the phosphoric acid as the chemical activating agent. There are many advantages of using chemical activation rather than physical activation because chemical activation are low energy cost and usually it only need the low temperature compare to the physical activation. Besides that, the chemical activation also will produce high yield and has better development of the porous structure (Prahas *et al*., 2007). Table 2.3 shows the advantages and disadvantages of using chemical and physical method.

Table 2.3: Advantages and disadvantages of using physical and chemical method

Types of method	**Advantages**	**Disadvantages**
Chemical	1. Reduce activation times 2. Activated carbons obtained in one step. 3. Produce high yield 4. High surface area of activated carbon 5. Lower temperature of	1. Involve a washing process 2. High cost 3. Corrosiveness the process 4. Inorganic impurities

	pyrolysis 6. Well-developed microporosity. 7. Decreasing of the natural mineral content.	
Physical method	1. A washing process is not involved. 2. low cost 3. The process is not corrosive 5. Prevents the incorporation of impurities coming from the activating agent.	1. The activated carbons obtained in two steps. 2. Higher temperature of activation. 3. Poorer control of the porosity.

2.11 Type of Acid Use in Modification of Adsorbent

There are many acids that can be used in the modification of the adsorbent. Firstly, phosphoric acid and zinc chloride is commonly used as the activation of the lignocellulose material that contained in the agricultural waste that will be using in this experiment. Other than that, the potassium hydroxide is metal compound and it can be used in the activation of the coal precursors and chars. Every chemical that used in the modification of the adsorbent have its advantages and disadvantages. For example, when compared to the zinc chloride, phosphoric acid is better because of the environmental value. Zinc chloride disadvantages compare to the phosphoric acid is the product that obtained cannot be used in the food industries and pharmaceutical because it may contaminate the product (Prahas *et al.*, 2007). In this study phosphoric acid and zinc chloride was used as chemical to soak the raw material. Table 2.4 shows some example of activating agents used in the synthesis of activation carbon for wastewater treatment.

Table 2.4: Example of activating agent used in synthesis of activated carbon

		Common activating agent	
Pollutants	Precursors	Physical	Chemical
Heavy metals	Nut shells, Peanut husk, Date pits, Oak cups pulp, Olives stones, Cane sugar bagasse, African palm pit, Chestnut shell, Coconut shell, Hazelnut shells, Maize stalks	Air, $CO_{2,}$ $SO_{2,}$ H_2O, $O_{2.}$	$AlCl_{3,}$ HCl, KOH, NaOH, $ZnCl_{2,}$ $HNO_{3,}$ $FeCl_{3,}$ $CaCl_{2,}$ $K_2CO_{3.}$
Dyes	Pecan nutshells, Orange seed, Cane bagasse, Olive seeds, Rice straw, Bamboo, Coffee husk, Jackfruit peel, Pistachio shell, Apricot shell, Coconut shells.	$CO_{2,}$ H_2O, Air	$H_3PO_{4,}$ $HNO_{3,}$ KOH, $CaCl_{2,}$ $FeCl_{3,}$ $ZnCl_{2.}$
Organic and inorganic pollutants	Oak cups pulp, Cashew nut shell, Almond shell, Cedar wood, Brewer’s spent grain lignin, Oil palm fruit bunch.	$CO_{2,}$ H_2O	$ZnCl_{2,}$ $ZrOCl_{2,}$ KOH, $HNO_{3,}$ $H_2SO_{4,}$ $K_2CO_{3.}$

2.12 Types of Adsorbents

There are two categories of adsorbents which are natural adsorbent and synthetic adsorbents. Natural adsorbents are usually abundant in supply, cheap and have great potential for modification of its material into the good adsorbent by enhance their adsorption capabilities. Examples of the natural adsorbents are zeolites, clay, clay mineral, ores and charcoal.

Synthetic adsorbents are usually made up from agricultural products and wastes, households waste, sewage sludge and polymeric adsorbents (Mohamed Nageeb., 2013) and in this study we will be using the agricultural waste as adsorbents. Examples of the synthetic adsorbents that made up from the agricultural waste are coconut husk, saw dust, tamarind fruit shell, bagasse fly ash and oil palm shell.

2.13 Factors that affect the rate of adsorption

2.13.1 Surface area of adsorbent

Surface area actually is related with the particle size, particle morphology, surface texturing and porosity (Trunschke., 2013). For the lower surface area (smaller pore size) materials had higher rate of adsorption for low boiling point and for adsorbing the contaminants at low concentration or in the small amount. The materials that have higher surface area can adsorb large amount concentration of contaminant due to the larger pore volumes (Mangun *et al*., 1997). So that's mean that smaller the size of material the higher the surface area and it can adsorbs large amount of contaminants since it have larger size of pores to adsorbs the contaminants. The material with the powdered size is more effective in adsorbs the contaminants compared to the material that produced in granule sized.

2.13.2 Developing Pore Size

The activation process is also carried out with effort made towards developing a high surface area carbon with desired pore size by optimizing the process parameters; such as the activation time, activation temperature, and impregnation ratio. Study of various parameters by Ahmadpour and Do revealed that the most important variable to porosity of activated carbon development is the ratio of the chemical agent to the precursor. The other operation variables with a direct effect on the porosity development are activation temperature and method of mixing. Nevertheless, it has been found that ordinary (sample—activating agent mixture) impregnation-method is the best method of mixing compared with physical and acid washed method (Prahas *et al*., 2007).

2.14 Jackfruit peel

Jackfruit scientific name is *Artocarpus heterophyllus* and it is very popular in India. Jackfruit also used as a cuisine in the South and the Southeast Asia. Jackfruit does not have a specific season and it can be harvested all year long. Jackfruit peel does not have any commercial value and it also contribute to the waste problem in the environment. The jackfruit peel become commercial value since it can be used as a low cost adsorbent. The jackfruit peel consist of lignocellulose (Prahas *et al*., 2007) which can be turned into carbocenous when treated with phosphoric acid and used as an adsorbent (Stephen Inbaraj & Sulochana, 2004).

CHAPTER 3

MATERIALS AND METHODS

3.1 Collection and Preparation of Activated Carbon.

The raw material that used in this study is jackfruit peel as shown in Figure 3.1. The jackfruit peel was collected from the market in Machang, Kelantan. Jackfruit peel cleaned by removing the carpel fibers and washed several times with distilled water to remove impurities. Next, the jackfruit peel was dried in oven at 105° C until constant weight was reached. Then the jackfruit peel was sieved or crushed into smaller size. The activating agent was used to treat the jackfruit peel, which is zinc chloride. This activating agent was used to activate the raw material. 10 g of raw material was impregnated by certain amount of 85 wt. % concentration zinc chloride with occasional stirring. The amount of phosphoric acid and zinc chloride used was adjusted to give a certain impregnation ratio of 1:3. The treated raw material was put in the furnace at constant temperature 300°C but at different time which are 5 min, 10 min, 30 min and 1 hour for the phosphoric acid and zinc chloride respectively. So, the four sample of activated carbon was obtained from same temperature but at different of activation time.

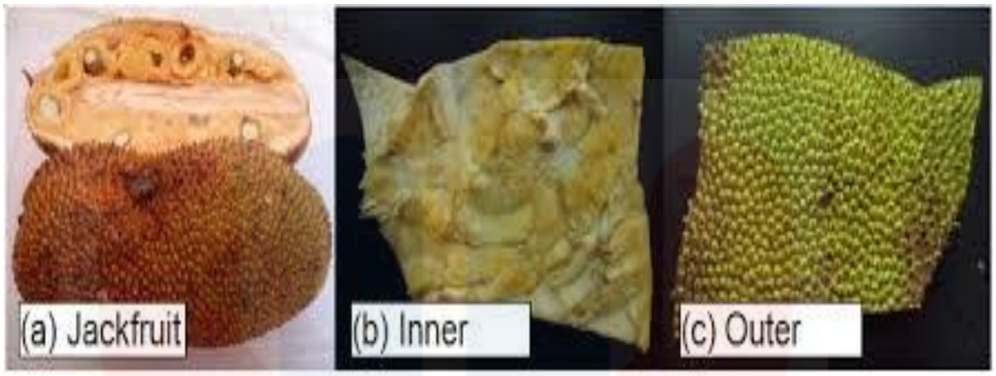

Figure 3.1: Image of jackfruit part (sources: Koh *et al*., 2014)

3.2 Washing process of activated carbon

The eight sample of activated carbon obtained was rinsed with hot distilled water by using vacuum pump or manual method until the pH 7 was obtained. pH can be measured by take the water that was obtained from the washing process and test by using pH meter or pH paper. The washing process is very important to remove chemical activating agents and can help a good development of porosity.

3.3 Oven dried

Activated carbon was dried at 150°C in the oven till their weight became constant. The activated carbon was stored into sterilize container until for the next use in the experiments.

3.4 Preparation of stock solution

The stock solution of 1000 mg/L of dye was prepared by dissolving 1g of Congo red dye in 1000 ml water. This stock solution was used to make different concentration needed in this experiment.

3.5 Calibration graph

A number of standard solutions which are 100 ppm were made from the stock solution in concentration range 0 to 12 ppm and a calibration curve was drawn by measuring the absorbance at λmax = 497nm using visible spectrophotometer.

3.6 Effect of contact time.

After the best activated carbon time was chosen, and then time was varied from 30 min to two hours to choose the optimum time. The concentration of dye solution was fixed with 10 ppm which was prepared from the 100 ppm standard solution. This dye solution was poured into two conical flasks with 0.5 g of best activated carbon was added into it. Then the solution was shaken at 100 rpm for 30 minutes and two hour at room temperature. Then, the conical flasks were taken out from the orbital shaker during the interval time, and were analyzed by UV-spectrophotometer. The percentage removal was calculated and the best optimization of time was chosen to proceed in the next parameter experiment.

3.7 Effect of Different Activation Time and Activating Agent.

A dye solution of 10 ppm was set by diluting the stock solution. This 10 ppm solution was added to four types of activated carbon (0.5 g) in the conical flask. Then the solution was shaken at 100 rpm for one hour at room temperature. Then, the four samples were taken out from the orbital shaker, and were analyzed by UV-spectrophotometer. The flow chart to determine the best activated carbon can be refer in the Figure 3.2.

The percentage removal was calculated and only the best activated carbon with the highest percentage removal was chosen to proceed the parameter test. The removal efficiency is calculated as:

$$\text{Percentage removal } \% = \frac{\text{Initial concentration} - \text{Final concentration}}{\text{Initial concentration}} \times 100$$

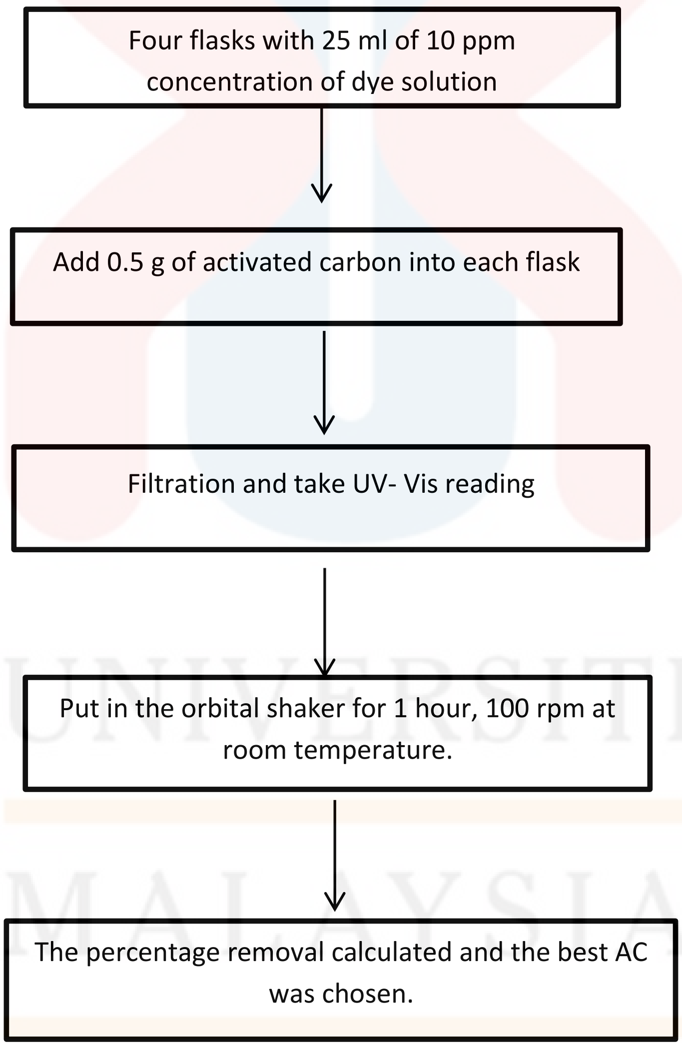

Figure 3.2: Experimental steps to determine the best activated carbon

3.8 Study of Initial dye concentration.

Dye solutions with varied concentrations of 10 ppm, 25 ppm, 50 ppm and 100 ppm were prepared by dilution from stock solution to be used as working solutions. To all these solutions, 0.5 gram activated carbon was added as adsorbent. Then, this solution was shaken in the orbital shaker at 100 rpm with the best optimization time at room temperature. Then, the solution was taken out from the orbital shaker and prepared for the uv-vis reading. The percentage removal was calculated based on the uv-vis reading.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Calibration graph.

In this experiment calibration graph was prepared from concentration of 100 ppm Congo red dye solution and the concentration was diluted to i.e. 0 ppm, 2 ppm, 4 ppm, 6 ppm, 10 ppm and 12 ppm respectively. The plotted calibration graph was shown as in the Figure 4.1 below:

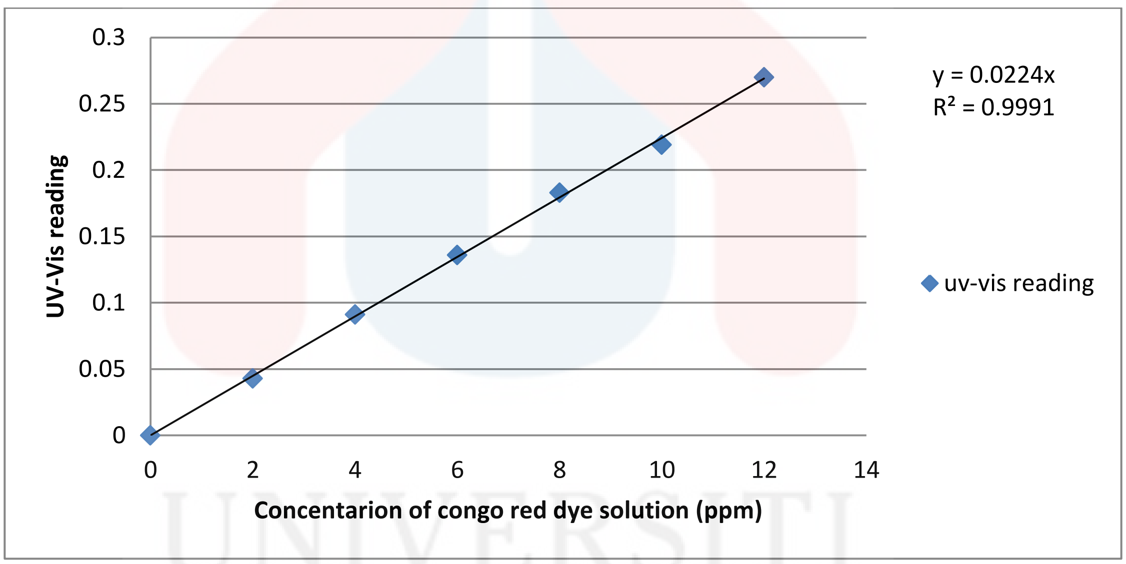

Figure 4.1: Calibration graph

Calibration graph was plotted in this experiment with the concentration range from 0 ppm until 12 ppm. This calibration graph is very important when we want to calculate the percentage removal. The new calibration graph needs to be plotted every day because the Congo red dyes solution concentration only last for one day. The concentration of Congo red dyes solution easily to degrade and it will affect the uv-vis reading and the calibration graph that we get may not accurate enough. Based on the figure 4.1, it was found that calibration graph plotted is accurate enough because the R^2 value equal to 0.9991. A perfect line would have R^2 value equal to one and normally R^2 value is more than 0.95(Huffman *et al*, 2009). This is because the uv-vis reading was done immediately after the solution of Congo red dye prepared, so the solution does not have time to degrade. Calibration graph is very important in most research work before the further calculation done. If the point on the line is not line properly, then it must have error when prepared the serial dilution or with the uv-vis reading. When this situation occur the serial dilution need to be repeat until the calibration become linear.

4.2 Effect of different activation time.

Figure 4.2 shows the effect of different activation time for the preparation of activated carbon using zinc chloride with constant furnace temperature of 300°C.

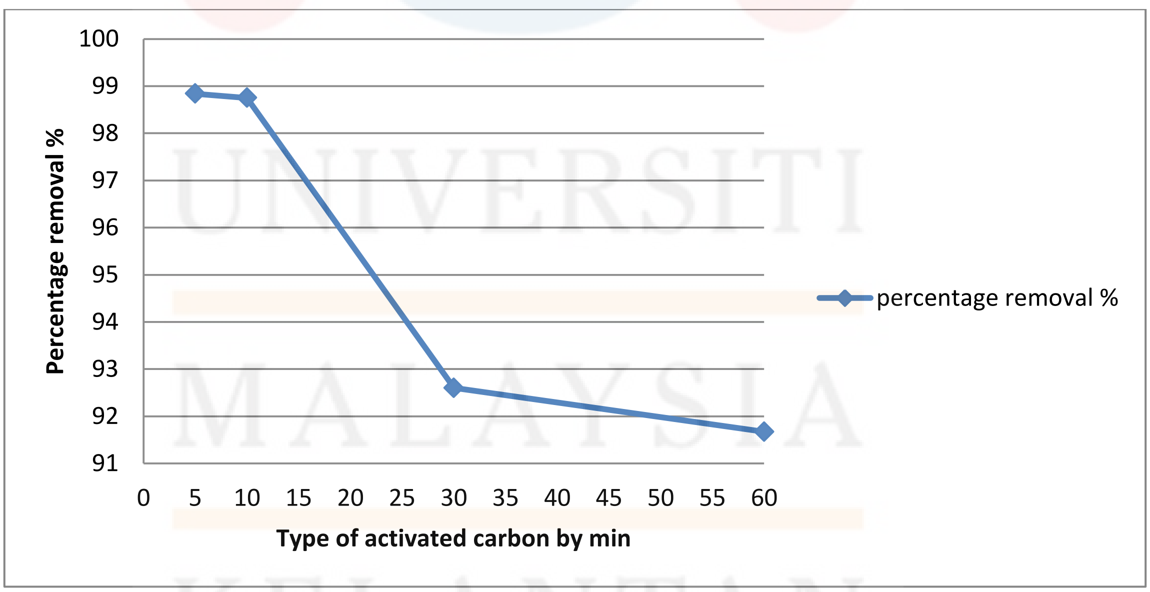

Figure 4.2: Effect of different activation time of activated carbon by using zinc chloride.

From the graph in Figure 4.2, it was shown that as the carbonizing time increase, the percentage removal percentage was decreased. Based on the graph plotted, the percentage removal of 5 and 10 min activation time was slightly decreasing. However, it was rapidly decreasing for the activation time of 30 min and 1 hour. The highest percentage removal was obtained for 5 min activation time.

This can be explained by relate the type of the chemical used as activating agent and the heating rate time during the carbonization process. Zinc chloride can be considered as one of the famous chemical used as an activating agent because it will produce good yield and well- developed porosity (Caturla *et al*., 1991). In this experiment jackfruit peel was treated with zinc chloride. The temperature used in chemical activation is lower than the physical activation and it only take shorter time to produce activated carbon. In the previous study, the heating rate used range from 2.5 min until 10 minutes (Brooks , 2015). So, it is not possible to get the best activated carbon at the heating rate time for only 5 minutes. Besides that, from the result obtained it can be seen as the heating rate increased the efficiency of the activated carbon to remove the dye also decreased. As the zinc chloride provides well developed porosity, the porosity rapidly developed when the heating time only 5 minute. As the heating time increased the porosity of the jackfruit peel cannot be well developed anymore.

Other than that, the impregnation ratios also need to be considered in process production of activated carbon. The complete saturation of the lignocellulosic precursor must be ensured to developed porosity of the material (Olivares-Marin *et al*., 2006). In this study, jackfruit peel was impregnated with the zinc chloride with the ratio 3:1. This ratio was used because to dissolved the jackfruit peel completely with the zinc chloride, so that the porosity of the jackfruit peel can be well developed. In the previous study, it was said that the maximal pore volume at impregnation ratio 3:1 is much higher compare to the maximal pore volume value at impregnation ratio of 2:1 (Uner et al., 2015). So, it can be concluded that the best activated carbon obtained using 5 min activation time with percentage removal of dye is 98.84%.

4.3 Effect of the contact time

In this experiment, the dosage of the adsorbent was 0.5 gram from the best activated carbon. The speed of the orbital shaker was also kept constant. The initial concentration of Congo red dye constant with 10 ppm and only the varied into 30 minutes, 60 minutes and 120 minutes. The result can be seen in the graph illustrated in Figure 4.3.

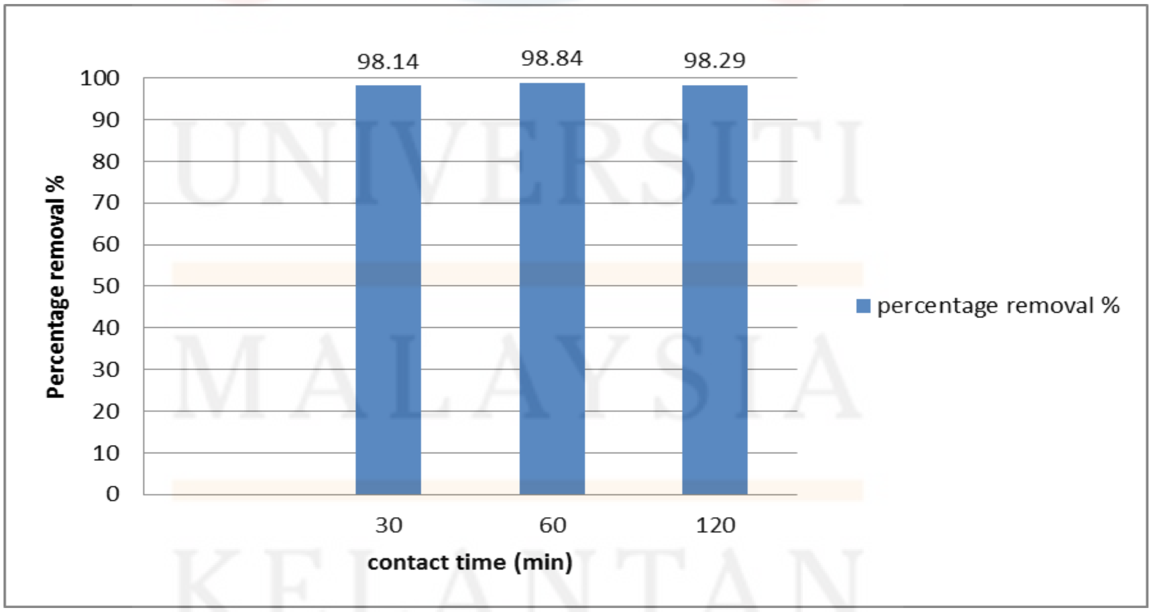

Figure 4.3: Graph for the effect of the contact time

The best activated carbon was chosen with 5 min activation time and was treated with the zinc chloride. For the test of the effect of the contact time, this activated carbon was used with the same dosage 0.5 g but with the time varied into 30 min, 1 hour and 2 hour respectively. From the graph illustrated in the Figure 4.4, it was found that the percentage removal was 98.14% after 30 min contact time. The removal percentage was slightly increased until 1 hour time and slightly decreased when the time increase to two hour. From the 30 min contact time the percentage removal is slowly increased until the 1 hour time and during this interval it can be said that the activated carbon was reached the equilibrium point in the process to remove Congo red dye. It can be related with the strong force attraction between the cation of the solution and the anion of the activated carbon sites.

For the two hour contact time the percentage was decreased because the activated carbon was reached its equilibrium state at 1 hour contact time. So, that is why the percentage removal decreases because the activated carbon cannot remove more dyes from Congo red dye solution because the site of the activated carbon has become saturated (El-Sayed *et al*., 2014). Equilibrium state means that no further change will occur after the equilibrium state had reached because during this time, even there are external force comes this state will become dynamic because it has reached their best state. By this, it can be considered that 1 hour as equilibrium time where there was slow of adsorption process occurs after this 1 hour. The equilibrium time is one of the essential parameters as it will contribute to economy efficiency in wastewater treatment system (Saifuddin & Kumaran, 2005). So, it can be concluded that one hour contact time is the best optimization time for the activated carbon removal of dye.

4.4 Effect of the initial concentration

In this experiment, the dosage of the adsorbent was 0.5 gram with the optimize contact time of 1 hour. The speed of the orbital shaker was also kept constant. The initial concentration of Congo red dye varied i.e. 10 ppm, 25 ppm, 50 ppm and 100 ppm respectively. From the graph illustrate in Figure 4.4, it can be explained that the percentage removal was slowly decreased with the increasing concentration of Congo red dye solution.

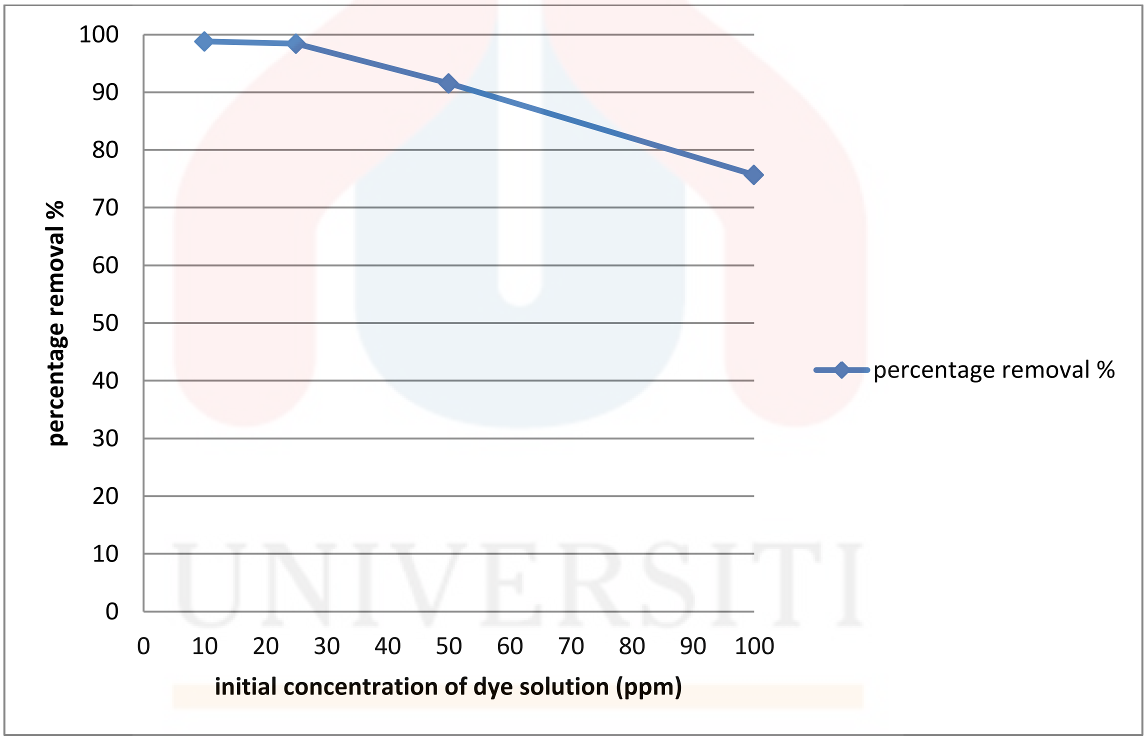

Figure 4.4 Effect of initial concentration dye

From the graph, it can be said that the highest percentage removal was at 10 ppm concentration and slightly decreasing at 25 ppm. The drastically decreasing pattern of the graph can be seen at 50 ppm and 100 ppm concentration of Congo red dye solution respectively. The decreasing pattern of the percentage removal with the increasing of the initial concentration of Congo red dye solution can be explained by the active site of the adsorbent. Since the mass of the adsorbance was fixed with 0.5 g, so the amount of the actives sites that function to remove the dye also remained constant. When the initial dyes concentration dyes increase, that's mean the activated carbon need to provide more active site for the removal of dyes, but when the dosage is constant and the initial concentration increase active site of the adsorbent will become more saturated and there will be decreasing in percentage of dye removal because the activated carbon cannot bear with the further removal of congo red dye solution. Similar result has been shown in the literature review (El-Sayed *et al.*, 2014).

Other than that, increasing of congo red dye solution in each flask also will increase the total moles remain in the liquid or aqueous medium after the adsorption process since a given constant of the activated carbon would adsorbs almost the same amount of the congo red dye (Jain , 2013). So, it can be proved that the adsorption of congo red dye was highly depends on the initial concentration of congo red dye solution.

If the initial concentration of congo red dye increase the dosage of the adsorbent also need to be increase. It was reported that, the increasing percentage removal of dye will be increasing with the increasing of the dosage adsorbent. This is explained by the number of actives site available when the amount activated carbon increase. Since the active sites of the adsorbent increase it will be capable to remove the higher initial dye concentration.

4.5 Data analysis

4.5.1 Effect Different of Activation Time

ANOVA analysis is very important to determine if there are significance differences or no significance differences in the experiment. Besides it is also important to decide either to accept the null hypothesis or reject the null hypothesis. Referred to the ANOVA table in Figure 4.5 below, the significance value is equal to 0.000. Normally, the first thing that researcher will see in ANOVA table is the significant value column because this is the exact significance level.

ANOVA

Percentage removal

	Sum of Squares	df	Mean Square	F	Sig.
Between Groups	134.476	3	44.825	185484.540	.000
Within Groups	.002	8	.000		
Total	134.478	11			

Figure 4.5: ANOVA table

The significance value is equal to zero but it can not to be interpret that the significance level is exactly zero because some number are too small to be detected and presented in the SPSS output. From the result above the 'Sig' value (p=0.00) is less than 0.05 and there is a statistical significance difference between the mean of activation time and the percentage removal. The critical value of alpha that set in this experiment is α=0.05. If the significant value is more than 0.05 than the result is not statistically significant and if the significant value is less than 0.05 the result is statistically significant.

The multiple comparisons table is available only when the result is statistical significant in order to find which group is different. This result was done by using Tukey test. From the result illustrated in the Figure 4.6, it can see there are statistical significant different in activation time between the group of 5 minutes and 10 minutes which *p=0.02* as well as between group of 5 minutes, 10 minutes, 30 minutes and 60 minutes which is *p=0.00*.

Multiple Comparisons

Dependent Variable: Percentage removal

Tukey HSD

(I) Time	(J) Time	Mean Difference (I-J)	Std. Error	Sig.	95% Confidence Interval	
					Lower Bound	Upper Bound
5 minutes	10 minutes	.07333*	.01269	.002	.0327	.1140
	30 minutes	6.22667*	.01269	.000	6.1860	6.2673
	60 minutes	7.17000*	.01269	.000	7.1294	7.2106
10 minutes	5 minutes	-.07333*	.01269	.002	-.1140	-.0327
	30 minutes	6.15333*	.01269	.000	6.1127	6.1940
	60 minutes	7.09667*	.01269	.000	7.0560	7.1373
30 minutes	5 minutes	-6.22667*	.01269	.000	-6.2673	-6.1860
	10 minutes	-6.15333*	.01269	.000	-6.1940	-6.1127
	60 minutes	.94333*	.01269	.000	.9027	.9840
60 minutes	5 minutes	-7.17000*	.01269	.000	-7.2106	-7.1294
	10 minutes	-7.09667*	.01269	.000	-7.1373	-7.0560
	30 minutes	-.94333*	.01269	.000	-.9840	-.9027

*. The mean difference is significant at the 0.05 level.

Figure 4.6: Multiple comparison table

4.5.2 Effect Contact of Time.

From the Figure 4.7 below, it can be seen that the significance value is equal to 0.00. When significance value is less than critical value, α=0.05 then this result is significant. This result is involves the dependent variable, percentage of removal and the factor is contact of time. So when the significance value 0.00 ≤ 0.05 that's mean the null hypothesis is rejected and there are differences of means between the percentage of removal and the group of time contact. The different specific group can be identified in the multiple comparison tables which contain the results of Tukey post hoc test.

ANOVA

Percentage removal

	Sum of Squares	df	Mean Square	F	Sig.
Between Groups	.857	2	.429	476.333	.000
Within Groups	.005	6	.001		
Total	.863	8			

Figure 4.7: ANOVA table

In the Figure 4.8, it can be see that there are statistical different significant in percentage removal between the group of time contact 30 minutes and 120 minutes which *p=0.004*. There also statistical significant different in percentage removal between all three group which are 30 minutes, 60 minutes and 120 minutes (*p=0.000).*

Multiple Comparisons

Dependent Variable: Percentage removal

Tukey HSD

(I) Time contact	(J) Time contact	Mean Difference (I-J)	Std. Error	Sig.	95% Confidence Interval	
					Lower Bound	Upper Bound
30 minutes	60 minutes	-.71000*	.02449	.000	-.7852	-.6348
	120 minutes	-.13000*	.02449	.004	-.2052	-.0548
60 minutes	30 minutes	.71000*	.02449	.000	.6348	.7852
	120 minutes	.58000*	.02449	.000	.5048	.6552
120 minutes	30 minutes	.13000*	.02449	.004	.0548	.2052
	60 minutes	-.58000*	.02449	.000	-.6552	-.5048

*. The mean difference is significant at the 0.05 level.

Figure 4.8: Multiple comparison table

4.5.3 Effect Initial Concentration of Congo Red Dye (ppm)

From the ANOVA table that illustrated in Figure 4.9, the significance value between groups is equal to 0.000. This result mean 0.000 ≤ 0.05 which is the result is significance and there is statistic different significance in percentage removal between the groups of different initial concentration which can be identified by further Turkey post hoc test. The null hypothesis was rejected because the result was significant and alternative hypothesis was accepted.

ANOVA

Percentage removal

	Sum of Squares	df	Mean Square	F	Sig.
Between Groups	1055.911	3	351.970	1279892.717	.000
Within Groups	.002	8	.000		
Total	1055.914	11			

Figure 4.9: ANOVA table

From the multiple comparisons table in the Figure 4.10, it can be concluded that there are statistic significant different in percentage removal between all the group which are 10 ppm, 25 ppm, 50 ppm and 100 ppm which the *p=0.000.*

Multiple Comparisons

Dependent Variable: Percentage

Tukey HSD

(I) Initial concentration	(J) Initial concentration	Mean Difference (I-J)	Std. Error	Sig.	95% Confidence Interval	
					Lower Bound	Upper Bound
10 ppm	25 ppm	.41333*	.01354	.000	.3700	.4567
	50 ppm	7.29667*	.01354	.000	7.2533	7.3400
	100 ppm	23.17333*	.01354	.000	23.1300	23.2167
25 ppm	10 ppm	-.41333*	.01354	.000	-.4567	-.3700
	50 ppm	6.88333*	.01354	.000	6.8400	6.9267
	100 ppm	22.76000*	.01354	.000	22.7166	22.8034
50 ppm	10 ppm	-7.29667*	.01354	.000	-7.3400	-7.2533
	25 ppm	-6.88333*	.01354	.000	-6.9267	-6.8400
	100 ppm	15.87667*	.01354	.000	15.8333	15.9200
100 ppm	10 ppm	-23.17333*	.01354	.000	-23.2167	-23.1300
	25 ppm	-22.76000*	.01354	.000	-22.8034	-22.7166
	50 ppm	-15.87667*	.01354	.000	-15.9200	-15.8333

*. The mean difference is significant at the 0.05 level.

Figure 4.10: Multiple comparison table

As a conclusion, ANOVA test is another way to test the hypothesis by tests whether the result is significant or not significant. This can be decide by find the significance level and compare with α value. If the significance level is less than α value then the result can be decided to be real and if not its vice versa.

CHAPTER 5

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

In this study it was highlighted the use of the use of agriculture waste as activated carbon to remove the Congo red dye pollution. The activated carbon produced from this experiment can be considered as successful because it has high percentage removal of Congo red dye solution. The highest percentage removal was 98.84% that was tested by using 10 ppm solution and the best activated carbon was obtained from five minute carbonization time and treated with zinc chloride.

Besides that, adsorption tends to be increase with the time of contact. This is because the free active sites will be decreasing as the time for adsorption process was continued until the active site was saturated. The optimum time in this study was found to be 1 hour of contact time. Increasing of time more than this will cause the percentage removal decreasing.

From this study, it can be seen that the increasing of initial concentration will cause the percentage removal become decreasing. It was proven by the result obtained from this experiment where the concentration use were 10 ppm, 25 ppm, 50 ppm and 100 ppm and the result was decreasing from 98.84% until the lowest 75.67%. It shows that the active site available to remove the dye is not enough with the higher concentration since the dosage of activated carbon is fixed 0.5 g.

Lastly, it also can be concluded that the activating agent and impregnation ratio used in this experiment is very important because it can affect the production of activated carbon. Zinc chloride was used in this experiment since it can produce high yield and environmental friendly and impregnation ratio used was 3:1 which is enough for activation of pore.

5.2 Recommendations

Agriculture waste that used as a low cost adsorbent potential need to be exploring so it can be one of the technologies that can be used in the future. There are many benefits that people can gain from this adsorbent since it can reduce the environmental pollution. The further testing for other raw material that can be used to remove the Congo red dye pollution need to be investigated since not all country have jackfruit peel to produce the same activated carbon.

Other than that, it will be the best if this activated carbon can be tested to filter the water that was polluted and produced the clear water that can be used as drinking water. It also can be used as one of the water filtered if the efficiency is proven to be successful. The using of the inexpensive chemical and shorter time to produced activated carbon also needs the further testing.

In this study, the developing size of pore does not observed. So, in the next experiment type and size pore developed in the activated carbon need to be observed in order to identify does the pore is fully developed or not. This analysis can be done by BET analysis.

Lastly, the application of jackfruit peel as activated carbon need to be explored as a long term technology so it can be used not only to remove the congo red dye but also valid for removed the other organic pollutants that caused water pollution problem.

REFERENCES

Ahmad, A. L., Harris, W. A., & Ooi, B. S. (2012). Removal of dye from wastewater of textile industry using membrane technology. *Jurnal Teknologi*, *36*(1), 31-44.

Awaleh & Soubaneh. (2014). The Concept and Current Technologies. *WasteWater Treatment in Chemical Industries*, *5*(1); 12.

Bekçi, Z., Seki, Y., & Cavas, L. (2009). Removal of malachite green by using an invasive marine alga Caulerpa racemosa var. cylindracea. *Journal of Hazardous Materials*, *161*(2), 1454-1460.

Bhatnagar, A., Hogland, W., Marques, M., & Sillanpää, M. (2013). An overview of the modification methods of activated carbon for its water treatment applications. *Chemical Engineering Journal*, *219*, 499-511.

Bharathi, K. S., & Ramesh, S. T. (2013). Removal of dyes using agricultural waste as low-cost adsorbents: a review. *Applied Water Science*, *3*(4), 773-790.

Chequer, F. M. D., Angeli, J. P. F., Ferraz, E. R. A., Tsuboy, M. S., Marcarini, J. C., Mantovani, M. S., & de Oliveira, D. P. (2009). The azo dyes Disperse Red 1 and Disperse Orange 1 increase the micronuclei frequencies in human lymphocytes and in HepG2 cells. *Mutation Research/Genetic Toxicology and Environmental Mutagenesis*, *676*(1), 83-86.

Caturla, F., Molina-Sabio, M., & Rodriguez-Reinoso, F. (1991). Preparation of activated carbon by chemical activation with ZnCl2. *Carbon*, *29*(7), 999-1007.

Demirbas, E., Dizge, N., Sulak, M. T., & Kobya, M. (2009). Adsorption kinetics and equilibrium of copper from aqueous solutions using hazelnut shell activated carbon. *Chemical Engineering Journal*, *148*(2), 480-487.

El-Sayed, G. O., Yehia, M. M., & Asaad, A. A. (2014). Assessment of activated carbon prepared from corncob by chemical activation with phosphoric acid. *Water Resources and Industry*, *7*, 66-75.

Huffman, M. L., & Venton, B. J. (2009). Carbon-fiber microelectrodes for in vivo applications. *Analyst*, *134*(1), 18-24.

Kant, R. (2012). Textile dyeing industry an environmental hazard. *Natural science*, *4*(1), 22.

Mangun, C. L., Daley, M. A., Braatz, R. D., & Economy, J. (1998). Effect of pore size on adsorption of hydrocarbons in phenolic-based activated carbon fibers. *Carbon*, *36*(1), 123-129.

Owa, F. W. (2014). Water pollution: sources, effects, control and management. *International Letters of Natural Sciences*, *3*.

Olivares-Marín, M., Fernández-González, C., Macías-García, A., & Gómez-Serrano, V. (2006). Preparation of activated carbon from cherry stones by chemical activation with ZnCl 2. *Applied Surface Science*, *252*(17), 5967-5971.

Ozhukil Kollath, V., Van den Broeck, F., Fehér, K., Martins, J. C., Luyten, J., Traina, K., ... & Cloots, R. (2015). A modular approach to study protein adsorption on surface modified hydroxyapatite. *Chemistry–A European Journal*, *21*(29), 10497-10505.

Patel, H., & Vashi, R. T. (2012). Removal of Congo Red dye from its aqueous solution using natural coagulants. *Journal of Saudi Chemical Society*, *16*(2), 131-136.

Pereira, L., & Alves, M. (2012). Dyes—environmental impact and remediation. In *Environmental protection strategies for sustainable development* (pp. 111-162). Springer Netherlands.

Prahas, D., Kartika, Y., Indraswati, N., & Ismadji, S. (2008). Activated carbon from jackfruit peel waste by $H_3\,PO_4$chemical activation: pore structure and surface chemistry characterization. *Chemical Engineering Journal*, *140*(1), 32-42.

Reddy, D. H. K., & Lee, S. M. (2012). Water pollution and treatment technologies. *Journal of Environmental & Analytical Toxicology*, *2012*, 2(5); 1-3.

Regina, Saraswathy, Kamal, Karthik & Muthukumaram. (2015). Removal of Nickel (II) Ions From Wastewater Using Low Cost Adsorbent: A Review. *Journal of Chemical and Pharmaceutical Sciences*, *8*(1); 6.

Rashed, M. N. (2013). *Adsorption technique for the removal of organic pollutants from water and wastewater*. INTECH Open Access Publisher.

Sabiiti, E. N. (2011). Utilising agricultural waste to enhance food security and conserve the environment. *African Journal of Food, Agriculture, Nutrition and Development*, *11*(6).

Salman, J. M., Amrin, A. R., Hassan, F. M., & Jouda, S. A. (2015). Removal of congo red dye from aqueous solution by using natural materials, Mesop. *Environ. j*, *1*(3), 82-89.

Saifuddin, M., & Kumaran, P. (2005). Removal of heavy metal from industrial wastewater using chitosan coated oil palm shell charcoal. *Electronic journal of Biotechnology*, *8*(1), 43-53.

Saxena, S., & Raja, A. S. M. (2014). Natural dyes: sources, chemistry, application and sustainability issues. In *Roadmap to Sustainable Textiles and Clothing* (pp. 37-80). Springer Singapore.

Sharma, J., & Janveja, B. (2008). A study on removal of congo red dye from the effluents of textile industry using rice husk carbon activated by steam. *Rasayan J Chem*, *1*(4), 936-942.

Stephen Inbaraj & Sulochana (2004). Carbonised jackfruit peel as an adsorbent for the removal of Cd(II) from aqueous solution. *Bioresource Technology, 94(1)* ; 49-52.

Trunschke. (2013). Surface area and pore size determination. *Modern Methods in Heterogeneous Catalysis Research.*Vol.1

Üner, O., Geçgel, Ü., & Bayrak, Y. (2015). Preparation and characterization of mesoporous activated carbons from waste watermelon rind by using the chemical activation method with zinc chloride. *Arabian Journal of Chemistry*.