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Production of regenerated cellulosic fiber from banana pseudo stem  
using copper amine solution

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By

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A Thesis

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## DECLARATION

I declare this thesis work intitled with **“Production of Regenerated Cellulosic Fiber from Banana Pseudo Stem Crosslinking with copper amine solution”** submitted for requirement for master degree at Addis Ababa university is my original work and has not previously been submitted for degree at this or any other university, and that all resources of materials used for this thesis has been fully acknowledged /referred.

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## **Acronyms**

|       |  |
|-------|--|
| ANOVA | Analysis of variance                   |
| DP    | Degree of polymerization               |
| Cuam  | Copper amine                           |
| FTIR  | Fourier transform infrared radiation   |
| TGA   | Thermogravimetric analysis             |
| LIDI  | Leather Industry Development Institute |
| Pred. | predicted                              |
| RF    | Raw Fiber                              |
| MF    | Mercerized Fiber                       |
| RCF   | Regenerated Cellulosic Fiber           |

## Abstract

Banana fiber is one of natural fiber and can be produced from pseudo stem with mechanical and chemical treating mechanism. The aim of this paper is producing regenerated cellulosic fiber from banana pseudo stem by using copper amine solution. Banana pseudo stem was collected around Jima zone and the fiber was extracted mechanically. The pseudo stem has around 6.5 w% of fiber and the fiber has a moisture content of 59.3 w% and has a cellulose content of 57-64 %. The analysis was taken which used to know the properties of raw fiber, mercerized fiber and regenerated fiber. Water absorption capacity is one useful technique to determine hydrophilicity of fiber. The water absorption of raw fiber, mercerized fiber and regenerated fiber was 4.85, 3.8 and 1.1 respectively. And the result shows that hydrophilicity of fiber decreases probably because of the hydroxyl group is replaced by sodium ion for mercerization and amine group for regenerated fiber. Other characterization techniques were also proceeded; such that FTIR analysis with a wave length between 4000 and 400  $\text{cm}^{-1}$ , thermal stability of regenerated fiber from a temperature 25 °C to 700 °C with a great mass loss from 200 to 400°C. And also, the crystallinity of regenerated fiber was calculated by taking reference polymers and gives around 72% crystallinity. The optimum factors obtained from design are time of 60.23 hours, copper amine volume of 66.96ml and concentration for regenerating compound is 1.07M for responses of water absorption capacity is 1.09 and yield of 1.4 gram.

**Key words:** *Banana Pseudo Stem; Raw Fiber; Mercerized Fiber; Regenerated Cellulosic Fibe*

# 1. Introduction

## 1.1. Background

Nowadays the need of textile products increased. The raw materials used for these textile products are fibers. Fibers are classes of materials that are continuous filaments or are in discrete elongated pieces, similar to lengths of thread. They can be spun into filaments, string or rope, used as a component of composite materials or matted into sheets to make products such as paper or felt. Fibers are often used in the manufacture of other materials mainly in composites, textiles and products which uses regenerated fiber as a raw material (Kalia *et al.*, 2009).

Generally, there are two types of fibers. Natural fiber and Manmade or synthetic fiber. Natural fibers include those produced by plants, animals, and geological processes. They are biodegradable over time. They can be classified according to their origin. Synthetic or man-made fibers generally come from synthetic materials such as petrochemicals. But some types of synthetic fibers are manufactured from natural cellulose. Banana fiber is a kind of new-type natural fiber. Banana fiber is extracted from bark of banana tree, it belongs to bast fiber. The appearance of banana fiber is similar with natural original bamboo fiber and ramie fiber, but fineness and spin ability of banana fiber is better than natural original bamboo fiber and ramie fiber. The chemical composition of banana fiber is mainly cellulose, hemicellulose, and lignin(Rashid *et al.*, 2015).

There are characteristics with high strength, small elongation, good luster, light weight, strong moisture absorption, fast moisture absorption and release, easy degradation as well as environmental protection etc. Banana fiber can be made into garment, curtain, towel, bedsheet etc. due to its characteristics with light, good luster and moisture absorption.

The rising interest in natural fibers in the composites field is undeniable, mainly due to sustainability, but also because of their good mechanical properties and low cost. The differences observed among different natural fibers are due to their chemical composition, origin, climate conditions, *etc.* On average, vegetable fibers are made of 60%–70% of cellulose, 10%–20% of hemicellulose, 5%–15% of lignin and up to 2% of pectin and waxes. Banana fiber is obtained from the superimposed leaves forming the pseudo stem of the plant, which currently has no use, apart from a low percentage dedicated to cattle feed. It is important to highlight that fibers are obtained

from the pseudo stems of the plant once the fruit has been harvested, and that each plant only bears fruit once; this is one of the main benefits of banana fibers in comparison with other natural fibers, as obtained from an agricultural residue. The use of natural fibers as reinforcement of polymeric parts has been widely studied, specially focusing on treating with chemical methods. As some researchers described, banana fibers are made of cellulose (65%), hemicellulose (17%), lignin (11%) and other substances (such as pectin, wax, 7%). Chemical methods for fiber extraction are usually performed with NaOH. Mechanical means are not able to remove the non-cellulosic constituents (lignin). An alternative is the use of biological processes, such as the immersed or solid-state fermentations. Enzymatic means are considered more environmentally friendly, and also avoid the fibers breakage. (Basel *et al.*, 2016), (Mohapatra *et al.*, 2010).

But when we see biological method of treatment, the difficulty is controlling enzyme parameters because of sensitivity of enzyme on temperature and this mechanism alters the property of fibers. So, the most common method of enhancing natural fiber is using chemical treatment with alkaline metals to avoid non-cellulosic parts of enzyme and cross-linking with acid aldehyde or amine groups (Lewin *et al.* 2006). Surprisingly regenerated cellulosic fiber sometimes called artificial silk can be produced from cellulosic fiber and a copper complex product with ammonia called copper tetraamine solution. The easy chemical way to obtaining hydroxide tetraamine, if on small particles of copper to act ammonia (ammonium hydroxide  $\text{NH}_4\text{OH}$ ), the result of chemical reaction is blue solution. Getting a nice blue solution of hydroxide tetraamine is the basis of a new process for the production of regenerated cellulosic fiber (Weed, 2014), (Kinnear, 2000).

In this paper chemical treatment mechanism was taken as for fiber intermediate processing with sodium hydroxide and then the treated fiber was performed a reaction with copper tetraamine solution and further injected in to sulfuric acid solution to form regenerated cellulose fiber. For characterize raw fiber, mercerized fiber and regenerated cellulosic fiber was measure its water absorbability, yield, density, functional group analysis, thermal stability and other related properties.

## 1.2. Statement of Problem

Fiber synthesis and fiber-based product are emphasized in most researchers many times. In recent, the areas for research based on fibers is going on natural fibers which is advisable. But when we see those researches that performs, most of them provides only at the intermediate step that is mechanically extracting and treating with alkaline solutions, and this is not enough for value adding of banana fiber. By further processing it is better to produce regenerated cellulosic fibers. And some researchers perform cross linking reactions on fiber but with epichlorohydrin (high toxic and expensive chemical) which can be substituted amine solutions.

And when we see problems other than research-based problems, there are resource related problems as follows. In most cases the use of natural fibers for textile is predetermined and restricted to conventional products. It is a well-known fact that cotton, silk, wool played a predominant role in textiles. Though there are other materials such as jute, ramie, sisal, banana fibers known to in human. Their uses are restricted to non-textile material such as packing material, ropes etc. The developments in the use of latest technology in adopting this non-apparel fiber have led to introduction of new diversified material. That is why; the present investigation focuses on enhancing and utilization of banana fibers for best suited end-uses. And banana fibers are completely biodegradable and naturally occurring, the banana fiber products are expected to be in great demand in the national markets as they pose no toxic effects to man and environment. Moreover, in our country banana pseudo stem is waste material which is a problem for farmers for their farm areas of cultivation. To overcome this problem converting of waste banana stem to valuable fiber as a resource for many products is un suspected idea.

In Ethiopia there are many traditional cloth producers with in their home. The raw material for their product is obviously cotton. And there are also products such as; mat, bag, handicraft, shipping rope etc. so, treated banana fiber will be a best substitution for their use.

It is known that, there are many wood processing cellulose related industries. So, to avoid deforestation it is better to change source of cellulose from waste material like banana stem fiber.

### **1.3. Objectives of The Study**

#### **1.3.1. General Objective**

The main objective of this thesis work is to produce regenerated cellulosic fiber from banana fiber extracted from banana pseudo stem using copper amine solution.

#### **1.3.2. Specific Objectives**

The specific objectives of the research were

- ✚ Extraction of raw banana fiber from banana pseudo stem.
- ✚ To treat the raw fiber using sodium hydroxide solution for removal of impurities and non-cellulosic components (lignin, pectin, wax and hemicellulose).
- ✚ To investigate the effect of treatment time and volume of copper amine solution on viscos formation.
- ✚ To characterizing yield, water absorptivity and other characteristics of raw mercerized and enhanced regenerated fiber
- ✚ To investigate the effect of mass of fiber to volume of copper tetra amine solution ratio with dissolving time and concentration of regenerating agent at atmospheric temperature and their interaction effect.

## 1.4. Significance of The Study

The significance of this work can be shown as the following perspectives,

- ✚ Promotes using bio degradable substitute for synthetic related composites, packaging materials and other cellulose related products.
- ✚ Since banana stem is a waste material, using this resource for further processing is one way of waste minimizing mechanism.
- ✚ It uses as a raw material for traditional cloth, mat, rope makers.
- ✚ It uses as blending with cotton or substituting for textile industries which uses also silk as a raw material for their products.
- ✚ It minimizes deforestation by substituting wood cellulose, thus protecting our ecology and environment.

## 1.5. Scope of The Study

This thesis work was proceeded from the raw banana stem plant to final enhanced fibers. Through this gap there are tasks for fulfillment of the work. Such as: raw fiber was extracted through mechanical wise and treated with alkaline solution and further gone up to producing regenerated fiber via cross-linking reaction of fiber with cross linking agent under different conditions; such as mass of fiber to volume of copper tetraamine solution, time of dissolving and concentration of regenerating agent. then characterizing of treated fiber was proceed and optimum conditions were analyzed.

## **2. Literature Review**

### **2.1. The need of banana fiber**

Mankind has been strongly dependent on plant fibers for all kind of purposes. In earlier days, natural fibers served a crucial role mitigate the everyday needs in a wide range of uses. But in recent years the arrival of synthetic products is dominating over the natural fiber, due to the low cost. But the synthetic fibers are nondegradable and causing serious pollution problems. Banana is one of the earliest and important fruit crops cultivated by man in tropical parts of the world. Banana is one of the important fruit crop cultivated in tropical parts of the world. Banana farming generates huge quantity of biomass all of which goes as waste and the above ground parts like pseudo stem and peduncle are the major source of fiber. Banana fiber can be used as raw material for industry for production of range of products like paper, cardboards, tea bags, currency notes and reinforced as polymer composite in high quality dress materials(Mohapatra *et al.*, 2010).

Banana fiber is a kind of new-type natural fiber. Banana fiber is extracted from bark of banana tree, it belongs to bast fiber. The appearance of banana fiber is similar with natural original bamboo fiber and ramie fiber, but fineness and spin ability of banana fiber is better than natural original bamboo fiber and ramie fiber(Preethi & G, 2016).

### **2.2. Types of Fibers**

#### **2.2.1. Manmade/Synthetic**

Synthetic fibers are attracted materials to develop composite materials and had resulted in tremendous growth of these materials. Recently, due to environmental concerns and growing cost of synthetic materials, the focus of research on natural fibers and their applications have created an enormous interest. Particularly, the study of lingo cellulosic fibers had attracted researchers due to their comparable properties at certain instances to glass, polymeric, carbon fibers ( Stud *et al.*, 2016).

#### **2.2.2. Natural Fibers**

Natural fibers are produced by plants, animals, and geological process. These fibers can be classified according to their origin, namely vegetable fibers, wood fibers, animal fibers and mineral fibers. Natural fibers occur in huge quantity in nature, which have properties which can be used in

many applications. From animal natural fibers silk is one of expensive and advanced fiber obtained from silk worms, it is formed naturally from the worm or artificially from cellulosic fibers. Nowadays there are two types of silks in the world, these are natural silk and artificial silk (Koschella & Friedrich, 2005).

#### **2.2.2.1. Natural Silk**

White shiny ball - this is the finest thread natural silk. It is built by the caterpillar silkworm. The caterpillar has two antennae on the head, which (as glands) that produce secretions, which harden and become silk thread. Silkworm builds a cocoon around themselves with a diameter of about 2 cm and a length of 3.5 cm. The thickness of the silk thread is only 0.02 mm, which is several times thinner than hair. If unwinding the cocoon of the silkworm, we get a thin thread approximately 3.5 km (3500 m). With such a thickness thread, of course, to work is not convenient, so usually before to work curl several threads into one. Natural silk is a complex organic substance with protein origin. It mainly consists of residues of molecules amino acids, tyrosine and aniline. Chemical formula of natural silk  $C_{15}H_{23}N_5O_6$ , which corresponds to chemical substance fibroin (from the Latin - fibra (fiber))(Kinnear, 2000).

#### **2.2.2.2. Coir Fiber**

Coir is a natural fiber extracted from the husk of coconut and used in products such as floor mats, doormats, brushes, mattresses, etc. Technically, coir is the fibrous material found between the hard, internal shell and the outer coat of a coconut. Other uses of brown coir (made from ripe coconut) are in upholstery padding, sacking and horticulture. White coir, harvested from unripe coconuts, is used for making finer brushes, string, rope and fishing nets. Mature brown coir fibers contain more lignin and less cellulose than fibers such as flax and cotton, so are stronger but less flexible. Coir fibers have higher strength and resistance towards saltwater makes it a better substitute for synthetic fibers; when the coir composites are used in construction industry.

#### **2.2.2.3. Jute Fiber**

Jute is a long, soft, shiny vegetable fiber that can be spun into coarse, strong threads. Jute is one of the most affordable natural fibers and is second only to cotton in amount produced and variety of uses of vegetable fibers. Jute fibers are composed primarily of the plant materials cellulose and lignin. It is thus a ligno-cellulosic fiber that is partially a textile fiber and partially wood. It falls into the bast fiber category. The industrial term for jute fiber is raw jute.

The fibers are off-white to brown and 1–4 meters long. It has high tensile strength, low extensibility, and ensures better breathability of fabrics. Therefore, jute is very suitable in agricultural commodity bulk packaging. Advantages of jute include good insulating and antistatic properties, as well as having low thermal conductivity and moderate moisture regain.

#### **2.2.2.4. Wool Fiber**

Wool is the textile fiber obtained from sheep and certain other animals; including cashmere from goats, mohair from goats, camel from animals in the camel family and angora from rabbits. Wool has several qualities that distinguish it from hair or fur: it is crimped, it is elastic and it grows in staples (clusters). Wool fabrics have a greater bulk than other textiles, and retain air, which causes the product to retain heat. Insulation also works both ways. Because of the crimp, wool fabrics have a greater bulk than other textiles, and retain air, which causes the product to retain heat. Wool fibers are hydrophilic, they readily absorb moisture, but are not hollow. Wool can absorb moisture almost one-third of its own weight. Wool based Composites can enhance both thermal and acoustical insulation ( Stud *et al.*, 2016).

#### **2.2.2.5. Banana Fiber**



*Figure 2-1 Extracted dried banana fiber*

Banana fiber is a kind of new-type natural fiber. Banana fiber is extracted from bark of banana tree, it belongs to bast fiber. The appearance of banana fiber is similar with natural original bamboo fiber and ramie fiber, but fineness and spin ability of banana fiber is better than natural original bamboo fiber and ramie fiber. The chemical composition of banana fiber is mainly cellulose, hemicelluloses, and lignin. There are characteristics with high strength, small elongation, good luster, light weight, fast moisture absorption and release, easy degradation as well as environmentally friendly etc. It has its own physical and chemical characteristics and many other properties that make it a fine quality fiber. The qualities such as strong, low elongation, lightweight, biodegradable etc. makes it suitable for composite application (Chinta *et al.*, 2012).

### **2.3. Structure and Properties of Banana Fiber**

The chemical composition of banana fiber is mainly cellulose, hemicellulose, and lignin. There are characteristics with high strength, small elongation, good luster, light weight, strong moisture absorption, fast moisture. Absorption and release, easy degradation as well as environmental protection etc. Banana fiber can be made into garment, curtain, towel, bedsheet etc. due to its characteristics with light, good luster and moisture absorption. Banana fiber average fineness is 2380Nm, 4.1dtex, average fiber length is 60mm, the feeling of banana fiber is better than ramie, fiber, there is no feeling of urtication. The appearance of banana fiber and banana series added a kind of new-typed natural and eco-friendly material for textile industries, banana fiber has played an important role in development of textile industry(Bhatnagar *et al.*, 2015).

### **2.4. Characteristics of Banana Fibers**

Banana fiber has its own physical and chemical characteristics and many other properties that make it affine quality fiber.

- Appearance of banana fiber is similar to that of bamboo fiber and ramie fiber, but its fineness and spin ability is better than the two.
- The chemical composition of banana fiber is cellulose, hemicellulose, and lignin.
- It is highly strong fiber.
- It has smaller elongation.

- It has somewhat shiny appearance depending upon the extraction & copper amine process.
- It is light weight.
- It has strong moisture absorption quality. It absorbs as well as releases moisture very fast.
- It is bio- degradable and has no negative effect on environment and thus can be categorized as eco-friendly fiber.
- Its average fineness is 2400Nm.
- It can be spun through almost all the methods of copper amine including ring-copper amine, open-end copper amine, bast fiber copper amine, and semi-worsted copper amine among others(Allam, 2015)

Thermal stability of raw banana fiber was studied by different authors, from many of them “Helena Becker” shows that there are three stages in thermogravimetric analysis stage, the first stage of mass loss, between 30 °C and 100 °C (11% mass variation) as shown the figure below, with a maximum rate of elimination at around 100 °C, was associated with the dehydration of surface water molecules. Thereafter, water could be removed by drying the samples at 150 °C. At the second stage, ranging from 200 to 400 °C, a larger mass loss was observed of approximately 55%, which can be attributed to hemicelluloses decomposition, breakdown of cellulose linkages (Becker *et al.*, 2013).

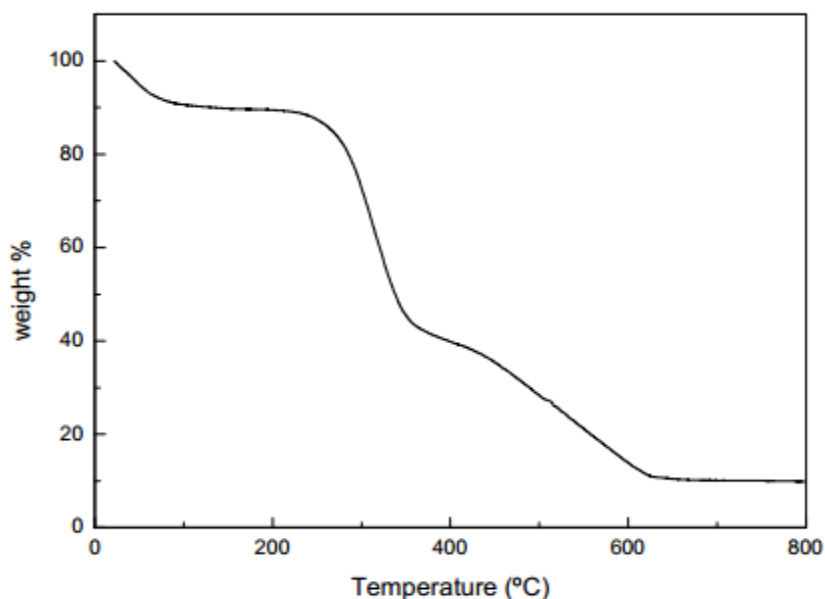


Figure 2-2 Thermogravimetric curve for pseudo-stem banana fibers using 5.0 mg of sample with a heating rate of 10 °C min<sup>-1</sup> under a nitrogen flow of 50 mL min<sup>-1</sup>(Becker *et al.*, 2013).

### **2.4.1. Components of Banana Fiber**

Banana fiber is obtained from banana plants consist mainly of three organic compounds: cellulose, hemicellulose, and lignin. These compounds are also major components of natural lignocellulosic materials. Cellulose molecules arrange regularly, gather into bundles, and determine the framework of the cell wall. Fibers are filled with hemicellulose and lignin. There is different bonding among cellulose, hemicellulose, and lignin. Cellulose and hemicellulose or lignin molecules are mainly coupled by a hydrogen bond. In addition to the hydrogen bond, there is the chemical bonding between hemicellulose and lignin, which results in the lignin, isolated from natural lignocelluloses, always contains a small amount of carbohydrates. cellulose is a long chain polysaccharide made up of glucose monomer units, which are alternately rotated to 180 degrees. Cellulose molecules align to form micro fibrils of diameter of about 3–4 nm. The micro fibrils have both crystalline and non-crystalline regions that merge together. The hemicelluloses, lignin etc. bound the cellulose into fibril aggregates of diameter roughly 10–25 nm. Hemicellulose binds to the surface of the cellulose micro fibrils, while lignin cross-links the hemicellulose molecules of adjacent micro fibrils. Hemicelluloses are short chain, amorphous polysaccharides with monomer units with acidic groups. They include xyloglucans, xylans, glucomannans and galacto glucomannans. Lignin is an amorphous, complex phenolic compound (Chen, 2014), (Kalia *et al.*, 2009).

## **2.5. Natural Fiber Processing**

### **2.5.1. Mechanical Processing**

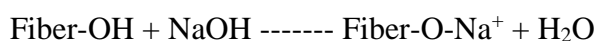
The extraction of the natural fiber from the plant required certain care to avoid damage. Initially the banana plant sections were cut from the main stem of the plant and then rolled lightly to remove the excess moisture. Impurities in the rolled fibers such as pigments, broken fibers, coating of cellulose etc. were removed manually by means of comb, and then the fibers were cleaned and dried. This mechanical and manual extraction of banana fibers was tedious, time consuming, and caused damage to the fiber. Consequently, this type of technique cannot be recommended for industrial application. A special machine was designed and developed for the extraction of banana fibers in a mechanically automated manner. It consisted mainly of two horizontal beams whereby a carriage with an attached and specially designed comb, could move back and forth. The fiber extraction using this technique could be performed simply by placing a cleaned part of the banana

stem on the fixed platform of the machine and clamped at the ends by jaws. This eliminated relative movement of the stem and avoided premature breakage of the fibers. This was followed by cleaning and drying of the fibers in a chamber at 200 °C for three hours. These fibers were then labeled and ready for lamination process. After fiber is collected, the process goes to yarn copper amine. The researcher investigated the traditional process, which use the filament yarns in weaving banana fabric. The finding showed that the convention process was very time-consuming, thus not appropriate for today's use. (Alam, 2015).

### 2.5.2. Chemical Processing

Chemical treatment of fibers is a common method of cleaning and modifying the fiber surface in order to lower the surface tension and enhance the interfacial adhesion between a natural fiber and a polymeric matrix. The most chemical treatment mechanism is Alkali treatment, removes fiber constituents including hemicellulose, lignin, pectin, fat and wax which exposes cellulose and increases surface roughness/area providing for improved interfacial bonding. Alkali treatment also modifies cellulose structure; modest treatments have been seen to bring about increased cellulose crystallinity considered to be due to removal of materials that could obstruct cellulose crystallinity, whereas at harsher treatments crystalline cellulose has been converted to amorphous material. Improvement of fiber strength has also been obtained using alkali treatment(Pickering *et al.*, 2016a).

#### 2.5.2.1. Alkali-Treatment



Alkali treatment removes fiber constituents including hemicellulose, lignin, pectin, fat and wax which exposes cellulose and increases surface roughness/area providing for improved interfacial bonding. Alkali treatment also modifies cellulose structure; modest treatments have been seen to bring about increased cellulose crystallinity considered to be due to removal of materials that could obstruct cellulose crystallinity, whereas at harsher treatments crystalline cellulose has been converted to amorphous material. Improvement of fiber strength has also been obtained using alkali treatment. (Pickering *et al.*, 2016b).

As (Sreenivasan, Ravindran, Manikandan, & Narayanasamy described that The SCFs is soaked in a stainless-steel vessel containing 10% sodium hydroxide solution for 1 h. The fibers were washed thoroughly with water to remove the excess of sodium hydroxide on the fibers. Final washing was

done with water containing a little acetic acid. Fibers were dried in an air oven at 70 °C for 3 h (Sreenivasan et al., 2012). (Bledzi and Gassan 1999) show that the dry fiber was treated with 5% solution of NaOH for 2 h to remove the unwanted soluble cellulose, hemi cellulose, pectin, lignin, etc. from the fiber. The fiber to solution weight ratio was maintained at 1:25. After 2 h the fiber was washed thoroughly in distilled water to remove excess of NaOH and dried at 60°C for 24 h.

### **2.5.2.2. Artificial Silk Preparation**

Artificial silk was first described in the scientific work of one of the French scientists rene Antoine Reaumur) in the mid-18th century. Studying insects, it is suggested that it is possible chemically to create a solution that would allow to extract from him a thin thread, suitable for machine. But practically his idea was realized only in the second half of the 19th century by French chemist (Louis Chardonnay)! Raw materials for the manufacture of artificial silk was cellulose and another complex chemical substance polysaccharide. Acting with nitric acid to these materials got nitrocellulose, this substance can be dissolved in a mixture of alcohol and ether, and extract from the thin filaments. The resulting streams fell into the cold water and froze there in thin threads. These silk threads automatically wound on the spool. When added to a solution of nitrocellulose any dye you can receive artificial silk with different colors. Nitrocellulose is a substance that ignites and burns perfectly, and also has explosive properties. This substance may exist in different modifications. Depending on the number of nitro groups that are included in the nitrocellulose, there are the following substances: colloxylin (with a small number of nitro groups), pyroxylin (with the increased number of nitro groups). The resulting substance is widely used for the manufacture of dynamite and smokeless gunpowder. It is known that the clothes are made from nitrocellulose instantly ignited and burned (Kinnear, 2000)

So, with combustible properties of tissue was developed a method to transfer of the cellulose in solution without of nitric acid! This method is based on copper-ammonium solution (call it copper amine solution). But interestingly, copper-ammonia solution does not contain a single molecule of the ammonia and does not contain copper! This solution was got in the mid-19th century by Swiss scientist-chemist Matthias Edward Schweizer. He is defined how to get the artificial silk from copper-ammonia solution. The composition of the solution -  $[\text{Cu}(\text{NH}_3)_4](\text{OH})_2$  copper hydroxide tetraamine. Beautiful dark-blue solution has very unusual properties: at room temperature it can dissolve cotton fabric and paper, wool, silk, and, what is especially interesting even dissolves hair!

In the mid-19th century was found rather easy chemical way to obtaining hydroxide tetraamine  $[\text{Cu}(\text{NH}_3)_4](\text{OH})_2$ . If on small particles of copper to act ammonia (ammonium hydroxide  $\text{NH}_4\text{OH}$ ), the result of chemical reaction was got the same substance - blue solution. It was also important to consider that the reaction proceeded with a sufficient amount of oxygen (e.g., outdoors). Getting a nice blue solution of hydroxide tetraamine is the basis of a new process for the production of artificial fibers in industry (Hearle & Woodings, 2001).

### **2.5.2.3. Dissolution of And Regeneration of Cellulose**

Cross-linked cellulose fibers and processes for making them have been described in many authors for many years (Tesoro, Cross-Linking of Cellulose, in *Handbook of Fiber Science and technology*, Vol. II, M. Lewis and S. B. Sello eds. pp 1-46, Mercel Decker, New York (1993)). The cross-linked cellulose fibers are typically made by reacting cellulose with polyfunctional agents that are capable of reacting with the hydroxyl groups of the anhydrous glucose repeating units of the cellulose either in the same chain, or in neighboring chains simultaneously. Cross-linked cellulose fibers generally are characterized by their high resiliency in the wet and dry states (Lewin, 2006).

Being a linear homopolymer, cellulose does not appear as a complex structure but its dissolution is in fact very complicated. Many factors contribute to make the cellulose macromolecule insoluble in most common solvents. Cellulose in its native form is often a very long polymer and high DP inevitably leads to decreased solubility due to decreased entropic gain in the dissolution process as already discussed. Glucose, cellobiose and any oligomer of cellulose structure up to around a DP of less than 10 is soluble in simple solvents such as water and cellulose is not, so this is indeed a relevant factor. Furthermore, the interactions between the cellulose molecules provide a challenge. To break these, solvents with high hydrogen bonding capacity are necessary. Quite a few liquids including water are able to swell, but not to dissolve cellulose. Swelling is defined as a process where the liquid molecule penetrates cellulose structure by interacting with the polymer to a certain extent, leaving the volume and physical properties of the cellulose significantly changed but the solid, or semi-solid state remains. Dissolution, on the other hand, completely destroys the supramolecular structure due to superior interaction between solvent and polymer, and results in a clear solution where the polymers are molecularly dispersed. The same liquid may act as a solvent or merely a swelling agent, depending on the conditions used in the experiment (Hearle, 2008).

#### **2.5.2.4. Solvents for Cellulose**

The efficient dissolution of cellulose is a long-standing goal in cellulose research and development. Dissolved cellulose is an indispensable prerequisite for Characterization (e.g., molecular weight and molecular weight distribution), Shaping (e.g., fiber copper amine of pulp) and Homogeneous phase chemistry of the polymer and of great commercial importance. From the commercial point of view, the viscose process, i.e., the cellulose xanthate, which is prepared by treating cellulose with CS<sub>2</sub> and NaOH, is by far the most important route to cellulose solutions. In addition, aqueous solutions of metal complexes like copper ethylene diamine or copper ammonium hydroxide are cellulose solvents known for a rather long time. The copper amine process uses the solubility of cellulose in cupric hydroxide or a mixture of a basic copper salt and concentrated ammonia. Copper amine into water regenerates the cellulose and in addition, an acidic bath for regeneration is applied. The copper amine process still keeps its importance for production of filaments, staple fibers, and membranes. Moreover, solvents of this type are applied for the determination of the molecular weight of cellulose by viscometer (Koschella & Friedrich Schiller, 2005).

#### **2.5.2.5. Systematization of Cellulose Solvents**

Several studies have been made in recent years to systemize the cellulose solvents known today. The classification according to the two categories of non-derivatizing and derivatizing solvents is considered to be most appropriate. The term "non-derivatizing" denotes systems dissolving the polymer by intermolecular interactions only. Aqueous transition metal complex solvents are conventionally included in this category despite an often very strong interaction, as no covalent interactions occur. The group of "derivatizing" solvents comprises all the systems where dissolution occurs in combination with formation of "unstable" ether, ester, or acetal derivatives. A specification within the large group of solvents acting via the formation of covalent derivatization of the polymer is given by the criterion that the derivative formed in a so-called derivatizing solvent is easily decomposed to regenerate cellulose by changing the medium (e.g., non-aqueous to aqueous) or the pH-value of the medium. Both categories of solvents comprise aqueous and non-aqueous media. Before presenting new results in the field of cellulose solvents, a systematic description of important cellulose solvents shall be given following the classification

mentioned above. and identifies three types of systems, namely non-derivatizing and derivatizing ones subdividing in aqueous and non-aqueous systems(Lewin, 2006).

### **Aqueous non-derivatizing solvents**

Aqueous solvents for cellulose, e.g., solutions of inorganic salts and complex compounds, were widely used for cellulose regeneration. The best known solvents of this group are copper ammonium hydroxide and copper ethylenediamine hydroxide. Despite the challenges associated with the dissolution of cellulose, numerous options are available. Here the focus lies on direct solvents, i.e. solvents that do not form covalent bonds with the polymer but merely interact by physical means. This distinction between no derivatizing and derivatizing solvents must be made in order to choose the correct solvent for a specific use. Solvents may further be classified as aqueous or non-aqueous, or as containing halogens or not or whatever else suits the user's purposes. Solvents can be further classified based on their number of components. Both unicomponent, bicomponent and tricomponent solvent systems are known and including solvent mixtures the range is even broader. Since many non-derivatizing, or direct, solvents for cellulose includes water, as bulk solvent or as bound water in a salt or organic hydrate, it is not fully satisfactory to make a binary distinction between aqueous and non-aqueous solvents. Instead, solvents could be placed on a continuous scale that represents the amount of water in the system. Cross-linking of fibers in fluff form is believed to improve the physical and chemical properties of the fibers in many ways, such as improving the stiffness, increasing resiliency (in the dry and wet state), reducing wrinkling, and improving shrinkage resistance. Any method of applying the cross-linking agent(s) to the fiber can be used in carrying out the cross-linking method of the invention. Acceptable methods include, for example, spraying, dipping, impregnation, and the like. Preferably, the fiber is impregnated with an aqueous solution of cross-linking agent. Impregnation usually creates a uniform distribution of cross-linking agent on the sheet and provides a better penetration of cross-linking agent into the interior part of the fiber (Hearle & Woodings, 2001).

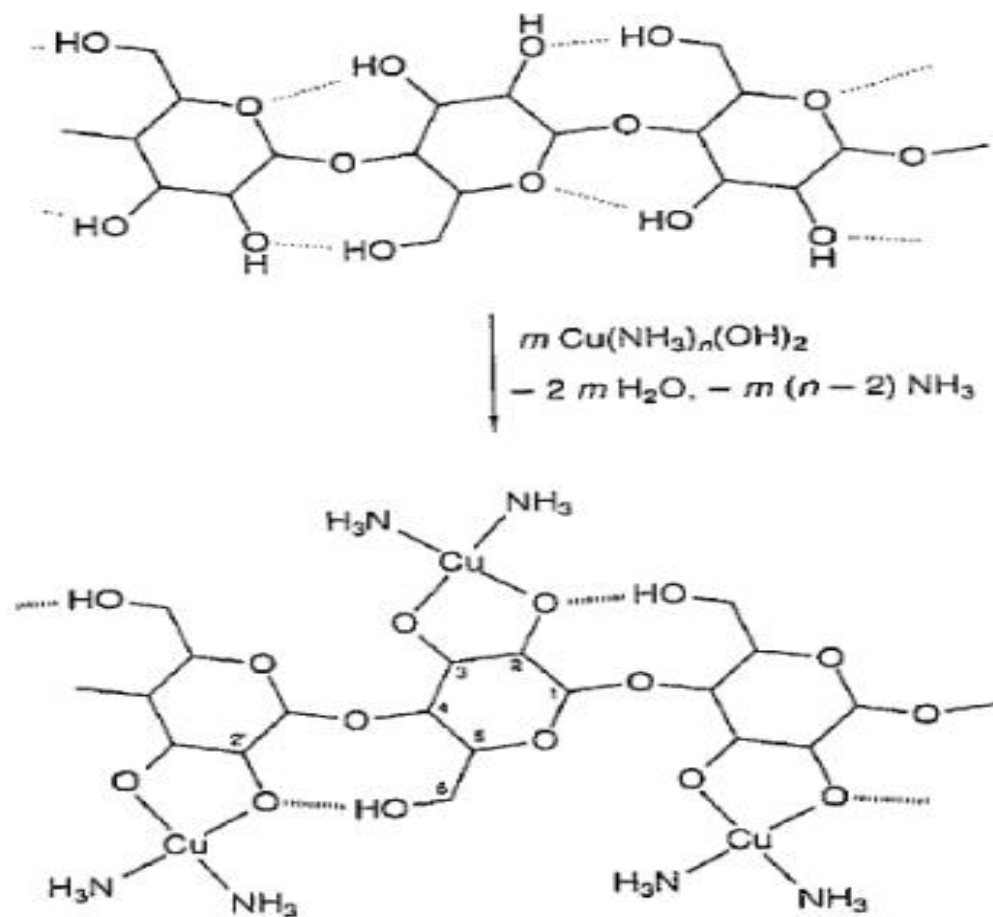


Figure 2-3 Cross linking of cellulose with copper amine solution

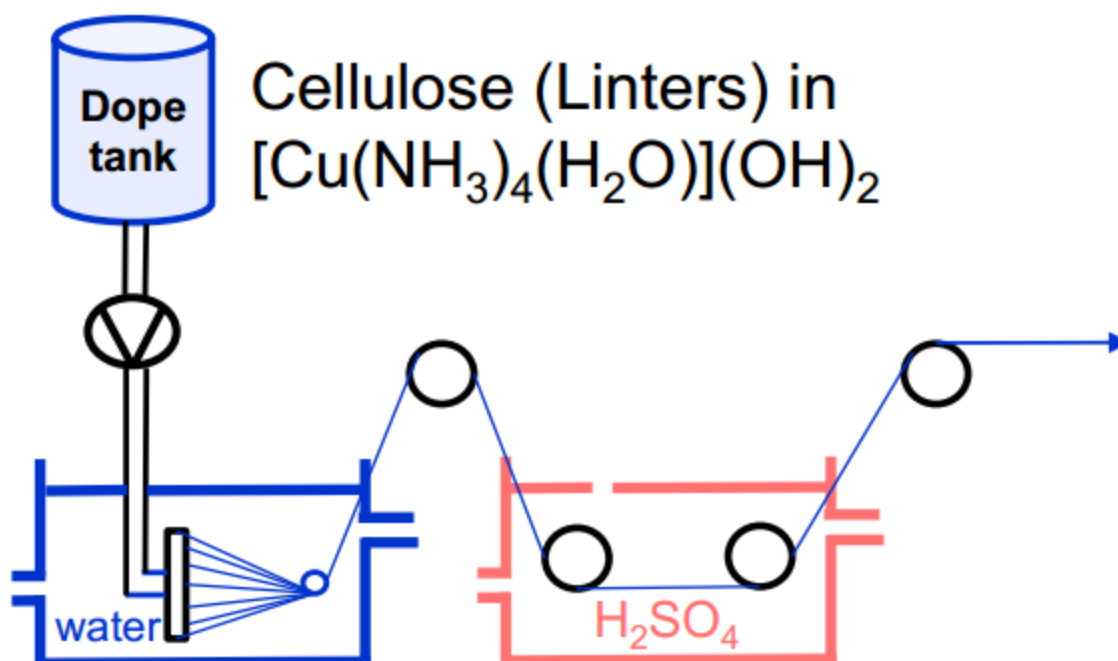


Figure 2-4 process of crosslinking and regenerating (Hearle & Woodings, 2001)

### 2.5.3. Biological/Enzymatic Processing

the use of biological processes, such as the immersed or solid-state fermentations. Enzymatic means are considered more environmentally friendly, and also avoid the fibers breakage, while altering the properties of the cellulosic fibers. There are different parameters which affect the enzyme choice, such as the type of substrate, composition, size, lignin content, etc. Previous studies show Pectinase and Xylanase as the most suitable ones for fiber extraction. Enzymatic treatments have been applied to hemp, flax or pineapple for fiber refining. Cellulases are used to remove fibrils from the surface and increase the smoothness of the fiber, although this treatment can also damage the fibers and reduce their mechanical properties. Pectinases are used in the textile industry for retting and degumming fiber crops, as they are capable of breaking down complex molecules of plant tissues into simpler ones, such as galacturonic acid; on the other hand, endoglucanases only act on amorphous celluloses. Hemicellulases are able to reduce water absorption by pentosan hydrolysis; xylanase and mannanase are used to dissolve hemicellulose (mainly xylan and glucomannan, respectively) (Basel *et al.*, 2016).

## **2.6. Factors Affecting Quality of Fiber**

### **2.6.1. Pre-Process Factors**

The properties of natural fibers are affected by many factors such as variety, climate, harvest, maturity, retting degree, decortications, disintegration (mechanical, steam explosion treatment), fiber modification, textile and technical processes (copper amine and carding). To understand the properties of natural fiber related products, it becomes necessary to know the mechanical, physical and chemical properties of natural fibers. The mechanical, chemical and physical properties of plant fibers are strongly harvest dependent, influenced by climate, location, weather conditions and soil characteristics. These properties are also affected during the processing of fiber such as retting, scotching, bleaching and copper amine(Kalia *et al.*, 2009).

Generally, the following are general factors that affect the property of natural fibers

Plant growth stage; this parameter includes, Species of plant, Crop Cultivation, Crop Location, Fiber location in plant and Local climate.

Harvesting stage; Fiber ripeness, which affects: Cell wall thickness, Coarseness of fibers Adherence between fiber and surrounding structure.

### **2.6.2. Post Process**

Fiber extraction, treatment mechanisms and conditions; Decortication process, the type of chemical used to treat is one factor to determine the quality of fiber. Not only the type of chemical but also the concentration, temperature and time of treatment are main factors for value of fibers(Basel *et al.*, 2016).

The amount of moisture absorption in the cellulose fibers can be dramatically reduced through chemical modification, such as acetylation, of some of the hydroxyl groups present in the fibers.

## 2.7. Uses and Applications

Most of the Banana fibers produced today is used for ropes and cordage. The resistance of the fiber to the sea-water and its natural buoyancy has created a ready market for it in the manufacture of shipping cables. It is also widely used for making power transmission ropes and cordage, wall drilling cables, fishing nets, lines and other types of cordage. Bast fibers such as flax, jute, hemp, and pineapple etc. plant fibers are all made up of thick walled cell tissue and they are bonded together by natural gums and support the branches, stems, leaves and fruits. Although banana plants and fibers are available in tropical regions in abundance, their application potential has not been exploited fully. At present, other companies make the limited application of banana fiber, for example, in making ropes, mats, and some other fields such as the composite materials. In recent years, more and more plant fibers were considered to be "environmentally friendly" fiber sources, and many countries are emphasizing the utilizing of these fibers (CEA, 2009).

Banana fiber is one of natural cellulosic fibers and present many advantages compared to synthetic fibers which make them attractive as reinforcements in composite materials. They come from an abundant and renewable resources at low cost which ensures a continuous fiber supply and a significant material cost saving to the plastics industry. Cellulose fibers, despite their low strength, can lead to composites with high specific properties because of their low densities. Unlike brittle fibers, such as glass and carbon fibers, cellulose fibers are flexible and will not fracture when processed over sharp curvatures. This enables the fibers to maintain the desired aspect ratio for good performance. Their non-abrasive nature permits a high-volume fraction of filling during processing, and this results in high mechanical properties without the usual machine wear problems associated with synthetic fibers especially glass and ceramic. Cellulose fibers are also non-toxic, easy to handle and present no health problems like glass fibers that can cause skin irritations and respiratory diseases when the fibrous dust is inhaled. They offer a high ability for surface modification, are economical, require low amounts of energy for processing, and are biodegradable. In terms of socio-economic issues, the use of cellulose fibers as source of raw materials is beneficial (Vigneswaran *et al.*, 2015). The best thing about these fabrics is that they are biodegradable, finally broken down into water and carbon dioxide by microorganisms in the soil.

### **3. Materials and Methods**

#### **3.1. Materials, Chemicals and Equipment**

##### **3.1.1. Materials**

The raw material used for the experimental work was banana plant (*Musa spp*) species. Banana pseudo stem was cut as soon as the fruits were harvested and transported using car from Jimma zone to Addis Ababa Institute of Technology School of Chemical and Bio Engineering laboratory class. Banana fiber was extracted traditionally with sharp blades from banana pseudo stem. After the raw fiber was extracted, there is a need of treating fiber to enhance its viability for further use.

##### **3.1.2. Chemicals**

Major Chemicals that were used in the experiment were;

Sodium hydroxide solution (NaOH) for treatment of raw fiber such that for removal of non-cellulosic components of raw fiber.

Copper hydroxide  $\text{Cu}(\text{OH})_2$  and Ammonia solution  $\text{NH}_4\text{OH}$  were used for preparation of a complex compound and mercerized fiber was dissolved in this compound.

Sulfuric acid  $\text{H}_2\text{SO}_4$  was used to regenerate cellulose from viscous solution.

##### **3.1.3. Equipment's/ Facilities**

Drying chamber (oven), mixing chamber, magnetic stirrer, filter paper, syringe, cutter or blade, washing Bowl, pH meter, conductivity meter, viscometer, water bath, test tubes, Fourier transform infrared radiation (FTIR) analyzer, thermogravimetric analysis (TGA) and beakers are the major equipment's used for experiment.

#### **3.2. Extraction of Raw Fiber**

The plant was cut down as soon as the fruits were harvested. The outer most covers were peeled and Brown-green skin was thrown away retaining the cleaner or white portion which can processed into knotted fibers. The extraction process of the fiber was proceeded by cutting the pseudo stem at the bottom to avoid non-fibrous materials as shown in (fig 3.1 a and b) with a sharp blade. fibers were extracted with a medium load to avoid breakage of fibers. Since the extracted fibers hade moisture, it was dried with sun for three days to remove moisture.

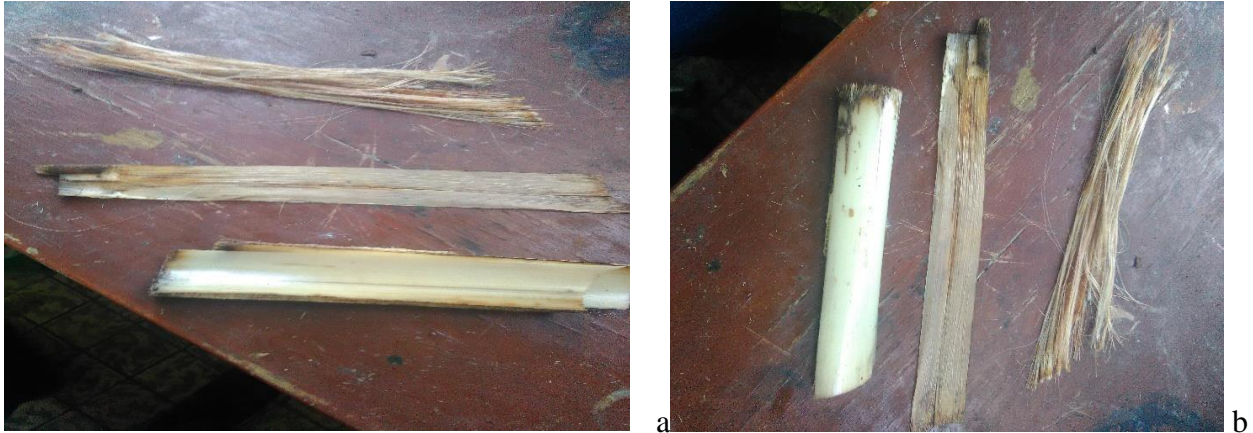


Figure 3-1 Extraction of fiber from pseudo stem a) starting from front cover and b) from back side cover

### 3.2.1. Estimation Yield of Fiber from Pseudo Stem

The yield of fiber present in stem was calculated by measuring one segment of the stem before extraction of fiber and measuring of extracted fiber and then the two masses were compared.

$$\% \text{yield} = \frac{\text{mass of fiber extracted}}{\text{mass of stem bundle}} * 100 \dots \dots \dots \text{eq 3.1}$$

### 3.3. Characterization of Raw Fiber

#### 3.3.1. Moisture Content

The moisture content of the raw fiber was measured with the principle of the mass difference between mass of fiber before dried and after dried. The process was done by drying the wet raw fiber up to constant weight, the decrease in the fiber weight can be calculated in the following formula (Ebisike *et al.*, 2013).

$$\text{moisture content}\% = \frac{\text{initial mass of fiber} - \text{mass of dried fiber}}{\text{initial mass of raw fiber}} * 100 \dots \dots \dots \text{eq 3.2}$$

#### 3.3.2. Cellulose and Non-Cellulose Content

Cellulose in banana fiber have been used to further processing, it is enough to know cellulose content and non-cellulose content. The cellulose content of raw banana fiber was analyzed by soaking the raw fiber with 7.5% sodium hydroxide solution for 2 hours at a temperature of 80 °C. After the process, soluble components were removed and the fiber was washed with distilled water

to the neutral condition happened and dried. The mass difference of raw fiber before and after soaking was taken to calculate the cellulose and non-cellulose content of the fiber by gravimetric analysis.

### 3.4. Mercerization of Raw Fiber Treatment

Mercerization is a treatment of fiber with dilute sodium hydroxide solution. The first chemical treatment chosen for this research was sodium hydroxide solution, which was obtained from chemical stores in Addis Ababa. 7.5% weight by volume Sodium hydroxide solution was prepared and dried raw fibers were treated with the alkaline treatment by soaking for 2 hours at a temperature of 80 °c as shown in fig 3.2a. sodium hydroxide soluble components were removed and the fibers were washed several times with distilled water to remove NaOH sticking to the surface to the pH becomes neutral. The fiber was dried at room temperature for 48 hours followed by oven at 70 oC for two hours and the dried mercerized fiber was stored for further use.



*Figure 3-2 mercerization of raw fiber (a fiber with sodium hydroxide solution b) washing with distilled water*

### 3.5. Experimental design

In this work there were three factors with three levels, these factors are volume of copper amine solution, treating time and concentration of regenerating agent( $H_2SO_4$ ). At constant fiber mass of 0.75gram, the volume of Schweizer's reagent was 40ml, 80ml and 120ml; a treating time of 24

hours, 48 hours and 72 hours with the concentration of regenerating agent( $H_2SO_4$ ) was leveled at concentration of 0.5M, 1M and 1.5M. Box Behnken design was selected to perform the analysis with a total number of 17 experiment in which 5 center point.

Table 3-1 factors and levels for experimental analysis

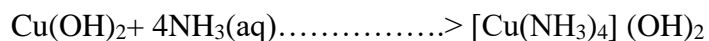
| Factors                              | Low point | Center   | High point | Responses        |
|--------------------------------------|-----------|----------|------------|------------------|
| Treatment Time                       | 24 hours  | 48 hours | 72 hours   | Water absorption |
| Copperamine volume                   | 40 ml     | 80 ml    | 120 ml     | Yield            |
| H <sub>2</sub> SO <sub>4</sub> conc. | 0.5 M     | 1 M      | 1.5 M      |                  |

these values were formulated to design expert box Behnken design method with proceeding responses to water absorption and yield with confidence interval of 95%. ANOVA out puts was manipulated and the effect of each factor on the response and also the interaction effect was justified on the responses water absorption and yield of regenerated cellulosic fiber.

### 3.6. Production Regenerated Cellulosic Fiber by Using Schweizer’s Reagent

#### 3.6.1. Schweizer’s Reagent Preparation

Copper tetra amine which known Schweizer’s reagent (copper amine complex compound) was prepared from copper hydroxide and 25% ammonia solution. The reaction was proceeded at ambient temperature. At the reaction time color changes was observed. The first one was a blue color and the last one was dark blue color. As the French scientist Schweizer described the deep blue black color tells the formation of the complex compound(James D. Martin†, 2013). The dark blue solution of hydroxide tetraamine copper  $[Cu(NH_3)_4](OH)_2$ , was obtained. This is also known as copper amine solution (Hearle & Woodings, 2001).



### 3.6.2. Dissolution and Regeneration of Fiber with Schweizer's Reagent and Dilute Sulfuric Acid Solution

The dried mercerized fiber was added to the solution as shown in fig 3.3a. Then, after reaction time the fiber was dissolved and a fairly viscous solution was formed probably because of crosslinking reaction of cellulose with Schweizer's reagent. The viscous solution was poured in the syringe and added to dilute sulfuric acid (fig 3.3b). finally, sulfuric acid and copper solution was reacted and regenerated fiber was precipitated(3.3c). then after, the regenerated fiber was washed with water(3.3d) and dried.



Figure 3-3 process of production of regenerated cellulosic fiber a) reaction of mercerized fiber to copper amine solution, b) injecting to sulfuric acid solution, c) precipitation of regenerated fiber and d) cleaned and put to be dried

### 3.7. Characterizing the Properties of Regenerated Cellulosic Fiber, Raw and Mercerized Fiber

#### 3.7.1. Viscosity of Dissolved Fiber Solution

The viscosity of fiber with Schweizer’s solution was measured by using universal viscometer at room temperature. and also, the complex compound itself and after the mercerized fiber dissolved by copper tetra amine solution was measured for comparison. The instrument was read the dynamic viscosity. And it can be converted to relative and specific viscosity by the following formulas (Berat, Kitosan, Kaedah, & Intrinsik, 2013).

$$\eta_r = \frac{\eta}{\eta_0} \dots \dots \dots \text{eq 3.3}$$

$$\eta_{sp} = \frac{\eta - \eta_0}{\eta_0} \dots \dots \dots \text{eq 3.4}$$

where;  $\eta$  = *viscosity of fiber to schweizer's reagent after reaction*

$\eta_0$  = *viscosity of schweizer's reagent before reaction*

#### 3.7.2. Water Absorption of Raw, Mercerized and Regenerated Cellulosic Fiber

Water absorption test was done by soaking grams of dried fiber with water for 24 hours and finally the soaked fiber was measured. After immersion, the excess water on the surface of the fiber was removed up using a filter cloth and the final weights of the specimens were then taken. The increase in the weight of the fiber was calculated using the following equation (Ebisike et al., 2013).

$$\text{fiber water absorptio}\% = \frac{\text{final weight of fiber} - \text{initial weight of fiber}}{\text{initial weight of fiber}} * 100 \dots \dots \dots \text{eq 3.5}$$

The water absorption capacity of three kinds of fibers of raw fiber, mercerized fiber and regenerated fiber were measured according to this principle. This value was calculated using equation 3.5.

#### 3.7.3. Density of Raw, Mercerized and Regenerated Fiber

Density was measured using Archimedes principle by using water as liquid. The mass of raw fiber, mercerized fiber and regenerated fiber was measured and immersed to a measured volume of the

water. This shows that the volume of the fiber was probably the volume difference of water after immersion and before immersion of fiber. And density was calculated with mass over volume formula (Amiri *et al.*, 2017).

$$\text{density} = \frac{\text{mass of fiber}}{\text{volume of fiber}} \dots \dots \dots \text{eq3.6}$$

#### 3.7.4. Crystallinity of Regenerated Fiber

Crystallinity was determined by using other complete crystalline and amorphous polymers. The two polymers that was taken for reference are polyethylene and polystyrene as crystalline and amorphous respectively. And the crystallinity of enhanced fiber was determined by the following formula.

$$\% \text{crystallinity} = \frac{g_c(g_s - g_a)}{g_s(g_c - g_a)} * 100 \dots \dots \dots \text{eq 3.7}$$

Where;

$g_c = \text{density of completely cristalline polymer}$

$g_s = \text{density of completely amorphous polymer}$

$g_a = \text{density of fiber.}$

The reference polymers that were used for this paper were poly ethylene for completely crystalline polymer and polystyrene for completely amorphous polymer (Derrick, 2010).

#### 3.7.5. Fourier Transform Infrared (FTIR) Spectroscopy

The chemical compositions of raw banana fiber, mercerized fiber and regenerated fiber were determined qualitatively using FTIR spectroscopy (FTIR-65, Perkin-Elmer) with the more conventional KBr pellets. FTIR spectroscopy is an important method to determine the presence and absence of particular bands of functional groups. FTIR is most useful for identifying chemicals that are either organic or inorganic. The instrument used was able to record spectra from wave numbers of 4000 to 400  $\text{cm}^{-1}$ . Spectrum is produced as a result of the absorption of infrared radiation. The functional group was determined based on the interpretation of the infrared spectrum obtained by comparing it with the standard spectrum group frequencies. The raw banana fiber, mercerized and regenerated samples for the analysis was first milled in a ceramic pestle and mortar to powdery conditions. The powder was then mixed with KBr pellets to make fiber plate and suitable for analysis. The mixture was then pressed to a small thickness, slightly below 1mm,

required for FTIR analysis. FTIR Analysis conducted in Addis Ababa University, College of Natural Science in the department of Chemistry. The FTIR data was graphed by origin pro 8.0 software for FTIR procedures.

### **3.7.6. Thermo Gravimetric Analysis (TGA)**

The thermal stability of regenerated cellulosic fiber was tested by measuring the mass loss during a heating ramp rate by thermogravimetric analyzer (TA instrument, SDT Q600 V20.9 Build 20) using temperature programming from 25 to 700°C at the heating rate of 20°C/min to which the sample was heated. For the thermo gravimetric analysis to be carried out, small amount of the sample was placed in a vial, which was present in the TGA analyzer. This vial is connected to sensors which detects the weight of the sample at all times. Testing was carried out under inert atmosphere (N<sub>2</sub>) with a flow rate of 2 ml/min to remove all corrosive gases and avoid thermo oxidative degradation and the retention time of the sample at the maximum temperature. These values were used as basis for the analysis. TGA was performed in Leather Industry Development Institute (LIDI) laboratory. The thermal degradation onset temperature and the thermal degradation weight loss of regenerated cellulosic fiber were recorded and analyzed using origin pro 8.0 software.

## **4. Results and Discussions**

### **4.1. Physical and Chemical Properties of Banana Fibers**

#### **4.1.1. Yield of Fiber from Pseudo Stem**

The yield of raw fiber from the pseudo stem was analyzed by taking a bar of stem with known weight and taking the wet mass of fiber after extracted. 3 replicates were recorded and calculated with the equation 3.1. From the calculation the average percentage yield of fiber in the pseudo stem is around 6.5 percent of fiber. (Vigneswaran et al., 2015) describes the yield of banana stem fiber from bundle stem was around 10% which is almost approximated with experimental result of 6.5% and the reason in the decrement of yield was probably the extraction process was not use modern machine and simply with knife.

#### **4.1.2. Moisture Content of Raw Fiber**

The moisture content of raw fiber used to determine amounts of water in the raw banana stem fiber was measured by taking three replicates of wet fibers Then the moisture content was calculated using equation 3.2. The average percentage moisture content obtained was around 59.3 weight percent which is less moisture content than the work of Ebisike K. in 2013 which shows the moisture content of fresh banana fiber was around 94% which is higher than the value 59%. This may probably cause by the removal of moisture during transportation and the extraction process (Ebisike *et al.*, 2013).

#### **4.1.3. Cellulose and Non-Cellulose Content**

Since cellulose part of banana fiber have used to further processing, it is enough to know cellulose content and non-cellulose content. Three replicates were taken for test and the percentage cellulose content was around 57%-64% which matches with the authors: (Ravi Bhatnagar *et al.*, 2014) describe the cellulose content of banana pseudo stem fiber was around 63-64%. And the other authors were also quantified the cellulose content of banana stem fiber between 60 and 65%.

#### **4.1.4. Water Absorption of Raw Fiber and Mercerized Fiber**

The water absorption capacity of fiber was measured for raw fiber, mercerized fiber and regenerated fiber. The absorption capacity of raw fiber was around 4.85. This large number is because of hydroxyl group of raw fiber. And for mercerized fiber the water absorption of 3.8 which is less than raw fiber probably because of some hydroxyl groups are substituted by sodium during mercerizing process (Kalia *et al.*, 2009).

#### **4.1.5. Density of Raw, Mercerized and Regenerated Cellulosic Fiber**

Density was measured using Archimedes principle by using water as liquid. The density of the raw fiber was 0.3gm/cm<sup>3</sup>, 0.38gm/cm<sup>3</sup> for mercerized fiber and 1.6gm/cm<sup>3</sup> for regenerated cellulosic fiber. The results described the density was increased as the fiber treated more because of more polymerization gives higher molecular arrangements from raw to regenerated. And also, at mercerization less dense components such that lignin and hemicellulose was removed and denser component (cellulose) was probably describes the content of mercerized fiber.

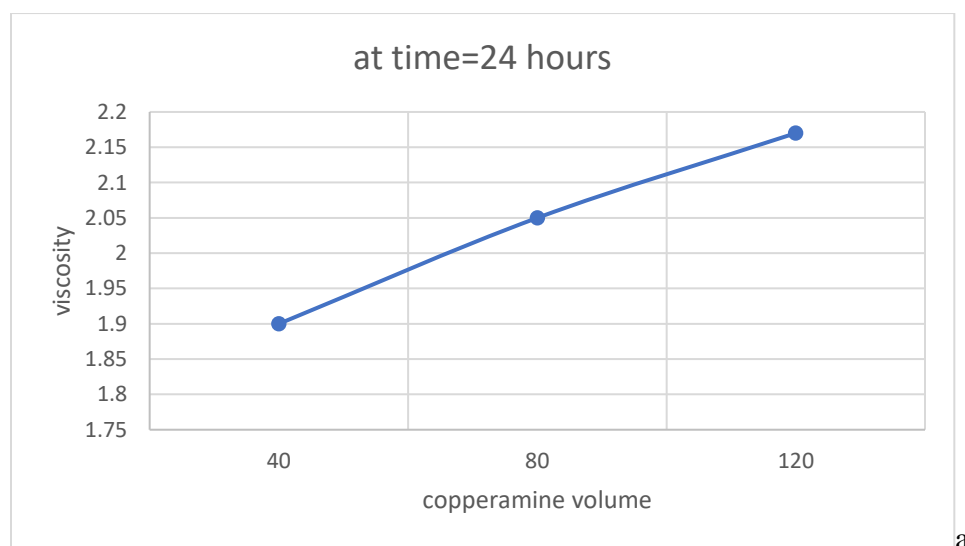
#### **4.1.6. Viscosity of Dissolved Fiber Solution**

The fiber to copper amine solution viscosity before regenerated of fiber used to relate degree of polymerization of regenerated cellulose in which, Polymerization of mercerized fiber with copper amine solution increases viscosity whereas degradation decreases viscosity. As shown below in table 4.1 high viscosities were obtained at time is equal to 48 hours and volume of 80 ml and this is probably because of better crosslinking reaction proceeds at this stage.

Table 4-1 treatment time and copper amine volume effect on viscosity of dissolved fiber solution

| Std | Treatment time (h) | Copper amine volume(ml) | Viscosity (Mpa.s) |
|-----|--------------------|-------------------------|-------------------|
| 1   | 24                 | 40                      | 1.9               |
| 2   | 72                 | 40                      | 1.5               |
| 3   | 24                 | 120                     | 2.17              |
| 4   | 72                 | 120                     | 1.51              |
| 5   | 24                 | 80                      | 2.05              |
| 6   | 72                 | 80                      | 2.09              |
| 7   | 24                 | 80                      | 2.05              |
| 8   | 72                 | 80                      | 2.09              |
| 9   | 48                 | 40                      | 3.5               |
| 10  | 48                 | 120                     | 1.6               |
| 11  | 48                 | 40                      | 3.65              |
| 12  | 48                 | 120                     | 2.86              |
| 13  | 48                 | 80                      | 3.7               |
| 14  | 48                 | 80                      | 3.85              |
| 15  | 48                 | 80                      | 3.9               |
| 16  | 48                 | 80                      | 3.7               |
| 17  | 48                 | 80                      | 3.8               |

the result was fitted with the general overview of polymerization technique which states, polymerization increases viscosity whereas degradation decreases viscosity (Ebewele, 2000). With a copper amine solution 80ml of solution and 48 hours of time there was a polymerization step and after this stage there was a degradation stage. the intrinsic viscosity can relate to degree of polymerization by calculating intrinsic viscosity from specific viscosity and concentration of solution.



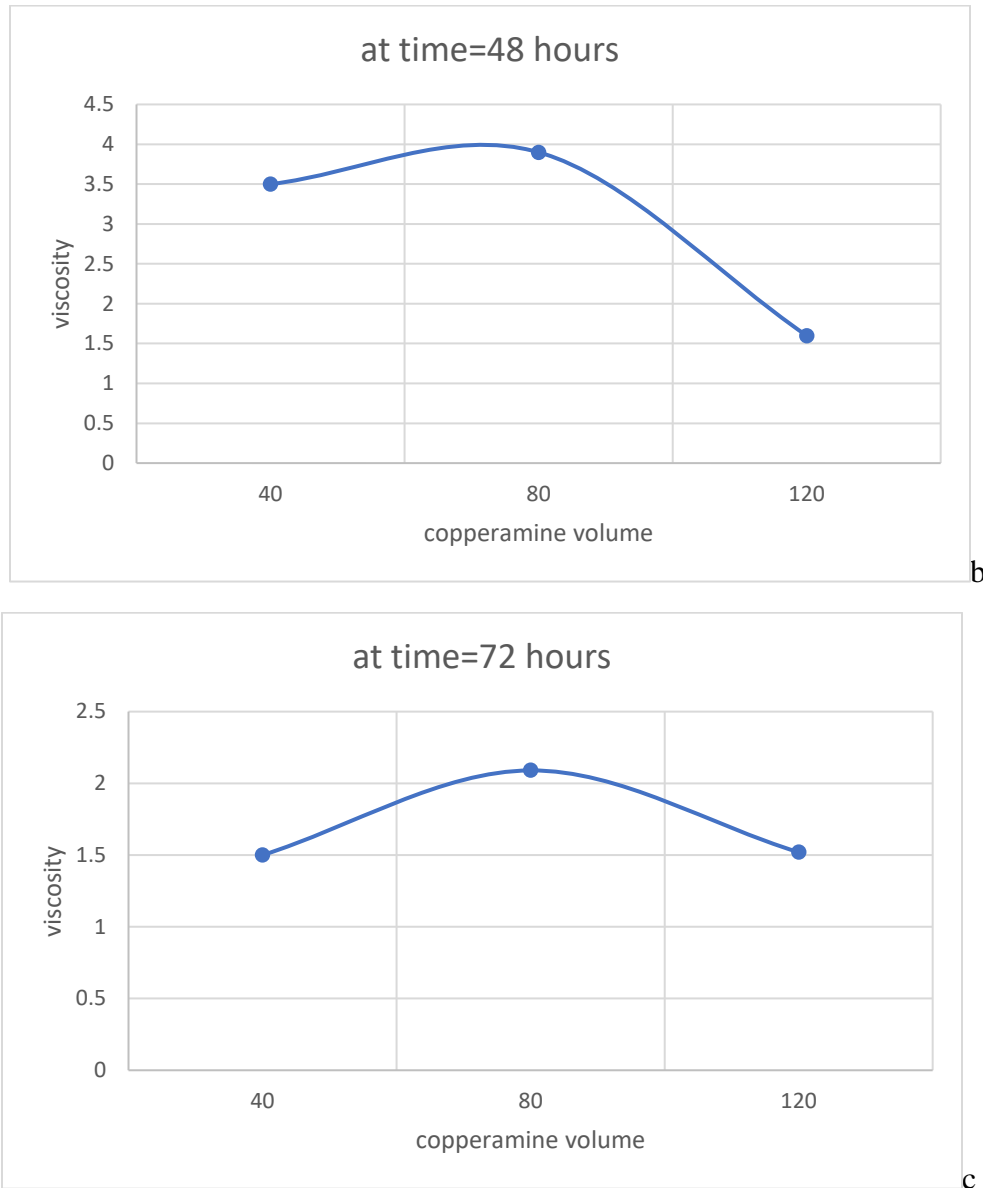


Figure 4-1 The effect of copper amine volume on the viscosity of dissolved solution at reaction time of a) 24 hours, b) 48 hours and c) 72 hours.

As shown in the figure 4.1a at a reaction time of 24 hours the viscosity was increase as the volume of copper tetra amine increase probably because of crosslinking phase didn't vanish with this time interval and needs high volume of copper amine solution for minimum reaction time. And fig 4.1b shows effect copper amine at 48 hours reaction time which drives the better viscosity at a volume of 80ml copper amine solution. The result shown above describes almost crosslinking reaction was

followed by depolymerization step after a reaction time of 48 hours with a reaction volume 80ml. fig 4.1c shows the decrement of viscosity compared to reaction time of 48 hours.

## **4.2. Statistical Analysis on Factors Affecting for the Production of Regenerated Fiber**

regenerated fiber production is affected by different treating parameters including types of fiber, types of compound used for treatment, time of treating, amount of treating compound and concentration of regenerating compound concentration. The experimental design selected for this study was response surface method and the response measured was the water absorption of regenerated fiber and yield of this product. The three variables studied were volume of Copper amine with mass of fiber, time for cross linking and concentration of sulfuric acid for regeneration. The Design Expert 6.0.8 and 7.0.0 software was used in analysis of variance (ANOVA). The actual experimental data including water absorption and yield of regenerated fiber at different parameters was actually recorded in lab and given in appendix B. the responses recorded was used to develop a mathematical model that correlates the fiber property to the treating process variables. The model fit summary for responses water absorption and yield with statistical analysis of the ANOVA are given in appendix C.

### **4.2.1. Development of Empirical Model**

Empirical models for the output responses of water absorption and yield of regenerated fiber in terms of the crosslinking and regenerating parameters in actual and coded factors were developed by using Box Behnken Design methodology. The sequential model Sum of Squares, Lack of Fit Tests and Model Summary Statistics are given at Appendix C and was found that quadratic model was the most suitable model for the present study, because quadratic model had high R-squared, adjusted R-squared and predicted R-squared for the two responses.

### **4.2.2. Model Adequacy**

It is essential to confirm whether the fitted model provides an adequate approximation of actual values or not. The model was tested for adequacy by analysis of variance. The regression model was found to be highly significant with the correlation coefficients of determination of R Squared (R), adjusted R-Squared and predicted R-Squared having a value of 0.9847, 0.9651 and 0.8492, respectively for water absorption and 0.9922,0.9821 and 0.9201 for response yield as indicated in

appendix B. The quality of the model developed could be evaluated from their coefficients of correlation. The value of R-squared for the developed correlation is 0.9847 and 0.9922 for water absorption and yield respectively: which implies that 98.47% and 99.22 of the total variation in water absorption and yield of fiber and is attributed to the experimental variables studied.

The "Pred R-Squared" of 0.8492 is in reasonable agreement with the "Adj R-Squared" of 0.9651 for water absorption and The "Pred R-Squared" of 0.9204 is in reasonable agreement with the "Adj R-Squared" of 0.9821 for yield of regenerated fiber.

"Adeq Precision" measures the signal to noise ratio. A ratio greater than 4 is desirable. a ratio of 20.885 indicates an adequate signal for the model on water absorption. And a ratio of 29.665 indicates an adequate signal of the model on yield response of fiber. Those two models can be used to navigate the design space.

The adequacy of the model was further checked with analysis of variance (ANOVA) as shown at the end of appendix B and the graph of the predicted values obtained using the developed correlation versus actual values is also shown in Figure 4.2.

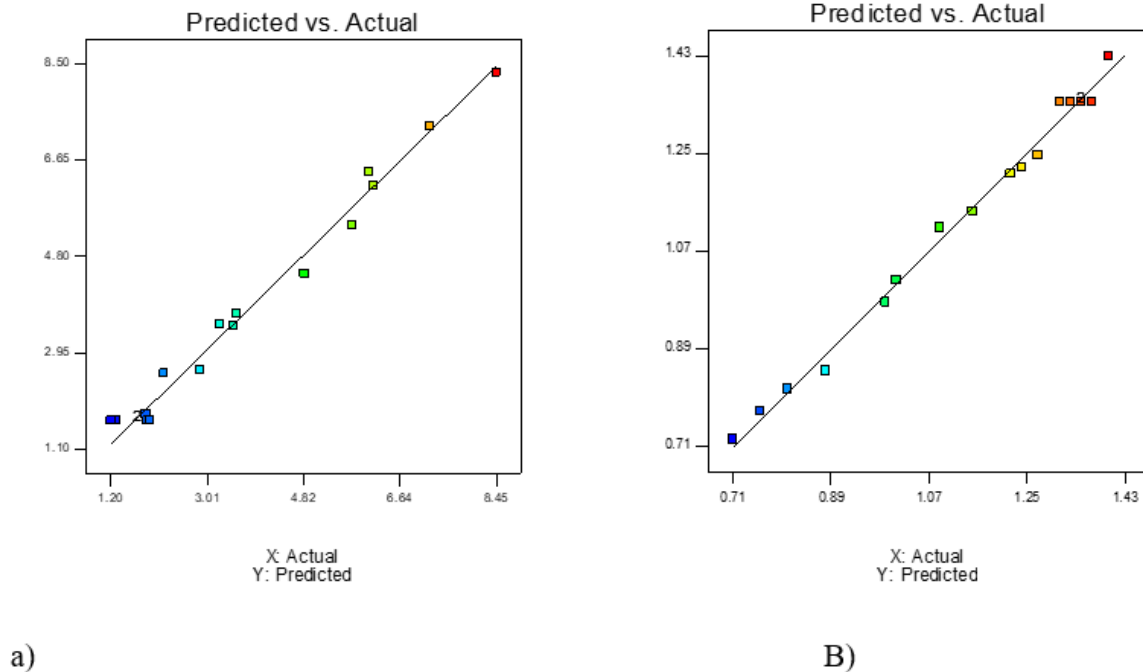


Figure 4-2 predicted vs actual plot (a) % water absorption and (b) yield

This plot represents the line of perfect fit with points corresponding to zero error between predicted values and actual values and demonstrated that the regression model equation provided accurate description of the experimental data, in which all the points are close to the line of perfect fit. This result indicates that it was successful in capturing the correlation between the three treatment variables for both the water absorption and yield of regenerated fiber.

As shown Table 4.3 below, values of "Prob > F" less than 0.0500 indicates model terms are significant. In this case A, B, C, A<sup>2</sup>, B<sup>2</sup>, C<sup>2</sup>, AC, BC are significant model terms. This shows that the time, volume of Copper amine, regenerating concentration, interaction between time and regenerating concentration and interaction between volume of copper amine and regenerating concentration affects the response water absorption much significantly and A, B, C, A<sup>2</sup>, B<sup>2</sup>, C<sup>2</sup>, BC for response yield where each designation is the same to the above. Each treatment variable effects and their interaction effects on water absorption and yield of regenerated fiber are discussed with a help of figures in the subsequent section.

Table 4-2 Analysis of Variance (ANOVA) for the Regression Model Equation and Coefficients for response water absorption.

| Source                   | Sum of Squares | DF | Mean Square | F Value | Prob > F | Remark          |
|--------------------------|----------------|----|-------------|---------|----------|-----------------|
| Model                    | 78.71          | 9  | 8.75        | 50.15   | < 0.0001 | Significant     |
| A: time                  | 28.01          | 1  | 28.01       | 160.62  | < 0.0001 | >>              |
| B: Copper amine volume   | 6.35           | 1  | 6.35        | 36.44   | 0.0005   | >>              |
| C: conc. Of regenerating | 1.62           | 1  | 1.62        | 9.29    | 0.0186   | >>              |
| A2                       | 20.38          | 1  | 20.38       | 116.88  | < 0.0001 | >>              |
| B2                       | 5.52           | 1  | 5.52        | 31.67   | 0.0008   | >>              |
| C2                       | 5.55           | 1  | 5.55        | 31.8    | 0.0008   | >>              |
| AB                       | 0.84           | 1  | 0.84        | 4.8     | 0.0646   | Not significant |
| AC                       | 4.16           | 1  | 4.16        | 23.86   | 0.0018   | Significant     |
| BC                       | 3.1            | 1  | 3.1         | 17.76   | 0.004    | >>              |
| Residual                 | 1.22           | 7  | 0.17        |         |          |                 |
| Lack of Fit              | 0.7            | 3  | 0.23        | 1.81    | 0.2855   | not significant |
| Pure Error               | 0.52           | 4  | 0.13        |         |          |                 |
| Cor Total                | 79.93          | 16 |             |         |          |                 |

As described in design expert, values greater than 0.1000 indicate the model terms are not significant. This shows that the interaction between treating time and Copper amine volume did not affect the water absorption property of regenerated cellulosic fiber. The "Lack of Fit F-value" of 1.81 implies the Lack of Fit is not significant relative to the pure error. There is a 28.55% chance that a "Lack of Fit F-value" this large could occur due to noise. Not significant lack of fit is good because we want the model to fit. Therefore, from above results the developed second order quadratic model provides an adequate approximation of the actual values.

*Table 4-3 Analysis of Variance (ANOVA) for the Regression Model Equation and Coefficients for response yield.*

| Source                 | Sum of squares | DF | Mean square | F value | Prob > F | Remark          |
|------------------------|----------------|----|-------------|---------|----------|-----------------|
| Model                  | 0.86           | 9  | 0.095       | 98.71   | < 0.0001 | Significant     |
| A: time                | 0.3            | 1  | 0.3         | 311.67  | < 0.0001 | >>              |
| B: copper amine volume | 7.20E-03       | 1  | 7.20E-03    | 7.47    | 0.0292   | >>              |
| C: conc. Regener.      | 0.074          | 1  | 0.074       | 76.91   | < 0.0001 | >>              |
| A <sup>2</sup>         | 0.062          | 1  | 0.062       | 64.77   | < 0.0001 | >>              |
| B <sup>2</sup>         | 0.23           | 1  | 0.23        | 239.78  | < 0.0001 | >>              |
| C <sup>2</sup>         | 0.039          | 1  | 0.039       | 40.9    | 0.0004   | >>              |
| AB                     | 4.00E-04       | 1  | 4.00E-04    | 0.42    | 0.5399   | Not significant |
| AC                     | 6.25E-04       | 1  | 6.25E-04    | 0.65    | 0.4471   | >>              |
| BC                     | 0.11           | 1  | 0.11        | 113.02  | < 0.0001 | Significant     |
| Residual               | 6.75E-03       | 7  | 9.64E-04    |         |          |                 |
| Lack of Fit            | 4.03E-03       | 3  | 1.34E-03    | 1.97    | 0.2602   | not significant |
| Pure Error             | 2.72E-03       | 4  | 6.80E-04    |         |          |                 |
| Cor Total              | 0.86           | 16 |             |         |          |                 |

The Model F-value of 98.71 implies the model is significant. There is only a 0.01% chance that a "Model F-Value" this large could occur due to noise. Values of "Prob > F" less than 0.0500 indicate model terms are significant. In this case A: time, B: Copper amine volume, C: regenerating concentration, A<sup>2</sup>, B<sup>2</sup>, C<sup>2</sup>, BC: Copper amine volume to regenerating concentration are significant model terms. Values greater than 0.1000 indicate the model terms are not significant. In this case AB: interaction b/n time and Copper amine volume and AC: interaction b/n time and regenerating

concentration had not significant effect on the response yield. The "Lack of Fit F-value" of 1.97 implies the Lack of Fit is not significant relative to the pure error. There is a 26.02% chance that a "Lack of Fit F-value" this large could occur due to noise. Non-significant lack of fit is good, because we want the model to fit.

Therefore, from above results the developed second order quadratic model provides an adequate approximation of the actual values on response yield too.

#### 4.2.2.1. Residual Plots for Percentage Absorption

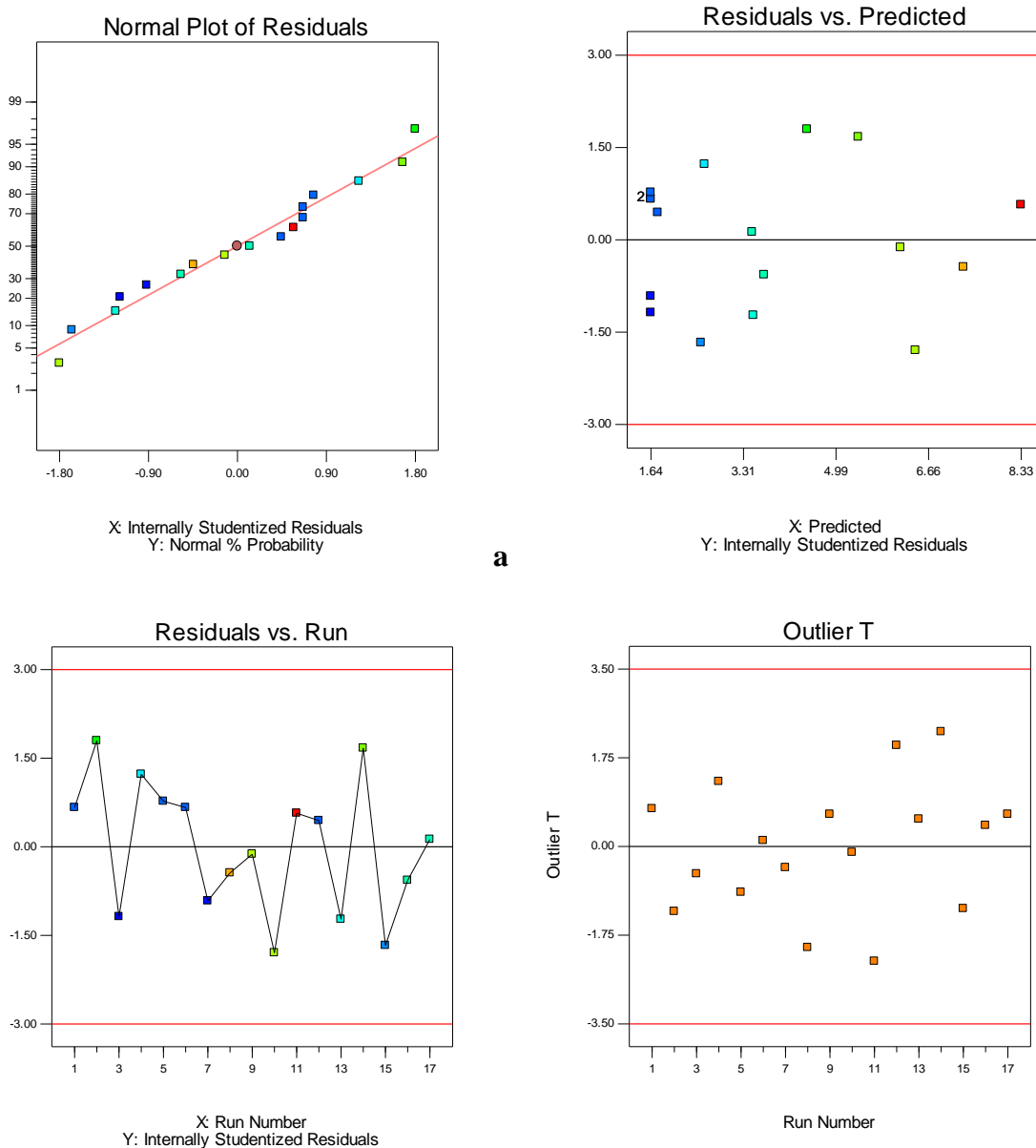


Figure 4-2 residual plots, a) normal plot of residues and residual vs predicted, b) residual vs run and outlier T

As we have seen both the graphs 4.3 above and 4.4 below i.e. normal plot of residues, residual vs predicted, residual vs run and outlier T, tells the validity of the model with fitted data. So, for both responses; the normal probability plot shows each data are fitted to straight line implies that the responses water absorption and yield of regenerated cellulosic fiber gotten from experimental data agreed with the assumption of regression models is that the error is normally distributed with a mean of zero. That is why the data are normally distributed and make straight along a line.

The residual vs predicted plot shows that no regular shape b/n the experimental data and the predicted value which shows that the data taken was collected randomly in the experiment.

Residual vs run order plot is useful however only if data have been collected in randomized run order or some other order that is not increasing or decreasing in any of the predictor variables used in the model. If the data have been collected in a time order that is increasing or decreasing predictor variables then any drift in the process may not be able to be separated from the functional relationship b/n the predictor and responses this is why randomization is emphasized in experiment design. And the residual vs run order graph of yield below there no functional relationship b/n the predictor with percentage absorption and yield which tells as the experimental data of yield was done in randomized order.

Outliers T vs run number graph tells the bad data in the experiments. Whether the data have been done in the appropriate condition or not. As shown below in the graph there is no any point gone out from the boundary limit, this shows that there is no bad data which needs to be repeated.

### 4.2.2.2. Residual Plots for Yield

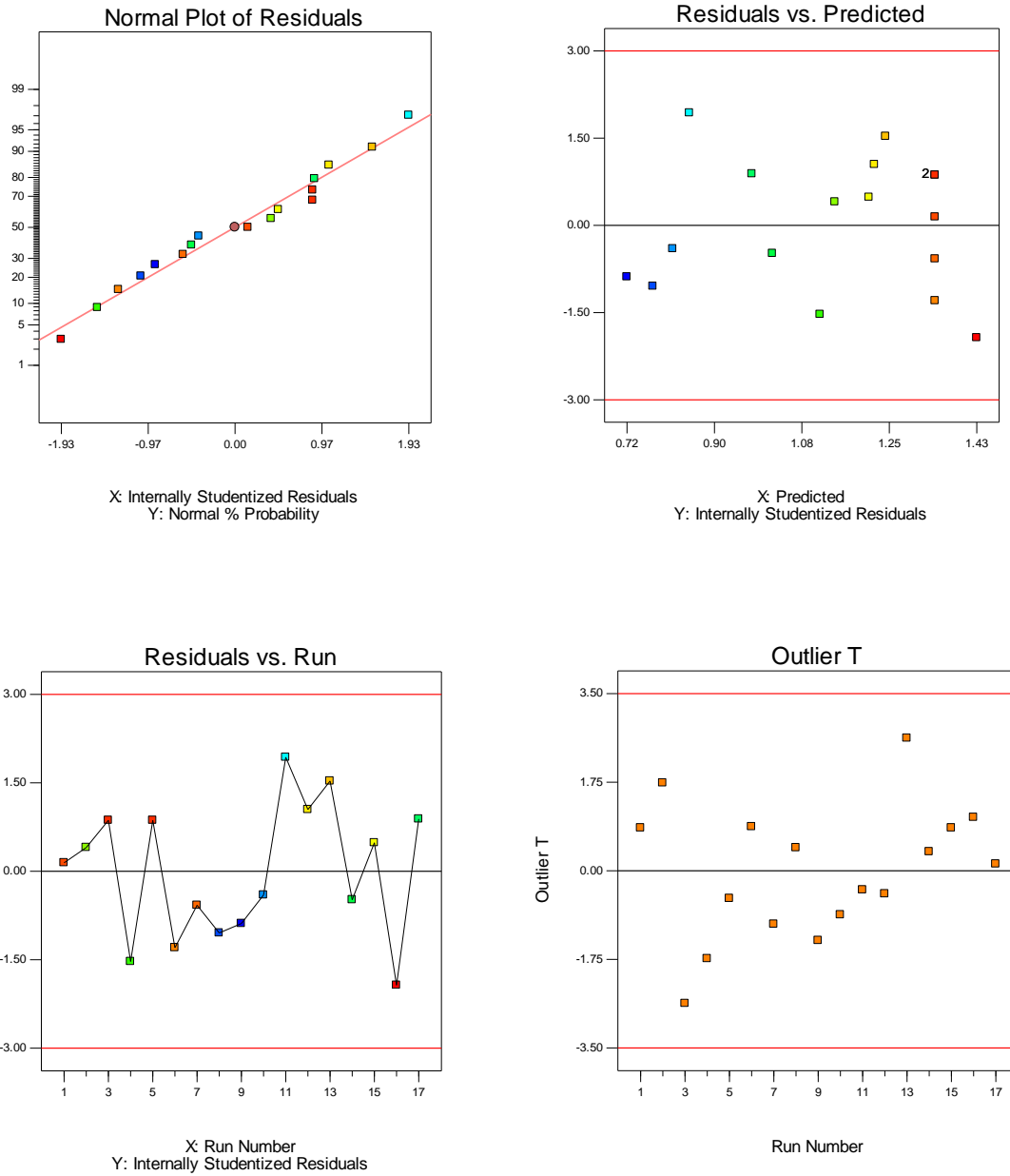


Figure 4-3 . residual plots for response yield

### **4.3. Effect of Treating Variables on Preparation of Regenerated Fiber**

Response surface design methodology was used to estimate the effect of three treating variables on the percentage water absorption of fiber and yield of fiber. interaction plots were drawn by using Box Behnken Design to investigate the effect of all the factors on the responses. Based on the analysis of variance, the treating process was significantly affected by various interactions between the process variables. On the other hand, individual process variables that affect the water absorption and yield of fiber significantly are treating time, volume to mass interaction and concentration of regenerating.

#### **4.3.1. Direct Effects of Treatment Process Variables (Time in Hours, Copper Amine Volume in ml And H<sub>2</sub>SO<sub>4</sub> Conc in M) on Water Absorption**

##### **4.3.1.1. Effect of Time on Water Absorption of Regenerated Cellulosic Fiber**

Treatment time is one of the major effect in cross linking reaction. Because in this reaction there are two major phases which have affected by treatment time; these phases are polymerization phase and degradation phase (Ebewele, 2000). In polymerization phase cellulose is crosslink with its cellulose unit and also with amine group. During this stage hydrophilic structure of cellulose was changed into hydrophobic structure because of OH group in the cellulose was substituted by amine structure (Hearle & Woodings, 2001). As shown in the graph below the hydrophobicity of the regenerated cellulosic fiber is increased (hydrophilicity is decreased) as the time of treating is increased up to 53 and around hours. And almost constant up to 58 hours and hydrophobicity decreased but hydrophilicity increased with the next time intervals as shown below. This shows that polymerization phase is up to 53 hours and the second phase or degradation was started with after 60 hours (fig4.5). So, the optimum water absorption capacity is around 53 hours which have around 1.2 absorption capacity and almost constant up to 60 hours and increased with the next time intervals.

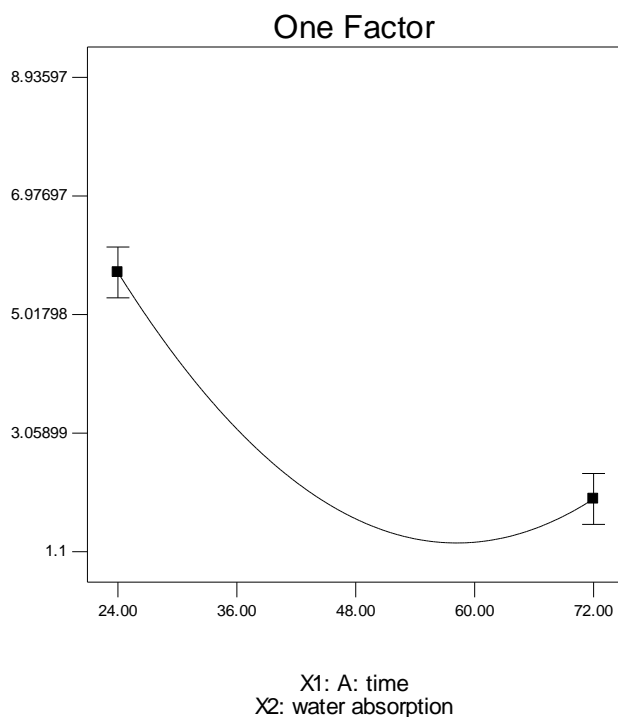
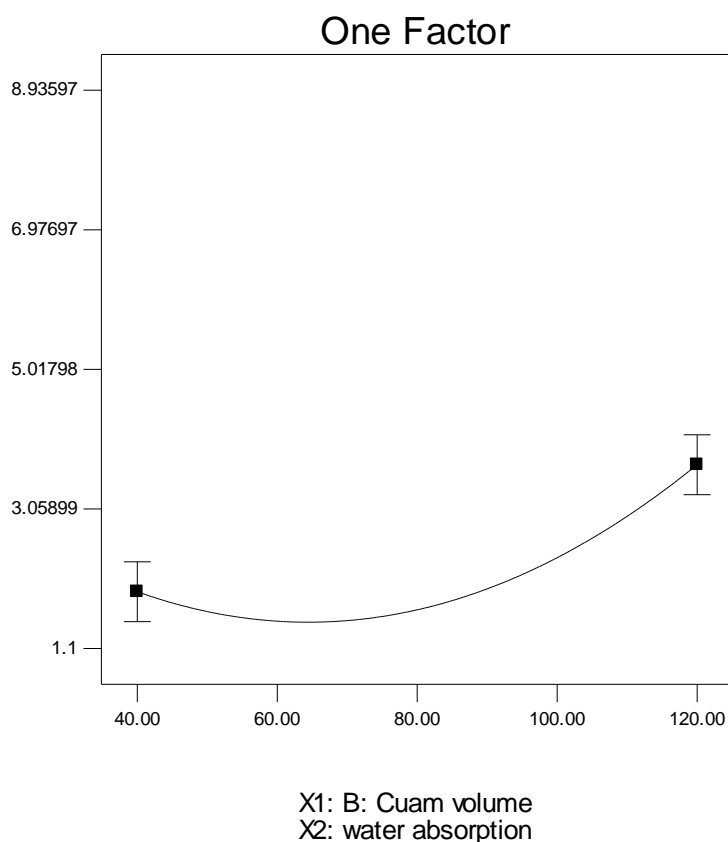


Figure 4-4 effect of time on water absorption of regenerated cellulosic fiber

#### 4.3.1.2. Effect of Copper Amine Volume (volume of Schweizer's reagent) on Water Absorption of Regenerated Cellulosic Fiber

The effect of copper amine volume is inter-related with mass of fiber with volume mass transfer and as the same time with chemical reaction. Since the mass of fiber in the process is constant, the mass to volume ratio is affected by the volume of the copper amine reagent. During the first stage the mass to volume ratio is most probably fitted to polymerization process. For the gap between 40ml and 50ml the water absorption capacity was decreased. And the volume of copper amine goes to 75ml and around the water absorption is almost constant(fig4.6). This was happened because of lack of high crosslinking b/n cellulose and copper amine agent when the volume is much less and as the same condition when the volume is much more the degradation step is faster than polymerization step. That is why water absorption capacity is high at initial and final stage. The optimum water absorption capacity was obtained around 60 to 77ml of Schweizer's reagent.



*Figure 4-5 effect of Copper amine volume on water absorption of regenerated cellulosic fiber*

#### **4.3.1.3. Effect of Regenerating Concentration ( $H_2SO_4$ ) on Water Absorption of Regenerated Cellulosic Fiber**

Regenerating concentration is the other factor which affect the water absorption properties of regenerated fiber. As described through methodology, the production of regenerated cellulosic fiber proceeds two steps, one is polymerization and the other is regenerating, in the regenerating step the viscos solution that loss its crystallinity changed to its structure and the excess copper amine solution is also reacted with sulfuric acid. as shown below in the graph as the concentration of acid increased the percentage water absorption decreased up to 1M, and constant up to 1.2M and again increased up to 1.5M (fig4.7). during the first phase, since sulfuric acid is very dilute acid lacks fast reaction with copper solution to separate completely the regenerated fiber. And in the last stage the acid itself may proceeds hydrolysis with fiber and percentage absorption is increased.

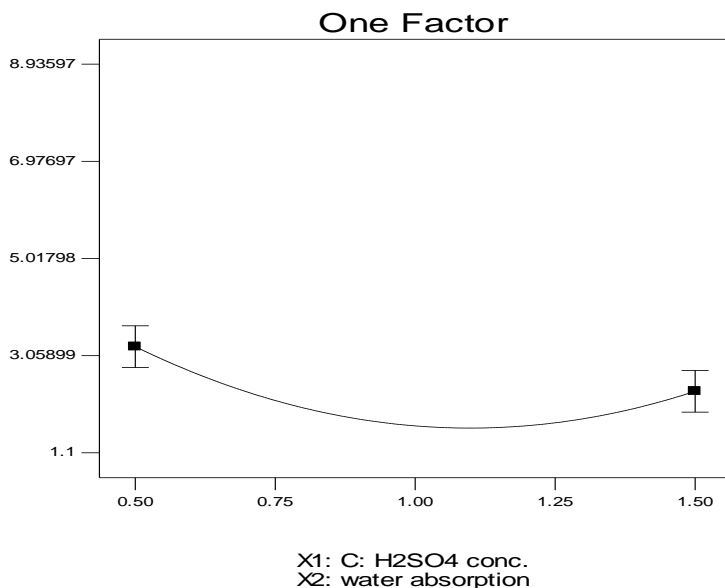


Figure 4-6 effect of  $H_2SO_4$  concentration on water absorption of regenerated cellulosic fiber

#### 4.3.1.4. Interaction Effect b/n Time of Treating and Concentration of Regenerating $H_2SO_4$ On Water Absorption of Regenerated Cellulosic Fiber

As shown below in the fig4.8 of interaction between time and concentration of acid, during the first stage both minimum concentration and maximum concentration of acid has similar effect with increasing time up to 55 hours to go. After that the two concentrations effect on water absorption cross each other. With 1.5 M concentration of acid the optimum percentage water absorption was obtained around a treatment time of 48 hours whereas at a concentration of 0.5 M the optimum percentage water absorption was obtained around a treatment time of 62 hours. And as shown in the interaction plot with combination of treatment time and regenerating concentration the optimum value of percentage water absorption was obtained around a treatment time 48 hour and a regenerating concentration of 1.1M.

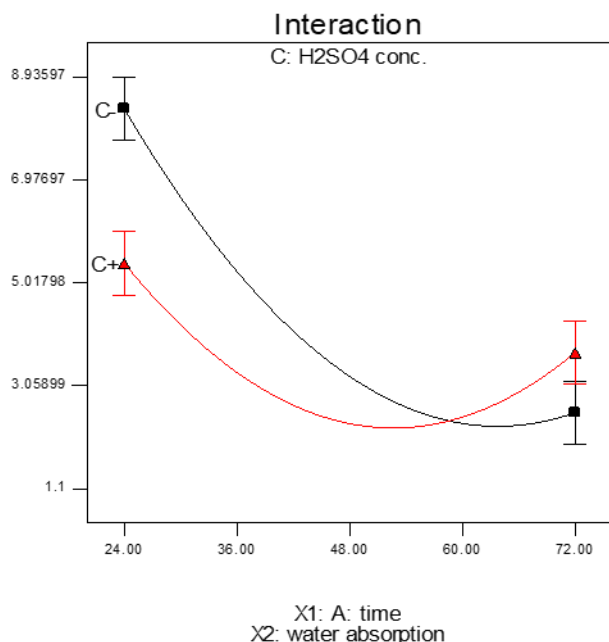


Figure 4-7 time and H<sub>2</sub>SO<sub>4</sub> concentration Interaction graph on water absorption

#### 4.3.1.5. Interaction Effect B/N Volume of Copper Amine and Concentration of Regenerating Compound(H<sub>2</sub>SO<sub>4</sub>) on Water Absorption

The other interaction effect on the water absorption capacity is volume of copper amine solution to concentration of regenerating compound. As we have seen before in chapter three, after the polymerization step proceeds, the regenerating step continues to get regenerating fiber. It is better to be minimum volume of copper amine solution for simple regeneration of fiber. But when the volume is much little the copper amine was not substitute the hydroxyl group perfectly and that is why the minimum percentage water absorption was not obtained at 40ml of copper amine solution in both minimum and maximum concentration of regenerating agent. The minimum percentage absorption was obtained at 80 ml of copper tetra amine solution and at 1 M concentration solution of sulfuric acid as shown in interaction plot fig 4.9.

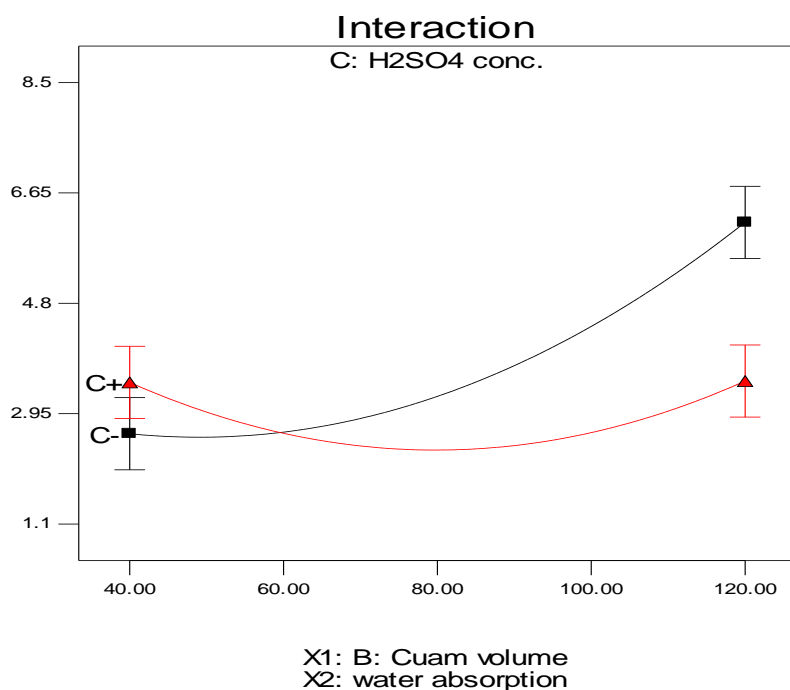


Figure 4-8 copper amine volume and H2SO4 concentration interaction graph effect on water absorption

### 4.3.2. Direct Effects of Treatment Process Variables on Yield of Regenerated Cellulosic Fiber

#### 4.3.2.1. Effect of Time

Corresponding to response yield, Treatment time is one of the major effect. Because in this reaction there are two major phases which have affected by treatment time; these phases are polymerization phase and degradation phase. In polymerization phase cellulose is crosslink with its cellulose unit and also with copper amine group. After the polymerization step there is degradation step. So, to get high yield we must stop after polymerization step facilitated and before the degradation step started. As shown in the graph 4.10 below the yield of the regenerated fiber is increased as the time of treating is increased up to approximately 65 hours as shown in fig4.10. And almost constant up to approximately 69 hours and yield of regenerated fiber was decreased with the next 2 or 3 hours of time intervals as shown below. So, the optimum yield of regenerated fiber was at a time around 65 hours which have around 1.4gram yield.

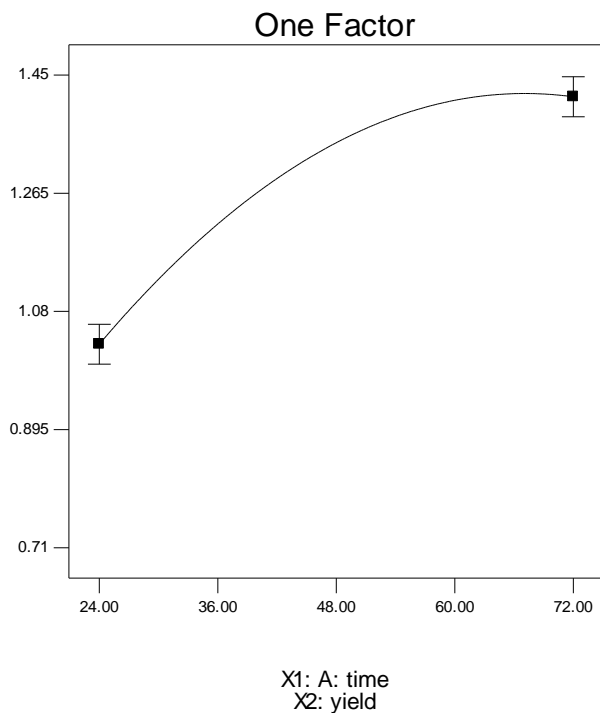
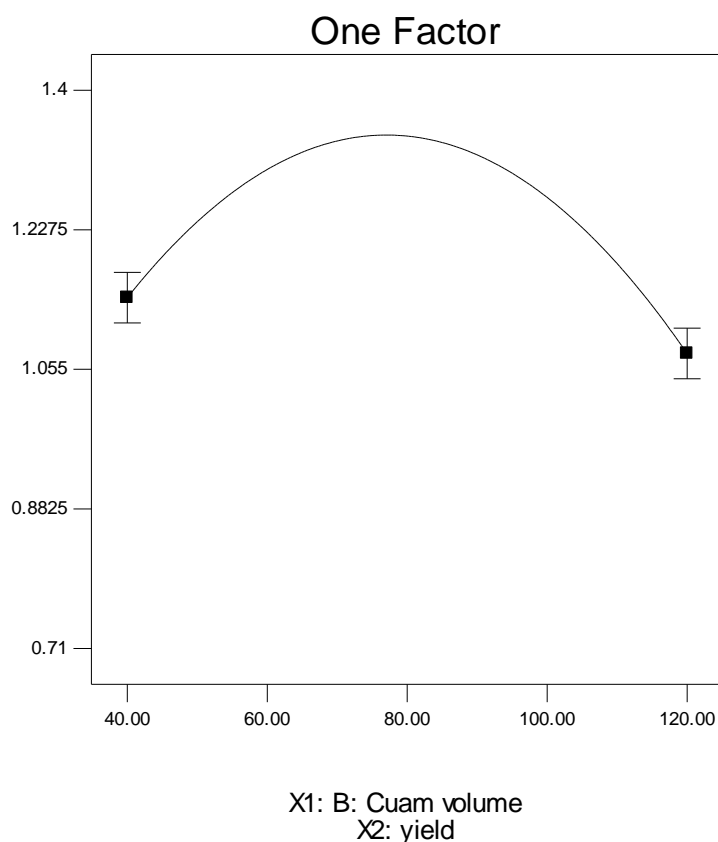


Figure 4-9 effect of time on yield of regenerated cellulosic fiber

#### 4.3.2.2. Effect of Copper Amine Volume

As described in 4.5.5.2. the volume of copper amine solution affects not only water absorption but also the yield of regenerated fiber. The yield of regenerated fiber increased as the volume increased to 80 ml. and constant for 2 or 3 ml difference. After that the yield of regenerated fiber was decreased with increasing volume. In this case the maximum yield was obtained at a volume of 80 ml of copper amine solution (fig 4.11). The reason at which high volume affects yield negatively is probably the degradation phase is dominate the polymerization step and also with high volume it is difficult to regenerate crystalline fiber.



*Figure 4-10 effect of copper amine volume on yield of regenerated cellulosic fiber*

#### **4.3.2.3. Effect of Regenerating Concentration**

Regenerating concentration is also the other factor which affect the yield of regenerated fiber. as shown below in the graph as the concentration of acid increased the yield of regenerated fiber also increases up to 1.1M and constant up to 1.3M and again decreased up to 1.5M (fig4.12). during the first phase acid was not separate completely the crosslinked and the copper amine solution. And in the last stage the acid itself proceeds hydrolysis with fiber and there was no complete separation, so, the yield was decreased.

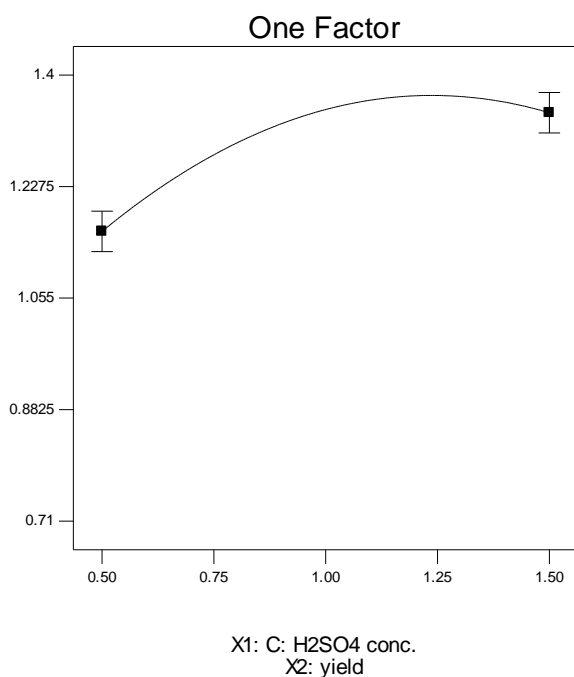


Figure 4-11 effect of H<sub>2</sub>SO<sub>4</sub> concentration on yield of regenerated cellulosic fiber

#### 4.3.2.4. Interaction Effect b/n Copper Amine Volume and Concentration of Regenerating Compound

The only interaction effect on the yield of regenerated fiber is volume of copper amine solution to concentration of regenerating compound. It is better to be minimum volume of copper amine solution for simple regeneration of fiber. But when the volume is much little the copper amine was not proceeding a crosslinking reaction with cellulosic mercerized fiber perfectly. and that is why the minimum amount of yield was obtained at 40ml of copper amine solution in both minimum and maximum concentration of regenerating agent. The maximum yield of regenerated fiber was obtained at 80ml of copper tetra amine solution and at 1M concentration solution of sulfuric acid as shown in interaction graph(fig4.13). And after that the yield of regenerated fiber was decreased very fast when the interaction becomes high volume and low concentration, because degradation step is dominated the polymerization step and as the same manner low concentration of acid lacks to regenerate and some solution of fiber was lost with copper amine solution.

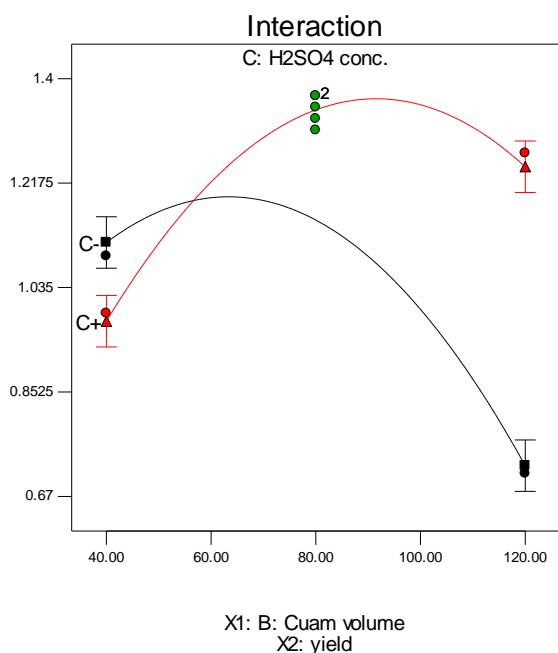


Figure 4-12 copper amine volume and H<sub>2</sub>SO<sub>4</sub> con. interaction effect graph

#### 4.4. Optimization of Process Treatment Variables for Two Responses

The results above have shown that the three process variables and the interaction among the parameters affect the percentage water absorption and yield of regenerated cellulosic fiber. Therefore, the next step is optimizing the process variables in order to obtain the highest yield and the lowest percentage water absorption using the developed model regression. In optimizing the enhancing fiber process; the cross-linking time, copper amine volume and regenerating concentration are a set of process variables held to be "in range" while percentage water absorption and yield are set of responses that need to be "minimized" and "maximized" respectively.

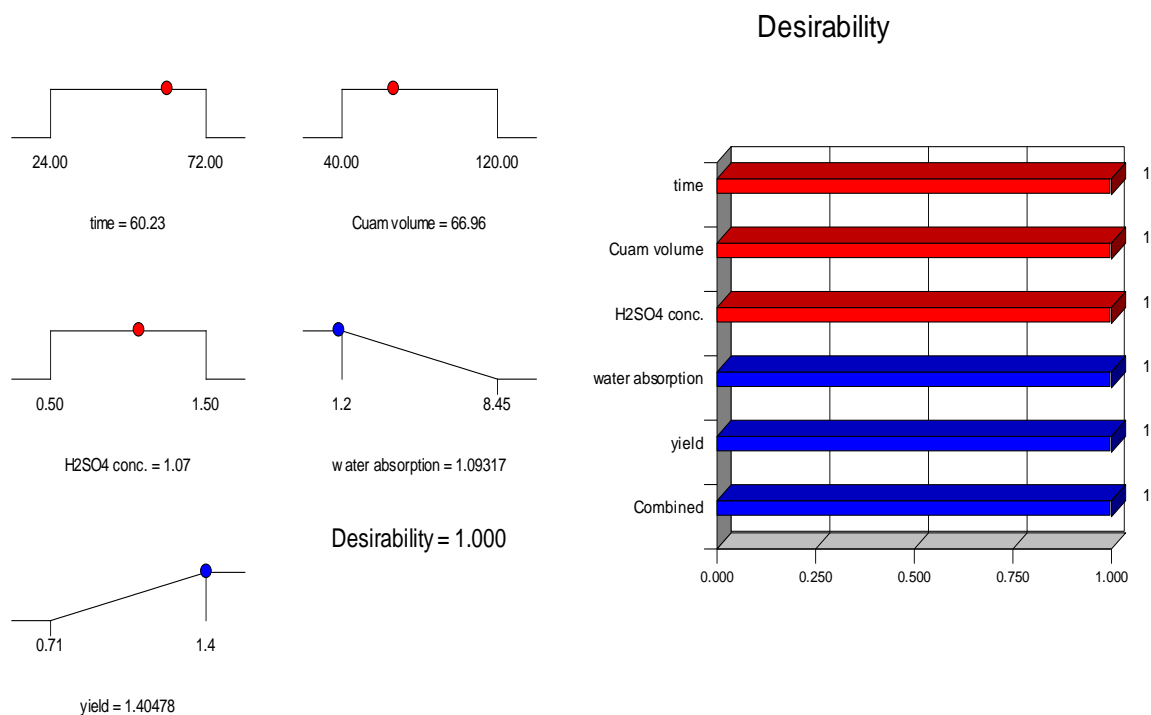


Figure 4-13 desirability for optimization of factors and responses of regenerated cellulosic fiber

As shown fig4.14 the optimal value above the desirability of the expert factors and levels was 100% for both responses for each factor and reasonable related to the predicted desirability of 1. And the optimum values are; the time taken at 60.23 hour, the volume taken was at 66.96ml of copper amine solution and a regenerating concentration of 1.07M. And in general, the optimal value of responses at the above optimal factors was 1.09 and 1.4gram for water absorption capacity and yield respectively.

#### 4.5. Validation of Developed Model

To confirm the developed model, the actual validation experiment was carried out at optimum conditions that was obtained in design expert. These optimum points were treatment time of 60.23hours, Copper amine volume of 66.96ml and regenerating concentration of 1.07M. For validation, actual experimental value, predicted value and the percentage of error is required and listed in Table 4-5 below. As observed from confirmation result in table4.5, the percentage accuracy 0.973 (97.3%) is higher than predicted R-Squared value of 96.51% for water absorption and 98.6% is greater or equal to 98.21% as indicated in appendix C. Therefore, the developed model is much accurate as the percentage of accuracy is higher than predicted R-Squared value.

Table 4-4 checking of model validation

|                  | Treatment time | Copper amine volume | Regenerating concentration | Actual value | Predicted value | % error | % Accuracy |
|------------------|----------------|---------------------|----------------------------|--------------|-----------------|---------|------------|
|                  | 60.23 hrs      | 66.96ml             | 1.07M                      |              |                 |         |            |
| Water absorption |                |                     |                            | 1.12         | 1.09            | 2.7     | 97.3       |
| Yield            |                |                     |                            | 1.38         | 1.4             | 1.4     | 98.6       |

#### 4.6. Crystallinity

The crystallinity of the regenerated fiber was calculated using eqn. 3.6 using density of two reference polymers, one complete crystalline and the other is complete amorphous. For complete crystalline poly Ethylene was taken with density of 0.9g/cm<sup>3</sup> and for complete amorphous polystyrene with density of 1gm/cm<sup>3</sup> was taken. The density of regenerated fiber was measured by Archimedes principle and it was around 1.4-1.5g/cm<sup>3</sup>. After calculation was proceed the percent crystallinity of regenerated fiber becomes about 72 percent. This result tells really what we have seen from the texture of the product.

#### 4.7. Fourier Transform Infrared Radiation(FTIR)

Spectrum 65 FTIR (PerkinElmer) was applied in the range 4000-400 cm<sup>-1</sup> using KBr pellets. Natural fiber powder samples of each species of small mg were dispersed in a matrix of KBr, followed by compression to form pellets. The sample collection was in the range of 4000 to 400 cm<sup>-1</sup>, at a resolution of 4 cm<sup>-1</sup>. Three samples such that; raw fiber, mercerized fiber and regenerated fiber measurements were evaluated, and shown at the graph below. The figure 4.2 below shows the FTIR spectra of the untreated pseudo-stem banana fiber, mercerized and regenerated fiber as shown in fig4.15. The absorptions bands at 3600-3100 cm<sup>-1</sup> can be assigned to stretching vibrations and other polymeric associations of hydroxyl groups and this result is fitted with in “Helena Becker” result for raw fiber of absorption band is b/n 3600-3100(Becker et al., 2013),(Fan *et al.*, 2012). Symmetric stretching at 3000 cm<sup>-1</sup> for raw and mercerized fiber, and around 3300 for regenerated fiber assigned to the present of CH<sub>2</sub> groups. Angular deformations of C–H linkages of aromatic groups were observed at 858, 761, 668 and 576 cm<sup>-1</sup>. An overlapping of peaks was observed between 1700–1125 cm<sup>-1</sup> due to C–C, C=C, OH, CO, CH<sub>n</sub>, CH, and C–O–C vibrations for raw fiber and mercerized fiber. in addition to these vibrations, the amine to hydrogen and to carbon vibration probably analyzed for regenerated fiber between 1700-1600 and 1150-1000 cm<sup>-1</sup> (Yang, Dou, Liang, & Shen, 2005).

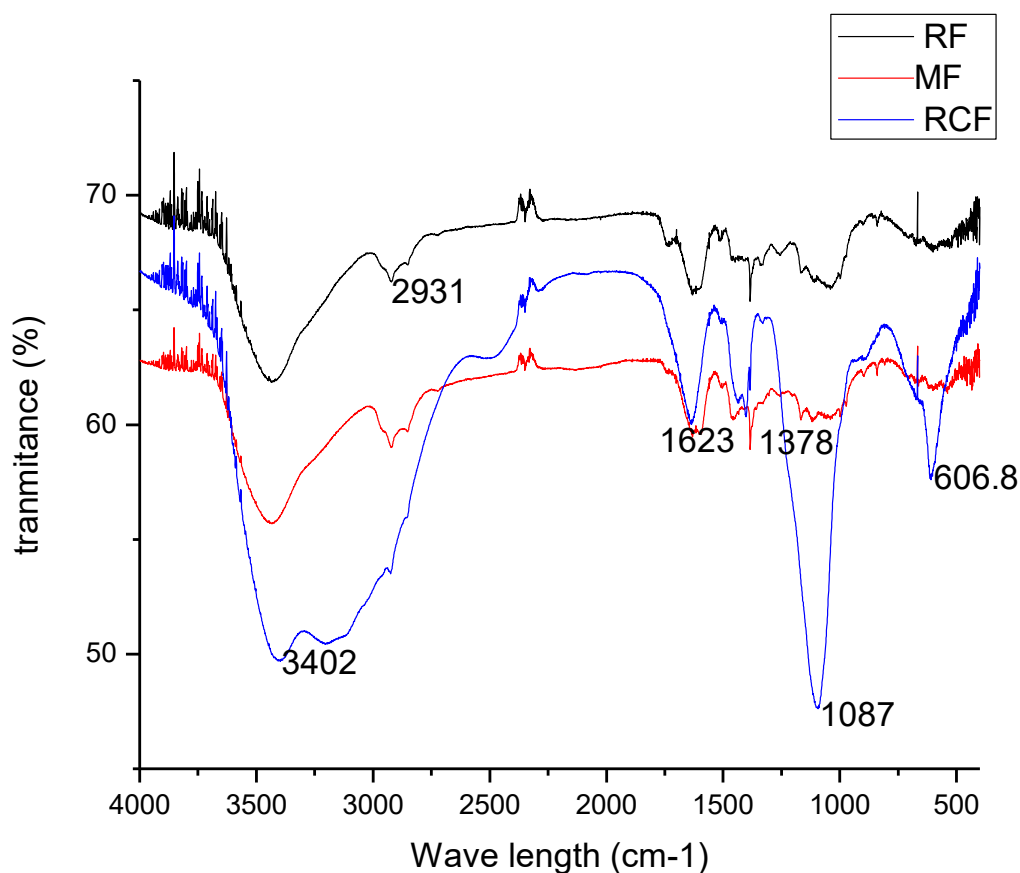


Figure 4-14 FTIR analysis graph for raw mercerized and regenerated fiber.

#### 4.8. Thermo Gravimetric Analysis(TGA)

Thermogravimetric analysis of banana regenerated fiber specimens was analyzed to know the degradation characteristics with specified temperature as shown in fig4.16. The regenerated fiber present three main weight loss regions. The initial weight loss of 10.2% in the region 25–100 °C is mainly due to moisture evaporation.

The broad peak weight loss of around 68% in the region from 200–380 °C is contributed by the degradation of cellulose takes place between 280 oC and 398 oC. This result is agreeing and more better than with B. Deepa, E. Abraham, Rekha Rose Koshy, L. A. Pothan, and Sabu Thomas results that was done on raw fiber and acid treated fiber (Barreto et al., 2010) and the 3<sup>rd</sup> phase from 400 to 700 degree centigrade 50% weight loss was due to degradation of some lignin impurities and decomposition of amine complexes (Ii, Gorbunov, Royal, & Springfield, 1974),(Xu, Xiong, Tan,

& Zhang, 2015). When we compare the thermal stability of this regenerated cellulosic fiber with cotton the values are approximate in first and second stage weight losses as Dr. Noureddine Abidi and Dr. Eric Hequet describe the thermal stability of cotton; The initial weight loss (region I) is located between 37°C and 150°C and is followed by a plateau region before the major weight loss occurs in region II, located between 225°C and 425°C (Abidi, Noureddine Noureddine Abidi , Lubbock & Hequet, 1947). And the stability of my product was a little bit better than cotton stability.

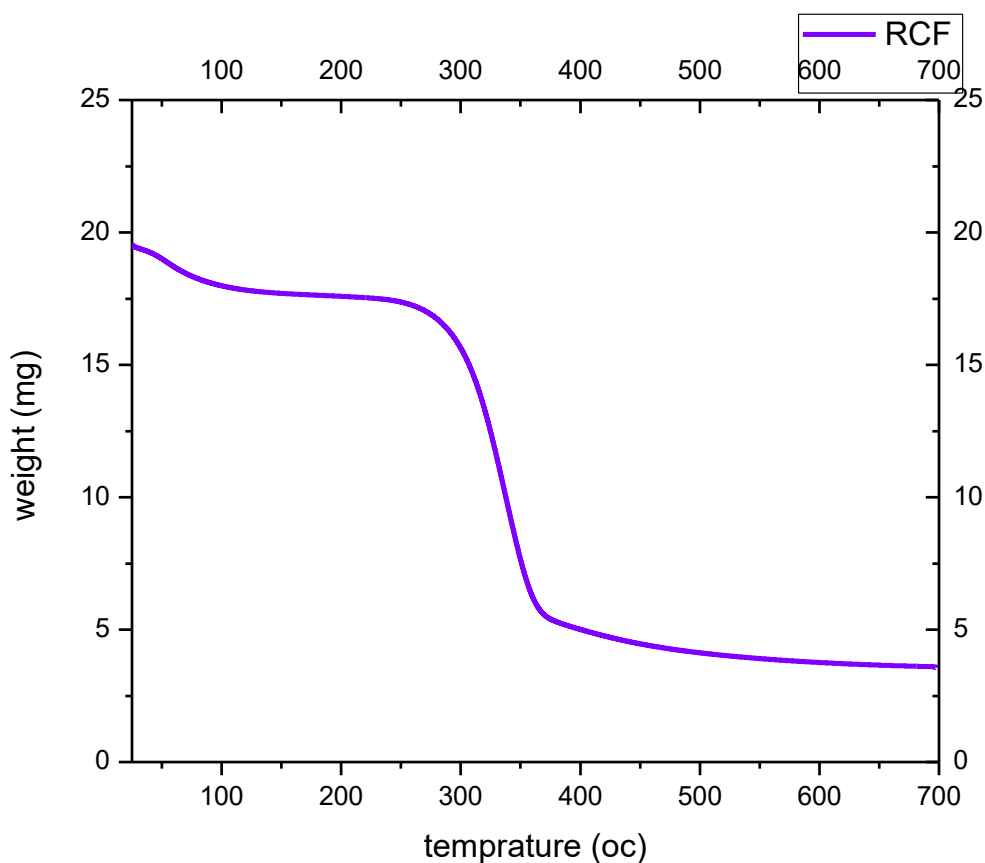


Figure 4-15 thermogravimetric analysis graph for regenerated fiber

## 5. Conclusion and Recommendation

### 5.1. Conclusion

Banana stem is a waste material after the fruit was collected in most areas of Ethiopia. This research was insight the ability to change this waste material in useful regenerated cellulosic fiber which can be used as a raw material for many industries.

In this research, the production of regenerated cellulosic fiber from banana pseudo stem was done by proceeding consecutive steps. Decortication of the stem was followed by extraction of raw fiber and then mercerization which was a pre-condition for dissolving cellulose was facilitated for production of regenerated cellulose. Three factors with three levels were used in design expert to study the effect of those factors on water absorption and yield for production regenerated cellulose fiber.

After optimum condition was found, the regenerated fiber crystallinity, density thermogravimetric analysis was characterized. Based on experimental results obtained, it is found that all the process parameters showed direct significant effect and some interaction effects on the water absorption and yield of regenerated cellulosic fiber. These effects were analyzed successfully using design experiment. Among the three variables studied, the minimum water absorption capacity of 1.09 and the maximum yield of 1.4 was suggested by design expert at optimum treatment time 60.23 hours, optimum Copper amine volume 66.96ml and optimum regenerating concentration of 1.07M. for validation this optimum condition, experiment was proceeded at these points and its percentage error was agreed to the predicted R-squared value.

Generally Crosslinking reaction was affected by reaction time and volume of Schweizer's reagent. The water absorption capacity of regenerated cellulosic fiber is less than that of raw fiber and mercerized fiber. The yield of regenerated cellulosic fiber increases to the optimum parameters and decrease elsewhere. During crosslinking, viscosity increases in polymerization step and decreases in degradation step; Density was increased from raw, mercerized to regenerated cellulosic fiber.

## 5.2. Recommendation

Depending on results investigated in this study, the following works which should be done for further works would be recommended.

- ✚ Samples should be collected from d/t directions to get the best cellulosic fiber.
- ✚ It is better to extract the fiber using machine.
- ✚ Further studies should be studied on dissolving of cellulosic fiber.
- ✚ Different chemicals can be used for dissolving and also for regenerating such as xanthate, formaldehydes and others.
- ✚ Degree of polymerization should be measured to know molecular weight of the regenerated cellulosic fiber.
- ✚ There should be a known size nozzle like instrument for regeneration purpose of viscous solution at preferable spannable sizes.
- ✚ The regenerated cellulosic fiber should be blended with cotton and other fibers to be projected in textile and other industries which used cellulose based fibers for their product.

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## Appendix

### Appendix A

#### A1. 7.5% sodium hydroxide solution preparation.

For this calculation the first thing that was taken in to account is knowing the volume of solution. And this was done by mass of fiber to volume of sodium hydroxide solution 1gm of fiber:20ml of solution as stated in methodology. So, for example the first sample was 2gm of raw fiber which leads to 40ml of solution.

$$\% \left( \frac{W}{V} \right) = \left( \frac{\text{mass of solute}}{\text{volume of solution}} \right) * 100$$

$$7.5\% = \frac{X \text{ gm NaOH}}{40 \text{ ml solution}} * 100 \quad \text{this gives that, mass of NaOH} = 3 \text{ gm}$$

This formula uses for all other samples which are done in mercerization step.

#### A2. 0.5M, 1M and 1.5M H<sub>2</sub>SO<sub>4</sub> solution preparation.

For 1M of sulfuric acid preparation

$\text{molarity} = \frac{\text{mole}}{\text{liter of solution}}$  and number of mole is mass over molecular weight which shows 100gm of H<sub>2</sub>SO<sub>4</sub> is 1mole. Density of 98% H<sub>2</sub>SO<sub>4</sub> is around 1.84 gm/ml implies that 100gm will occupy 54.3ml of concentrated sulfuric acid.

And using 1000ml elementary flask distilled water was added up to some stage first and then 54.3 ml of concentrated acid and then the distilled water was filled.

A3. Preparation of copper tetra amine solution was by reacting copper hydroxide and 25% ammonia solution. By using theoretical balancing equation 97.56 grams of Cu(OH)<sub>2</sub> was reacted with 140.18 grams of NH<sub>4</sub>OH. Since ammonia solution is liquid it is better to use its volume by converting mass to volume using density of 0.9 gm/ml. This gives 155.75ml of ammonia solution.

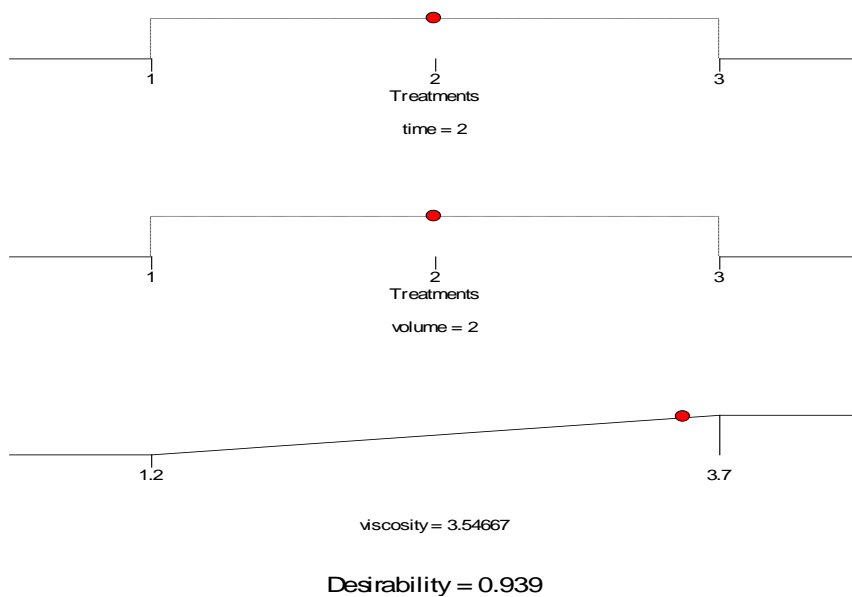
#### A4 D optimal analysis.

The D optimal experimental analysis method is used to approve the selected levels of factors or to get the optimum value for main design. This is response surface mechanism. Viscosity was taken

as a response with corresponding of factors of treatment time and volume of copper amine solution. The levels were 24, 48, and 72 hours for time and 40, 80 and 120 ml of copper amine solution.

Table A1 experimental viscosity results for D optimal analysis

| Std | Run | Factor A<br>treatment time | Factor B<br>Copperamine<br>volume | Response<br>Viscosity |
|-----|-----|----------------------------|-----------------------------------|-----------------------|
| 1   | 8   | 24                         | 40                                | 2.11                  |
| 2   | 3   | 48                         | 40                                | 3.2                   |
| 3   | 6   | 72                         | 40                                | 1.55                  |
| 4   | 7   | 24                         | 80                                | 2.3                   |
| 5   | 9   | 48                         | 80                                | 3.7                   |
| 6   | 5   | 72                         | 80                                | 2.09                  |
| 7   | 4   | 24                         | 120                               | 1.9                   |
| 8   | 2   | 48                         | 120                               | 2.5                   |
| 9   | 1   | 72                         | 120                               | 1.48                  |



As shown above in figure above the D optimal method above the desirability to do with such factors and levels was around 93.9% and reasonable related to the predicted desirability of 1. And the optimum values are the time taken at point 2 which is 48 hours and the volume taken also at point 2 which is 80 ml of copper amine solution. And in general overview viscosity is

related to my responses, such that polymerization gives increasing viscosity and degradation minimizes viscosity.

## Appendix B

### B1 experimental data

- a) Dynamic viscosity relative viscosity and specific viscosity of dissolved fiber solution before regeneration

| Std | Treatment time (Hrs) | Copperamine volume(ml) | Viscosity (Mpa.s) | Relative viscosity | Specific viscosity |
|-----|----------------------|------------------------|-------------------|--------------------|--------------------|
| 1   | 24                   | 40                     | 2.11              | 1.48               | 0.48               |
| 2   | 72                   | 40                     | 1.5               | 1.06               | 0.05               |
| 3   | 24                   | 120                    | 2.17              | 1.53               | 0.52               |
| 4   | 72                   | 120                    | 1.51              | 1.06               | 0.06               |
| 5   | 24                   | 80                     | 2.05              | 1.44               | 0.44               |
| 6   | 72                   | 80                     | 2.09              | 1.47               | 0.47               |
| 7   | 24                   | 80                     | 2.05              | 1.44               | 0.44               |
| 8   | 72                   | 80                     | 2.09              | 1.47               | 0.47               |
| 9   | 48                   | 40                     | 3.5               | 2.46               | 1.46               |
| 10  | 48                   | 120                    | 1.6               | 1.12               | 0.12               |
| 11  | 48                   | 40                     | 3.65              | 2.57               | 1.57               |
| 12  | 48                   | 120                    | 2.86              | 2.01               | 1.01               |
| 13  | 48                   | 80                     | 3.7               | 2.60               | 1.60               |
| 14  | 48                   | 80                     | 3.85              | 2.71               | 1.71               |
| 15  | 48                   | 80                     | 3.9               | 2.74               | 1.74               |
| 16  | 48                   | 80                     | 3.7               | 2.60               | 1.60               |
| 17  | 48                   | 80                     | 3.8               | 2.67               | 1.67               |

## b) Experimental data of water absorption and yield of regenerated cellulosic fiber

|     |     | Factor 1 | Factor 2          | Factor 3                              | Response 2 | Mass of immersed fiber | Response 1       |
|-----|-----|----------|-------------------|---------------------------------------|------------|------------------------|------------------|
| Std | Run | A: time  | B: copper amine V | C:H <sub>2</sub> SO <sub>4</sub> con. | Yield      |                        | water absorption |
|     |     | Hrs      | ml                | M                                     | Gm         | gm                     |                  |
| 1   | 11  | 24       | 40                | 1                                     | 0.81       | 5.71                   | 6.05             |
| 2   | 16  | 72       | 40                | 1                                     | 1.24       | 3.54                   | 1.86             |
| 3   | 7   | 24       | 120               | 1                                     | 0.76       | 6.23                   | 7.2              |
| 4   | 14  | 72       | 120               | 1                                     | 1.15       | 6.71                   | 4.84             |
| 5   | 13  | 24       | 80                | 0.5                                   | 0.88       | 8.31                   | 8.45             |
| 6   | 8   | 72       | 80                | 0.5                                   | 1.22       | 3.90                   | 2.2              |
| 7   | 12  | 24       | 80                | 1.5                                   | 1.01       | 6.80                   | 5.74             |
| 8   | 3   | 72       | 80                | 1.5                                   | 1.4        | 6.39                   | 3.57             |
| 9   | 4   | 48       | 40                | 0.5                                   | 1.09       | 4.218                  | 2.87             |
| 10  | 10  | 48       | 120               | 0.5                                   | 0.71       | 5.06                   | 6.13             |
| 11  | 6   | 48       | 40                | 1.5                                   | 0.99       | 4.45                   | 3.5              |
| 12  | 2   | 48       | 120               | 1.5                                   | 1.27       | 5.38                   | 3.24             |
| 13  | 9   | 48       | 80                | 1                                     | 1.31       | 3.78                   | 1.89             |
| 14  | 5   | 48       | 80                | 1                                     | 1.33       | 3.05                   | 1.5              |
| 15  | 17  | 48       | 80                | 1                                     | 1.35       | 3.90                   | 1.89             |
| 16  | 1   | 48       | 80                | 1                                     | 1.37       | 4.01                   | 1.93             |
| 17  | 15  | 48       | 80                | 1                                     | 1.37       | 3.01                   | 1.2              |

Table B1 original data from experiment for design expert data analysis

**B2 ANOVA result for water absorption**

Response: water absorption

ANOVA for Response Surface Quadratic Model  
Analysis of variance table [Partial sum of squares]

| Source                   | Sum of Squares | DF | Mean Square | F Value | Prob > F | Remark          |
|--------------------------|----------------|----|-------------|---------|----------|-----------------|
| Model                    | 78.71          | 9  | 8.75        | 50.15   | < 0.0001 | Significant     |
| A: time                  | 28.01          | 1  | 28.01       | 160.62  | < 0.0001 | >>              |
| B: Copper amine volume   | 6.35           | 1  | 6.35        | 36.44   | 0.0005   | >>              |
| C: conc. Of regenerating | 1.62           | 1  | 1.62        | 9.29    | 0.0186   | >>              |
| A2                       | 20.38          | 1  | 20.38       | 116.88  | < 0.0001 | >>              |
| B2                       | 5.52           | 1  | 5.52        | 31.67   | 0.0008   | >>              |
| C2                       | 5.55           | 1  | 5.55        | 31.8    | 0.0008   | >>              |
| AB                       | 0.84           | 1  | 0.84        | 4.8     | 0.0646   | Not significant |
| AC                       | 4.16           | 1  | 4.16        | 23.86   | 0.0018   | Significant     |
| BC                       | 3.1            | 1  | 3.1         | 17.76   | 0.004    | >>              |
| Residual                 | 1.22           | 7  | 0.17        |         |          |                 |
| Lack of Fit              | 0.7            | 3  | 0.23        | 1.81    | 0.2855   | not significant |
| Pure Error               | 0.52           | 4  | 0.13        |         |          |                 |
| Cor Total                | 79.93          | 16 |             |         |          |                 |

The Model F-value of 50.15 implies the model is significant. There is only a 0.01% chance that a "Model F-Value" this large could occur due to noise.

Values of "Prob > F" less than 0.0500 indicate model terms are significant. In this case A, B, C, A2, B2, C2, AC, BC are significant model terms.

Values greater than 0.1000 indicate the model terms are not significant.

If there are many insignificant model terms (not counting those required to support hierarchy), model reduction may improve your model.

The "Lack of Fit F-value" of 1.81 implies the Lack of Fit is not significant relative to the pure error. There is a 28.55% chance that a "Lack of Fit F-value" this large could occur due to noise. Non-significant lack of fit is good -- we want the model to fit.

|           |       |                |        |
|-----------|-------|----------------|--------|
| Std. Dev. | 0.42  | R-Squared      | 0.9847 |
| Mean      | 3.76  | Adj R-Squared  | 0.9651 |
| C.V.      | 11.12 | Pred R-Squared | 0.8492 |

PRESS 12.05 Adeq Precision 20.885

The "Pred R-Squared" of 0.8492 is in reasonable agreement with the "Adj R-Squared" of 0.9651.

"Adeq Precision" measures the signal to noise ratio. A ratio greater than 4 is desirable. Your ratio of 20.885 indicates an adequate signal. This model can be used to navigate the design space.

| Factor               | Coefficient Estimate | DF | Standard Error | 95% CI Low | 95% CI High | VIF  |
|----------------------|----------------------|----|----------------|------------|-------------|------|
| Intercept            | 1.64                 | 1  | 0.19           | 1.20       | 2.08        |      |
| A-time               | -1.87                | 1  | 0.15           | -2.22      | -1.52       | 1.00 |
| B-volume copperamine | 0.89                 | 1  | 0.15           | 0.54       | 1.24        | 1.00 |
| C-H2SO4 con.         | -0.45                | 1  | 0.15           | -0.80      | -0.10       | 1.01 |
| A2                   | 2.20                 | 1  | 0.20           | 1.72       | 2.68        | 1.01 |
| B2                   | 1.15                 | 1  | 0.20           | 0.66       | 1.63        | 1.01 |
| C2                   | 1.15                 | 1  | 0.20           | 0.67       | 1.63        | 1.00 |
| AB                   | 0.46                 | 1  | 0.21           | -0.036     | 0.95        | 1.00 |
| AC                   | 1.02                 | 1  | 0.21           | 0.53       | 1.51        | 1.00 |
| BC                   | -0.88                | 1  | 0.21           | -1.37      | -0.39       | 1.00 |

**Final Equation in Terms of Coded Factors:**

$$\begin{aligned} \text{water absorption} &= \\ &+1.64 \\ &-1.87 * A \\ &+0.89 * B \\ &-0.45 * C \\ &+2.20 * A^2 \\ &+1.15 * B^2 \\ &+1.15 * C^2 \\ &+0.46 * A * B \\ &+1.02 * A * C \\ &-0.88 * B * C \end{aligned}$$

**Final Equation in Terms of Actual Factors:**

$$\begin{aligned} \text{water absorption} &= +24.86500 - 0.56780 * \text{time} - 0.071119 * \text{volume} \\ \text{copperamine} &- 10.64200 * \text{H2SO4 con.} + 3.81988\text{E-}003 * \text{time}^2 + 7.15781\text{E-}004 * \text{volume} \end{aligned}$$

copperamine2 + 4.59100 \* H2SO4 con.2 + 4.76562E-004 \* time \* volume copperamine + 0.085000 \* time \* H2SO4 con. -0.044000 \* volume copperamine \* H2SO4 con.

### Diagnosics Case Statistics

| Std | Actual Value | Predicted Value | Residual | Leverage | Student Residual | Cook's Distance | Outlier t | Run Order |
|-----|--------------|-----------------|----------|----------|------------------|-----------------|-----------|-----------|
| 1   | 6.05         | 6.42            | -0.37    | 0.750    | -1.796           | 0.968           | -2.264    | 11        |
| 2   | 1.86         | 1.77            | 0.092    | 0.750    | 0.443            | 0.059           | 0.416     | 16        |
| 3   | 7.20         | 7.29            | -0.093   | 0.750    | -0.443           | 0.059           | -0.416    | 7         |
| 4   | 4.84         | 4.46            | 0.38     | 0.750    | 1.796            | 0.968           | 2.264     | 14        |
| 5   | 8.45         | 8.33            | 0.12     | 0.750    | 0.569            | 0.097           | 0.539     | 13        |
| 6   | 2.20         | 2.55            | -0.35    | 0.750    | -1.670           | 0.837           | -1.994    | 8         |
| 7   | 5.74         | 5.39            | 0.35     | 0.750    | 1.670            | 0.837           | 1.994     | 12        |
| 8   | 3.57         | 3.69            | -0.12    | 0.750    | -0.569           | 0.097           | -0.539    | 3         |
| 9   | 2.87         | 2.61            | 0.26     | 0.750    | 1.227            | 0.452           | 1.282     | 4         |
| 10  | 6.13         | 6.16            | -0.026   | 0.750    | -0.126           | 0.005           | -0.117    | 10        |
| 11  | 3.50         | 3.47            | 0.026    | 0.750    | 0.126            | 0.005           | 0.117     | 6         |
| 12  | 3.24         | 3.50            | -0.26    | 0.750    | -1.227           | 0.452           | -1.282    | 2         |
| 13  | 1.89         | 1.64            | 0.25     | 0.200    | 0.664            | 0.011           | 0.635     | 9         |
| 14  | 1.30         | 1.64            | -0.34    | 0.200    | -0.916           | 0.021           | -0.904    | 5         |
| 15  | 1.89         | 1.64            | 0.25     | 0.200    | 0.664            | 0.011           | 0.635     | 17        |
| 16  | 1.93         | 1.64            | 0.29     | 0.200    | 0.771            | 0.015           | 0.746     | 1         |
| 17  | 1.20         | 1.64            | -0.44    | 0.200    | -1.183           | 0.035           | -1.225    | 15        |

Proceed to Diagnostic Plots (the next icon in progression). Be sure to look at the:

- 1) Normal probability plot of the studentized residuals to check for normality of residuals.
- 2) Studentized residuals versus predicted values to check for constant error.
- 3) Outlier t versus run order to look for outliers, i.e., influential values.
- 4) Box-Cox plot for power transformations.

If all the model statistics and diagnostic plots are OK, finish up with the Model Graphs icon.

### B3 ANOVA result

Response: yield

ANOVA for Response Surface Quadratic Model

Analysis of variance table [Partial sum of squares]

| Source         | Sum of Squares | DF | Mean Square | F Value | Prob > F |                 |
|----------------|----------------|----|-------------|---------|----------|-----------------|
| Model          | 0.86           | 9  | 0.095       | 98.71   | < 0.0001 | significant     |
| A              | 0.30           | 1  | 0.30        | 311.67  | < 0.0001 |                 |
| B              | 7.200E-003     | 1  | 7.200E-003  | 7.47    | 0.0292   |                 |
| C              | 0.074          | 1  | 0.074       | 76.91   | < 0.0001 |                 |
| A <sup>2</sup> | 0.062          | 1  | 0.062       | 64.77   | < 0.0001 |                 |
| B <sup>2</sup> | 0.23           | 1  | 0.23        | 239.78  | < 0.0001 |                 |
| C <sup>2</sup> | 0.039          | 1  | 0.039       | 40.90   | 0.0004   |                 |
| AB             | 4.000E-004     | 1  | 4.000E-004  | 0.42    | 0.5399   |                 |
| AC             | 6.250E-004     | 1  | 6.250E-004  | 0.65    | 0.4471   |                 |
| BC             | 0.11           | 1  | 0.11        | 113.02  | < 0.0001 |                 |
| Residual       | 6.745E-003     | 7  | 9.636E-004  |         |          |                 |
| Lack of Fit    | 4.025E-003     | 3  | 1.342E-003  | 1.97    | 0.2602   | not significant |
| Pure Error     | 2.720E-003     | 4  | 6.800E-004  |         |          |                 |
| Cor Total      | 0.86           | 16 |             |         |          |                 |

The Model F-value of 98.71 implies the model is significant. There is only a 0.01% chance that a "Model F-Value" this large could occur due to noise.

Values of "Prob > F" less than 0.0500 indicate model terms are significant.

In this case A, B, C, A<sup>2</sup>, B<sup>2</sup>, C<sup>2</sup>, BC are significant model terms.

Values greater than 0.1000 indicate the model terms are not significant.

If there are many insignificant model terms (not counting those required to support hierarchy), model reduction may improve your model.

The "Lack of Fit F-value" of 1.97 implies the Lack of Fit is not significant relative to the pure error. There is a 26.02% chance that a "Lack of Fit F-value" this large could occur due to noise. Non-significant lack of fit is good -- we want the model to fit.

|           |       |                |        |
|-----------|-------|----------------|--------|
| Std. Dev. | 0.031 | R-Squared      | 0.9922 |
| Mean      | 1.13  | Adj R-Squared  | 0.9821 |
| C.V.      | 2.74  | Pred R-Squared | 0.9204 |
| PRESS     | 0.069 | Adeq Precision | 29.665 |

The "Pred R-Squared" of 0.9204 is in reasonable agreement with the "Adj R-Squared" of 0.9821.

"Adeq Precision" measures the signal to noise ratio. A ratio greater than 4 is desirable. Your ratio of 29.665 indicates an adequate signal. This model can be used to navigate the design space.

| Factor                | Coefficient |    | Standard Error | 95% CI |             | VIF  |
|-----------------------|-------------|----|----------------|--------|-------------|------|
|                       | Estimate    | DF |                | Low    | High        |      |
| Intercept             | 1.35        | 1  | 0.014          | 1.31   | 1.38        |      |
| A-time                | 0.19        | 1  | 0.011          | 0.17   | 0.22        | 1.00 |
| B-volume copperamine- | 0.030       | 1  | 0.011          | -0.056 | -4.049E-003 | 1.00 |

|                |             |   |       |        |        |      |
|----------------|-------------|---|-------|--------|--------|------|
| C-H2SO4 con.   | 0.096       | 1 | 0.011 | 0.070  | 0.12   | 1.00 |
| A <sup>2</sup> | -0.12       | 1 | 0.015 | -0.16  | -0.086 | 1.01 |
| B <sup>2</sup> | -0.23       | 1 | 0.015 | -0.27  | -0.20  | 1.01 |
| C <sup>2</sup> | -0.097      | 1 | 0.015 | -0.13  | -0.061 | 1.01 |
| AB             | -1.000E-002 | 1 | 0.016 | -0.047 | 0.027  | 1.00 |
| AC             | 0.012       | 1 | 0.016 | -0.024 | 0.049  | 1.00 |
| BC             | 0.17        | 1 | 0.016 | 0.13   | 0.20   | 1.00 |

**Final Equation in Terms of Coded Factors:**

$$\begin{aligned} \text{yield} &= \\ &+1.35 \\ &+0.19 * A \\ &-0.030 * B \\ &+0.096 * C \\ &-0.12 * A^2 \\ &-0.23 * B^2 \\ &-0.097 * C^2 \\ &-1.000E-002 * A * B \\ &+0.012 * A * C \\ &+0.17 * B * C \end{aligned}$$

**Final Equation in Terms of Actual Factors:**

$$\begin{aligned} \text{yield} &= \\ &-0.31500 \\ &+0.028156 * \text{time} \\ &+0.014925 * \text{volume copperamine} \\ &+0.25650 * \text{H2SO4 con.} \\ &-2.11372E-004 * \text{time}^2 \\ &-1.46406E-004 * \text{volume copperamine}^2 \\ &-0.38700 * \text{H2SO4 con.}^2 \\ &-1.04167E-005 * \text{time} * \text{volume copperamine} \\ &+1.04167E-003 * \text{time} * \text{H2SO4 con.} \\ &+8.25000E-003 * \text{volume copperamine} * \text{H2SO4 con.} \end{aligned}$$

**Diagnostics Case Statistics**

| Std | Actual Value | Predicted Value | Residual | Leverage | Student Residual | Cook's Distance | Outlier t | Run Order |
|-----|--------------|-----------------|----------|----------|------------------|-----------------|-----------|-----------|
|-----|--------------|-----------------|----------|----------|------------------|-----------------|-----------|-----------|

|    |      |      |             |       |        |       |        |    |
|----|------|------|-------------|-------|--------|-------|--------|----|
| 1  | 0.81 | 0.82 | -6.250E-003 | 0.750 | -0.403 | 0.049 | -0.377 | 11 |
| 2  | 1.24 | 1.22 | 0.016       | 0.750 | 1.047  | 0.329 | 1.055  | 16 |
| 3  | 0.76 | 0.78 | -0.016      | 0.750 | -1.047 | 0.329 | -1.055 | 7  |
| 4  | 1.15 | 1.14 | 6.250E-003  | 0.750 | 0.403  | 0.049 | 0.377  | 14 |
| 5  | 0.88 | 0.85 | 0.030       | 0.750 | 1.933  | 1.121 | 2.621  | 13 |
| 6  | 1.22 | 1.21 | 7.500E-003  | 0.750 | 0.483  | 0.070 | 0.455  | 8  |
| 7  | 1.01 | 1.02 | -7.500E-003 | 0.750 | -0.483 | 0.070 | -0.455 | 12 |
| 8  | 1.40 | 1.43 | -0.030      | 0.750 | -1.933 | 1.121 | -2.621 | 3  |
| 9  | 1.09 | 1.11 | -0.024      | 0.750 | -1.530 | 0.702 | -1.737 | 4  |
| 10 | 0.71 | 0.72 | -0.014      | 0.750 | -0.886 | 0.235 | -0.870 | 10 |
| 11 | 0.99 | 0.98 | 0.014       | 0.750 | 0.886  | 0.235 | 0.870  | 6  |
| 12 | 1.27 | 1.25 | 0.024       | 0.750 | 1.530  | 0.702 | 1.737  | 2  |
| 13 | 1.31 | 1.35 | -0.036      | 0.200 | -1.297 | 0.042 | -1.377 | 9  |
| 14 | 1.33 | 1.35 | -0.016      | 0.200 | -0.576 | 0.008 | -0.547 | 5  |
| 15 | 1.35 | 1.35 | 4.000E-003  | 0.200 | 0.144  | 0.001 | 0.134  | 17 |
| 16 | 1.37 | 1.35 | 0.024       | 0.200 | 0.864  | 0.019 | 0.847  | 1  |
| 17 | 1.37 | 1.35 | 0.024       | 0.200 | 0.864  | 0.019 | 0.847  | 15 |

Proceed to Diagnostic Plots (the next icon in progression). Be sure to look at the:

- 1) Normal probability plot of the studentized residuals to check for normality of residuals.
- 2) Studentized residuals versus predicted values to check for constant error.
- 3) Outlier t versus run order to look for outliers, i.e., influential values.
- 4) Box-Cox plot for power transformations.

If all the model statistics and diagnostic plots are OK, finish up with the Model Graphs icon.

## Appendix c

### C1 FIT SUMMERY

Water absorption

| source           | Sum of squares | DF       | Mean square  | F value      | Prob >F            |                 |
|------------------|----------------|----------|--------------|--------------|--------------------|-----------------|
| Mean             | 239.89         | 1        | 239.89       |              |                    |                 |
| Linear           | 35.99          | 3        | 12.00        | 3.55         | 0.0450             |                 |
| 2FI              | 8.10           | 3        | 2.70         | 0.75         | 0.5454             |                 |
| <u>Quadratic</u> | <u>34.63</u>   | <u>3</u> | <u>11.54</u> | <u>66.19</u> | <u>&lt; 0.0001</u> | <u>sugusted</u> |
| Cubic            | 0.70           | 3        | 0.23         | 1.81         | 0.2855             | alliased        |
| Residual         | 0.52           | 4        | 0.13         |              |                    |                 |

"Lack of Fit Tests" Want the selected model to have insignificant lack-of-fit.

Model Summary Statistics

| Source    | Std. Dev. | Adjusted R-Squared | Predicted R-Squared | R-Squared | PRESS |           |
|-----------|-----------|--------------------|---------------------|-----------|-------|-----------|
| Linear    | 1.84      | 0.4502             | 0.3233              | 0.1251    | 69.94 |           |
| 2FI       | 1.89      | 0.5515             | 0.2824              | -0.1572   | 92.50 |           |
| Quadratic | 0.42      | 0.9847             | 0.9651              | 0.8492    | 12.05 | Suggested |
| Cubic     | 0.36      | 0.9935             | 0.9741              |           |       | Aliased   |

"Model Summary Statistics": Focus on the model maximizing the "Adjusted R-Squared" and the "Predicted R-Squared".

yield

| Source           | Sum of squares | DF       | Mean square | F value       | Prob >F            |                 |
|------------------|----------------|----------|-------------|---------------|--------------------|-----------------|
| Mean             | 21.82          | 1        | 21.82       |               |                    |                 |
| Linear           | 0.38           | 3        | 0.13        | 3.44          | 0.0490             |                 |
| 2FI              | 0.11           | 3        | 0.037       | 0.99          | 0.4376             |                 |
| <u>Quadratic</u> | <u>0.36</u>    | <u>3</u> | <u>0.12</u> | <u>126.08</u> | <u>&lt; 0.0001</u> | <u>sugusted</u> |
| Cubic            | 4.025E-003     | 3        | 1.342E-003  | 1.97          | 0.2602             | alliased        |
| Residual         | 2.720E-003     | 4        | 6.800E-004  |               |                    |                 |

"Sequential Model Sum of Squares": Select the highest order polynomial where the additional terms are significant and the model is not aliased.

**Lack of Fit Tests**

| Source     | Sum of Squares | DF | Mean Square | F Value | Prob > F |           |
|------------|----------------|----|-------------|---------|----------|-----------|
| Linear     | 0.48           | 9  | 0.053       | 78.17   | 0.0004   |           |
| 2FI        | 0.37           | 6  | 0.061       | 90.31   | 0.0003   |           |
| Quadratic  | 4.025E-003     | 3  | 1.342E-003  | 1.97    | 0.2602   | Suggested |
| Cubic      | 0.000          | 0  |             |         |          | Aliased   |
| Pure Error | 2.720E-003     | 4  | 6.800E-004  |         |          |           |

"Lack of Fit Tests": Want the selected model to have insignificant lack-of-fit.

**Model Summary Statistics**

| Source    | Std. Dev. | Adjusted R-Squared | Predicted R-Squared | R-Squared | PRESS |           |
|-----------|-----------|--------------------|---------------------|-----------|-------|-----------|
| Linear    | 0.19      | 0.4423             | 0.3136              | 0.0894    | 0.79  |           |
| 2FI       | 0.19      | 0.5697             | 0.3116              | -0.1433   | 0.99  |           |
| Quadratic | 0.031     | 0.9922             | 0.9821              | 0.9204    | 0.069 | Suggested |
| Cubic     | 0.026     | 0.9968             | 0.9874              |           |       | Aliased   |

"Model Summary Statistics"0+: Focus on the model maximizing the "Adjusted R-Squared" and the "Predicted R-Squared".

**C2 Desirability**

**Constraints**

| Name               | Goal        | Lower Limit | Upper Limit | Lower Weight | Upper Weight | Importance |
|--------------------|-------------|-------------|-------------|--------------|--------------|------------|
| time               | is in range | 24          | 72          | 1            | 1            | 3          |
| volume copperamine | is in range |             | 40          | 120          |              | 1          |
| H2SO4 con.         | is in range | 0.5         | 1.5         | 1            | 1            | 3          |
| water absorption   | minimize    | 1.2         | 8.45        | 1            | 1            | 3          |
| yield              | maximize    | 0.71        | 1.4         | 1            | 1            | 3          |

**solution.**

---

| Number | Time  | volume<br>copperamine | H2SO4<br>con. | water<br>absorption | Yield   | Desirability |          |
|--------|-------|-----------------------|---------------|---------------------|---------|--------------|----------|
| 1      | 59.04 | 72.82                 | 1.01          | 1.08911             | 1.40917 | 1.000        | Selected |
| 2      | 60.67 | 68.75                 | 1.05          | 1.08274             | 1.41032 | 1.000        |          |
| 3      | 60.84 | 74.85                 | 1.01          | 1.14638             | 1.4166  | 1.000        |          |
| 4      | 59.49 | 75.16                 | 1.06          | 1.16911             | 1.42043 | 1.000        |          |
| 5      | 63.69 | 72.37                 | 0.95          | 1.14802             | 1.4115  | 1.000        |          |
| 6      | 61.88 | 67.88                 | 0.98          | 1.03526             | 1.40419 | 1.000        |          |
| 7      | 61.89 | 71.69                 | 0.97          | 1.09053             | 1.41043 | 1.000        |          |
| 8      | 60.67 | 71.27                 | 1.11          | 1.18955             | 1.42084 | 1.000        |          |
| 9      | 61.53 | 72.20                 | 1.05          | 1.14663             | 1.42069 | 1.000        |          |
| 10     | 63.05 | 75.35                 | 0.93          | 1.19418             | 1.40597 | 1.000        |          |