



Preparation and Characterization on Natural Dyes Based on Neem, Henna and Turmeric for Dyeing on Cotton with Superhydrophobic Coating

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Abstract: This study is presents the preparation and characterization on natural dyes based on neem, henna and turmeric for dyeing on cotton with superhydrophobic coating. Natural dyes were used to reduce the usage of the synthetic dyes in dyeing process and superhydrophobic coating to maintain the colour of the dyes on the fabric from faded and acts as a self-cleaning. The methodology involved for this study was the extraction process of natural dyes from the neem, henna and turmeric. There are eight different concentrations consists of 0.25g/ml, 0.5g/ml, 0.75g/ml, 1.0g/ml, 1.25g/ml, 1.50g/ml, 1.75g/ml and 2.0g/ml. The mechanical test involved is abrasion test for evaluation the resists wear caused by flat rubbing contact with another materials. Henna coated cotton fabrics have lower percentage weight loss compared to neem and turmeric coated cotton fabrics and uncoated cotton fabrics. The weight percentage loss for henna coated cotton fabrics at 2.0g/ml was 3.32%, for neem coated cotton fabrics at 2.0g/ml was 3.33% and turmeric at 2.0g/ml concentration was 3.34%. On the other hand, the value for the henna uncoated cotton fabric at 2.0g/ml was at 3.41%, neem at 2.0g/ml concentration was 3.42% and the value for uncoated turmeric at 2.0g/ml was 3.43% respectively. The porosity decreases when the concentration of dyes and superhydrophobic coating applied on the cotton fabrics.

Keywords: Natural dyes, superhydrophobic coating, extraction, fabrics.

1. Introduction

Textile materials are mainly from synthetic dyes where it was produced from petrochemical which are not renewable and not biodegradable [1]. Around 30 million tonnes of dye was estimated for the global consumption for textiles and was expected to grow up to 3% per annum while 70,000 tonnes of dyes were released to the environment [2]. The population of people who will have allergic to the chemicals will grow up to 60 per cent by year 2020 which was stated in a business week [3,4]. In addition, approximately 280,000 tons of textile dyes are discharged annually worldwide and approximately 10,000 different dyes have been produced and an estimated 8×10^5 tons of synthetic dyes

are used in the textile industry worldwide [4,5]. Natural dyes become an alternative way to the industries when synthetic dyes caused severe problems such as visible residues in the effluent [5] and toxic amines were created [6]. On the other hand, natural dyes can return to nature at the end of use, such as wastewater from dyeing process. Therefore, researchers are interested in natural dyes because it is more compatible with the environment and has an antibacterial and ultraviolet protection functions [6,7]. Synthetic dye production involves a large number of cancer-causing synthetic chemicals and effluents that are released into the water or produced into the air resulting in contamination. Besides that, interest in the usage of natural dyes has been revived worldwide due to the toxic nature and effect of synthetic dyes on all forms of life. There are many examples of natural dyes such as henna which was used in textile fabrics [7] and it contains red-orange pigment known as 2-hydroxyl-1, 4-naphthaquinone [8]. Henna leaves have many names, such as hina, henna tree, mignonette tree, and *Lawsonia Inermis*, scientifically derived from Egyptian privet [9]. Furthermore turmeric was used for medicinal properties and curcumin is the colouring agent which produce yellow colour [10]. The scientific name for turmeric is called the rhizome *Curcuma longa* and the family name is *Zingiberaceae* [11]. Turmeric dyed cotton fabric has an excellent UV protection properties and good home laundering longevity. Moreover neem is a renewable source for soap industry, medicines and textile industry where azadirachtin produce green colour [12,13]. The scientific name for neem plant is *Azadiractha Indica*, a plant belonging to the Mahogany family known as *Meliaceae* [14].

Cotton fabrics have cellulose and abundant hydroxyl groups structure where it will make hydrophilic properties where the fiber can provide an appropriate environment for microorganism growth when in contact with water and sweats. Besides that, cellulose as 94%, 1.3% protein, 0.9% pectin, 1.2% oil, 0.6% wax and 2% other substances are the general composition of cotton fibers. Cotton fabric is widely used in various application due to their biodegradability, renewability and environmental friendly where it does not contain any harmful composition [15]. On the other hand, superhydrophobic coating acts as self-cleaning and protection to the products. Coating technology has desirable performance such as mechanical properties, water resistance, antibacterial properties, UV resistance and can be used in food, clothing, paper packaging and other applications. Hydrophobic materials have been incorporated into the fabric layer with paraffin waxes and paraffin emulsion, resulting in inadequate air and vapor permeability and poor comfort. Nano-scaled particles such as silicon dioxide (SiO_2), titanium dioxide (TiO_2) and zinc oxide (ZnO) were then used to coat and improve the textile fiber surface roughness [16]. In this research natural resources such as neem, henna and turmeric have been used to replace the artificial dye where synthetic dye can lead to severe contamination of wastewater and human health effects. On the other hand, the superhydrophobic coating was used in this research to enhance the mechanical properties for the fabric where superhydrophobic coating was synthesized from the titanium dioxide. The main objectives of this research is to evaluate the abrasion strength and porosity for the uncoated and coated dyed cotton fabrics.

2. Materials and method

The materials were natural resources based on neem, henna and turmeric to dye on cotton fabric for dyeing. Henna is known as *Lawsonia Inermis* which belong to the Lythraceae's family and produce orange-red colour. Apart from that, neem plant was known as *Azadiractha Indica* was from Mahogany family known as *Meliceae*. Turmeric also known as *Curcuma Longa* from rhizome and *Zingiberaceae* is the family name for the turmeric plant. The neem, henna and turmeric were cut from plant and washed thoroughly with water for five times [17]. Then the neem, henna and turmeric were dried in the oven for 57°C for 24 hours. After that, the dried leaves were grind by using grinder into powdered form for effective extraction. The particle size for the powdered form was $0.05\mu\text{m}\pm 0.01$. Then the powdered samples were sieved with a sieve size $0.05\mu\text{m}$ with 270 meshes to remove some stalk that is not well ground [18]. The methods used in extracting dye for neem, henna and turmeric were cold water extraction. Later the powdered dyes were weighed into 10g, 20g, 30g, 40g, 50g, 60g, 70g and 80g. Then 40ml of distilled water were added into the divided powdered dyes respectively. Moreover the water based dye were filtered using filter paper Whitman No 1, $6\pm 1\mu\text{m}$. Table 1 shows the concentration of dyes.

Table 1 - Concentration of dyes

Powdered dye (g)	Distilled water (ml)	Concentration of dye (g/ml)
1	4	0.
2	4	0.
3	4	0.
4	4	1.
5	4	1.
6	4	1.
7	4	1.
80	40	2.00

2.1 Process of superhydrophobic coating

The superhydrophobic coating has three stages which are alcoholysis, condensation and purification. For stage alcoholysis the 50.3% of solvent which is fluorocarbon was used and mixed with 80ml of crosslinker isopropyl alcohol. Then second stage was condensation step which 2.4% (wt/wt%) was used to produce the binder. Third step is purification process. The solution that was produced from first stage and second stage were undergoing mixing and stirring process for 45 minutes at 70°C. Then the DI water was doped with TiO₂ powder. The solutions will heat at 1000 for 60 minutes. Lastly the superhydrophobic coating will pour into the spray and gun and spray on the dyed cotton fabrics [19]. The method of coating was spraying technique. The standard environment for drying time of air dry was 23±20C while the relative humidity was 50±5% under diffuse sunlight. The angle for spray gun and the coated fabric was 45° by swing the gun back and forth across the surface. The coating layer for each concentration dyed cotton fabrics were five layers.

2.2 Abrasion test (ASTM 4966-98)

Abrasion test was conducted to test the ability to resist wear caused by flat rubbing contact with another material. Abrasion test was inspected for wear and tear resistance of the fabrics. The ASTM 4966-98: Standard Test Method for Abrasion Resistance for Textile Fabric (Martindale Abrasion Tester Method). Figure 1 show the schematic diagram of abrasion test where the abrasion machine was pulled out the fabrics and loaded with the weight [20].

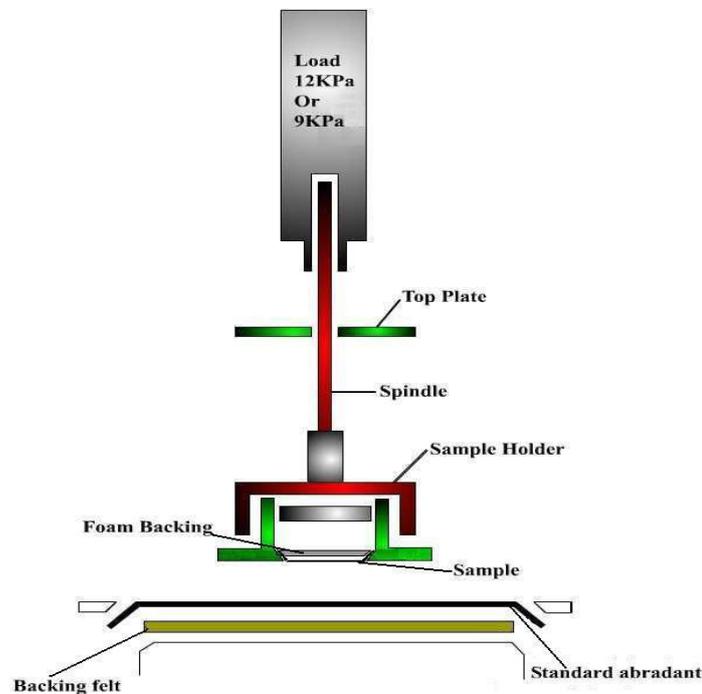


Fig. 1- Schematic diagram of abrasion tester [20]

Martindale abrasion tester was used for this study. The test procedure is four pieces of abrasive materials were cut into 140mm diameter. Then, the fabrics sample with coated and uncoated dyed cotton fabrics were cut into 38mm diameter. All the dyed cotton fabrics and uncoated dyed fabrics for three types were weighed and the 12kg of load was put on the samples. The rubbing cycle was 5000 times and the weight of the fabric before and after was weighed [21]. The percentage of weight loss was calculated.

2.3 Porosity test (ASTM A276)

The porosity test was conducted using Olympus Metallographic Microscope and Image Analysis Software. Moreover the standard method was Standard Test Method for Porosity on Textile Fabric (Metallographic Image Analysis). The fabric size was 5cm x 5cm according to the standard method. Eight different concentrations based on neem, henna and turmeric fabrics with coated and uncoated were used for this test. Olympus microscope was the tool for measuring the area fraction and number of pores on cross-sectional surfaces and coating fabrics. Apart from that, the software was used threshold method to differentiate between the pores and the substrate on colour. Then the pore

density and the size of the largest pore were calculated for every selected region of interest on the whole image. Besides that, the magnification lens was x5.

3. Results and Discussion

3.1 Abrasion test

From figure 2 the percentage of weight loss for the coated dyed fabrics are lower than the uncoated dyed cotton fabrics. The percentage of weight loss for the 1000 rub cycles coated henna dyed cotton fabrics are lower compared to the neem and turmeric. The percentage of weight loss for the coated henna dyed cotton fabrics are from 3.32% to 3.38% from 0.25g/ml to 2.0g/ml concentrations. The weight loss for the coated neem dyed cotton fabrics are from 3.33% to 3.39% for the concentration 0.25g/ml to 2g/ml and coated turmeric dyed cotton fabrics have 3.34% to 3.40% respectively. On the other hand, the weight loss for the concentration 0.25g/ml to 2g/ml for the uncoated henna was 3.48% to 3.41%, uncoated neem was 3.49% to 3.42% and uncoated turmeric was 3.5% to 3.43% respectively.

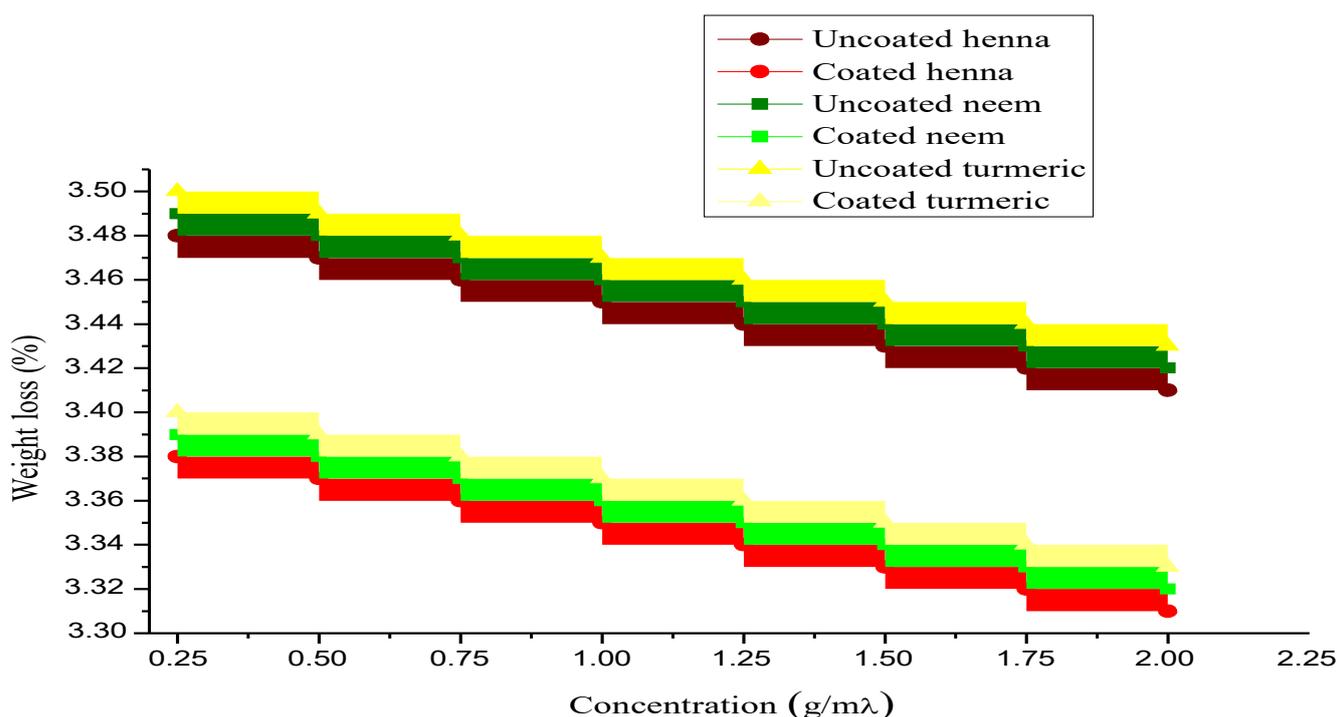


Fig. 2 - Weight loss (%) versus concentration (g/ml)

There are differences in weight loss for the coated and uncoated dyed cotton fabrics. The differences between the coated and uncoated value for 0.25g/ml for the henna was 0.1%, for neem was 0.1% and for henna as 0.1%. The higher the concentration the lower the weight losses for the abrasion test. This is due to the strong covalent bond that forms between the hydroxyl group in henna structure and coating particles in the fabrics [22]. These covalent bonds enhance the strength of fibers in the fabrics [23]. Apart from that henna dye have higher affinity towards cotton fabrics because it has hydroxyl group and bonded by covalent bond [24]. Moreover the abrasion results was good for henna coated fabrics due to happens of Lawsone which are functional groups of henna dye. This Lawsone functional group is capable in forming covalent bonds with active site in the fiber such as hydroxyl group in the cotton and the bond formation was bonded between the functional groups and the substrate results in high abrasion strength and other as moderate [25]. The neem dye can form intermolecular attraction between the hydrogen structure and hydroxyl bond in the coating particles and it has slightly low abrasion value as compared to the henna dye [26]. Neem and turmeric dyed fabrics have weak energy of adsorption between surface dispersant and abrasive causes more fibers to be thorn during the abrasion process on the other material surfaces. Therefore free hydroxyl groups in the neem and turmeric may promote at a high concentration where it decrease the covelant bond between the coating particles and fibers. Due to the increase, residual abrasive particles are significantly reduced for the neem and turmeric coated fabrics. During the dyeing cycle, temperature may reduce the effect on the dye penetration process into the cotton fibers. Apart from that, it can be concluded that fabrics with superhydrophobic coating have slightly effect on abrasion resistance properties as compared to the uncoated fabrics. This is due to the particles in the coating will penetrate inside the fibers and it will form a stronger layer on the top of fabrics [27]. The weight loss of fabric is affected the superhydrophobic coating layer

on the fabric, the rubbing action tends to damage the layer of the fabrics first then only the dyed surface [28]. Each type of natural based on neem, henna and turmeric can used for different types of application even though it gives different abrasion values.

3.2 Porosity test

Figure 3 shows the results that were obtained from Olympus microscope for the porosity of uncoated dyed cotton and coated dyed cotton fabrics for eight different concentrations for three types of dyes which were neem, henna and turmeric. Turmeric dyed uncoated cotton fabrics showed the porosity percentage for 0.25g/ml was 11.33 % and 2.0g/ml for 10.47 % whereas for the coated dyed fabrics shown 9.98 % for 0.25g/ml and 6.13 % for 2.0g/ml respectively. The porosity value for the henna uncoated dyed cotton fabrics were 18.34 % for 0.25g/ml and 10.55 % for 2.0g/ml whereas for henna coated dyed cotton fabrics were 9.46 % for 0.25g/ml and 6.31 % for 2.0g/ml respectively. Besides that, neem uncoated dyed cotton fabrics were shown results as 16.45 % for 0.25g/ml and 12.67 % for 2.0g/ml while for the coated cotton fabrics were 11.93 % for 0.25g/ml, and 10.01 % for 2.0 g/ml respectively.

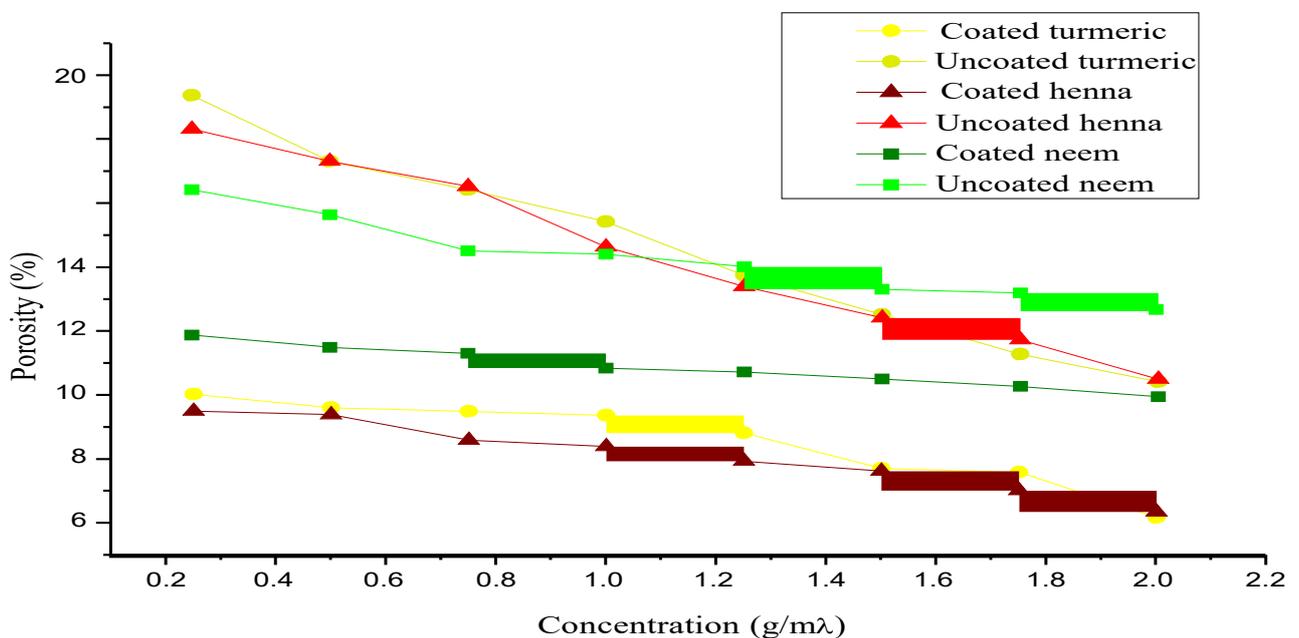


Fig. 3 - Porosity (%) versus concentration (g/ml)

The porosity value was decreased as the concentration increases with superhydrophobic coating. The porosity for the uncoated dyed cotton fabrics was higher than coated dyed cotton fabrics. This is because the particles in the superhydrophobic coating were fully covered all the pores in the fabrics and the structure of the fabrics was compact [29]. Moreover increasing amount of dyes during the dyeing process will resulted in higher rate of dye adsorption. It will increase the surface area and decrease the porosity on the fabrics. The ions in the henna, neem and turmeric will attract the coating ions and will form a larger molecules which can closed the pores between the cotton fibers. Another researcher explained there were low surface energy for the coated fabric samples and therefore more attraction of dye particles to the running liquid than to the solid surface itself [30]. Another reason was that the cellulosic fibers will combine with the dyes mainly by hydrogen bonding as well as Van der Waals forces and do not have any identifiable dye sites. As a result there is usually no specific limit to the amount of dyes where the fibers will absorb the maximum dye molecules during the dyeing process and coating molecules [31]. Apart from that, the natural dyes have good dye attraction towards the fabrics due to the strong covalent bonds between the dye molecules and the fibre on the fabrics while there were some uncovered pores in the fabrics. Besides that, the cotton fabrics were absorbed more dyes and particles from superhydrophobic coating than other fabrics because cotton is made up from cellulose fibres [32]. The current study of porosity is using capacitance sensing approach for the frost growth on heat pumping [33].

4. Conclusion

The preparation of natural dyes based on neem, henna and turmeric were successfully obtained and was dye on the fabrics. The present research was studied to determine the abrasion strength and porosity for the uncoated and coated dyed cotton fabrics. The results of this research showed the decreasing of weight loss at 2.0g/ml for the coated henna was 3.32%, neem at 3.33% and turmeric at 3.34% compared to the uncoated dyed cotton fabric. It reveals that the abrasion strength was good at 2.0g/ml compared to other concentrations. The porosity results were shown that 6.13% for the coated turmeric, henna at 6.31% and neem at 10.01% and it explained that the number of pores in the cotton fabrics decreases when the concentration of dyes increases with the superhydrophobic coating. The findings from this research make several contribution to the current literature. Further experiments and studies that involved natural dyes are strongly recommended. There are a lot of natural resources which have not discovered yet. Moreover more active researcher is needed to build a knowledge base and database with the production of appropriate studies for different textiles. It would be interesting to assess the effect of UV exposure on the uncoated and coated dyed cotton fabrics.

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