Preparation of Azo Dye from *Acacia catechu* and its Application on Silk Fabrics

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Abstract— Recently customers are going to be more conscious about the quality of garments products and there is an increasing interest in textile industry for the development of eco-friendly dyeing process. In the present study widely used azo dye has been synthesized by using the polyphenolic compounds from a natural source. The polyphenolic compounds of the *Acacia catechu* is used for diazotization reaction and formation of dye containing the azo group was confirmed by FTIR and UV-VIS spectra. The prepared dye has been applied on silk fabrics and effects of sunlight, detergent, acid and alkali have been investigated. Fair to moderate fastness values have been recorded.

Keywords: Azo dye, *Acacia catechu*, Diazotization, Wash Fastness.

I. INTRODUCTION

Dyes can generally be described as colored substances that have affinity to the substrates to which they are being applied. Azo dyes are the most versatile class of dyes and in 1858 first preparation of aromatic diazo compounds was reported by Peter Griess [1]. Traditionally azo dyes have been most widely used in textile industry [2-4]. Recently advanced applications in organic synthesis and some diversified applications such as biomedical studies, ink-jet printer, liquid crystalline displays, electro-optical devices etc. have been reported [5-7].

One side of the cromophoric azo group is always connected with an aromatic ring or heterocyclic nucleus and the other side of the group can be linked with an unsaturated molecule of heterocyclic or aliphatic molecules. Due to the possibility of connecting with unlimited number of different molecules, large numbers of azo compounds are available and possible to be synthesized [8]. The huge application and good tinctorial strength as well as stability make azo compounds important dyestuff and continuously receiving attention in scientific research. Huge applications of azo dye create a great environmental problem because synthesis of azo dye requires lagre quantity of carcinogenic aryl amines and naphthols [9]. Moreover, degradation of azo dyes sometimes produces harmful aromatic amine [10-11]. Regarding these environmental and toxicological concerns extended research is going on for new diazotization reaction to prepare new azo compounds.

Over the last two decades emphasis has been given to the synthetic dye industry to developed non-toxic or less toxic dyes thorough health, safety and environmental standards. However, there are still companies making carcinogenic dyes or those laced with harsh chemicals. Today, many researches have been carried out to rediscover the joy of achieving color through the use of renewable, non-toxic, natural sources. Generally azo dyes do not occur in nature and are produced only through chemical synthesis [12]. But recently many azo dyes have been prepared by using some natural source to reduce the negative impacts. Ashitosh B. Pawar *et al.* have prepared azo dye by coupling Kasim Kaaram (Ancient natural dye) with diazonium salts of primary amines [13]. A. Akhter *et al.* have used the extract of green coconut shell as a source of polyphenolic compounds to synthesis coconut shell extract based azo dye [14].

Acacia catechu is a small to moderate sized plant widely distributed throughout Asia and hardwood of Acacia catechu is anciently used as natural dye. The main origin of this plant is Bangladesh, India, Pakistan and Thailand. It contains polyphenolic components, tannins, alkaloids, carbohydrates, flavonoids [15]. In this study the *Acacia catechu* is chemically modified by coupling it with diazonium salts of primary amines to prepare azo dye and its dyeing capacity on silk fabrics has been investigated.

II. MATERIALS AND METHODS

2.1 Collection of Raw Materials:

Aniline, hydrochloric acid, sodium nitrite and sodium hydroxide were purchased from Merck Germany and used without any further purification. *Acacia catechu* powder was collected from the local market of Chargaht bazaar, Rajshahi, Bangladesh.

2.2 Diazotization reaction:

5.0 ml Aniline and concentrated HCl were added in a beaker and cooled on an ice bath for half an hour. Previously cooled 4% 100 mL $NaNO_2$ solution was added slowly to the beaker with continuous stirring for the formation of diazonium salt. In another beaker 15g of raw *Acacia catechu* powder was dissolved in 250 mL 0.1M NaOH solution and cooled in ice bath. The insoluble materials were separated from the solution through filtration. After that the solution of Acacia catechu was slowly added to the beaker of diazonium salt keeping in an ice bath.

2.3 Fourier transform-infrared (FTIR) and UV-VIS spectroscopy:

Fourier transform-infrared **(**FTIR) absorption spectra of synthesized azo dye and raw *Acacia catechu* powder were taken using the IR Prestige 21 Shimadzu Spectrometer. Dilute solution of the prepared azo dye in distilled water was prepared and absorbance maximum was determined with Shimadzu UV-3600.

2.4 Application on Silk Fabrics:

The silk fabrics used for dyeing in the present work were collected from sericulture industry, Rajshahi. The fabric of size 15 x 20 cm was heated with 1% detergent solution at 80˚C for 30 minutes in order to degumming. Finally the fabric was washed thoroughly 2-3 times with distilled water. Then the fabric was heated at 80˚C with 0.05% acetic acid solution for 30 minutes. The dyeing bath was prepared with 3% of *Acacia Catechu* based azo dye and the temperature of the dye bath was 80˚C. Finally the silk fabric was dipped in the dyeing bath and allowed to stand for 1.5 hours with occasional stirring. After dyeing, the dyed material was washed with cold water for 2-3 times and dried at room temperature.

2.5 Light fastness study:

The dyed sample was divided in five pieces and one of them was exposed to sunlight for 6 hours a day for three consecutive days. The changes of shades after every day were measured by means of SDC-3305 ISO Grey Scale. The results are shown in Table 1.

2.6 Wash Fastness study:

One piece of the dyed fabric dipped in 1% standard detergent solution and allowed to stand for 30 minutes at 45˚C temperature. After 30 minutes the fabric was rinsed with distilled water, squeezed and allowed to dry at defused sunlight. The washing procedure was repeated for 2 more times and after every wash the dried fabrics were compared with the controlled sample. The change in color was assessed by means of SDC-3305 ISO Grey Scale. The results are shown in Table 1.

2.7 Action of Acid and Alkali:

The color changes by the action of dilute acid and alkali were investigated with 0.01 N HCl and 0.01 N NaOH solutions. The other two pieces of the fabrics were dipped in 0.01 N HCl and 0.01 N NaOH solutions and allowed to stand for 30 minutes at room temperature. This procedure was repeated for 2 more times and after every wash the dried samples were compared with the controlled sample. The change in color was assessed by means of Grey Scale (Table 1).

III. RESULTS AND DISCUSSION

In presence of alkali, phenols and diazonium salts can undergo diazotization reaction. In this study the polyphenolic compounds of *Acacia catechu* like catechin, quercetin can undergo diazocoupling reaction. Alkaline extract of Acacia catechu may not contain a single compound and consequently may not give a pure azo dye. The probable structure of one azo dye [16] that may present in the mixture is presented in fig. 1.

The FTIR spectra of *Acacia catechu* and *Acacia Catechu* based azo dye are presented in fig. 2. The sharp peak with medium intensity at 1620 cm^{-1} for Acacia catechu and broad absorption band in the region of wave number 3500-3100 cm^{-1} were found due the presence of aromatic compounds and the O-H stretching vibration respectively (Fig. 2) [13]. The absorption band at 1320 cm^{-1} for *Acacia catechu* powder is due to O-H bending vibrations. The new absorption band at 1450 cm^{-1} for the synthesized *Acacia catechu* based azo dye was appeared due to the formation of azo (-N=N-) group. Azo colorants selectively reflect, transmit or scatter light in the visible spectrum [14]. The UV-VIS spectrum of *Acacia catechu* based azo dye is presented in fig. 3. Two characteristic absorption bands were found in the spectrum of the azo dye. In the visible range and UV range the maximum absorptions (λ_{max}) were found at 480 nm and 392 nm respectively.

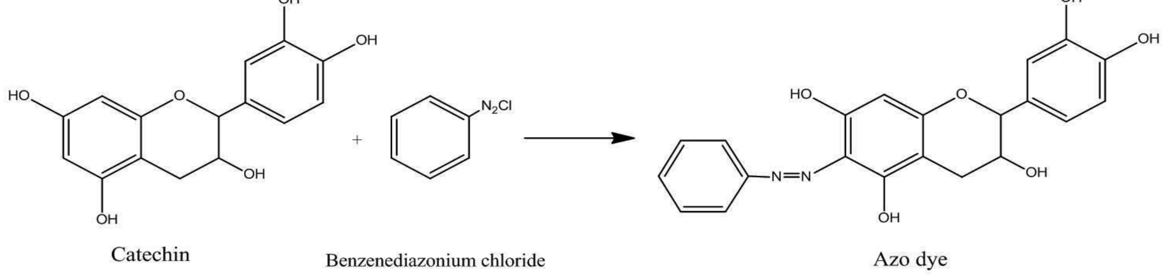

Figure 1: Proposed reaction for the preparation of *Acacia catechu* based azo dye

Dyeing is the aqueous application of color to the textile substrate and the prepared azo dye is completely water soluble. The natural source *Acacia catechu* based azo dye was fixed on silk fabrics without any mordant and gives brown-orange shade to silk (Fig. 4a). The dye can be washed away from the clothing by dissolving in the washing liquid. Dissolving in water is a physical change results in the color alteration or fading of clothes. On washing with normal tap water the color was found stable. Fading through exposure to sunlight is a chemical change in which ultraviolet rays can break down the chemical bonds and thus fade the color of fabrics. The fastness values with respect to sunlight, detergent, dilute acid and alkali are shown in Table 1. During 3 times washing with 1% standard detergent and 0.01 N NaOH solutions, moderate fastness were observed in the both cases and the final fastness values were found to be 3. Appreciable alterations of color were observed in both cases [17]. Very slight alterations in color were observed after exposure to sunlight and acid treatment. After exposure to sunlight for 6 hours a day for three consecutive days and after 3 times washing with 0.01N HCl the final fastness values were found to be 4. The images of dyed silk with Acacia catechu based azo dye and the altered shades of the silk by sunlight, detergent, dilute acid and alkali are presented in fig. 4.

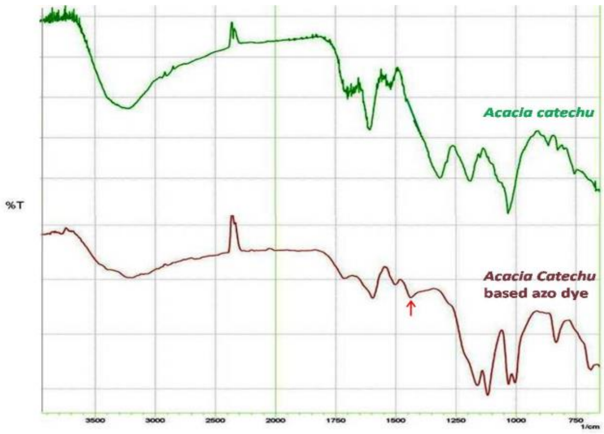

Figure 2: FTIR spectra of *Acacia catechu* and *Acacia Catechu* based azo dye

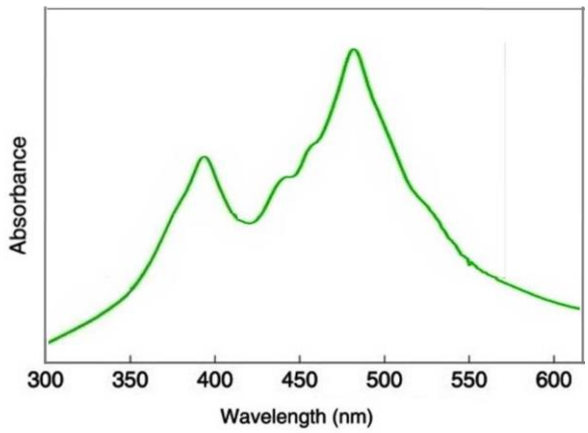

Figure 3: UV-VIS spectrum of *Acacia catechu* based azo dye

Table 1: Fastness reading of silk fabrics dyed with *Acacia Catechu* based azo dye

	Day-1	Day-2	Day-3
Light fastness	4/5	4	4
Wash fastness	4	3/4	3
Action of acid	4/5	4/5	4
Action of alkali	4	3	3

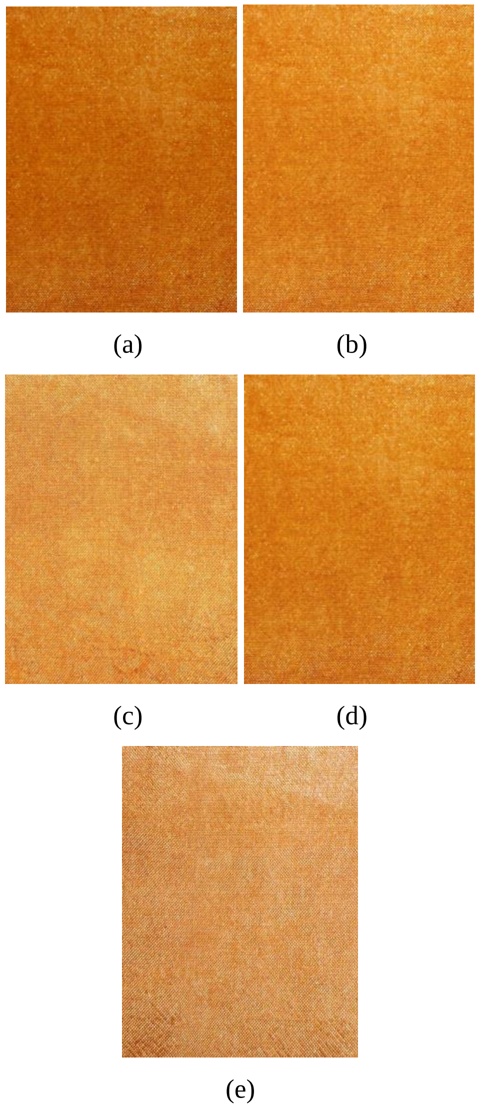

Figure 4: Images: (a) dyed silk with *Acacia catechu* based azo dye and the altered shades of the silk fabrics by (b) sunlight, (c) detergent, (d) acid, (e) alkali

IV. CONCLUSION

Azo dye was prepared by using the natural polyphenolic compounds of *Acacia catechu* through diazotization reaction. Formation of dye containing the azo group was confirmed by FTIR and UV-VIS spectra. The prepared dye was applied on silk fabrics and appreciable alterations in color were observed after the treatments with detergent and alkali. The color was found quite stable against sunlight and dilute acid. There might be a chance to reduce the environmental problem associated with azo dye by using the natural source based azo dye. However the environmental impact and toxicity of the dye should be investigated before its application.

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