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A MICROSCOPIC EXAMINATION TO MONITOR THE HISTORICAL PAPER DYEING TECHNIQUES

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ABSTRACT

The study aims to determine the type of technique used in dyeing old Arab paper (ca. 8th century A.D.) by examining a number of colored historical manuscripts and comparing them with modern samples by using a microscopic examination. Studying these historical references not only adds a new strategy to the identification of dyes and materials used in dyeing paper manuscripts under study, but also can help to preserve the colors by providing a better understanding of their composition, manufacture and use. SEM microscopy of archaeological dyed papers in different shades were evaluated and compared with the morphological structure of known dyed samples were dyed in different techniques. The type of dyeing used in dyed papers was determined according to the results of surface morphology investigation. SEM images confirmed that there are dramatic differences between dyed paper by beating and dyed by dipping, whereas, the dye molecules linked on the outer surface of the fiber in the dipping method. On the other hand, in the beating method, fibers were affected dramatically and were destructive. This work proved that dipping was the method used in the dyeing of Arabic manuscripts.

KEYWORDS: dipping dyeing, hearting, natural dyes, mordant, SEM, old manuscripts

1. INTRODUCTION

Paper made out of plant-like fibres was invented by the Chinese Cai Lun, who in 105 AD mixed textile fibers from the bark of the mulberry in water and produced sheets of paper from that. The invention of paper was one of the reasons of the successes of early China, with ease in governing the country, while it was, also, carried by Buddhist monks throughout the Central Asia. By the late 8thcentury, paper was being manufactured in Baghdad(the capital of the Abbasid caliphate in central Iraq), its use and manufacture was soon disseminated throughout the empire. Paper was introduced to Syria ca. 800, and it quickly spread throughout the Arab Mediterranean lands replacing papyrus and parchment, the two portable and flexible writing supports that had been used in the region for millennia after centuries the Muslims carried papermaking and paper to the Mediterranean region, and European Christians there learned how to make it by the twelfth Century (Adam, 2015; Adamo et al., 2003).The conversion of raw plants or textile waste into a pulp of cellulose fibers suitable for papermaking requires not only the raw materials but also a considerable amount of physical effort and time. The fibers are first washed to clean them and then soaked, fermented, or cooked to soften them; they are then beaten in water until they break down into a uniform pulp (stuff) that can be suspended in water for the actual formation of the sheet. Papermaking therefore requires an adequate and steady supply of pure water for manufacture (Adams, 2011).As the only way of bleaching fibers before the discovery of chlorine in the eighteenth century was to expose them to the sun, the preparation of white paper required either clean white fibers or a sunny climate. Many early Arab papers are decidedly tan or even brown. Although the Chinese character for paper suggests that its discovery was related to textile waste, for centuries the Chinese had used bast fibers extracted from such semi-tropical plants as papermulberry, jute, hemp, bamboo, and rattan, for papermaking, and papermakers in East Asia followed suit. For example, Japanese papermakers typically used kozo, mitsumata, gampi, and other plants that provided long fibers, which when seen under a microscope resemble "buttered spaghetti" and give the paper its characteristic strength and feel. Such plants did not grow in the arid climate of Central Asia, however, and papermakers there apparently discovered (or rediscovered) not only that paper could be made from rags and waste from textiles made from such plants as cotton, flax, and hemp, but that it was easier to make paper out of fibers that had already been processed and bleached in the sun. Muslim Arab papermakers consequently learned to make paper from both baste fibers and rags (Adelantado et al., 2005; Afsharpour et al, 2011).We should note that paper is amulti-component material, which is made of wood derived fibres composed of cellulose, hemicellulose, lignin and additives such as starch, gelatine, minerals, natural dyes or synthetic pigments. Paper production is divided into two historical periods: the first is the paper made before the 19th century which includes hand-made papers, and, the second is the paper made after the 19th century with the introduction of machine-made papers. Hand-made paper is manufactured from cotton, linen and hemp rags (Idoneet al., 2012). These materials are composed of pure cellulose fibres and sizing materials including starch and gelatin. The sizing materials are used for smoothing paper and make it ready for writing and prevent smearing of writing inks. Hand-made papers have a high degree of polymerization (DP) for the production of higher quality paper. The ranges of color of ancient papers vary from a creamy to a light brown there was due absence of a bleaching process, so. The demand for paper increased after the invention of the printing press in the 16th century by Gutenberg. When the Hollander beater was developed in 1680 AD, the quality of the paper sheet produced becomes a concern for paper conservators (Anders, 2013 and Angel et al., 2014). Hence, during the 19th century, machine-made papers were produced from wood pulp sources in which the earlier sizing materials are replaced by alum (potassium-aluminum sulfate), which increases acidity and accelerates the paper ageing over time. In ancient era people have been tried to add color to the world around them. They used natural matter to stain hides, fabrics, decorate shells and feathers, and paint their story on the walls of ancient caves. Scientists may never know where the art of dyeing originated, and the first people to dye their papers. It was mentioned that by 3000 B.C., dyeing was a well-developed art in Egypt and Yemen (Bloom & Blair, 2009, 302; Li 2005, 332). Early frag-ments, historical documents, and archaeological artifacts give evidence of the skill with which dyes of antiquity were used. Dyes and pigments as colorants are widely used in the textile, pharmaceutical, food, cosmetics, plastics, paint, ink, photographic and paper industries. A color additive is a substance capable of imparting its color to a given substrate, such as paint, paper or cotton, in which it is present. Dyes and pigments are the most important colorants used to add a color or to change the color of something. A dye must be soluble in the application medium, usually water, at some points during the coloration process. It will also usually exhibit some resistance for the material being dyed and be absorbed

from the aqueous solution. On the other hand, pigments are the colorants composed of particles that are insoluble in the application medium (Bogaard, 2017; Paual, 2001). Dyes were used in Persian manuscripts by two ways: a) to dye the paper support, a practice relatively common in Islamic manuscripts but rare in Western manuscripts; b) to impart delicate hues to particular details in miniatures. For what concerns paper dyeing, different colors could be obtained, frequently inside the same manuscript: dark blue (obtained with indigo), pale green (a mixture of indigo and a yellow dye), red (henna) and yellow. The practice of dyeing paper can be possibly explained with the desire of highlighting text over a proper background, such as gold text on a dark blue surface. For what specifically concerns miniatures, it has been found a wide use of anthraquinone dyes, these seem to be of animal origin, though using non-invasive techniques only it has not been possible to distinguish among the different scale insects possibly employed (Casoli et al., 2013). However, historical considerations make Armenian cochineal or lac dye, imported from India, the most suitable candidates. In yellow painted areas, the evidence of saffron was found, which is not surprising considering that Crocus sativus L. was cultivated in the Iranian area since Sasanian times (Bastone et al. 2017; Moropoulou and Zervos 2003; Zou et al., 1993).

Colorants can be divided into two groups: dyes and pigments. Dyes are generally defined as compounds that undergo chemical interaction with substrates such as paper and textiles (Giovanni et al., 2016; Goodwin, 1982).A dye molecule has two principal chemical groups: a chromophore and an auxochrome. The former usually incorporates an aromatic ring, which is associated with the coloring property, and the latter helps the dye molecule to combine with the substrate. Pigments are composed of solid and insoluble particles, which are suspended in media. They are responsible for maintaining the optical properties of the color (Gulrajani, 1992; Hilde and Piet, 2011). Both pigments and dyes are used to provide color to all sorts of substances and have been important to humans since the dawn of history. The difference between the two is that dyes are soluble in the substrate and thus disperse at a molecular level, while pigments are insoluble and are dispersed as particles. Dyes provide brighter color than conventional pigments, but they are less light stable and less permanent (Hassan, 2015).

Natural dyes, as lake pigments, have been widely applied in painting. For example, anthraquinones and their Hydroxy derivatives have been used as red dyes and pigment lakes from prehistoric times, and we can find written accounts of the use of anthraquinone reds and purples as dyes in ancient Egypt; anthraquinone lakes (e.g. madder red) were also very popular with Impressionist painters, including Vincent van Gogh. Lake pigments can be prepared by precipitating the dye extract with aluminium or other inorganic salts, such as alum. Pure dyes such as indigo were also used as painting materials, e.g. in medieval illuminations (Hassan, 2016).

Papermakers probably started early on to dye the paper pulp during processing in order to achieve more intense colors. The blue textile dyes; Woad, and especially logwood and litmus, were commonly used in paper coloring.

In the Islamic mediaeval era, papermakers were producing paper ranging in color from cream to dark cream and either grey or off-white in tone according to its production inputs, there is no record of color being added to the pulp during papermaking manufacturing process (Hassan, and Sabry, 2017). The copied original manuscript such as Al Safti, 1851AD indicated that the dyes were applied directly to the sheet of paper.

In the Islamic countries Paper was easier to color. This was usually done by immersing prepared sheets of paper in a vat of dye solution. The 11th century Zirid prince and calligrapher ibn Badis (1007-61) noted that color could also be to the size during the process of smoothing the surface.

Most European colored their papers by coloring the paper pulp in a vat of dye solution before making the paper sheet. Most of the paper dyeing techniques were carried out on un sized paper, because soaking a sheet in a dye solution or marbling would remove the sizing. These types of paper were sized after dyeing, followed by polishing. Recipes and texts describing paper dyeing in Iran dates from the 15$_{th}$ century. The main vegetable dyes were saffron, turmeric, safflower, lac, other red dyes, sapanwood, henna, pomegranate bark, indigo, and sunflower-croton. Pigments include verdigris, orpiment and blue vitriol. Mordants were often used to fix the colors during dyeing and to improve color fastness of dyes (Hassan, and Mansour, 2018).

Despite fundamental literature on the dyeing of textiles, there is a lack of research about dyeing and coloring paper pulp for paper conservation purposes. Papers dyed with natural dyes have the most effective antifungal properties. This new knowledge will assist paper conservators in making better choices to ensure the long term preservation of paper materials, with a greater resistance to fungal attack (Giorgi et al., 2002).

In spite of the vast amount of colored manuscripts, it is not known exactly how to add those dyes. Is it added after the formation of paper sheet or was added during the beating process .The present study attempts to identify the technology of adding dyes

into archaeological paper through microscopic examination of colored paper samples and comparing them to modern standard samples implemented in two ways of adding colors: first samples, the dye was added during beating and another was added after the formation of paper sheet.

2. MATERIAL AND METHODS

The objectives of this experimental study are: dyeing new paper samples (Cotton linter & Whatman) with different application methods (dipping & beating) by the natural dyes from local Egyptian market as a standard reference; using SEM in order to identify the method of dyeing in a rare colored manuscripts dating back to Islamic era.

2.1 Archaeological samples

Microscopic examination was carried out on three colored manuscript 1) a manuscript entitled: Supply of Fatah, explaining the light of clarification and the survival of spirits of the famous Arab Golf Sharnblali and carries a history dating back to 1745 AD - consisting of 233 pages and written for carbon ink and red vermillion on blue paper–the manuscript returns to the manuscripts Aziz Suryal Library of Alexandria University, The second manuscript is a poetry collection in Persian, which includes the Eastern-Persian language group of the traveler "Libar", which dates back to 1508 AD. It consists of 196 pages of iron ink on paper dyed in yellow- Library of Alexandria University, the third is a book of prayers and Award date back to 1600 AD, Othman - era with inventory number: 841/3444 in Al-Azhar Library, Cairo .The text block consists of 16 dyed paper leaves (blue and yellow) (see, Table 1 for all information of these manuscripts).

Table 2. shows all data concerned the studied manuscripts

Archaeological samples	Code of the manuscripts	Type of pulp	Type of dye	Date	Type of ink
A manuscript entitled: "Supply of Fatah, explaining the light of clarification and the survival of spirits	463	cotton	indigo	1745 AD	Carbon ink and red vermillion
A manuscript entitled: a poetry collection in Persian	80F	cotton	Turmeric	1508 AD	Iron ink
prayers and Award	841/3444	cotton	Indigo and turmeric	1600 AD	Iron ink, gold leaves and cinnabar

2.2. Modern samples and dyes

The samples analyzed are: A) Whatman filter paper (100% of pure cotton fibers – which similar the archaeological rag paper) no. 4 was Model quantitative filter Whatman ash less, grade 41 paper diam.55 mm was obtained from Sigma (Sigma–Aldrich, Mo, St. Louis, USA), B) Cotton linter pulp from Egyptian National Library and Archives (Dar al Kutub).

C) Dyes: two natural dyes were used indigo plant was obtained from Sigma (Sigma–Aldrich, Mo, St. Louis, USA)& turmeric roots sources were bought from Harraz (Agricultural, Seeds Medicinal plant Company) (see Fig.**1**).

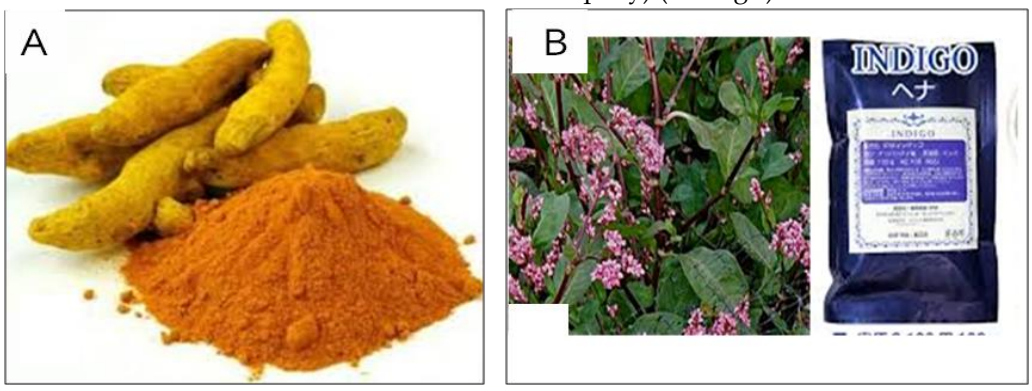

Figure 2. The natural dyes which used in the experimental part: (A) turmeric roots& (B) indigo plant.

2.3. Extraction of coloring material from dried turmeric roots

Aqueous extract from dried Turmeric roots (see Fig.3). In this method, plant dyestuff extraction was carried out by boiling the plant materials in deionized water for 1hour with stirring, maintaining a liquor ratio of 1g plant material to 20 mL of deionized water (Baglioni et al., 2009). Following this ratio, 20 g of turmeric powder was placed into beaker and 400 mL deionized water was added so that the plant material was completely covered. The contents

were brought to boiling and simmered on a hotplate for one hour. The dyeing liquor was allowed to cool and then the insoluble residue was removed by sedimentation and filtration through Whatman Cellulose Filter Paper, and then used for dyeing.

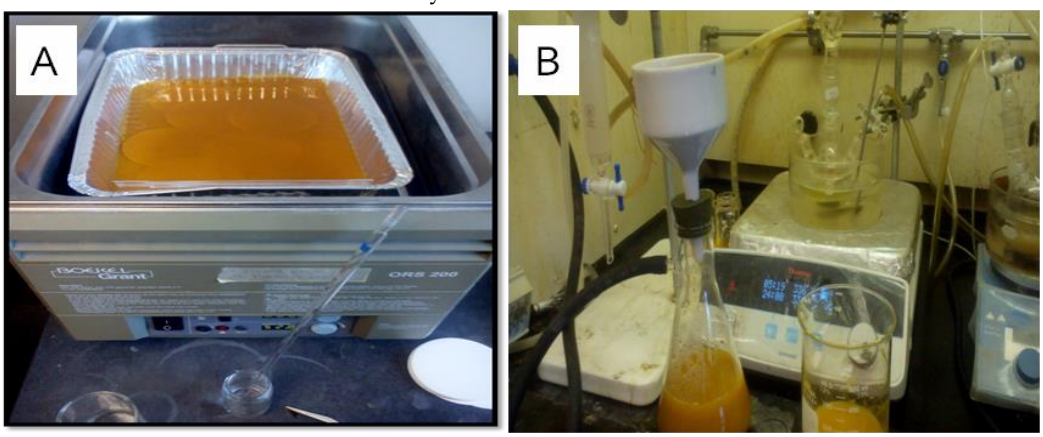

Figure 4. (A) & (B) Aqueous extract from of color from dried Turmeric roots.

2.4. Dyeing of what man filter paper

Paper sheet was entered into the dye bath containing the calculated amount of dye; water and required chemicals (Fig. 5). Treatment in the dye bath contains Mordant 3% at Temperature of 60°C for 30 minutes (60 ml of lemon juice was added to 600 ml of distilled water). Then the paper samples were treated with dye Mordants and dried, according to (Zou et al., 1994).

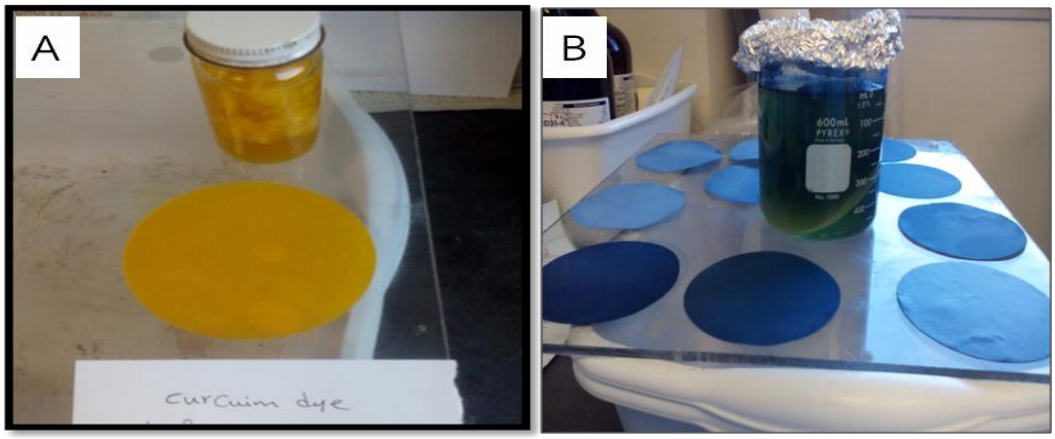

Figure 3. The paper sheets during & after dyeing with turmeric (A) and indigo dye (B) by immersion.

2.5. Dyeing cotton linter pulp

When the standard samples were prepared and dyed according to the specialized references in this field, heating was applied, thus, the pulp was dyed and the paper sheet was formed.

Dyeing of pulp

Dyeing was done using a liquor ratio of 1:55 mL/g so we used about 1.8 g of oven-dried pulp, and 100 mL of water) at various combinations of temperature (from 25 to 80 ◦C and from 15 to 60 min) according to Papadaki et al., 2014). Cotton linter pulp was put into a glass bottles and the dyeing process was performed using water bath (Fig.6).

the dye was dissolved in 1 litter of hot water then pour into pulp slurry while stirring (about 15-20 minutes). We let pulp stand for 1-2 hours and test for bleed. Finally strain process for the colored pulp and rinse by pouring several gallons (litters) of water through it.

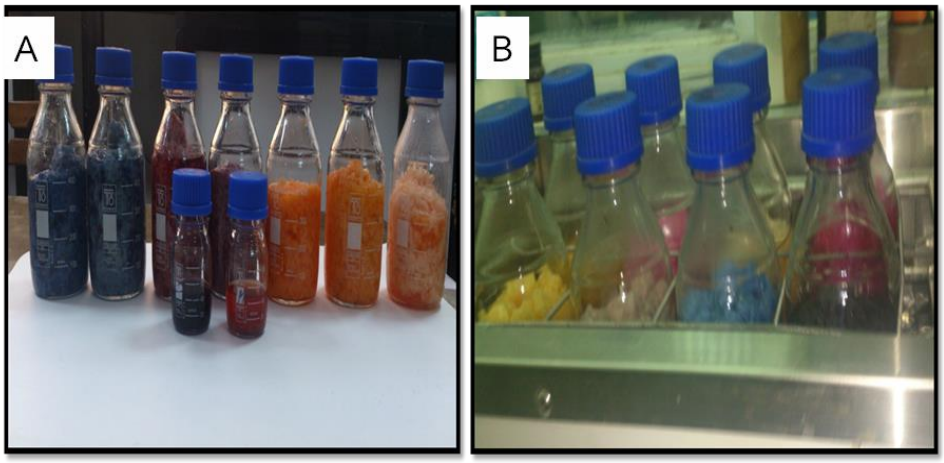

Figure 7. A and B show paper pulp during dyeing process.

Sheet formation

Pulp was beaten to 40° SR in a Jokro mill beater according to the Swedish Standard Method (SCA). Paper sheets of basis weight 80 g/m² were formed using leaf cast instrument.

The paper sheets (Fig.8) were prepared according to the S.C.A standard, using the model S.C.A sheet former.

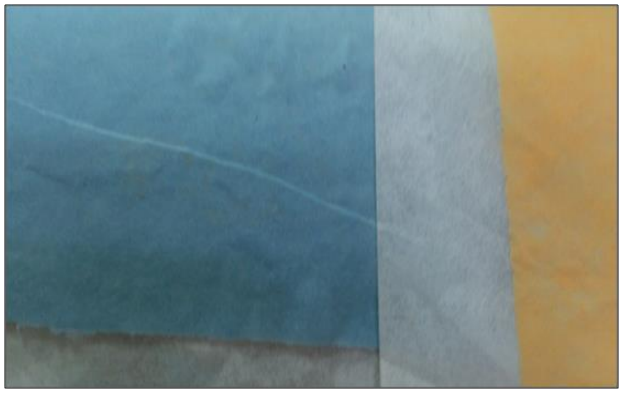

Figure 9. Paper sheets prepared on leaf-casting machine.

Artificial ageing

The natural ageing process of paper samples is too slow to observe the changes in paper materials in a manageable time scale (Othman et al., 2014). In order to simulate long-term deterioration processes over a manageable time scale, a moist heat artificial ageing test was performed on the dyed paper samples dyed with selected dyes. The paper samples were subjected to artificial ageing at 70°C and 65% RH in a climatic chamber for 12 days according to the Technical Association of Pulp and Paper Industry (TAPPI) Standard Test Method T 544 cm-08 (TAPPI 2008) (Poletto, et al., 2014). The standard test temperature was altered from 80 °C to 70 °C to accommodate the limitations of the climatic chamber.

2.6 SCANNING ELECTRON MICROSCOPY (SEM)

To detect the differences between dyeing methods for dyed modern and archaeological samples measurements were carried using an Electron Microscope Survey Nanoscience instrument FEI Company MVE00001162, FP 3950/00 at NCSU. All the samples examined were prepared according to the standard procedures. The samples were coated with gold using EMITECH K450X sputter coater to avoid charging. The SEM has a scan area of 50 mm x 50 mm as standard. Optionally, the motorized scan area can be upgraded to 100mmx100mm. The examination of samples was performed at the SEM Laboratory, Department of Polymer and Color Science, the University of North Carolina.

3. RESULTS AND DISCUSSION

Dyeing of the Arab paper is one of the artistic secrets of the Arab civilization where there are a limited number of historical sources to describe the dyes used , the most important references are : al-Safti's text (13th–15thcentury) and Simi Nishapuri's text (Tehran 15thcentury though they give 15 recipes for paper dyeing such as Henna , Red Onion Skins, Turmeric etc., but no information presented about method of application of these dyes (Derman, 1998). Hence, many scientific studies have emerged to identify the methods used in the dyeing of paper, but most of them agreed that the fabrics (cotton rags) were colored with various dyes then it used in making pulp and the others reported that the dyes were added during the beating process of pulp (Zidan et al., 2017; Abdel-Maksoud and al-Saad, 2009). But those studies relied on historical sources and therefore during the current experimental study we can identify these methods simply, based on concrete physical evidence so, we show the observations of the microscopic examination as follows:

Modern dyed paper by beating

From Fig.10 one can note that the surface is uneven and not much different from the reference cotton fiber with many particles on the surface of the fibers in addition to the apparent breakage of fibers furthermore, there are a dramatic difference between turmeric dye and indigo dye in the penetration mechanism where the turmeric dye solution is penetrated completely in the primary cells of cotton fibers while in dyed paper by indigo dye we can observe the notable deposition of the dye on the outer surface of the fibers and between the porous spaces of the pulp fibers, The cracks may have arisen from the effect of accelerated aging on paper fibers.

Examination of dyed modern samples by dipping

The microscopic image (Fig. 11) indicated that application of the dye by immersion leaves a clear color on the surface of the fibers where the surface is covered with a homogeneous layer of dye molecules We also note the distinctive light color for fiber as a result of the presence of deposits of dye molecules.

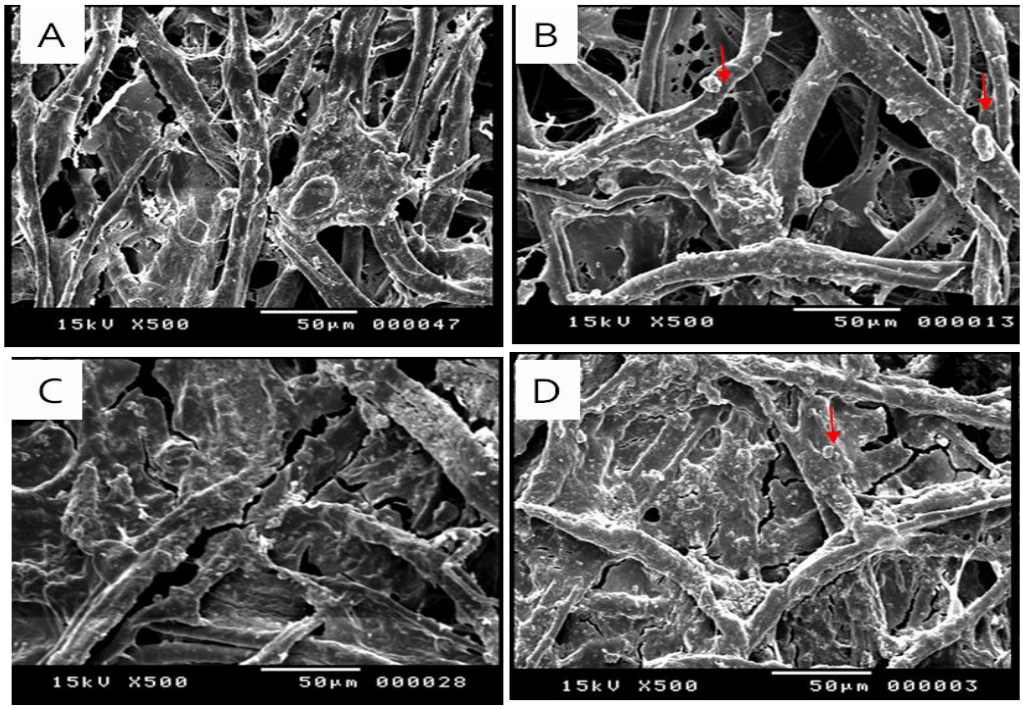

Figure 12. Micrographs of dyed paper by the beating with pulp (A&B) turmeric dye and (C&D) indigo. One may observe the destruction and random distribution of the fibre structures, the erosion of the fibres and many bores. There was total deformation of the surface morphology, attributed to the dye kneading processes with the pulp along with the physical weathering factors.

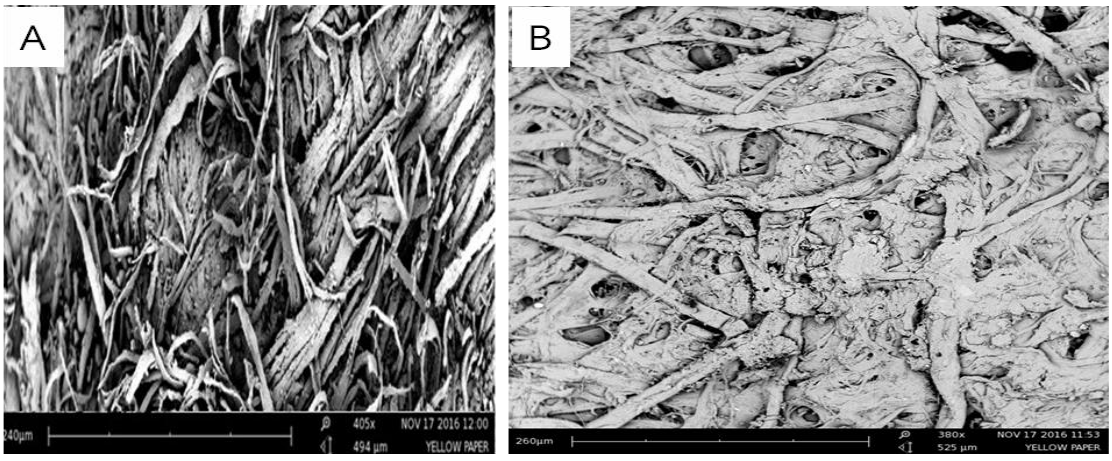

Figure13. Micrographs of dyed paper by the dipping (A) turmeric dye and (B) indigo dye. Fundamental differences are noted between the indigo dye and the turmeric dye, where the density of the indigo dye particles is high with a thick coverage, unlike turmeric dye.

Archaeological samples

Microscopic graphs of yellow dyed paper (Fig. 14 and Fig. 15) clearly indicate the archaeological paper is similar to the aged modern that performed in an dipping method in terms of the shape of the fiber and the nature of the presence of dye molecules on the outer surface of the fiber indicating that the yellow color was applied in the style of dipping and not dyed pulp paper. In addition to the great color brightness of the archaeological samples as previously observed in modern samples (Fig. 16) that carried out by dipping.

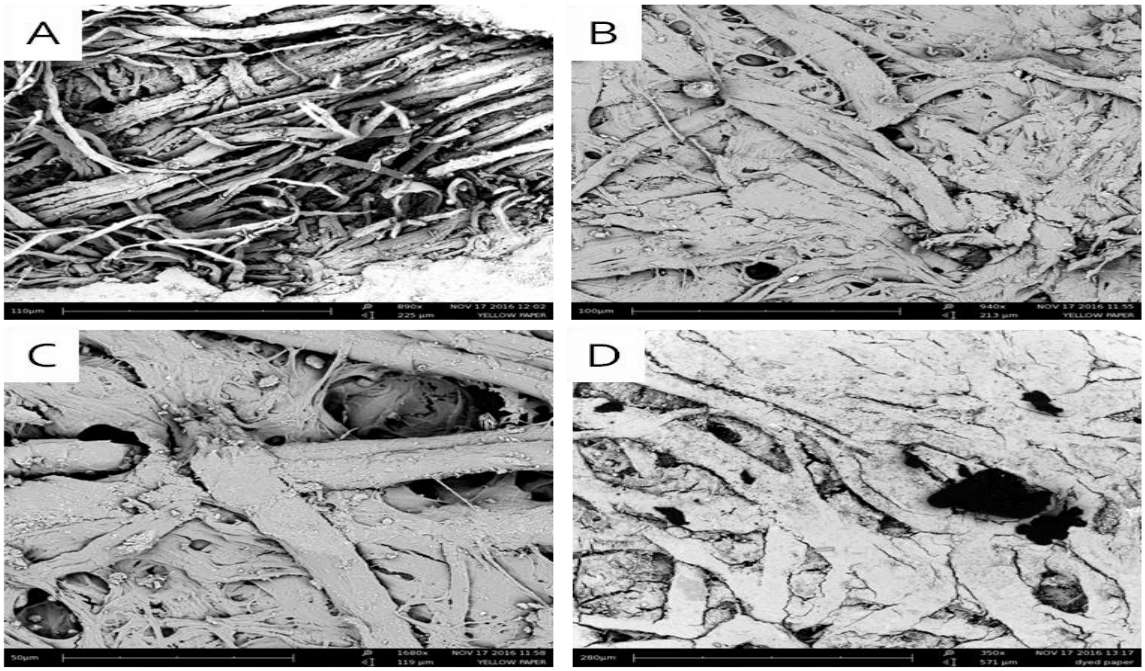

Figure 17. Micrographs of archaeological yellow dyed paper (A&B) dyed paper from a manuscript N. 2 (a poetry collection in Persian) and (C&D) dyed paper from a manuscript N. 3(a book of prayers and Award).The shape did not differ with the blue archaeological samples where the characteristic features of the dipping method appeared on the paper fibers, which manifested in the brightness and the dye layer which cover the walls of paper cells.

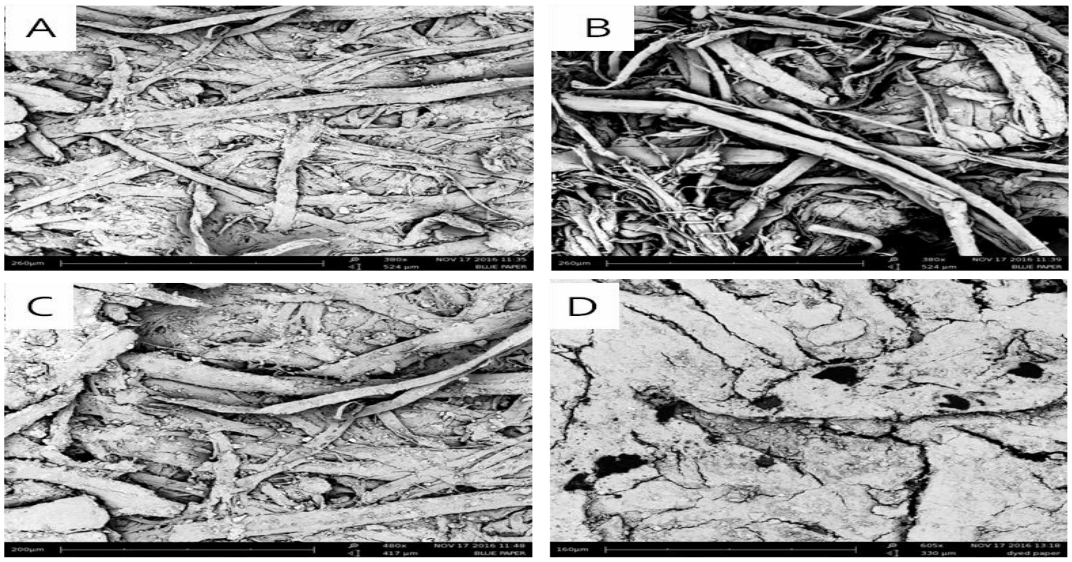

Figure 18. Micrographs of archaeological blue dyed paper (A&B) dyed paper from a manuscript N. 1 (entitled: Supply of Fatah, explaining the light of clarification) and (C&D) dyed paper from a manuscript N. 3(a book of prayers and Award) (NB: The figure shows that the manuscript paper was probably made from cotton, because cotton fibres are long; the length ranges between 10 and 65 nm. Some contaminations (Fig. 9B) from stains and dusts were noticed on the surface of the original paper. Damages caused by physical factors (Fig. 9D) appeared in the form of bores, and the tearing of paper fibres and deformation of the paper appearance were also noted. Image 9D also revealed the dye molecules that completely cover the fibers).

4. CONCLUSION

The microscopic study showed much absent information about the technology of dyeing paper in ancient era; it can be summarized in the following points:

i) Fibers of dyed modern samples by beating with pulp were affected dramatically; the fiber is destructive and the sprains were stretched along the fiber. Some fibers have a noticeable shrinkage due to pulping process, In addition to fragmentation and divisions and extended cracks. As for the archaeological sample, the fiber was less fragmented and may be attributed to the nature of primitive beating machines that were used in ancient times.

ii) In dipping dyeing method , the surface of fibers are completely covered with dye particles even the pores not detected due to the deep covering , unlike the beating the dye with pulp, the dye granules are mixed with fiber walls and are no longer noticeable.

iii) Microscopic evidence has shown that the archaeological paper in the third manuscripts, whether yellow (imprinted with turmeric) or blue (indigo dye) were carried out by dipping. Our investigations agreed with (Derman, 1998) who explained the steps and materials used in Ottoman times; the dye was boiled in water which was then poured into a vat. Then paper sheet was soaked in the dye solution until it colored with desired shades and finally let the paper sheet to dry.

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