

Application of Frankincense and Rice Starch as Eco-Friendly Substances for the Resizing of Paper as a Conservation Practice

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Smart, environmentally friendly alternatives, *i.e.*, frankincense and rice starch, are recommended for usage in modern paper conservation processes during the re-sizing process treatments. Different concentrations of frankincense and rice starch were applied to paper samples before and after ageing. Multiple analysis methods were performed to ensure the effectiveness of these materials. Promising results were found, but at varying degrees according to the type and concentration of the materials. Scanning electron microscopy illustrated that the frankincense particles were completely absorbed into the cell walls after ageing. Results indicated that there was no considerable change in pH before and after treatment or ageing; the best results for decreasing the acidity utilized a treatment with a mixture of frankincense and rice starch in a 2 to 1 ratio (F₂S₁). Fourier transform infrared spectroscopy illustrated an increased CH₂ region and decreased OH stretching as a result of the bonds formed from the starch and crystals formed by frankincense, which agreed with the increased coating and strength of the paper fibers. The total color change values of all the treated samples after ageing were less than 4.5. Frankincense was found to provide strength in supporting wood fibers.

Keywords: Wood pulp; Paper; Resizing; Ageing; Frankincense; Starch; FTIR; SEM

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INTRODUCTION

Plant sources such as cotton, flax, jute fibers, ramie, and finally wood-derived fibers have been the main source for paper making from the beginning of its invention until the present time (Dillen *et al.* 2016). However, its method of manufacture is quite different, since paper forms as a layer when a suspension of disintegrated paper pulp is allowed to settle, and the water is drained off (Taha *et al.* 2019; Hassan *et al.* 2021). The invention of paper is credited to the Chinese (Hassan *et al.* 2020). Plant fibers are the most commonly used material for paper manufacturing and at present, these come from wood pulp. However, before the 20th century, linen and cotton rags were used. Wood pulp is the principal fibrous raw material from which paper is made, of which cellulose, the major

component of papermaking fibers, contributes 40% to 45% of the dry weight of the wood. In addition, wood paper is composed of cellulose, lignin, hemicellulose, and some extractives (Bajpai 2018).

Paper exposes many factors contributing to deterioration factors, and these can be divided into three categories: 1) physical factors, which include moisture, temperature, and light (Hassan *et al.* 2020). These effects affect paper during droughts and humid periods, which results in fragility, microorganism growth, acceleration of chemical reactions, changes in dimensions, and shrinking and swelling (Moncmanová 2007; Hassan 2015); 2) chemical factors, which include gaseous pollutants and salts that increase the degree of acidity and results in oxidation-reduction reactions (Hassan *et al.* 2020); and 3) biological factors, *e.g.*, insects, rodents, fungi, and bacteria (Bankole 2010; Mansour *et al.* 2017). The effect of biological factors is summarized in cellulose hydrolysis, pigment stain, as well as the loss of portions of the paper and holes (Björdal *et al.* 1999; Björdal 2012; Liu *et al.* 2018). Since these deterioration factors occur in paper, there are natural materials, *e.g.*, gelatin, natural extracts, plant biomass, and glue, and synthetic materials, *e.g.*, methyl cellulose, polyvinyl alcohol, and methyl hydroxy ethyl cellulose, that are used for the resizing of paper (Evetts *et al.* 1989; Spitzmueller 1992; Ravines and Faurie 1993; Kolbe 2004; Ardelean *et al.* 2007; Baker 2007; Hummert *et al.* 2013; Ciolacu *et al.* 2017; Mohamed *et al.* 2019; Salim *et al.* 2020; Hassan *et al.* 2021; Mansour *et al.* 2021a,b; Taha *et al.* 2021). The term re-sizing means that the historical manuscripts and documents are covered with an aqueous solution of the sizing agent. The original sizing agents, applied to paper during the initial manufacturing, tend to decompose with time. Therefore, conservators routinely replace or supplement the lost sizing agent by natural or synthetic materials called resizing agents.

Gelatin is a protein obtained from the hard and soft connective tissues of animals. It became used for paper treatment in the second half of the 13th century (Pataki 2009). It was used to prevent the bleeding of inks, as well as for its ability to improve various properties of paper, *e.g.*, its strength and resistance to degradation and contamination. It also plays a beneficial role at the molecular level by neutralizing the rate of cellulose degradation due to ageing (Missori *et al.* 2006; Irvine *et al.* 2015; Huang *et al.* 2018).

Animal gums are produced from a wide range of animal tissues, *e.g.*, leather, bones, *etc.* The gums are proteins that consist of many amino acids. There are also types of vegetable glue, and a 25% solution of vegetable glue in water was used as a sizing solution for paper. These substances also have been used in painting media, coatings and grounds, in the gilding of illuminated manuscripts, and pastel fixatives (Missori *et al.* 2006). Paraloid B-72 (Acryloid) is one of the most frequently used acrylic polymers, employed for several object arts, as well as to wood and paper to improve their mechanical characteristics (Pataki 2009; Mansour *et al.* 2015; Mansour and Salem 2015; Vinçotte *et al.* 2019; Abo Elgat *et al.* 2020; Kovács *et al.* 2021).

Some synthetic chemicals are useful for the sizing of paper. The idea is that these synthetic chemicals dissolve in organic solvents and do not affect the ink. These synthetic chemicals include soluble nylon, which is a form of nylon called methyl methoxynylon. This chemical is soluble in methyl alcohol, ethyl alcohol, or methylated spirits, hence the name soluble nylon (Agrawal and Barkeshli 1997). Methylcellulose, unlike glue and gelatin, is soluble in cold water. It is also soluble in some organic solvents, *e.g.*, dimethylformamide and dimethylsulphoxide. One great advantage of methylcellulose is its resistance to the growth of fungus (Nasatto *et al.* 2015). Methyl hydroxyethyl cellulose, which has some very desirable properties, can be dissolved in water and has also been used

as a resizing material (Agrawal and Barkeshli 1997). The mechanical properties between paper components can be improved by increasing the crosslinking between the cellulose fibers, which has been achieved with several polymers, *e.g.*, cellulose acetate, polymethyl acrylate, and phenol-formaldehyde resin solutions (Manjunath and Sailaja 2014; Saha *et al.* 2016; Meesorn *et al.* 2017; Kumar *et al.* 2021). Cationic starch, polyacrylamide, and polyvinyl alcohol (PVA) have also been used for the sizing of paper (Liu *et al.* 2010; Hassan and Mohamed 2017; Seo *et al.* 2020; Xia *et al.* 2020).

Currently, with trends heading toward green methods that are utilized to help preserve the environment, the use of eco-friendly materials is more prevalent. These materials maintain the health of conservator specialists, are harmless to the environment, and are safe to use. As such, rice starch and frankincense have been considered for study in terms of paper resizing.

Pure starch generally contains, by weight, 20% to 25% amylose and 75% to 80% amylopectin (Brown and Poon 2016). Historically, a pure extracted wheat starch paste was possibly used in Ancient Egypt to glue papyrus. More recently, it has been used in the paper industry for its strong bonding properties between the pulp fibers. The starch works to strengthen the paper and provide it resistance to water absorption, in addition to increasing paper smoothness and improving printing properties. As such, it is used for the surface treatment of paper (Hunter 1978; Hubbe and Bowden 2009).

The structure and the properties of starch are well known, and there is considerable usage throughout history of starch as an eco-friendly resizing material. One of the disadvantages of using starch as a re-sizing material is that it is an attractive substance for microbiological deterioration. In addition, it can also cause color change.

Frankincense obtained from *Boswellia* trees (family Burseraceae), particularly *Boswellia sacra* Flueck. is primarily composed of a mucus-like cluster (12% to 23%), essential oil (5% to 15%), and a lipophilic part (55% to 66%). This composition has shown quite a bit of variation, depending on the species and grade of the resin. Frankincense contains volatile compounds, with more than 20 monoterpenes and 28 sesquiterpenes. The lipophilic parts are composed of terpenoids, including Boswellia acids (BAs), which are the chemotaxonomic markers of *Boswellia* (Ali *et al.* 2008; El-Nagerabi *et al.* 2013; Al-Harrasi *et al.* 2019; Rajabian *et al.* 2020). This substance has been used throughout history in medicine, herbalism, *etc.*, but until now it has not been used in the field of archaeology. In view of these different and promising components of frankincense, it was presented as a re-sizing agent in this study, as it contains resinous materials similar in properties to gum Arabic, which can provide the protection coating and strengthening required for damaged paper, in addition to its anti-oxidation properties (Al-Harrasi *et al.* 2013). As such, it has been chosen for this project to determine whether it is suitable for use as an eco-friendly re-sizing material applied to paper. Furthermore, many studies have reported its anticancer, anti-inflammatory, immunomodulatory, and antimicrobial properties (Al-Yasiry and Kiczorowska 2016). Re-sizing of ancient paper depends on the use of gelatin or industrial cellulose derivatives, each of which carries many defects. Therefore, the current study is promising in terms of incorporating new natural components in the field of manuscript restoration, and even in the field of papermaking. Thus, frankincense and rice starch were employed for first time in such treatments. This may effectively address the defects of traditional resizing materials and enhance the chemical properties of wood paper fibers.

The aim of this study is to propose and evaluate new materials for resizing paper manuscripts using eco-friendly materials and to demonstrate the effectiveness of these materials by using a set of examinations, *i.e.*, digital and scanning electron (SEM)

microscopy, in addition to Fourier transform infrared spectroscopy (FTIR), pH measurements, and color change measurements.

EXPERIMENTAL

Samples and Artificial Ageing

A group of sulfate pulp paper sheets (40 g/m²) were brought from the Egyptian National Library and Archives and were exposed to artificial ageing at the National Institute of Standards (NIS), Giza, Egypt for this study. The artificial ageing process (both before and after treatment) was at a temperature of 80 °C and a relative humidity of 65% that lasted for 5 days. This was equivalent to 25 years of natural ageing according to ISO standard 5630-3 (1996). Afterwards, six samples were cut from the paper sheets with 5 cm × 10 cm dimensions.

Preparation of the Iron Gall Ink and Gum Arabic

The ink was prepared by adding gallic acid and tannic acid with some grains of ferrous sulfate to distilled water, after which gum Arabic was added (Jančovičová *et al.* 2007). The gum Arabic was prepared by grinding the crystals and then melting them in a hot water bath at a temperature of 80 °C and stirring until complete melting; the mixture was then filtered to remove impurities (Hidalgo *et al.* 2018). The iron gall ink was applied to the six previously cut samples with a brush to the one half of the samples, and the other half was left without ink. All samples were left out until they become completely dry, after which the different concentrations of the sizing materials were applied on the paper samples by brush (Figs. 1 and 2).

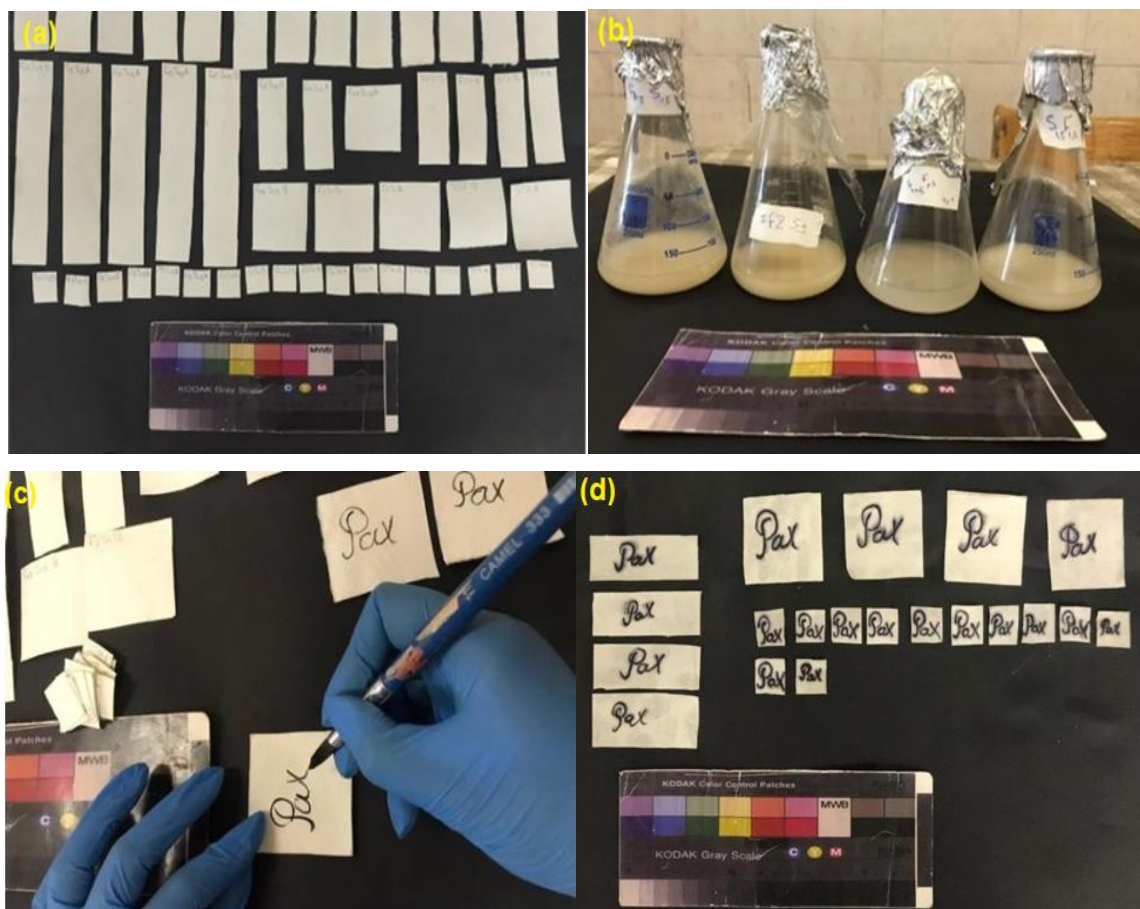
Preparation of the Starch and Frankincense

Rice starch was used as a resizing material and was produced by Al Mashreq Gardens for Trading and Distribution, Cairo, Egypt. It contains 5.95 g of protein, 1.42 g of fats, 80.13 g of carbohydrates, 10 g of calcium, and 0.35 g of iron. In addition to using rice starch as a resizing material, frankincense (*Boswellia serrata*) gum was also used, which was produced by Herbal House Centers Company.

Six rice starch (S) and frankincense (F) solutions were prepared with different concentrations (as shown in Table 1). Frankincense and starch were mixed to prepare different solutions according to the proportions in the table. The samples were prepared by applying various concentrations in 100 mL of distilled water and melted in a hot water bath by stirring these substances until complete melting and then filtered to remove any impurities. It should be noted that mixtures of rice starch and frankincense were employed without using either material alone for two reasons: 1) the common belief about the rapid decomposition of starch; and 2) the authors tested several primary samples with only starch and found the paper fibers became brittle, especially at high concentrations. Therefore, only mixtures of rice starch (S) and frankincense (F) were used. Note also that the highest concentration was 2%. This was based on preliminary experiments. There was a pre-test to determine if a higher concentration would be better, but the test showed that concentrations higher than 2% for this material resulted in an increase of the viscosity of the solutions, and this hinders the application process and penetration into the paper structure.

Table 1. Different Concentrations Used as Resizing Materials for the Treatment of Paper samples

Sample Number	Frankincense Concentration (gm)	Rice Starch Concentration (gm)	Dissolving solution (mL)	Surface area for each concentration
1	0.5	1.5	All resizing materials were dissolved in 100 mL of distilled water to produce different concentrations.	Each concentration in 100 mL water applied on surface 1.84 m ²
2	1	1		
3	1	2		
4	1.5	0.5		
5	1.5	1.5		
6	2	1		
7	1.75	0.25		

**Fig. 1.** Steps of the experimental study: (a) the cutting of samples; (b) the preparation of the treated materials; (c) writing with iron gall ink; and (d) the shapes of the samples after treatment.

After the application of ink to one side of the paper samples, the samples were exposed to heat for 3 h to determine the best treatment concentrations. The results showed that the samples with a high concentration of rice starch were missing water, resulting in solid content. Therefore, the concentrations $F_{1.5}S_{1.5}$, $F_{1}S_{2}$, and $F_{1}S_{1}$ were excluded (Egharevba 2019), while the concentration $F_{2}S_{1}$ showed the best result among other concentrations ($F_{2}S_{1}$ - $F_{1.5}S_{0.5}$ - $F_{0.5}S_{1.5}$). In addition, extra attention was given to the $F_{1.75}S_{0.25}$ treatment. Therefore, four concentrations were used to treat the paper samples (Fig. 1).

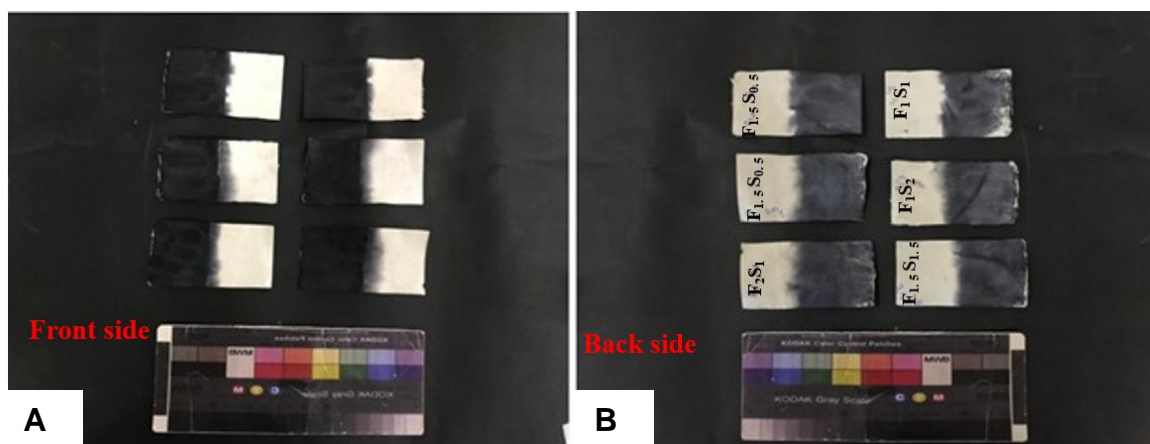


Fig. 2. The 6 samples after the application of ink and the treatment materials

Next, 2 cm × 2 cm samples were cut from wood paper to be analyzed *via* SEM and infrared analysis. Samples with 5 cm × 5 cm dimensions were cut and used in the universal serial bus (USB) microscope and color change analysis. In addition, the samples used for the pH analysis were cut to 2.5 cm × 7.5 cm. After that, the samples all had a Latin word (*Pax*, *i.e.*, peace) written on them with iron gall ink. The treatment materials were applied at different concentrations (F_2S_1 , $F_{1.75}S_{0.25}$, $F_{1.5}S_{0.5}$, and $F_{0.5}S_{1.5}$), which were allowed to completely dry, and then half of the samples were exposed to artificial ageing (Fig. 2).

The concentrations used are as follows: F_2S_1B , $F_{1.75}S_{0.25}B$, $F_{1.5}S_{0.5}A$, F_2S_1A , $F_{0.5}S_{1.5}A$, $F_{0.5}S_{1.5}B$, $F_{1.75}S_{0.25}A$, and $F_{1.5}S_{0.5}B$. A USB microscope and SEM microscope were used to examine the samples and to detect any changes that occurred in the treated samples before and after ageing. In addition, FTIR analysis, pH measurements, and color change measurements were in this study.

Universal Serial Bus (USB) Digital Microscope

A USB digital microscope (200x) (PZ01, Shenzhen Super Eyes Co., Ltd., Guangdong, China) was used for the visual assessment of the experimental samples.

Scanning Electron Microscope

The paper samples were examined using a JEOLJSM 5400LV EDX Link ISIS-Oxford Detector High Vacuum), and the samples were coated with gold.

pH Measurement

The pH meter model, made in Romania, is a waterproof temp pocket tester with a replaceable probe. It is located at the lab of manuscripts, Faculty of Archaeology, Cairo University. The samples were cut into 50 g pieces for each treatment and placed into 40 mL of distilled water (a pH of 7) for 1 h to measure the paper acidity using the cold extraction method. However, the pH value was initially measured *via* a method at room temperature according to ASTM standard D778 – 97 (1997).

Color Change Using the CIELAB System

The colors of the samples were measured with an Optimatch 3100 (CE 3100, SDL, United Kingdom). All samples were measured in the visible region, *i.e.*, in a wavelength range of 400 to 700 nm, with an interval of 10 nm, using a D65 light source and an observed

angle of 10° . The colorimetric coordinates L , a , and b of the CIELAB color space were used to express color change. The CIELAB color space is organized in a cubic form. The L axis runs from top to bottom. The maximum for L is 100, which represents white, while the minimum for L is zero, which represents black. The a and b axes have no specific numerical limits; positive a is red, negative a is green, positive b is yellow, and negative b is blue (Sehlstedt-Persson 2005). The total color change of all the treated paper samples was expressed as ΔE , which was calculated according to Eq. 1,

$$\Delta E = \sqrt{(\Delta L)^2 + (\Delta a)^2 + (\Delta b)^2} \quad (1)$$

where $(\Delta L)^2$, $(\Delta a)^2$, and $(\Delta b)^2$ are the differences between the values of the respective color indices before and after treatment.

Fourier Transform Infrared (FTIR) Analysis

Fourier transform infrared analysis was used to monitor the chemical composition and changes that occurred in the paper due to treatment. The samples were analyzed with an FTIR spectrometer (Model 6100, Jasco, Tokyo, Japan). The spectra were obtained in transmission mode with a triglycine sulfate (TGS) detector via the KBr method and represent 2 mm/s co-added scans in the spectral region from 4000 to 400 cm^{-1} , with a resolution of 4 cm^{-1} (Salim *et al.* 2020).

Water Absorption

The samples were cut into 3 cm \times 3 cm pieces (Fig. 3), and then the dry weight of the sample was recorded. After that, the pieces were placed in Petri dishes and 12 mL of distilled water was added (at a temperature of $23^\circ\text{C} \pm 1^\circ\text{C}$ (73.4°F)) and left for 120 sec. Afterwards its wet weight was recorded, according to TAPPI standard T441 om-09 (2013). The water absorption was estimated by using three samples from the same concentration, and the average was calculated and according to Eq. 2,

$$\text{Water Absorption (\%)} = \frac{WW - WD}{WD} \times 100 \quad (2)$$

where WW is the wet weight (g), and WD is the dry weight (g).

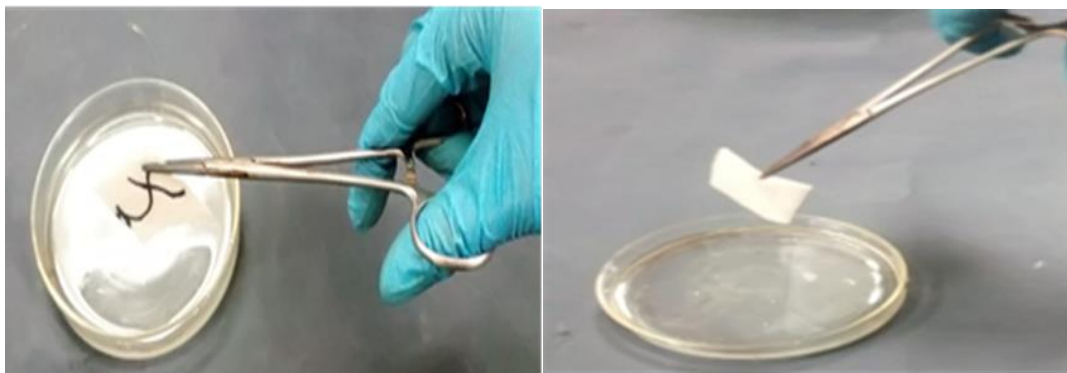


Fig. 3. Preparing the samples and immersing them in distilled water before weighing

Statistical Analysis

The water absorption data was statistically measured using one-way ANOVA, and the comparison among means was performed using LSD at a 0.05 level of probability (SAS 2001).

RESULTS AND DISCUSSION

Universal Serial Bus (USB) Digital Microscope

At first, no visual changes were detected by eye, but when comparing the standard samples before and after the ageing process using USB Digital Microscope, it was found that the pores in the paper were larger after undergoing artificial ageing. In addition, it was found that the paper used was poorly manufactured due to the appearance of stains and dirt (Fig. 4).

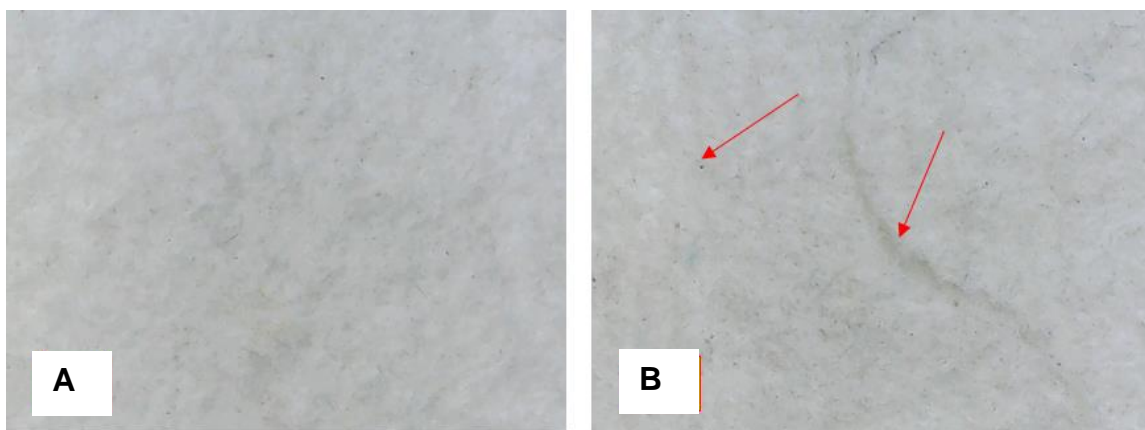


Fig. 4. Surface of standard sample by USB microscope before and after ageing: (A) before; (B) after

Using the treated materials at different concentrations, it was found that frankincense and rice starch gave best results at three concentrations ($F_{1.5}S_{0.5}$, $F_{0.5}S_{1.5}$, and $F_{2}S_{1}$). It was apparent by the naked eye that the increase in frankincense concentration after treatment gave the surface of the paper an unwanted shine. In addition, it was found that by increasing the rice starch concentration, the surface became slightly whiter, as shown in Fig. 5. After performing the microscopy examination, it was found that the three best concentrations were $F_{1.5}S_{0.5}$, $F_{0.5}S_{1.5}$, and $F_{2}S_{1}$. After the treatment, it was observed that one of the $F_{1.75}S_{0.25}$ samples was accidentally exposed to some drops of distilled water and after seven days, it showed green fungal stains. This hinted at the possibility of the treated material being susceptible to fungal attack (Fig. 6), although some studies mentioned that frankincense oil has antifungal properties (Al-Yasiry and Kiczorowska 2016). Furthermore, it was observed that higher concentration of rice starch compared to frankincense ($F_{0.5}S_{1.5}$) made the sample slightly whiter than the standard sample after artificial ageing (Fig. 7).



Fig. 5. Images show that with an increase of rice starch concentration, the surface became slightly whiter. Part (A) shows a specimen with low concentration of starch (sample F_{1.5}S_{0.5}), while (B) shows results with a high concentration of starch (sample F_{0.5}S_{1.5}).

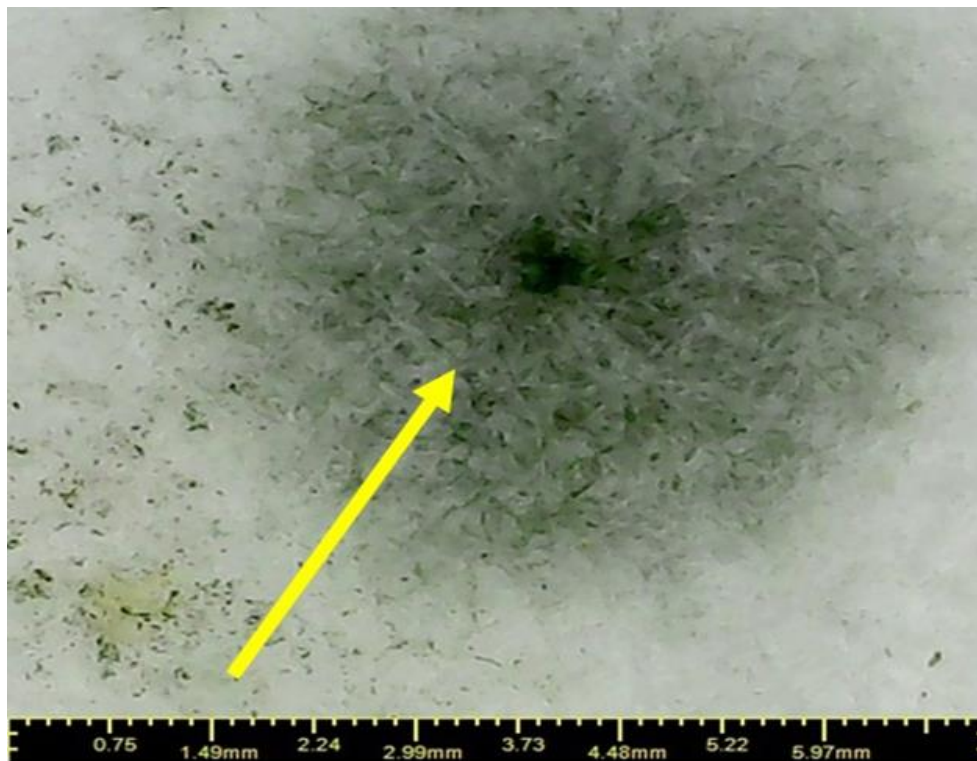


Fig. 6. Fungal stains on sample F_{1.75}S_{0.25} due to the exposure of the treated material to water

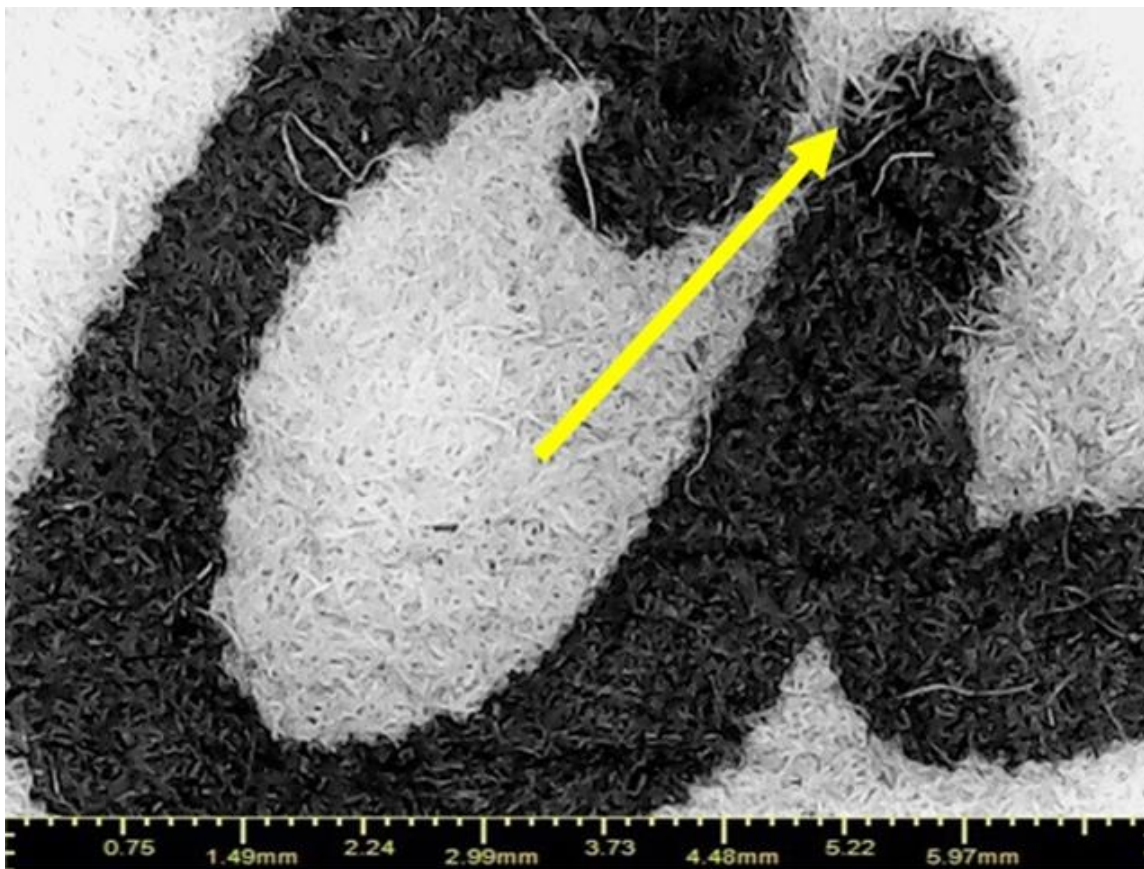


Fig. 7. Fibers curl in the $F_{0.5}S_{1.5}$ sample; the presence of a higher concentration of rice starch compared to frankincense made the sample slightly whiter than the standard sample after artificial ageing

Scanning Electron Microscope

Scanning electron microscopy was used to detect the shape of the wood fibers, the effect of artificial ageing on the fibers, and the effect of the treatment materials. Figure 8 shows the effect of the artificial ageing process on the wood fibers, *i.e.*, its deterioration in the form of the loss of water content and shrinkage, in addition to the weakness of the fibers and gaps (Zervos 2010). It was also observable that rice starch created connections and bridges of starch molecules between the cellulose bundles. This supported the observation that viscosity of the starch at the time of application was suitable for easily penetrating the walls of the wood cells. However, the frankincense treatment led to the formation of crystals that coated the fibers, which were not noticeable in the standard sample (Fig. 9), as they melted due to the heat of the artificial ageing process (Edwards and Falk 1997; Cruz-Cañizares *et al.* 2005; Shi *et al.* 2012) (Fig. 10). It was observed that the frankincense particles were completely absorbed by the cell walls (Fig. 10c and 10d) (Amagliani *et al.* 2016). Therefore, frankincense utilizes its full strength in supporting the wood fibers with exposure to the ageing process, which is a qualitative advantage of frankincense. It can be seen from Fig. 11 that when the concentration of frankincense to starch was increased to 1.5%, the bonding between the fiber bundles was strong and a clear improvement appeared in the structure of the paper, as the thickness of the fibers increased.

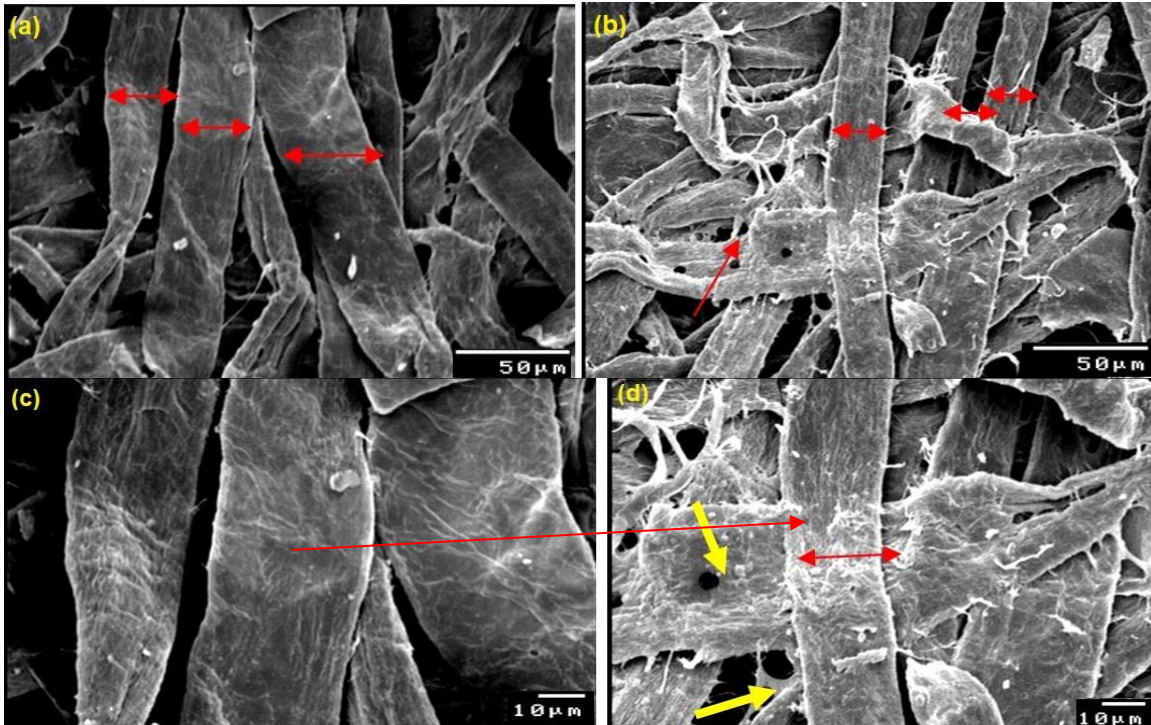


Fig. 8. Deterioration of the wood fibers due to artificial ageing in the form of shrinkage and fragility in wood fiber (b and d) in comparison to a standard sample before artificial ageing (a and c). By comparing the images c and d, a clear shrinkage in the fibers was apparent, as shown by the big red arrows.

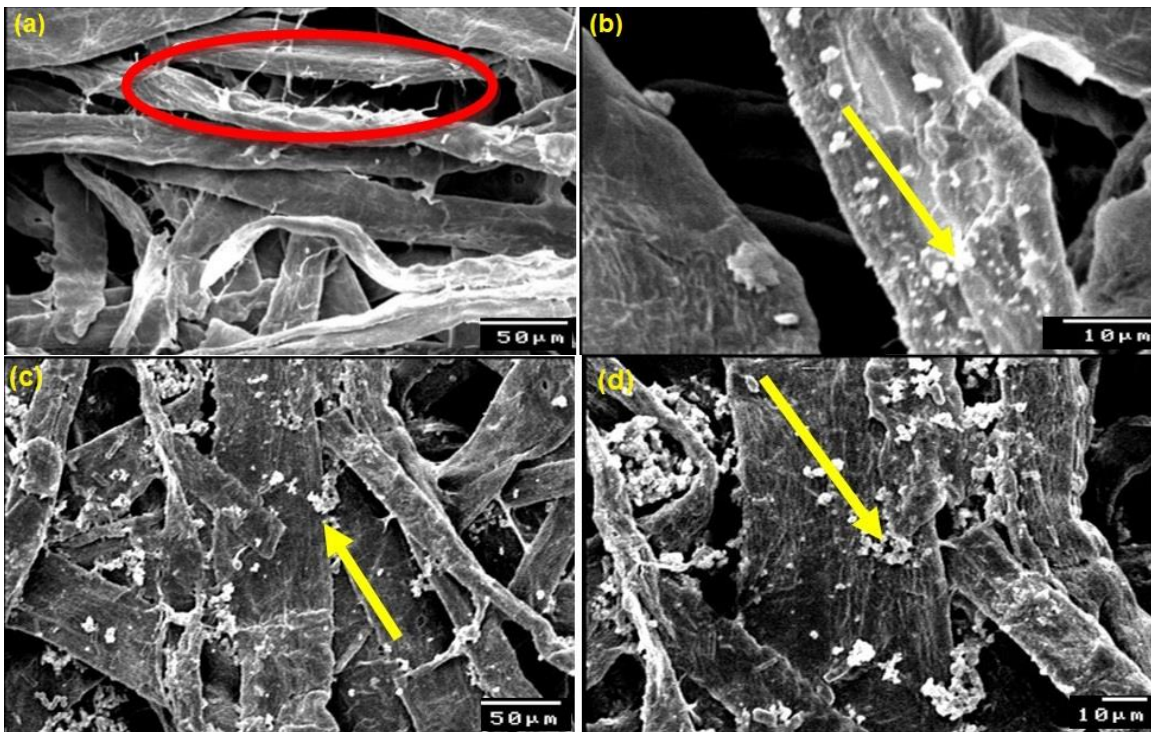


Fig. 9. The size of fiber fibrils increased after treatment by rice starch, which resulted in more linking between fibers (a); and the small frankincense crystals in sample $F_{1.75}S_{0.25}$ that appeared after the ageing process (b, c, and d).

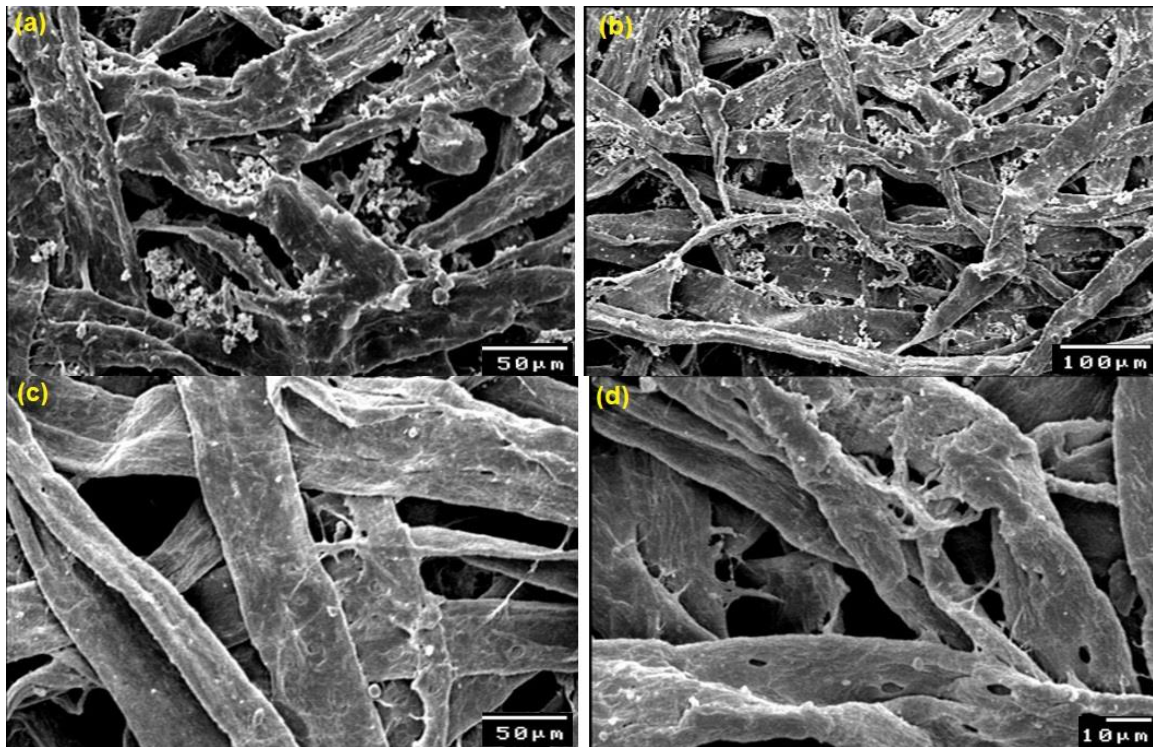


Fig. 10. Frankincense crystals in sample F_{1.5}S_{0.5} that appeared before the ageing process (a and b) but disappeared after the ageing process (c and d)

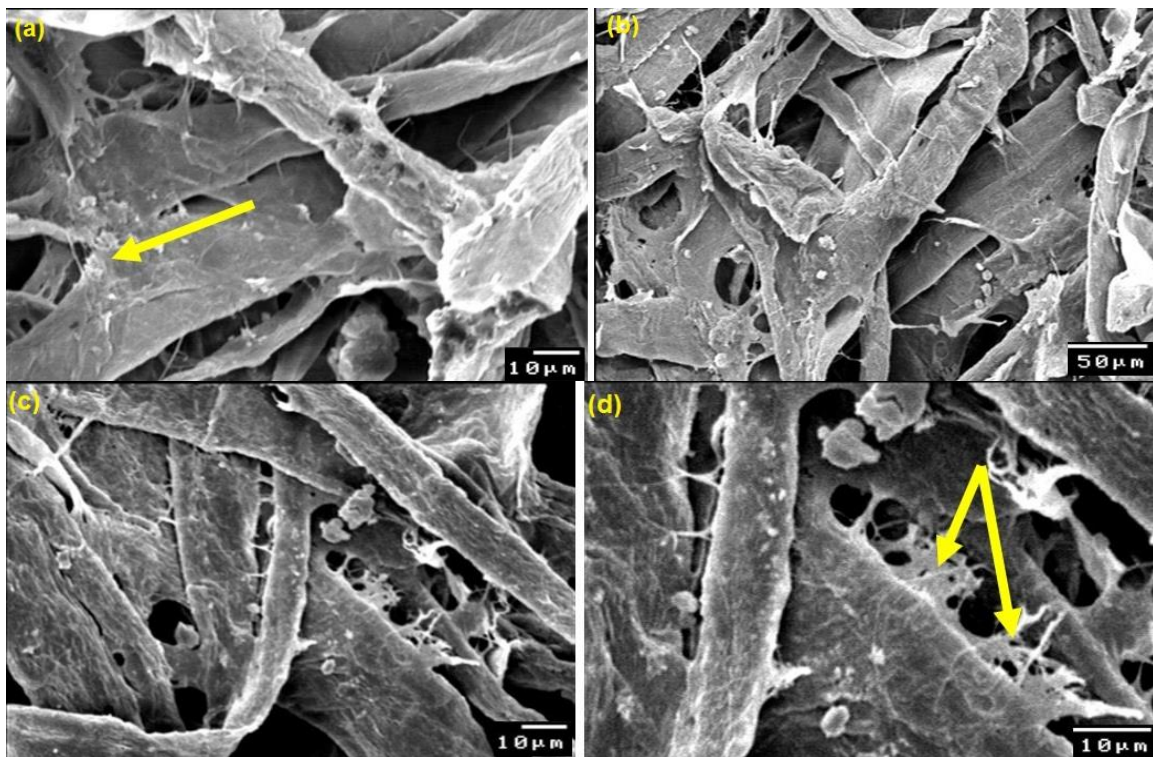


Fig. 11. Bridges between the wood fibers that appeared in sample F_{0.5}S_{1.5} after the artificial ageing process

pH Measurement

Under accelerated ageing of cellulose, the main chemical pathways for cellulose degradation are hydrolysis and oxidation. In the hydrolysis process, the water in cellulose cells spontaneously ionizes into H_3O^+ and hydroxyl groups (OH^-) (Hassan 2016; Speight 2018; Noshay *et al.* 2021). In the presence of dust particles (basic salt), free radicals in salt such sodium or copper react very little with the hydroxide ions (OH^-), whereas hydronium ions combine with the acetate ions to produce acetic acid (CH_3COOH), which reduces the pH of cellulose fibers. It should be noted that when preparing the samples for pH measurement, a white layer deposited around the ink surface was observed (Fig. 12) in sample $\text{F}_{0.5}\text{S}_{1.5}$ (which had a high concentration of frankincense). This showed that when the treated samples with a high concentration of frankincense were exposed to water, it caused whiteness to appear around the ink, which is an advantage in the process of displaying opaque inks in a safe way. From the pH measurement results (Table 2), it was found that there were no considerable changes before and after treatment.



Fig. 12. A digital microscope image of the $\text{F}_{0.5}\text{S}_{1.5}$ sample after cold extraction shows a white layer deposition around the ink surface

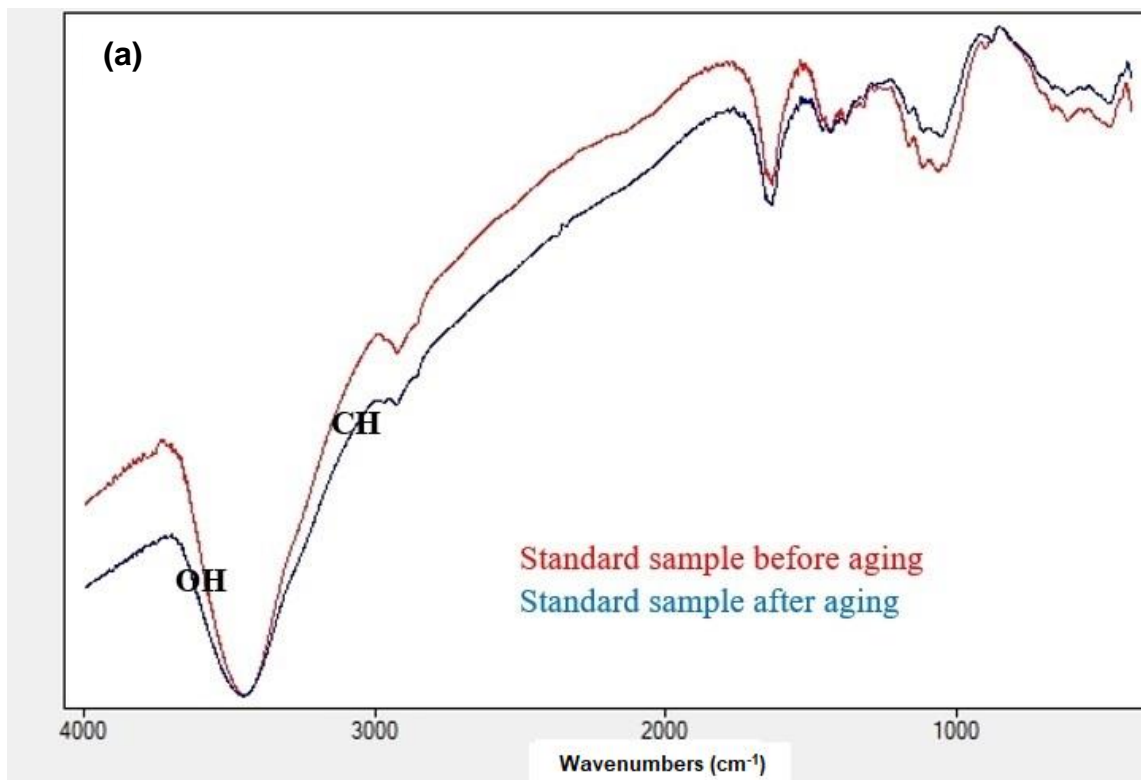
Table 2. Results of the pH Meter at Different Treatment Concentrations Before and After the Ageing Process

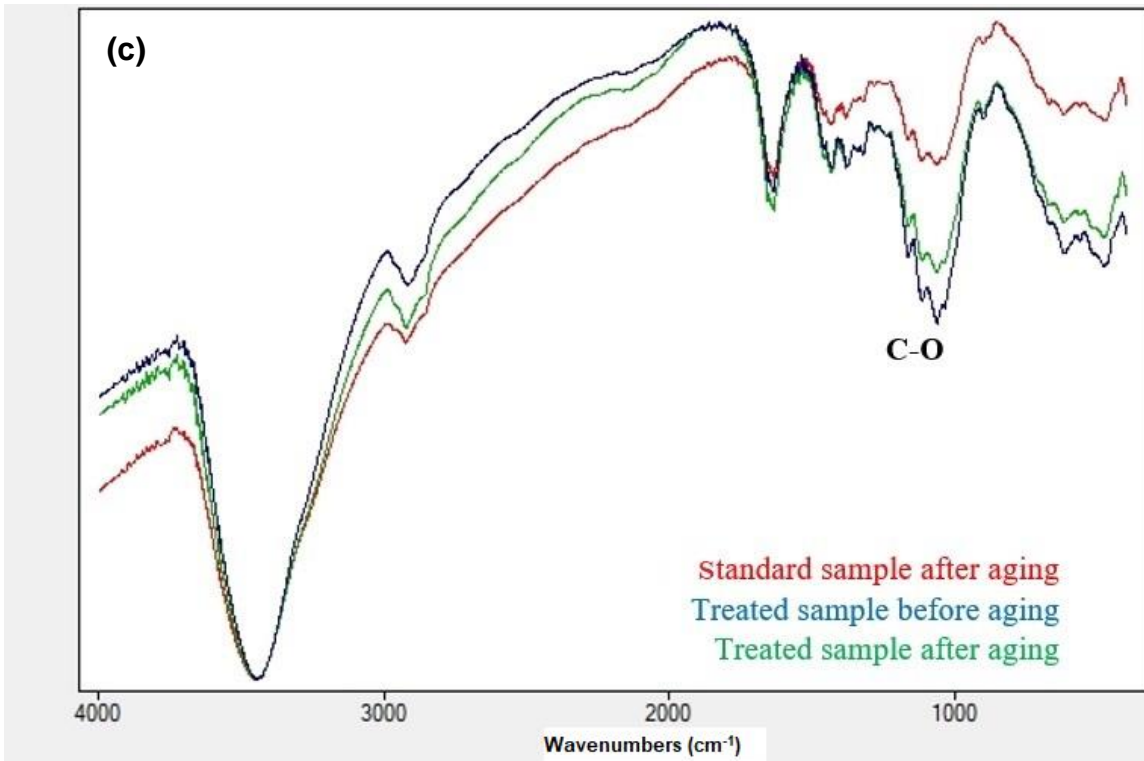
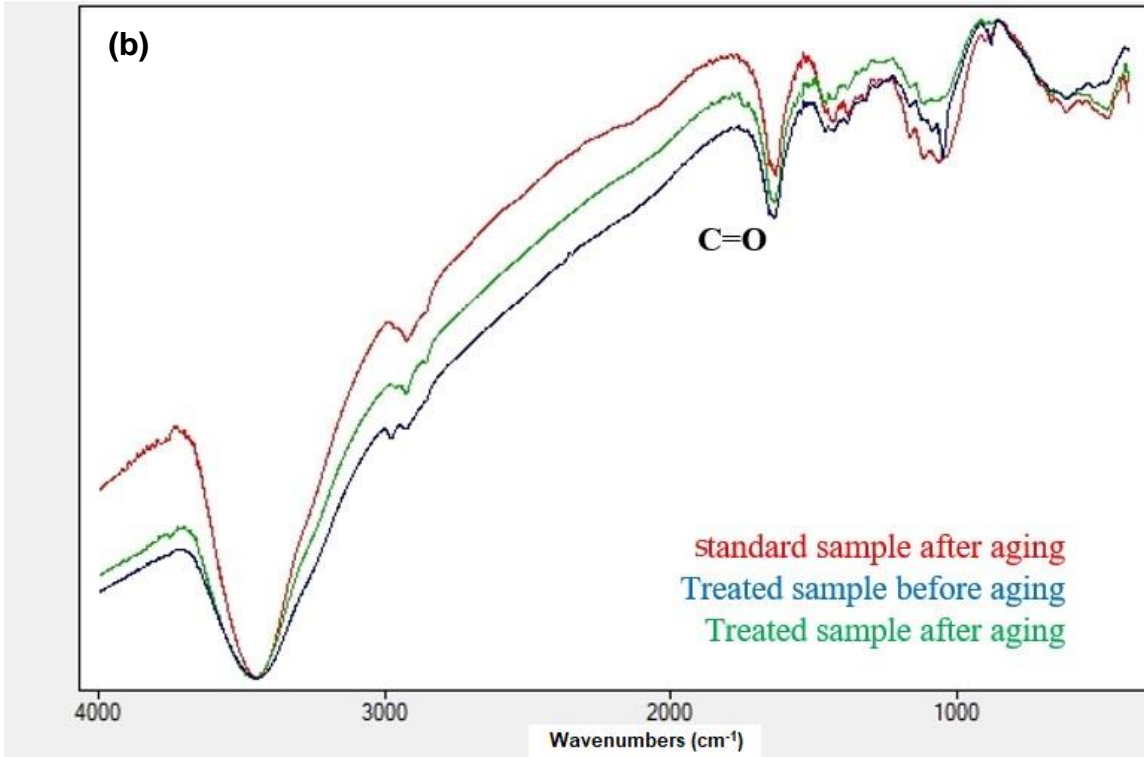
Sample	pH After Cold Extraction	
	Before Ageing	After Ageing
Standard	6.5	6.2
$\text{F}_{1.5}\text{S}_{0.5}$	6.4	6.2
F_2S_1	6.6	6.3
$\text{F}_{0.5}\text{S}_{1.5}$	6.4	6.4
$\text{F}_{1.75}\text{S}_{0.25}$	6.4	6.3

Fourier Transform Infrared (FTIR) Analysis

Figure 13a shows the FTIR spectra of the standard samples before and after the artificial ageing process. It was found that there was not an observable change in the OH region of all the samples. Furthermore, after the ageing process, new bands appeared at 900.6, 910.3, and 952.5 cm^{-1} (in comparison to the unaged sample). This may be due to the formation of an ester (C-O) group as well as C-OH stretching resulting from the decomposition of the pyranose ring. Furthermore, multiple carbonyl groups were formed as a result of the oxidation of cellulose molecules found in the C-OH groups, where the oxidation process of cellulose can easily occur (Hassan 2021).

From Table 3, a dramatic increase in OH stretching in all the treated sample was observed, especially in sample F₂S₁, which supports the role of frankincense in terms of increasing the hydration of the treated sample. In addition, a shift in the C=O bond (from 1634 to 1639 cm^{-1}) was detected, which explains the role of the frankincense molecules in terms of changing the structure of the carbonyl groups, thus reducing degradation processes even when subjected to accelerated ageing (Table 3). Furthermore, a notable increase in CH₂ stretching was observed in the treated samples, even after undergoing the ageing process; the intensity of these peaks remained higher than the control peak (Fig. 12b through 12d). The stretching of the C-H band, which is evidence of complete water desorption, can increase and improve the crystalline of paper (Hassan 2015; Mansour *et al.* 2021a). In addition, it was observed that the absorption bands located at 1428, 1367, 1334, 1027, and 896 cm^{-1} belong to the stretching and bending vibrations of the -CH₂, -CH, -OH, and C-O bonds in cellulose and were increased, especially in the treated sample F_{1.5}S_{0.5} (Fig. 12b).





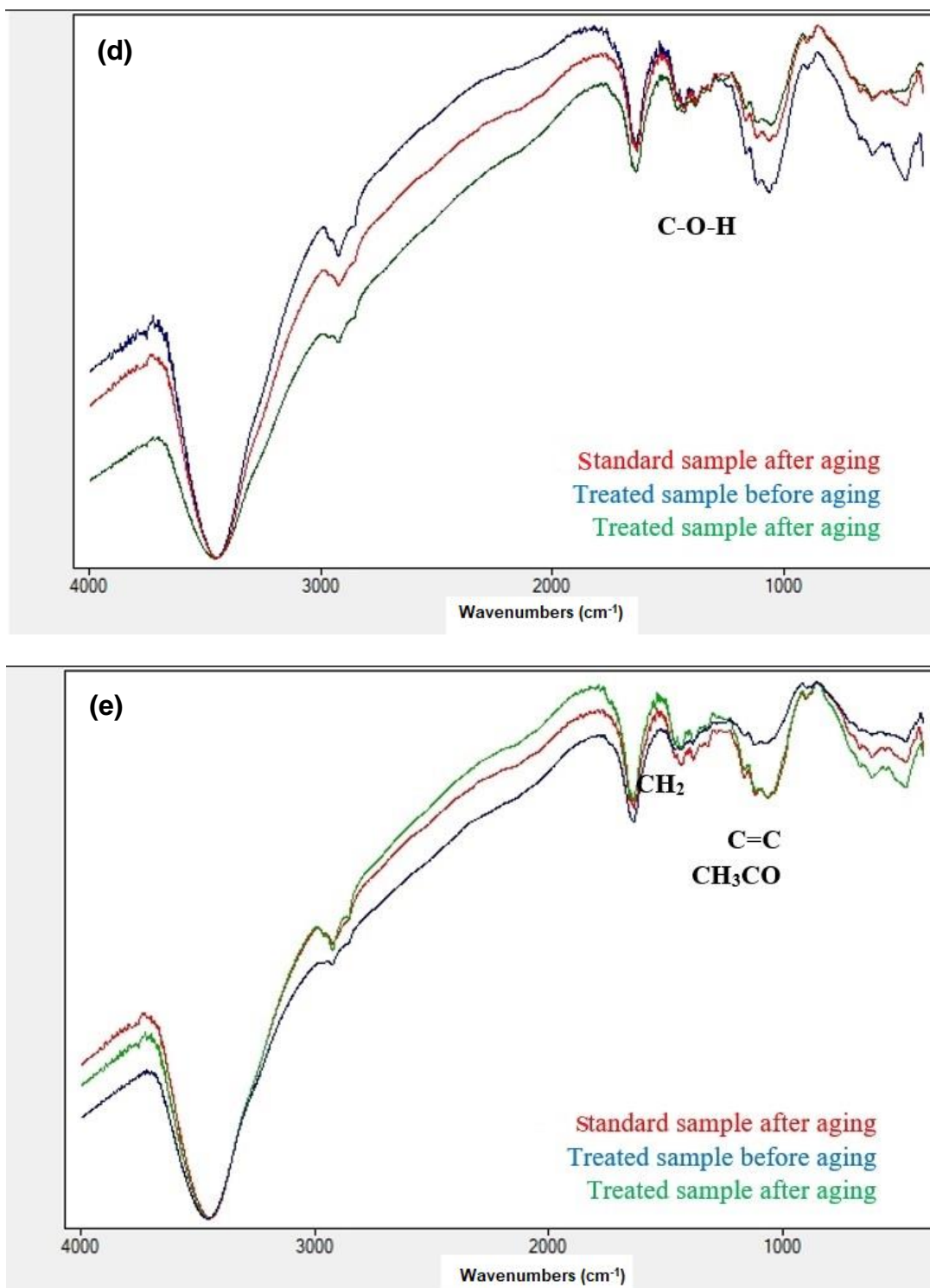


Fig. 13. (a) FTIR spectra of the standard samples before and after the artificial ageing processes; (b) a comparison between the standard sample and the treated sample $F_{1.5}S_{0.5}$, where an obvious improvement can be seen in the treated sample by the increase in the intensity of the bending vibrations of $-CH_2$ and $-CH$, $-OH$, and $C-O$ bonds and decrease of $C=O$; (c) a comparison between the standard sample and the treated sample $F_{0.5}S_{1.5}$, which shows an decrease in the OH group; (d) a comparison between the standard sample and the treated sample $F_{1.75}S_{0.25}$, which shows an increase in the CH_2 peak; and (e) a comparison between the standard sample and treated sample F_2S_1 , which shows an increase in the peak of the $C=O$ group.

It should be noted that there was a slight shift in OH stretching in treated sample F_{0.5}S_{1.5} (Fig. 13c), where the OH stretching shifted from 3452 to 3443 cm⁻¹. This shift could be due to the effect of the temperature on the paper and starch, which caused a loss in water content of the samples.

From Fig. 13d, which shows the peaks diagram of the F_{1.75}S_{0.25} sample, it was concluded that with an increase in concentration of frankincense in the mixture, the intensities of the topical variations of cellulose also dramatically increased. In addition, the variations responsible for the crystalline regions in cellulose (the bands at approximately 1420, 1330, and 1430 cm⁻¹) especially increased. This was due to the oils in the chemical composition of frankincense; for treated sample F₂S₁ there was an increase in the bands at approximately 1420 and 1430 cm⁻¹ associated with the amount of the crystalline structure of the cellulose, as shown in Fig. 13e (Al-Harrasi *et al.* 2018). From FTIR results, it was confirmed that the F₂S₁ and F_{1.75}S_{0.25} samples were the best concentrations for resizing paper.

Table 3. Changes in Functional Groups of the Treated Samples

Sample	Functional Groups (cm ⁻¹)						
	OH *	C-H *	C=O ester carbonyl	CH ₂ **	C=C and CH ₃ CO (frankincense)	C-O-H **	C-O stretching band
Standard	3452	2921	1634	1322	1427	1163	1056
F _{1.5} S _{0.5} B	3451	2930	1635	1327	1433	1163	1049
F _{1.5} S _{0.5} A	3449	2927	1638	1330	1440	1160	1052
F ₂ S ₁ B	3452	2927	1636	1324	1439	1154	1066
F ₂ S ₁ A	3455	2930	1639	1327	1442	1148	1069
F _{1.75} S _{0.25} B	3451	2930	1636	1327	1433	1163	1062
F _{1.75} S _{0.25} A	3452	2933	1639	1330	1436	1160	1057
F _{0.5} S _{1.5} B	3449	2921	1633	1322	1430	1160	1060
F _{0.5} S _{1.5} A	3443	2924	1636	1324	1433	1163	1063

Note: B = before ageing; A = after ageing; * denotes stretching; and ** denotes banding

Color Change in the CIELAB System

Colorimetric testing was used to investigate the color changes induced by the treatment of the paper samples, both by the resizing products and the artificial accelerated ageing tests. Based on the color parameter measurements, the ΔE values were calculated. The ΔE scale in organic material conservation is shown in Table 4 (Darwish 2013).

Table 4. Relationship between ΔE and the Qualitative Degree of Color Change

Very small difference	$\Delta E < 0.5$
Small difference	$\Delta E < 2$
Fairly perceptible difference	$\Delta E < 3$
Perceptible difference	$\Delta E < 6$
Strong difference	$\Delta E < 12$
Different colors	$\Delta E > 12$

It was also previously mentioned that starch and frankincense were used in resizing the paper samples at different concentrations, and the paper was subjected to artificial ageing using moisture and heat. Therefore, it was clear from the results of the color change values when comparing the standard sample before and after artificial ageing that the color

change value was 0.7. The ageing process caused a small difference in color change in the paper samples that were manufactured from wood pulp (Table 5). In addition, when comparing the samples treated with frankincense and starch before artificial ageing with the standard sample, it was observed that the lowest samples in terms of color change values was the treated sample $F_{1.75}S_{0.25}$, with a ΔE value of 0.8.

The ΔE value for the treated sample F_2S_1 before undergoing the ageing process was 0.9, while the treated samples $F_{0.5}S_{1.5}$ and $F_{1.5}S_{0.5}$ had ΔE values before undergoing the ageing process of 1.5. After exposing the treated samples to the ageing process, it was found that the ΔE value increased in all samples, while the sample that had the least color change was treated sample $F_{1.75}S_{0.25}$, where an increase of 0.9 occurred. The value of the color change was 1.7, and the sample that had the greatest increase in color change was the treated sample $F_{1.5}S_{0.5}$, which had an increase of 3.1° , resulting in a ΔE of 4. The color change that occurred in the samples can be attributed to the chemical changes in the samples as a result of the treatment with starch and frankincense. These chemical changes were identified through FTIR analysis, which showed the occurrence of oxidation and hydrolysis in the samples, involving a dramatic increase in the carbonyl groups (C=O). Therefore, it was evident through FTIR analysis that with an increase in the percentage of frankincense, the values of the color change also increased, and all the values of the color change in the samples after the ageing process were less than 4.5. This indicated that all the treatment concentrations were within the permissible limits, but it is preferable to use the lowest concentrations because no noticeable color change occurred (Table 5).

Table 5. ΔE Measurements for Treated Samples with Different Concentrations

Name of Samples	<i>L</i>	<i>a</i>	<i>B</i>	ΔE
Standard before ageing	89.4	0.4	5.7	-
Standard after ageing	88.9	0.3	6.1	0.7
$F_{1.5}S_{0.5}$ before ageing	88.2	0.3	6.5	1.5
$F_{1.5}S_{0.5}$ after ageing	84.3	0.4	6.6	4.6
F_2S_1 before ageing	88.6	0.3	6.9	0.9
F_2S_1 after ageing	88.4	0.3	7.7	2.3
$F_{1.75}S_{0.25}$ before ageing	88.7	0.4	6.8	0.8
$F_{1.75}S_{0.25}$ after ageing	88.1	0.4	6.8	1.7
$F_{0.5}S_{1.5}$ before ageing	88	0.4	6	1.5
$F_{0.5}S_{1.5}$ after ageing	85.1	0.9	4.9	4

Water Absorption

When comparing the results of the standard sample before and after the ageing process, it became clear that there was an increase in water absorption (WA) by 26% in the aged sample, reaching 621% after ageing compared to the 595% before ageing. This was because the thermal ageing process resulted in the breaking of the crystallized areas in the paper and thus these areas increased the WA of the paper samples, leading to dryness of the samples. This loss of water content resulted in the increases in WA when the sample was exposed to water again. Zervos (2010) indicated that during thermal ageing, in both humid and dry ovens, the degradation of pure cellulose paper is strictly hydrolytic. Due to this knowledge, when comparing the samples treated before ageing with the standard sample, it became clear that when the concentration of frankincense was 2%, 1.75%, 1%, and 0.5%, it was found that the frankincense in the sample reduced the WA percentage

because it is a resin material that is insoluble in water and the chemical composition of frankincense contains oils and acids. Thus, the frankincense covers the fibers and prevents them from absorbing water (Al-Harrasi *et al.* 2019; Johnson *et al.* 2019).

In the treated sample F₂S₁, there was an increase in WA by 379.7%, where the WA was 370.3% in the treated sample. In addition, in the treated sample F_{1.75}S_{0.25}, there was an increase in the WA by 379.7%, where the WA rate reached 359.2% in the treated sample. There was also an increase in the WA in the treated sample F_{1.5}S_{0.5} after ageing by 443.2%, where the WA percentage was 372.2%.

The treated sample F_{0.5}S_{1.5} had a decrease in water absorption of 186%, reaching a percentage of absorption of 409.8% before ageing, and reached 617.2% in the aged sample. When comparing the sample that had the highest percentage of frankincense with the sample that had the highest percentage of starch, it was found that the higher the percentage of starch, the greater the WA, due to the occurrence of starch hydrolysis. From these comparisons, it was concluded that the optimal samples were F₂S₁ and F_{1.75}S_{0.25}, due to the decrease in WA in these samples because of the increased percentage of frankincense compared to the samples with a high percentage of starch.

Table 6. Degree of Change in Water Absorption for All Treated Samples before and After the Ageing Process

Treatment	Ageing	Water Absorption (%)
Standard	B	595.32±50.26 a*
	A	621.82±67.60 a
F _{1.5} S _{0.5}	B	372.22± 63.09 b
	A	443.22± 24.11 b
F ₂ S ₁	B	370.28 ± 34.72 b
	A	379.72± 58.21 b
F _{1.75} S _{0.25}	B	359.17± 85.70 b
	A	406.25± 78.31 b
F _{0.5} S _{1.5}	B	409.83± 66.55 b
	A	617.22± 76.76 a
LSD	-	107.79

Note: B = before ageing; A = after ageing; * denotes the same letter (A-B) are not significantly different according to LSD at a 0.05 level of probability

CONCLUSIONS

1. The two substances, frankincense and rice starch, were able to achieve the objective of this study, which was to reduce the percentage of water absorption and make the paper less susceptible to damage. However, this reduction occurred in varying proportions according to their respective concentrations. The best concentrations were found in samples F_{1.75}S_{0.25} and F₂S₁.
2. The results of the pH measurement demonstrated that there were no noticeable changes in the pH values, but the treatments had an effective role in raising the pH values after the accelerated ageing process compared to the untreated samples. Therefore, these treatments are considered safe for both ancient and modern papers.

3. It should be noted that in the case of a high concentration of frankincense, the best results were obtained. However, there was a qualitative advantage in the case of sample F_{0.5}S_{1.5}, where the high proportion of starch led to form white a layer around the letters of the writing during exposure to water, which may be a promising method for revealing inks.
4. The results of the digital microscopy analysis showed that there was fungal growth on one of the treated paper samples after being exposed to an environment suitable for microbiological growth. This confirmed the inefficiency of frankincense in protecting the paper from microbiological infection.
5. The water absorption examination results showed a decrease in the water absorption percentage of the treated paper because frankincense coated the fibers. Therefore, in conjunction with the results of the previous experiments, it was concluded that the best concentrations that were applied were in samples F_{1.75}S_{0.25} and F₂S₁.
6. It can be concluded from the FTIR analysis that with an increase in concentration of frankincense, the intensities of topical variations of cellulose also dramatically increased. This was especially true for the variations responsible for the crystalline regions in cellulose, *i.e.*, the bands at approximately 1420, 1330, and 1430 cm⁻¹, which were due to the oils in the chemical composition of frankincense.
7. The results of the color change analysis showed that artificial ageing caused a slight color change in the treated samples. After comparing the standard samples with the samples treated with frankincense and starch, it was found that the lowest color change value occurred in treated sample F_{1.75}S_{0.25}, which had a ΔE value of 0.8. In addition, the most frequently reported samples with an increase were the F_{1.5}S_{0.5} samples, where an increase of 3.1° occurred; the evidence for this was the occurrence of oxidation, which was identified through FTIR analysis that showed an increase in the carbonyl and hydroxyl groups, which indicated chemical changes. Therefore, it was concluded that increasing the percentage of frankincense increased the rate of color change due to the oils and acids included in its composition, but those changes are within the permissible range.

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