AN EXAMINATION OF THE CHARACTERISTIS OF CELLULOSIC HANDSHEETS TREATED WITH GLUTARALDEHYDE

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The COVID-19 pandemic has influenced the demand for products that are considered hygienic, thereby increasing the production rate and variety of hygienic products. Researching new antimicrobial materials is gaining importance with increasing awareness of the topic of infectious diseases caused by various microorganisms. In the present work, cellulosic handsheets were produced and then coated with coatings having different glutaraldehyde concentrations by a roller bar technique. The surface water absorption capacity of the sample groups and their structural and strength characteristics were analyzed. Also, the cross-linking effect of glutaraldehyde was determined by FTIR analysis. The results not only showed that, after being exposed to glutaraldehyde on their surface, the handsheets presented a higher hydrophilic structure and higher tensile strength properties, but also confirmed that coatings containing 1–5% glutaraldehyde lessened fungal activity on their surfaces.

Keywords: coating, cellulosic handsheet, glutaraldehyde, antifungal

INTRODUCTION

Paper is a structurally layered polymeric material. This material essentially consists of individual cellulose fibres bonded together by van der Waals forces and hydrogen bonds. Cellulose is the most abundant polysaccharide in nature, a non-toxic, renewable, biodegradable, biocompatible biopolymer.[1,2] As a functional material, paper is composed of cellulose fibres, filler, binders and other components, depending on its end uses *etc*.[3] Paper has been used for many different purposes from the past to the present and has been differentiated and redeveloped, improved and transformed with the development of technology. Paper-based products are versatile materials that are widely used in the fields of communication, education, packaging,[4] 3D printing processes,[5] healthcare and cleaning.[6,7] Paper material can be produced under different production conditions and with different content of specific components, depending on the final usage areas and the features it is expected to present.

Like most polysaccharides, cellulosic-based paper products have structural pores and hydrophilic surfaces that can provide reasonably fair conditions for microbial growth in terms of nutrition, temperature and humidity.[8-13] For paper-based materials, this property is not particularly desirable. Especially because of the effects of Covid-19, the demand for paper-based materials increased rapidly in a short period of time.[14] The most prominent among these products are hygiene paper, food packaging and medical speciality papers.[15] It is believed that the invention of innovative paper products that dry easily, are clean and resist the growth of bacteria will bring great benefits to the paper industry and society around the world.[16]

Coatings are frequently used in industrial settings due to their beneficial qualities and usability. By depositing material in either the liquid phase (solution) or the solid phase (powder or nanoparticles), a thin layer of material can be incorporated into a paper substrate.[17] The coating techniques can be customized and applied in a way that satisfies the needs of end products. The mechanical, surface and barrier properties of paper-based products can be improved by coating formulations containing antiseptic chemicals. Due to the increasing need to control the spread of possible infections, this preliminary work was able to test a functional and innovative antiseptic paper with a coating colour formulation containing glutaraldehyde.

Glutaraldehyde is a colourless oily liquid, with a strong aroma, which is miscible with benzene, alcohol and water under room conditions.[18] There are numerous applications for glutaraldehyde. The usage areas include paper mill process water systems, water floods, metalworking fluids, water-based conveyor lubricants, air washers, and industrial scrubbers. Specifically, these areas include oil storage tanks, drilling muds, drilling, completion and workover fluids, packer fluids, gas production and transmission pipe systems, gas storage wells and systems.[18] Also, in human medicine, glutaraldehyde is mostly employed to disinfect inanimate surfaces (in varying amounts, according to the formulation).[18] Likewise, in hospitals, devices are sterilized using antiseptic solutions containing glutaraldehyde (GA), which possesses viral inactivation and sterilization properties.[19-21]

This study will function as a pilot project. We have chosen to use glutaraldehyde as an antibacterial chemical and carboxymethylcellulose as a coating colour binder. The main research has focused on examining the impact of newly developed coating formulations on handsheets composed of pure cellulose produced in the laboratory. The reason of avoiding the utilization of chemicals in the manufacture of the handmade papers used as the base paper in this study was to investigate the changes induced by the glutaraldehyde surface treatment in the structural and chemical properties of papers created directly from pure cellulose. As generally known, fillers, retention aids, optic bleaching agents, sizing chemicals, and other chemical compounds are normally utilized in the structure of mass-produced paper sheets. However, measuring the effects of all independent characteristics is challenging because they can all affect the outcome. Thus, the purpose of this work has been to investigate in depth the interaction of different amounts of the glutaraldehyde chemical with pure cellulose.

EXPERIMENTAL

Pulping and making of handsheets

One of the main goals of this study was to possibly control the variables that can cause variations in paper stock and to deal with paper components and manufacturing parameters. To that end, experimental handsheets were produced as basis papers under specified normal conditions.

All handsheets were manufactured from bleached pine kraft pulp. The pulp-beating process was run in a Valley beater for 60 minutes in accordance with ISO 52641: 1979 22. Then, the fractionation process was performed in accordance with Tappi T275 23, to control sufficiently long fibre length and eliminate fine fibres from the pulp feed. The drainage resistance of the beaten pulp suspension was characterised by a Schopper–Riegler device according to ISO 5267–1:1999 24. Also, the swelling ability of the pulp suspension was evaluated by using Equation 1:

$$WRV = \frac{(\mathrm{Mw} - \mathrm{Md})}{\mathrm{Md}} \qquad (1)$$

where WRV is the water retention value, MW is the wet mass (g) and Md is the oven-dry mass (g) of the pulp.

Handsheets were formed as base papers from fines-free beaten pulp, using a Standard Handsheet Former, by following the principles of Standard ISO 5269–1:2005 25. Their grammage was determined as 60 g/m². The handsheets were pressed by a type of hydraulic press, raising its pressure gradually up to 345 kPa. The pressed handsheets were settled in drying rings having metal surfaces and allowed for 48 h for the drying process.

Preparation of coating colour formulations

The mixture was prepared containing distilled water, commercial Alfasol Glutaraldehyde chemical (GA 50%) and carboxymethyl cellulose (CMC 2%) used as a binder. Table 1 lists the distinguishing characteristics of the chemical substance Alfasol Glutaraldehyde (GA 50%) and carboxymethyl cellulose (CMC 2%). The distilled water and CMC (2%) solution was heated and continuously mixed at 50 °C until completely dissolved. By adding glutaraldehyde chemical (in prescribed amounts) to the modified solutions containing 2% CMC, additionally, these prepared uniform coating colour formulations were sonicated for an adequate time. These solutions (1, 2, 3, 4 and 5), prepared separately with different contents of glutaraldehyde and cooled at room temperature, were applied to the paper surface when their temperature reached 23 °C. The composition of each coating formulation is given in Table 2. Equal solution amounts of 100 mL were taken for calculations, with 2% CMC used as binder in all formulations. Viscosity values and pH measurements of the solutions prepared at 23 °C were determined with a Brookfield type viscometer and a pH measurement kit.

Application of coating colour formulations

The prepared coating colours were applied to the paper surfaces with the help of a cylindrical glass rod. During the application, the same amount of the formulation was distributed on both surfaces of each sample with a glass rod. In addition, care was taken to pull the rod by applying constant speed and pressure for homogeneous distribution of the solution on the surface.

Table 1
Characteristics of Alfasol Glutaraldehyde chemical (GA 50%) and carboxymethyl cellulose (CMC)

Glutaraldehyde (GA 50%)	
Molecular formula	$C_5H_8O_2$
Molecular weight	100.117 g/mol
Exterior	Colourless, strong odor, liquid
Density	1.06 g /cm^3
Melting point	-14 ºC, 7 ºF, 259 ºK
Boiling point	187 ºC, 369 ºF, 460 ºK
Solubility	soluble in water, mix and react
Carboxymethyl cellulose (CMC)	
Molecular formula	$C_6H_7O_2(OH)_2OCH_2COO_2$
Molecular weight	6400 ± 1000 g/mol
Exterior	Slightly yellow color, odorless, hygroscopic, powder
Density	1050 g/cm^3
Solubility	soluble in water

Table 2
Coating colour formulations

Coating formulation	Glutaraldehyde (%)	Binder (CMC) (%)
1	0	2
2	1	2
3	5	2
4	10	2
5	20	2

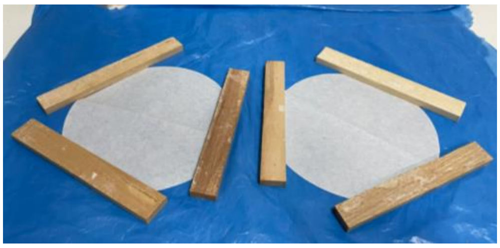


Figure 1: Drying of surface-treated paper specimen

Table 3
Codes given to paper specimens

Specimen code	Coated formulation
C	-
C*	1
G1*	2
G5*	3
G10*	4
G20*	5

After the surface coating process, the treated paper samples were left to dry naturally for 24 hours, under normal room conditions. After coating and drying one side of the paper sample, the other side was also coated, so that both sides were surface treated with glutaraldehyde. In order to prevent curling, which may occur during drying, wooden weights were used on the edges of the samples as shown in Figure 1.

The coated papers were denoted as a function of the coating colour formulation applied, as shown in Table 3.

Preparation of coated papers for testing

Before testing, each sample was conditioned according to ISO 187:2022[26] standard for 24 hours at 50% ±2 relative humidity and 23 ±1 °C, and prepared for physical and mechanical tests. Five paper samples were stacked for each sample group and the thickness of the papers was measured on different randomly selected points, using an electronic micrometer, according to ISO 534:2011[27] standard. Results are defined in μm. The area of each paper sample was defined as 100 cm². The masses of these samples were determined with a balance sensitive to 0.001 g, according to the relevant standard ISO 536:2019[28] and their grammage was calculated in g m^{-2} units. The density values of the papers were calculated in accordance with ISO 534:2011[27] standard. These values are expressed in units of g cm^{-3}.

Wettability and water retention tests

The surface wettability and water retention qualities of the produced samples were evaluated using the contact angle test and the Cobb test.

An Attension Theta optical tensiometer, with an autonomous liquid pump system, was utilized to characterize specimen wettability. As the probe fluid, pure and degassed water was used. The contact angle of a drop of liquid with a solid surface on the bottom was measured simply by finding the tangent (angle). Changes in the contact angle of the droplets were measured over time.

The Tappi T441[29] standard method was used to measure changes in the water retention capacity of the paper, before and after surface treatment with glutaraldehyde. To do this, a circular sample was cut to the standard specified dimensions and weighed using a precision balance. Since both sides of the paper were coated, a randomly selected paper side came into contact with water and was then clamped in the test mechanism. At a determined time in the experiment, 100 mL of distilled water was transferred into the Cobb cylinder simultaneously. The Cobb test assembly was flipped after contacting the paper surface with water for approximately 1 minute. Then, the excess water held in the coated paper was removed onto a special blotter with a casting roller that was gently moved over it. Paper samples were reweighed while still moist. The weight differences of the samples before (m1) and after (m2) testing were used to calculate Cobb's value:

$$COBB = 100 * (m2 - m1) \quad (2)$$

Tensile test

Tensile tests were carried out on a Hybrid Rheometer Discovery HR-2 Machine, which can be used as an alternative to the Universal Test Machine, where standard tensile tests are performed.[30] Test samples were prepared with the dimensions of 5 x 50 mm. The distance between the pulling jaws was determined as 40 mm and the samples were pulled at a speed of 0.6 mm/min. The stress-strain curve of each sample was recorded separately. The modulus of elasticity values were calculated from the slope of the steepest parts of each stress-strain curve of the samples. To represent the mechanical properties of the samples, the tensile strength, tensile index and modulus of elasticity values were calculated by considering the ISO 1924-2:2008[31] standard method.

FT-IR ATR spectral analyses

A Thermo Scientific Nicolet iS50 FT-IR Spectrometer, with an ATR unit (Universal ATR Diamond Zn/Se), was used to conduct the FT-IR investigations. The pressure that is applied to a specimen affects the quality of the spectra.[32] As such, pressure was controlled using a single reflection ATR setup that included a diamond crystal.[33] For the effective implementation of this technique, the ATR unit was in direct contact with all the samples. The test area of the specimens of 40 x 40 mm^2 was used to record each FT-IR spectrum. The wavenumber range for the FT-IR spectra was 650-4000 cm^{-1}. Utilizing the OriginLab software (8.1), the baseline adjustment was made for the cross-sectional area of specimens to lessen the impacts of light scattering.

Bacterial and fungal tests

Control and coated handsheets were tested for bacterial and fungal activity against microbial samples present on the surface of unwashed human hands and unwashed laboratory workbenches. Bacterial and fungal growth was monitored on the paper surfaces using the Hytech Slide® hygiene test kit. C, G1*, G5*, G10*, G20* coded samples were contacted with the growth medium in the test kits for 5-6 seconds, then the samples were kept in an oven at 35 °C for 48 hours for observing bacterial growth, or in a conditioned test room at 25 °C for 120 hours for observing the development of molds and fungi.

RESULTS AND DISCUSSION

In the present work, the variables employed in papermaking are reduced to a minimum. This method has ensured the possibility to keep all other handsheet-making factors the same. This makes it easier to see how simply varying the glutaraldehyde content in the handsheets has changed the impact of the surface coating.

Characteristics of pulp

The paper material has a heterogeneous nature, due to its structure and the manufacturing method used. The morphological characteristics of the fibres and the fundamental characteristics of the pulp suspension they create are crucial as they will impact the mechanical, chemical, and structural characteristics of the paper that will be produced.[34] All the changes that can result from the raw materials used and the variation in the process parameters have been reduced, to make a scientific comparison in the conclusion of the study. All the measured details of these pulp and fibre characteristics are reported in Tables 4 and 5.

Characteristics of coating colour formulations

The thickness of the coating paint that is applied to the paper surface depends on the coating solution's viscosity. According to the literature, coating solutions with viscosities of 1-6 cp are acceptable.[35,36] As can be seen from the results in Table 6, the thickness and grammage values of the G10* and G20* specimens increased because solutions with higher viscosity and high glutaraldehyde content (formulations 4 and 5) attached better to the surface of the handsheets.

The pH and viscosity values of coating colour formulations containing glutaraldehyde in different concentrations are summarized in Table 6. The pH values of the produced solutions range from 5 to 8, as shown in Table 6. Since the pH values decreased as glutaraldehyde concentrations increased, it was concluded that the solutions turned from neutral to acidic.

Table 4
Schoper-Riegler (°SR) and water retention value (WRV) of pulps

Beating time (min)	°SR		WRV	
	WP	FF	WP	FF
0	14	11	1.80	1.60
60	17	13	2.50	2.35

WP: Whole pulp, FF: Fines-free pulp

Table 5
Fibre parameters of beaten and fractionated fines-free pulp

Beating time (min)	Length λ (mm)	Width ω (μm)	Coarseness (mg/m)
60	2.1	20.08	0.143

Table 6
pH and viscosity values depending on contents of glutaraldehyde in coating formulations

Coating formulation	Glutaraldehyde (%)	pH	Viscosity (cp)
1	0	8	3.5
2	1	8	3.8
3	5	7	4.2
4	10	6	4.7
5	20	5	6.3

Characteristics of handsheets

The structural characteristics of handsheets were analysed and the results are given in Table 7. The standard deviations of the test measurements were used to calculate the error bars in the following figures. The density and grammage values of the handsheet specimens, having thickness varying between 110 μm and 123 μm, were given in Figure 2. The density of the handsheets was determined in accordance with ISO 534:2011[27] standard. The density results are given as g cm^{-3}.

The use of CMC as a binder has not appeared to cause any increase in the density of the handsheets. On the contrary, a slight decrease was observed in the density value, since the thickness of the handsheet was increased after the surface treatment with coating formulation 1 and the grammage of the handsheet was not influenced. As can be observed in Figure 2, the increase in the glutaraldehyde concentration enhanced the grammage and density of the coated specimens. The density and the grammage of the control handsheets increased by 11% and 21%, respectively, with the application of the coating formulation with 20% glutaraldehyde. Naturally, this feature also affects the mechanical characteristics of the coated handsheets.

Wettability and water retention test results

The contact angle measurements demonstrated the effect of glutaraldehyde on the hydrophobicity or hydrophilicity of the handsheet samples. Each specimen's mean contact angle was assessed as a function of time. The contact angle images in Figure 3 show the mean values of contact angles within the first second of the test duration. The hydrophilic character of the paper material was confirmed by the sample G5*, which also had the smallest contact angle values in this test and quickly absorbed the water by trapping it from the surface inside. The uncoated control (C) sample had a very high contact angle value, but this value was lowered when glutaraldehyde coatings were applied as surface treatment. The glutaraldehyde coating method clearly rendered the specimens more hydrophilic, as shown by the data. The Cobb results discussed below supported the same conclusion.

The Cobb test was used to evaluate the absorption quality of the specimens. We investigated how the glutaraldehyde content affected the water absorption capabilities of the handsheets. For a predetermined time, a column of water was put on the paper, and the amount of water absorbed was measured; the results are given in Figure 4.

Table 7
Structural characteristics as a function of coating formulation

Specimen code	Grammage (g/m²)	Density (g/cm³)	Thickness (μm)
C	60.06	0.53	110
C*	60.15	0.54	113
G1*	61.50	0.53	113
G5*	62.12	0.55	113
G10*	68.33	0.58	118
G20*	73.09	0.59	123

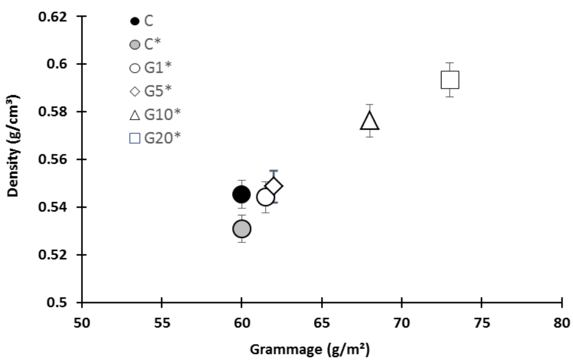


Figure 2: Grammage and density changes as a function of surface treatments with glutaraldehyde

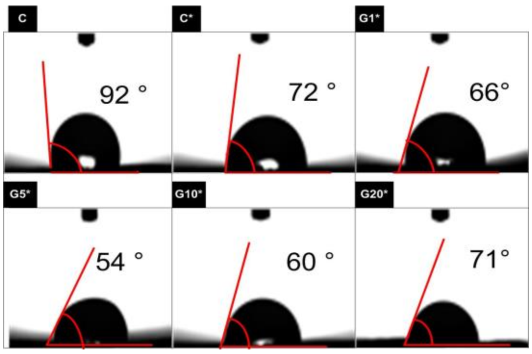


Figure 3: Variation in contact angle values and photos of water droplets

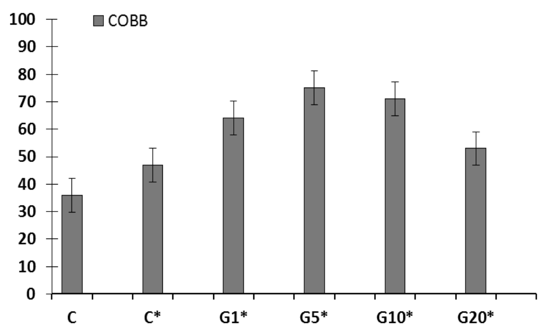


Figure 4: Changes in Cobb values for different handsheets groups

The application of coating formulations to the surface of the handsheets led to an increase in their ability to absorb water. Specimen G5* has the highest Cobb score, calculated to be 75. In comparison with the initial values belonging to the control samples, the Cobb values of the papers treated with 5 and 10% glutaraldehyde increased about twice. The Cobb values of the surface-treated handsheets with glutaraldehyde concentrations between 1 and 5% present an increasing trend, as compared to the other groups of handsheets. However, this trend is not linear, since the Cobb value of the handsheets to which 10–20% glutaraldehyde surface treatment was applied did not support it. Nonetheless, the Cobb values of all surface-treated handsheets are higher than those of the control sample C. This reveals that the handsheets have a more hydrophilic structure after surface treatment with glutaraldehyde.

Tensile results

The impact of glutaraldehyde contents on the tensile properties of the handsheets was examined using coated handsheets as a base paper. To establish the effect of the binder chemical, a comparison of the control sample with the CMC-coated sample alone was considered. Figure 5 displays the load and elongation curves of each specimen. Observing the sample groups, the data in Figure 6 show how the tensile index and the elastic modulus have changed. These numbers are given in Nmg^{-1} and MPa units, respectively.

Examining the tensile test results reveals that the tensile load, tensile index, and elasticity values of the handsheets increased after surface treatment (Figs. 5 and 6). Figure 5 demonstrates that the paper G5* have the lowest elongation percentage (about 5.5%) before the sample deforms, and a higher tensile index value than the other surface-treated handsheet groups. The handsheets coated with solutions containing 5% glutaraldehyde concentration (Fig. 6) have the highest tensile values. The tensile index value enhanced from 30 to 56 Nmg^{-1}, (by about 86%), after coating the initial paper with the glutaraldehyde-containing formulation with a concentration of 5%. The handsheets coated with solutions containing 20% glutaraldehyde concentration (Fig. 6) have the highest elastic modulus. The elastic modulus value of the control specimens enhanced from 5 to 37 MPa with the application of coating formulation 5. Certainly, the disparity in the grammage of the uncoated and coated handsheets is a contributing factor to the variation of the tensile test results.[37]

FT-IR results

The handsheet C, produced without the application of any coating, was considered as a control sample. Figure 7 depicts the IR bands that are characteristic of all handsheet groups. The control sample (C) and other samples coated with coating formulations containing glutaraldehyde (0, 1, 5, 10, 20%) and CMC are labelled as C*, G1*, G5*, G10* and G20*. The interaction between CMC and glutaraldehyde results in significant modifications in the functional groups of cellulose, as seen in the FTIR spectra of uncoated and coated handsheets with CMC and glutaraldehyde in Figure 7. Cellulose has been used as the primary raw material in all the handsheets, as stated above. To calculate the relative absorbance and make baseline corrections for all spectra, the distinctive cellulose peak, located at 1160 cm^{-1},[38] was chosen as a reference point.

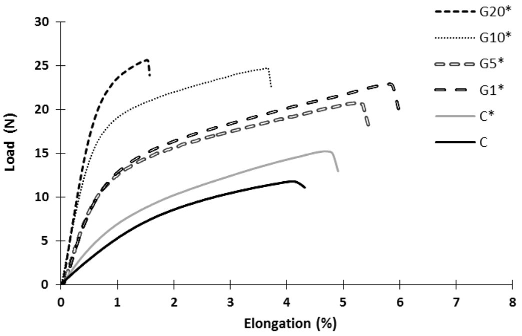


Figure 5: Load and elongation curves of the handsheet groups

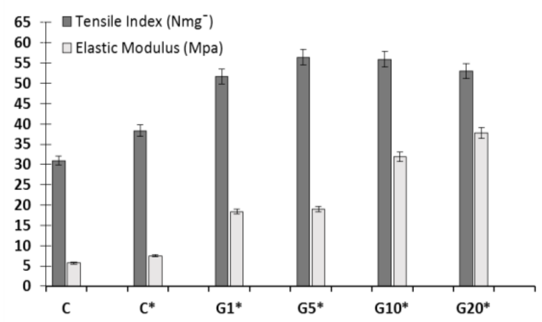


Figure 6: Tensile index and elastic modulus values of the handsheet groups

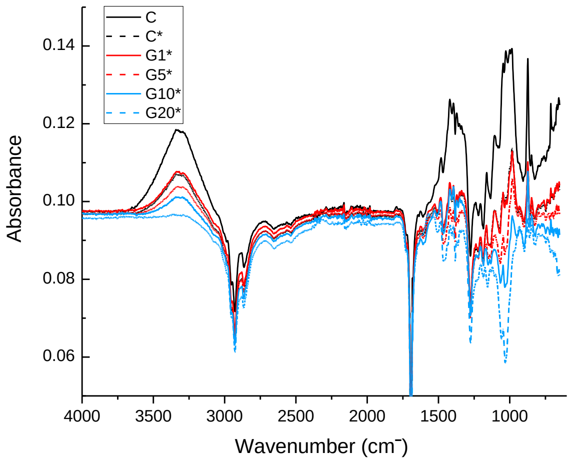


Figure 7: FTIR absorbance of handsheet groups in the range of 4000 and 650 cm^{-1}

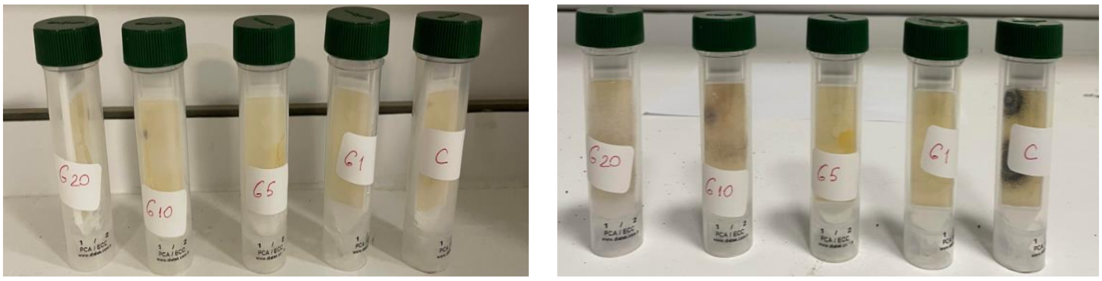
The presence of cellulose in the samples has been demonstrated by the characteristic peaks at 1160 cm^{-1} (C-O-C asymmetric valence vibration), 1030 cm^{-1} (C-OH stretching vibrations of secondary and primary alcoholic groups), and at approximately 1100 cm^{-1} (glycosidic C-O-C stretching vibration).[38-40] Also, the peak at 1055 cm^{-1} is apparent; it is connected to the hydroxyl (OH) and ether groups' C-O stretching modes in cellulose for all specimens.[41,42] However, the cellulose peaks showed significant differences in their absorbance values, especially in the G10* and G20* samples, when the glutaraldehyde percentage in the coating formulation was increased. By increasing the glutaraldehyde content, a decline in the related absorbance values is seen. The IR spectra reveal that the CH aliphatic group's absorption has been reduced to 2952 cm^{-1}. The addition of glutaraldehyde, on the other hand, causes a significant change in the absorption between 1250 and 1000 cm^{-1}, confirming the reaction of the glutaraldehyde with the CMC that creates the hemiacetal structure.[43] The vibrational spectra of the carbonyl groups (C=O) and C-O are located around at 1740 cm^{-1} and 1055 cm^{-1}, respectively. The crosslinking processes were seen to reduce the C=O band's intensity at 1740 cm^{-1}. The coating treatments may be responsible for the decrease in the C=O band's intensity by consuming additional free carbonyl groups. The wide band of OH^- groups around 3350-3400 cm^{-1} can be attributed to absorbed water, secondary alcohols (CMC), and hydrogen bonds between cellulose fibres.[44,45]

Bacterial and fungal test results

Hytech Slide® hygiene test kits were inspected on the second and fifth days after the initiation of the bacterial and fungal tests. The C* specimens were eliminated from the test kit inspection since only the impact of glutaraldehyde on bacterial and fungal development was examined.

Although, macroscopically, bacterial activity has not been observed very frequently in the Hytech Slide® hygiene test kits (Fig. 8a), fungal activity has (Fig. 8b). Because of this, no type of diagnosis for bacteria has been fulfilled; nevertheless, basic macroscopic diagnostic application has been carried out for fungi, which are quite clearly seen in the Hytech Slide® hygiene test kits. A detailed microscopic view of the growth of *Aspergillus niger* colonies on the Hytech Slide® hygiene test kits of C specimens is shown in Figure 9.

There are numerous species of *Aspergillus* that live indoors and outdoors. The aspergillum-like conidial-bearing structure, or conidial head, is known as this genus' primary microscopic defining feature.[46] After examining the microorganism's activity in the growth media, a similar conidial structure was found under the microscope (in Fig. 9). According to these data (in Figs. 8a and 8b), the use of glutaraldehyde coating colors as a sterilizing agent in concentrations between 1 and 5% can be beneficial, since it prevents fungal growth.



a) b)

Figure 8: Growth medium and microorganism activities; (a) bacterial activities, b) fungal activities for different handsheet groups

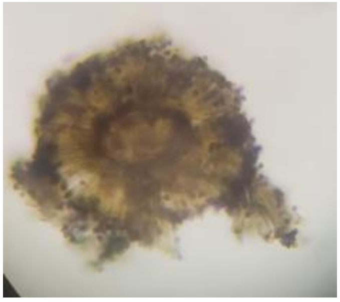


Figure 9: A detailed microscopic view of the growth of *Aspergillus niger* colonies

CONCLUSION

To assess the benefits and drawbacks of glutaraldehyde-containing coatings in practical application, coating formulations with various glutaraldehyde contents were applied on handsheets and the final products were characterised and tested under laboratory conditions.

In brief, paper made from cellulosic materials has structural pores and hydrophilic surfaces that can offer rather favourable conditions for microbial development, in terms of nutrition, temperature and humidity. The objective of this work was to avoid this. Since, in this investigation, the surface pores of the paper samples were filled with coatings containing glutaraldehyde, improvements in paper density and, consequently, tensile characteristics were observed. However, no similar enhancement in the hydrophobicity of specimens was seen; rather, with the application of the coatings, there were recorded rises in the water retention values and declines in the contact angle values for the specimens, as it was noted from the contact angle and Cobb test results. Fungal growth on the paper surface was also greatly reduced as a result of the coating procedure and the specific antiseptic property of the glutaraldehyde (specifically, when used in 1-5% concentrations in the coatings). In other words, by treating papers with glutaraldehyde, a novel material that is both more water-absorbing and resistant to tensile stress and fungal development has been produced. Thus, the data from this study can serve as a starting point in the future development of a distinctive product that will meet the requirements for hygienic papers for specific applications.

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