

# IMPACT OF GLUTARALDEHYDE CROSSLINKING AGENT ON THE PROPERTIES OF NONWOVEN VISCOSE FABRIC–POLYVINYL ALCOHOL FLEXIBLE COMPOSITES

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In recent years, researchers and industries have made efforts to decrease the reliance on fossil fuel-based materials due to the rise in environmental consciousness. A potential environmentally friendly alternative to petroleum-based, non-biodegradable polymeric materials is known to be natural fiber-reinforced polymer composites. Polyvinyl alcohol (PVA) is a biodegradable synthetic polymer that is water-soluble, easy to use, having film-formation property and good potential as a biodegradable matrix in environmentally friendly composites. The objective of this work has been to create a versatile, lightweight, flexible bio-composite based on nonwoven viscose fabric using PVA and a crosslinking agent (glutaraldehyde), and to analyze the effects of the crosslinking agent and of the nonwoven viscose filler in the PVA matrix on the mechanical and UV properties of the composite. The structural, chemical and mechanical properties of the composite were investigated using FTIR, and tensile and UV testing. The results showed that the composite exhibited high tensile strength, but with a drop in elongation. FTIR verified the interaction of the two polymers in the composites. The flexible bio-composite can be potentially used for food packaging applications.

**Keywords:** nonwoven viscose, PVA, composite, FTIR, glutaraldehyde crosslinking, UV spectroscopy

## INTRODUCTION

Polyvinyl alcohol (PVA) has the form of white granular particles, which are odorless, colorless and flammable. It is water soluble, and has a molecular weight between 26000 and 30000, and a degree of hydrolysis of 85.5–89%. PVA is very versatile, it is used in wood, paper and textile industries, as well as in agribusiness.<sup>1</sup> PVA is non-carcinogenic and non-immunogenic, which means it is not involved in antibody creation.<sup>2</sup> The high gas barrier properties of PVA are explained by its closely packed small crystallites. Due to this, PVA is often used as a barrier film in food packaging applications.<sup>3</sup> The advantageous properties of PVA, such as its ability to form films, its biodegradability, ease of use and solubility recommend it for being used as matrix in environmentally friendly composites.<sup>4</sup>

The compatibility between PVA and cellulose can be increased by using different crosslinking

agents, which can contribute to improving the mechanical and chemical properties of the composite.<sup>5</sup> Crosslinking agents, such as polycarboxylic acids (citric acid, polymaleic, succinic anhydride and citric acid), acetals (1,1,5,5 tetramethoxybutane and 1,1,4,4-tetramethoxybutane), phosphorus derivatives (triethyl phosphate and phosphoric acid) and dialdehydes are known for their biodegradability and low toxicity, being suitable for various applications.<sup>6</sup> In water solutions, glutaraldehyde acts as a hydrate, improving the ability to crosslink cellulose. The resistance to hydrolysis can be increased due to the cyclic structure of the acetal of glutaraldehyde and cellulose.<sup>7</sup> By forming polygluteraldehydes in aqueous solutions, glutaraldehyde can easily polymerize or condensate, it forms products with high thermal stability, which can be acetylated.<sup>8</sup>

The pad-dry-cure procedure is a practical and simple method to prepare thin and dense layers for PVA-modified non-woven viscose fabric.<sup>9</sup> The membrane is immersed into a casting solution that contains a pre-polymer, a monomer and a polymer.<sup>1</sup> When removing the viscose fabric from the casting solution, a thin layer of solution adheres to it. After vaporization of the solvent, when crosslinking occurs at a certain temperature, the layer is immobilized on the the substrate.<sup>10</sup> PVA immobilizes on the surface of the nonwoven fabric, followed by the formation of PVA-modified non-woven fabric.<sup>6</sup> The acetal bridge is formed when the reaction between the aldehyde group of glutaraldehyde and the hydroxyl group of PVA takes place. The reaction is simple and the product formed is stable and non-toxic.<sup>11</sup>

In recent years, researchers have developed thin membrane-like, completely biodegradable composites based on microfibrillated cellulose (MFC) by blending the MFC suspension with polyvinyl alcohol (PVA).<sup>12</sup> Glyoxal was used to perform the chemical crosslinking of PVA, which resulted in improved mechanical and thermal properties of the composites.<sup>12</sup> By comparing the thermal, mechanical, physical and chemical characteristics of generated films with those of neat cellulosic films, the effect of PVA in the composition on cellulose was assessed. In contrast to the cellulosic film, the water absorption, thickness swelling and water vapor permeability of the composite films have been improved. In another study, the development of a hydrogel film for application in food packaging, using PVP (polyvinylpyrrolidone) and CMC (carboxymethyl cellulose), was reported. The biodegradability of the prepared film was investigated and it was found that the material was biodegradable and breathable, and very suitable for food packaging.<sup>3</sup>

In the current research, polyvinyl alcohol was used as matrix, glutaraldehyde (GA) – as crosslinking agent and nonwoven viscose fabric – as reinforcement for developing environmentally friendly and biodegradable packaging materials.

## EXPERIMENTAL

### Materials

Laboratory grade polyvinyl alcohol (with a molecular weight of 86.09 g/mol and 86-89% degree of hydrolyzation) and glutaraldehyde (GA) of 25% concentration were purchased from Fine-Chem S.D., Mumbai. Aqueous solution of PVA and glutaraldehyde was used as matrix, while nonwoven viscose fabric, of 80 g/m<sup>2</sup>, was used as reinforcement in the development of the composite.

### Processing parameters and statistical analysis

The concentrations of PVA solution (w/v%) and GA solution (v/v%), as well as temperature (°C), were varied in order to study their effects on the properties of nonwoven viscose-PVA composites. As listed in Table 1, three different concentrations, *i.e.*, 0.5%, 1% and 1.5% of PVA solutions were used. PVA solution was prepared by continuous stirring at 90 °C for 2 hours. Also, three different concentrations of GA, *i.e.*, 1.25%, 3.13% and 5.0%, were used.

Box Behnken design was used, with 3 levels for each factor, for investigating the effects of the processing parameters, *i.e.* concentration of PVA, concentration of glutaraldehyde and temperature, and for optimizing them. The three levels considered for each process parameter are shown in Table 2. This statistical tool was employed in order to attain the most suitable combination of the selected factors with respect to the tensile properties of the composites.

### Fabrication of nonwoven viscose-PVA-GA composite

Nonwoven viscose-PVA-GA composites were fabricated by the pad dry cure method described in the literature.<sup>11</sup> This involved dipping the nonwoven viscose fabric into a PVA-glutaraldehyde solution, prepared as per the Box Behnken design. The procedure is illustrated in Figure 1. Nonwoven viscose fabric was immersed into the PVA-GA solution for an hour at different temperatures, to allow crosslinking of the PVA/GA with the fabric.<sup>10</sup> Care was taken to avoid the formation of air bubbles during the fabrication of the composite. The PVA soaked viscose fabric was removed from the solution and, after removing the excess solution, the fabric was oven dried at 60 °C for 25 min, and then cured at 120 °C for 5 minutes to ensure the complete formation of the PVA film and the development of crosslinking.

Table 1  
Process parameters and their levels

Factors	Levels		
	Low (-1)	Medium (0)	High (+1)
A – PVA concentration (%)	0.5	1	1.5
B – GA concentration (%)	1.25	3.13	5.0
C – Temperature (°C)	40	60	80

Table 2  
Experimental runs as per Box Behnken Design

Run	Factor A (PVA%)	Factor B (GA%)	Factor C (Temp.)
1	0.5	5.0	60
2	1.0	3.13	60
3	1.0	3.13	60
4	1.5	3.13	80
5	1.0	3.13	60
6	0.5	1.25	60
7	1.0	3.13	60
8	1.5	1.25	60
9	0.5	3.13	40
10	1.5	3.13	40
11	1.0	5.0	40
12	1.0	5.0	80
13	1.0	1.25	80
14	0.5	3.13	80
15	1.5	5.0	60
16	1.0	1.25	40
17	1.0	3.13	60

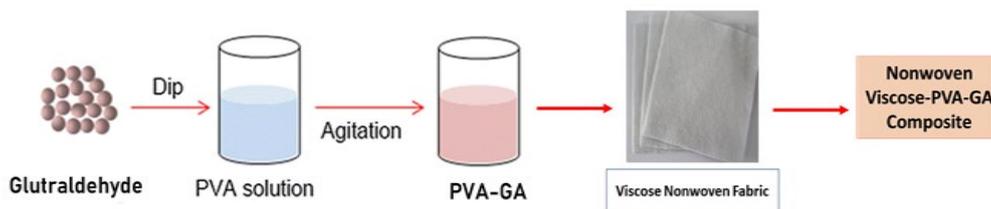


Figure 1: Schematic representation of the fabrication procedure for obtaining nonwoven viscose-PVA-GA composites

## Characterization

### FTIR analysis

An Agilent Technologies Carry 630 FTIR Spectrometer was used for the analysis of nonwoven viscose fabric, and nonwoven viscose-PVA-GA composites in the 400-4000  $\text{cm}^{-1}$  spectrum range, at a resolution of 4  $\text{cm}^{-1}$  by accumulating 24 scans.

### Tensile properties

The tensile properties of the viscose-PVA-GA composites were measured on a Universal Testing Machine (Aimil), by following ASTM D5035. For each sample, three replicates were used.

### UV blockage property

A UV/Visible Spectrophotometer (Shimadzu, UV-2600) was used for UV spectroscopy of untreated nonwoven viscose fabric and nonwoven viscose-PVA composite, in the range of 200-500 (nm) wavelengths. To achieve the average value for determination, three specimens were scanned for each sample.

## RESULTS AND DISCUSSION

### Tensile properties

Table 3 shows the tensile properties of composites with different concentrations of PVA

and GA. The tensile strength for the nonwoven viscose is 6.60  $\text{N/mm}^2$ . The tensile strength of the initial viscose nonwoven fabric is very low, however, after adding PVA and GA to the viscose fabric, the tensile strength significantly enhanced. Also, the tensile strength values of the composites were noted to increase with increasing concentrations of PVA and GA. Thus, the relative increase in the strength of the composites reached 400% to 500%, with respect to that of the initial nonwoven fabric. The effect of increasing temperature on the tensile strength of the composites should also be remarked. Thus, the solutions of PVA/GA were prepared at different temperatures (40, 60 and 80  $^{\circ}\text{C}$ ), and it has been observed that better tensile strength results are obtained at higher temperature. It can be concluded that higher temperature benefits the crosslinking between GA and PVA. The reaction between PVA and GA occurs between the hydroxyl group of PVA and the aldehyde group of GA for the formation of an acetal bridge.

The elongation of initial non-woven viscose fabric is around 103%, but for the composites, a

decreasing trend is observed with higher concentrations of PVA and GA, and temperature. This can be explained by the fact that there is a strong interaction between the components in the composite, which restricts the movement of fibres in the nonwoven viscose, which is ascribed to the presence of hydroxyl groups in both viscose and PVA.<sup>6</sup> This results in decreasing the deformability of the film, which results in an increase in tensile strength, as discussed above, and a reduction in elongation, for the composites, in contrast to the neat viscose fabric.<sup>13</sup>

The variation of tensile strength as a function of the concentration of PVA and GA, as well as temperature, is depicted in Figure 2. Other authors have also reported higher tensile strength with increasing concentrations of GA and PVA, and for higher reaction temperature.<sup>5</sup> The reason for this can lie in the fact that higher temperatures

favor enhanced crosslinking of PVA and GA, contributing to an improved reinforcement effect on the nonwoven viscose fabric.<sup>2,14</sup> The relation between the elongation and the concentration of PVA/GA is presented in Figure 3. The elongation of the composite can be seen to decrease with an increase in the concentrations of PVA and GA.<sup>2</sup> The improved tensile strength, as well as the lower elongation, of the composites can be explained by the fact that, while in the untreated viscose fabric, the fibres are loose, which allows for greater elongation, but poor strength, with the addition of PVA and GA, these immobilize the fibres in the fabric, binding them,<sup>12</sup> which decreases the deformability of the composite, and hence its strength increases, while its elongation decreases.<sup>15,16</sup>

Table 3  
Tensile test results for nonwoven viscose fabric composites at different concentrations of PVA/GA and temperature

Run	PVA conc. (%)	GA conc. (%)	Temp (°C)	Tensile strength (N/mm <sup>2</sup> )	Elongation (%)	Pick-up (%)
1	0.5	5.0	60	10	25.5	4.3
2	1.0	3.13	60	15.3	4.75	9.4
3	1.0	3.13	60	17.5	3.86	12.6
4	1.5	3.13	80	28.5	3.2	17.1
5	1.0	3.13	60	14.65	11.04	12
6	0.5	1.25	60	8	65.1	6
7	1.0	3.13	60	16.6	12	12
8	1.5	1.25	60	15	13	4.4
9	0.5	3.13	40	8.3	34.47	4.4
10	1.5	3.13	40	15.3	20.2	12.5
11	1.0	5.0	40	18.6	3.25	25.3
12	1.0	5.0	80	25.5	2	30
13	1.0	1.25	80	15	20.9	4
14	0.5	3.13	80	10.5	29.5	4.5
15	1.5	5.0	60	20	3.8	20.6
16	1.0	1.25	40	16.15	10.7	13.2
17	1.0	3.13	60	14.5	15.94	10.4

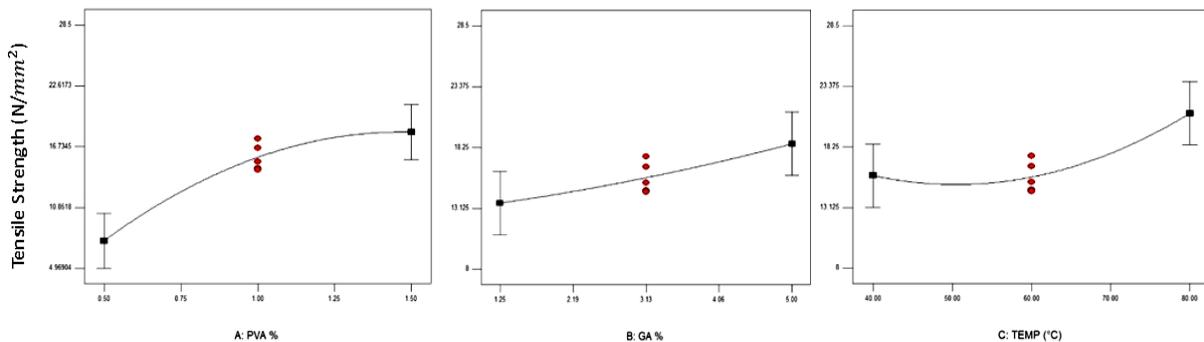


Figure 2: Tensile strength as a function of (A) PVA concentration, (B) GA concentration and (C) Temperature (°C)

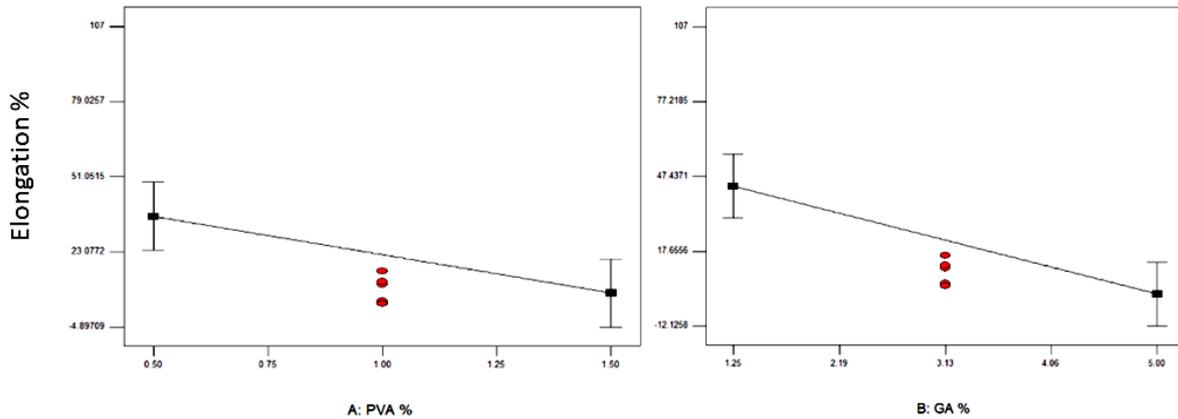


Figure 3: Elongation % as a function of (A) PVA concentration, and (B) GA concentration

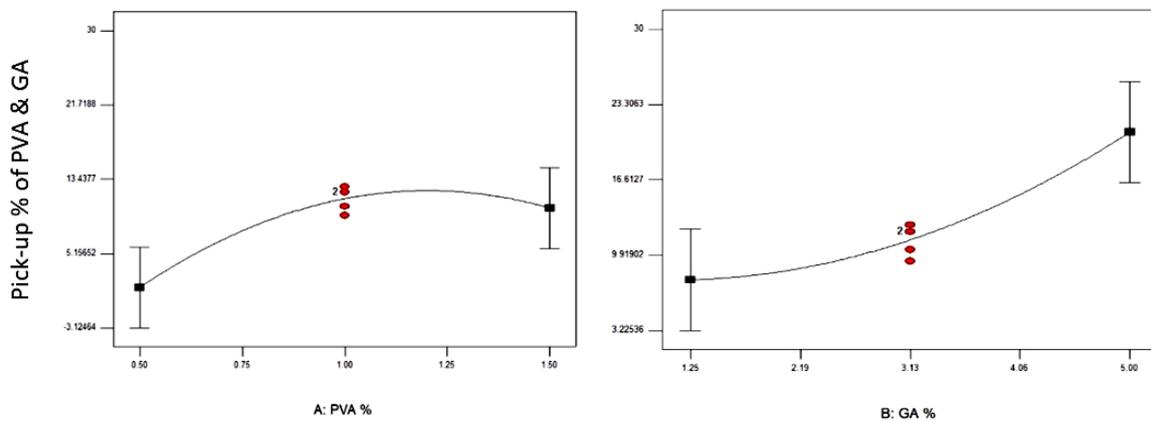


Figure 4: Pick-up % of PVA and GA as a function of (A) PVA concentration, and (B) GA concentration

The pick-up percentage of PVA and GA in the fabric was remarked to rise for higher PVA and GA concentrations, as can be seen in Figure 4. The reason for an increase in the pick-up percentage with the addition of higher PVA and

GA concentrations is ascribed to the synergistic effect of the crosslinking of PVA and GA, which favoured at higher concentrations.<sup>17,18</sup>

Table 4  
Analysis of variance of tensile strength results

Source	Sum of squares	DF	Mean square	F value	Prob > F
Model	418.46	6	69.74	13.89	0.0002
A	220.50	1	220.50	43.92	< 0.0001 (Significant)
B	49.75	1	49.75	9.91	0.0104
C	55.92	1	55.92	11.14	0.0075
A <sup>2</sup>	35.85	1	35.85	7.14	0.0234
C <sup>2</sup>	29.62	1	29.62	5.90	0.0355
AC	30.25	1	30.25	6.03	0.0340
Lack of fit	42.75	6	7.13	3.82	0.1075 (Not significant)
Pure error	7.45	4	1.86		
Cor. total	468.66	16			

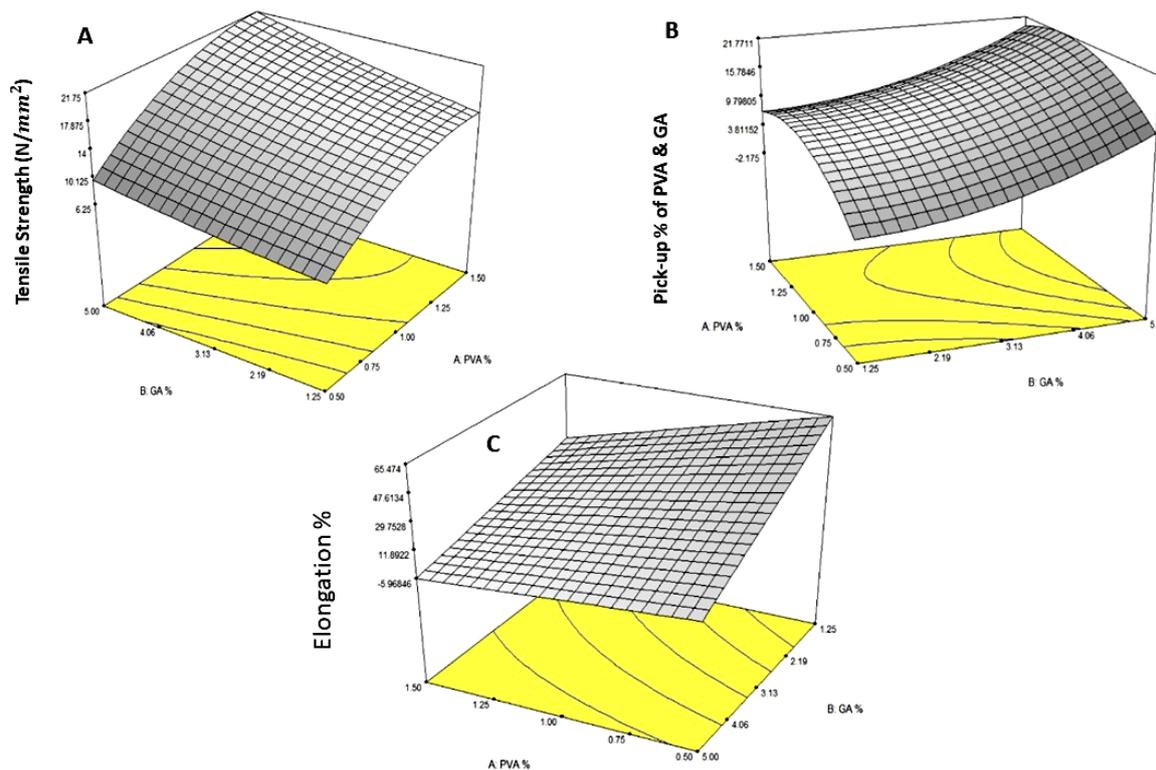


Figure 5: 3D surface plots displaying effects of PVA and GA concentration on (A) tensile strength, (B) pick-up % of PVA and GA, and (C) elongation % of composites

### Statistical analysis of tensile strength

The impact of PVA and GA concentration and temperature on the tensile strength of the nonwoven viscose-PVA-GA composites has been explored by using Box-Behnken Design. The significant results and the two factor interaction effects obtained by ANOVA are shown in Table 4.

The Model F-value of 13.89 suggested that the model is significant. Also, the p-values of concentration of PVA (A), concentration of glutaraldehyde (B), and temperature (C) are statistically significant. The “Lack of Fit F-value” of 3.82 indicates that the lack of fit is not significant relative to the pure error. Only 10.75% chance that a “Lack of Fit F-value” could occur due to noise. Non-significant lack of fit is good.

Figure 5 envisages the effects of PVA and GA concentrations on tensile strength, pick-up % of PVA and GA onto the nonwoven fabric and elongation % of the composites. A drastic increase in tensile strength with the increase in PVA concentration can be seen from the surface plot in Figure 5 (A), meanwhile, the surface plot reveals that the influence of GA concentration on tensile strength is also positive, but less pronounced.<sup>19</sup> The pick-up % also increased with the increase in PVA and GA concentrations (Fig.

5 (B)). The decrease in the elongation % of the composites for higher PVA and GA concentrations is illustrated in Figure 5 (C), and it can be ascribed to the stiffness induced by higher levels of PVA and GA.

### FTIR analysis

Figure 6 shows the spectra of the nonwoven viscose-PVA/GA composite and of the initial nonwoven viscose fabric, allowing comparison between them. Also, the assignments of the absorption bands noted on the spectra of both materials are listed in Table 5. The peak near 3270 cm<sup>-1</sup> is assigned to O-H stretching of hydroxyl groups.<sup>20</sup> It is present in both spectra, however, it should be remarked that the peak corresponding to hydrogen bonds has lower intensity in the spectrum of the nonwoven viscose-PVA/GA composite. The peaks at 2920, 1375 and 1088 cm<sup>-1</sup> are attributed to the C-H elongation, C-H bending and C-O elongation of PVA, respectively.<sup>2</sup> C-H stretching vibration (2892 cm<sup>-1</sup>) is shifted to 2920 cm<sup>-1</sup>, which indicates the breakage of intermolecular hydrogen bonds and the tendency of forming new hydrogen bonds with PVA for the remaining free hydroxyl groups. Transmittance intensity of the bands pertaining to 1500–1000 cm<sup>-1</sup> regions are

appreciably decreased.<sup>21</sup> There is a marginal shift in the position of CH<sub>2</sub> bending vibration, owing to the transmittance band at 1425 cm<sup>-1</sup> (representing

the crystallinity band), which indicates a decrease in the degree of crystallinity of the composite.<sup>22</sup>

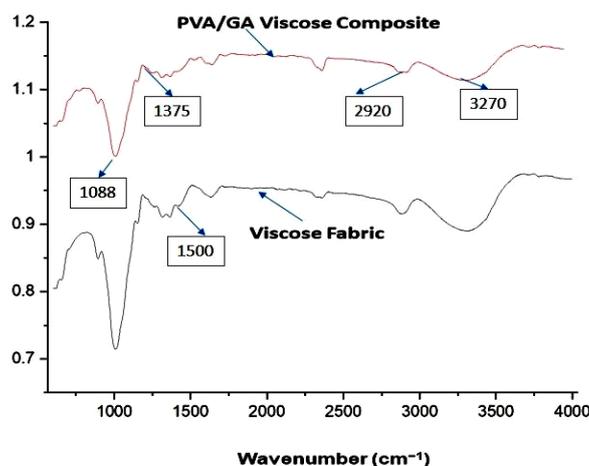


Figure 6: FTIR spectra of initial nonwoven viscose fabric and its composite

Table 5

Assignments of absorption bands in FTIR spectra of nonwoven viscose fabric and its composite

Wavenumber (cm <sup>-1</sup> )	Viscose fabric	PVA/GA composite
OH	3255	3550-3220
CH	2892	3000-2840
C=O	-	1750-1735
CH <sub>2</sub>	-	1460-1470
COO, δCH	1368	1373
C—O	-	1259
C—O, δOH	1017	1150-1085

The reaction between PVA and GA resulted in a significant decrease in the intensity of the O-H peaks, as remarked in Figure 6, indicating the development of acetal bridges in the composite.<sup>23</sup> The FTIR spectrum of the composite material discloses two main bands at 2855 cm<sup>-1</sup> and 2760 cm<sup>-1</sup>, corresponding to C-H stretching connected to aldehydes, a duet absorption related to the alkyl chain.<sup>24</sup> The bands from the carbonyl group were confirmed at 1720-1750 cm<sup>-1</sup> (C=O). The crosslinking of PVA with GA shows a reasonable reduction in the O-H stretching vibration peak (3330-3360 cm<sup>-1</sup>), when compared to that of the pure non-woven viscose.<sup>25</sup> The existence of a peak corresponding to aldehydes in the sample could indicate insufficient reaction of GA with the O-H groups of PVA during the network formation when crosslinking occurs.<sup>3</sup>

#### UV blocking property of PVA/GA composite films

The use of transparent plastic packaging has attracted increasing interest in the last two

decades. The reason for this is the low weight, low cost and design flexibility of transparent plastic packaging, as well as the fact that it allows presenting products more effectively.<sup>17</sup> However, compared to conventional packaging, transparent plastic packaging offers less protection against UV light.<sup>26</sup> Though UV light can sanitize products and delay microbial growth on fruits and vegetables, being thus quite necessary, complete transmittance can cause premature loss of total chlorophyll and a decrease in the Chl a/Chl b ratio in fruits.<sup>27</sup>

The UV blocking ability of the developed composite was measured by the light transmitted through the composite in the wavelength range of 200-500 nm.<sup>28</sup> Three specimens for each sample of initial nonwoven viscose fabric and its composite with PVA/GA were tested in terms of their UV absorbency and the results were compared. Figure 7 shows the transmittance vs. wavelength graph for the composite and the initial fabric. The UV transmittance of the composite is higher, compared to that of the nonwoven viscose

fabric.<sup>29</sup> The maximum transmittance % of the viscose fabric in the range of 450-500 nm is 30.7%, while for the composite, it is 33%. The minimum transmittance % obtained in the range of 200-250 nm was 5.32% for the viscose fabric, and 0.96% for the composite. Hence, the UV blocking ability of the composite is not complete, the minimum transmittance through the composite being of approximately 1%. The surface plot

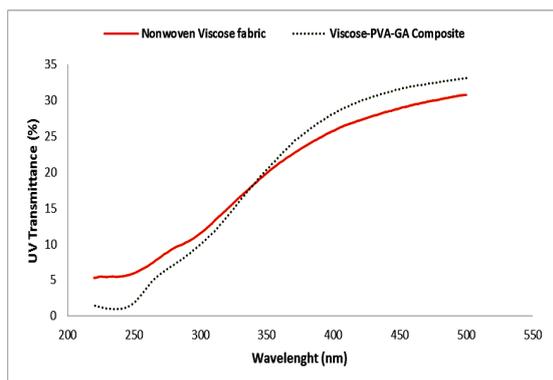


Figure 7: UV transmittance (%) vs. wavelength (nm) for non-woven viscose fabric and viscose-PVA-GA composite

## CONCLUSION

A biodegradable composite has been prepared by using nonwoven viscose fabric as reinforcing element with PVA and GA. Nonwoven viscose fabric was treated by dipping methods to immobilize the PVA/GA on the surface of the fabric. It was found that the use of PVA/GA on the surface of viscose can significantly modify the surface properties of the initial fabric, altering its physical and chemical properties. The interaction of PVA and GA with the nonwoven viscose in the composites was confirmed by FTIR results. The structure of the composite was examined in terms of the appearance of specific functional groups, as confirmed by FTIR analysis. The FTIR spectra indicated that chemical changes occurred in the structure of the nonwoven fabric, the polar groups C–O and C–O–C were introduced to the surface of the fabric after applying PVA/GA. The developed composite exhibited higher tensile strength, compared to the initial viscose fabric, while also showing a drastic reduction in elongation. Tensile strength results were validated through statistical tools. The UV transmittance of the composite was higher than that of the nonwoven viscose fabric. Thus, the obtained flexible biocomposite based on nonwoven viscose

shown in Figure 8 illustrates the effect of PVA and GA concentrations on the UV transmittance of the composites. From the 3D surface plot, it can be noted that, with the increase in the PVA concentration in the formulation, the transmittance percentage decreases, whereas it rises when the GA % increases (Fig. 8). The results of the study agree with others previously reported in the literature.<sup>15</sup>

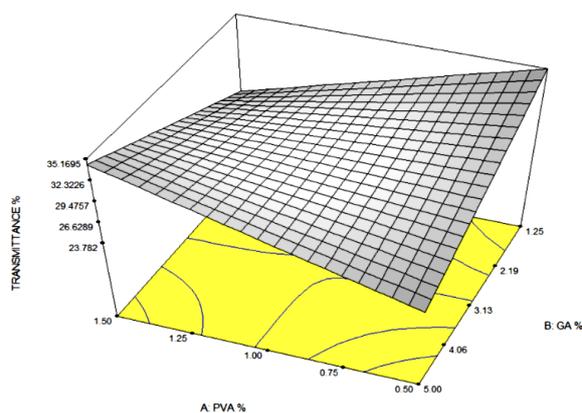


Figure 8: Surface plot of the effects of PVA and GA concentrations on UV transmittance of viscose-PVA-GA composites

and PVA/GA can potentially be used for food packaging application.

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