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Phosphate crosslinking of gum Arabic with sodium trimetaphosphate as a sustainable biopolymer

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Abstract

Gum Arabic (GA), a natural biopolymer, is highly hydrophilic and mechanically unstable, limiting some applications. This study investigates modification of GA using sodium trimetaphosphate (STMP) and changes in its physicochemical and thermal properties. A custom factorial design assessed STMP concentration (2 %, 5 %, and 8 %), pH (10, 11.5, and 13), and temperature (30, 40, and 50 °C), with viscosity and water absorption as responses. Analysis of variance (ANOVA) yielded an R^2 of 0.7519 for the viscosity model and 0.5006 for water absorption. Conditions selected for viscosity were 8 % STMP, pH 13, and 40 °C, producing 4.56 cP (predicted: 4.601 cP) against a maximum of 6.25 cP. Conditions selected for water absorption were 2 % STMP, pH 11.5, and 40 °C, producing 17.85 % (predicted: 12.47 %) against a maximum of 32.57 %. Fourier transform infrared spectroscopy (FTIR) indicated phosphorylation and hydroxyl substitution through P–O–C and P=O stretching vibrations at 976 and 1256 cm^{-1}, respectively. Thermogravimetric analysis (TGA) showed improved thermal stability in the viscosity-optimised sample, with a peak degradation temperature of 382 °C, degradation weight loss of 72.89 %, and residual mass of 23.47 %. This sample showed the most pronounced chemical modification and greatest thermal stability. STMP-crosslinked GA may serve as a sustainable, locally available biopolymer for concrete admixtures, drug-delivery encapsulation, and beverage stabilisers.

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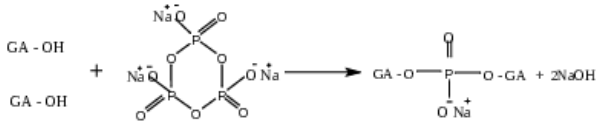
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1. Introduction

Gum Arabic (GA), also known as acacia gum, is a natural biopolymer composed of arabinogalactan (AG), arabinogalactan-protein (AGP), and glycoprotein (GP) fractions, with AGP playing a key role in emulsion stability [1]. Its chemical composition varies with location, species, climate, and processing. GA is light-coloured, odourless, tasteless, and water-soluble, forming viscous colloidal solutions [2]. It possesses both hydrophilic and hydrophobic properties making it an excellent stabiliser for use in emulsions, ink, beverages, pharmaceuticals, and concrete-based construction industries. Native GA's high hydrophilicity poses a serious limitation in certain applications. Previous studies which highlighted the application of GA were carried out using native GA without any form of modification [3, 4].

GA's low moisture resistance and modest mechanical performance are major limitations in most of its applications especially when used as an admixture in concrete. This can

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Scheme 1: Crosslinking reaction of GA with sodium trimetaphosphate [5]

be improved by chemical modification, particularly by the use of chemical cross-linkers such as sodium trimetaphosphate (STMP). Crosslinking reduces the number of free hydroxyl groups present within the GA polysaccharide backbone by phosphorylation. The reaction agrees with the GA crosslinking studies carried out by Ribeiro *et al.* [5] as shown in Scheme 1.

The crosslinked GA exhibits decreased hydrophilicity and water permeability and better workability when used as an admixture in cementitious mixes during building construction. STMP (in contrast to other toxic chemical modifying agents such as phosphorus chloride, epichlorohydrin and citric acid) is an efficient, non-hazardous and non-toxic solid crosslinking agent for starch, forming phosphate ester bonds that improve granule stability and reduce retro-gradation (see Zhao *et al.* [6]). Crosslinking mainly affects the amorphous regions without altering crystalline structure [7], but increases compactness and reduces solubility. STMP-crosslinked GA offers stronger covalent bonds, and higher density [8]. Optimising dosage and conditions using methods like Response Surface Methodology (RSM) could maximise its benefits and applicability [9]. GA's multifunctional nature, coupled with sustainable sourcing and modifiability, makes it a promising green additive across various industries. The purpose of this study is to modify the inherent properties of GA in order to tailor its physical and chemical properties towards a given end usage. The

modification of GA through physical (plasma treatment), chemical (acetylation, esterification), or crosslinking with agents such as STMP enhances properties such as its mechanical strength, hydrophobicity, and reduced water retention, and increases its applicability in various end products [1]. Statistical optimisation methods such as Response Surface Methodology and other Bayesian optimisation are highlighted as tools for determining optimal processing conditions, improving efficiency, and minimising trial-and-error in GA modification and application in the pharmaceutical industry, beverages and other materials [10].

2. Materials and methods

2.1. Sample collection and preparation

The raw GA biopolymer was sourced from a local market in Kano State, Nigeria, while STMP ($Na_3P_3O_9$, ≥ 95 % purity) was supplied by Sigma-Aldrich, absolute ethanol C_2H_5OH (99.5 % purity), acetone $(CH_3)_2CO$, 99 % purity and potassium sulphate (99 % purity) were supplied by JHD, China. Sulfuric acid (H_2SO_4, 99.5 % purity) was supplied by Loba Chemie, India, and sodium hydroxide (NaOH, 98.9 % purity) was supplied by Sigma-Aldrich.

A viscous solution of the raw GA exudate was prepared by dissolving the gum in distilled water followed by intermittent stirring to ensure uniform dispersion and its complete dissolution. The solution was subsequently filtered using a fine linen mesh to remove visible sediments, bark particles, and other insoluble impurities associated with the crude gum exudate [2]. The filtered gum was spread to air dry on clean glass slides measuring 30 *imes* 30 cm (length *imes* breadth). The dried GA was collected and then dried to constant weight in a desiccator. It was pulverised and kept in sealed plastic bags in a desiccator for further use.

Table 1: Independent variables for the custom factorial design.

Independent variable	Units	Low (-1)	Mid*	High (+1)
STMP	%	2	5	8
Temperature	°C	30	40	50
pH		10	11.5	13

2.2. Experimental design

The experimental design was carried out using a custom factorial design and the runs generated using Design Expert v13 statistical package were used in crosslinking the GA at STMP concentration (2, 5, 8 %), temperature (30, 40, 50 °C), and pH (10, 11.5, 13) on GA properties (Table 1). A total of 25 runs were generated to study the viscosity and water absorption as responses (Table 2), while Response Surface Methodology (RSM), was used to model and optimise the responses obtained from the experimental runs.

2.3. Chemical modification of GA

The chemical modification of GA was carried out according to the method reported by Ribeiro *et al.* [5] with slight modifications. GA solutions (20 % w/v) were homogenised, adjusted to target pH using NaOH, and reacted with STMP at set concentrations and temperatures in a water bath. After 4 h, the product was adjusted to pH 7 using 1 mol dm^{-3} H2SO4, washed with ethanol–acetone (1:1), and dried at room temperature (Scheme 1). The dried modified GA samples were further pulverised and stored over silica gel in a desiccator for further use.

2.4. Characterisation of the STMP-modified GA samples

Water absorption (WA) was evaluated for modified GA films, while the viscosity of its corresponding emulsions was measured.

2.4.1. Water absorption

Equilibrium moisture uptake was calculated by measuring the weight change between the dry and moisture-saturated GA films at a relative humidity of 97.3 %, following the guidelines of American Standard for Testing and Materials, ASTM

Table 2: Experimental design matrix with variables expressed in actual units.

Run	A: Conc. (%)	B: Temperature (°C)	C: pH
1	8	50	11.5
2	2	50	13
3	2	40	13
4	2	40	10
5	5	40	13
6	2	50	11.5
7	8	30	13
8	5	30	10
9	5	30	13
10	8	30	11.5
11	5	40	10
12	5	40	11.5
13	8	40	10
14	8	50	13
15	8	50	10
16	2	40	11.5
17	5	50	10
18	2	30	10
19	5	50	13
20	5	50	11.5
21	2	30	13
22	2	30	11.5
23	8	30	10
24	8	40	11.5
25	8	40	13

E104-20a [11]. To fabricate the test specimens, the unmodified GA (as control) and Modified GA samples from the pulverised products were prepared as 30 % w/v solutions and cast onto glass slides. The cast solutions were dried at 50 °C for 24 hours. The resulting films were cut into 50 mm × 13 mm strips and placed in a desiccator until they achieved a constant dry mass. For humidity exposure, the pre-weighed dry films were placed inside a sealed desiccator containing a saturated K_2SO_4 salt solution to maintain a 97.3 % relative humidity atmosphere. Finally, the mass variance between the dry state and the fully saturated state was calculated using Eq. (1).

$$\% \text{ moisture absorption} = \frac{W_2 - W_1}{W_1} \times 100, \tag{1}$$

where W_1 and W_2 are the initial mass of the dried GA sample and the final mass of the moisture-saturated GA sample, respectively.

2.4.2. Viscosity

Following the protocol outlined by Ribeiro *et al.* [5], the viscosity of the modified GA samples was evaluated. To prepare 1 % (w/v) test solutions, 1 g each of the pulverised unmodified (control) and modified GA samples were dissolved in 100 cm^3 of water. Viscosity measurements were carried out at room temperature (25 °C) using a rotational viscometer (NDJ-8S Lonroy, China) fitted with a type 1 spindle, operating at a speed of 60 rpm. All tests were performed in triplicate, and the mean values were reported.

2.5. Statistical analysis of data

2.5.1. Model development

An empirical equation of fitted model terms from Design-Expert was applied to define the relationship between the independent (temperature, pH and STMP concentration) and response variables (water absorption and viscosity). The equation in terms of coded factors can be used to make predictions about the response for given levels of each factor (Eq. (2)). By default, the high levels of the factors are coded as +1 and the low levels are coded as -1. The coded equation is useful for identifying the relative impact of the factors by comparing the factor coefficients. The coefficient estimate represents the expected change in response per unit change in factor value when all remaining factors are held constant. The intercept in an orthogonal design is the overall average response of all the runs. The coefficients are adjustments around that average based on the factor settings. This is necessary in order to correlate the relationship between the experimentally obtained data values and the RSM estimate values obtained on numeric optimisation. This ensures the validity of the optimisation process.

$$\begin{aligned} \text{Response} = \text{intercept} &\pm \text{CE}\,A[1] \pm \text{CE}\,A[2] \\ &\pm \text{CE}\,B[1] \pm \text{CE}\,B[2] \\ &\pm \text{CE}\,C[1] \pm \text{CE}\,C[2], \end{aligned} \tag{2}$$

where ±CE denotes the coefficient estimate (positive for high levels or negative for low levels)

Response: water absorption, viscosity,

A: STMP concentration,

B: Temperature,

C: pH.

2.5.2. Analysis of variance (ANOVA)

To assess the statistical importance of the interactions between independent and dependent variables, an analysis of variance (ANOVA) was conducted on the optimised design at a threshold of p = 0.05. This analytical technique represents a standard practice within custom factorial design, response surface methodology, and optimisation research [12].

2.5.3. The optimisation process

At the end of the factorial experiment and the results obtained, the statistical analysis and numerical optimisation was performed using the desirability function approach [12] as shown in Eq. (3).

$$\begin{aligned} D &= (d_1 \times d_2 \times \cdots \times d_m)^{1/m} \\ &= \left(\prod_{i=1}^{m} d_i \right)^{1/m}, \end{aligned} \tag{3}$$

where D is the desirability function representing how desirable or well-matched the dependent variables or responses were at a selected level of independent variables. In this context, m represents the total count of responses factored into the optimisation process, while d_i reflects the individual desirability score corresponding to each specific response.

2.5.4. Confirmation process

In order to confirm the validity of the optimisation process, the experimentally obtained results from the responses were compared with the model predicted/expected response at the chosen settings. The mean and standard deviation values (Eqs. (4) and (5), respectively) of the measured responses were calculated from the confirmation runs and compared with the actual mean responses with the predicted values [13].

The mean ($\bar{x}$) of confirmation run values $X_1, X_2, \ldots, X_n$ was calculated using Eq. (4).

$$\bar{x} = \frac{1}{n}\sum_{i=1}^{n} x, \tag{4}$$

where $X_1, X_2, \ldots, X_n$ represents the expected or average values of the target operating condition of the monitored process. The sample standard deviation (s) was used (not the population standard deviation) since the variability from a sample of runs were being estimated according to Eq. (5).

$$S = \sqrt{\frac{1}{n-1}}\sum(x_i - x)^2. \tag{5}$$

From the values of the actual (experimental) and predicted interval of the mean and standard deviation, if the actual mean response falls within the prediction interval, the optimisation is considered validated. Also, the residuals were examined for differences between predicted and actual values to check for normality. This ensured that the model assumptions still held at the optimal point. Once confirmation was successful, the model was valid and was used for process control or implementation.

2.6. Determination of the optimised samples from experimental runs results

At the end of the numeric optimisation using RSM, each of the responses measured (water absorption and viscosity) produced a sample that is best for water absorption and for viscosity. All samples were optimised with the goal of the lower-the-better for both water absorption and viscosity. These optimally modified GA samples were further characterised.

2.7. Characterisation of optimised samples

For this study, the optimally modified GA samples were characterised for chemical changes using Fourier Transform Infrared Spectroscopy (FTIR) and thermal stability by Thermogravimetric Analysis (TGA).

2.7.1. Fourier transform infrared spectroscopy (FTIR) of GA

FTIR spectra of approximately 1–2 mg of each of the unmodified and modified GA samples were recorded using an Agilent Cary 630 spectrometer (USA) fitted with a universal attenuated total reflectance (ATR) accessory. Each sample was scanned in the range of 4000 cm^{-1} to 650 cm^{-1} with 32 scans collected at a resolution of 8 cm^{-1} to improve the signal-to-noise ratio.

2.7.2. Thermogravimetric analysis (TGA) of GA

Thermogravimetric Analysis (TGA) was carried out on the optimised GA samples and the unmodified GA to evaluate the thermal stability and decomposition behaviour of the samples by monitoring the change in mass as a function of temperature. The analysis was performed using a TGA 550 Thermogravimetric Analyzer. Approximately 5–10 mg of the dried sample was weighed into a clean alumina crucible and heated from room temperature to 1000 °C at linear controlled heating rates of 0.01–150 °C/min under a nitrogen atmosphere at a 20 mL min^{-1} to prevent oxidative degradation. The instrument's low-mass furnace facilitates cooling between runs. The resulting TGA and derivative thermogravimetric (DTG) curves were used to determine the thermal degradation stages such as the onset decomposition temperature, peak degradation temperature, and residual mass.

3. Results and discussion

Two responses were studied from the modification as carried out following the variables in Table 2. The results of the responses are presented in Table 3. These results were further optimised to obtain the best performing samples of GA in terms of the best combination of modification parameters that will give the lowest viscosity and water absorption. The "predicted versus actual" values in Table 3 show that the model can accurately predict GA behaviour based on the input factors (temperature, pH and concentration)[14, 15].

3.1. Influence of chemical modification on the viscosity and water absorption of GA

The influence of chemical modification on the viscosity of GA is presented in Table 3. From the results, the highest viscosity value of 6.25 ±0.13 cP was obtained for uGA, while the lowest value (4.55 ±0.06 cP) was obtained for a GA sample modified at 5 % STMP concentration, pH 13 and 40 °C temperature. The temperature and pH of modification had the most influence on the viscosity of modified GA samples as compared to the STMP concentration. Generally, an elevation in temperature of modification tends to decrease the viscosity of a biopolymer solution [16]. This could be due to the fact that most polysaccharides have a higher molecular mass and increased temperature, increases the molecular motion of the dissolved biopolymer in solution. As temperature increases, the kinetic energy of the polymer molecules and solvent molecules increases.

This increased molecular motion results in a decline in the thickness of the mix. Furthermore, heightened molecular activity weakens the attractive intermolecular interactions including van der Waals forces and hydrogen bonding that hold the biopolymer chains together. Also, the viscosity of GA and its STMP-modified forms is significantly influenced by pH. Neutral to slightly alkaline conditions favour higher viscosity due to optimal ionisation and crosslinking efficiency, while extreme pH levels can lead to reduced viscosity through chain contraction or degradation [17].

Results from the influence of chemical modification on the water absorption of GA are also as shown in Table 3. It was observed that the sample obtained from 5 % STMP concentration, pH 10, modification conditions of 30 °C had the highest water absorption value of 32.57 % ±0.07, while the least WA value (12.12 % ±0.03) was seen in the sample modified at 2 % STMP concentration, pH 11.5 and a temperature of 50 °C. Studies on other STMP-modified polysaccharides have reported similar increases in water absorption, suggesting a consistent effect of STMP crosslinking on enhancing hydrophilic properties. This indicates that STMP crosslinking can enhance the hydrophilic properties of polysaccharides, although excessive crosslinking may reduce water absorption due to network densification [18]. The introduction of phosphate groups through STMP modification enhances the hydrophilicity of GA leading to increased water absorption (WA).

Also, the temperature of modification has the most impact on the water absorption properties of the modified GA, as the samples modified at high temperatures (50 °C) were observed to have the least water absorption values of 12.12 % ±0.03 (Table 3). The results clearly show that water absorption of the biopolymer samples decreases with increase in temperature of modification. GA exhibits unique water interaction properties that are significantly influenced by temperature changes. This decrease in water absorption at increased temperatures could be attributed to several interrelated factors since heating induces conformational changes in the GA polysaccharide chains, reducing the availability of hydrophilic binding sites. Isothermal experiments between 50 and 60 °C reveal that higher temperatures lead to reduced mass loss, indicating that less water is held by the gum [19]. Research indicates that thermal treatment promotes structural rearrangement of GA molecules, potentially forming more compact configurations that are less accessible to water molecules [20]. This structural reorganisation at elevated temperatures directly impacts the polysaccharide's ability to bind and retain water molecules.

3.2. *Optimisation of crosslinking conditions using response surface methodology*

The optimisation of the responses was carried out using RSM from Design Expert v13 software[15] at different temperatures, pH values and STMP concentrations (Table 2) to obtain the optimum conditions for the cross-linking. Table 3 shows the experimental values (Actual) and the predicted values of the responses as generated by the software. The lowest experimental and predicted response values for viscosity were at run number 5 (4.55 cP and 4.68 cP respectively), at STMP concentration of 5, pH 13 and 40 °C (Table 3). The highest experimental and predicted response values was the uGA (6.25 cP and 6.57 cP respectively). Kersten *et al.* [21]assert that GA solution viscosity correlates with molecular weight. The higher the molecular weight, the higher the viscosity. Unmodified GA can show higher viscosity as also observed in this study. STMP concentration and alkaline conditions affect rheological (viscosity) behaviour of polysaccharides. Therefore, specific STMP, pH and temperature combinations can produce lower or higher viscosities [22].

The lowest experimental and predicted response values for water absorption were at run number 6, at 12.12 and 12.50 %, respectively, at STMP concentration of 2 %, pH 11.5 and 50 °C. While, the highest experimental and predicted response values for WA were at run number 8 (32.57 and 24.93 respectively) at STMP concentration of 5, pH 10 and 30 °C. It is established that STMP concentration and processing conditions (such as pH and temperature) are commonly varied and have proven to influence water absorption in films and hydrogels. Typical STMP cross-linking procedures use alkaline conditions (approx. pH 11), which affects the degree of phosphorylation and hence the observable differences in water uptake [23]. According to Almonaityte *et al.* [24], natural starches are unstable to changes in pH and temperature. Therefore, minor changes in pH and temperature in the presence of STMP could influence its degree of cross-linking thereby bringing about the variations observed in the viscosity and water absorption of the mGA.

3.3. *Statistical optimisation using RSM*

Statistical optimisation was carried out for each of the responses studied so as to obtain the optimal combination factors of GA modification based on the independent variables selected for this study (pH, temperature and STMP concentration). The quality of fit for each outcome's regression model was evaluated using the coefficients of determination (R^2 and adjusted R^2), which quantify the proportion of variance in the measured data attributable to the main variables and their joint effects. The value closer to 1.0 shows a better fitting of the model with the actual data and a good correlation between experimental and predicted values. The optimisation assessed the significance of the input variables and their interactions, with different confidence levels for each response. Regression models were developed to relate variables to responses, and desirability functions optimised multiple outcomes simultaneously.

3.3.1. *Statistical optimisation for viscosity of modified gum Arabic*

Statistical significance for the model is demonstrated by its F-value of 8.59, with a negligible 0.02 %, a probability that a value of this magnitude is the result of random experimental noise. With p-values below 0.05 signalling key model terms, both Temperature (B) and pH (C) show a significant effect. Furthermore, as detailed in Table 4, pH exerts a greater influence on the viscosity of the modified GA than temperature, reflected by their relative F-values of 21.39 and 4.09, respectively. p-values greater than 0.1 indicate that the model term is not significant.

Table 3: Experimental and predicted response values from the factorial experimental design.

Run	Conc. (%)	pH	Temp. (°C)	Visc. actual (cP)	Visc. pred. (cP)	WA actual (%)	WA pred. (%)
GA	0	0	0	6.25	6.57	15.71	16.88
1	8	11.5	50	5.44	5.42	14.15	14.95
2	2	13	50	5.20	5.03	19.36	14.53
3	2	13	40	4.84	4.79	13.61	14.49
4	2	10	40	5.3	5.15	15.68	14.11
5	5	13	40	4.55	4.68	19.94	19.51
6	2	11.5	50	5.38	5.61	12.12	12.50
7	8	13	30	4.68	4.68	22.15	22.73
8	5	10	30	5.08	5.13	32.57	24.93
9	5	13	30	4.77	4.76	27.87	25.30
10	8	11.5	30	5.31	5.25	16.3	20.71
11	5	10	40	5.32	5.05	13.86	19.13
12	5	11.5	40	5.07	5.26	14.68	17.49
13	8	10	40	4.68	4.97	16.02	16.56
14	8	13	50	4.72	4.85	14.84	16.97
15	8	10	50	5.29	5.22	17.64	16.60
16	2	11.5	40	5.4	5.36	17.85	12.47
17	5	10	50	5.17	5.30	17.03	19.17
18	2	10	30	5.06	5.23	12.31	19.90
19	5	13	50	4.89	4.93	17.51	19.55
20	5	11.5	50	5.78	5.51	19.15	17.53
21	2	13	30	4.94	4.86	18.08	20.28
22	2	11.5	30	5.35	5.44	17.53	18.26
23	8	10	30	5.21	5.05	27.65	22.35
24	8	11.5	40	5.28	5.17	17.03	14.91
25	8	13	40	4.56	4.60	14.81	16.93

This is the case with STMP concentration (p-value = 0.1448). Its F-value of 2.17 also suggests that it exerts little or no influence on the viscosity of the modified GA samples.

The empirical link between viscosity and the process parameters expressed via coded factors is detailed in Eq. (6) for the modified GA, featuring a standard deviation of 0.1743 and intercept of 5.11, relative to the average viscosity.

$$\begin{aligned}\text{Viscosity} = 5.11 &+ 0.0952A[1] - 0.0057A[2] \\ &- 0.0313B[1] - 0.1083B[2] \\ &+ 0.0552C[1] + 0.2587C[2],\end{aligned} \tag{6}$$

where A, B and C represent the input factors of STMP concentration, Temperature and pH respectively, while the corresponding values are their coefficient estimates. The coefficient estimates for each input factor represent the expected change in viscosity per unit change in pH and temperature values (which are the most significant factors), when the remaining factor (STMP Conc.) is having little or no impact on the viscosity. This is observed from its low coefficient estimates. The viscosity ultimate equation (Eq. (6)) in terms of coded factors is as generated by the Design Expert software.

3.3.2. *Confirmation of the viscometry statistical model*

Statistical values as obtained from the ANOVA (Table 5) further establish that the predictive model described by Eq. (2) for the viscosity results agree with the experimental data obtained for the modified GA. This is represented in Figures 1 and 2.

3.3.3. *ANOVA confirmation from "predicted versus actual" plot*

From the predicted versus actual values of the viscosity of mGA (Table 5), the model's reliability and accuracy are highlighted by the strong relationship between predicted and experimental values. An observable tight clustering along the diagonal line indicates a strong model fit [15]. A good "predicted versus actual" plot shows points close to the 45° line (line of perfect fit where predicted = actual), with an even distribution along the line with no systematic deviation or curving pattern. The "predicted versus actual" plot (Figure 1) means the model can accurately predict GA behaviour based on the input factors (temperature, pH and concentration)[14, 15].

ANOVA confirmation from the "residuals versus run" plot

The residuals versus run plot of the viscosity of mGA samples showed scattered points randomly above and below zero (Figure 2). This shows no visible trend, curvature, or clustering, with an even spread of residuals across all run numbers. An observed random scatter in the plot indicates good model validity [25]. This confirms that the experimental results are not biased by time or order of runs and the assumption of independence is valid. This supports that the ANOVA results (F-

Table 4: Viscosity ANOVA for the selected factorial model.

Source	Sum of squares	df	Mean square	F-value	p-value	Significance
Required	Intercept = 5.11					
Model	1.57	6	0.2609	8.59	0.0002	Significant
A-Conc.	0.1319	2	0.0659	2.17	0.1448	
B-Temperature	0.2485	2	0.1242	4.09	0.0355	
C-pH	1.30	2	0.6499	21.39	< 0.0001	
Residual	0.5165	17	0.0304			
Cor Total	2.08	23				
Std. Dev.	0.1743		R^2		0.7519	
Mean	5.11		Adjusted R^2		0.6643	
C.V. %	3.41		Predicted R^2		0.5026	
			Adeq. Precision		9.8598	

Table 5: Statistically predicted versus actual viscosity response values for modified GA.

Run order	Actual	Predicted	Residual	Leverage	Std. order
1	5.44	5.42	0.0182	0.264	16
2	5.20	5.03	0.1662	0.301	23
3	4.84	4.79	0.0541	0.301	20
4	5.30	5.15	0.1450	0.301	6
5	4.55	4.68	-0.1350	0.301	21
6	5.38	5.61	-0.2264	0.301	14
7	4.68	4.68	0.0017	0.301	19
8	5.08	5.13	-0.0511	0.301	2
9	4.77	4.76	0.0080	0.301	18
10	5.31	5.25	0.0591	0.301	10
11	5.32	5.05	0.2660	0.264	4
12	5.07	5.26	-0.1875	0.301	12
13	4.68	4.97	-0.2903	0.301	5
14	4.72	4.85	-0.1292	0.301	22
15	5.29	5.22	0.0717	0.301	8
16	5.40	5.36	0.0415	0.264	11
17	5.17	5.30	-0.1320	0.301	7
18	5.06	5.23	-0.1720	0.301	1
19	4.89	4.93	-0.0429	0.264	24
20	5.78	5.51	0.2745	0.301	15
21	4.94	4.86	0.0771	0.264	17
22	5.35	5.44	-0.0855	0.301	9
23	5.21	5.05	0.1626	0.264	3
24	5.28	5.17	0.1062	0.301	13
25	4.56	4.60	0.0410	0.264	25

tests, p-values) are reliable, hence the model assumptions hold well [26].

3.3.4. Numerical optimisation of the viscosity results

The optimisation process carried out using RSM was based on a goal of achieving a modified sample with minimum and suitable viscosity. From the constraint as outlined on Table 6, the numeric optimisation selected a sample with 8 % STMP, pH 13 and 40 °C as the best sample from 25 combinations of categoric factor levels (Table 7). The selection was based on the desirability values as obtained for each set of combination factors deduced from the RSM software from the runs' experimental results of a given response. These desirability values determine the optimum combination of the input factors for the crosslinking conditions of GA that leads to the maximised response while satisfying the restriction of the model [27]. The best desirability value of 0.958 was obtained for the best sample optimised at 8 % STMP, pH 13 and 40 °C. An overall desirability closer to 1 indicates better optimisation [15]. Thus, these modification conditions were selected as the best for viscosity (Table 6).

Solutions for 25 combination of categoric factor levels

The R^2 coefficient of 0.7519 (Table 4), recorded for viscosity reflects how closely the experimental results align with the

Table 6: Numerical optimisation constraints for viscosity.

Name	Goal	Lower limit	Upper limit	Lower weight	Upper weight	Importance
A:Conc.	is in range	2	8	1	1	3
B:Temperature	is in range	30	50	1	1	3
C:pH	is in range	10	13	1	1	3
Viscosity	minimise	4.55	5.78	1	1	3

Table 7: Numerical optimisation solutions for viscosity.

No.	STMP (%)	Temp. (°C)	pH	Viscosity (cP)	Desirability	Selection
1	8	40	13	4.601	0.958	Selected
2	8	30	13	4.678	0.896	
3	5	40	13	4.685	0.890	
4	5	30	13	4.762	0.828	
5	2	40	13	4.786	0.808	
6	8	50	13	4.849	0.757	
7	2	30	13	4.863	0.746	
8	5	50	13	4.933	0.689	
9	8	40	10	4.970	0.658	
10	2	50	13	5.034	0.607	
11	8	30	10	5.047	0.596	
12	5	40	10	5.054	0.590	
13	5	30	10	5.131	0.528	
14	2	40	10	5.155	0.508	
15	8	40	11.5	5.174	0.493	
16	8	50	10	5.218	0.457	
17	2	30	10	5.232	0.446	
18	8	30	11.5	5.251	0.430	
19	5	40	11.5	5.258	0.425	
20	5	50	10	5.302	0.389	
21	2	40	11.5	5.358	0.343	
22	8	50	11.5	5.422	0.291	
23	2	30	11.5	5.436	0.280	
24	5	50	11.5	5.506	0.223	
25	2	50	11.5	5.606	0.141	

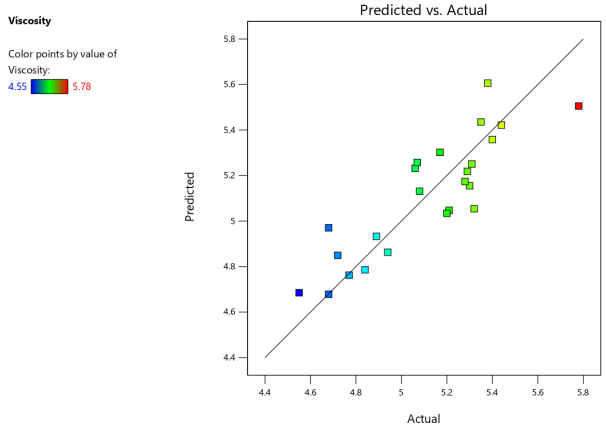


Figure 1: Predicted versus actual viscosity values for modified GA.

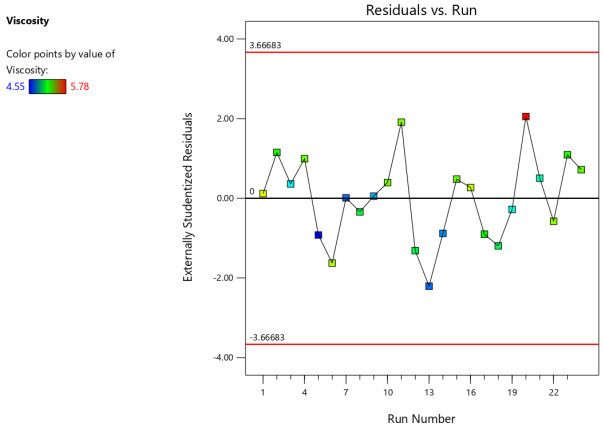


Figure 2: Residuals versus run values of viscosity for modified GA.

predicted values. It demonstrates a robust relationship between the observed data for mGA and its predicted model. Furthermore, the predicted R^2 value of 0.5026 aligns well with the adjusted R^2 of 0.6643, as the variance between the two remains below the 0.2 threshold. Adequate precision evaluates the signal-to-noise ratio. According to Stat-Ease [15], a ratio exceeding 4 is considered ideal. Thus, the adequate precision value of 9.8598 achieved in this research confirms a sufficient signal. This outcome indicates that the established model can effectively navigate the design space. Consequently, after excluding non-significant terms, the resulting empirical relationship between viscosity and the process variables (expressed via coded factors A, B, and C) for the modified GA is defined by Eq. (6), featuring a standard deviation of 0.1743 cP and intercept of 5.11 relative to the mean viscosity.

From all the factors plot (Figure 3), increase in both temperature and STMP concentration decreases the viscosity. Non-parallel lines (intersecting or diverging) mean that an interaction exists. That is, the response changes differently depending on the combination of factor levels, an indication that the combined effect is not additive, and both factors must be considered together when optimising or interpreting results [14].

The experimental results show that increase in STMP concentration decreases the viscosity, hence higher viscosity values were mainly obtained within the lower to medium STMP concentrations, (5.78 cP as obtained at 5 % STMP, pH 11.5). This is in agreement with a study carried out by Ribeiro *et al.* [5].

3.4. Statistical optimisation for water absorption of modified gum Arabic

An analysis of variance was conducted to evaluate the water absorption characteristics of the modified GA, with detailed outputs presented in Table 8. The selected model demonstrated statistical significance, as evidenced by an F-value of 2.84 (p-value = 0.0421), with merely a 4.21 % (p-value = 0.0421) likelihood of a value this high arising from random noise. Model terms were deemed significant at p-values < 0.0500, highlighting temperature (factor B) as a significant term (Table 8), whereas terms with p-values exceeding 0.05 were considered non-significant. In this case, B (temperature) is the most significant model term, having the greatest influence on the water absorption. This is reflected by its F-value of 5.03. p-values greater than 0.05 indicate that the model term is not significant. This is the case with STMP concentration (p-value = 0.0854), its F-value of 2.85 also suggests that it exerts little or no influence on the water absorption of the modified GA samples. The same applies to changes in pH (p-value = 0.6017, and F-value = 0.5235).

The empirical link between water absorption and the process parameters expressed via coded factors is detailed in Eq. (7) for the modified GA, featuring a standard deviation of ±4.13 % and intercept of 18.12, relative to the average water absorption.

$$\begin{aligned}\text{Water absorption} = {} & 18.12 - 2.49A[1] + 2.53A[2] \\ & + 3.85B[1] - 1.94B[2] \\ & + 0.4235C[1] - 1.22C[2],\end{aligned} \tag{7}$$

where A, B and C represent the input factors of STMP concentration, Temperature and pH respectively, while the corresponding values are their coefficient estimates. The coefficient estimates for each input factor represents the expected change in water absorption per unit change in temperature value (which is the most significant factor), when the remaining factors (STMP Conc. and pH) are having little or no impact on the water absorption, this is observed from their low and negative coefficient of estimate values. The water absorption ultimate equation in terms of coded factors is as generated by the Design Expert software (Eq. (7)).

3.5. Confirmation of the water absorption statistical model

Statistical values as obtained from the ANOVA (Table 9) further establishes that the predictive model described by Eq. (7) for the water absorption results agree with the experimental data obtained for the modified GA. This is further represented in Figures 4 and 5.

3.5.1. ANOVA confirmation from "predicted versus actual" plot

A reliable "predicted versus actual" plot (Table 9) shows points close to the 45° line with an even distribution along the line on either side with no systematic deviation or curving pattern as seen in Figure 4. When the observed data align closely with the forecasted outcomes, it demonstrates that the model possesses a high degree of precision and dependability. An observable tight clustering along the diagonal line indicates a strong model fit [15]. This means that the model predictions closely match the experimental data. This therefore means that the model can accurately predict GA behavior based on the input factors (temperature, pH and concentration)[14, 15].

ANOVA confirmation from "residuals versus run" plot

A reliable "residuals versus run" plot will show scattered points randomly above and below zero, shows no visible trend, curvature, or clustering, with an even spread of residuals across all run numbers as seen in the residuals versus run plot for WA (Figure 5). An observed random scatter in the plot indicates good model validity [25], confirming that the experimental results are not biased by time or order of runs and the assumption of independence is valid. This supports that the ANOVA results (F-tests, p-values) are reliable, hence the model assumptions hold well [26].

3.5.2. Numerical optimisation of water absorption results

The optimisation process as carried out using RSM was based on a goal of achieving a modified sample with the lowest and suitable water absorption. Based on the constraints as outlined on Table 10, the numeric optimisation selected a sample prepared with 2 % STMP concentration, pH 11.5 and at a temperature of 40 °C as the best sample from 25 combinations of

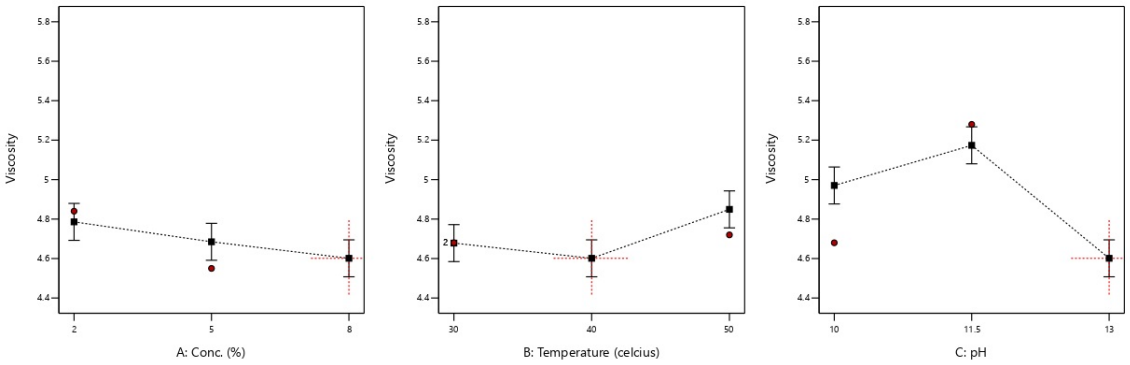


Figure 3: All-factors plot of the optimised viscosity selection.

Table 8: Water absorption ANOVA for the selected factorial model.

Source	Sum of squares	df	Mean square	F-value	p-value	Significance
Required	Intercept = 18.12					
Model	290.58	6	48.43	2.84	0.0421	Significant
A-Conc.	97.31	2	48.65	2.85	0.0854	
B-Temperature	171.44	2	85.72	5.03	0.0193	
C-pH	17.86	2	8.93	0.5235	0.6017	
Residual	289.93	17	17.05			
Cor Total	580.51	23				
Std. Dev.	4.13		R^2		0.5006	
Mean	18.12		Adjusted R^2		0.3243	
C.V. %	22.79		Predicted R^2		0.0107	
			Adeq. Precision		5.7561	

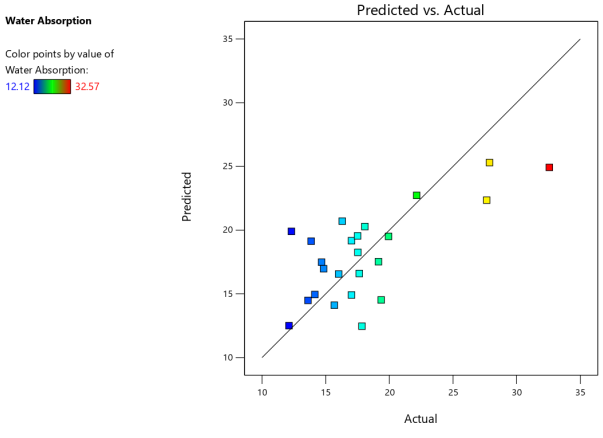


Figure 4: Predicted versus actual water absorption values for modified GA.

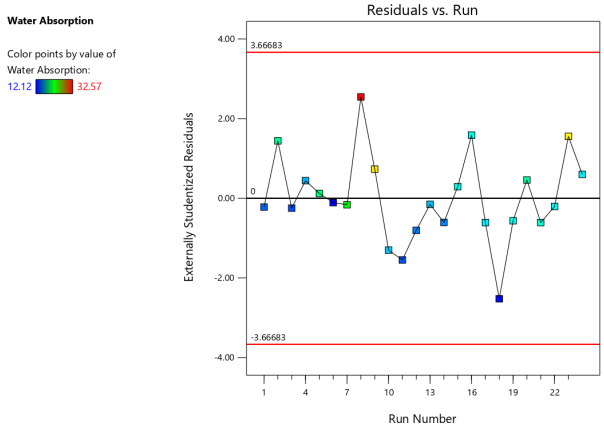


Figure 5: Residuals versus run values of water absorption for modified GA.

categoric factor levels (Table 11). The selection was based on the desirability values as obtained for each set of combination factors deduced from the RSM software from the runs' experimental results of a given response. These desirability values determine the optimum combination of the input factors for the crosslinking conditions of GA that leads to the maximised response while satisfying the restriction of the model [27]. The best desirability value of 0.983 was obtained for the best sample optimised at 2 % STMP concentration, pH 11.5 and a temperature of 40 °C. An overall desirability closer to 1 indicates better optimisation [15]. Thus, these sample modification conditions were selected as the best for water absorption (Table 11).

Solutions for 25 combinations of categoric factor levels.

The experimental results showed that increase in tempera-

Table 9: Predicted versus actual water absorption response values for modified GA.

Run order	Actual	Predicted	Residual	Leverage	Std. order
1	14.15	14.95	-0.8010	0.264	16
2	19.36	14.53	4.83	0.301	23
3	13.61	14.49	-0.8782	0.301	20
4	15.68	14.11	1.57	0.301	6
5	19.94	19.51	0.4296	0.301	21
6	12.12	12.50	-0.3838	0.301	14
7	22.15	22.73	-0.5782	0.301	19
8	32.57	24.93	7.64	0.301	2
9	27.87	25.30	2.57	0.301	18
10	16.30	20.71	-4.41	0.301	10
11	13.86	19.13	-5.27	0.264	4
12	14.68	17.49	-2.81	0.301	12
13	16.02	16.56	-0.5393	0.301	5
14	14.84	16.97	-2.13	0.301	22
15	17.64	16.60	1.04	0.301	8
16	17.85	12.47	5.38	0.264	11
17	17.03	19.17	-2.14	0.301	7
18	12.31	19.90	-7.59	0.301	1
19	17.51	19.55	-2.04	0.264	24
20	19.15	17.53	1.62	0.301	15
21	18.08	20.28	-2.20	0.264	17
22	17.53	18.26	-0.7282	0.301	9
23	27.65	22.35	5.30	0.264	3
24	17.03	14.91	2.12	0.301	13
25	14.81	16.93	-2.12	0.301	25

Table 10: Numerical optimisation constraints for water absorption.

Name	Goal	Lower limit	Upper limit	Lower weight	Upper weight	Importance
A:Conc.	is in range	2	8	1	1	3
B:Temperature	is in range	30	50	1	1	3
C:pH	is in range	10	13	1	1	3
Water Absorption	minimise	12.12	32.57	1	1	3

ture decreases the water absorption of modified GA samples. The least water absorption values were observed at temperatures above 30 °C (12.12 % at 50 °C), while the highest WA value (32.57 %) were obtained at the least modification temperature of 30 °C. This is because, at elevated temperatures (>25 °C), GA's branched polysaccharide protein complexes (arabinogalactan-protein) undergo structural changes like hydrophobic clustering in which the protein-rich regions aggregate, limiting the hydrogen bonding sites for water [28]. GA at this temperature also exhibits decreased hygroscopicity as phase separation disrupts the polysaccharide network, reducing water retention within the polysaccharide backbone [29].

From the water absorption all factors plot (Figure 6), increase in temperature of modification decreases the water absorption properties of the modified GA. Non-parallel lines (intersecting or diverging) mean that an interaction exists. These enable changes in the observed responses depending on the combination of factor levels.

The subsequent characterisation and response summary are illustrated in Figures 7–9 and Tables 12 and 13.

3.6. Influence of chemical modification on properties of GA

Natural GA exhibits high water absorption due to the presence of hydrophilic, hydroxyl and carboxyl groups [28]. While crosslinked GA hydrogels display pH-dependent swelling, blends improve moisture barrier properties in films and packaging. Chemical crosslinking of GA with STMP significantly altered its physicochemical behavior. From this study, the results show that increasing STMP levels decreased viscosity. This is attributed to polymer network disruption and reduced chain entanglement [5]. For water absorption, STMP enhanced hydrophilicity by introducing phosphate groups, improving water retention, although excessive crosslinking reduces absorption due to denser networks.

3.6.1. Influence of input factors on the viscosity of modified GA

Viscosity of the modified GA samples was observed to be most influenced by temperature and pH rather than STMP

Table 11: Numerical optimisation solutions for water absorption.

No.	STMP (%)	Temp. (°C)	pH	Water absorption (%)	Desirability	Selection
1	2	40	11.5	12.465	0.983	Selected
2	2	50	11.5	12.504	0.981	
3	2	40	10	14.112	0.903	
4	2	40	13	14.488	0.884	
5	2	50	13	14.527	0.882	
6	8	40	11.5	14.913	0.863	
7	8	50	11.5	14.951	0.862	
8	8	40	10	16.559	0.783	
9	8	50	10	16.598	0.781	
10	8	40	13	16.935	0.765	
11	8	50	13	16.974	0.763	
12	5	40	11.5	17.488	0.738	
13	5	50	11.5	17.526	0.736	
14	2	30	11.5	18.258	0.700	
15	5	40	10	19.134	0.657	
16	5	50	10	19.173	0.655	
17	5	40	13	19.510	0.639	
18	5	50	13	19.549	0.637	
19	2	30	10	19.905	0.619	
20	2	30	13	20.281	0.601	
21	8	30	11.5	20.705	0.580	
22	8	30	10	22.352	0.500	
23	8	30	13	22.728	0.481	
24	5	30	10	24.927	0.374	
25	5	30	13	25.303	0.355	

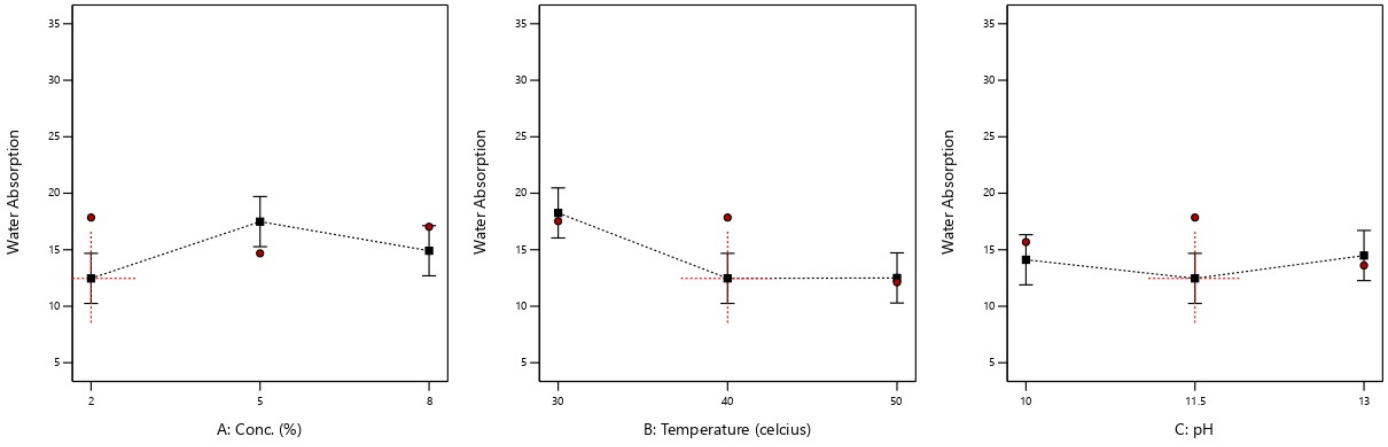


Figure 6: All-factors plot of the optimised water absorption selection.

Table 12: Onset and peak decomposition temperatures of unmodified and modified GA.

Sample	Initial mass loss (%)	Onset temperature (°C)	Peak temperature (°C)	Degradation loss (%)	Residue (%)	Decomposition steps
uGA	4.10 and 5.88	220 and 439.22	No distinct peak	74.52	15.50	2 distinct degradation steps
WA (2% pH 11.5)	8.47	296 and 363.54	367	87.91	3.62	2 distinct degradation steps
Viscosity (8% pH 13)	4.32	319.4	382	72.89	23.47	One sharp step

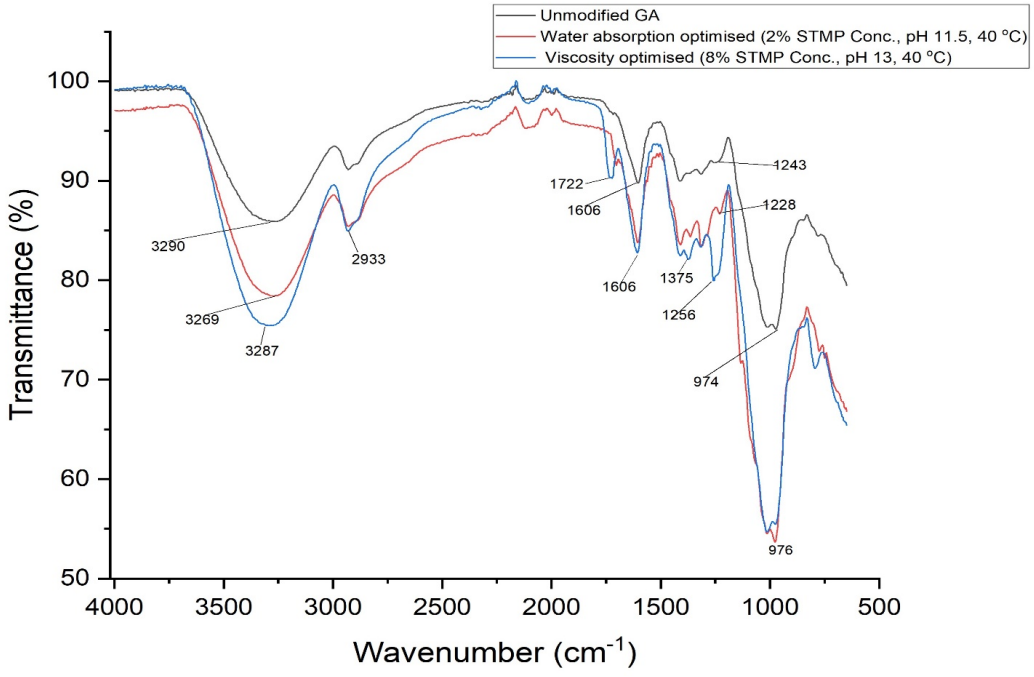


Figure 7: FTIR spectra of unmodified and modified GA samples.

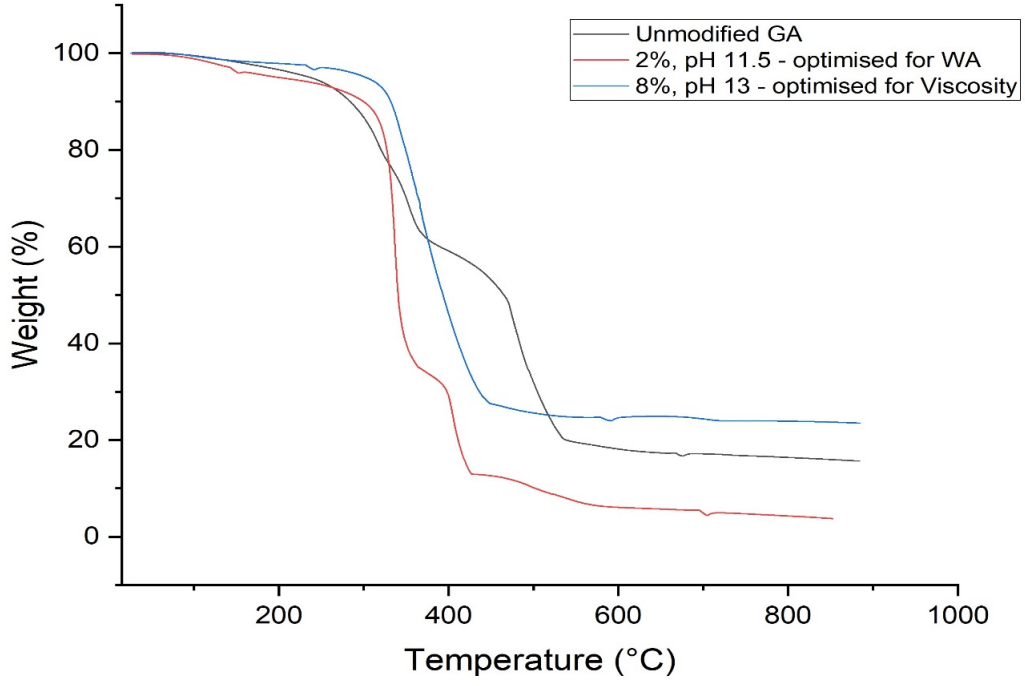


Figure 8: TGA curves of unmodified GA and water-absorption-optimised (2 % STMP, pH 11.5) and viscosity-optimised (8 % STMP, pH 13) samples at 40°C.

Table 13: Summary of responses from optimised samples.

Response	STMP (%)	Temp. (°C)	pH	Observed response	Predicted response	Desirability	ANOVA R^2
Viscosity	8	40	13	4.56 cP	4.601 cP	0.958	0.7519
Water absorption	2	40	11.5	17.85 %	12.465 %	0.983	0.5006

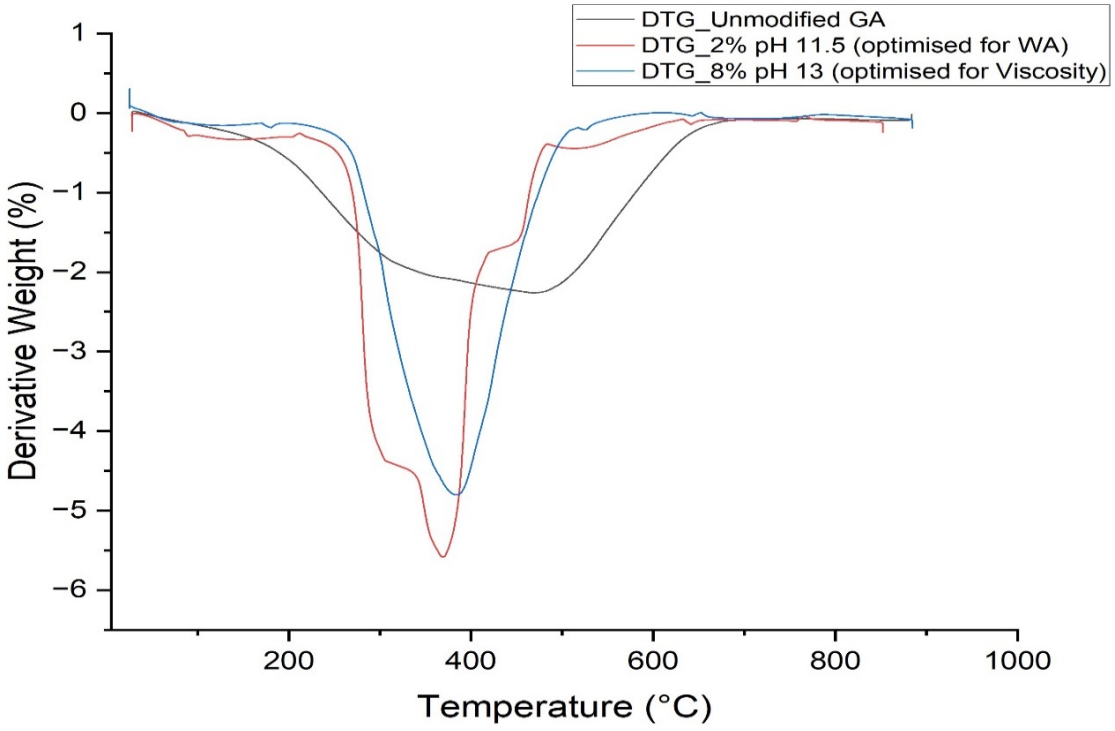


Figure 9: DTG curves of unmodified GA and water-absorption-optimised (2% STMP, pH 11.5) and viscosity-optimised (8% STMP, pH 13) samples at 40°C.

concentration. Rising temperature decreased viscosity because higher molecular motion weakened hydrogen bonding and intermolecular forces. pH also played a critical role since at alkaline conditions, deprotonation of carboxyl groups increased electrostatic repulsion and solubility, lowering viscosity. ANOVA for viscosity (Table 4) confirmed the statistical significance of the model (F-value 8.59, $p < 0.05$). Optimisation identified the best condition as 8 % STMP, pH 13, and 40 °C, which yielded the viscosity of 4.56 (predicted: 4.601 cP) with high desirability of 0.958 (Table 7).

3.6.2. Influence of input factors on the water absorption of modified GA

Temperature emerged as the most significant factor controlling water absorption. Elevated temperatures reduced water uptake due to conformational rearrangements, loss of free volume, and weakened hydrogen bonding. This aligns with Flory-Huggins theory and FTIR evidence of hydrogen bond disruption at high thermal conditions [30]. ANOVA results (Table 8) confirmed significance ($F = 2.84$, $p = 0.0421$), with temperature showing the strongest effect. Optimisation selected the sample prepared at 2 % STMP, pH 11.5, and 40 °C as the best condition, yielding a water absorption value of 17.85 % (predicted: 12.465 %) as against the highest observed experimental value of 32.57 % with desirability value of 0.983 (Table 10).

3.7. Characterisation of optimised samples

The optimal samples for viscosity and water absorption as obtained from the optimisation process using RSM along with unmodified GA were further analysed for FTIR and TGA.

3.7.1. Fourier transform infrared spectroscopy (FTIR) analysis

FTIR spectroscopy revealed significant chemical changes in GA after modification (Figure 7). Unmodified GA displayed strong hydroxyl (-OH) stretching bands, characteristic of natural polysaccharides. FTIR spectra of the sample modified at 2 % STMP concentration, pH of 11.5, and a temperature of 40 °C showed P-O-C stretching vibration at around 976 cm^{-1} and a mild band weakening with a shift at 3269 cm^{-1} and 1228 cm^{-1}, indicating slight oxidation and phosphorylation respectively. The observable changes were minimal, suggesting low modification at this pH and concentration. Sample modified at 8 % STMP concentration, pH of 13 and 40 °C showed the most substantial chemical alteration, with major decreases in O-H stretching intensity and new bands related to phosphate ester formation. This indicates significant hydroxyl group consumption and crosslinking. The key modifications observed include a shift of O-H bands from 3290 cm^{-1} to 3287 cm^{-1}, confirming hydroxyl group replacement or loss [31]. Also, the appearance of P-O-C stretch at 976 cm^{-1} and P=O stretching vibration at 1256 cm^{-1} respectively confirms phosphorylation by STMP [32], (Figure 7).

3.7.2. Thermogravimetric analysis (TGA)

The thermal stability tests provided complementary evidence of chemical modification (Figures 8 and 9). The unmodified GA sample showed high initial weight loss (9.98 %) from

the two steps due to evaporation of moisture and volatile matter. Two major decomposition events can also be seen; one at low-to-mid temperatures resulting from dehydration or loss of volatile matter and a second at a higher temperature (220.0 °C and 439.2 °C) arising from the degradation of the polymer backbone. A low char/residual mass (15.5 %) reflected lower thermal stability [19]. The sample optimised for water absorption (2 % STMP concentration and pH of 11.5, 40 °C) displayed moderate stability with an initial weight loss of 8.47 %.

A two-step degradation from 296.0 °C to 363.5 °C was also observed, but with a higher degradation weight loss (87.91 %) and 3.62 % residual mass, confirming slight modification [19]. The sample optimised for viscosity (8 % STMP concentration, pH of 13 and 40 °C) showed the best performance with a minimal initial loss of 4.32 %. A single sharp decomposition event was observed between 250.0 °C and 473.0 °C with delayed onset at 319.4 °C. A lower degradation weight loss of 72.89 % and higher residual char of 23.47 % was also seen at 800.0 °C. This is because, as chemical modification increases, the peak decomposition temperatures and residual char also increase [33]. These are summarised in Table 12.

This single-step degradation pattern indicates a more uniform, crosslinked structure compared to the multi-step breakdown of unmodified GA and the sample optimised with 2% STMP concentration at a pH of 11.5 and 40 °C temperature [34], (Figures 8 and 9). GA sample modified at 8 % STMP concentration, pH of 13 at 40 °C, showed higher decomposition temperatures (Tp), enhanced crosslinking, stronger intermolecular forces, greater char yield with reduced moisture sensitivity, indicating enhanced thermal stability [19].

3.8. Summary of the responses studied

From results obtained from the physicochemical characterisation of modified and unmodified GA samples, the sample prepared with 8% STMP, at pH 13 and a temperature of 40 °C revealed a minimal initial weight loss which was < 5 % with a peak decomposition temperature of 382 °C while having the highest residual char (23.47 %) at 900 °C from its TGA, while its FTIR results revealed P-O-C stretch at 976 cm^{-1} and P=O stretching vibration at 1256 cm^{-1} respectively confirming phosphorylation by STMP. Also, its minimal water absorption and viscosity values (Table 13) proved that this sample appears to be the most chemically modified and was selected as the best out of the two responses studied.

4. Conclusion

The study demonstrated that chemical modification of GA via STMP crosslinking can be systematically tuned by adjusting concentration, temperature, and pH. Viscosity and water absorption responded differently to these factors, with temperature governing water interactions. The study observed that the degree of modification is strongly dependent on pH and STMP concentration, with high alkalinity (pH 13) and higher STMP concentration (8 %) being the basic requirement to optimise GA modification, yielding a material with reduced hydrophilicity since the modification process reduced the free hydroxyl groups and delayed thermal decomposition. Results from FTIR and TGA of the unmodified and modified samples show that uGA retained its natural structure but had high moisture content, stepwise degradation, and low stability. Unmodified GA had higher viscosity (6.25 cP), while the viscosity of mGA reached as low as 4.55 cP.

ANOVA showed that pH and temperature significantly influenced viscosity and STMP concentration had less direct effect. The factorial design successfully predicted GA behaviour. The model for viscosity showed a fit (R^2 = 0.7519). pH had the strongest statistical influence on modified GA properties. Sample modified at 2 % STMP concentration, pH of 11.5 and 40 °C exhibited slight modification, thermal performance and high weight loss. The sample prepared with 8 % STMP concentration, pH of 13, at 40 °C appears to be the most chemically modified and thermally stable, with its FTIR showing extensive hydroxyl substitution, phosphorylation, crosslinking, and superior thermal resistance.

Overall, chemical modification of GA biopolymer and the optimisation of the modification parameters result in tailoring its inherent properties for various applications such as admixtures in concrete technology, encapsulation in drug delivery, stabilisers in beverages, a food additive (E414), binding and suspending agents, printing and lithography, with GA being a sustainable and locally available raw material for these various applications.

Data availability

All data generated and analysed during this study are included in this manuscript. However, other raw data supporting the findings of this study are available from the corresponding author upon request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this manuscript.

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