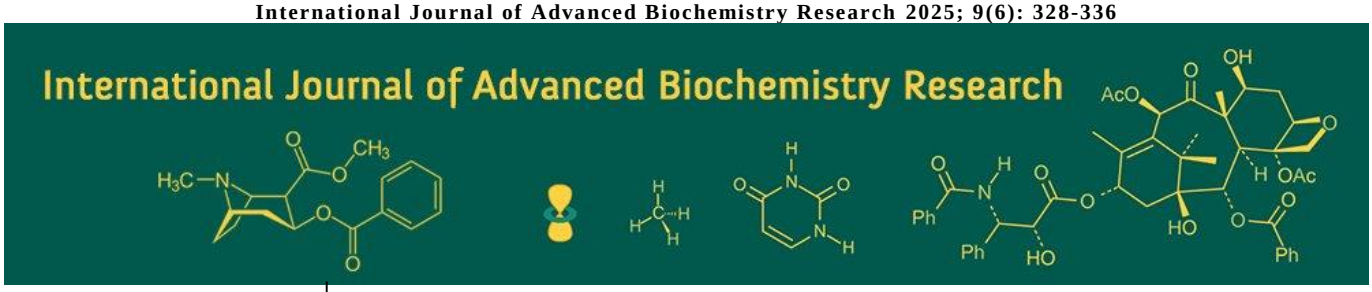

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Optimization of the process conditions for preparing methylcellulose from sugarcane bagasse using response surface methodology (RSM)

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Abstract
Methylcellulose is a cellulose ether derivative obtained through the chemical modification of cellulose via the methylation process. Sugarcane bagasse, a rich source of lignocellulosic biomass, provides an abundant supply of cellulose suitable for methylcellulose production. The properties of methylcellulose are significantly influenced by three key methylation process variables: NaOH concentration, methylation temperature, and the type of solvent used. In this study, Response Surface Methodology (RSM) was employed to investigate and optimize these process conditions for the preparation of methylcellulose from alpha-cellulose extracted from sugarcane bagasse. A Central Composite Design (CCD) was prepared to analyze the effects of independent variables, including, NaOH concentration (30%, 40%, and 50%), methylation temperature (40 °C, 50 °C, and 60 °C), and solvent type (acetone, toluene, and 2-propanol) on the response variables, namely, degree of substitution (DS), methoxy content (%), and solubility (%). All three factors had a statistically significant influence on the responses. The regression models exhibited a coefficient of determination (R^2) greater than 0.8, indicating a good fit for the experimental data. The optimal conditions identified through RSM were 40% NaOH concentration, a methylation temperature of 50 °C, and acetone as the solvent.

Keywords: Methylcellulose, sugarcane bagasse, response surface methodology, degree of substitution, solubility, methoxy content

Introduction
Methylcellulose (MC) is one of the most common and basic cellulose ether derivatives, in which methyl groups (-CH_3) substitute the hydroxyl groups at the C2, C3, and C6 positions of the anhydroglucose unit (AGU) of cellulose. MC belongs to a family of hydrocolloids and is a hydrophilic chemical compound obtained from cellulose. MC is produced through the chemical modification of alkali cellulose using etherifying agents. MC is non-digestible, non-toxic, and non-allergenic, making it suitable for use in food, pharmaceutical, and cosmetic applications. It is globally approved as a food additive [1]. The European Union designates it as E461, recognizing its role as an emulsifier and texturizer. Similarly, the Food Safety and Standards Authority of India (FSSAI) recognizes methylcellulose (INS 461) as a permitted food additive functioning as an emulsifier, stabilizer, and thickener in various food products. MC is primarily used as a food additive (E461) or in cosmetics and pharmaceuticals for its thickening and emulsifying properties. In the food industry, MC is mainly used as food additives in adhesives or thickening agents, viscosity control agents, binders, thickeners, and stabilizers, and also in the pharmaceutical industry as a matrix to cover the release of drugs as well as it is also used as protective coatings, toiletries and protection in paint formulations [2]. In recent years, the application of methylcellulose (MC) as a binder in plant-based burgers has gained significant attention [3]. Additionally, in products like mayonnaise and sauces, MC contributes to maintaining viscosity and enhancing emulsion stability. It is also used to stabilize foams in cold beverages, providing improved texture and mouthfeel [4].
To date, the industrial production of food-grade methylcellulose (MC) relies on high-purity cellulose, typically derived from wood pulp or cotton linters. These conventional sources are preferred due to their high cellulose content and low levels of impurities. However, with rising demand and growing concerns over the availability and sustainability of woody biomass, there is increasing interest in utilizing cellulose from non-woody biomass for MC synthesis [5].

Agricultural residues have emerged as promising alternatives due to their abundance, renewable and biodegradable nature, low toxicity, and their potential to enhance the value of agro-industrial by-products. The synthesis of MC from cellulose extracted from such agro-waste not only supports waste valorization but also offers a sustainable and cost-effective approach for the MC industry. This shift could reduce reliance on forest resources while promoting circular economy principles within agro-based sectors [6]. Several previous studies have reported that cellulose extracted from sugarcane bagasse [8] and rice husk [8] is suitable for the commercial preparation of methylcellulose. To achieve the desired quality of MC, it is crucial to optimize the conditions for cellulose chemical modification. Key parameters affecting MC quality include NaOH concentration during mercerization, the temperature of the chemical reaction, and the solvent used. Previous studies have investigated the impact of solvents [9] and etherification agents [10] on MC properties from woody biomass, as well as NaOH concentration effects on MC from pineapple leaves [2]. Response Surface Methodology (RSM), incorporating Central Composite Design (CCD), is effective for determining optimal MC preparation conditions [11]. However, there are no studies optimizing methylcellulose from sugarcane bagasse. This study will analyze the effects of NaOH concentration, methylation temperature, and solvent on MC properties, optimizing the preparation process using RSM.

2. Materials and methods

2.1 Sample preparation: Sugarcane bagasse was collected from a local sugar mill (Thanjavur, Tamil Nadu, India) and then dried to reduce the moisture content to below 10% on a dry basis. The dried biomass was pulverized, and the particles that passed through a 250-micron sieve were used for the study.

2.2 Alpha-cellulose extraction: The alpha-cellulose was extracted from the sugarcane bagasse as per the method described by Rasheed *et al.* [11] with some modifications. The dried biomass was pretreated with a 2N NaOH solution (1:10, w/v) for 4 hours at 80°C. After this, the mixture was autoclaved at 121±2°C for 1 hour under a pressure of 15 psi. The pre-treated biomass was then washed with distilled water and filtered, the remaining solid residue was mixed with 10% hydrogen peroxide (1:20, w/v). The pH of this mixture was adjusted to 4 using 10% acetic acid, and it was heated at 80°C for 4 hours. Following this, the mixture was filtered and washed with distilled water until to get the cellulose pulp with a pH of 7. The resulting cellulose was dried in a hot air oven at 50°C for 6 hours. After drying, it was pulverized and sieved. The cellulose fibers that passed through a 250-micron sieve were selected for further analysis.

2.3 Preparation of methylcellulose (MC)

MC was prepared from sugarcane bagasse cellulose according to the method described by Suhaimi *et al.* [2], with some modifications. The cellulose was mixed with a high concentration of NaOH (1:20, w/v) for 1 hour at room temperature (27±2℃). The mixture was then filtered, and the solid residue was combined with acetone (1:9, w/v). This mixture was heated at 50℃ for 1 hour, during which dimethyl sulfate (DMS) was added dropwise (1:3, w/v). This process was repeated twice, replacing the aqueous solution (acetone, DMS) with fresh materials each time. After 3 hours of processing, the mixture's pH was adjusted to neutral using 10% acetic acid, followed by washing and filtering. The final product of MC was dried at 50℃ and stored in high-density polyethylene bags for further use.

2.4 Experimental design for optimization of process conditions: A statistical response surface methodology (RSM) was employed using Design Expert Software (Stat-Ease Inc., version 13.0, USA) for the optimization of process conditions of chemical modification of cellulose. The three-factor three-level Central Composite Design (CCD) was carried out to optimize the process condition of chemical modification [12]. Two numerical factors and one categorical factor, and a Quadratic Model (QM) consisting of 39 experimental runs were employed, including 5 replicates at the center point. The design variables were the low and high levels of 3 principal factors of NaOH concentration (30%, 40%, and 50%), Temperature (40℃, 50℃, and 60℃), and Solvents (Acetone, Toluene, and 2-Propanol) while the response variables were the degree of substitution (DS), methoxy content (%), and solubility of MC in water (at 28±2℃). Coded and actual levels for the independent variables are shown in Table 1. At the center point, five experiments were conducted under identical conditions to calculate repeatability of the data (13). The regression equation was developed based on the design matrix.

Table 1: Coded and actual levels for the independent variables in the CCD.

Numerical factors					
	Coded levels				
Independent variables	-1.681	-1	0	+1	+1.681
NaOH concentration (%)	25	30	40	50	55
Temperature (℃)	35	40	50	60	65
Categorical factor					
Solvent	Acetone		Toluene	2-propanol	

The numerical optimization procedure was performed to obtain the desired property of MC. The maximum values of DS and methoxy content were determined as 2.2 and 33% based on the commercial requirements. The maximum range of solubility of MC in water was taken as the required criterion. The graphical technique utilized contour plots along with a 3D response surface to visualize the relationship between the response and the experimental levels of each variable. In this method, one variable was held constant at the center point while the other two variables were varied within their experimental ranges.

The adequacy of the developed models for response was verified through experiments conducted under the optimal conditions suggested by the software. The experimental and predicted values of the response were compared, and the error was calculated in order to check the validity of the predicted models [14].

2.5 Properties of MC

2.5.1 Degree of substitution (DS) and methoxy content of MC: The percentage of methoxy group (%OCH_3) in MC was calculated using the Zeisel-viebock-Schwappach method as mentioned in Ria *et al.* (15). The degree of substitution (DS) of MC was calculated using the following Eq. (1),

$$DS = \frac{1.62 \times \%OCH_3}{31 - (0.14 \times \%OCH_3)}$$

Here, %OCH_3 represents the percentage of methoxy groups in MC.

2.5.2 Solubility

The solubility of the samples was analysed in inorganic solvent (water) as described by Ria *et al.* [15]. Two grams of MC samples were added with 20 mL of solvents, stirred continuously, and kept at room temperature (28±2 ℃) for 8 h. The mixture was filtered, and the filtrate was dried at 105 °C for 6 h. Based on the weight loss, the percentage solubility of the MC samples was calculated.

2.6 Statistics

The experimental data obtained from the CCD were analysed by the second-order regression polynomial equation to describe the relationship between the predicted response variable and the independent variables. The generalized second-order polynomial model proposed for the optimization of process conditions is shown in Eq. (2).

$$Y_i = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{11} {x_1}^2 + \beta_{22} {x_2}^2 + \beta_{33} {x_3}^2 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_2 + \beta_{23} x_2 x_3$$

Where Y_i is the desired response, β_0 is the constant term, β_1, β_2, and β_3 are the regression coefficients for linear effect terms, β_{11}, β_{22}, and β_{33} are quadratic effects, and β_{12}, β_{13}, and β_{23} are interaction effects. In this model, x_1, x_2, and x_3 are the process variables like NaOH concentration (%), temperature (℃), and solvent, respectively.

The analysis of variance (ANOVA) analysis of the experimental runs provided the regression coefficients for linear, quadratic, and interaction terms, individually were carried out individually. The significance of each term for the response was also evaluated by observing the F value, where the probability (p) is less than 0.05 (significant) or 0.01 (highly significant). The adequacy of the model was determined using model analysis and coefficient of determination (R^2) analysis. The experimental design matrix, data analysis, and optimization procedure were carried out using the software Design Expert Version 13.0.0. The mathematical model was considered satisfactory only when the ANOVA data exhibited a high level of statistical significance.

3. Results and discussion

The degree of substitution (DS) and overall quality of methylcellulose (MC) are significantly influenced by the conditions applied during the mercerization and chemical modification steps. To systematically investigate these effects, cellulose extracted from sugarcane bagasse was selected as a representative sample. Three different levels of process variables were considered: NaOH concentration during mercerization (30%, 40%, and 50%), Reaction temperature during chemical modification (40℃, 50℃, and 60℃), and Type of organic solvent used in the methylation step (Acetone, Toluene, and isopropanol). These variables are known to influence cellulose swelling behavior, the accessibility of hydroxyl groups, and the reaction kinetics, thereby affecting the efficiency of the etherification reaction and the resulting MC properties [16]. The mercerization process was optimized using response surface methodology (RSM) with a central composite design (CCD). The degree of substitution (DS), methoxy content (%), and water solubility (%) of the synthesized MC were selected as the response variables for model development and analysis.

3.1 Optimization of the process condition of methylation

The experimental results associated with the interaction between independent variables, experimental values, and predicted responses are shown in Table 2. The experimental results showed that: DS values ranged from 0.18 to 0.72, Methoxy content ranged from 3.39% to 12.89%, and Solubility ranged from 11.72% to 31.79% (Table 2).

The quadratic model created by RSM to predict the response is shown in Eqs. (3), (4), and (5). The impact of each component on the characteristics of MC is displayed by the linear coefficient for the reaction variables of NaOH concentration, temperature, and solvent. Conversely, the interaction effects on the responses are represented by the coefficient, which is the product of two components. The quadratic effect is represented by second-order terms. An antagonistic effect is shown by a negative sign, while a synergistic effect is indicated by a positive sign [17].

DS = +0.63 + 0.021NaOH concentration + 0.036Temperature + 0.037Acetone - 0.00058Toluene - 0.067NaOH concentration^2 - 0.13Temperature^2 + 0.016NaOH concentrationTemperature - 0.017NaOH concentrationAcetone + 0.022NaOH concentrationToluene - 0.036temperatureAcetone - 0.00822TemperatureToluene	…(3)
Methoxy content (%) = +11.51 + 0.36NaOH concentration + 0.65Temperature + 0.63Acetone - 0.00053Toluene - 1.16NaOH concentration^2 - 2.13Temperature^2 + 0.28NaOH concentrationTemperature - 0.30NaOH concentrationAcetone + 0.39NaOH concentrationToluene - 0.65temperatureAcetone - 0.15TemperatureToluene	…(4)
Solubility (%) = +23.79 + 0.48NaOH concentration + 0.55Temperature + 6.53Acetone - 1.89Toluene - 0.83NaOH concentration^2 - 1.69Temperature^2 + 0.29NaOH concentrationTemperature - 0.34NaOH concentrationAcetone + 0.18NaOH concentrationToluene - 1.52TemperatureAcetone + 0.58TemperatureToluene	…(5)

Table 2: Quadratic model design for chemical modification of cellulose, and experimental values of response variables.

Run	Model parameters			Experimental			Predicted		
	NaOH con. (%)	Temperature (℃)	Solvent	DS	Methoxy content (%)	Solubility (%)	DS	Methoxy content (%)	Solubility (%)
1	40	50	2-Propanol	0.57	10.40	19.07	0.60	10.88	19.14
2	40	50	Toluene	0.62	11.26	20.51	0.63	11.51	21.90
3	25	50	2-Propanol	0.41	7.54	15.65	0.44	8.18	16.58
4	40	50	Acetone	0.72	12.89	31.79	0.67	12.15	30.32
5	40	65	2-Propanol	0.48	8.82	18.99	0.45	8.30	17.88
6	50	60	Toluene	0.55	10.05	23.09	0.52	9.57	21.45
7	40	50	2-Propanol	0.57	10.40	19.07	0.60	10.88	19.14
8	40	50	Acetone	0.72	12.89	31.79	0.67	12.15	30.32
9	55	50	Toluene	0.51	9.35	20.17	0.56	10.25	21.17
10	40	50	Toluene	0.62	11.26	20.51	0.63	11.51	21.90
11	25	50	Toluene	0.41	7.58	19.59	0.44	8.12	19.31
12	40	65	Toluene	0.43	7.93	21.00	0.41	7.59	20.11
13	40	50	Toluene	0.62	11.26	20.51	0.63	11.51	21.90
14	40	50	2-Propanol	0.57	10.40	19.07	0.60	10.88	19.14
15	30	40	Toluene	0.48	8.82	20.24	0.38	7.05	17.87
16	40	50	Acetone	0.72	12.89	31.79	0.67	12.15	30.32
17	30	60	2-Propanol	0.51	9.35	16.99	0.45	8.30	17.19
18	40	35	2-Propanol	0.18	3.39	11.72	0.22	4.21	13.65
19	50	40	Acetone	0.47	8.64	28.53	0.46	8.45	28.63
20	40	50	Toluene	0.62	11.26	20.51	0.63	11.51	21.90
21	50	60	2-Propanol	0.50	9.17	18.82	0.51	9.40	19.04
22	30	60	Acetone	0.37	6.86	24.93	0.45	8.33	26.40
23	50	40	2-Propanol	0.41	7.57	17.69	0.32	5.95	15.48
24	50	60	Acetone	0.41	7.58	25.43	0.49	9.02	27.26
25	40	50	Acetone	0.72	12.89	31.79	0.67	12.15	30.32
26	40	50	Acetone	0.72	12.89	31.79	0.67	12.15	30.32
27	50	40	Toluene	0.54	9.87	22.01	0.43	8.00	18.62
28	25	50	Acetone	0.47	8.64	27.09	0.53	9.73	28.46
29	40	35	Acetone	0.27	5.05	25.16	0.41	7.51	28.31
30	40	50	2-Propanol	0.57	10.40	19.07	0.60	10.88	19.14
31	40	65	Acetone	0.46	8.47	25.43	0.41	7.53	25.57
32	40	50	2-Propanol	0.57	10.40	19.07	0.60	10.88	19.14
33	40	50	Toluene	0.62	11.26	20.51	0.63	11.51	21.90
34	30	40	Acetone	0.58	10.57	29.95	0.49	8.88	28.91
35	30	60	Toluene	0.41	7.58	20.39	0.41	7.51	19.56
36	30	40	2-Propanol	0.38	7.04	16.44	0.32	5.97	14.78
37	55	50	2-Propanol	0.48	8.82	17.07	0.49	8.94	18.39
38	40	35	Toluene	0.23	4.32	15.48	0.33	6.16	16.92
39	55	50	Acetone	0.54	9.87	28.56	0.54	9.92	28.87

The significance of linear, interaction, and quadratic model terms was analyzed using an F-test and a t-test, and the results are presented in Table 3. Based on the statistical analysis, P-values < 0.05 indicate that the corresponding model terms are statistically significant, while P-values < 0.01 denote highly significant effects. Notably, the models for DS, methoxy content, and solubility were found to be highly significant, each with a P-value < 0.0001, confirming the robustness and reliability of the models in predicting MC quality attributes (Table 3). The quadratic relationship between temperature and NaOH concentration had a significant impact on the DS and methoxy content. DS and methoxy content of MC are unaffected by the interactive phrases. The solvent's linear term and the temperature's quadratic effect have a significant impact on solubility. Solubility is significantly impacted by temperature and the solvent interaction term.

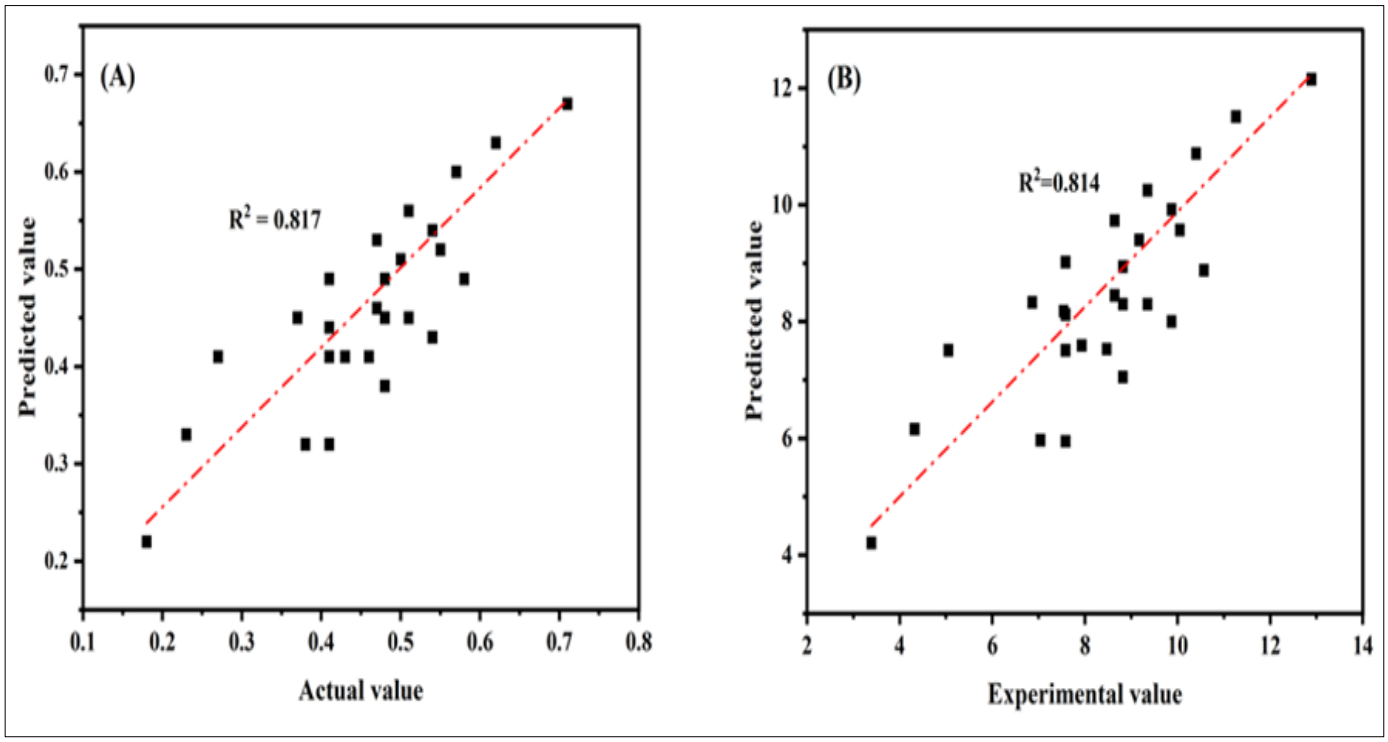
ANOVA was used to assess the accuracy of the resulting model and confirm the statistical significance of the mean square variation to the mean square residual error ratio. The statistical parameters of regression models like standard deviation (SD, %), R^2 values, coefficient of variation (CV), and adequate precision are shown in Table 3. In all response values, the experimental R^2 values were in reasonable agreement with the adjusted R^2. The R^2 values of all models are above 0.80, which ensures satisfactory fitness of the regression models. The model's repeatability is further demonstrated by the low SD value of the models. Adequate precision of more than 4 is desired for the model fitting. In this study, all three quadratic models exhibit more than 4 adequate precisions. The ANOVA results indicate that the process conditions of methylation significantly affect the property of MC.

The predicted values of responses obtained by the empirical model and experimental responses are plotted linearly, as shown in Figure 1. As can be observed from the three plots, the predicted values for DS, methoxy content, and solubility are fairly close to their actual/experimental values. In that, the actual value of solubility is very close to the predicted value, which was explained by the high R^2 value of 0.92. Whereas the DS and methoxy content quadratic models developed by RSM exhibit the R^2 value of 0.817 and 0.814, respectively.

Table 3: Analysis of variance (ANOVA) for response surface quadratic model.

Source	DS			Methoxy content			Solubility		
	F value	**P value**	**Significance**	**F value**	**P value**	**Significance**	**F value**	**P value**	**Significance**
Model	10.86	< 0.0001		10.72	< 0.0001		30.98	< 0.0001	
A	2.30	0.141		2.30	0.1413		1.90	0.1792	
B	7.11	0.013		7.42	0.0112		2.51	0.1246	
C	3.93	0.032		3.76	0.0363		151.36	< 0.0001	
A^2	21.46	< 0.0001		20.43	0.0001		4.92	0.0352	
B^2	81.79	< 0.0001		80.98	< 0.0001		20.46	0.0001	
AB	0.68	0.415		0.68	0.4182		0.34	0.5663	
AC	0.73	0.490		0.72	0.4954		0.23	0.7942	
BC	3.06	0.063		3.10	0.0615		4.85	0.0159	
Lack of fit	46.96	<0.0001		46.18	<0.0001		11.58	0.0002	
Statistical parameters									
	DS			Methoxy content			Solubility		
SD (%)	0.066			1.18			1.71		
R^2	0.815			0.813			0.927		
Adj R^2	0.740			0.737			0.897		
Predicted R^2	0.479			0.471			0.810		
Mean	0.51			9.37			22.24		
CV	12.92			12.54			7.68		
Adeq. precision	12.217			12.810			17.610		

Significant (P<0.05). Highly significant (P<0.01). SD: Standard deviation (%), CV: Coefficient of variation.



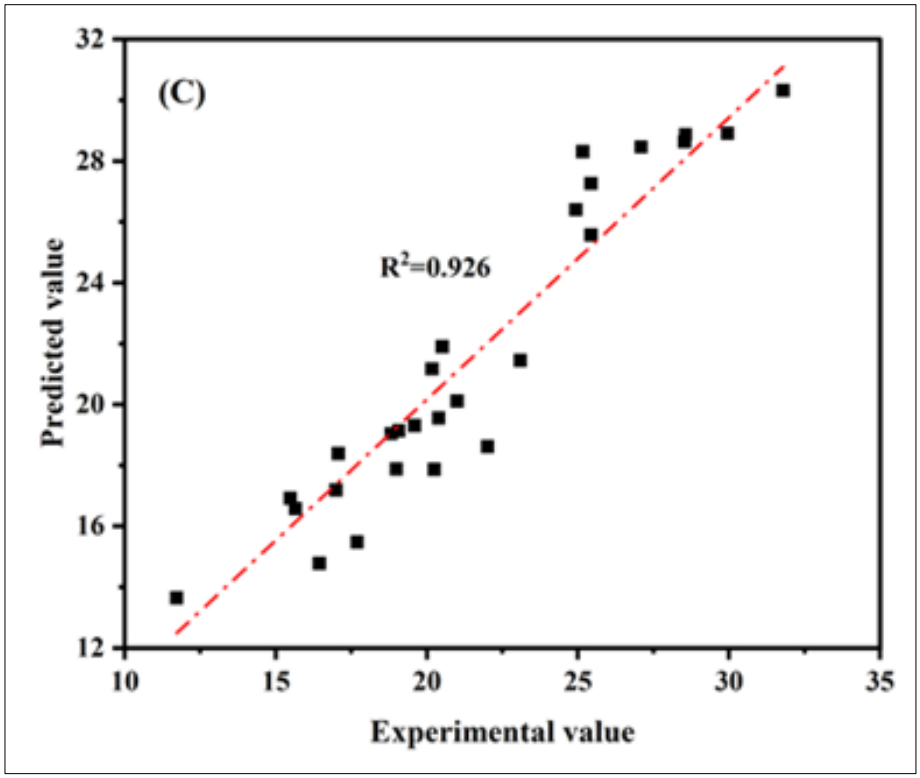


Fig 1: Predicted versus actual (A) Degree of substitution (DS), (B) Methoxy content (%), and (C) Solubility (%) of methylcellulose (MC).

3.2 3D surface contour plots to analyze the relation between the independent and response variable

The combined effect of NaOH concentration and reaction temperature on the degree of substitution (DS), methoxy content, and solubility of methylcellulose (MC), while the type of organic solvent (acetone, 2-propanol, and toluene) was held constant at the center point of the experimental design. The results demonstrate a synergistic interaction between NaOH concentration and temperature, which significantly influences all three response variables. However, between NaOH concentrations of 35% to 45% and temperatures of 45 °C to 55 °C, DS does not significantly change while using solvents like acetone and toluene. The results suggest that acetone and toluene facilitate a more stable and efficient etherification under moderate alkaline and thermal conditions. In contrast, with 2-propanol, the highest DS is observed at the center point of the other two independent variables (Figure 2).

Acetone reacts with the cellulose's original structure by modifying hydrogen bonds between the chains, which helps to reduce the amount of inter-chain hydrogen bonds and the formation of segment and inter-segment packing during methylation [18]. Meanwhile, toluene reacts with methylated cellulose by modifying the interaction between the hydrophobic blocks formed during the methylation. The main function of toluene is to avoid the hydrophobic interaction between the methylated blocks, so the addition of toluene at the beginning of the methylation is not significant due to its little interaction with the hydroxyl groups [19]. This might be a reason for the low DS in the MC prepared using the solvent toluene. Both acetone and toluene prevent gel formation during methylation [18]. The usage of the solvent isopropanol has the ability to form hydrogen bonds with the hydroxyl groups of cellulose, which may hinder the methylation and result in a low DS [20]. The swelling behavior of cellulose in the organic solvent also affects the accessibility of the hydroxyl groups during methylation. The swelling of cellulose in isopropanol solvent is much lower compared to toluene and acetone. Acetone can provide high swelling. Similar to these findings, several previous studies reported that acetone provides a high DS of MC prepared from corn cob cellulose (DS = 1.14) (18) and rice husk cellulose (DS = 0.86) [8]. Ke *et al.* [21] reported the MC from cotton linters using NaOH/urea aqueous solution as a methylation solvent. The prepared MC provided a high DS value of 1.20 and had better solubility in water and organic solvents. The methoxy content represents the percentage of the cellulose molecule substituted with a methoxy group ($-OCH_3$) during methylation. The methoxy content depends on the DS and affects the solubility of MC. The percentage of methoxy groups in MC facilitates the hydrophobic interaction between molecules, which highly affects the solubility behavior of MC [5]. The hydrophobic interaction during methylation can be favored by high temperature [22].

For methoxy content and solubility, a notable decline is observed when using acetone as a solvent at NaOH concentrations above 45% and temperatures above 55 °C. This sharper decline, compared to toluene and 2-propanol, can be attributed to acetone's partial miscibility with water and its higher polarity, which may disrupt the reaction environment at elevated temperatures and alkaline conditions. This can lead to degradation of the cellulose backbone or reduced availability of reactive sites for methylation, thereby lowering the methoxy content and ultimately reducing solubility. Results denote that the substitution of the methoxy group during methylation is facilitated in the temperature range of 40 °C to 55 °C, but not above 55 °C. The usage of NaOH concentration above 45% can provide better swelling of cellulose; however, during methylation, it leads to the formation of a gel-like structure and hinders the methylation process. In previous studies, a

higher yield of MC was also reported at 40% NaOH concentration compared to 50% [2].

The solubility of the MC depends on the DS and methoxy content; the MC polymer with DS between 1.4 and 2.2 may have better solubility in water, which may be used to prepare the solutions in low concentration [18]. The solubility of MC further depends on how uniformly the methoxy groups are distributed along the polymeric chain [19]. Results indicate that the NaOH concentration during methylation, methylation temperature, and selection of solvent for the chemical reaction significantly affect the property of MC.

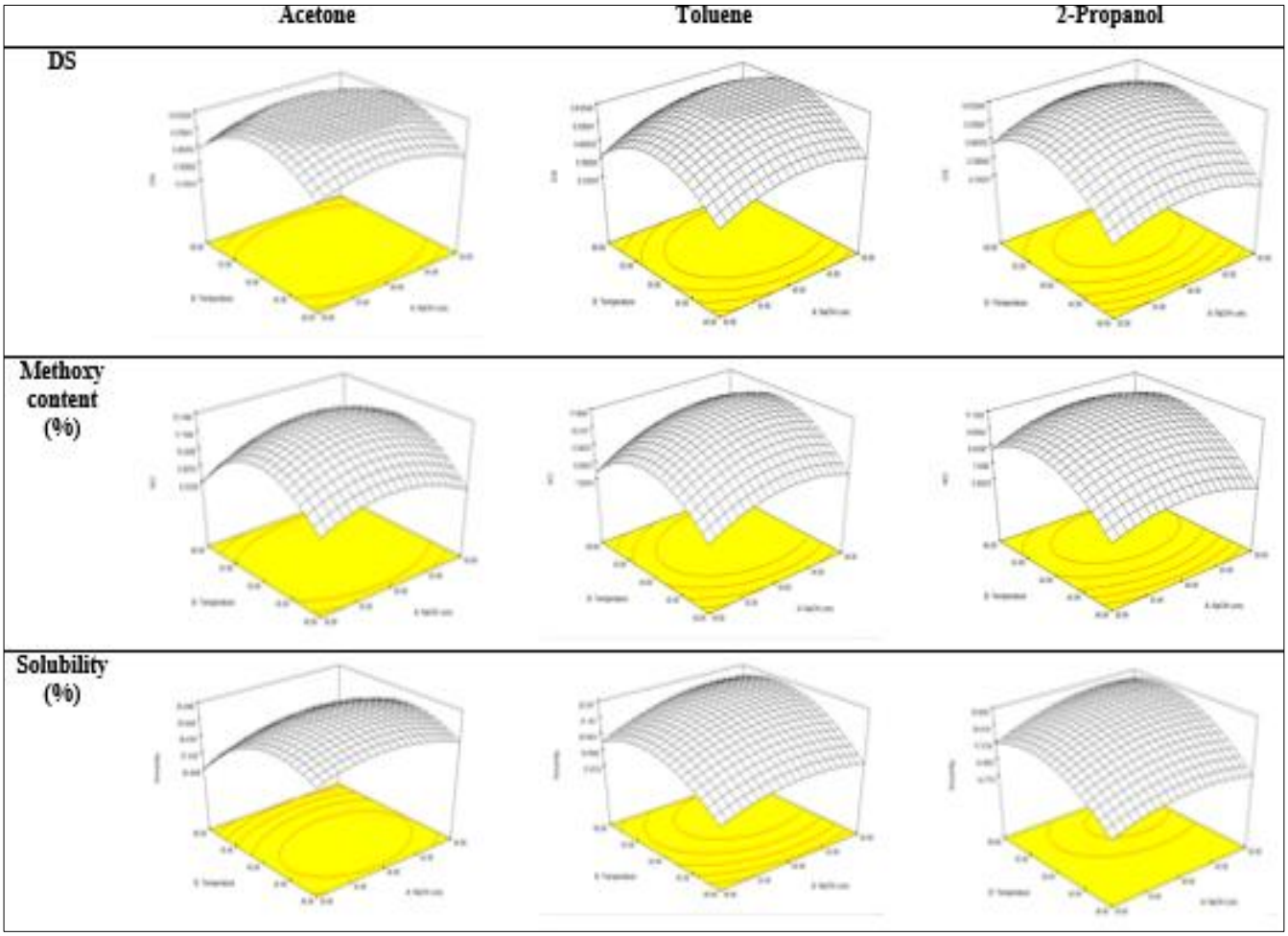


Fig 2: A response 3D surface plot illustrates the interaction effects of various process variables on the different response variables (Choice of solvent is kept at the center point for each plot).

3.3 Optimization of process variables in the preparation of MC: The quality and commercial application of methylcellulose is determined by its properties, like DS, methoxy content (%), and solubility (%). In the experimental design matrix, a strong positive correlation was observed between DS and both methoxy content and solubility, indicating that an increase in DS enhances the methoxy groups, which in turn improves solubility. This is consistent with the known structure-property relationship of MC, where a higher DS typically indicates more methoxy substitution on the cellulose backbone, reducing hydrogen bonding among cellulose chains and increasing hydrophilicity.

To achieve an optimal balance among these response variables, numerical optimization was performed using the desirability function in the RSM. For this, the target criteria were set as: DS in the range of 2.2, methoxy content in the range of 33, and maximum value of solubility. These values were chosen based on the desired performance characteristics of MC for industrial applications such as film formation, thickening, and emulsification, where high water solubility and sufficient substitution levels are critical. Meanwhile, the value of independent variables is set in the ranges being studied. From RSM, the optimized condition obtained to prepare the enhanced property of MC is 40% NaOH concentration, 50℃ of methylation temperature, and the usage of acetone.

3.4 Model verification

To assess the reliability of the developed model in predicting the optimal response values, methylcellulose (MC) samples were synthesized using the process parameters identified as optimal. The experimental outcomes were then compared with the corresponding values predicted by the regression model. The percentage deviations between the actual and predicted results are summarized in Table 4. Under these optimized conditions, the synthesized MC exhibited a degree of substitution (DS) of 0.715, a methoxy content of 12.89%, and a water solubility of 31.78%. The close agreement between the predicted and experimental data, with only minor error, validates the effectiveness and predictive accuracy of the optimization model.

Table 4: Optimized process conditions for the preparation of MC.

NaOH concentration (%)	Temperature (℃)	Solvent		Predicted	Experimental	Error
40	50	Acetone	DS	0.67	0.715	0.07
			Methoxy content (%)	12.15	12.89	1.24
			Solubility (%)	30.32	31.78	1.80

4. Conclusion

The study aimed to optimize the process conditions for the methylation of cellulose derived from sugarcane bagasse to produce methylcellulose (MC). Three independent variables were considered for optimization: NaOH concentration, methylation temperature, and the type of solvent used in the methylation process. Using Response Surface Methodology (RSM), the optimal conditions were determined to be 40% NaOH concentration, a methylation temperature of 50 °C, and acetone as the solvent. The close agreement between the predicted and experimental values indicates that the RSM regression models are reliable for predicting MC properties under varying process conditions. Under optimal conditions, the resulting MC exhibited a degree of substitution (DS) of 0.72, a methoxy content of 12.89%, and a solubility of 31.8%. These results confirm the feasibility of producing MC from sugarcane bagasse.

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