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Research Article

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# Optimization of cellulose yield from oil palm trunks with deep eutectic solvents using response surface methodology

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**Abstract:** Oil palm trunk (OPT) is widely studied as an underutilized material with large cellulose and lignin contents. As an environmentally friendly solvent, deep eutectic solvents (DESs) can be employed to extract cellulose. The main objective of this study was to evaluate both basic and acidic DES and to optimize cellulose yield from OPT extraction. The type of DES used were choline chloride (ChCl)–urea, ChCl–lactic acid, ChCl–levulinic acid, and ChCl–glycerol. For the optimization, response surface methodology with three-level factorial Box–Behnken design was used with mass ratio solid/liquid in the range of 5–10 g sample/g DES, temperature between 80 and 110°C, and extraction time in the range of 1–4 h. The results showed that the optimal condition for ChCl–levulinic acid was a mass ratio of 5.05%, a temperature of 104.2°C, and a reaction time of 3.76 h with a cellulose yield of 91.29%. It is shown that acidic DESs produce a higher cellulose yield than basic DES. This experiment also offers an important understanding of fractionation and optimization to improve OPT utilization with green solvents.

**Keywords:** cellulose, deep eutectic solvent, green processing, response surface methodology

## 1 Introduction

Cellulose is the most common renewable polymer, with a yield of about 75 billion tons per year, and accounts for 35–50% of the total natural fibre composition, making it a highly sustainable resource [1]. Cellulose can be sourced from various natural materials, such as grass, agricultural waste (e.g., wheat straw, rice husk, corn cob, banana peel, sugarcane bagasse), and plantation waste (e.g., poplar, bamboo, oil palm kernel shell [OPKS], oil palm trunk [OPT]), reducing dependence on fossil-based materials. Cellulose fibres are widely employed as a material for textiles, paper, packaging, biocomposites, and others because of their unique qualities, non-toxicity, biocompatibility, and biodegradability [2]. Cellulose has also been utilized as a reinforcing agent with favourable outcomes across various sustainable sectors, including pharmaceutical, medicine, packaging, environmental, energy, and electronic applications [3].

There are many obstacles to extracting cellulose from its source, including the complex structure of lignocellulosic biomass due to cross-linking between lignin and polysaccharides [4]. Lignin acts as a binder for hemicellulose and cellulose through strong covalent bonds [5]. These bonds keep the plant sturdy but pose a challenge in cellulose utilization. Several methods are used to remove lignin in lignocellulosic materials, including biological, physical, chemical, and physicochemical treatments.

There is a need to select an extraction process that is efficient, environmentally sustainable, and cost-effective. Most conventional solvents are organic, volatile, flammable, and typically toxic, such as benzene, chloroform, diethyl ether, and dichloromethane [6], with the potential to be persistent in the environment. Therefore, such liquids must be replaced with green alternatives. Over the past two decades, deep eutectic solvents (DESs) have been explored and promoted as green solvents. DES was developed as a renewable and low-cost component to address concerns about biodegradability, biocompatibility,

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and the cost of ionic liquid solvents [7]. DES consists of two mixture components, one acting as a hydrogen bond donor (HBD) and the other acting as a hydrogen bond acceptor (HBA). In several studies, this solvent has been shown to have considerable efficiency in cellulose extraction through the delignification process [2,8,9].

Viscosity and pH are critical for the performance of recently developed technologies such as biomass pre-treatment, extraction, and solubility improvement [10]. Beeler et al. [11] investigated the types of DES, classified into strong acid, strong base, weak acid, and weak base, to remove lignin and hemicellulose from grapevine agricultural by-products. The results showed that DES of strong acid (choline chloride [ChCl]–lactic acid) and strong base (potassium carbonate–ethylene glycol) had the highest lignin removal efficiency. Lignin removal was correlated to the amount of cellulose yield in the solids. Strongly acidic and basic DES have a superior ability to increase cellulose yield and dissolve lignin.

Extraction variable conditions are crucial in the extraction process. During the extraction process, cellulose yield is affected by the ratio of biomass material to DES as it relates to the contact surface area between solids and solvent. Inadequate solvent volume might lead to inadequate extraction, whereas excessive volume may result in waste and complicate the separation process [12]. The extraction temperature influences cellulose yield by altering molecule kinetic energy and diffusion rate [13]. Extraction time influences the extraction process because a short time may result in an inadequate reaction and decrease process efficiency. Conversely, extending the extraction time can cause oxidative degradation and increase the cost of the process. Therefore, process variables such as the mass ratio of biomass sample to solvent, extraction temperature, and extraction time are important to obtain optimum cellulose yield. Those parameters are dependent on the type of biomass and solvent.

As a source of biomass products with high cellulose content, OPT was selected as a potential biomass for the lignocellulosic biomass extraction process. The amount of OPT cellulose is 39.4%, which is higher than other palm oil wastes such as OPKS, oil palm decanter cake, oil palm fibre, empty fruit bunches, and oil palm sewage sludge [14]. OPTs exhibit lower density and dimensional stability relative to other components of the oil palm, resulting in limited utilization [15]. Whereas if properly and appropriately utilized, old felled OPTs can be a significant source of biomass [16]. OPT waste increased from 13.9 to 59.7 Mt/year in 2020 [17]. Oil palm plantations are one of the largest and abundant plantations in Indonesia and tropical and subtropical regions in Southeast Asia. Response surface methodology (RSM) with Box–Behnken design (BBD) is a methodology used to optimize cellulose yield from OPT and is effective for analysing the relationship between a response and process factors [11,18,19]. Through effective experimental design, RSM provides a wealth of information and can be used for process optimization in various situations [20]. Analysis of variance (ANOVA) is used to identify input variables that significantly affect a specific response.

Various literature investigated the optimization of biomass pre-treatment using DES [21–24]. However, few studies have focused on agricultural or plantation residue biomass with acid-based DES. Rodrigues et al. [25] presented a study on corn cobs pre-treatment for optimizing lignin extraction, cellulose to glucose conversion, and hemicellulose removal with ChCl/lactic acid. Teo et al. [26] conducted a cellulose yield optimization study of raw oil palm leaves using ChCl–lactic acid. An optimization study of OPTs with the selection of several types of DES has not been reported to the best of our knowledge.

Environmentally friendly, efficient, sustainable, and cost-effective strategies to extract cellulose are promising research areas nowadays. New solvents, innovative methodologies, and optimization are the most expected goals. With these prospects in mind, this study was planned. The objective of this research was to explore cellulose extraction from OPT using four types of acidic and basic DES, followed by optimizing the extraction operating conditions by RSM. It was assumed that the efficient strategy could potentially be applicable on a commercial scale.

## 2 Materials and methods

### 2.1 Materials

OPT was sourced from South Sumatra (Indonesia) oil palm plantations. OPT was reduced in size to 120 mesh, followed by washing and drying at 105°C until moisture content <10%. OPT powder was stored at room temperature for further use. Choline chloride ($C_5H_{14}ClNO$) > 99.0% purity was obtained from Himedia (India). Urea ($CO(NH_2)_2$) ≥ 98% purity, lactic acid ($C_3H_6O_3$) 85% purity, levulinic acid ($C_5H_8O_3$) 98% purity, and glycerol ($C_3H_8O_3$) 85% purity were obtained from Sigma–Aldrich (China).

### 2.2 DESs preparation

DESs were synthesized with a fixed molar ratio (HBA:HBD = 1:2). ChCl as HBA was combined with four types of HBDs (urea, lactic acid, levulinic acid, and glycerol). The mixture of HBA and HBD was put into a 250 mL flask, heated at 80°C, and stirred for 2 h until a stable liquid was obtained characterized by a clear and transparent colour. The result was that colourless and homogenous liquids were referred to as DES, and to preserve them for later use, they were stored in a vacuum desiccator with silica gel. If no precipitate formed after 24 h of cooling at room temperature, the DES preparation was successful.

## 2.3 Extraction of lignocellulose

In a three-neck flask, OPT was extracted using DES with mass ratio (5–10 g sample/g DES), temperature (80–110°C), and time (1–4 h) variations. The mixture was separated using vacuum filtration and washed to remove the remaining DES. The solid phase was homogenized by ultrasonication (600 W, 40 kHz, 15 min), then centrifuged (5,500 rpm, 20 min), and freeze-dried (24 h) before being used for proximate analysis.

## 2.4 Determination of hemicellulose, cellulose, and lignin

The procedure analysis was conducted to calculate hemicellulose, cellulose, and lignin content using the Chesson-Datta method [27]. 1 g OPT ($w_1$) and 150 mL $H_2O$ were heated at 100°C for 1 h in the water bath. The solution was filtered, and the residue was washed with heated water and dried until the constant weight ($w_2$). $H_2SO_4$ 1 N was added to the residue and refluxed at 100°C for 1 h. The residue was dried following filtration and washing to neutrality, and the constant weight was recorded ($w_3$). The residue was later soaked with $H_2SO_4$ 72% at ambient temperature for 4 h. Subsequently, 150 mL of $H_2SO_4$ 1 N was added to the solution and heated at 100°C for 1 h. After filtration, the final residue was neutralized with 400 mL of water until it reached a neutral pH. The residue was heated at 105°C in the oven until its weight stabilized ($w_4$). The residual solid was then transformed into ash ($w_5$) after being heated at 600°C in the furnace. The calculation of chemical composition, cellulose yield, and percentage of delignification were shown as follows:

$$\text{Hemicellulose (\%)} = \left(\frac{w_2 - w_3}{w_1}\right) \times 100\% \tag{1}$$

$$\text{Cellulose (\%)} = \left(\frac{w_3 - w_4}{w_1}\right) \times 100\% \tag{2}$$

$$\text{Lignin (\%)} = \left(\frac{w_4 - w_5}{w_1}\right) \times 100\% \tag{3}$$

$$\text{Ash (\%)} = \left(\frac{w_5}{w_1}\right) \times 100\% \tag{4}$$

$$\text{Cellulose yield (\%)} = \left(\frac{C_r}{C_u}\right) \times 100\% \tag{5}$$

$$\text{Delignification (\%)} = \left(\frac{L_u - L_t}{L_u}\right) \times 100\% \tag{6}$$

where $C_r$ is the cellulose recovered (g), $C_u$ = cellulose content of untreated OPT (g), $L_u$ = lignin content of untreated OPT (g), and $L_t$ = lignin content of treated OPT (g).

## 2.5 Design of experiment using BBD–RSM

Based on the DES screening experiment, the selected DES was used for cellulose yield optimization in BBD–RSM by adjusting the mass ratio (5–10%), temperature (80–110°C), and time (2–6 h). BBD was used due to its efficient and reliable method utilizing three levels (−1, 0, +1), allowing for investigating interaction effects among selected parameters [28]. Experimental runs of 15 (12 middle nodes and 3 central nodes) with different sets of independent parameters (three components and three levels). Cellulose yield was used as the response value, and the extraction process was optimized with Design Expert 13.0. To analyse the significance of each factor and its interaction, a one-way ANOVA with a 95% confidence interval was used.

The experimental results and the model were fitted with the coefficient of determination ($R^2$). The correlation between the independent parameters and the response variable in the experimental results can be stated through a second-order polynomial equation (Eq. 7)

$$\begin{aligned} \text{CY} = \beta_0 \; - \; \beta_1 A + \beta_2 B + \beta_3 C + \beta_{12} AC + \beta_{23} BC + \beta_{13} AC \\ + \beta_{11} A^2 + \beta_{22} B^2 + \beta_{33} C^2 \end{aligned} \tag{7}$$

where CY is the response variable (cellulose yield); $A$, $B$, and $C$ are the independent variable; $AB$, $BC$, and $AC$ are the parameter interaction; $A^2$, $B^2$, and $C^2$ are quadratic independent parameters; $\beta_0$ is a constant term; and $\beta_1$, $\beta_2$, $\beta_3$, $\beta_{11}$, $\beta_{23}$, $\beta_{13}$, $\beta_{11}$, $\beta_{12}$, $\beta_{13}$ are the regression coefficients.

## 2.6 Lignocellulose characterization

### 2.6.1 Fourier transform infrared spectroscopy (FTIR)

A Thermo Nicolet iS10 infrared spectrophotometer with a diamond Attenuated Total Reflectance was used to record

FTIR spectra that can analyse various functional groups in samples. The investigations were carried out in transmittance mode with 32 scans for each sample in the 500–4,000 $cm^{-1}$ range.

### 2.6.2 Thermogravimetric analysis (TGA)

NETZSCH STA 449F3 was used in an inert atmosphere to determine the thermal stability of OPT and treated samples. A 10 mg sample was heated from room temperature to 800°C, with a flow of nitrogen 20 $mL\cdot min^{-1}$ and a heating rate of 10°C·$min^{-1}$. The mass change was recorded during each run.

## 3 Results and discussion

### 3.1 Physical and chemical content of OPT

The proximate analysis of OPT indicated 7.08% moisture, 1.80% ash, 11.23% fixed carbon, and 76.45% of volatile matter. The ash, fixed carbon, and volatile matter were assessed on an oven-dried basis. OPT was analysed for chemical composition and found hemicellulose 28.84% ± 0.17%, cellulose 35.55% ± 0.02%, and lignin 26.73% ± 0.05%.

### 3.2 Screening DES for OPT delignification

Four different DES were used to evaluate the cellulose yield and delignification of OPT extraction. Parameters such as DES ratio, temperature, extraction time, solid/liquid mass ratio, and stirring speed were kept constant. The DES preparation ratio is 1:2, making it a popular choice for various applications, including biocatalysis and extraction processes [29–32]. The temperature used in this study ranges from 80 to 110°C to save energy and avoid potential cellulose degradation with various DES [33,34]. The highest cellulose yield from the four DESs was achieved in the following order: ChCl–levulinic acid, ChCl–lactic acid, ChCl–urea, and ChCl–glycerol (Figure 1).

Strongly acidic DES have higher performance than weakly acidic DES due to their high conductivity, low pH, and lower viscosity [35]. Higher acidity (ChCl–levulinic acid) results in more efficient cleavage of ester and ether bonds in biomass cell walls, allowing lignin and xylan to be broken down. Acidic DES with carboxylic groups is an effective compound in dissolving lignin and hemicellulose (53–86%), higher than hydroxyl and amide groups [36]. Xia et al. [37] reported that the lignin removal results using ChCl–glycerol exhibited limited fractionation efficiency in biomass pre-treatment. A mechanism of inefficiency was identified as a result of weak competitive interactions. In this study, ChCl–glycerol has weak acid properties, but Zhao et al. [38] evaluated ChCl–glycerol as a neutral DES. Neutral DES and weak bases have little effect on the pre-treatment of agricultural waste. The ChCl–levulinic acid also has a lower viscosity than the other DES. Low viscosity can increase the mobility of solvent molecules and accelerate the processing of lignocellulosic biomass because it increases mass and heat transfer [39].

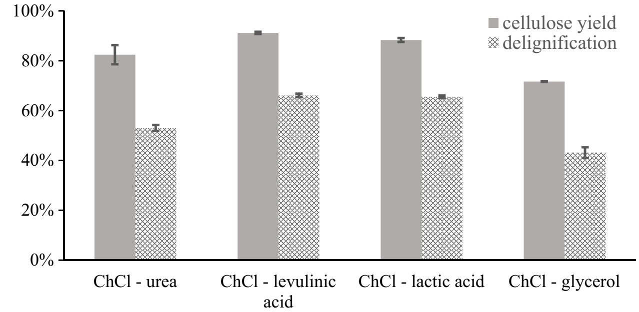

**Figure 1:** Cellulose yield and delignification of OPT with different DES (temperature: 110°C, mass ratio: 5%, DES ratio: 1:2).

The functional group contained in DES acid influences the delignification process due to its association with proton transfer/donation. Tan et al. [40] observed that carboxylic acids resulted in greater delignification compared to polyol-based acids. ChCl–levulinic acid contains a carboxylic group and a carbonyl group at the gamma position, classified as a gamma-keto acid. The variation in functional groups influences the intensity of its hydrogen bonding interaction with lignin [41].

The cellulose yield correlated with the effectiveness of the delignification process during extraction. The cellulose yield range was 71.64–91.13%, and the delignification range was 43.12–66.05%. Based on this wide range, it can be proven through a one-way ANOVA test that different types of DES have a significant effect on cellulose yield and % delignification ($p < 0.05$). ChCl–levulinic acid was chosen as the DES for the further optimization process because it produced the highest cellulose yields.

### 3.3 Optimization of cellulose yield from OPT

#### 3.3.1 Extraction optimization using BBD

Determining the main extraction process parameters has a significant influence on the response. Therefore, BBD

**Table 1:** Factor levels of BBD effect surface

| Factors | Low<br>−1 | Central<br>0 | High<br>+1 |
|---|---|---|---|
| Mass ratio (%) | 5 | 7.5 | 10 |
| Temperature (°C) | 80 | 95 | 110 |
| Reaction time (h) | 2 | 4 | 6 |

**Table 3:** ANOVA in the quadratic model of ChCl–levulinic acid treatment

| Source | Sum of squares | df | Mean square | *F*-value | *p*-value |
|---|---|---|---|---|---|
| Model | 928.72 | 9 | 103.19 | 5.11 | 0.0436 |
| *A*-mass ratio | 5.88 | 1 | 5.88 | 0.2914 | 0.6125 |
| *B*-temperature | 433.36 | 1 | 433.36 | 21.47 | 0.0057 |
| *C*-time | 45.51 | 1 | 45.51 | 2.25 | 0.1935 |
| *AB* | 0.0000 | 1 | 0.0000 | 0.0000 | 1.0000 |
| *AC* | 45.70 | 1 | 45.70 | 2.26 | 0.1928 |
| *BC* | 148.60 | 1 | 148.60 | 7.36 | 0.0421 |
| $A^2$ | 17.65 | 1 | 17.65 | 0.8746 | 0.3926 |
| $B^2$ | 128.46 | 1 | 128.46 | 6.36 | 0.0530 |
| $C^2$ | 104.44 | 1 | 104.44 | 5.17 | 0.0720 |
| Residual | 100.93 | 5 | 20.19 | | |
| Lack of fit | 74.27 | 3 | 24.76 | 1.86 | 0.3687 |
| Pure error | 26.66 | 2 | 13.33 | | |
| Cor total | 1,029.65 | 14 | | | |

centred on three different extraction parameters with three levels using the selected DES. The number of experiments ($N$) is $N = 2n(n-1) + C_0$, where $n$ is the factor number, and $C_0$ is the central point number. From 15 experimental runs, nodes −1 to +1 with a centre value of 0 are presented for parameters of extraction (Table 1).

Table 2 shows the actual and predicted yield differences under pre-treatment conditions to maximize cellulose yield as the response variables in the design of experiment analysis. The $R^2$ models of ChCl–levulinic acid were found to be 0.9020. The highest cellulose yield with ChCl–levulinic acid was obtained in run 2 (mass ratio 7.5%, temperature 95°C, and extraction time 4 h).

ANOVA for the developed model on ChCl–levulinic acid corresponds to the significance model ($F = 5.11$ and $p < 0.05$) (Table 3). Prediction model validation is required to examine the fitted model and ensure it adequately approximates the actual case. The linearity of actual and predicted response values confirmed the significant model correlation (Figure 2). In addition, the effect of each parameter, such as mass ratio, temperature, and time, is significant in maximizing cellulose yield ($p < 0.05$). The individual parameter that had a significant effect was temperature ($F = 21.47$), while mass ratio and extraction time were not significant ($p > 0.05$). Only the interaction between temperature and time was significant ($F = 7.36$), while the other interactions were not significant. Lack of fit

**Table 2:** BBD of OPT pre-treatment for maximizing cellulose yield

| Run | *A*: mass ratio (%) | *B*: temperature (°C) | *C*: time (h) | ChCl–levulinic acid | |
|---|---|---|---|---|---|
| | | | | Cellulose yield (%) Actual value* | Cellulose yield (%) Predicted value** |
| 1 | 7.5 | 80 | 2 | 80.10 | 78.52 |
| 2 | 7.5 | 95 | 4 | 92.59 | 88.62 |
| 3 | 7.5 | 95 | 4 | 85.41 | 88.62 |
| 4 | 10 | 95 | 6 | 79.90 | 78.86 |
| 5 | 5 | 95 | 2 | 84.31 | 85.35 |
| 6 | 10 | 80 | 4 | 79.08 | 76.69 |
| 7 | 5 | 95 | 6 | 91.31 | 87.34 |
| 8 | 7.5 | 95 | 4 | 87.85 | 88.62 |
| 9 | 7.5 | 80 | 6 | 58.13 | 61.56 |
| 10 | 7.5 | 110 | 6 | 86.89 | 88.47 |
| 11 | 10 | 95 | 2 | 86.42 | 90.39 |
| 12 | 10 | 110 | 4 | 91.95 | 91.41 |
| 13 | 7.5 | 110 | 2 | 84.48 | 81.05 |
| 14 | 5 | 80 | 4 | 77.86 | 78.40 |
| 15 | 5 | 110 | 4 | 90.73 | 93.12 |

*Actual value from the experiments. **Predicted value from a software of Design Expert 13.

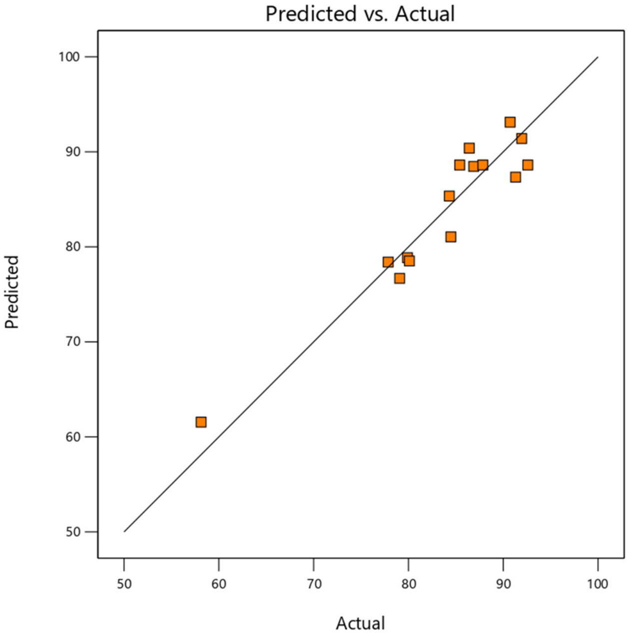

**Figure 2:** Actual vs predicted response for cellulose yield with ChCl–levulinic acid.

confirms that it is not significant ($F = 1.86$), and there is a 36.87% possibility of lack of fit due to noise.

The second-order polynomial equation (Eq. 7) provides a model for the response variable and the different parameters that affect cellulose yield

$$\mathrm{CY} = 88.62 - 0.8575A + 7.36B - 2.38C - 3.38AC + 6.09BC + 2.19A^2 - 5.90B^2 - 5.32C^2$$

It is widely assumed that the model must have a minimum $R^2$ value of 80% to be considered well-fitted [42]. These equations had regression coefficients ($R^2$) of more than 90%. The model's satisfactory $R^2$ values for cellulose yield using ChCl–levulinic acid is 90.2%.

### 3.3.2 Plots of response surface

The three-dimensional (3D) surface plot shows the correlation between each parameter and its interaction with cellulose yield. The 3D surface graphic demonstrates the influence of each independent variable on the response value and illustrates the interactions among the independent variables [43].

Figure 3 shows that at constant mass and time, an increase in temperature leads to an increase in cellulose yield. The viscosity of DES decreased as the temperature increased. Therefore, at a greater temperature, a better mass transfer between OPT and DES was achieved and ultimately affected the efficiency of the extraction process [44,45]. Increasing the initial temperature from 80°C to 110°C can increase the dissolution of lignin and the breaking of ether and ester bonds in OPT. Kinematically, the protonation rate and diffusion rate also increase.

When reaching the optimal point, similar trends in time and temperature were observed, but no noticeable drop in the curve above 110°C (Figure 3a and c). If the experiment is continued for higher temperatures, the cellulose yield may increase until it eventually decreases. The temperature in this experiment was taken at the exact upper limit of 110°C to determine the effect of cellulose yield optimization according to temperature in screening using four different DES. But overall, from the graph, it can be seen that a too high temperature will cause a decrease in cellulose yield. Suopäjarvi et al. [46] conducted delignification experiments on various DES using ChCl mixtures with variations of lactic acid, malic acid, levulinic acid, glycerol, and glutaric acid. The result was that the highest cellulose yield was obtained in ChCl–levulinic acid at 100°C. A similar phenomenon was reported by Jančíková et al. [47], who conducted delignification in the temperature range of 80–160°C and reached optimum conditions at 111°C with a lignin content of 16.44%. Further increases in temperature resulted in the degradation of alpha-cellulose, thus reducing the yield of solids. Temperature was the only significant individual parameter on cellulose yield in using DES ChCl–levulinic acid. In contrast, the other parameters (mass ratio and reaction time) had no significant effect (Table 3).

Figure 3b illustrates that at a constant temperature and mass ratio, cellulose yield increases during a specific time. The longer reaction times result in a greater contribution of protons, hence enhancing the cleavage of ether bonds in lignin and glycosidic bonds in polysaccharides [48]. Conversion increases significantly and reaches equilibrium at sufficient reaction time [49]. However, if the extraction process continues, the opposite phenomenon will occur a reduction of cellulose yield. Research by Chen et al. [50] indicated that carboxylic acid-based DES pre-treatment efficiently eliminates lignin within 3–24 h at 60–150°C.

A significant interaction effect is the interaction between temperature and time (Table 3). As illustrated in Figure 3c, at 6 h, the cellulose yield increased significantly with increasing temperature. At 2 h, the cellulose yield increases from 80°C to 95°C, followed by a decrease at 110°C. Longer extraction times result in a greater proton contribution, thus increasing the breakdown of ether bonds in lignin and glycosidic bonds in polysaccharides. Studies conducted by Chen et al. [50] have shown that treatment with DES efficiently eliminates lignin within 3 to 24 h at temperatures ranging from 60°C to 150°C.

### 3.3.3 Model validation

RSM was applied to determine the optimal conditions for maximizing cellulose yield from OPT, with experimental results serving as validation. From the optimization formula in Table 4, the mass ratio, temperature, and time were optimized within 5–10%, 80–110°C, and 2–6 h. The cellulose yield response was optimized with the maximum target and the highest level of importance. Response with a certain desirability is obtained. Desirability is a value that determines the accuracy degree of the optimal solution [51].

Figure 4 indicates that the desirability value was lower in the region characterized by low temperature at the specified mass ratio when keeping the time at 3.76 h. The optimum area at the top of the plot, indicated by a desirability value of 1, means the closeness to the target response (Figure 4a). The analysed results of the desirability ramp shown in Figure 4b indicate that each point

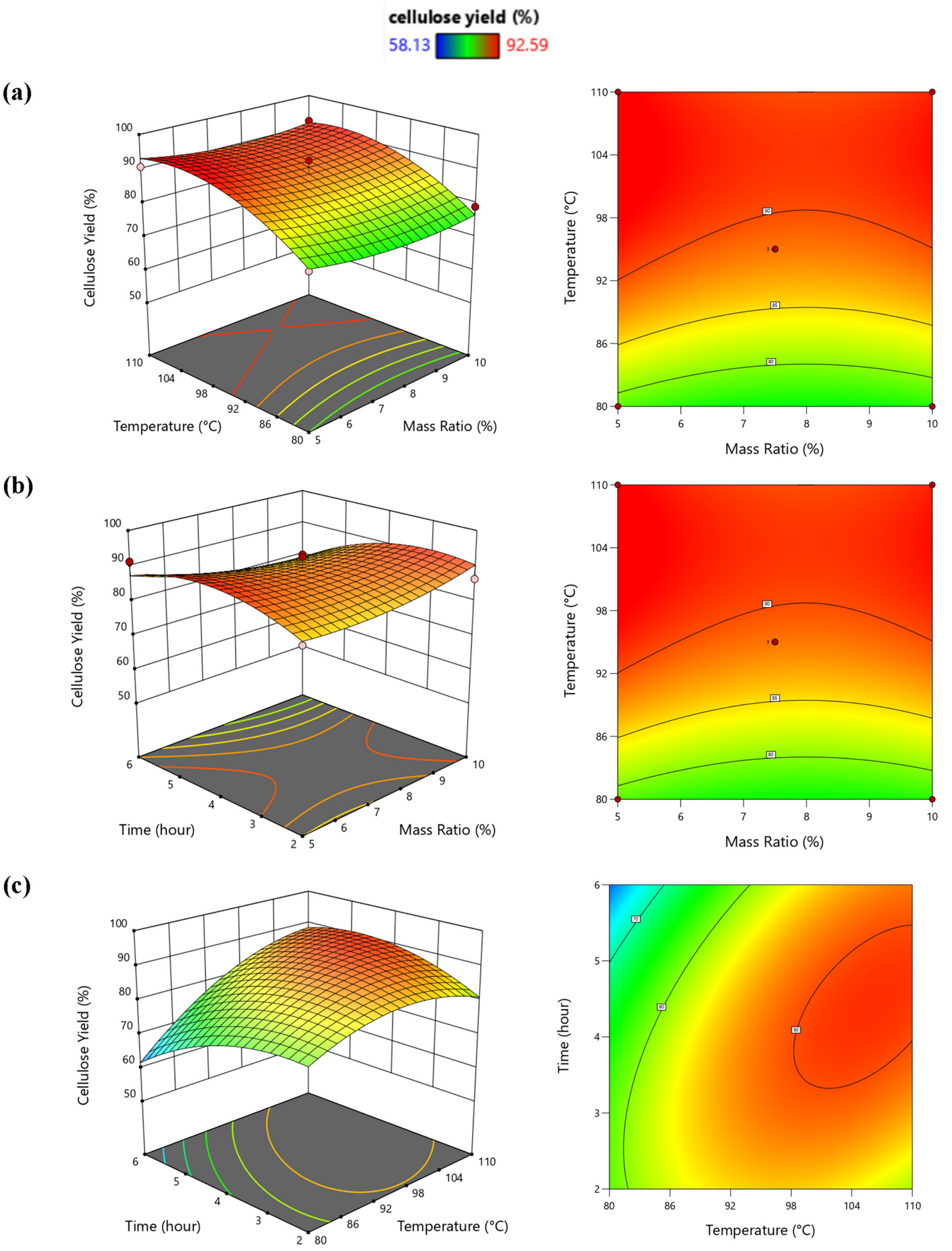

**Figure 3:** Response surface diagrams of cellulose yield with ChCl–levulinic acid on the interaction (a) temperature and mass ratio, (b) time and mass ratio, and (c) time and temperature.

**Table 4:** Response components and importance at the formula optimization

| Parameter | Goal | Lower level | Upper level | Importance |
|---|---|---|---|---|
| *A*: mass ratio | Is in range | 5 | 10 | +++ |
| *B*: temperature | Is in range | 80 | 110 | +++ |
| *C*: time | Is in range | 2 | 6 | +++ |
| Cellulose yield (ChCl–levulinic acid) | Maximize | 58.13 | 92.59 | +++++ |

on the ramp corresponds to a specific factor setting or response prediction. The optimal extraction parameters were identified as a mass ratio of 5.05%, a temperature of 104.2°C, and a duration of 3.76 h, resulting in a predicted cellulose yield of 93.22%. Experiments were performed to confirm the optimal predicted value of the response variable based on the specified independent parameters. The experimental results showed cellulose yield values of 91.29%. A 95% confidence interval between the experimental results and the RSM-predicted value indicates that the extraction parameters were adequate, precise, and reliable enough to be used in practice.

The research has obtained optimal parameters for the extraction process, including types of DES, temperature, time, and ratio using DES ChCl–levulinic acid. A further study, such as a life cycle assessment [52], can explore the sustainability and environmental effects of using DES. Techno-economic studies are needed for an efficient process and cost-effectiveness [53]. Actually, there is another parameter that can influence the extraction process, which is particle size. Ratnakumar et al. [54] investigated the effect of rice straw particle size on nanofibres' yield and

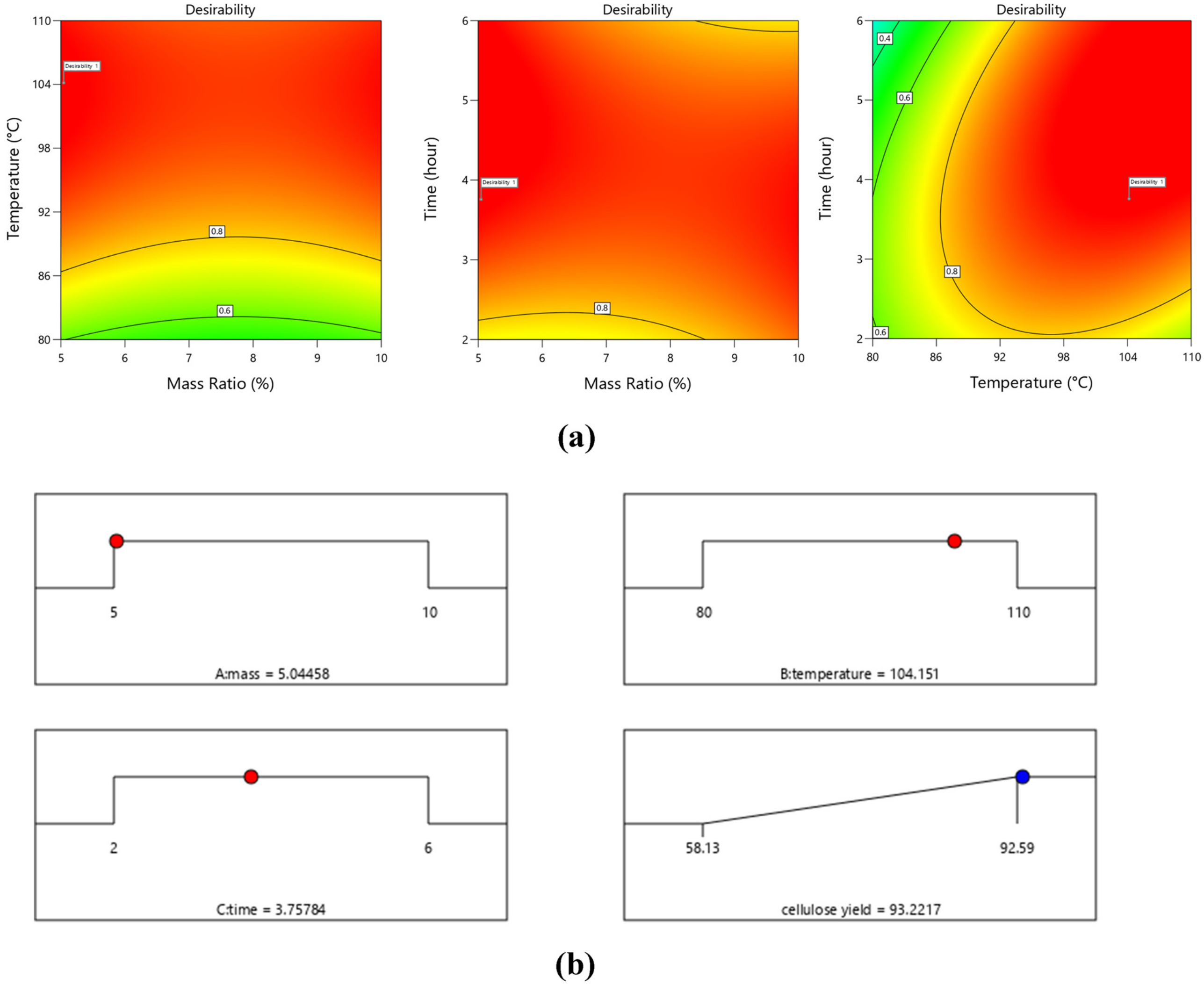

**Figure 4:** (a) Contour plots of the desirability surface graph and (b) desirability ramp.

**Table 5:** Lignocellulosic content of untreated and treated OPT

| OPT | Cellulose (%) | Hemicellulose (%) | Lignin (%) | Cellulose yield (%) | Delignification (%) |
|---|---|---|---|---|---|
| Untreated | 38.26 ± 0.03 | 31.04 ± 0.18 | 28.76 ± 0.06 | — | — |
| Treated with ChCl–levulinic acid | 47.34 ± 3.72 | 32.25 ± 1.82 | 17.53 ± 2.34 | 83.80 ± 8.58 | 58.08 ± 10.19 |

final characteristics. It was shown that particle size affects product properties, with larger particle sizes resulting in longer fibres. Sheng et al. [55] confirmed that smaller particles produce elevated extraction rates due to enhanced solvent contact and reduced mass transfer resistance. In this research, particle size was fixed under 120 mesh.

## 3.4 Characterization of treated OPT

Changes in lignocellulose content after pre-treatment with DES are shown in Table 5. After delignification, the lignin content was reduced by 39% of the initial content. The cellulose content increased with an average cellulose yield of 83.8%. Acidic conditions break the lignin–carbohydrate complex bonds and dissolve the lignin into the solvent [56]. The elimination of lignin is controlled by proton dissociation capacity, which is affected by hydrogen bonds between molecules. The hydrogen bonds influence the lignin ether bonds, decreasing the energy necessary for their cleavage [57].

To correlate each analysis, FTIR and TGA characterization was conducted. FTIR determines the functional groups in opt before and after treatment (Figure 5). Raw OPT and treated OPT are divided into two peaks in the range of 500–1,750 $\text{cm}^{-1}$ and 2,850–3,500 $\text{cm}^{-1}$. These areas are consistent with the research of Al Ragib et al. [58]. The 3,343 $\text{cm}^{-1}$ absorption band occurs due to the –OH group stretching vibration between hydrogen bonds, while 2,918 $\text{cm}^{-1}$ is due to the –CH group of cellulose. Acid DES-treated cellulose fibres have weaker absorption band intensity because the cellulose molecular chains are less exposed due to aggregation inside [59].

The peaks at 1,593 and 1,733 $\text{cm}^{-1}$ in the hemicellulose and lignin spectra correspond to C═C stretching vibrations of the aromatic ring in lignin, as well as C═O vibrations from acetyl ester and uronic acid groups. A verified lignin absorption band is observed at 1,234 $\text{cm}^{-1}$, attributed to the C–O–C bond of the ether [60]. This peak signifies that the raw OPT comprises a complicated cellulose, hemicellulose, and lignin arrangement. Following the treatment of OPT with DES, a reduction in the strength of the absorption peaks for hemicellulose and lignin was observed. The peak near 1,034 $\text{cm}^{-1}$, typically linked to the stretching of the C–O–C pyranose ring, the beta-1,4-glycosidic bond, and the asymmetric vibrational motion of C–O–C in cellulose, may exhibit variations in intensity or position indicative of alterations in the cellulose structure due to DES treatment [61]. DES is known for effectively delignification of lignocellulosic biomass, resulting in the rupture of hydrogen bonding and elimination of amorphous components from the OPT raw material.

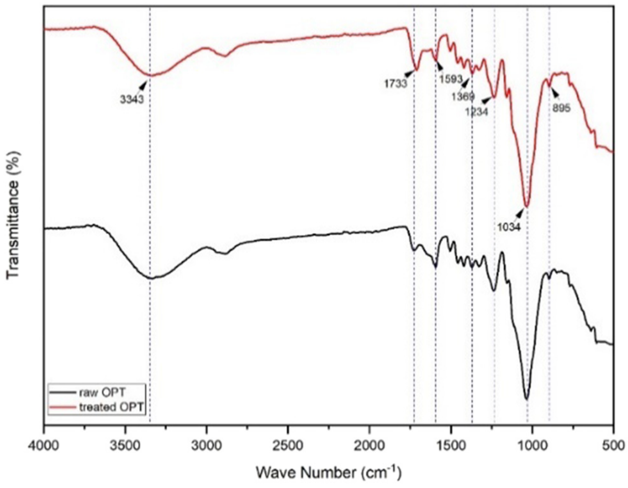

**Figure 5:** FTIR curve for raw and treated OPT.

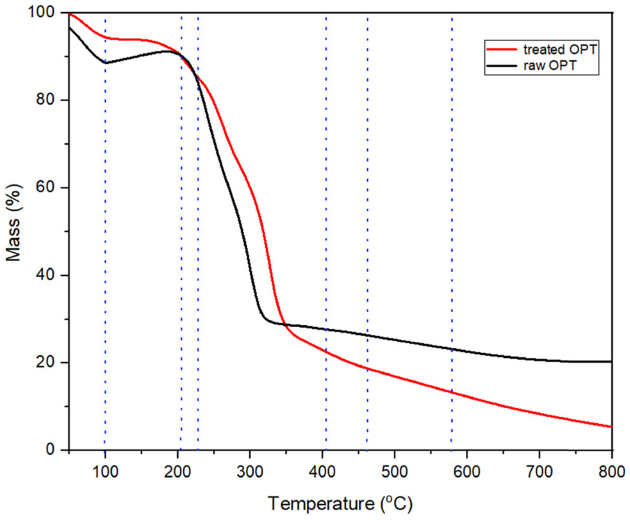

**Figure 6:** TGA curve for raw and treated OPT.

**Table 6:** Thermal properties of raw and treated OPT

| Sample | Initial weight loss (%) | $T_{onset}$ (°C) | $T_{max}$ (°C) | $T_{final}$ decomposition (°C) | Main weight loss (%) | Char residue weight (%) |
|---|---|---|---|---|---|---|
| Raw OPT | 15.13 | 206.1 | 301.5 | 403.5 | 64.64 | 20.23 |
| Treated OPT | 9.98 | 225.7 | 325.7 | 461.4 | 84.61 | 5.41 |

The decomposition of raw and treated OPT is shown in Figure 6. There are three stages of weight loss that can be observed. The first stage is at 50–100°C, which represents weight loss caused by the evaporation of water molecules. Furthermore, the weight loss is insignificant at 100–200°C due to the evaporation of other volatile compounds in the sample. The second stage occurs at 200–350°C, where the crystalline part of the cellulose decomposes rapidly. The third stage is at 350–500°C, where weight reduction occurs due to the decomposition of the amorphous component. At temperatures above 500°C, the final oxidation of carbonaceous char residues with aromatic polycyclic structures [60].

Table 6 shows the results of TGA analysis on the OPT sample before and after treatment. The comparison of initial weight loss is related to water evaporation, where treated OPT is smaller than raw OPT due to the reduction of hydrophilic properties, which is consistent in previous literature [62]. The thermal analysis data shows that raw OPT has a lower degradation temperature ($T_{onset}$ and $T_{max}$) than treated OPT. In treated OPT, there is an increase in cellulose crystals, which causes higher degradation temperature, which is accomplished by chemically eliminating lignin and hemicellulose [63]. The amount of char residue of raw OPT is larger because it contains more lignin. Lignin generates the highest quantity of char residue among lignocellulosic materials due to its benzene ring configurations, while hemicellulose contributes to char formation through the catalysis of inorganic mineral impurities [58].

## 4 Conclusion

DESs based on ChCl and carboxylic acid effectively extract cellulose from OPT. The RSM method was used to obtain the optimal conditions of cellulose yield for OPT extraction using ChCl–levulinic acid. Optimal conditions were achieved with a mass ratio of 5.05%, a temperature of 104.2°C, and a reaction time of 3.76 h. These results were validated with experimental resulting in a cellulose yield of 91.29%. The FTIR and TGA data indicated an increase in cellulose content after treatment.

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**Data availability statement:** The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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