

Supplementary Materials

3.1. Evaluation of the rotary experiment

Table S1. Obtained group analysis of variance in terms of lignin content.

Indication	s	f	s ²	F	F _k
S ₁	282.77	3	94.26	249.38	5.41
S ₂	201.44	6	33.57	88.26	4.95
S _e	1.89	5	0.38	1	-
S _{LF}	22.90	5	4.58	12.12	5.05

S₁ - the variability component characterized by the terms of the linear part of the regression equation,

S₂ - the variability component characterized by quadratic and interaction terms regression equation,

S_e - the component characterizing the experimental variability,

S_{LF} - the component characterizing the mathematical inaccuracy of the regression equation,

F_k - the component characterizing the critical value of the F criterion when using a significance level of 0.05.

Furthermore, we obtained a set of regression equations, the regression coefficients of which we summarized in Table S2. Using the method of least squares, we subsequently obtained the calculated coefficients of the quadratic model.

Table S2. Obtained coefficients of regression equations when monitoring the efficiency of delignification and content of lignin, cellulose, holocellulose and ash.

Coefficients	Delignification efficiency (%)		Lignin content (%)		Cellulose content (%)		Holocellulose content (%)		Ash content (%)	
	b _i	b _{crit.}	b _i	b _{crit.}	b _i	b _{crit.}	b _i	b _{crit.}	b _i	b _{crit.}
b ₀	31.83	2.31	19.01	0.64	59.75	1.13	75.48	1.60	4.50	0.36

b ₁	-12.97	1.53	3.62	0.43	-0.16	0.75	-2.34	1.06	0.15	0.24
b ₂	-3.23	1.53	0.90	0.43	0.74	0.75	-1.48	1.06	0.03	0.24
b ₃	9.37	1.53	-2.61	0.43	0.98	0.75	2.66	1.06	0.01	0.24
b ₁₁	-12.45	1.49	3.47	0.42	-5.59	0.73	-4.04	1.03	0.06	0.23
b ₁₂	0.64	2.00	-0.18	0.56	3.05	0.98	-0.77	1.39	-0.02	0.31
b ₁₃	5.46	2.00	-1.52	0.56	3.41	0.98	1.94	1.39	-0.50	0.31
b ₂₂	-0.95	1.49	0.26	0.42	-0.18	0.73	-0.21	1.03	-0.34	0.23
b ₂₃	0.28	2.00	-0.08	0.56	3.67	0.98	0.47	1.39	-0.68	0.31
b ₃₃	-3.99	1.49	1.11	0.42	0.95	0.73	-0.83	1.03	0.06	0.23

Marking $b_{crit.}$ represents the critical values of the given coefficient at a probability of 95%. We have marked regression coefficients that are statistically significant in bold. At the same time, we obtained equation (S1) from these coefficients describing the lignin content with the included coefficients for the temperature, time and ratio parameters monitored by us:

$$Lignin\ content = 19.01 + 3.62 x_1 + 0.90x_2 - 2.61 x_3 - 0.18 x_1x_2 - 1.52 x_1x_3 - 0.08 x_2x_3 + 3.47 x_{11} + 0.26 x_{22} + 1.11 x_{33} \quad (S1)$$

while the evaluated parameter is the lignin content, the regression coefficients are characterized by the obtained numerical values and the encoded factor levels represent x_i (x_1 represents temperature parameter, x_2 time parameter and x_3 biomass to solvent ratio parameter). Subsequently, we performed an analysis to monitor the influence of factors on the resulting lignin content. Our task was to test the significance of the effects of these factors by calculating the critical ones values of the coefficients shown in Table S2 as $b_{crit.}$. We used the equation (S2) to calculate them:

$$b_{crit.} = s_b * t_k \quad (S2)$$

where s_b represents the root mean square error for the respective coefficient and t_k is characterized by the tabular critical value of Student's t-distribution at the given level reliability and number of degrees of freedom.

The next step in evaluating the planned experiment is checking the adequacy of the model. To check the adequacy, we used a mathematical model and evaluated it using the S_{LF} variability component using the equation (S3):

$$S_{LF} = S_R - S_E \quad (S3)$$

where the residual component of variability is characterized by S_R and S_E represents part of the experimental (independent) variability.

Table S3. Optical microscopy images on samples after delignification using DES-like mixtures in a composition of choline chloride and lactic acid in a molar ratio of 1:5 at magnifications of 230X, 480X, and 930X (experiment 1 to 3).




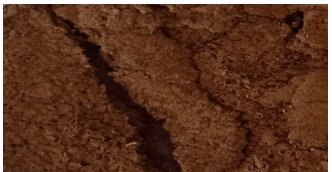
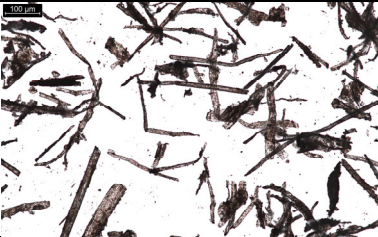
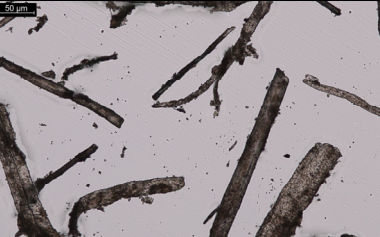


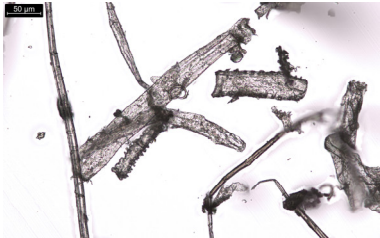
Number of experiment	Magnification 230X	Magnification 480X	Magnification 930X
1			
2			
3			

Table S4. Optical microscopy images on samples after delignification using DES-like mixtures in a composition of choline chloride and lactic acid in a molar ratio of 1:5 at magnifications of 230X, 480X, and 930X (experiment 4 to 6).



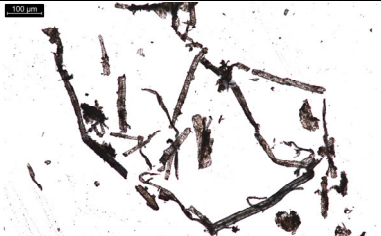


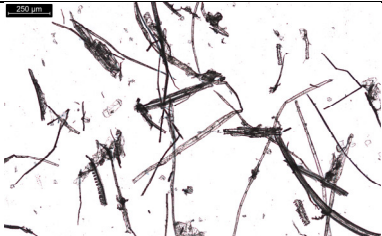

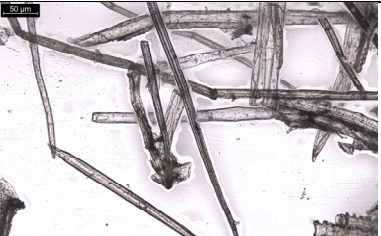

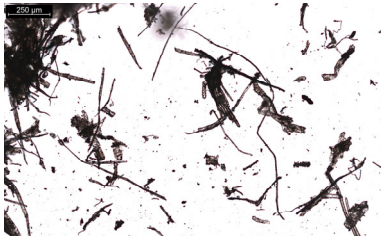

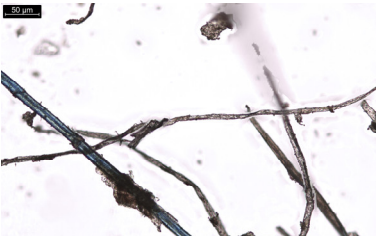
Number of experiment	Magnification 230X	Magnification 480X	Magnification 930X
 4			
 5			
 6			

Table S5. Optical microscopy images on samples after delignification using DES-like mixtures in a composition of choline chloride and lactic acid in a molar ratio of 1:5 at magnifications of 230X, 480X, and 930X (experiment 7 to 9).


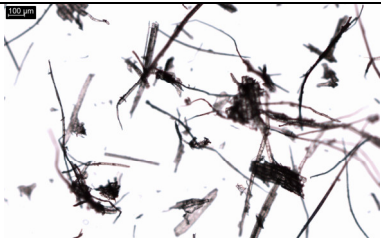

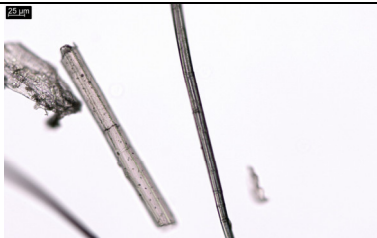
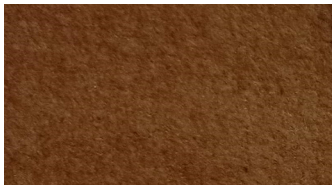


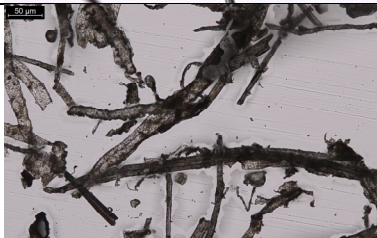
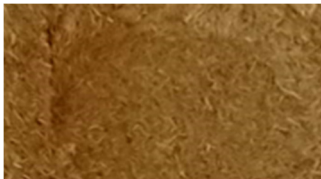

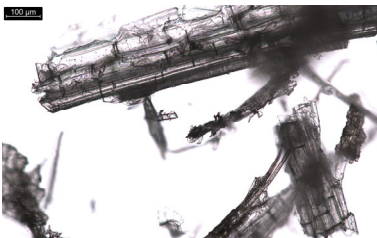
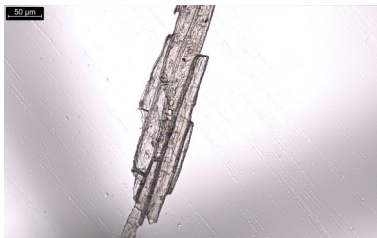
Number of experiment	Magnification 230X	Magnification 480X	Magnification 930X
 7			
 8			
 9			

Table S6. Optical microscopy images on samples after delignification using DES-like mixtures in a composition of choline chloride and lactic acid in a molar ratio of 1:5 at magnifications of 230X, 480X, and 930X (experiment 10 to 12).

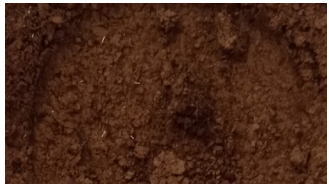
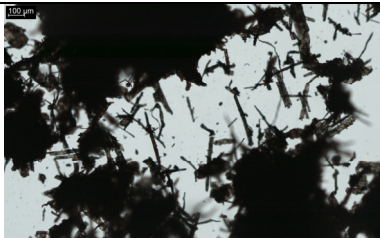
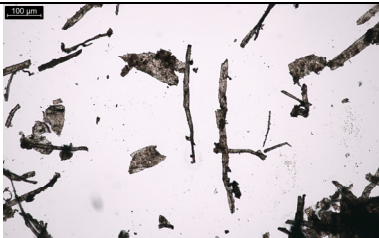
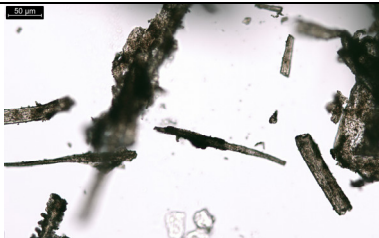
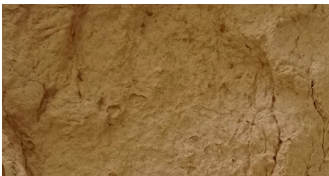


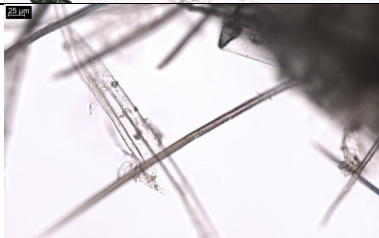
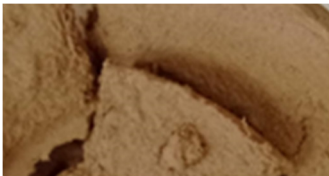

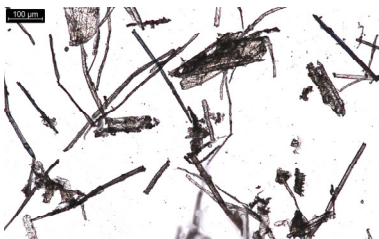

Number of experiment	Magnification 230X	Magnification 480X	Magnification 930X
 10			
 11			
 12			

Table S7. Optical microscopy images on samples after delignification using DES-like mixtures in a composition of choline chloride and lactic acid in a molar ratio of 1:5 at magnifications of 230X, 480X, and 930X (experiment 13 to 15).

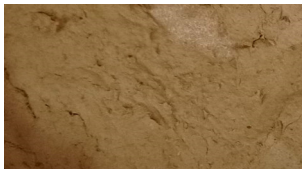
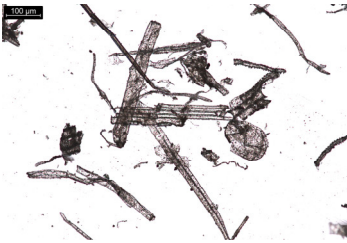
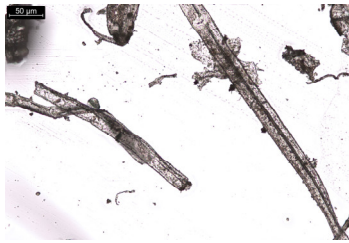

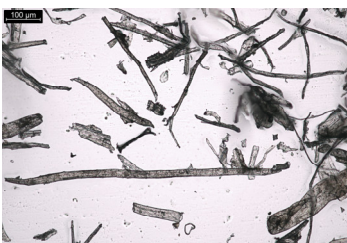
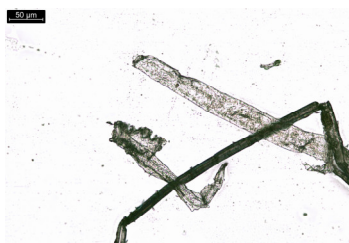


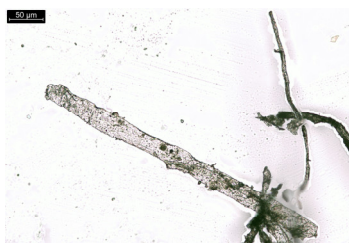
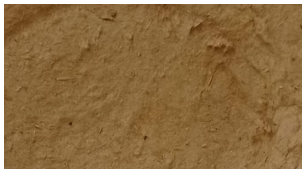
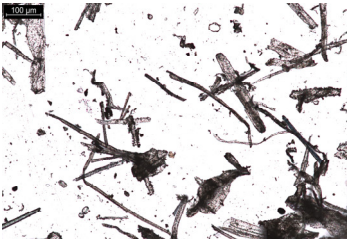
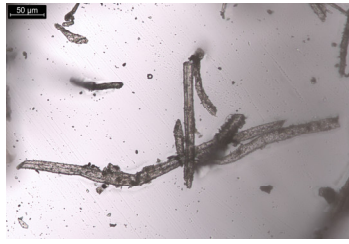
Number of experiment	Magnification 230X	Magnification 480X	Magnification 930X
13			
14			
15			

Table S8. Optical microscopy images on samples after delignification using DES-like mixtures in a composition of choline chloride and lactic acid in a molar ratio of 1:5 at magnifications of 230X, 480X, and 930X (experiment 16 to 20).

Number of experiment	Magnification 230X	Magnification 480X	Magnification 930X
16			

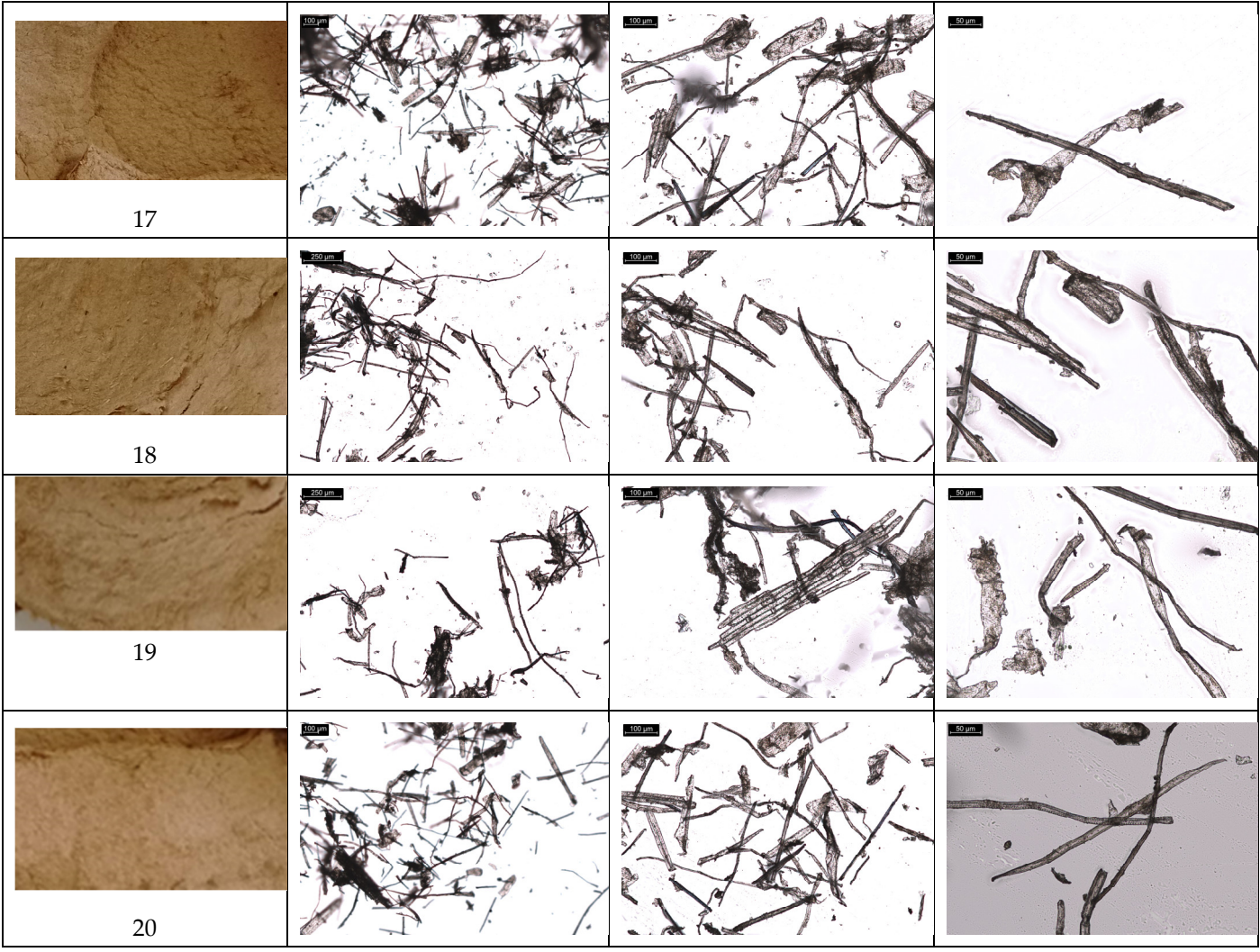
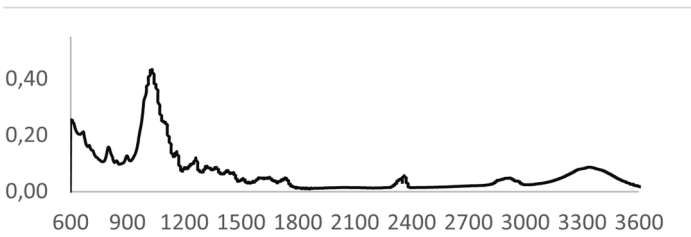
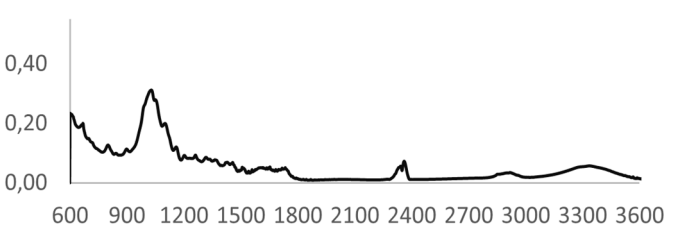
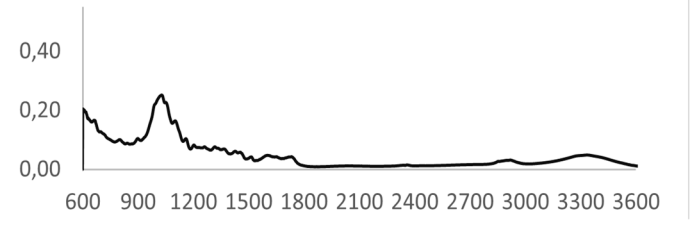
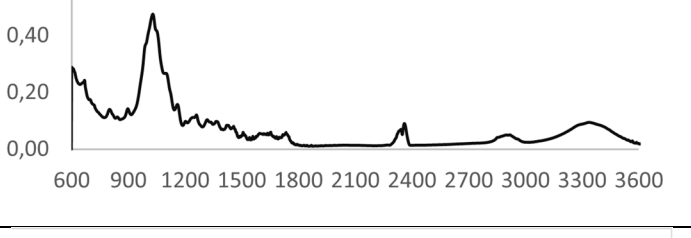
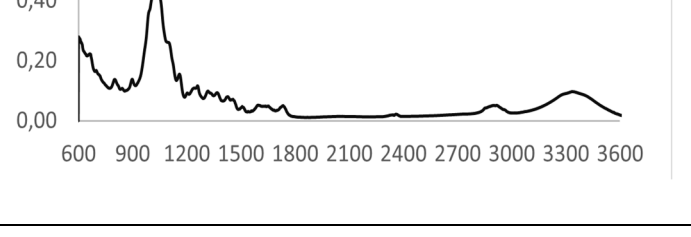
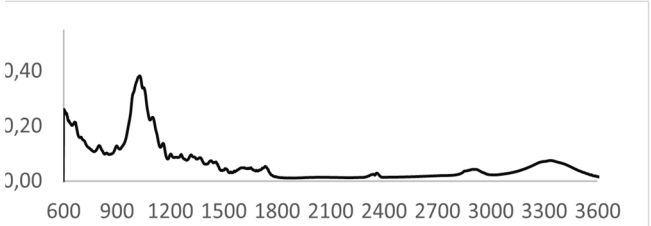
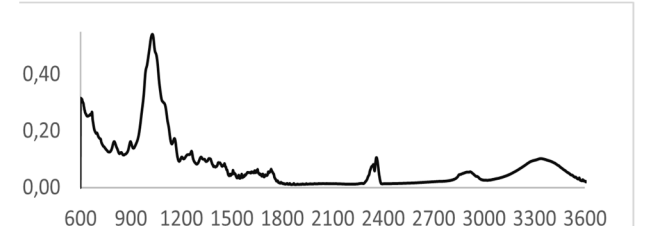
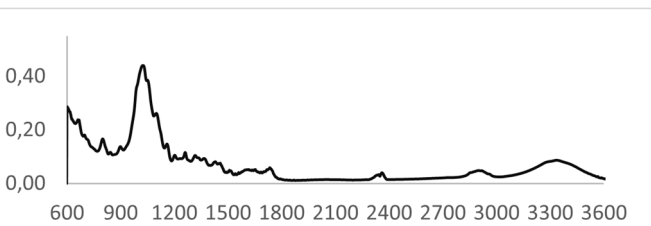
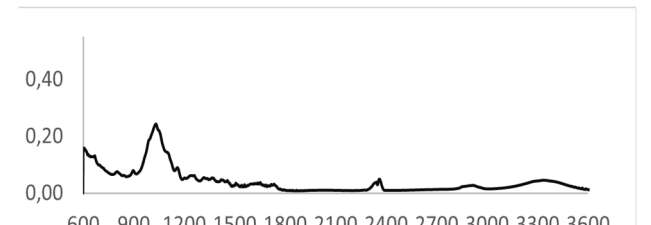
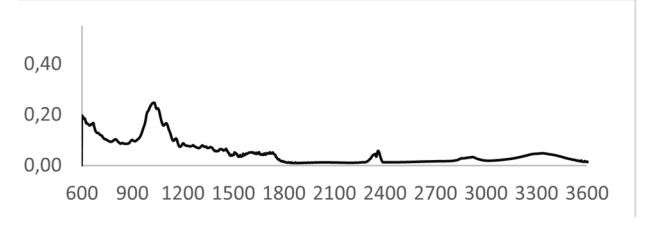

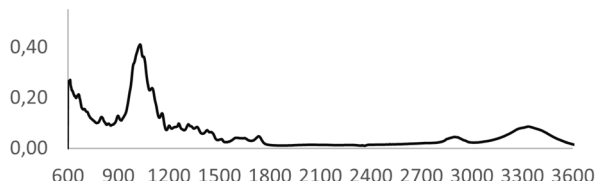
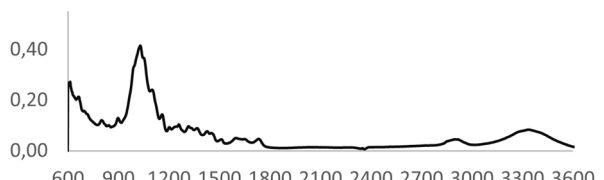
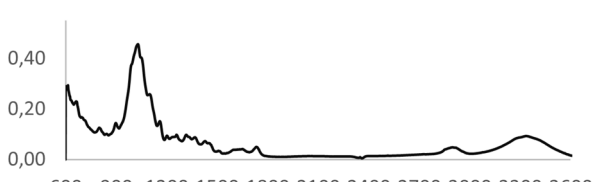
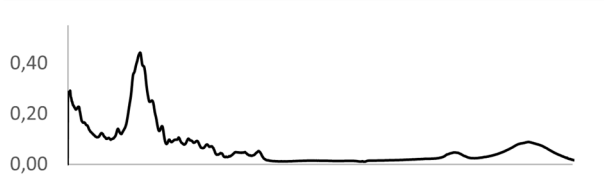


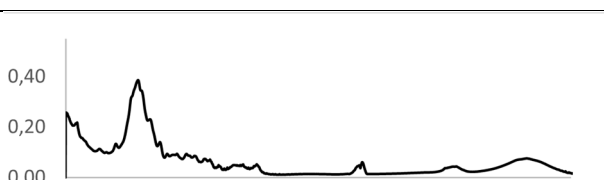
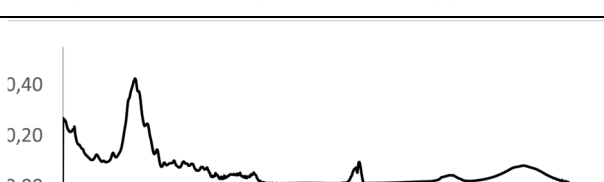
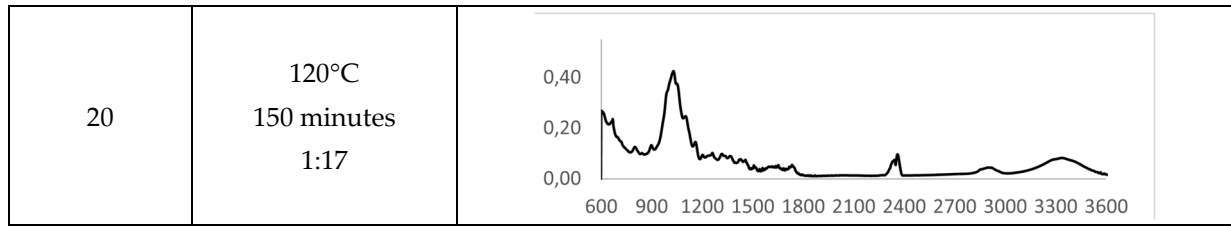


Table S9. FT-IR spectra of pulps obtained after delignification according to the rotary experiment using DES-like mixtures based on choline chloride and lactic acid in a molar ratio of 1:5.

Number of experiment	Delignification conditions (temperature, time, biomass/solvent ratio)	FT-IR spectrum
1	96°C 97 minutes 1:12	 The FT-IR spectrum for experiment 1 shows a prominent peak at approximately 1030 cm⁻¹ with an intensity of about 0.40. There are smaller peaks around 1200 cm⁻¹ and 1500 cm⁻¹. The x-axis ranges from 600 to 3600 cm⁻¹, and the y-axis ranges from 0.00 to 0.40.
2	144°C 97 minutes 1:12	 The FT-IR spectrum for experiment 2 is very similar to experiment 1, with a major peak at 1030 cm⁻¹ (intensity ~0.35) and smaller features at 1200 and 1500 cm⁻¹. The axes are the same as in experiment 1.
3	96°C 204 minutes 1:12	 The FT-IR spectrum for experiment 3 shows a peak at 1030 cm⁻¹ with a lower intensity of approximately 0.25 compared to experiments 1 and 2. The x-axis ranges from 600 to 3600 cm⁻¹, and the y-axis ranges from 0.00 to 0.40.
4	144°C 204 minutes 1:12	 The FT-IR spectrum for experiment 4 shows a peak at 1030 cm⁻¹ with an intensity of about 0.45, which is slightly higher than in experiment 1. The x-axis ranges from 600 to 3600 cm⁻¹, and the y-axis ranges from 0.00 to 0.40.
5	96°C 97 minutes 1:30	 The FT-IR spectrum for experiment 5 shows a peak at 1030 cm⁻¹ with an intensity of about 0.45, similar to experiment 4. The x-axis ranges from 600 to 3600 cm⁻¹, and the y-axis ranges from 0.00 to 0.40.

6	144°C 97 minutes 1:30	 <p>DSC thermogram for sample 6. The x-axis represents temperature in Kelvin (K) from 600 to 3600, and the y-axis represents heat flow from 0,00 to 0,40. A prominent endothermic peak is observed at approximately 1050 K, reaching a maximum heat flow of about 0,35. Smaller peaks are visible at higher temperatures, around 2400 K and 3300 K.</p>
7	96°C 204 minutes 1:30	 <p>DSC thermogram for sample 7. The x-axis represents temperature in Kelvin (K) from 600 to 3600, and the y-axis represents heat flow from 0,00 to 0,40. A prominent endothermic peak is observed at approximately 1050 K, reaching a maximum heat flow of about 0,45. Smaller peaks are visible at higher temperatures, around 2400 K and 3300 K.</p>
8	144°C 204 minutes 1:30	 <p>DSC thermogram for sample 8. The x-axis represents temperature in Kelvin (K) from 600 to 3600, and the y-axis represents heat flow from 0,00 to 0,40. A prominent endothermic peak is observed at approximately 1050 K, reaching a maximum heat flow of about 0,45. Smaller peaks are visible at higher temperatures, around 2400 K and 3300 K.</p>
9	80°C 150 minutes 1:17	 <p>DSC thermogram for sample 9. The x-axis represents temperature in Kelvin (K) from 600 to 3600, and the y-axis represents heat flow from 0,00 to 0,40. A prominent endothermic peak is observed at approximately 1050 K, reaching a maximum heat flow of about 0,25. Smaller peaks are visible at higher temperatures, around 2400 K and 3300 K.</p>
10	160°C 150 minutes 1:17	 <p>DSC thermogram for sample 10. The x-axis represents temperature in Kelvin (K) from 600 to 3600, and the y-axis represents heat flow from 0,00 to 0,40. A prominent endothermic peak is observed at approximately 1050 K, reaching a maximum heat flow of about 0,25. Smaller peaks are visible at higher temperatures, around 2400 K and 3300 K.</p>
11	120°C 60 minutes 1:17	 <p>DSC thermogram for sample 11. The x-axis represents temperature in Kelvin (K) from 600 to 3600, and the y-axis represents heat flow from 0,00 to 0,40. A prominent endothermic peak is observed at approximately 1050 K, reaching a maximum heat flow of about 0,40. Smaller peaks are visible at higher temperatures, around 2400 K and 3300 K.</p>

12	120°C 240 minutes 1:17		
13	120°C 150 minutes 1:10		
14	120°C 150 minutes 1:60		
15	120°C 150 minutes 1:17		
16	120°C 150 minutes 1:17		
17	120°C 150 minutes 1:17		
18	120°C 150 minutes 1:17		
19	120°C 150 minutes 1:17		



2.6. Analysis methods

2.6.1.1 Determination of cellulose content

For the determination of cellulose content in original biomass samples and samples after delignification we used the modified Kurschner-Hoffer method. This method consisted of from weighing 1 g of fibers into a 250 ml Erlenmeyer flask, to which we added 100 ml of ethanol, 25 ml concentrated nitric acid and boiling stones. We subsequently closed the bank with a smaller one with a cooking glass so that the generated vapors do not escape into the surroundings. We boiled the fibers prepared in this way in a boiling water bath for one hour. Subsequently, we removed the flask from the water bath, cooled and the fibers were filtered through a frit with porosity S2. After washing them with a liter of cold water of distilled water we transferred them using 150 ml of distilled water back to the flask that we reheated in a boiling water bath for 30 minutes. After this time, we bank removed from the water bath, cooled, and filtered the fibers again on frit S2 using one-liter distilled water. After suctioning off the water, we closed the drain of the frit and filled the fibers with 75 ml of ethanol. After ten minutes, ethanol was aspirated, and alkaline extraction was performed. Prepared in advance lye mixture consisting of 4% sodium hydroxide, 4% sodium sulfite and distilled water, we are in three intervals they watered the threads. For the first time, we covered the fibers with 40 ml of lye mixture. After five minutes we added another 50 ml of lye mixture. After 30 minutes, we added another 50 ml of lye of the mixture, which has been active for only 5 minutes. After this, the lye mixture was sucked off and the fibers on the frit were removed washed with a liter of distilled water. We neutralized the residual lye by adding 100 ml of 2% acetic acid. Finally, we washed the fibers with at least another liter of distilled water to neutral pH. We dried the fibers processed in this way in a drying oven to a constant weight, while we determined the percentage of cellulose in the samples using to the equation (S4):

$$\text{Content of cellulose} = \frac{A}{m_{a.s.}} * 100 \% \quad (S4)$$

where A is a weight of samples (absolutely dry) after treatment in g, and $m_{a.s.}$ is a weight of samples (absolutely dry) before determination in g.

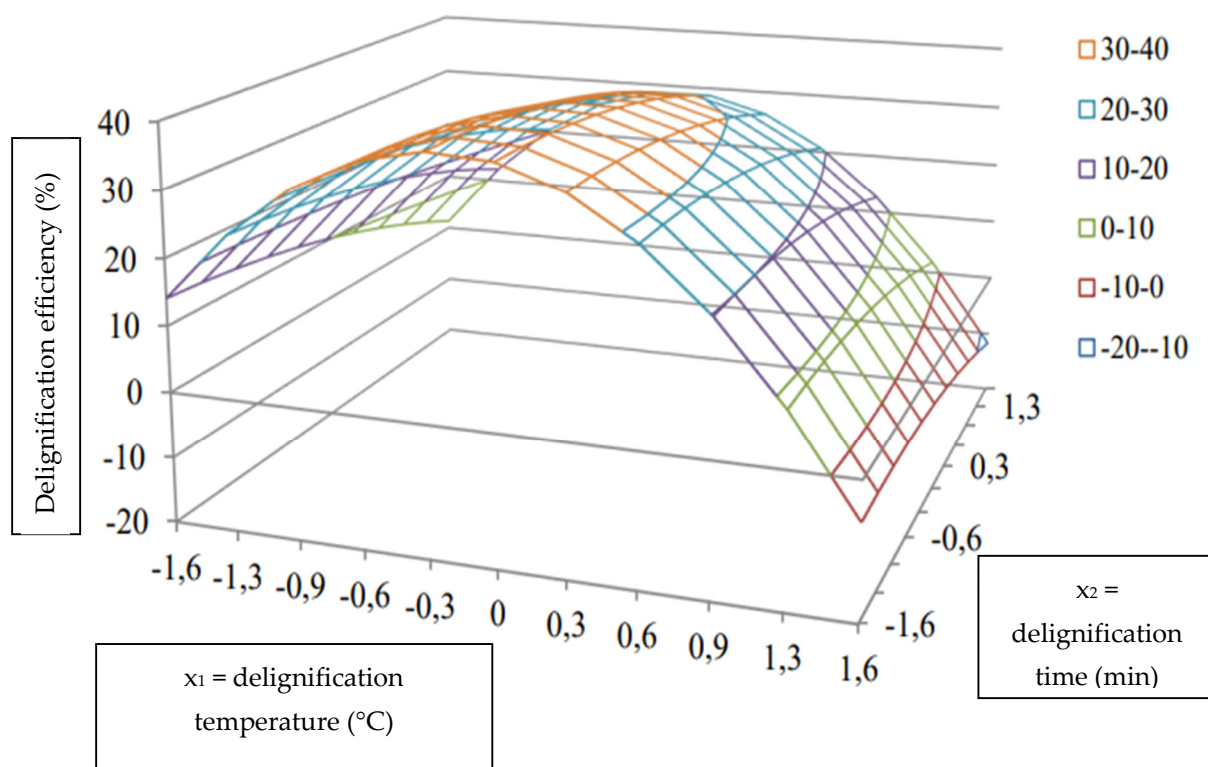


Figure S1. Dependence of delignification efficiency on delignification time and temperature.