The impact of molded pulp product process on the mechanical properties of molded BCTMP

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Abstract

This study was carried out using bleached softwood Chemi-Thermo-Mechanical Pulp to evaluate the influence of Molded Pulp Products' manufacturing process parameters on the finished products' mechanical and hygroscopic properties. A Taguchi table was done to make 8 tests with specific process parameters such as moulds temperature, pulping time, drying time and pressing time. The results of these tests were used to obtain an optimized manufacturing process with improved mechanical properties and a lower water uptake after sorption analysis and water immersion. The optimized process parameters allowed us to improve the Young' Modulus after 1h immersion of 58% and a water uptake reduction of 78% with the first 8 tests done.

Keywords: Cellulose fibers, Molded Pulp Product, mechanical properties, barrier properties

1. Introduction

As single use plastic packaging is becoming more controversial in the current environmental context, industrials have been studying other materials and manufacturing processes to propose new products with a lower environmental impact. Some industrials have chosen to turn to wood cellulose fiber historically used for the pulp and paper industry.

There are several processing methods known to extract cellulose fibers from wood chips. These processes either use mechanical technique, chemical technique, thermal technique or a combination of these techniques. A widely known and used technique in the paper industry is called kraft process [1,2] and uses a combination of thermal, mechanical and mostly chemical products to obtain cellulose fibers free of hemicellulose, lignin, pectin and other wood molecules.

Another fiber extraction method used is called Chemi-Thermo-Mechanical Pulping process or CTMP [2,3]. This process is considered to be a chemi-mechanical processing method as most of the wood molecules (lignin, hemicellulose) are kept in the final pulp. It has replaced some chemical pulps
for several applications as it is a less-expensive pulp to produce with a much higher yield of pulp produced.

In this method, the wood chips are first pretreated with about 2% of sodium sulfite or hydrogen peroxide, then they are sent to specific refiners with high pressure and steam to free the fibers. The high temperature and presence of steam allow the fibers to soften and reduce their cutting and fines formation. This pulp may be further bleached (BCTMP) to obtain a whiter pulp even if the result cannot be as white as for kraft pulps since lignin is maintained in the CTMP process [2].

Processed pulp (kraft or BCTMP) or recycled paper are used as raw material to make 3D products using a specific manufacturing process called Molded Pulp Products process (MPP process). The first patent of a molded pulp processing was published in 1890 [4], whereas a patent for a manufacturing machine was published in 1903 by Mr. Martin L. Keyes [5] . This process has been developed and modified but its use in industry was first limited to the manufacturing of egg trays. The MPP process can vary depending on the final destination of the manufactured product. According to the International Molded Fiber Association (IMFA) [6], there are 4 categories of MPPs depending on the process used [7], which are described as followed:

- “Thick-wall”: made using one mould resulting in a smooth side (in contact with mould) and a rougher side. The piece is oven dried, has a thickness of 5 to 10 mm and is mainly used to transport and protect heavy products (vehicle parts, motors, ...)

- “Transfer Molded”: made using two moulds, a forming one (same as the "thick-wall" process) and a transferring one. The surface in contact with the forming mould is smoother than the other surface. The resulting piece is also oven dried, 3 to 5 mm thick and is used to make egg trays or other product packaging to protect objects such as cellphones, electrical appliances and drink trays

- “Thermoformed” or “Thin-wall”: made using a number of moulds to press and dry the piece. The resulting product is 2 to 4 mm thick, denser than for previous processes and gives a smooth surface to both sides. Making a "thin-wall" product allows to obtain a piece with a similar finish to thermoformed plastic products and may be used to make drink trays, cellulose tableware such as plates, bowls or food trays that require a good product visual

- “Processed”: This type is referring to molded fiber products that require some type of secondary or special treatment other than simply being molded and cured, such as a coating, printing or cutting
on the finished piece. This process may be used for food applications that require a specific barrier
that cannot be done by the cellulose product alone

In this study, the "thin-wall" method was tested with 4 moulds used: a forming one, a transferring
one and two moulds to press and dry the piece. In order to better understand the influence of the MPP
process on the properties of the resulting product, 6 different process parameters were analyzed by
changing temperature or specific step time. The result of these experiments will allow us to obtain a
manufacturing process with specific parameters that gives higher mechanical properties and limited
water uptake following water immersion. Tensile tests in different conditions and water uptake analysis
were carried out to compare the influence of the process parameters tested on the structure and
properties of the manufactured MPPs. Water uptake tests were made following 1h water immersion and
sorption analysis 5 different water activities ($a_w$). Tensile tests included initial analysis as well as after
1h water immersion and sorption analysis in the same conditions as water uptake tests.

As 6 process parameters were chosen to be tested at 2 levels each, a total of 64 tests should be done
to correctly analyze all process possibilities. To lower the number of tests, a design of experiment was
done following the Taguchi method, allowing us to lower the number of tests needed to 8. With the
experimental results of these 8 specific tests, the Taguchi Table allowed us to obtain theoretical results
of all the other tests possibilities. 2 final tests with optimized process parameters from the initial 8 were
done to confirm the Taguchi method used for this study.

2. Materials and methods

2.1. Material and production of MPP

Bleached Chemi-Thermo-Mechanical Pulp (BCTMP) of softwood was purchased from Rottneros
(Sweden) and used with no further modification to the fibers. The preparation of the test samples was
done in several steps:

- 3D pieces manufacturing using the molded pulp process
- Samples cutting in the shape of shouldered bars using a punch cutting object
- Conditioning of the samples depending on the test to be done

The first step uses a molded pulp processing machine whose diagram is shown in Figure 1.
Figure 1: Diagram of the manufacturing process of molded pulp products (MPPs)

In this process 6 parameters, detailed in Table 1, were tested to observe and analyze their influence on the water uptake and the mechanical properties of the finished product. These parameters are easily changed in a production process depending on the desired production properties and objectives (cycle time, finished product water content).

Table 1: Process parameters tested

| Parameter tested       | Level 1         | Level 2         |
|------------------------|-----------------|-----------------|
| Time of pulping        | 10 minutes      | 40 minutes      |
| Mould B temperature    | 20 °C           | 120 °C          |
| Moulds C-D temperature | 220 °C          | 250 °C          |
| Dehydration time       | 10 seconds      | 30 seconds      |
| Drying time moulds C-D | 0 second        | 10 seconds      |
| Pressing time moulds C-D | 15 seconds   | 45 seconds      |

2.2. Design of experiment

Taguchi methods were used to optimize and reduce the number of experiments without neglecting the experimental possibilities. Table 2 shows the experimental card used and the parameters that were tested in the manufacturing process as well as the levels chosen to analyze the manufacturing process. In order to study 6 parameters with 2 levels and 1 interaction, the L8 Taguchi’s matrix is used. The pulping time was the most restraining parameter on the production time and was affected to the first column of the table. Other parameters had roughly the same influence on the process time. No preference was attributed and one interaction was analyzed. We have chosen the interaction between Moulds C-D temperature and dehydration time.

In order to correctly compare the factors tested and their influence on the analyzed properties, their effect was calculated following the Equation 1 for the level 1 of each factor and Equation 2 for level 2 of
each factor. $Y_t$ is the average result of all the results for the analyzed factor. The index $k$ is the studied parameter. 1 and 2 correspond to the level of the parameter (in the studied design of experiment, level 1 corresponds to the lower value of the parameter and level 2 to the higher value).

| Test n° | Pulping time | Mould B Temperature | Moulds C-D Temperature | Dehydration time | Drying time moulds C-D | Pressing time moulds C-D | Result |
|---------|--------------|---------------------|------------------------|-----------------|------------------------|--------------------------|--------|
| 1       | 10           | 20                  | 220                    | 10              | 0                      | 15                       | Y1     |
| 2       | 10           | 20                  | 220                    | 30              | 10                     | 45                       | Y2     |
| 3       | 10           | 120                 | 250                    | 10              | 0                      | 45                       | Y3     |
| 4       | 10           | 120                 | 250                    | 30              | 10                     | 15                       | Y4     |
| 5       | 40           | 20                  | 250                    | 10              | 10                     | 15                       | Y5     |
| 6       | 40           | 20                  | 250                    | 30              | 0                      | 45                       | Y6     |
| 7       | 40           | 120                 | 220                    | 10              | 10                     | 45                       | Y7     |
| 8       | 40           | 120                 | 220                    | 30              | 0                      | 15                       | Y8     |

All experiments are done 5 times to allow the optimization of the robustness of the model developed for the process. At the end, a confirmation test is done. In this study, we used the tests that are supposed to optimize the mechanical properties and the water uptake.

\[
E_{1,k} = \frac{\text{Sum of the results, } Y_t \text{ for which the parameter } k \text{ has a level of 1}}{\text{Number of results considered in the above sum}} - Y_t \quad [1]
\]

\[
E_{2,k} = \frac{\text{Sum of the results, } Y_t \text{ for which the parameter } k \text{ has a level of 2}}{\text{Number of results considered in the above sum}} - Y_t \quad [2]
\]

Once we obtained the theoretical results with the help of Taguchi method, we made MPPs using process parameters that theoretically gave higher mechanical properties and lower water uptake and compared with the results obtained experimentally. For this comparison, tests were done with samples immersed in distilled water for 1 hour at 23 °C (± 2°C). Samples were weighed once prior to their test conditioning and then after being taken out of water and slightly wiped, then tensile testing was done. This comparison allowed us to test the Taguchi table’s efficiency and conclude on the theoretical results given.

2.3. Characterization techniques of Molded Pulp products

Tensile tests were made using an MTS 10kN machine with a load cell of 250N, at a speed of 1 mm/min. At least 5 samples were done for each condition tested. A shouldered testing bar shaped punch cutter was used to obtain the testing samples from the 3D product with the desired shape and size as shown in Figure 2 and with dimensions given in NF EN ISO 527-2 by specimen 5A type [8]. With this
cutting method, the samples obtained had exactly the same shape, thus reducing the possible errors in length or width measures. However, the thickness could be different and was measured on all samples prior to their testing.

Initial tensile tests were carried out on unmodified samples. For the analysis of sorption behavior, samples were kept in desiccators at a specific water activity ($a_w$) and at 23°C ($\pm$ 2°C), as shown in Table 3. Samples were weighed at their initial condition, then oven dried at 105°C for 48h, weighed again and put in controlled humidity chambers at 23 °C ($\pm$ 2°C) until equilibrium was reached. Once sorption equilibrium was reached, samples were once again weighed and tensile tests were performed.

![3D model of the food tray used for the study](image)
![Sample cutting zones used for the base and edge of the food tray](image)

Figure 2 : (a) 3D model of the food tray used for the study, (b) sample cutting zones used for the base and edge of the food tray

The samples’ water uptake was calculated with the following formula:

$$\tau \ (%) = \frac{W_f - W_o}{W_o} \times 100$$  \hspace{1cm} [3]

Where $W_f$ is the sample’s weight after sorption test and $W_o$ the dry sample’s weight.

**Table 3: $a_w$ of saturated salts used to test sorption behavior of MPPs**

| $a_w$  | 0.10 | 0.33 | 0.50 | 0.75 | 0.98 |
|--------|------|------|------|------|------|
| Saturated salt | KOH | MgCl2 | Climatic chamber | NaCl | K2SO4 |

GAB model was used to better understand the sorption behavior of the materials tested as the Equation 4 shows. This model is an evolution of BET model made successively by Anderson [9], De Boer [10] and Guggenheim [11] to improve the isotherm for high relative pressure ($a_w$ close to 1).

Sorption analysis are often made on food products and living substances [12,13] as their weight depends on the relative humidity in air. Parker et al [14] made a review on the sorption studies previously done on cellulose, paper and pulp and shows that GAB model gives accurate results when compared to experimental results. Bedane et al [15] studied the paper modification with additives and coating on the moisture sorption and showed that GAB equation is effective to model the paper’s sorption behavior.
It is interesting to analyze the effect of relative humidity (or \( a_w \)) on their weight and mechanical properties. \( \tau \) is the weight gain after test and \( a_w \) is the water activity in the desiccator. \( K, C \) and \( \tau_m \) are variables that are specific to each sample tested and may be obtained using solver parameter in excel. \( \tau_m \) is the water content adsorbed on the first layer, also known as the monolayer moisture content. \( C \) is a constant related to the chemical potential’s difference of the sorbate molecule (water for this study) in the upper layer and the monolayer whereas \( K \) is a constant related to the multilayers’ heat properties [16]. These 2 constants vary with the temperature. \( C \) gives an indication on the isotherm type given by IUPAC and if \( 0 > C > 2 \), the isotherm is type III whereas if \( C > 2 \) it is a type II isotherm [17].

\[
\tau (\%) = \frac{\tau_m \times C \times K \times a_w}{(1 - K \times a_w) \times (1 + (C - 1) \times K \times a_w)} \quad [\text {4}]
\]

Young Modulus was calculated using the test results following the Equation 5. \( S_{\text{max}} \) is the maximum slope of the force - elongation curve (with the force given in N, and the elongation given in mm/mm), \( l \) is the initial length of the sample in mm, \( b \) is the sample width in mm and \( t \) is the sample thickness in mm.

\[
E (\text{MPa}) = \frac{S_{\text{max}} \times l}{b \times t} \quad [\text {5}]
\]

3. Results and discussions

3.1. The influence of process parameters on the sorption properties of MPPs

The resulting effects of each processing parameter tested for the sorption behavior is described in Table 4, the effects improve the property when it is negative since the objective is to limit the water uptake. We observe that for all \( a_w \) tested, the effects of each factor are all less than 1 which means, they have a small impact on the weight gain of the samples tested. However, when we observe which level of each parameter has the impact limiting the weight gain, we note that they are different depending on the process parameter observed and the \( a_w \) tested.

A longer pulping time gives a lower water uptake for \( a_w \) of 0.33, 0.5 and 0.75 with effects closer to zero as opposed to the effects for 0.10 and 0.98 where the water uptake is limited for shorter pulping time. Varying the pulping time may have an influence on the morphology of cellulose fibers with a possible reduction of fibers length and modification of surface fibrillation. Chen et al [18] studied the influence of beating on the properties of cellulose fibers and they observed that a longer pulping time...
results in a higher water retention value due to a higher swelling ability of the pulp as well as a lower degree of crystallinity.

Table 4: Resulting effect for each factor tested on water uptake of samples after sorption

| $A_w$ Level | Pulping time | Mouuld B temperature | Moulds C-D temperature | Dehydration time | Drying time moulds C-D | Pressing time moulds C-D |
|-------------|--------------|-----------------------|-------------------------|-----------------|-----------------------|-------------------------|
| 0.10        | -0.31        | -0.20                 | 0.03                    | -0.05           | 0.07                  | -0.04                   |
| 0.33        | 0.31         | 0.20                  | -0.03                   | 0.05            | -0.07                 | 0.04                    |
| 0.50        | 0.08         | -0.08                 | 0.17                    | 0.004           | 0.07                  | 0.12                    |
| 0.75        | -0.08        | 0.08                  | -0.17                   | -0.004          | -0.07                 | -0.12                   |
| 0.98        | 0.003        | -0.10                 | 0.22                    | -0.06           | 0.06                  | 0.05                    |
| 0.75        | -0.003       | 0.10                  | -0.22                   | 0.06            | -0.06                 | -0.05                   |
| 0.98        | -0.07        | -0.19                 | -0.05                   | -0.13           | -0.10                 | -0.11                   |
| 0.98        | 0.28         | 0.52                  | -0.36                   | 0.29            | -0.35                 | 0.16                    |
| 0.98        | 0.28         | -0.52                 | 0.36                    | -0.29           | 0.35                  | -0.16                   |

A shorter pulping time allows the fiber to maintain their degree of crystallinity and thus a lower swelling ability and lower water uptake. In their study, Chen et al. [18] also showed that when a longer beating time is applied to cellulose fiber, the pore size and volume are increased. Park et al. [19] studied the influence of pore size on cellulose fibers' drying properties and observed that a higher sample's moisture content is explained by the presence of wider pores. Beg et Al. [20] studied the influence of beating time on fibers morphology and showed that a longer pulping time shortens the fibers' length.

The mould B temperature has a better impact at lower temperature for $a_w$ of 0.1 to 0.5, however, the effect is better at higher temperature for $a_w$ of 0.75 and 0.98. For the moulds C and D temperature, we observe that the effect is optimized at higher temperature for all $a_w$ expect 0.98 where a lower moulds' temperature is more effective. As the temperature of the moulds C and D is applied at the same time as pressure, the drying phenomena of the MPP is different from the mould B where only temperature is applied. The MPP is in contact with mould B for about 10 seconds, the product is not dry when it is transferred to the mould C. However, this short contact time with the mould B at 120°C allows the finished product to have a limited water uptake at high $a_w$.

Several studies [21,22] show that the Water retention value of cellulose pulp is lower when a higher temperature is applied to dry the product. A higher process temperature limits the ability of cellulose fibers to absorb water molecules. Chen et Al [21] also observed that a higher drying temperature increases the amount of lactones produced which can be translated by a lower swelling ability of the dried fiber. Norgren et al. [23] studied the influence of temperature on the density of produced materials
and observed that a higher temperature increases the product’s density. With a higher density, they observed that the tensile strength is also improved. Marta et al [24] studied the influence of drying temperature on cellulose properties and observed that a higher drying temperature resulted in a lower water content value translated by a fiber hornification thus reducing the available water adsorption sites and swelling capability.

The drying time of moulds C and D is a process step during which no pressure is applied to the MPP as opposed to the pressing time of moulds C and D where a pressure of 40 MPa is applied. For low \(a_w\) of 0.1 to 0.5, a longer drying time allows the MPP to reduce its water uptake. For 0.75 and 0.98 \(a_w\), a shorter drying time is preferred to limit the water uptake. This difference observed between low \(aw\) and high \(aw\) may be explained by the monolayer and multilayer specific adsorption behaviors.

The pressing time of moulds C and D has more impact at 15 seconds for \(a_w\) of 0.10 and 0.75 whereas a longer pressing time has a better impact on limiting the water uptake for all other \(a_w\) tested.

Figueiredo et al [25] tested the effect of pressure on the dried morphology of pulps and showed that a higher pressure applied to the sample increases the crystallinity of cellulose fibers. They also showed that it allows the fiber to have a higher swelling recovery translated by a higher water retention value and better flexibility compared to fibers that are not submitted to high pressure. Applying a pressure for a longer time on MPPs increases the water uptake, as observed in our tests with a longer pressing time on moulds C and D.

Hunt et al. [26] studied the influence of pressure on cellulose fibers properties and observed that a drying in a hot press strongly limits the material’s shrinkage as well as reduces the product’s thickness and water content. Joelsson et al. [27] observed that a higher processing temperature and a higher pressure increase the material’s density, thus resulting in a lower pores size. Reducing the pores size limits the water uptake and fiber swelling while increasing the cellulose’s degree of crystallinity.

The analyzed factors had different effects depending on the \(a_w\) tested as the resulting effects (level 1 or level 2) varied when \(a_w\) was modified. In order to better understand the influence of the factors on the water uptake after sorption analysis, the GAB equation was used. When modifying the variables in the GAB equation, we can observe that depending on the variable analyzed, the resulting slope will either have a high or a low shift as shown in Figure 3. The control slope has the variables set as followed: \(C = 10, K = 0.85\) and \(\tau = 4\).
When C is varied between 5 and 15 the resulting slope is close to the control with a slight modification at lower \(a_w\), showing that C is a factor translating the water uptake for \(a_w\) between 0 and 0.5. The variation of K between 0.8 and 0.9 shows bigger modifications in the slope as we observe a similar slope to the control for lower \(a_w\). However, when \(a_w\) is higher than 0.6, we can observe that the slope with \(K=0.9\) diverges from the control and results in a higher water uptake (39% at 1 compared to 25% for the control slope). This variable translates the behavior of the water multilayer on the cellulose fibers surface. Bedane et al [15] showed that the sorption slope is different when comparing the moisture content on the monolayer and the multilayer. They observed that a lower monolayer content is preferred to maintain the product’s optimized properties.

With a higher K, the cellulose fiber is better able to adsorb water molecules thus increasing the multilayer and water uptake. A lower K indicates that the product is less capable of adsorbing water molecules thus limiting the water uptake at higher \(a_w\). \(\tau_m\) is the product’s monolayer uptake, above this water content, the bilayer is created.

With a lower \(\tau_m\), we observe that the slope is lower for all \(a_w\), indicating that with a lower monolayer limit the multilayer is also limited. This may be explained with higher inaccessible sites for water molecules to be adsorbed on thus reducing the product’s water uptake for both the monolayer and the multilayers.

The variables \(\tau_m\), C and K were analyzed to understand the influence of the process parameters on the global sorption behavior for all water activities as shown in Table 5.
Table 5: Effect of process parameters on GAB model Variables $\tau_m$, C and K

| Level | Pulping time | Mould B temperature | Moulds C-D temperature | Dehydration time | Drying time moulds C-D | Pressing time moulds C-D |
|-------|--------------|----------------------|-------------------------|------------------|-------------------------|-------------------------|
| 1     | 0.20         | 0.07                 | 0.14                    | 0.07             | -0.06                   | -0.001                  |
| 2     | -0.20        | -0.07                | -0.14                   | -0.07            | 0.06                    | 0.001                   |
| C     | -8.00        | -2.51                | 1.10                    | -5.02            | 5.20                    | -0.16                   |
| 1     | 8.00         | 2.51                 | -1.10                   | 5.02             | -5.20                   | 0.16                    |
| 2     | -0.005       | -0.001               | -0.006                  | -0.005           | 0.004                   | -0.001                  |
| K     | 0.005        | 0.001                | 0.006                   | 0.005            | -0.004                  | 0.001                   |

When comparing the results of the process effects (Table 5) and the slope modification (Figure 3), we can observe that in order for C to have a higher effect result on the GAB equation modification compared to K and $\tau_m$, it needs to be a higher number. It means that a small modification of K and/or $\tau_m$ has a much higher impact on the equation than C. Moreover, each constant has a different effect depending on the parameter tested. In order to limit the water uptake, C, K and $\tau_m$ must have the lowest result.

A lower drying and pressing time in moulds C and D and higher temperatures (moulds B, C and D), pulping time and dehydration time lower $\tau_m$ result. When $\tau_m$ is reduced, the cellulose-water monolayer is filled at a lower $a_w$. This may be explained by a limited access to cellulose surfaces with the presence of smaller pores or a lower cellulose surface fibrillation. As there is less availability for water molecules to be fixed to cellulose, its’ monolayer is more rapidly filled. With a lower $\tau_m$ the resulting MPPs are then less hydrophilic, thus reducing the global water uptake for all $a_w$.

When the pulping time, dehydration time, pressing time and mould B temperature are at level 1, and Moulds C-D temperatures and drying time are at level 2, C is reduced. This may be explained by a higher degree of crystallinity as several studies [21,28] showed that a higher crystallinity results in a lower water uptake. The cellulose amorphous phase is more hydrophilic with more available sites when compared to crystalline phase.

In order for K to be lower, all process parameters were at level 1 except for the drying time of moulds C and D. As K is a factor for multilayers and mostly impacts the equation at higher $a_w$ [15,16], this lower result shows that for the given parameters the water vapor adsorption saturation may be reached at a lower water content. One explanation can be the reduction of pores size with the specific process parameters chosen.
This morphology modification given by the analysis of the process parameters allows us to infer that the mechanical properties of the MPPs may also be changed and optimized with the correct choice of parameters.

### 3.2. Mechanical properties depending on the process parameters

The initial tensile tests were performed to compare the influence of the MPPs production parameters without adding any other specific condition to the finished products. These results were compared in Table 6 with the tensile tests done after water sorption with $a_w$ at 0.98 as it is the state in which MPPs had the lower mechanical property over all the $a_w$ spectrum.

**Table 6: Resulting effect for each factor tested on the mechanical properties of samples**

| test  | Level | Pulping time | Mould B temperature | Moulds C-D temperature | Dehydration time | Drying time moulds C-D | Pressing time moulds C-D |
|-------|-------|--------------|---------------------|------------------------|-----------------|------------------------|--------------------------|
| Initial | 1     | 48.22        | -45.12              | -31.09                 | -88.08         | 17.02                  | -40.08                   |
|        | 2     | -48.22       | 45.12               | 31.09                  | 88.98          | -17.02                 | 40.08                    |
| $a_w$ 0.98 | 1     | -35.90       | -49.86              | 11.05                  | -18.70         | 0.64                   | -0.10                    |
|        | 2     | 35.90        | 49.86               | -11.05                 | 18.70          | -0.64                  | 0.10                     |

The initial mechanical test, without any specific condition given to the samples prior to their testing, shows that the process parameters tested have an impact on the resulting MPPs’ Young Modulus. A lower amount and size of pores as well as longer fibers given by a shorter pulping time is beneficial for a higher Young’s Modulus as observed by several studies \cite{18,29} with a resulting higher tensile index. A higher mould B temperature improves the MPP’s Young’s Modulus for both initial and after water sorption. Several studies \cite{23,24,27} observed that a higher processing temperature allows the tensile index to be improved as it allows to have a lower WTR and swelling capacity \cite{21,24,30}.

Moreover, an even higher temperature used decreases the mechanical properties as given by the results in the drying time of moulds C and D as well as the moulds C and D temperature for the sorption analysis. Several Studies \cite{24,28,31} observed that a high drying temperature reduces the paper breaking length and tensile strength. However, when pressure is applied in combination with high temperature, the cellulose fibers show a different behavior with a higher Young’s Modulus for a longer pressing time on moulds C and D. It has been shown \cite{23,27,32} that a high pressure and high temperature drying improve the mechanical properties with higher tensile strength as the average pore size is reduced and the density improved. It was also observed that the crystallinity is improved with high pressure and high temperature, which is translated by a lower water uptake and higher mechanical properties.
These mechanical and hygroscopic results show that with a specific manufacturing process, we may obtain MPPs with a lower water uptake and higher mechanical properties. As only 8 tests were done on the 64 possible, 2 more tests were performed to confirm the theoretical results given by the Taguchi table used.

### 3.3. Process manufacturing optimization to improve MPPs properties

The Taguchi table allowed us to obtain theoretical results on the water uptake and mechanical properties tested experimentally. With these results, 2 process parameters gave good results with a limited water uptake and higher mechanical properties. We decided to make MPPs following the specific process parameters given in Table 7.

**Table 7: Optimized process parameters from Taguchi model**

| Test n° | Pulping time | Mould B Temperature | Moulds C-D Temperature | Dehydration time | Drying time moulds C-D | Pressing time moulds C-D |
|---------|--------------|---------------------|------------------------|-----------------|------------------------|-------------------------|
| 9       | 40           | 120                 | 220                    | 30              | 0                      | 45                      |
| 10      | 40           | 120                 | 250                    | 30              | 0                      | 45                      |

We then tested the samples and compared the theoretical results with the experimental results, shown in Figure 4. We observe that for test n°9, the experimental results are better than the theory, with a lower water uptake and a higher Young’s Modulus in both initial state and after 1h water immersion. For the test n°10, the initial Young’s Modulus is higher than the theory, however the water uptake and Young’s Modulus after immersion are not better than the theory even if the results are close.

With experimental results close to the theory for tests 9 and 10, we infer that the Taguchi Table gives conclusive results and allows us to rapidly and efficiently improve the MPPs processing with specific process manufacturing parameters. With the results of these 2 tests, we were able to obtain improved mechanical and hygroscopic properties on the MPPs made with bleached CTMP. When comparing the results of the tests 9 and 10 with the first 8 tests, we observe an optimization in the water uptake and initial mechanical properties. We obtain an average decrease of the water uptake of 15% for test 9 and over 78% for test 10 whereas the overall Young’s Modulus after 1h of water immersion is improved by 19% for test 9 and 58% for test 10. These specific process parameters used together efficiently improve the hygroscopic and mechanical properties of the MPPs made.
4. Conclusion

This study allowed us to optimize the hygroscopic and mechanical properties of BCTMP without using additives and by optimizing the manufacturing process.

We observed that pulping lowers the cellulose crystallinity thus increasing the swelling and WTR of cellulose fibers. With higher moulds’ temperature, we showed that Swelling and WTR decreased as crystallinity increased and we also obtained lower pore size. When pressure is applied, the product’s density increased and pore size reduced.

As a result, these modifications in the samples morphology either improved or decreased the mechanical properties. We were able to further understand the MPPs sorption behavior and improved the water uptake with the help of Taguchi table and GAB model. Depending on the process parameters analyzed, the GAB variables could be modified thus reducing the water uptake after sorption test.
It was interesting to analyze the process parameters by making optimized tests. These tests showed that with specific parameters, we were able to further improve the hygroscopic and mechanical properties of the MPPs. With test n°10 the water uptake was on average 78% lower than all 8 tests made with the Taguchi table as well as a Young's modulus after 1h immersion 58% higher.

**Abbreviations**

CTMP: Chemi-Thermo-Mechanical Pulp, BCTMP: Bleached Chemi-Thermo-Mechanical Pulp, MPP: Molded Pulp Product, $a_w$: water activity, $\tau_m$: Water uptake in the monolayer, E: Young Modulus

5. Declarations

**Availability of data and materials**

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request

**Competing interests**

The authors declare that they have no competing interests

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**Authors’ contributions**

Claire Dislaire carried out the work and drafted the original manuscript. Yves Grohens is the supervisor of the PhD thesis, and Bastien Seantier is the co-supervisor of the PhD thesis. The authors read and approved the final manuscript.

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