Research article

Challenges in computational materials modelling and simulation: A case-study to predict tissue paper properties

Flávia P. Morais a,*, Joana M.R. Curto a, b

a Fiber Materials and Environmental Technologies (FibEnTech-UBI), Universidade da Beira Interior, R. Marquês de D’ Ávila e Bolama, 6201-001, Covilhã, Portugal
b Chemical Process Engineering and Forest Products Research Centre (CIEPQPF), University of Coimbra, R. Sílvio Lima, Polo II, 3004-531, Coimbra, Portugal

ARTICLE INFO

Keywords:
3D fiber-based simulator
Artificial neural network
Multiple linear regression
Tissue functional properties
Tissue paper materials

ABSTRACT

The growing demand for tissue papers worldwide encourages the paper industry to find new approaches to optimize the raw materials furnish management, and simultaneously to improve tissue paper performance. Softness, strength, and absorption are the key tissue properties that enhance the attention of both industry and consumers. Fiber morphology, fiber modification process steps, and structural properties affect these functional properties, and, therefore, the efforts to evaluate them and establish the relationship or models that describe them constitute a multifactorial challenge. For this purpose, we aimed to investigate the trade-off between the input variables (morphological, suspension, and structural properties) and the final properties. Key variables like the type of furnish raw materials, including the fiber mixture, mechanical and enzymatic treatments, additives incorporation, and the type of industrial base tissue papers were taken under consideration. To achieve these relationships, we used different data-driven modeling approaches including multiple linear regression (MLR), artificial neural networks (ANN), and a 3D fiber-based simulator. The MLR and ANN models were built by data collected from an experimental design, and isotropic laboratory structures were prepared and tested for changes in structural and functional properties. Moreover, a 3D fiber-based simulator was used to investigate the influence of fibers on structural properties. These results indicated that the realistic predictions enabled us to link fiber and tissue structure characteristics. In conclusion, this work has revealed that this computational modeling approach can be used to model the effect of fiber pulps parameters with final end-use tissue properties, allowing to design innovative tissue products.

1. Introduction

The identification of challenges and improvements in the field of material computer science, in particular its applications to structured materials, such as the case of tissue papers, constitutes the motivation to this work. The need within this industry for methods for the rational analysis of the fiber materials’ behavior has been emphasized, and meaningful work has been done in the modeling of fiber materials. The morphology of fiber-based materials has a deep impact on their functional properties, therefore detailed structural and morphological investigations are needed [1]. The analysis of the fiber properties with an impact on the tissue properties is complex since it presents a series of dependencies between them. Not only the raw material but also the conditions of the overall process determine the suitability of the fibers to produce tissue products. Moreover, the tissue’s final end-use properties are dependent on the final application. All these tissue properties are fundamental to obtain a product of high quality. The main limitation in the prediction of tissue paper properties is the absence of mathematical descriptors to describe the trade-off between fiber features and materials properties. The possibility of having computational tools with predictive capacity for these properties would be essential to define which are the key pulp fiber parameters that influence the type of tissue paper desired [2, 3, 4, 5].

1.1. Mathematical and computational models to predict paper properties

With the development of several computer-based papermaking simulations, the impact of fiber characteristics on paper properties has been a focus of research interest [6, 7, 8, 9, 10]. Computational simulations of paper structures predict and determine the structural and mechanical properties according to the constituent fibers, which are deposited independently and randomly, and the structure of the fibrous...
Different computational models of tissue materials predicted a non-linear relationship between sheet basis-weight and thickness, with higher density variation [14, 15]. Furthermore, many investigations have emerged to predict the softness, strength, and absorption properties of tissue products [16, 17, 18]; however, the different authors have not proposed any mathematical model that relates the fiber morphological properties with these functional properties. Page (1969) proposed a general mathematical equation [19] that assesses the relative bond strength compared to the fiber strength, describing the strength properties of paper sheets with uniform random fiber orientation, such as tensile index (T), as follows in Eq. (1),

\[
\frac{1}{T} = \frac{9}{8Z} + \frac{12 \cdot g \cdot C}{P \cdot l \cdot b \cdot RBA}
\]  

where the contribution of fibers is described by the zero-span tensile index (Z), and the contribution of bonding is merged into a gravitational constant (g), fiber coarseness (C), fiber perimeter (P), fiber length (l), fiber-fiber bond strength (b), and relative bonded area (RBA). At a low level of bonding (low Page RBA), bonding is the controlling factor for strength, as is the case of tissue paper materials. Therefore, when fiber strength is higher than bond strength, the assumption approaches the value where fibers do not fracture (9/8Z = 0) [20]. However, some of these parameters are difficult to determine due to the complexity of analyzing highly nonlinear problems. For this reason, over the years, data-driven models have been considered alternatives to forecast and optimize advanced processes. Examples of these modeling methods are multiple linear regressions (MLR) and artificial neural networks (ANN).

MLR model’s application has demonstrated great benefit in the pulp and paper industry. Rיסin, Hultén, and Paulsson (2004) reported that network strength is influenced by fiber length, lignin content, specific surface area, and total charge, for a given pulp [21]. Another approach proposed by Chagaev and Zou (2007) proved that strength development during mechanical pulp refining can be monitored through indexes based on the fine content/coarse fibers ratio, considering pulp quality and fiber development [22]. The relationship between the fiber pulps’ morphological and chemical characteristics and the paper’s structural and mechanical properties has been constantly investigated using these mathematical models, concluding that the quantitative assessment of different types of fibers for paper applications is effective through theoretical-experimental methods [23, 24, 25, 26]. Similarly, the application of ANN models in the paper industry has been investigated [27, 28, 29]. Ciesielski and Olejnik (2014) proved that an additional ANN approach can be applied in a quality control system in a paper mill, predicting different properties including apparent density, breaking length, tear strength, water retention value (WRV), fine content, and fiber length [30]. Additionally, Almonti et al. (2019) also developed an ANN model to optimize a fiber refining system. It was found that additives’ incorporation and process parameters can be optimized through fiber length prediction, in order to obtain detailed properties of the final end-used products [31]. Overall, a neural network provides a predictive model, but not a fundamental understanding of the underlying process mechanism that produced the data, which is not superior to the prediction obtained from a well-designed MLR study [32]. Neural networks can be a complement to the statistical tools of regression analysis to model the effects of fibers on paper parameters [33, 34].

1.2. Tissue paper materials

Tissue paper materials are lightweight creped papers used for cleaning, hygiene, and cosmetic purposes, including facial papers, toilet papers, towel papers, napkins, diapers, facial masks, among others. The properties required for these materials, such as softness, strength, and absorption, depend on fiber properties and their relationship with the process [35]. The tissue materials production requires several steps that can affect the tissue’s key properties, such as the selection of raw materials, the method and conditions of pulps used, the refining degrees, the pulp enzyme performance, and the amount of additives added [36, 37, 38, 39, 40, 41, 42, 43, 44].

1.2.1. Tissue paper materials requirements

Softness is one of the most important tissue paper properties, in which the perception of this property varies from individual to individual. However, this characteristic is quite subjective and complex, combining several parameters: superficial softness which is related to the smoothness felt when the product touches the skin; bulk softness related to the perception of softness when wrinkling and denting the paper; and flexibility, which is the softness associated with paper malleability. These three parameters are influenced by bulk, flexibility, and rigidity properties [48]. Therefore, the properties of the fibers can influence the softness of the final products depending on the type of fiber (eucalyptus and softwood fibers), the pulp production process, and, consequently, fiber properties, such as coarseness, curl, kinks, fine content, among others [35].

Tensile strength is an important feature for tissue papers, as it maintains the tissue sheet integrity, and ensures the paper machine runnability. This property is influenced by the fiber intrinsic strength and the strength of the inter-fiber bonds because when more bonds are present, more energy is needed to separate these bonds [17]. The mechanical properties of tissue paper sheets can be developed by optimizing the mechanical and/or enzymatic refining, additives incorporation, and changing stocks. The pulp refining improves the inter-fiber bonds and, consequently, the strength properties; however, this treatment can result in fiber damage [36]. These treatments promote an increase in fines content, fiber flexibility, and collapsibility, enhancing the inter-fiber bonds, and consequently the strengths [17, 49].

Water absorption is related to the water absorption capacity and absorption speed, which are influenced by similar properties as raw materials, type of fiber network structure, stock preparation, fine content, bulk, sheet formation, pressing, additives, among others [50]. The absorption capacity includes the total water content that the paper can absorb in its structure, while the absorption speed consists of the time that a paper takes to absorb the water [16, 51]. Porosity is one of the most important factors in liquid absorption. A more porous structure promotes higher water absorption, as there will be more empty spaces for water interaction. The structure porosity is influenced by the size of the empty spaces, which depend on the fiber dimensions, the collapse degree, and the inter-fiber bonds [51]. On the other hand, the water capillary rise is a physical phenomenon that depends on the adhesion and cohesion forces of the water molecules. These molecules can ascend through the fiber walls or between the pores [52]. Hence, if the pores are too large, the intermolecular bonding between the fibers and the water molecules will not be sufficient to counteract the force of gravity, which impairs the capillary rise. In the case of smaller pores, water progression will be more restricted by the limited space for flow [52]. The structure bulk also influences water absorption properties. Papers with higher bulk (more open fiber structure) will present higher absorption since there will be more bonding points, wider channels in the paper structure, and less strength to flow [16, 53].

1.2.2. Effect of fiber and network characteristics on tissue paper materials

The fiber properties influence the tissue’s final end-use properties, as the type of fiber and the type of pulp production process can obtain fibers with different properties that influence the structure bulk and, consequently, the softness, strength, and absorption properties. Bulk is improved by thicker fibers, with more resistance to collapse. Rigid fibers promote more porous and open structures, with less contact area and bonding between fibers. In contrast, fibers with a higher width and less thickness are more susceptible to collapse, resulting in denser structures, with less bulk, softness, and absorption, and more strength [35, 38, 39]. Coarseness is also related to the fiber wall thickness, diameter, and tissue sheet density. Softwood fibers with lower coarseness present better
formation, higher softness, tensile strength, absorption, less tearing, energy spent to obtain good inter-fiber bonds, among other properties [54]. Eucalyptus fibers with higher coarseness present tissue papers with higher bulk and softness, as these fibers resist to collapse and have low flexibility, enhancing the formation of bulky and soft products [35, 38, 39]. The fiber curl and kinks contribute to improve the pulp performance to produce tissue papers and appear through mechanical processes. Page et al. (1985) and Kerekes and Schell (1995) reported that higher curl indexes promote higher porosity, bulk, softness, absorption capacity, wet elongation, drainage, and tear rate. Moreover, fibers with a higher kink index negatively influence tensile strength and tearing and promote bulk development [55, 56]. The fiber length also influences the tissue's mechanical properties, enhancing the several inter-fiber bonds [17]. The inter-fiber bonds, and consequently the tissue paper strength, are also influenced by the fines content. The pulp fine content is a limiting factor in the paper quality, contributing negatively to the softness. To produce tissue papers, the use of pulps with the lowest possible fine content should be preferred, to avoid increasing the pulp consistency and drainage difficulties [57]. Additionally, the suspension properties are often used to monitor fiber changes by refining or enzymatic treatments. The \( SR \) is indicative of the water drainage capacity by the suspension pulp, which influences the formation, pressing, and drying of the paper sheet. The pulp drainability depends on the fiber strength to the water flow, which increases with refining, due to fiber fibrillation and fines content [40].

As the optimization of tissue paper materials requires the use of several computational tools, our approach is to combine experimental and computational planning to model the fibers and establish relationships between fiber and structural properties, process modification steps, and functional tissue properties. To the best of our knowledge, the development of models for tissue paper materials based on the influence of pulp fiber and structure characteristics for different process steps has not been reported in the literature. The main goal of this work is to present an innovative strategy for computer materials simulation and to identify the main achievements and challenges. The strategy to predict softness, strength, and absorption properties of several process steps, such as fiber furnish, mechanical and enzymatic treatments, and additives incorporation, uses a combination of computational tools, including MLR, ANN, and a 3D fiber-based simulator. The models were built by data collected from fiber morphological, suspension, and paper structural properties. The 3D fiber-based simulator, implemented and validated by Conceição et al. (2010) and Curto et al. (2011) [7,8], was used to model the fibers, obtaining 3D structural computational simulations with the predictive capability of the tissue paper structural properties.

2. Experimental

2.1. Materials

A wide range of samples from our previous works [38, 39, 40, 41, 42, 45, 46] was selected for the present study. These samples include eucalyptus pulps and softwood pulps, which were subjected to mechanical and enzymatic treatments. These industrial pulp samples present different cooking and bleaching histories. Nano/microfibrillated cellulose (NMFC) [41], a biopolymer designed to substitute cationic starch [42], and carboxymethylcellulose (CMC) were used as additives. A set of industrial base tissue paper samples were also selected for the studies, using data from ours and other previous studies [47]. These samples included industrial reels, toilet papers, paper towels, and napkins.

2.2. Methods

The fiber furnishes included different mixtures of eucalyptus fibers and softwood fibers, in different percentages (0–100%). More detailed information about these studies can be found in [41, 42].

Regarding mechanical treatments, different eucalyptus pulps were beaten in a PFI mill at different revolutions, under two refining intensities of 3.33 and 1.67 N/mm. To accomplish the enzymatic processes, eucalyptus fiber pulps were treated, using enzyme dosages between 10 and 100 g per ton of pulp, for 30 and 60 min. The assays were carried out at a consistency of 4%, pH 7, and 40 °C, with continuous mechanical agitation using an impeller. More detailed information about these studies can be found in [40]. Additionally, eucalyptus fiber pulps were also subjected to enzymatic and mechanical combination treatments. In these assays, enzyme doses between 0.25 and 1 kg per ton of pulp were used, from 30 to 120 min, with the same process conditions mentioned above. Subsequently, these pulps were beaten in a PFI mill at different revolutions, under a refining intensity of 3.33 N/mm. A pre-refining process followed by enzymatic treatment was also carried out under the same conditions.

The additives incorporation was performed with NMFC, at dosages between 1 to 10 %, a biopolymer, at 2%, and CMC, at 5%, in different fiber furnish formulations consisted of eucalyptus and softwood pulps. More detailed information about these studies can be found in [41, 42].

Each sample was analyzed for morphological properties using MorFi Fiber Analyzer (TECHPAP, Grenoble, France). The morphological characteristics evaluated were fiber length weighted by length, fiber width, coarseness, kinks, curls, and fine elements.

Additionally, the drainability suspension properties of each sample were also measured according to the Schopper-Riegler degree (‘SR’) method (ISO 5267-1).

Isotropic laboratory structures of 20 g/m² were prepared without pressing, according to an adaptation of ISO 5269-1. These structures were air-dried under atmospheric conditions, preconditioned at (23 ± 1) °C at a relative humidity of (50 ± 2) %, and evaluated for structural properties, such as bulk (ISO 12625-3), and porosity, according to the equation: Porosity = 1 – (density of the sheet/density of cellulose), and functional tissue properties, such as softness HF (calculated considering softness, smoothness and stiffness parameters with basis weight and thickness) and softness TS7 (superficial softness influenced by the presence of fibers in the Z-direction on paper) using Tissue Softness Analyzer (TSA Emtec) equipment, tensile index (ISO 12625-4), water absorption capacity (ISO 12625-8 adaptation), and capillary rise (ISO 8787 adaptation). The range of values for morphological, suspension, structural and functional tissue properties, for the furnish process and industrial base tissue papers, can be found in the supplementary data file (Tables S1).

3. Computational modeling

3.1. Multiple linear regression (MLR)

MLR modeling was used to predict the functional tissue properties of structures made from furnish process and industrial base tissue papers. Statistical software IBM SPSS Statistics 25 (Armonk, NY, USA) was used for MLR analysis. Tissue properties of softness HF, softness TS7, tensile index, water absorption capacity, and capillary rise were used as dependent variables (output variables, \( Y \)), and the morphological, suspension, and structural properties as independent variables (input variables, \( X \), as shown in Table 1. Additionally, the backward elimination procedure was used to build the models, since this method rejects statistically insignificant terms in the model. Only statistically significant linear regression equations (ANOVA, p-value ≤ 0.5 %) were reported.

Through a collection of statistical techniques, MLR allows building different empirical models needed in response surface methodology. In general, these models fit a linear equation to the experimental data, relating different independent variables with a dependent variable. Considering a set of \( n \) independent variables, namely \( x_1, x_2, ..., x_n \), associated with a value of the dependent variable \( y \), the linear regression model that describes this relationship is seen in Eq. (2),
Table 1. Dependent and independent variables used for MLR analysis.

| Dependent Variables (Outputs)          |          |
|----------------------------------------|----------|
| Y1                                     | Softness HF |
| Y2                                     | Softness TS7 |
| Y3                                     | Tensile Index (N.m/g) |
| Y4                                     | Water Absorption Capacity (g/g) |
| Y5                                     | Capillary Rise (mm at 10 min) |

| Independent Variables (Inputs)         |          |
|----------------------------------------|----------|
| X1                                     | Length weighted by length (mm) |
| X2                                     | Width (μm) |
| X3                                     | Coarseness (mg/100m) |
| X4                                     | Kinks (%) |
| X5                                     | Curl (%) |
| X6                                     | Fines content (% in length) |
| X7                                     | Schopper-Riegler degree (SR) |
| X8                                     | Bulk (cm³/g) |
| X9                                     | Porosity (%) |

Y = \beta_0 + \beta_1X_1 + \beta_2X_2 + \ldots + \beta_nX_n + \varepsilon \quad (2)

where the regression coefficients are the parameters \( \beta_j, j = 0, 1, \ldots, n \), and \( \varepsilon \) is a random error. In the \( n \)-dimensional space of the regressor variables \( (x_i) \), a hyperplane is described. When all the remaining independent variables \( x_i (i \neq j) \) are kept constant, the estimated change in response \( y \) per unit change in \( x_j \) is represented by the parameter \( \beta_j \) [32].

3.2. Artificial neural networks (ANN)

Statistical software IBM SPSS Statistics 25 (Armonk, NY, USA) was used for the configuration of the ANN models. The data of dependent variables considered (softness HF, softness TS7, tensile index, water absorption capacity, and capillary rise) were randomly divided into training (approximately 70%) and test (approximately 30%) sets. Morphological, suspension, and structural properties were used as input variables and functional tissue properties as output variables in the models.

Overall, in order to approximate the output data, ANN models perform a non-linear transformation of the input data. This modeling learns from examples of experimental data and exhibits some generalizability beyond the training data. A multilayer feedforward artificial neural network is the most commonly used ANN. Each variable in a layer, the so-called nodes, involves the input layer, which are the original predictors, the hidden layer, composed of a set of constructed variables, and the output layer, consisting of the responses. Additionally, an activation or transfer function achieves the output value of each node. This function can be presented as a sigmoid, a hyperbolic tangent, or an exponential [32]. Each of the \( n \) hidden layer nodes, \( d_i \), can be a linear combination of the input variables, as described in Eq. (3),

\[
a_i = \sum_{j=1}^{n} w_{ij}x_j + \theta_i
\]

where the \( w_{ij} \) are unknown parameters that must be estimated (weights), and \( \theta_i \) is a parameter that plays the role of an intercept in linear regression (bias node) [32].

3.3. 3D fiber-based modeling simulation

A 3D fiber-based simulator, the voxelfiber, developed, implemented, and validated by Conceição et al. (2010) and Curto et al. (2011) [7,8], was adapted and used to simulate different tissue structures with the fiber dimensions obtained experimentally. The simulator is based on a cellular automaton, in which there is a cell Cartesian division, making each fiber represented as a sequence of voxels, where each one occupies a pre-established volume [9]. This is open-source software, and the code is available at https://github.com/eduardotrincaoconceicao/voxelfiber. In general, this simulator allowed the modeling of tissue structures as planar random networks, through the fiber dimensions and properties, such as length/width ratio, fiber flexibility, fiber wall thickness, lumen thickness, and resolution [45, 46], as shown in Figure 1. The models proposed to predict the functional tissue properties were validated through the structural properties results of 3D computational modeled structures with this simulator. These computational studies were carried out using MATLAB® (R 2020a, 9.8.0.1323502, MathWorks, Natick, MA, USA).

4. Results and discussion

4.1. Multiple linear regression (MLR) analysis between morphological, suspension, and structural properties and functional properties of tissue furnish and papers

An MLR was run to predict softness HF, softness TS7, tensile index, absorption capacity, and capillary rise properties from different morphological, suspension, and structural properties, for tissue furnish process and industrial base tissue papers. All variables added statistically significantly to the prediction, \( p < 0.05 \). The higher the multiple
coefficients of determination ($R^2$) value, the better the models fit the data. The adjusted multiple coefficients of determination (adjusted $R^2$) explain how much variance in the results would be considered if the model had been derived from the samples, the population used to create the model. The higher the value, the better the adjustment data. The results of all MLR analysis can be found in the supplementary data file (Tables S2 and S3).

4.1.1. Tissue furnish process

Our experimental studies of the tissue furnish process were composed of a mixture of eucalyptus and softwood fiber pulp, fibers subjected to refining and enzymatic treatments, and additives incorporation, and their yee fiber and structure properties influence the tissue's functional properties. Therefore, the mathematical models developed can predict the softness, strength, and absorption properties of isotropic laboratory structures of 20 g/m², based on the fiber and structure modification furnish processes.

The MLR analysis showed that the width, curl, SR, and porosity variables statistically significantly predicted softness HF, $F (4, 109) = 89.677$, $p < 0.005$, $R^2 = 0.767$, adjusted $R^2 = 0.758$. The most significant MLR model developed between these variables is described in Eq. (4).

$$Y_1 = -31.616 - 2.708X_1 + 2.376X_2 - 2.365X_3 + 2.134X_4$$

(4)

Additionally, the same variables (width, curl, SR, and porosity) statistically significantly predicted softness TS7, $F (4, 109) = 94.428$, $p < 0.005$, $R^2 = 0.776$, adjusted $R^2 = 0.768$. The most significant MLR model developed between these variables is seen in Eq. (5).

$$Y_2 = 83.955 + 1.459X_1 - 1.199X_2 + 1.261X_3 - 1.272X_4$$

(5)

The softness of tissue paper sheets is achieved with the use of curly fibers, obtaining structures with higher porosity (or bulk) in which the bonding strength and bonding area among the fibers are not fully developed. Furthermore, the degree of refining expressed in SR influences this fiber bonding capacity [35].

The MLR models also reveal that the key factors which affect tensile index are all independent variables except the curl, $F (8, 105) = 47.255$, $p < 0.005$, $R^2 = 0.783$, adjusted $R^2 = 0.766$. The most significant MLR model developed between these variables is shown in Eq. (6).

$$Y_3 = -92.882 + 11.860X_1 - 1.402X_2 + 1.317X_3 - 0.404X_4 - 0.273X_5 + 0.764X_6 - 6.543X_7 + 1.802X_8$$

(6)

The strength of sheets is promoted by long fibers that present a higher capacity to form bonds with multiple fibers [35]. Coarseness is related to the fiber wall thickness, diameter, and density. Therefore, strengths are improved with the conformation/collapse degree of the fibers, and consequently, with the amount of fiber bonding. Fibers with higher diameter and smaller thickness collapse more easily, resulting in denser structures, with less bulk and improved strength. To develop this property, enzymatic treatment and/or refining processes are used, which subsequently improve the sheet formation and fiber drainability (SR), with fine element formation [36]. On the other hand, fibers with a higher kink index negatively influence tensile strength [55].

The MLR analysis showed that the width, kinks, curl, SR, bulk, and porosity variables statistically significantly predicted water absorption capacity, $F (6, 107) = 25.227$, $p < 0.005$, $R^2 = 0.586$, adjusted $R^2 = 0.563$. The most significant MLR model developed between these variables is presented in Eq. (7).

$$Y_4 = 28.473 - 0.067X_1 - 0.057X_2 + 0.316X_3 - 0.047X_4 + 1.125X_5 - 0.288X_6$$

(7)

Finally, the coarseness, curl, SR, and bulk variables statistically significantly predicted capillary rise, $F (4, 109) = 49.364$, $p < 0.005$, $R^2 = 0.644$, adjusted $R^2 = 0.631$. The most significant MLR model developed between these variables is presented in Eq. (8).

$$Y_5 = 139.284 - 2.044X_1 - 1.841X_2 - 1.500X_3 + 7.427X_4$$

(8)

The coarse, curly, and kinked fibers enhanced the formation of bulkier, porous, and absorbent sheets. The fiber and structure modification processes influence the drainability, promoting efficient fiber fibrillation and, consequently, a higher availability and accessibility for establishing water bonding and, consequently, a higher water retention capacity.

In general, the fiber deformations (curl and kinks), SR, and the structural properties of bulk and porosity were the dominating factors that influence all the tissue functional properties for the tissue furnish process.

4.1.2. Industrial base tissue papers

The quality requirements of tissue papers differ according to their purpose and consumer expectations, and the creping and converting processes are conveyed to the final consumer since these operations are encoded in the final tissue structure [35, 58, 59].

The MLR analysis between softness HF and morphological, suspension and structural properties of industrial base tissue papers have shown a non-significant correlation, and no regression model could be suggested. However, the length weighted by length, kinks, bulk, and porosity variables statistically significantly predicted softness TS7, $F (4, 18) = 3.304$, $p < 0.005$, $R^2 = 0.423$, adjusted $R^2 = 0.295$. The most significant MLR model developed between these variables is described in Eq. (9).

$$Y_2 = -161.562 + 24.248X_1 - 0.050X_4 - 1.487X_6 + 1.929X_9$$

(9)

This evidence can be explained by the different process operations used to produce each industrial base tissue paper. The creping process improves the product's softness, bulk, absorption, and stretch. During this process, the doctor blade scraps the sheet off from the Yankee dryer surface, consisting of the formation of macro and micro wrinkled on the paper web. Creping delivers the laminate layer and changes the fibers from machine direction to Z-direction, resulting in a breaking of interfiber bonding, and consequently, a higher softness and elongation capacity. Therefore, these changes that occur in each tissue paper depend on the process conditions used, which can be different in each situation, since the creping process is influenced by the sheet adherence to the Yankee, the blade geometry, and the difference between the Yankee and the paper reel speed [35, 58].

The MLR models also reveal that the key factors which affect tensile index are curl, bulk, and porosity, $F (3, 19) = 15.331$, $p < 0.005$, $R^2 = 0.708$, adjusted $R^2 = 0.661$. The most significant MLR model developed between these variables is seen in Eq. (10).

$$Y_3 = -79.336 - 1.804X_5 - 1.222X_9 + 1.287X_9$$

(10)

As reported above, collapsed fibers with less bulk, porosity, and curl, enhanced the tensile strength properties. The creping process also contributes to the tissue paper's mechanical properties, since it promotes elongation. These mechanical properties can also be influenced by raw materials, stock preparation, sheet formation, pressing, humidity content, and paper adhesion on the Yankee cylinder, and be developed by optimizing fiber modification processes and changing furnish stocks [35, 55].

The MLR analysis showed that only bulk variable statistically significantly predicted water absorption capacity, $F (1, 21) = 109.580$, $p < 0.005$, $R^2 = 0.839$, adjusted $R^2 = 0.832$. The most significant MLR model developed between these variables is shown in Eq. (11).

$$Y_4 = 5.107 + 0.426X_8$$

(11)

The creping and converting operations influence absorbency properties, enhancing a bulkier structure. The converting operations, including embossing, are performed on base tissue paper sheets to form a
single layer or multilayer finished product. Embossing patterns are applied to tissue materials to improve bulk, and consequently, absorption [59, 60].

The MLR analysis was unable to predict any model with significance for the capillary rise properties due to the data absence. In general, the fiber deformations and structural properties were the dominating factors.
that influence all the tissue functional properties for industrial base tissue papers, with creping and converting operations.

4.2. Artificial neural network (ANN) analysis between morphological, suspension and structural properties and functional properties of tissue furnish and papers

The ANN models were built by specifying the minimum (1) and maximum (50) number of units accepted in the hidden layer by default. ANN’s automatic architecture selection procedure identified the best prediction of the number of units in this layer through the hyperbolic tangent function, and in the output layer through the identity function. Morphological, suspension, and structural properties are represented as “covariates”, the predictor variables. The network training was improved with the choice of the standardized method for the covariates rescaling, since the batch training method minimizes the total error directly, on “smaller” datasets. Additionally, the stopping criteria of the network training allowed for a maximum step if the error was not further reduced. The ANN models accurately replicated the results using the same initialization value for the random number generator, the same order of data and variables, and the same procedural settings. The results of the ANN architecture analysis can be found in the supplementary data file (Tables S4 and S5). The model summary of Table 2 presents the results of the ANN training and testing for tissue furnish and papers. The relative error of each scale-dependent variable is associated to the sum-of-squares, where the predicted value for each case is the average value of the dependent variable. This relative error is defined as the error of the sum-of-squares of the dependent variable divided by the error of the sum-of-squares of the “null” model.

4.2.1. Tissue furnish process

The maximum relative error of all dependent variables (tissue functional properties) was 0.261 and the average relative error was 0.157, for the ANN training model of fiber furnish. Regarding the ANN testing model, the maximum relative error was 0.428 and the average relative error is 0.306 (Table 2). From the architectural point of view, there was a total of 9 independent variables in the input layer, 6 neurons in the hidden layer, and 5 dependent variables in the output layer (Table S4). Figure 2 shows this architecture of the ANN model for tissue furnish.

Figure 3 presents the relationship between the experimental values and predicted values for the ANN model of tissue furnish. The results indicated that the neural network prediction was very close to the measured values. The accuracy of the prediction models for functional properties was proved by the correlation coefficient values. As R²
approaches 1, prediction accuracy increases. The developed model includes a coefficient of determination of 0.880 for softness HF, 0.891 for softness TS7, 0.779 for tensile index, 0.680 for water absorption capacity, and 0.769 for capillary rise. These results found a good linear correlation between the measured and predicted values. Compared to MLR models, ANN models were better suited to predict softness and absorption properties, and these models were in the same range to predict strength properties. This suggested that the ANN results also complement the MLR analysis results.

Figure 4 presents the importance and normalized importance of each independent variable to determine the ANN model, based on the training and testing samples. The importance of an independent variable is quantified by changing the different values of the independent variable achieved by the ANN's model-predicted value. Furthermore, the normalized importance, expressed in percentages, is the ratio between the obtained importance values and the maximum importance values. However, these results do not identify the relationship between the independent variables and the predicted probability of default. This is why predictions from ANN models are complementary to well-designed MLR studies, despite the neural network's most noticeable limitations. For the tissue furnish process, the results revealed that SR, bulk, and width contribute most to the ANN model construction. Comparing both

Figure 4. Quantitative assessment of the importance of independent variables on the tissue furnish process, using an ANN model.

Figure 5. Predict values as function of experimental values of (a) softness HF, (b) softness TS7, (c) tensile index, and (d) water absorption capacity, using ANN models for industrial base tissue papers.
statistical models, “SR and structural properties are essential for the development of mathematical models. The furnish of different fibrous compositions, enzymatic and mechanical treatments, and additives modified the fiber refining degree, expressed in ‘SR, and consequently the structure bulk. Hence, these characteristics influenced the tissue functional properties.

4.2.2. Industrial base tissue papers

The maximum relative error of all dependent variables was 0.616 and the average relative error was 0.355, for the ANN training model of industrial base tissue papers. Regarding the ANN testing model, the maximum relative error was 0.897 and the average relative error was 0.573 (Table 2). From the architectural point of view, there was a total of 7 independent variables in the input layer, 3 neurons in the hidden layer, and 4 dependent variables in the output layer (Table S5 and Figure S1). Figure 5 presented the relationship between the experimental values and predicted values for tissue functional properties. Results indicated that the neural network prediction is very close to the measured values for tensile index and water absorption capacity, while for softness HF and softness TS7 it was not verified. Moreover, this analysis could not develop a model for capillary rise properties, as observed for MRL models. Regression analysis presented a coefficient of determination of 0.431 for softness HF, 0.517 for softness TS7, 0.743 for tensile index, and 0.807 for water absorption capacity. As mentioned above, the MLR analysis was unable to predict any model with significance for the softness HF properties, and the R² obtained with the ANN models was also very low. The same trend has also occurred for softness TS7. Additionally, both MLR and ANN models showed a good forecast accuracy to predict the tensile index and water absorption capacity. These results suggest that it was not possible to find any statistically accurate model to predict the softness properties of industrial base tissue papers.

Additionally, the results also explained the importance of curl, bulk, coarseness, length, porosity, and width in tissue functional properties of industrial base tissue papers (Figure 6). The structural properties were the key independent variables to develop the MLR and ANN models. The creping and converting processes improve bulk and porosity properties, which influence tissue functional properties.

4.3. Validation of models using experimental and 3D fiber-based modeling simulation approaches

The regression models of the relationship between softness HF, softness TS7, tensile index, water absorption capacity, and capillary rise (dependent variables) and morphological, suspension, and structural properties (independent variables) were validated using experimental and computational data. We presented an example of the tissue furnish process and industrial base tissue paper models’ validation. For this purpose, we selected a bleached eucalyptus kraft pulp, a biopolymer as additives, and an industrial base tissue paper for these studies. Note that these samples were not used to build the models. Additionally, the eucalyptus pulp was beaten in a PFI mill at 500, 1000, and 1500 revolutions under a refining intensity of 3.33 N/mm. The morphological properties of pulp fibers were determined using the MorFi Analyzer. To evaluate the impact of different fibers on paper properties, isotropic laboratory structures of 20 g/m² were prepared and tested regarding structural and tissue functional properties (ISO standards).

Using the 3D fiber-based simulator, it was possible to model the fibers individually, resulting in the 3D tissue computational structure. This approach is important to investigate the relationship between fibers and structures formed by them, predicting the influence of these fibers on the design of innovative tissue products and furnish optimization, saving laboratory and industrial resources [45, 46]. For example, a comparison

| PFI Revolutions | Experimental Data | Computational Data |
|-----------------|-------------------|--------------------|
| Thickness (μm)  | Bulk (cm³/g)      | Apparent Porosity (%) |
| 0               | 101               | 4.89               | 86.37 |
| 500             | 93                | 4.61               | 85.53 |
| 1000            | 85                | 4.18               | 84.03 |
| 1500            | 77                | 3.90               | 82.89 |
| Effective Thickness (μm) | Bulk (cm³/g) | Apparent Porosity (%) |
| 0               | 99                | 4.81               | 86    |
| 500             | 94                | 4.68               | 85    |
| 1000            | 85                | 4.19               | 84    |
| 1500            | 76                | 3.84               | 82    |

Figure 6. Quantitative assessment of the importance of independent variables on the industrial base tissue papers, using an ANN model.
of the simulated and experimental structures was performed to validate the predictive character of the structural properties of tissue papers (Table 3). The pulp fibers have been beaten in four beating levels (0, 500, 1000, 1500 PFI revolutions), and laboratory structures corresponding to a thickness of 101, 93, 85, and 77 µm, respectively. Following our experimental verification, the fiber flexibility and fiber effective thickness, related to the fiber coarseness, presented a crucial impact on the paper structure. A similar impact for the influence of these fiber properties on simulated structures was verified. This 3D fibrous material computational simulator simulates the structure’s effective thickness and apparent porosity, among other structural properties. The model proves to adjust to the experimental data. The bulk properties were obtained through the experimental and computational thickness. Through this methodology, it is possible to model the fibers and, consequently, predict the structural properties of tissue structures. Without the need for extensive experimental planning, the simulator evaluates different scenarios of interest quickly and accurately. Structural properties, such as the bulk, obtained indirectly by the computational effective thickness, and the computational apparent porosity, obtained directly from simulation, can be introduced into the mathematical models to predict the functional properties, saving laboratory resources. These results suggested that 3D fiber modeling proved to be an essential tool to investigate the influence of fiber properties on the structural properties of tissue products due to the good estimative of the simulated results with the experimental ones. This innovative approach results in higher speed and flexibility of response, without the need to produce laboratory structures.

Posteriorly, the developed models were validated. The percent deviation between experimental and the predicted values of softness HF, softness TS7, tensile index, water absorption capacity, and capillary rise of tissue furnish process and industrial base tissue papers using MLR models. The predicted data is within the experimental error.

5. Conclusion

Through the combination of an experimental and computational design, this work allowed establishing relationships between the variables of fiber and the softness, tensile strength, and absorption properties, in order to obtain predictive capacity, quantifying the changes that occur due to the use of different raw materials and tissue paper production process operations. An alternative proposed in this work was the development of statistical models allowing correlation of the tissue functional properties with morphological, suspension, and structural
characteristics, considering the tissue furnish process that included data from fiber mixtures, mechanical and enzymatic treatments, and additives incorporations, and industrial base tissue papers.

The predicted results of the relationship between these dependent and independent variables using MLR and ANN modeling were found to be highly satisfactory in terms of explanatory characteristics and validity of all models, except for softness and capillary rise properties of industrial base tissue papers. Additionally, using 3D fiber modeling to obtain 3D tissue structure computational simulations, it was possible to study the influence of fiber properties on the structural properties of tissue papers. The results indicated that these three approaches can be successfully used to model the effects of key fiber properties, fiber modification process steps, and tissue structural properties on softness, strength, and absorption parameters.

The developed models allow evaluating preliminary decisions regarding the usability of different pulp samples and process steps in which the final end-use tissue properties are important. This computational modeling approach can be used to design innovative tissue products with added value, meeting specific requirements, by changing the pulp fiber morphological characteristics. Furthermore, these methods are important tools to evaluate different scenarios of industrial interest, without the need for extensive experimental planning, contributing to raw materials furnish management by predicting the associated formulation costs.

Declarations

Author contribution statement

Flávia P. Morais: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Joana M.R. Curto: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Funding statement

This work was supported by Project InPaCTus – Innovative Products and Technologies from eucalyptus. Project N° 21 874 funded by Portugal 2020 through the European Regional Development Fund (ERDF) in the frame of COMPETE 2020 n° 246/AXIS II/2017 and by research unit Fiber Materials and Environmental Technologies (FibEnTech-UBI), on the project of the target research UIDB/00195/2020, funded by the Fundação para a Ciência e a Tecnologia, IP/MCTES through national funds (PIDDAC).

Data availability statement

The data that has been used is confidential.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

Supplementary content related to this article has been published online at https://doi.org/10.1016/j.helyon.2022.e09356.

References

[1] G. Gaiselmann, R. Thiedemann, I. Manke, W. Lehnert, V. Schmidt, Stochastic 3D modeling of fiber-based materials, Comput. Mater. Sci. 59 (2012) 75–86.

[2] R. Kerekes, D. McDonald, J. Zhao, Perspectives on deriving mathematical models in pulp and paper science, BioRes 15 (2020) 7319–7329.

[3] W.T. Wange, J. Li, W. Liu, Z.-K. Liu, Integrated computational materials engineering for advanced materials: a brief review, Comput. Mater. Sci. 158 (2019) 42–48.
