KnE Materials Science
FibEnTech21
1st International FibEnTech Congress (FibEnTech21) New opportunities for fibrous materials in the ecological transition
Volume 2022

Research article

Development of Sustainable Cellulose-Based Tissue Materials Using an Innovative Experimental and Computational Methodology

Flávia P. Morais[1]*, Ana M.5M.S. Carta[2], Maria E. Amaral[1], and Joana M.R. Curto[1,3]

[1]Fibre Materials and Environmental Technologies (FibEnTech-UBI), University of Beira Interior, R. Marquês de Ávila e Bolama, 6201-001 Covilhã, Portugal
[2]Forest and Paper Research Institute (RAIZ), R. José Estevão, 3800-783 Eixo, Aveiro, Portugal
[3]Chemical Process Engineering and Forest Products Research Centre (CIEPQPF), University of Coimbra, R. Sílvio Lima, Polo II, 3004-531 Coimbra, Portugal

Abstract.
In recent years, the tissue industry has been exposed to several challenges related to the growing demand for high-quality materials and sustainability. An approach that combined experimental and computational planning was implemented and presented in this work. For this purpose, a simulator that established relationships between the key fibre properties, the process steps that modify them, and the functional properties, named the *SimTissue*, was developed and validated. Different scenarios and a summary of the *SimTissue* research strategy are presented. The experimental planning design consisted of examining the influence of refining, enzymatic treatment, and incorporation of additives such as micro / nanofibrillated cellulose and biopolymers. The correlations between these tissue process inputs, and the softness, strength and absorption properties were established using the *SimTissue*. Overall, the *SimTissue* predicted and optimized several case studies for the management and optimization of sustainability formulations.

Keywords: 3D computational simulation, cellulose-based materials, furnish optimization, tissue materials

Corresponding Author: Flávia P. Morais; email: flavia.morais@ubi.pt

Published 10 August 2022

Publishing services provided by Knowledge E

Selection and Peer-review under the responsibility of the FibEnTech21 Conference Committee.

1. Introduction

Sustainable cellulose-based fibrous materials, such as tissue papers, are designed for personal hygiene and utility purposes. Examples of tissue papers include toilet papers, towel papers, napkins, facial tissue papers, among others. These materials are recognized for their creped and embossed structures, with low basis weight, and high porosity. Their key properties include bulk, softness, strength, and absorption, whose contribution and importance differ and depend on the type of final product [1]. Significant global growth in this sector is projected into the next decade, also related to the current outbreak of COVID-19. Consumption of tissue materials was estimated to grow by around 55% in Eastern Europe and 45% in Western Europe between 2012-2022 [2]. The tissue industry is exposed to several challenges related to the growing demand for sustainable products and furnish, namely the raw materials, process operations steps, and additives incorporation [1]. A commitment to sustainable development is necessary to guide process decisions, as well as to search for simultaneous strategies of economic prosperity, environmental responsibility, and social equity [3]. Raw materials, such as cellulose fibre pulp, are the main factor in tissue paper manufacture, accounting for 45% to 59% of the total manufacturing cost [4]. Therefore, finding strategies to optimize the use of these raw materials on the final product and offset the cost-effectiveness offers substantial opportunities to drive consumer demand for optimized tissue products with innovative functionality and added value.

Eucalyptus fibre pulps are traditionally used in tissue production because of their ability to provide formation and softness to the materials, while softwood fibre pulps are used to ensure strength and paper machine runnability [1]. Eucalyptus fibres present the potential to be optimized to improve their mechanical properties while maintaining existing benefits. A more recent example is the extended impregnation cooking process which has shown the potential to increase tensile strength, reduce refining energy through its physical and chemical properties, increase pulp production yield, with higher product quality, lower consumption of chemical products in cooking, and cost reduction [5]. In addition to these strategies in the pulping process development, fibre modification strategies along the process steps have also been extensively investigated in recent years [1,4,6-12]. Maximizing the *Eucalyptus globulus* fibres incorporation, and the total or partial replacement of softwood pulps consists of a challenge since the required adjustments represent consequences on the runnability of the paper machine and converting lines. Obtaining tissue products with the best combination of fibres, and maximization of *Eucalyptus globulus* fibres, suggests knowledge of the different pulps on the market and the consequences on the finished product properties [6,7]. Due to the fibres' properties, such as high coarseness, the eucalyptus fibre's structure can be modified through enzymatic treatment processes [10], refining, and incorporation of additives such as micro/nanofibrillated cellulose (MFC/NFC) and biopolymers [11,12], minimizing the influence on the strength and stiffness properties that the structure needs for the process operations [13,14].

This entire tissue furnish optimization process requires extensive research and methodologies to overcome the issues, which involves the design and analysis of laboratory and computational experiences. Computational methods for integrating all important variables for analysing the behaviour and modelling of fibre materials, process modification steps, and functional properties have been increasingly investigated in recent decades [8,9,14-19]. Computational tools allow us to significantly reduce experimental work in the development and optimization of new materials, to present a better synergy between experimental tests and simulation of specific properties. The development of advanced computational tools with predictive capability for furnish is a major milestone soon as it allows us to estimate which raw materials are most promising for a particular tissue product and which process operations allow us to obtain tissue with desired and optimized properties [20].

Therefore, an innovative design approach was implemented to create product differentiation. To develop and optimize these premium materials, we designed a simulator that allows us to compare various scenarios of furnish formulations, the *SimTissue*. This simulator establishes relationships between key fibre properties, the fibre modification process steps, and the tissue structural and functional properties. In this work, we present different scenarios and a summary of all the research that is based on the development of *SimTissue* for the management and optimization of sustainable furnish materials, using experimental and computational methodology approaches.

2. Material and methods

To implement the computational studies, initially, a characterization of the fibre dimensions in the paper structure was carried out, in the three dimensions, through the development of methodologies using SEM image analysis and advanced computational tools for fibre and structure modelling [8,9]. The experimental planning also consisted of the characterization and selection of different cellulosic pulps, obtained from hardwood (HW) and softwood (SW) species and different pulping processes of industrial interest [6,7]. The effect of fibre modification was also investigated, either by mechanical (refining) process, either by enzymatic treatment, or a combination of both [10], and the incorporation of additives such as MFC/NFC and commercial biopolymer (CBA) [11,12]. All experimental conditions used can be found in our referenced and cited publications. The properties of bulk (ISO 12625-3), softness (using Tissue Softness Analyzer (TSA Emtec) equipment), tensile strength index (ISO 12625-4), water absorption capacity (ISO 12625-8 adaptation), and capillary rise (ISO 8787 adaptation) were evaluated on laboratory structures of 20 g/m^2 (ISO 5269-1 adaptation). This was a proposed methodology to mimic the tissue-based sheets. The relationships between these properties were established, using different innovative computational tools, such as Lasso regression, robust multi-linear regressions (MLR), artificial neural networks

(ANN), among others, and using simulation and experimental data, to obtain predictive capacity. Then, the structures were 3D computationally simulated, to calibrate the tissue model and validate its predictive character [8,9,15,16]. This entire process allowed the computational implementation of the *SimTissue* through the calculation engine algorithm programming, database, and user interface integration, to be used in specific cases to support furnish's industrial management.

3. Results and discussion

For the *SimTissue* development, a methodology that combines an experimental and computational approach was designed (Figure 1). *SimTissue* establishes relationships between input variables of fibre properties and output variables of tissue functional properties. Through experimental and computational data, *SimTissue* quantifies the changes that occur due to the use of different pulps and process operations of tissue paper production. The validation of the final end-use properties' prediction was carried out with experimental and simulated structures, to develop this simulator for the fibrous materials and furnish optimization [21]. Different furnish scenarios with industrial interest are investigated including different fibre combinations and the design and optimization of each tissue product. In our previous works, this simulator was also used to obtain predictive capacity for the creping and embossing influence on the softness, strength, and absorption properties of industrial and laboratory structures produced with eucalyptus and softwood fibers [20]. Therefore, the computational implementation of *SimTissue* also supports industrial furnish management considering structures with creping and converting operations. In the present work, we are presenting only a furnish optimization approach, not considering these operations.

The results indicate that *SimTissue* predicts the influence of various types of raw materials used to produce tissue products, process operations, and additives at the micro and nanoscale. An important achievement is the comparison of structures produced with 100% eucalyptus fibres, with different percentages of other fibres, and additives. In this work, we present different comparative scenarios of case studies predicted by *SimTissue* to design new tissue paper formulations, compared to typical industrial formulations (75%HW+25%SW) with typical mill drainability values (≈25°SR). These formulations presented a softness of 59.9 HF, a tensile index of 18.5 Nm/g, a water absorption capacity of 7.5 g/g, and a capillary rise of 121 mm.

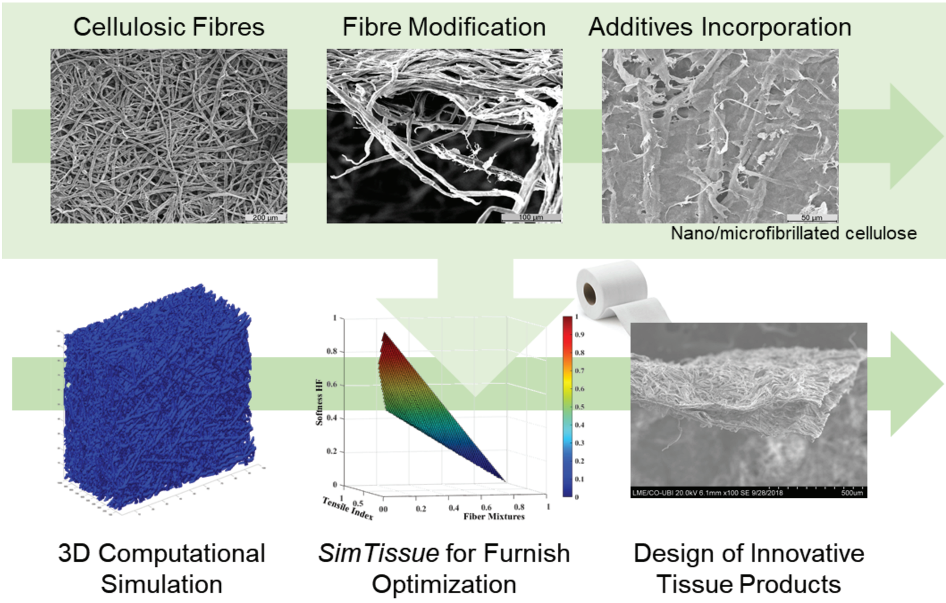

Figure 1: Outline of the methodology approach used in this work to develop the *SimTissue* for furnish management and optimization.

Figure 2 presents the percentage of variation in the final end-use tissue properties for formulations with different fibrous compositions and MFC/NFC incorporation. Formulations were simulated with a eucalyptus pulp (HW), and a softwood pulp (SW) with higher strength properties, in percentages of 1%, 5%, and 10% (Figure 2a). Formulations with MFC/NFC incorporation were also simulated, at the same percentages (Figure 2b). A formulation was further proposed that contained these three fibrous elements, namely, 90%HW+5%MFC/NFC+5%SW (Figure 2b). All formulations with HW and SW presented 25°SR, and formulations with MFC/NFC presented drainability between 26 and 44°SR.

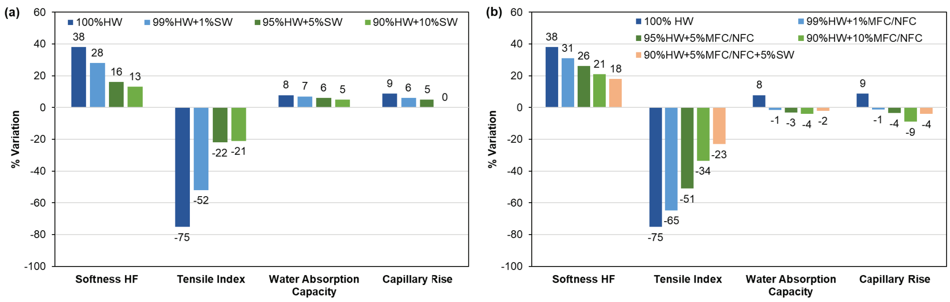

Figure 2: Percentage of variation in the tissue final end-use properties for formulations with (a) HW and SW; and (b) HW and MFC/NFC, compared to the typical industrial structures (75%HW+25%SW), and predicted using the *SimTissue*.

From these scenarios, the results indicated that the production of tissue structures with reduced SW incorporation or its replacement by MFC/NFC promoted an increase in softness properties. Structures with HW and SW contributed positively to the strength development compared to structures with MFC/NFC. The formulations whose balance between the softness and tensile index properties, and with the highest potential to replace this industrial reference, were 90%HW+10%SW and 95%HW+5%MFC/NFC. This evidence is suggested by comparison with industrial base tissue papers. Despite the 51% loss in the tensile index, structures with 5% MFC/NFC showed strength values in the range of these industrial tissue papers (6.9 - 12.0 Nm/g), and consequently, unsustainable breaks are expected to be unlikely to occur. The results also indicated that SW fibres remain an important element to confer strength and absorption properties, especially in products such as wet strength papers. MFC/NFC presents a good potential to be used in tissue products such as toilet and facial papers, produced with the eucalyptus maximization. The 90%HW+5%MFC/NFC+5%SW formulation also presented a good combination between softness, strength, and absorption, essential to produce premium materials [11].

Regarding fibre modification studies, a kraft eucalyptus pulp with higher tensile index values and a sulphite pulp with higher softness values were separately subjected to enzymatic treatments [10]. The information about fibre flexibility and biometry is determinant to predict tissue paper properties. These two raw materials correspond to the two extremes of eucalyptus pulps, regarding fibre wall thickness, degree of collapse, refining ability, flexibility values, and softness and strength properties. All these fibre properties and dimensions are essential to model them in 3D, resulting in a structure with optimized bulk, for example. Therefore, the results constitute valuable information applicable for other pulps of interest, with fibre dimensions in this range. Through this optimization, the variation of time and enzyme dosage can be predicted, using data-driven statistical mathematical models. Table 1 presents examples of simulated scenarios for the enzymatic treatment of these pulps. First, and regarding kraft pulp, *SimTissue* predicted softness and tensile index values in the same range as typical industrial formulations (softness: 60 HF; tensile index: 19 Nm/g; capillary rise: 121 mm). Therefore, *SimTissue* predicts the formulation properties with variation in these two process variables: reaction time and enzyme dosage. In these simulation studies, similar softness and strength properties are obtained with an enzyme reaction time of 30 minutes, and with a dosage of 50 g/ton, or also with twice the enzyme reaction time, and with a lower dosage of 20 g/ton. The same analysis can also be performed for other pulps of interest.

To obtain new formulations with 100% eucalyptus pulps, it is important to select the process conditions that allow a good trade-off between them, to adjust the furnish for the tissue properties development. A design was proposed that combines 80% of

DOI 10.18502/kms.v7i1.11606

Table 1: Scenarios for the enzymatic treatments of kraft and sulphite pulps, predicted by *SimTissue*.

	Enzyme Dosage (g/ton)	Reaction Time (minutes)	Softness HF	Tensile Index (Nm/g)	Capillary Rise (mm)
Simulation 1 (Kraft pulp)	50	30	53	18	108
Simulation 2 (Kraft pulp)	20	60	53	20	108
Simulation 3 (Sulphite pulp)	100	30	81	7	117
Simulation 4 (Sulphite pulp)	30	60	82	7	116

the enzymatically treated kraft eucalyptus pulp and 20% of the sulphite pulp with and without enzymatic treatment [10]. Figure 3a presents the percentage of variation in the final end-use tissue properties for formulations with different combinations, compared to formulations with 75%HW+25%SW. Overall, the simulated combinations contributed to increased softness with good tensile and absorption properties, ensuring the necessary strength to obtain premium tissue papers. Considering the inverse progression of softness HF and tensile index, the identification of different mixture conditions that allow the best balance between these two properties can be identified for different types of tissue papers. Despite the difference found between the strength of the combinations and the industrial reference, the tensile index values of the combinations (14 – 17 Nm/g) were higher than the range of values for industrial base creped tissue papers (6 – 11 Nm/g) [20].

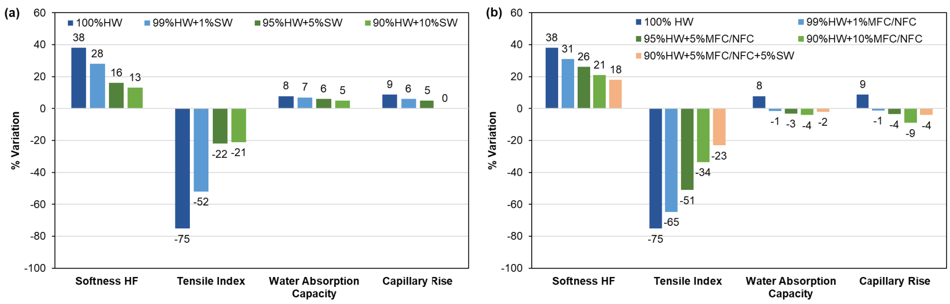

Figure 3: Percentage of variation in the tissue final end-use properties for fibre modification formulations, compared to the typical industrial structures (75%HW+25%SW) and predicted using the *SimTissue*. (a) Combination 1: 80% kraft pulp with 10 g/ton, 30 minutes + 20% sulphite pulp without treatment; Combination 2: 80% kraft pulp with 10 g/ton, 60 minutes + 20% sulphite pulp without treatment; Combination 3: 80% kraft pulp + 20% sulphite pulp, both with 100 g/ton, 30 minutes; Combination 4: 80% kraft pulp + 20% sulphite pulp, both with 100 g/ton, 60 minutes. (b) Simulation 1: kraft pulp treated with 50 g/ton, 120 minutes followed by mechanical treatment (500 PFI revolution); Simulation 2: kraft pulp treated with 350 g/ton, 120 minutes followed by 500 PFI revolution; Simulation 3: kraft pulp with 500 PFI revolution followed by 50 g/ton, 120 minutes; Simulation 4: kraft pulp with 500 PFI revolution followed by 100 g/ton, 120 minutes.

One of *SimTissue*'s features is also to allow the simulation of scenarios that combine different process operations, such as mechanical and enzymatic treatments. To this end, the same kraft pulp was subjected to enzymatic pre-treatment followed by refining, and vice versa. Examples of these simulations, compared to the industrial reference (75%HW+25%SW), are shown in Figure 3b. From this study, *SimTissue* predicted that similar properties were obtained with an enzymatic pre-treatment with an enzyme dosage of 350 g/ton, followed by refining (simulation 2) and a pre-refining followed by enzymatic treatment with an enzyme dosage 7 times less, 50 g/ton (simulation 1). With this approach, several scenarios for enzymatic and mechanical treatments with equivalent energy consumption can be foreseen. Overall, formulations were obtained with improved softness and absorption properties compared to the industrial reference, and with strengths in the commercial tissue paper range. Although a pre-refining followed by an enzymatic treatment presents a negative impact on the fibres, namely the decrease of fibre length and the increase of fines content, this procedure can be advantageous in obtaining pulps with strength characteristics. Therefore, a pulp with this fibre modification process can be used as an additive in pulps with good softness and absorption properties. Consequently, the production of tissue papers with a composition of 100% eucalyptus pulp and with optimized tissue properties can be achievable.

Regarding the incorporation of additives, in addition to MFC/NFC, *SimTissue* also allows the study of other additives such as CBA [12]. This computational platform predicted the tissue properties as a function of the CBA incorporation in eucalyptus pulps and compared with the industrial reference (Figure 4a). Overall, the incorporation of this additive increased the softness and absorption at the expense of decreasing strength, up to 3% incorporation. On the other hand, after the 4% incorporation, the softness and strength properties increased at the expense of absorption. Note also that the 4% CBA incorporation has practically the same tensile index value as the industrial reference. This means that this incorporation can allow the replacement of SW in terms of strength properties compared to 75%HW+25%SW.

Another planning design that included the selection of eucalyptus and softwood pulps, the combination of process operations and incorporation of two additives was carried out and foreseen by *SimTissue*. Two scenarios were simulated on our computational platform and compared to the industrial reference (Figure 4b). The simulations were carried out to study the effect of the conjugation of two additives on tissue properties and to study their implication in softwood pulp reduction [12]. In Simulation 1, the scenario of selecting 75%HW, 25%SW with mechanical refining treatment followed by enzymatic treatment of 30 g/ton for 60 minutes, and incorporation of 2% MFC/NFC

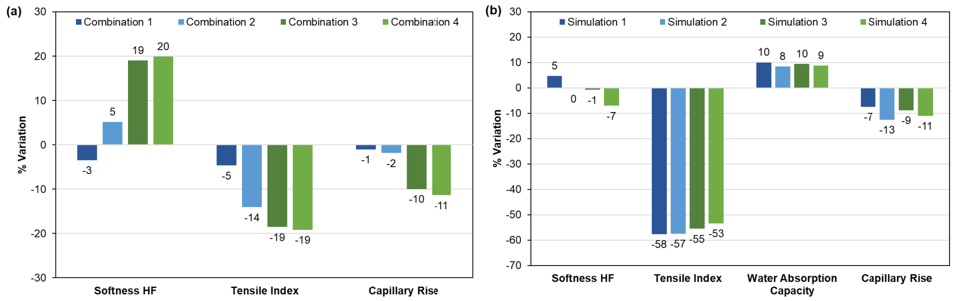

Figure 4: Percentage of variation in the tissue final end-use properties for formulations with (a) HW and CBA; and (b) HW, SW, MFC/NFC, and CBA, compared to the typical industrial structures (75%HW+25%SW), and predicted using the *SimTissue*. Simulation 1: 75%HW+25%+2%MFC/NFC+2%CBA; Simulation 2: 90%HW+10%+2%MFC/NFC+2%CBA.

and 2%CBA, as additives, was proposed. In Simulation 2, the same previous conditions of process operation and additives were proposed, but with a selection of 90%HW and 10%SW. Compared to 100%HW formulations, the addition of both additives improved strength at the expense of decreased softness and absorption. With the softwood pulp reduction, the additives' incorporation promoted an increase in softness and absorption, with good strength properties similar to commercial tissue papers. The understanding of the possibility of producing tissue structures with different additives, softwood pulp reduction, and fibre modification treatments, in the range of industrial drainage, was possible through these simulations.

4. Conclusions

With this research work, we present an experimental and computational methodology to develop sustainable cellulose-based tissue materials. A simulator with predictive capability, the *SimTissue*, was developed and validated to optimize innovative furnish formulations, saving laboratory, and industrial resources. *SimTissue* was applied to identify the more adequate eucalyptus and softwood pulps from a set of raw material pulps. The results indicated the optimized combination of pulps to produce toilet and facial papers, in which softness is the key property, as well as towel papers and napkins, in which strength and absorption are essential. Simulation studies were used to optimize different furnish formulations, predicting different scenarios with enzyme dosages reduction, extending its reaction time, translating into refining energy gains. The effects of incorporating additives, such as MFC/NFC or CBA, were also estimated by *SimTissue*. Several hypotheses of industrial interest were simulated, and the trade-off between softness, strength, and absorption properties was quantified in different scenarios. In conclusion, the *SimTissue* is a computational tool with predictive capacity useful for furnish management and sustainable process optimization.

Acknowledgments

This research was supported by Project InPaCTus – Innovative Products and Technologies from eucalyptus. Project Nº 21 874 funded by Portugal 2020 through European Regional Development Fund (ERDF) in the frame of COMPETE 2020 nº 246/AXIS II/2017. The authors are also very grateful for the support given by research unit Fibre Materials and Environmental Technologies (FibEnTech-UBI), on the extent of the project reference UIDB/00195/2020, funded by the Fundação para a Ciência e a Tecnologia, IP/MCTES through national funds (PIDDAC).

References

[1] de Assis T, Reisinger LW, Pal L, Pawlak J, Jameel H, Gonzalez RW. Understanding the effect of machine technology and cellulosic fibers on tissue properties – A review. BioResources. 2018;13(2):4593-4629. https://doi.org/10.15376/biores.13.2.DeAssis

[2] Tissue Word Magazine. Growth in the European tissue business moving from West to East. 2014, March 13. Available from: https://www.tissueworldmagazine.com/departments/marketissues/growth-in-the-european-tissue-business-moving-from-west-to-east/

[3] The Navigator Company. Sustainability policy. 2021. Available from: http://en.thenavigatorcompany.com/Sustainability/Sustainable-Forest/Sustainability-Policy

[4] Gotera ZFJ. Developing and understanding state-of-the-art technologies at nano- and macroscales to enhance fiber utilization in the hygiene tissue industry [Ph.D. thesis]. Raleigh: North Carolina State University (NCSU); 2021.

[5] Tissue World Magazine. Eucalyptus – Adding tensile strength and keeping formation and softness. 2020, December 3. Available from: https://www.tissueworldmagazine.com/special-features/focus-on/eucalyptus-adding-tensile-strength-and-keeping-formation-and-softness/

[6] Morais FP, Bértolo RAC, Curto JMR, Amaral MECC, Carta AMMS, Evtyugin DV. Comparative characterization of eucalyptus fibers and softwood fibers for tissue papers application. Materials Letter. 2019;10(4):100028. https://doi.org/10.1016/j.mlblux.2019.100028

[7] Morais FP, Bértolo RAC, Curto JMR, Amaral MECC, Carta AMMS, Evtyugin DV. Characterization data of pulp fibres performance in tissue papers applications. Data in Brief. 2020;29:105253. https://doi.org/10.1016/j.dib.2020.105253

[8] Morais FP, Carta AMMS, Amaral ME, Curto JMRC. Experimental 3D fibre data for tissue papers applications. Data in Brief. 2020;30:105479. https://doi.org/10.1016/j.dib.2020.105479

[9] Morais FP, Carta AMMS, Amaral ME, Curto JMRC. 3D Fiber models to simulate and optimize tissue materials. BioResources. 2020;15(4):8833-8848. https://doi.org/10.15376/biores.15.4.8833 8848

[10] Morais FP, Carta AMMS, Amaral ME, Curto JMRC. Cellulose fiber enzymatic modification to improve the softness, strength, and absorption properties of tissue papers. BioResources. 2021;16(1):846-861. https://doi.org/10.15376/biores.16.1.846 861

[11] Morais FP, Carta AMMS, Amaral ME, Curto JMRC. Micro/nano fibrillated cellulose (MFC/NFC) fibers as an additive to maximize eucalyptus fibers on tissue paper production. Cellulose. 2021;28(4):6587-6605. https://doi.org/10.1007/s10570 021039129

[12] Morais FP, Carta AMMS, Amaral ME, Curto JMRC. An innovative computational strategy to optimize different furnish compositions of tissue materials using micro/nanofibrillated cellulose and biopolymer as additives. Polymers. 2021;13(15):2397. https://doi.org/10.3390/polym13152397

[13] Ramezani O, Nazhad MM. The effect of coarseness on paper formation. African Pulp and Paper Week; 2004.

[14] Alava M, Niskanen K. The physics of paper. Reports on Progress in Physics. 2006;69(3):669-723. https://doi.org/10.1088/0034-4885/69/3/R03

[15] Conceição ELT, Curto JMR, Simões RMS, Portugal ATG. Computational modeling of objects represented in images. Barneva RP, Brimkov VE, Hauptman HA, Natal Jorge RM, Tavares JM, editors. Heidelberg: Springer; 2010.

[16] Curto JMR, Conceição ELT, Portugal ATG, Simões RMS. Three dimensional modelling of fibrous materials and experimental validation. Materialwissenschaft und Werkstofftechnik. 2011;42(5):370-374. https://doi.org/10.1002/mawe.201100790

[17] Lavrykov S, Lindström SB, Singh KM, Ramarao BV. 3D network simulations of paper structure. Nordic Pulp and Paper Research Journal. 2012;27:256-263. https://doi.org/10.3183/NPPRJ-2012-27-02-p256-263

[18] Raunio J-P, Ritala R. Simulation of creping pattern in tissue paper. Nordic Pulp and Paper Research Journal. 2012;27:375-381. https://doi.org/10.3183/NPPRJ-2012-27-02-p375-381

[19] Simon J-W. A review of recent trends and challenges in computational modeling of paper and paperboard at different scales. Archives of Computational Methods in Engineering. 2021;28:2409-2428. https://doi.org/10.1007/s11831-020-09460-y

[20] Curto JR, Vieira JC, Morais FP et al. Embossing influence on the 3D structure and key properties of tissue paper. Paper presented at: Proceedings of the XXV TECNICELPA Conferência Internacional da Floresta, Pasta e Papel XI CIADICYP 2021; 2021 Mar 9-12; WebConference, Tomar, Portugal.

[21] Morais FP, Curto JMR. Challenges in Computational Materials Modelling and Simulation. A case-study to predict tissue paper properties. Heliyon. 2022;8(5):e09356. https://doi.org/10.1016/j.heliyon.2022.e09356.