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Effectiveness of the H-factor for Controlling *Eucalyptus* Kraft Pulping

Efetividade do Fator H para o controle do cozimento kraft de *Eucalyptus*

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RESUMO

O objetivo deste trabalho foi avaliar a efetividade do fator H como uma ferramenta de controle de processo no cozimento de *Eucalyptus*. Foram utilizados cavacos de madeira de 5,5 anos de um híbrido de *Eucalyptus grandis* × *Eucalyptus urophylla* plantado no Brasil. As simulações dos cozimentos foram realizadas em um digestor laboratorial utilizando oito diferentes temperaturas máximas (140 °C, 145 °C, 150 °C, 155 °C, 160 °C, 165 °C, 170 °C, e 175 °C) e oito diferentes tempos na temperatura máxima (28 min., 46 min., 72 min., 114 min., 180 min., 284 min., 530 min., e 727 min.), com o objetivo de manter o mesmo fator H para todos os tratamentos. Álcali ativo aplicado, sulfidez e tempo de aquecimento foram constantes para todos os tratamentos. Os resultados evidenciam que a taxa de deslignificação foi similar para todos os tratamentos, apesar da variação de temperatura. Um incremento na temperatura associado à redução do tempo de cozimento leva a uma redução no rendimento depurado, aumento do teor de sólidos orgânicos, e maior geração de rejeitos. Essa tendência foi mais clara em temperaturas acima de 155ºC. A formação de ácidos hexenurônicos e o consumo alcalino foram similares para todos os tratamentos. Os resultados encontrados comprovam que o fator H é uma ferramenta confiável para predizer e controlar o grau de deslignificação e os resultados do processo de cozimento, e que o rendimento depurado é favorecido em temperaturas mais baixas.

Palavras chave: Temperatura de cozimento; Deslignificação; Rendimento; Folhosa

ABSTRACT

The objective of this work was to evaluate the effectiveness of the H-factor as a process control tool on *Eucalyptus* pulping. Wood chips from 5.5-year-old Brazilian hybrid *Eucalyptus grandis* × *Eucalyptus urophylla* were used. The pulping simulations were performed in a laboratory digester using eight maximum temperatures (140 °C, 145 °C, 150 °C, 155 °C, 160 °C, 165 °C, 170 °C, and 175 °C) and eight times at the maximum temperature (28 min., 46 min., 72 min., 114 min., 180 min., 284 min., 4530 min., and 727 min.), with the objective of maintaining the same H-factor for all of the treatments. The active alkali, sulfidity, and heating time remained constant. The results showed that the delignification rate was similar for all of the treatments, even with the temperature variation. An increased cooking temperature and lower pulping time led to a reduction in the screened yield, increase in the black liquor organic solids, and a higher reject formation. This behavior was more obvious for temperatures above 155 °C. The hexenuronic acid formation and alkali consumption were similar for all of the treatments. These results proved that the H-factor is a reliable tool to predict and control the delignification rate and pulping results, and the screened yield is favored by lower cooking temperatures.

Keywords: Pulping temperature; Delignification; Yield; Hardwood

INTRODUCTION

The Kraft pulping process was patented by Dahl in 1884 and the first kraft mill went into operation in 1890 (BIERMANN, 1996). Since the introduction in 1879 of sodium sulfate (Kraft process) instead of carbonate (soda process) as the make-up chemicals to cover the losses in the recovery cycle of the

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cooking chemicals cycle, a considerable improvement in the rate of pulping and the quality of the pulp were observed (RYDHOLM, 1969).

The Kraft process remains the dominant pulping process due to the many advantages that it presents when compared with other processes, such as adaptability to many lignocellulosic materials, production of high quality pulp, pulp with a high bleachability, and favorable energy and chemical recovery capabilities (GOMIDE et al., 1980; GUSTAFSON et al., 1983; ASSUMPÇÃO et al., 1988; SANTOS et al., 2011).

In Kraft pulping, many parameters affect the delignification rate, pulping yield, and pulp quality. These parameters include the wood species, chip quality, applied alkali, pulping time, and temperature (COLODETTE et al., 2002; MACLEOD, 2007; SEGURA et al., 2016). Vroom (1957) proposed a single variable combining the pulping time and temperature, named the H-factor, which represents the extent of the reaction. Although different temperatures may be used, the cooking degree can be accurately estimated by this method, provided that the other variables, such as the applied alkali, sulfidity, and liquor-to-wood ratio, remain constant (BIERMANN, 1996).

According to Sixta (2006), the H-factor (H) is defined as:

$$H = \int_{t_0}^{t} k_L dt \qquad (1)$$

where k_L is the relative reaction of the pulping, and t_0 and t are initial and final times, respectively.

Assuming activation energy of 134 kJ/mol, the H-factor can be expressed as:

$$H = \int_{t_0}^{t} \frac{k_{L(T)}}{k_{100°C}} dt = \int_{t_0}^{t} Exp\left(43.19 - \frac{16113}{T}\right) dt \qquad (2)$$

where T is the cooking temperature (K).

The H-factor is the area under the relative reaction *versus* time curve. This parameter was developed to predict the temperature or cooking time needed to achieve a given kappa number. This result is only possible when the other cooking conditions, such as the effective alkali concentration and liquor-to-wood ratio, are kept constant (SIXTA, 2006).

An H-factor of 1 represents the effect of pulping for 1 h at 100 °C, while cooking for 1 h at 171 °C will change the H-factor to 1000 (RYDHOLM, 1965).

The H-factor is a useful process control tool for Kraft pulp mills. Even the most modern plants use this parameter to control the pulp delignification rate, which is expressed as the kappa number (SEGURA et al., 2016).

Many empirical kinetic models have been derived from the H-factor (GUSTAFSON et al., 1983). For example, the model by Hatton (1973) predicts the kappa number and yield for various wood species, while the model by Bailey et al. (1969) uses five variables to predict the pulp delignification rate.

As the H-factor is a very usual tool to control delignification rate in pulp mills, this study aimed to determine its effectiveness as a process control tool, while testing different combinations of time and temperature, maintaining a constant H-factor, and evaluating some of the most important process parameters. Similar works were not published in scientific journals and only technical reports are available.

MATERIAL AND METHODS

Wood

Eucalyptus grandis × *Eucalyptus urophylla* wood chips from Brazilian trees that were 5.5 years in age were collected and screened with a laboratory screener. A thickness fraction of 4 mm to 6 mm was used in the experiments. The wood characteristics are shown in Table 1.

Regarding the wood characterization, the samples used in this study represent a typical Brazilian eucalyptus hybrid. Duarte (2007), Segura et al. (2016), and Vivian et al. (2017) describe similar *Eucalyptus grandis* × *Eucalyptus urophylla* wood characteristics.

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Table 1. Wood Characteristics
Tabela 1. Caracterização da madeira

Basic Density[1] (g/cm³)	Total Extractives[2] (%)	Total Lignin[3] (%)	Holocellulose[4] (%)
0.457	4.1	27.5	68.4

[1] Smith (SMITH, 1954); [2] TAPPI T204 cm-07 (TAPPI, 2007); [3] TAPPI T222 om-06 (TAPPI, 2006); [4] Difference between 100% and the sum of the total extractives and total lignin

Pulping

The Kraft pulping was performed in triplicate in a forced circulation digester (Technological Solutions Integrated (TSI), Piracicaba, Brazil) with a capacity of 10 L and heat exchangers for heating. The fixed pulping conditions are shown in Table 2.

The maximum pulping temperatures were varied. To maintain the same H-factor (825) for all of the treatments, the time at the maximum temperature was also varied for each treatment, which is shown in Table 3.

The pulping liquor was sampled every 165 H-factor points. Sampling points were varied for each treatment (Table 4).

Table 2. Fixed Pulping Conditions
Tabela 2. Condições fixas de cozimento

Parameter	Value
Active Alkali (%)	16.0
Sulfidity (%)	25.0
Heating time (min)	60
H-factor	825

Table 3. Pulping Variable Conditions
Tabela 3. Condições variáveis de cozimento

Treatment	A	B	C	D	E	F	G	H
Maximum temperature (°C)	140	145	150	155	160	165	170	175
Time at maximum temperature (min)	727	453	284	180	114	72	46	28

Table 4. Black Liquor Sampling Times
Tabela 4. Tempos de amostragem do licor negro

H-factor	165	330	495	660	825
A (140 °C; min)	200	347	493	640	787
B (145 °C; min)	145	237	329	421	513
C (150 °C; min)	112	170	228	286	344
D (155 °C; min)	91	128	165	203	240
E (160 °C; min)	78	102	126	150	174
F (165 °C; min)	69	85	101	117	132
G (170 °C; min)	64	74	85	95	106
H (175 °C; min)	61	68	74	81	88

After cooking, pulps were washed with distilled water to remove the liquor and to perform other analyses. The residual alkali in the liquor was determined according to SCAN-N 2:88 (SCAN, 1988), and the solids content was determined with TAPPI T625 cm-14 (TAPPI, 2014) and TAPPI T650 om-15 (TAPPI, 2015). The yields were calculated based on the pulp and wood dried masses. The kappa numbers and hexenuronic acids contents of the unbleached pulps were determined according to TAPPI T236 om-99 (TAPPI, 1999) and TAPPI T282 om-13 (TAPPI, 2013).

Data analysis

The main results were statistically analyzed using an analysis of variance, and the significant differences were determined using Tukey's test at 5% significance (Minitab 17, Minitab Inc., Pennsylvania, USA).

RESULTS AND DISCUSSION

The kappa number is defined as the volume (in mL) of 0.1N potassium permanganate solution consumed by one gram of moisture free pulp under standard conditions and is equivalent to approximately seven times the mass percentage of lignin (SALEEM; AKHTAR, 2002). In a pulp mill, the brownstock kappa number influences some of the most important parameters of pulp production, such as the pulping yield and quality, alkali charge, solids generation, and bleaching chemical consumption (SEGURA et al., 2016). The kappa number results are shown in Figure 1.

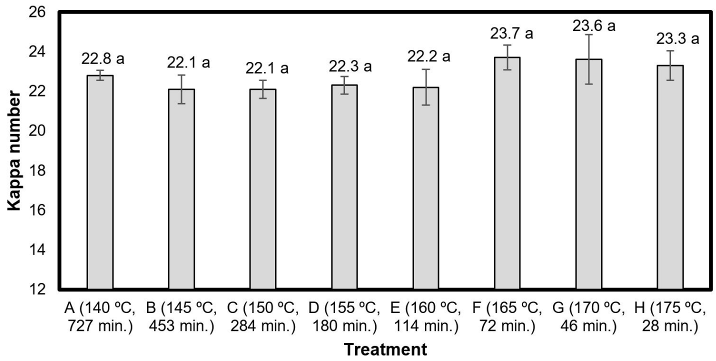

Figure 1. Kappa numbers for the different H-factors - Different letters indicate statistical significance (p-value ≤ 0.05) using Tukey's test

Figura 1. Números kappa para os diferentes fatores-H – letras diferentes indicam significância estatística (p ≤ 0.05) utilizando o Teste de Tukey

All of the treatments resulted in brownstock pulps with the same range of delignification. The kappa numbers ranged from 22.1 to 23.7, and no statistically significant difference was observed. These results showed that the H-factor is a useful tool to predict and control the delignification rate, even if different combinations of pulping time and temperature are used.

Figure 2 presents the total and screened yields, and Table 5 shows the statistical analysis of the screened yield results.

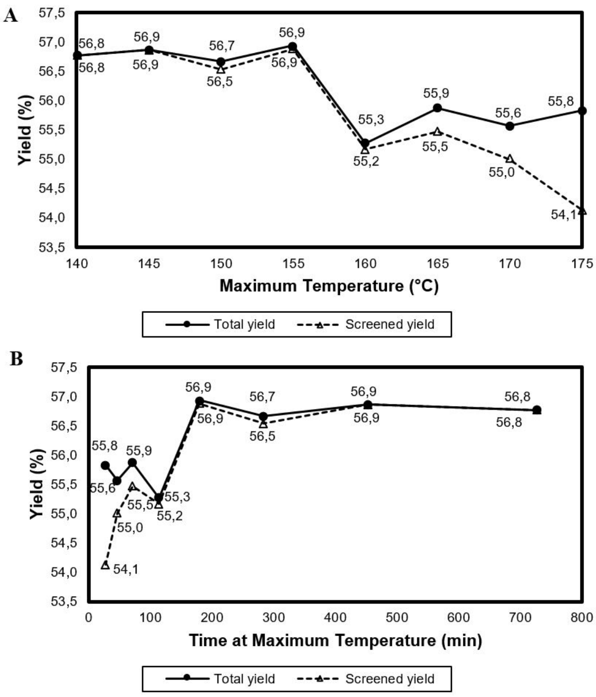

Figure 2. Total and screened yields as a function of the maximum temperature (A) and time at the maximum temperature (B)

Figura 2. Rendimento bruto e depurado em função da temperatura máxima (A) e do tempo a temperatura máxima (B)

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Table 5. Statistical Analysis of the Screened Yield
Tabela 5. Análises estatísticas do rendimento depurado

Treatment	Screened Yield (%)	Standard Deviation (%)
A (140 °C, 727 min)	56.8 a	0.8
B (145 °C, 453 min)	56.9 a	0.3
C (150 °C, 284 min)	56.5 ab	1.2
D (155 °C, 180 min)	56.9 a	0.6
E (160 °C, 114 min)	55.2 ab	0.4
F (165 °C, 72 min)	55.5 ab	1.1
G (170 °, 46 min)	55.0 ab	0.7
H (175 °C, 28 min)	54.1 b	1.4

Different letters indicate statistical significance (p-value ≤ 0.05) using Tukey's test.
Letras diferentes indicam significância estatística (p ≤ 0.05) utilizando o Teste de Tukey.

Even when the H-factor was constant, a decrease in the yields was observed with an increase in the maximum pulping temperature. A flat trend was observed for the temperatures between 140 °C and 155 °C. In this range, both the total and screened yields were constant, and the rejects generation was next to zero. A noticeable decrease in the pulp yield was observed between 155 °C and 160 °C; for the maximum temperatures of 160 °C to 175 °C, the total yields were relatively constant, but the screened yield tended to decrease with an increase in the temperature, which indicated an increase in the reject formation at the highest temperatures and consequently decreased the cooking times.

Some studies have analyzed the effect of the maximum pulping temperature on the process parameters and pulp characteristics. The results found in this work agreed with those of Colodette et al. (2002), who showed that a low cooking temperature leads to an increase in the pulping yield. At the same time, Colodette et al. (2002) found that higher pulping temperatures cause an increase in the pulp bleachability.

Rosli et al. (2009) analyzed the effect of different Kraft pulping variables on pulp and paper. It was confirmed that the applied alkali and sulfidity are the most important factors that affect the yield, while the maximum temperature has a moderate effect.

The hexenuronic acid results are presented in Figure 3.

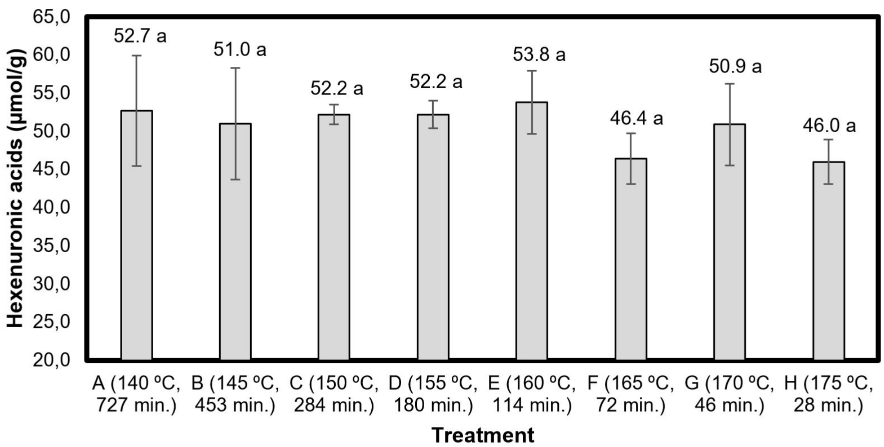

Figure 3. Statistical analysis of the hexenuronic acids - Different letters indicate statistical significance (p-value ≤ 0.05) using Tukey's test
Figura 3. Análise estatística dos resultados de ácidos hexenurônicos – letras diferentes indicam significância estatística (p ≤ 0.05) utilizando o Teste de Tukey

Hexenuronic acids are components formed during the alkaline cooking of wood and are part of the kappa number (COSTA; COLODETTE, 2002), which is dependent on many cooking variables (ROSLI et al., 2009) and the chemical composition of the wood (CARDOSO et al., 2002).

In the present work, all of the produced pulps presented a hexenuronic acids content that ranged from 46.4 µmol/g to 53.8 µmol/g, and there were no statistical differences. Along with a constant delignification level, cooking wood with different combinations of time and temperature and maintaining a constant H-Factor and other variables (wood sample, alkali charge, and liquor-to-wood ratio) will result in a constant amount of hexenuronic acids in the pulp.

The alkali consumption during pulping is presented in Figure 4. This consumption followed the H-factor, and the residual results were very similar for all of the treatments with different H-factor levels. The consumed alkali was also calculated in g/L and in kg/A.D. ton (Table 6), and was statistically similar for all of the tested pulping conditions.

The black liquor solids results for the different treatments are presented in Table 7.

Despite the constant H-factor, an increase in the cooking temperature caused an increase in the black liquor organic solids. However, the statistical analysis did not reveal any significant differences. This increase was caused by the decrease in the pulping yields, which was observed when the pulping temperature increased; a lower pulping yield leads to a higher formation of black liquor solids.

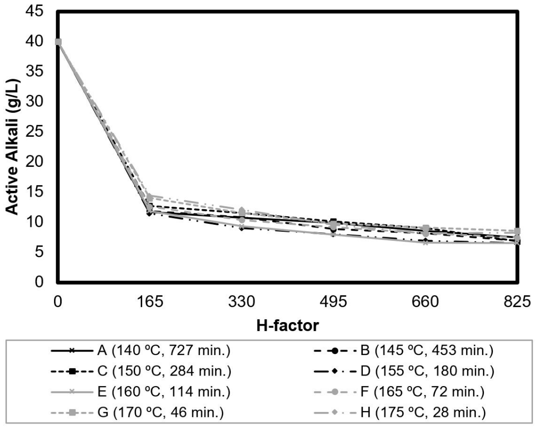

Figure 4. Residual alkali profiles
Figura 4. Perfis de álcali residual

Table 6. Consumed Alkali
Tabela 6. Álcali consumido

Treatment	Consumed Active Alkali	
	(g/L)	(Kg/A.D.ton)
A (140 °C, 727 min)	32.5 a	229 a
B (145 °C, 453 min)	33.1 a	233 a
C (150 °C, 284 min)	33.1 a	234 a
D (155 °C, 180 min)	33.5 a	236 a
E (160 °C, 114 min)	33.4 a	243 a
F (165 °C, 72 min)	32.5 a	235 a
G (170 °C, 46 min)	31.4 a	229 a
H (175 °C, 28 min)	31.8 a	235 a

Different letters indicate statistical significance (p-value ≤ 0.05) using Tukey's test.
Letras diferentes indicam significância estatística (p ≤ 0.05) utilizando o Teste de Tukey.

Table 7. Black Liquor Solids
Tabela 7. Sólidos do licor negro

Treatment	Total Solids (%)	Organic Solids (%)	Inorganic Solids (%)
A (140 °C, 727 min)	12.9 b	8.9 b	4.0 a
B (145 °C, 453 min)	13.7 ab	8.7 b	5.0 a
C (150 °C, 284 min)	13.1 ab	8.9 b	4.2 a
D (155 °C, 180 min)	14.5 a	10.3 a	4.2 a
E (160 °C, 114 min)	14.4 ab	9.9 ab	4.5 a
F (165 °C, 72 min)	14.2 ab	9.8 ab	4.4 a
G (170 °C, 46 min)	14.2 ab	9.7 ab	4.5 a
H (175 °C, 28 min)	14.0 ab	9.6 ab	4.4 a

Different letters in the same column indicate statistical significance (p-value ≤ 0.05) using Tukey's test.
Letras diferentes na mesma coluna indicam significância estatística (p ≤ 0.05) utilizando o Teste de Tukey.

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CONCLUSIONS

The pulp delignification degree, expressed as the kappa number, was constant with the same H-factor and different combinations of cooking times and temperatures;

The pulping yields tended to be higher when lower cooking temperatures were used. For temperatures higher than 155 °C, the yields tended to decrease with an increase in the temperature;

For temperatures higher than 155 °C, the reject generation tended to increase as the maximum cooking temperature increased, which resulted in a reduction of the screened yield. This increase in the reject content was probably because of the low pulping times;

The alkali consumption during pulping was constant as the H-factor increased with different combinations of pulping times and temperatures;

The total alkali consumption was constant when the same H-factor was applied;

The H-factor is a reliable tool to control and predict the delignification rate and pulping results for *Eucalyptus* wood.

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