Optimization of elemental chlorine-free bleaching for a softwood kraft pulp – part 2: economic analysis of chemical and steam consumption

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ABSTRACT: Our previous investigation [1] re-analyzed the data from Basta and co-workers (1992 TAPPI Pulping Conference) to demonstrate how oxidative alkaline extraction can be augmented and how these changes affect chlorine dioxide consumption with elemental chlorine-free (ECF) sequences. The current study manipulates extraction delignification variables to curtail bleaching costs with a conventional U.S. Southern softwood kraft pulp. The economic advantages of ~0.35% to 0.65% H_2O_2 peroxide reinforcement in a 70°C (EOP)-stage versus 90°C (EO)-stage are predisposed to the brightness targets, to short or long bleach sequences, and to mill energy costs. Minimized bleaching costs are generally realized when a 90°C (EO) is employed in D_0(EO)D_1 bleaching, whereas a 70°C (EOP) is economically advantageous for D_0(EOP)$D_1E_2D_2$ bleaching. The findings we disclose here help to clarify previous ECF optimization studies of conventional softwood kraft pulps.

Application: The findings presented exemplify how oxidative extraction variables can influence overall variable bleached production costs for a variety of economic scenarios. Operators, engineers, and scientists can apply this data as a guide to optimizing softwood pulp bleaching.

In the present day, global markets demand North American mills lower their kraft pulp production costs in order to remain economically viable. Wood typically accounts for 60% to 70% of the variable bleached production costs, with the remainder being comprised of chemical and energy costs. Approximately one-half to two-thirds of this chemical cost for bleached softwood kraft pulps is related to chlorine dioxide (ClO_2) consumption [1-7].

A major contributor affecting ClO_2 consumption in elemental chlorine-free (ECF) bleaching of softwood kraft pulp is the lignin content of the brownstock. The lower the brownstock kappa number is, the lower the amount of ClO_2 that is consumed in delignification (i.e., D_0-stage). This lower kappa number has to be balanced with the constraints of pulping selectivity, both in terms of pulp yield and strength. Oxygen delignification and/or kraft cooking modifications [2-7] can be employed to selectively lower the lignin content; however, the drawback of these approaches is the capital expenditures required to implement them if they are not already a part of the existing pulp mill. Minimizing black liquor carryover with the pulp through improved brownstock washing is another technique for lowering ClO_2 consumption [8-10]. Brownstock washing can be improved through the effective application of defoamers/drainage aids and shower water, as well as manipulating the dilution factor. Adjusting the dilution factor in brownstock washing has to be judiciously balanced such that any bleaching chemical savings are not outweighed in additional energy cost associated with concentrating a more diluted black liquor stream at the evaporators.

Another approach to lower bleaching costs, which has received limited attention, is to employ more aggressive conditions in the oxidative alkaline extraction stage (e.g., (EO) and (EOP)) [1,11-17]. These more aggressive extraction stages utilize hydrogen peroxide and/or higher extraction temperatures (>80°C) in conjunction with oxygen. Van Lierop et al. [15] investigated very aggressive (EO) and (EPO) stages, which employ high extraction temperatures (90°C – 110°C) and/or high peroxide dosages (≥1%). Their work suggested that an (EO)-stage conducted with a pressurized pre-retention tube at 110°C followed by atmospheric extraction was more economically effective at obtaining 3 to 4.5 post-extracted kappa numbers than utilizing high peroxide dosages in the (EPO)-stage, either at moderate or high temperatures (85°C to 110°C). This extraction optimization study primarily focused on bleaching delignification costs rather than overall bleaching costs; the authors assumed that a given brightness increase after extraction would require the same amount of ClO_2 at a fixed post-extraction kappa number for a given bleach sequence.

Interest has been stirred by these contemporary investigations, which manipulate (EO) and (EOP) variables to optimize ECF bleaching [5,13-15]. Most oxidative extraction modifications require little to no capital expenditures to implement with an existing bleach plant [11-15]. The costs of commodity bleaching chemicals, such as sodium chlorate, caustic, hydrogen peroxide and sulfuric acid, as well as energy costs, have been on the rise [10,16-18]. Some of these chemical costs have seen dramatic price fluctuations, such as caustic and sulfuric acid. In this age of volatile chemical and energy prices, is it more sensible to use hydrogen peroxide (H_2O_2) and/or steam in extraction versus applying ClO_2 in delignification and/or brightening? An item absent in previous (EO) and (EOP) studies is how extraction variables affect ClO_2 dosage and balancing in three- and five-stage bleach sequences.

Our earlier investigation [1] rigorously examined how ClO_2 dosages in delignification and brightening stages interact with oxidant-reinforced extraction variables, as well as how these interactions work in concert with each other to affect the overall consumption of bleaching chemicals. This ClO_2 stoichiometric data analysis was based on re-interpreting the extensive bleaching data reported by Basta and co-workers [19], who were focusing on methods to minimize adsorbable organic halide (AOX) generation during ECF bleaching. The intent of this investigation is to exploit the stoichiometric data analysis from our earlier work [1] to evaluate the economic impact of extraction stage variables on the overall bleaching costs with three- and five-stage bleach sequences for various brightness targets.

RESULTS AND DISCUSSION

Methodology

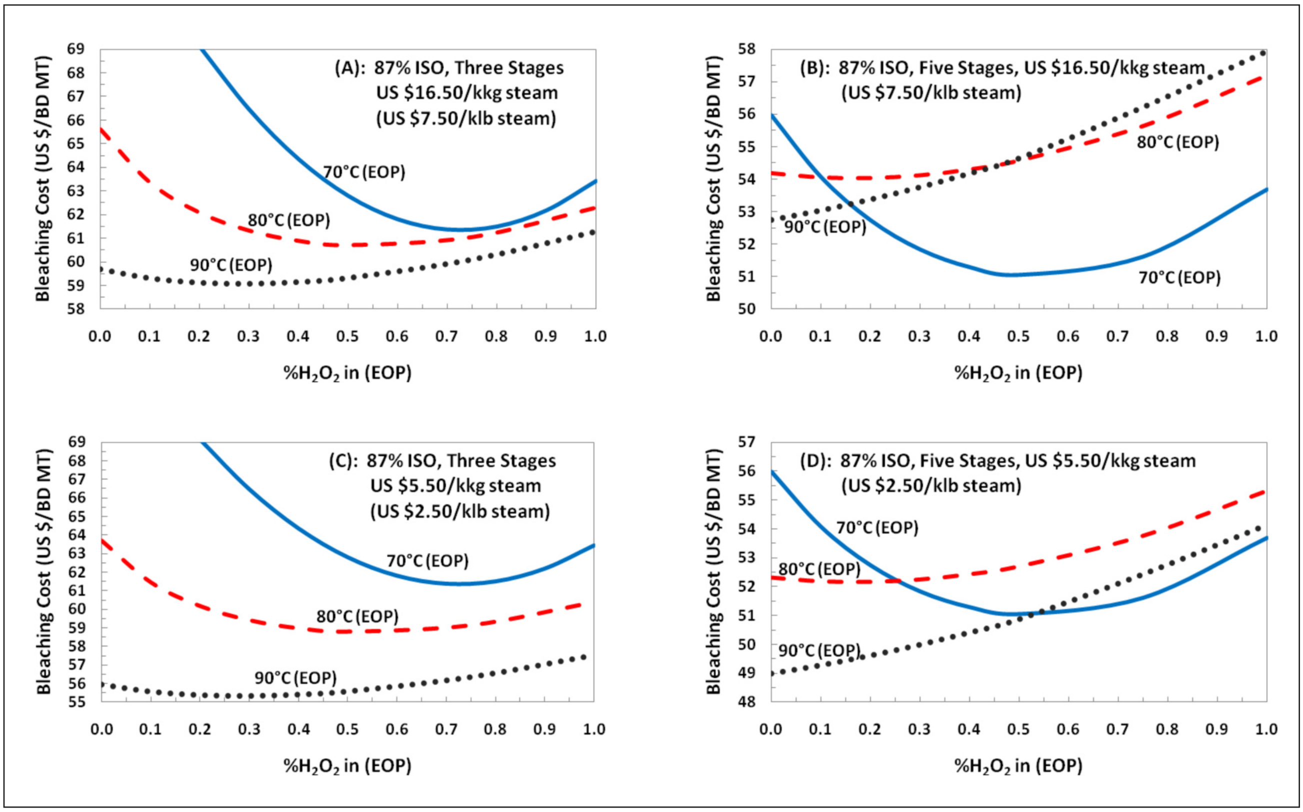
The ECF bleaching data of Basta et al. [19] are meticulously re-analyzed and re-interpreted in this investigation. The details from their experimental bleaching experiments are as follows. A conventional U.S. Southern pine kraft pulp was used that had a kappa number of 24, SCAN intrinsic viscosity of 1052 dm^3/Kg (~27.1 mP•s, TAPPI T-230) and brightness of 23% ISO. The range of conditions for each stage is summarized in Table I for obtaining 83% to 90% ISO brightness. The graphical data of Basta et al. [19] for each stage were re-analyzed by statistical regression to develop models (r^2>0.99), which were discussed in our earlier investigation [1]. These models were used to show how various H_2O_2 levels and reaction temperatures in the oxidative caustic extraction stage, (EO) or (EOP), affected ClO_2 consumption in delignification (D_0) and brightening stages (D_1 and D_2) for short and long bleach sequences.

Several assumptions were made for a hypothetical ECF bleach plant employing the statistical regression model of the previous study [1]. The amount of black liquor carryover with the brownstock was not considered in order to simplify the calculations. Likewise, carryover from ineffective washing from the previous stage was not evaluated. Black liquor carryover can significantly affect bleach chemical consumption, as commented by other investigators [8-10]. It was surmised that ~0.6% H_2SO_4 would be needed to adjust the end pH of the brownstock to ~3 once the ClO_2 reacted in the D_0-stage [2]. The amount of NaOH added in the (EO) or (EOP) was allowed to vary in accordance to the amount of ClO_2 applied in the D_0 stage as active chlorine (= KF•(unbleached kappa no.)); this NaOH:ClO_2 as active chlorine ratio was set at 0.40. It was also assumed that no $MgSO_4$ or bleach stabilizers were needed in extraction to mitigate pulp viscosity losses from the decomposition of H_2O_2 and/or O_2. The efficacy of bleach stabilizers in extraction is highly dependent upon metals profile of the pulp and has been examined by other investigators [15,21-23]

The residence time in the D_1 and D_2 stages was allowed to vary such that the majority of ClO_2 applied was consumed [1]. The amount of NaOH or H_2SO_4 needed to adjust the end pH of the D_1 or D_2 stages was estimated based on the ClO_2 charge into the brightening stage [2,3,20]. Pulp shrinkage (i.e., yield

Stage Conditions	D_0	(EO) or (EOP)	D_1	E_2	D_2
Consistency, %	3.5	10	10	10	10
Temperature, °C	50	70 – 90	70	70	70
Time, min	30 – 60	60	variable	60	variable
Kappa Factor (KF)	0.10 – 0.27	–	–	–	–
ClO_2, % on pulp	0.91 – 2.10	–	0.50 – 1.50	–	0.20 – 0.95
O_2, % on pulp	–	0.50 (at 0.22 MPa)	–	–	–
H_2O_2, % on pulp	–	0 – 1	–	–	–
NaOH, % on pulp	–	0.4•(KF)•(Kappa no.)	variable	0.5	variable
H_2SO_4, % on pulp	0.6%	–	variable	–	variable
End pH	~3	~10.5 to 11.0	~3	~11	~4

I. Conditions used in economic evaluation of three- and five-stage ECF bleach sequences for a 24 kappa Southern pine kraft pulp to reach target brightness levels of 83% to 88% ISO and 84% to 90% ISO, respectively.



1. Effect of steam cost on ECF bleaching cost to reach 87% ISO for various extraction delignification conditions. Calculations are based on stoichiometric data analysis of Brogdon [1] and the conditions in Table I for the following chemical costs held constant: ClO_2 \$1/kg, H_2O_2 \$1/kg, \$0.60/kg NaOH, \$0.10/kg O_2, and \$0.41/kg H_2SO_4.

loss) was postulated to be ~4.3% to 4.8% post-extraction (depending on extraction temperature) [13,14,24,25], in addition to a ~1% shrinkage after D_1 or $D_1E_2D_2$ [24,25].

The amount of steam added to the pulp prior to (EO)- or (EOP)-stage, if needed to attain the required extraction temperature, was calculated based on an incoming temperature of 70°C and 10% pulp consistency. It was further reasoned that the amount of low-pressure steam to raise the 10% consistency stock by Δ 10°C was ~162 Kg steam/metric ton pulp [26]. Calculations of bleaching cost include the chemicals used, as well as additional low-pressure steam consumed in extraction if the (EO) or (EOP) was conducted at >70°C. It was surmised in the calculations of the various circumstances that the mill is steam-limited and that any additional steam needed to operate a bleaching stage at >70°C would have to be generated by external fuel sources. Calculated bleaching costs in this hypothetical ECF bleach plant do not include electrical power consumption for process equipment.

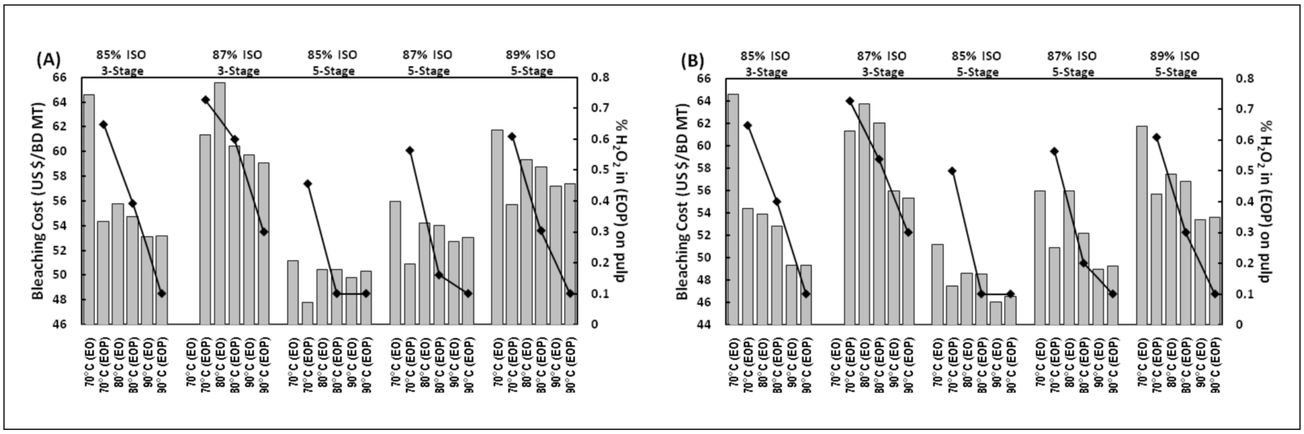
Several economic scenarios will illustrate how changes in (EO) or (EOP) variables favorably or adversely affect bleaching costs. We will scrutinize how steam costs influence the optimum temperature or peroxide level used in extraction. We will also examine what are the optimum extraction conditions as the ClO_2:H_2O_2 cost ratio is varied, in addition to how caustic cost affects extraction operation variables. These various scenarios will be assessed in terms of a three-stage or five-stage bleach sequence, as well as the final brightness target.

Influence of steam costs on bleaching economics

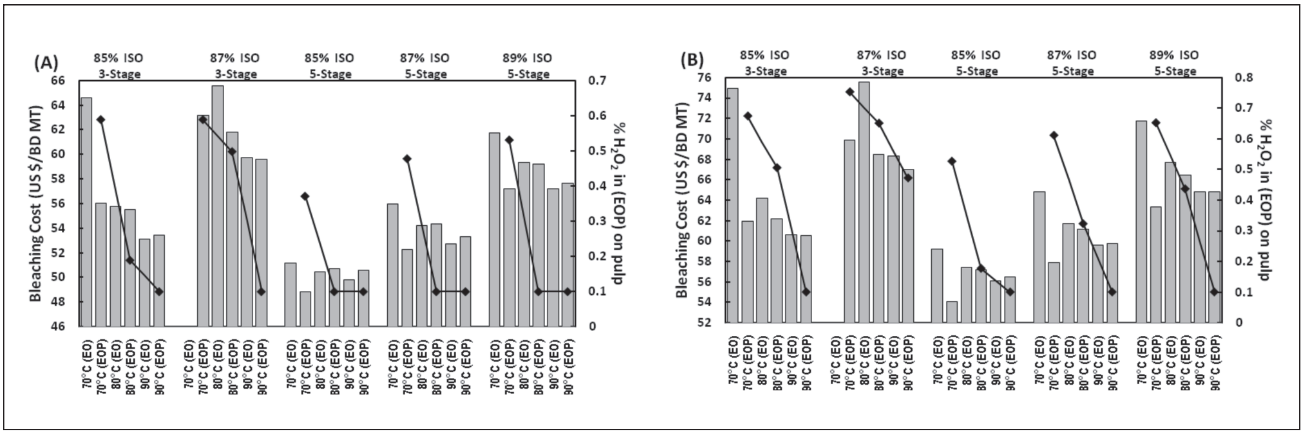
The cost to produce extra steam, if the mill is steam limited, is dependent upon the fuel burned in the auxiliary power boiler. These fuel sources can be hog fuels (e.g., bark, sawdust residues, etc.), fossil fuels (e.g., coal or oil) and natural gas. The cost of generating the steam can vary considerably, from ~\$4 to \$5.50/kkg generated from coal or purchased bark, ~\$16 to \$21/kkg generated from natural gas and ~\$20 to \$30/kkg generated from fuel oil. Typical average steam costs for U.S. mills tend to run \$16 to \$20/kkg, which are close to that observed for natural gas [27,28].

Figure 1 illustrates the impact on variable bleaching costs to reach 87% ISO brightness at two different steam costs for a given set of fixed chemical costs. Increasing the extraction temperature of D_0(EO)D_1 or D_0(EOP)D_1 sequence can effectively reduce bleaching costs if steam costs are low (e.g., \$5.50/kkg (\$2.50/klb)). Similar trends are seen with three-stage bleaching to 87% ISO as steam costs increase from \$5.50/kkg to \$16.50/kkg.

A different set of cost curves are observed when bleaching to 87% ISO utilizing a five-stage sequence (Figs. 1b and 1d).



2. Summarized ECF bleaching cost for various extraction delignification conditions for steam costs (a) \$16.50/kkg and (b) \$5.50/kkg. Calculations are based on stoichiometric data analysis of Brogdon [1] and the conditions in Table I for the following chemical costs held constant: ClO_2 \$1/kg, H_2O_2 \$1/kg, \$0.60/kg NaOH, \$0.10/kg O_2, and \$0.41/kg H_2SO_4.



3. Summarized ECF bleaching cost for various extraction delignification conditions for (a) ClO_2 \$1/kg and H_2O_2 \$1.25/kg, and (b) ClO_2 \$1.25/kg and H_2O_2 \$1/kg. Calculations are based on stoichiometric data analysis of Brogdon [1] and the conditions in Table I for the following costs held constant: steam \$16.50/kkg, \$0.60/kg NaOH, \$0.10/kg O_2, and \$0.41/kg H_2SO_4.

Increasing the extraction temperature from 70°C to 80°C generally results in higher bleaching costs. The higher cost of an 80°C extraction stage is diminished if the temperature is raised to 90°C when steam costs are low. A 70°C (EOP) stage is more favored than a 90°C (EO) stage when steam costs are high, whereas the converse is true when steam costs are low.

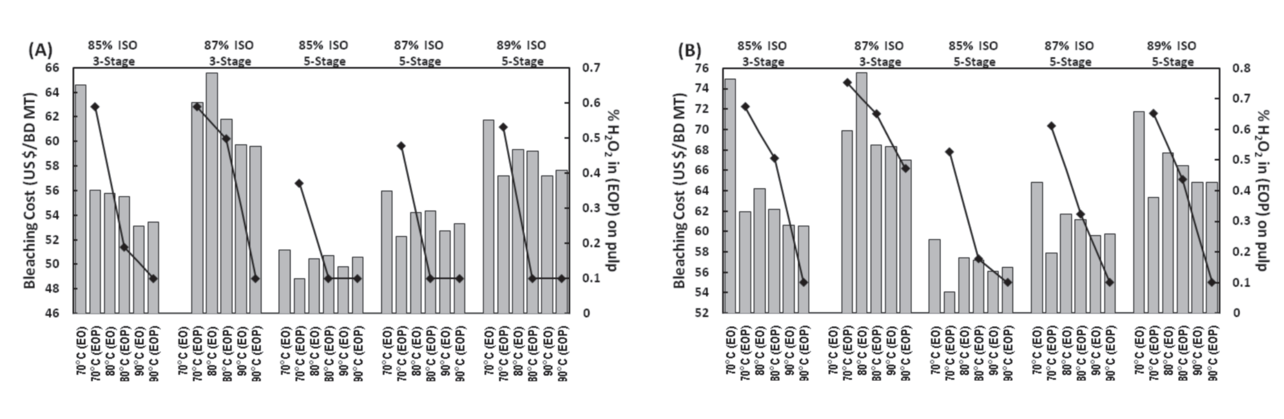
Figure 2 summarizes the effect of low and high steam costs on the economics of three- and five-stage bleach sequences for different brightness targets. Low steam costs tend to favor a 90°C (EO) over a 70°C (EOP) with either three- or five-stage bleach sequences. This preference shifts to favor the low temperature (EOP) for five-stage bleaching to 85% to 89% ISO brightness when steam costs are elevated.

Influence of chemical costs on bleaching economics

Our next case study considers how the cost of chlorine dioxide and peroxide, relative to each other, affect the optimum conditions of the extraction stage on overall bleaching economics. In this particular scenario, we will hold the cost of steam at \$16.50/kkg (\$7.50/klb). The cost of chlorine dioxide varies considerably from mill to mill, depending on the process employed for on-site generation, the negotiated contract price of raw materials (e.g., sodium chlorate, methanol, sulfuric acid, etc.), and the delivery cost of raw materials, just to name a few items [29,30]. Likewise, the price of hydrogen peroxide is variable from mill to mill, depending on the negotiated contracts for price and delivery. Historically, the ratio of ClO_2-to-H_2O_2 costs tends to range from ~0.7 to 1.4.

Figure 3a illustrates the effect of H_2O_2 price increase from \$1.00/kg to \$1.25/kg, while maintaining ClO_2 cost at \$1.00/kg. The bleaching costs are similar to those seen in Fig. 2b with some subtle differences. Higher peroxide cost tends to lessen the differences between operating a 70°C (EOP) as compared to a 90°C (EO). The other difference observed with the five-stage bleach sequence under these economic conditions is that the amount of H_2O_2 used in (EOP) drops down to 0.1% for stages operated above 70°C.

Figure 3b exemplifies the consequence of ClO_2 price increase from \$1.00/kg to \$1.25/kg, while sustaining H_2O_2



4. Summarized ECF bleaching cost for various extraction delignification conditions for (a) NaOH \$0.30/kg, and (b) NaOH \$0.90/kg. Calculations are based on stoichiometric data analysis of Brogdon [1] and the conditions in Table I for the following costs held constant: steam \$16.50/kkg, ClO_2 \$1/kg, H_2O_2 \$1/kg, \$0.10/kg O_2, and \$0.41/kg H_2SO_4.

costs at \$1.00/kg. Although the overall bleaching costs are higher than those seen in Figs. 2b or 3a, the trends in these economic scenarios are analogous. Short bleaching sequences yield lower overall bleaching costs when a 90°C (EO) is used, whereas the five-stage bleach sequence has lower overall bleaching cost when a 70°C (EOP) is employed. The higher cost of ClO_2 results in a small increase in the optimum amount of H_2O_2 applied in the 70°C or 80°C (EOP).

Our second case study explores the impact of caustic cost on the most economical extraction stage conditions. The amount of caustic consumed in ECF bleaching is significant and is generally the second most used chemical in comparison to ClO_2. Caustic prices tend to fluctuate depending on energy costs and industrial market demands. In the past, this particular bleaching chemical was a low-cost commodity product, which typically varies from ~\$0.20 to \$0.40/kg [16,17]. The cost of NaOH underwent some significant and erratic cost increases in 2008, from ~\$0.40 to ~\$1.00/kg, due to rapid increases in energy prices, and imbalances with supply/demand of chlor-alkali products from various industrial manufacturing sectors.

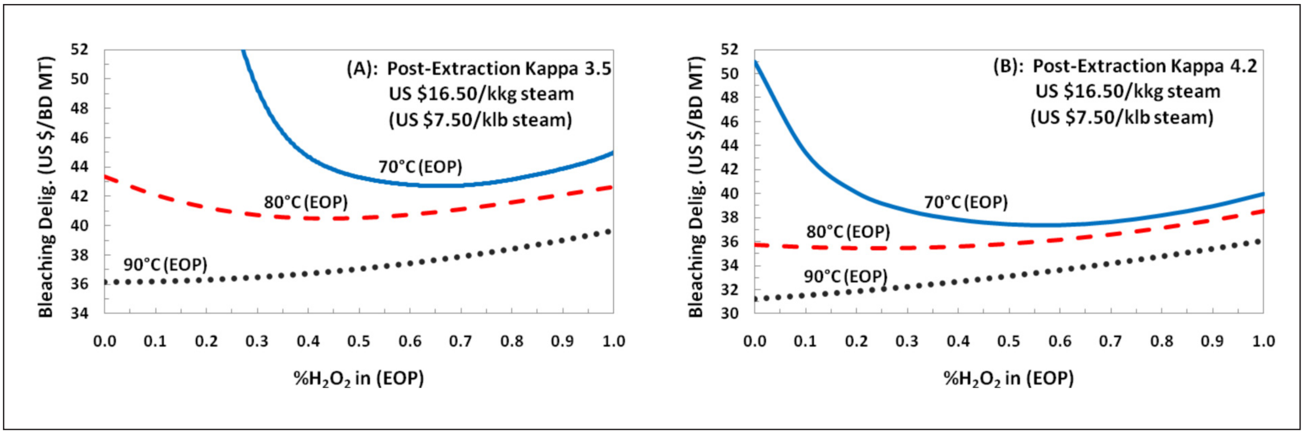
In this particular cost analysis, the prices for steam, H_2O_2, and ClO_2 were set at \$16.50/kkg (\$7.50/klb), \$1.00/kg, and \$1.00/kg, respectively. The cost of caustic was varied from \$0.30/kg to \$0.90/kg. **Figure 4** shows the results of this analysis. The relative trends for the optimum extraction conditions are similar to those observed earlier in Figs. 2 and 3 for changes in steam and ClO_2 costs, respectively.

General discussion

The results from these various assessments revealed some interesting general trends concerning the ECF bleaching of a 24-kappa U.S. Southern pine pulp. A hot (EO)-stage operated at ~90°C yielded lower overall bleaching costs for obtaining 83% to 87% ISO brightness with three-stage bleach sequences under various chemical costs and steam <\$22/kkg (\$11.50/klb) steam. On the other hand, a 70°C (EOP)-stage usually resulted in lower overall bleaching costs for reaching 84% to 90% ISO brightness with five-stage bleach sequences with >\$5.50/kkg (\$2.50/klb) steam. The optimum amount of H_2O_2 for the 70°C (EOP)-stages was ~0.35% to 0.65% on pulp for the D_0(EOP)$D_1E_2D_2$ sequence. More often than not in these various economic senarios, the operation of an 80°C (EO)- or (EOP)-stage was more expensive than either 70°C (EOP) or 90°C (EO).

The differences in the optimum extraction conditions for three- and five-stage bleaching sequences can be explained by considering how aggressive bleaching delignification needs to be. Short bleaching sequences require lower post-extraction kappa numbers entering into the D_1 stage versus longer sequences. This can be seen in our previous study for reaching 87% ISO [1]. The D_0(EO)D_1 or D_0(EOP)D_1 sequences generally require a post-extraction kappa number of ~3.5 opposed to ~4.2 for D_0(EO)$D_1E_2D_2$ or D_0(EOP)$D_1E_2D_2$. **Figure 5** illustrates the costs to delignify the unbleached Southern pine pulp to these post-extraction kappa numbers at various extraction conditions. The curves to reach a 3.5 post-extraction kappa mirror the total bleaching costs observed in Fig. 2 for D_0(EO)D_1 or D_0(EOP)D_1. Under these circumstances, it is generally harder to reach 3.5 kappa utilzing a 70°C or 80°C (EOP) when compared to a 90°C (EO). The five-stage sequences do not require the low post-extraction entering into the D_1; under these conditions, it is relatively easy to obtain a post-extraction kappa of 4 to 4.5 utilizing a 70°C (EOP) as it is to employ a 90°C (EO).

The results in this study help put into context previous reports of bleaching optimization by manipulating extraction delignification varibles [11-15,21-23,29,31]. Van Lierop et al. [14] concluded that a hot (EO) was more economically effective than an (EOP) for reaching >87% ISO brightness. These findings are confirmed by this investigation for three-stage short bleach sequences and can be generalized with such sequences used to obtain 83% to 87% ISO brightness. However, the conclusions of van Lierop et al. [14] are less applicable when ex-



5. ECF bleaching delignification costs to reach (a) 3.5 and (b) 4.2 post-extraction kappa number for various extraction delignification conditions. Calculations are based on stoichiometric data analysis of Brogdon [1] and the conditions in Table I for the following chemical costs: ClO_2 \$1/kg, H_2O_2 \$1/kg, \$0.60/kg NaOH, \$0.10/kg O_2, and \$0.41/kg H_2SO_4.

amining the five-stage bleach sequences, particularly where steam costs are not negligible. Renders and co-workers [29] suggested that an (EOP)-stage was economically advantageous when the ClO_2-to-H_2O_2 cost ratio was ≤0.86. Our study suggests that steam cost and the length of the bleach sequence tend to have a greater influence on the economics of H_2O_2 for ClO_2-to-H_2O_2 cost ratios ranging from 0.80 to 1.25.

Although the stoichiometric bleaching data analysis [1] employed in this economic study did not cover four-stage bleach sequences (i.e., sequences without a full-blown E_2-stage), some generalizations can be interpolated. The brightening efficacy of D_1D_2 is lower than $D_1E_2D_2$ while higher than that of a single D_1 stage [32,33]. Hence, it would be reasonable to deduce that a four-stage bleach sequence bleaching to 85% to 89% ISO brightness would be between that of the three- and five-stage sequences. Based on this reasoning, it is likely that a 70°C (EO) in D_0(EOP)D_1D_2 yields similar bleaching costs as a 90°C (EO) in D_0(EO)D_1D_2. The D_1D_2 brightening sequence that has neutralization at the bottom of the D_1-stage (i.e., D_N-stage) yields higher brightness than D_1D_2 without neutralization and approaches that of $D_1E_2D_2$ [32-34]. Hence, it would be expected that 70°C (EOP) in D_0(EOP)D_ND_2 affords lower bleaching cost, similar to that seen for the five-stage D_0(EOP) $D_1E_2D_2$.

CONCLUSIONS

The results from our investigation, based on the bleaching data of Basta and co-workers [19], illustrate how elevated extraction temperatures, H_2O_2 reinforcement, and combinations thereof can affect the overall bleaching costs in three- and five-stage ECF sequences for a Southern softwood kraft pulp. Lowest bleaching cost for a three-stage bleach sequence favors a hot 90°C (EO)-stage for obtaining 83% to 87% ISO brightness. Alternatively, a 70°C (EOP)-stage with ~0.35% to 0.65% H_2O_2 is more economically favored for five-stage sequences for obtaining 85% to 89% ISO brightness. The results from this economic analysis of various chemical and steam costs elucidate several past ECF optimization studies of conventional softwood kraft pulps in which extraction delignification variables were manipulated.

ACKNOWLEDGEMENTS

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ABOUT THE AUTHORS

Mill personnel seek to optimize their operations to lower production costs. Too often, managers and accountants focus on budgeted chemical allocations instead of individually adjusting each chemical to achieve maximum efficiency and economy for the entire bleach sequence. This "silo thinking" can limit process optimization and overall cost minimization, particularly in this age where chemical and energy costs are in flux. Our goal was to show how caustic extraction variables affect bleach plant costs from a holistic perspective. Very few scientists have investigated how extraction affects bleaching economics.

The challenges with this study were constructing a steady-state spreadsheet model of a hypothetical bleach plant based on our previous publication data analysis, and utilizing the model to evaluate how bleaching variables impacted bleach plant costs. Operators, engineers, and managers can use the information in this study and our earlier study as a guide of how to alter process conditions/targets when the cost of bleaching chemicals and energy fluctuate. Also, the information presented in these two studies can serve as a generalized reference for the ECF bleaching of conventional softwood kraft pulps.

We demonstrate how peroxide reinforcement and/or higher extraction temperatures affect bleach plant costs for different economic situations. In certain circumstances, it is advantageous to use additional peroxide reinforcement, and in other scenarios, increasing extraction temperature is beneficial.

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Many of our findings complement and clarify the conclusions from other ECF optimization studies, and we did uncover some surprises. In many of the cases we examined, a 90°C (EO) tended to be more economical than a 70°C (EOP) for short sequences (three-stage), and the converse was true for extended sequences (five-stage). A second revelation was that an 80°C oxidant-reinforced extraction was generally higher in costs than either a 70°C (EOP) or a 90°C (EO).

The next step in our research is to examine how caustic extraction variables affect chemical consumption and economics for ECF hardwood pulp bleaching.

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