

# PULP BLEACHING CONTROL AND OPTIMIZATION

Danielle MAJOR,\* Michel PERRIER,\*<sup>1</sup>  
Sylvain GENDRON,\*\* and Bernard LUPIEN\*\*\*

\* *Department of Chemical Engineering*  
*École Polytechnique de Montréal, Québec, Canada*  
\*\* *Pulp and Paper Research Institute of Canada*  
\*\*\* *Papier Masson Ltée, Québec, Canada*

Abstract: The control of bleaching and brightening reactions is important in the pulp and paper industry to ensure quality of the final product. This paper presents a review of the principal fundamental and practical aspects to consider when designing control strategies for bleaching and brightening operations. An approach is also suggested to minimize the operating costs of bleaching sequences.

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## 1. INTRODUCTION

Pulp used in the paper making process can be made whiter by several methods. These methods can be grouped in two categories: bleaching and brightening. The distinction is made between brightening and bleaching but the term bleaching is also often used to describe brightening operations.

Chemical pulps are bleached by the addition of chemicals such as chlorine, chlorine dioxide or hypochlorite. The bleaching process removes lignin from the pulp, which reduces the yield of a pulp produced from a given initial quantity of wood. Bleached pulps generally give stable high-brightness pulps.

Mechanical pulps are brightened by the addition of chemicals such as hydrogen peroxide or sodium hydrosulfite. These chemicals remove the chromophores that impart colour to the pulp while maintaining the high yield that is characteristic

of mechanical pulping processes. However, the resulting brightness can undergo significant reversion, thus limiting the use of brightened pulps, for example to newsprint grades.

The modelling, control and optimization of pulp bleaching processes are reviewed in this paper. The structure is organized as follows. An overview of existing bleaching and brightening technologies is given in terms of the variables that can be manipulated in control and optimization strategies. A special emphasis is given on bleaching and brightening kinetic models because they are particularly important for the determination of the dynamics of a process. The limitations of these kinetic models in the context of the development of control strategies are also explored further. A review of existing control strategies for brightening and bleaching processes is then given. Many studies have examined the implementation of control strategies to bleaching processes, but little has been published on the control of mechanical pulp brightening. The limitations of these control strategies for optimization purposes are

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<sup>1</sup> Corresponding author : Michel.Perrier@polymtl.ca

briefly outlined. Finally, an optimization strategy for multi-stage processes is introduced.

## 2. MODELING OF THE BLEACHING STAGES

The control of any process is generally preceded by a careful modeling of its behavior. When available, knowledge of the underlying principles (be they physical, chemical or mechanical) guide the investigator in choosing appropriate model structures for a given process. The stoichiometry and kinetics of many bleaching sub-processes have been investigated. Unfortunately, the mechanisms involved are extremely complex and rarely lend themselves to direct observation such that it is difficult to verify even well founded hypotheses. The degree of development of "bleaching models" is not uniform over the array of existing bleaching and brightening technologies. Roughly, one would expect the degree to which a given technology has been investigated to be proportional to its age and to the number of installations. Consequently, in the following review, the bleaching technologies have been loosely divided in "conventional" technologies (chlorine, chlorine dioxide, alkaline extraction, hypochlorite), "emerging" technologies (oxygen, ozone, peroxide) and brightening operations.

### 2.1 Variables Affecting Bleaching

The kinetic models discussed in the literature try to describe and/or explain the chemical and physical interactions between the bleaching agents and the pulp fibers. Besides the respective proportions of lignin and bleaching chemical, there are a number of variables which affect the reactions. Most of these effects are common to all bleaching/brightening stages but their importance or function may vary from one stage to another.

**Mixing** In any bleaching or brightening reaction, the homogeneity of the mixture composed of pulp (made up of liquid and solids) and bleaching agent (gas and/or liquid). This involves proper pulp consistency control and an appropriate choice of the mixing technology.

**Pulp pre-treatment** The behavior of a bleaching/brightening may be affected to varying degrees by the presence of pulping by products or metallic ions. Pulp washers solve in good part the potential problems associated to these elements. However, some stages (e.g. peroxide) may require special treatment of the pulp with chelating agents.

**Temperature** It is generally agreed that temperature affects positively the quantity of lignin that will be made to react with the bleaching

agent. Temperature generally enters the models as a nonlinear coefficient of a rate equation (Axegard *et al.* (1984); Germgard and Karlsson (1985a); Devenyns *et al.* (1995); Hsu and Hsieh (1991a); Myers and Edwards (1991)). Temperature was also recently shown to play a central role in peroxide bleaching. It has been established that bleaching with peroxide at a temperature beyond a certain threshold greatly intensifies the reaction rate, considerably reducing the time required to achieve a certain brightness increase.

**pH** The alkalinity or acidity of the solution as measured by its pH plays an important role in every bleaching stages (McDonough and Reeve (1991a); Reeve (1991); Daneault *et al.* (1995); Axegard and Tormund (1985); Gellerstedt *et al.* (1985); Kutney and Evans (1985a); Kutney and Evans (1985b); van Lierop *et al.* (1991); Hsu and Hsieh (1991b); Myers and Edwards (1991)). It may act as a lever for modifying the reaction rate or act as an observer of some internal mechanisms of the reaction. The optimal pH varies from one stage to another. This is discussed in greater details later.

**Pulp strength** Preserving the strength properties of the bleached pulp is an issue for every bleaching sequence. Attacking pulp with a bleaching chemical may actually weaken its fibers if the chemical is inclined to react with the carbohydrates and break the cellulose chain. Selectivity, a term used to describe how the bleaching chemical prefers reacting with lignin instead of carbohydrates varies widely from one chemical to another and will thus be individually discussed.

### 2.2 Variables Affecting Brightening

#### Process Conditions

Mechanical pulps are usually brightened with sodium hydrosulfite or with hydrogen peroxide for high brightness grades. New classes of brightening chemicals that are effective over a wide range of process conditions are also emerging, such as BBHPE, recently presented by Hu and James (2005). In this section, more emphasis is given on sodium hydrosulfite brightening because it presents many interesting control and optimization challenges. The brightening of mechanical pulps with hydrosulfite is accomplished by a reduction reaction under good mixing and in the absence of air. Compared to other chemicals, the addition of hydrosulfite to pulp results in lower brightness increments; however the popularity of hydrosulfite is ensured by its low cost, which makes it an ideal chemical for the brightening of newsprint grades.

Many mechanical pulp mills inject hydrosulfite at two locations in the process in order to control the

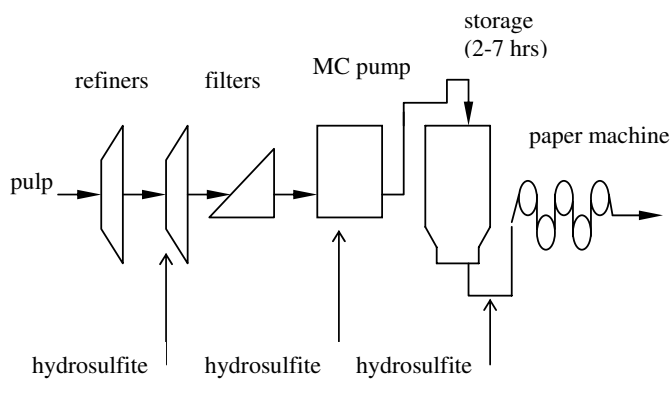


Fig. 1. Simplified diagram of a thermomechanical pulp process with three hydrosulfite injection points.

final brightness of the paper. The main injection point is located a few hours upstream from the paper machine where a relatively large amount of hydrosulfite is used, usually in an upflow tower. The second injection point is located half an hour from the paper machine and smaller amounts of hydrosulfite are used to quickly compensate for unexpected decreases in paper brightness. Some pulp mills also inject small amounts of hydrosulfite further upstream than the main injection point, such as in the refiner, in order to give an initial brightness to pulp that is particularly dark due to seasonal variations in wood quality. At any given injection point, pulp brightness increases as a function of hydrosulfite dosage up to a certain point, beyond which brightness reaches a plateau and any additional amount of hydrosulfite is wasted. A thorough discussion of the process conditions and equipment required for hydrosulfite brightening is given by Ellis (1996). An example of a thermomechanical pulp line that has three hydrosulfite injection points is given in Figure 1. It should be noted that the most important source of delay in this process is the storage tower, in which the residence time can vary between 2 and 7 hours. This residence time cannot always be predicted by the flowrates and the level in the tower because of channeling problems.

### Incoming Brightness and Brightening History

In order to develop control and optimization strategies for a hydrosulfite brightening process that has several injection points, better knowledge of the effect of incoming brightness on the brightness response at a given injection point is required.

Mill experience suggests that incoming brightness has a strong effect on the brightness response, but the literature on this topic remains unclear. Mongrain *et al.* (2004) used a brightness soft-sensor which estimated the resulting brightness

following the addition of hydrosulfite to pulp of a known measured brightness. The brightness gain used in the soft-sensor was chosen to be constant, despite fluctuations in incoming brightness. The use of a constant soft-sensor gain could therefore overestimate the calculated brightness beyond the maximum possible brightness.

Crawford (1969) reported that the brightness increment is independent of the initial brightness in his study of hydrosulfite brightening with 3% consistency radiata pine groundwood. In some hydrosulfite brightening studies, results are reported in terms of a brightness "gain", which is increment in number of brightness points that a pulp sample has undergone due to its exposure to hydrosulfite. The initial brightness is not always stated when such brightness increment results are given, which makes it difficult to quantify the effects of incoming brightness. For example, Ellis (1996) gives the brightness increment as a function of the hydrosulfite loading for thermomechanical pulp at 4% consistency without indicating the initial brightness. Similarly, Fluet *et al.* (1994) report brightness increment results under different laboratory methods without stating the initial brightness.

Nevertheless, some authors do give brightness increment results along with the initial brightness. These include Gupta and Mutton (1969) and Ingruber and Kopanidis (1967), but they do not discuss the effect of incoming pulp brightness. An example of a case where the changes in incoming brightness are acknowledged is McArthur *et al.* (1988) study of medium consistency brightening where the results are reported by averaging the brightness increments, regardless of incoming brightness variations. Another example is in Joyce and Mackie (1979) study with 4% groundwood which shows that the brightness increment at a given hydrosulfite charge is the same despite variation in initial brightness, except in the case of extremely bright incoming wood which will significantly reduce the possible brightness increment.

In the case of two-stage peroxide brightening, the brightness resulting from 1st stage bleaching can be used to predict the effects of bleaching in the second stage. For example, Hook and Wallin (1989) supply a constant hydrogen peroxide charge to the first stage and use the resulting brightness as an indicator of the bleachability of the pulp entering the second stage. Reliable pulp brightness sensors are required to successfully use this strategy.

### 3. BLEACHING KINETIC MODELS

In what follows, we survey the particularities of the kinetic models for individual bleaching stages.

**Chlorine** For a long time, elemental chlorine ( $\text{Cl}_2$ ) has been the choice chemical for attacking the remaining lignin of kraft pulp. Because of environmental concerns however, it is rapidly being displaced by chlorine dioxide ( $\text{ClO}_2$ ) (Pryke, 1991). Its main advantage over chlorine dioxide is its lower price. Chlorine attacks lignin directly by dissolving part of it and rendering an even greater part of it soluble in the subsequent stage (McDonough and Reeve, 1991b).

Chlorine is first dispersed in water as small bubbles and the gas carrying liquid then diffuses through the pulp slurry with the help of (typically static) mixers. The mixing of pulp with chlorine is immediately followed by a sharp decrease of the lignin content of the pulp. After a few minutes however, the delignification reaction dramatically slows down and eventually stops (Berry and Fleming (1987); McDonough and Reeve (1991b); Germgard and Karlsson (1985b); Ni *et al.* (1990) Ackert *et al.* (1975); Karter and Bobalek (1971)). The work of Berry and Fleming (1987) and Ni *et al.* (1990) provides insight into the fundamental chemical mechanisms involved but does not provide an explicit mathematical description of the delignification reaction. The hypotheses formulated to explain this apparent shift in delignification regime attribute it to chemical reactions concurrent to the expected reactions associated to delignification. The net effect of these concurrent reactions is to progressively deplete the amount of lignin amenable to delignification by chlorine. Ni *et al.* (1990) established a parallel between the delignification reaction proper and the production of methanol and identify aromatic substitution as the inhibiting mechanism. A corollary of these studies is that the effect dominating the reaction rate of chlorine with lignin is of a chemical nature as opposed to earlier hypotheses based on physical rate determining steps (Pryke and McDonough (1991); Karter and Bobalek (1971)). The latter studies propose that the delignification reaction is accompanied by the build up of physical barriers preventing chlorine to come in contact with unreacted lignin. Based on this assumption, Karter and Bobalek (1971) proposes an explicit mechanistic mathematical model of the delignification process. A consequence of these "blocking" mechanisms is that the extraction stage must play a dual role: 1) dissolving of the chlorinated lignin and 2) removal of the inhibitors to render the lignin available for further delignification in subsequent stages. Finally, simple empirical models are shown to well approximate experimental data in Edwards *et al.* (1973).

**Chlorine dioxide** While being more expensive than elemental chlorine, chlorine dioxide has the following advantages over the former: It reacts very little with carbohydrates and is thus more

capable of preserving pulp strength (Reeve (1991); Germgard and Karlsson (1985b)). It is a more powerful oxidizing agent than chlorine (Smook (1992); Pryke (1991)) and it is thought to have less detrimental effects on the environment than chlorine (Pryke, 1991). Chlorine dioxide was originally used in the end stages of a bleaching sequence to do the final removal of the remaining lignin. It was also substituted in small quantities to chlorine in the first stage to protect the cellulose (McDonough and Reeve, 1991b). With the recent drive toward elemental chlorine free (ECF) bleaching, chlorine dioxide is rapidly displacing chlorine as the main bleaching agent for both softwood and hardwood kraft pulps (Pryke and McKenzie, 1996). When chlorine dioxide is applied to the pulp, it is normally in the form of a dilute solution which is considerably cooler than the pulp. Consequently, part of the chlorine dioxide may vaporize. A particularity of chlorine dioxide bleaching then consists of building up a sufficient hydrostatic pressure in order to keep the  $\text{ClO}_2$  solution in close contact with the pulp. As with chlorine, the mixing of chlorine dioxide with pulp is immediately followed by a rapid decrease in the pulp lignin. The reaction then progressively shifts to a much slower delignification regime as is observed with chlorine. However, here appears a major distinction between chlorine and chlorine dioxide bleaching: the ultimate lignin content (i.e. the lignin as  $t \rightarrow \infty$ ) is substantially smaller with  $\text{ClO}_2$  bleaching than with  $\text{Cl}_2$  bleaching. This result appears in separate laboratory tests of chlorine dioxide bleaching (Teder and Tormund (1977); Rapson and Anderson (1985)). In particular, Rapson and Anderson (1985) show that all the lignin can be removed by a single D stage with sufficient  $\text{ClO}_2$  charge. Ni (1992) describes the chemistry of chlorine dioxide delignification in great details and attribute the reported effectiveness of  $\text{ClO}_2$  bleaching by the local generation of hypochlorous acid. However, no explicit mathematical model of delignification is proposed in this work. Teder and Tormund (1977) developed an exhaustive empirical model for  $\text{ClO}_2$  bleaching where the extent of bleaching is expressed as the concentration of chromophore groups. The latter study is also the only one which has attempted to quantify the interaction between  $\text{ClO}_2$  and  $\text{Cl}_2$  which are often combined in a bleaching stage. Finally, note that Parming and Backlund (1991) have studied the effect of the lignin in the liquor carry over from the previous stage on consumed  $\text{ClO}_2$ .

**Caustic extraction** Caustic extraction is the usual companion process to the chlorination (or  $\text{ClO}_2$ ) stage. Its purpose is to solubilize the reacted elements of the pulp from the previous stage and to "prepare" the pulp for further delignifica-

tion. While the chemistry of alkali treatment is not completely known (Singh and Atkinson, 1979), extensive experimental studies offer well documented qualitative and quantitative description of the process (van Lierop *et al.* (1991); Axegard (1970)). An interesting outcome of the study by van Lierop *et al.* is that the solubilization of reacted lignin is fully achieved provided that the end pH is greater than some threshold in the vicinity of 10. For various process conditions, the rate of lignin solubilization (as measured by Kappa number) was shown to be a monotonic decreasing function of time with a "fast" regime followed by a "slow" regime as is also found in other bleaching stages (Axegard, 1970).

**Hypochlorite bleaching** Hypochlorite is known to easily brighten pulp by attacking some specific chromophoric groups of lignin (Smook, 1992). Despite its brightening efficiency, its use is normally limited to a single intermediate stage in a multi stage sequence because it also voraciously attacks carbohydrates and may cause severe degradation of pulp strength. While an acidic solution ( $\text{pH} < 3$ ) favors the generation of elemental chlorine, an alkaline solution favors the generation of hypochlorite ions. Consequently, the pH of an hypochlorite stage is generally greater than 9. For the range  $\text{pH} > 9$ , the rate of chromophore removal is relatively insensitive to pH variations while increasing temperature may greatly accelerate chromophore removal (Axegard and Tormund, 1985). This study by Axegard and Tormund estimates empirical models not only for chromophore removal, but also for carbohydrate degradation (as expressed by the inverse of the degree of polymerization) and for shives removal.

**Emerging bleaching technologies** Increasing environmental concerns over the by products generated by chlorine and chlorine dioxide stages has driven the search for alternative bleaching agents. Furthermore, effluents containing even minute amounts of chlorine may cause corrosion problems and thus cannot be recovered. The bleaching chemicals listed below are friendlier to the environment and can be safely recovered thus offering the possibility for mill closure. Although not really new, these bleaching technologies are getting increasing attention because of recent process developments that make their industrial utilization more attractive. They are oxygen, ozone and peroxide bleaching.

**Oxygen** Developed in the late 60's, oxygen bleaching is being usually limited to a "pre bleaching" stage to remove some portion of the lignin of unbleached pulp (25–50%). Also, most installations introduce small amounts of oxygen in the caustic extraction stage. The net effect of using oxygen is to reduce the required amount of

chlorine and chlorine dioxide in subsequent stages (Smook, 1992). Oxygen delignification is usually not allowed to delignify more because of its poor selectivity, i.e. it is capable of breaking up cellulose chains and weakening the pulp (Smook (1992); McDonough and Reeve (1991b)). Metal ions present in the pulp (copper, iron, manganese) appear to act as intermediate catalyzing agents in the breakage of the cellulose chains. The kinetics of oxygen bleaching has been investigated in several studies (Olm and Teder (1979); Myers and Edwards (1991); Kutney and Evans (1985a); Hsu and Hsieh (1991a)). All studies identify temperature, alkalinity and partial pressure of oxygen as the factors influencing the rate of delignification. The study by Olm and Teder further investigates the effect of the above factors on the rate of cellulose chain breakage. There is no final agreement on the exact form of a model but all studies agree that the delignification reaction goes through a step of rapid delignification followed by a slow delignification regime. This is expressed in different ways: a sum of two (slow and fast) first order rate equations (Olm and Teder, 1979), a set of non linear rate equations active on different time segments (Hsu and Hsieh (1988); Hsu and Hsieh (1991a); Hsu and Hsieh (1991b)) or a sum of three (slow, fast and bottom level) first order rate equations (Myers and Edwards, 1991).

**Ozone** Ozone ( $\text{O}_3$ ) is a relatively new bleaching agent. Its main advantage is that within the array of non chlorine based bleaching agents it is the chemical most reactive with lignin (Chirat and Lachenal (1995); Lapierre *et al.* (1995)). It has been reported that the insertion of an ozone stage inside a sequence of peroxide treatments considerably enhances the brightening capability of the whole sequence. However, its poor selectivity and the relative inefficiency of ozone generating equipment have contributed so far to its limited industrial use in North America. The chemistry and kinetics of ozone delignification and its effect on pulp strength was recently investigated; the results are reported by van Lierop *et al.* (1996).

**Peroxide** Since hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) selectively attacks certain chromophoric groups of the fiber without destroying the lignin, it is often used for brightening mechanical pulps with little or no yield loss (Smook (1992); Daneault *et al.* (1995)). Under certain conditions, it may perform some delignification and be also used to bleach kraft pulps when total chlorine free (TCF) bleaching is wanted (Lapierre *et al.*, 1995). It has been recognized that the perhydroxyl ion ( $\text{OOH}^-$ ) and perhydroxyl radical ( $\text{OOH}\cdot$ ) both produced by the decomposition of peroxide are responsible for brightening ( $\text{OOH}^-$ ) may modify some chromophoric groups) or delignification ( $\text{OOH}\cdot$ ). On the other hand, the perhydroxyl radical is not se-

lective and will attack cellulose. Its concentration is accelerated by the presence of metal ions and it is thus critical to rid the unbleached pulp of these ions through chelation and washing prior to peroxide application.

The decomposition reaction towards OOH may be favored in the reaction tower by appropriate alkaline conditions (Daneault *et al.*, 1995) or by changing the partial pressure of oxygen in the reaction vessel (Stromberg, 1995). Careful control of the bleaching conditions must be ensured for if, for instance the pH of the solution is pushed too high, the reaction may favor the generation of oxygen (Daneault *et al.*, 1995) which may actually cause the pulp to darken. It has recently been found that peroxide bleaching of kraft pulps performed at temperatures greater than a threshold of (approximately) 100 °C substantially increases the reactivity of peroxide with lignin without having to excessively pressurize the reaction vessel. The proposed bleaching stage (labeled PHT) shows kinetic behavior similar to what is found with chlorine or chlorine dioxide, i.e. rapid delignification regime followed by a slower regime followed by an asymptotic limit lignin content (Reed and Colodette, 1995).

The chemistry of peroxide bleaching is not completely known and is an active research subject. Consequently few kinetics studies have been published so far with the noticeable exception of Devenyns *et al.* (1995) which proposes a first order chromophore removal reaction with the concentration of perhydroxyl ions and temperature (through Arrhenius' law) are multiplying coefficients of the rate equation.

#### 4. BRIGHTENING KINETIC MODELS

Information on hydrosulfite brightening kinetics is also required in order to develop control and optimisation strategies. In light of the widespread use of hydrosulfite in pulp brightening applications, it is surprising to find that there is limited information on kinetics. Most studies are performed on very specific conditions and do not attempt to explain the effects of operational conditions on kinetics. In contrast, the kinetics of mechanical pulp bleaching with hydrogen peroxide have received considerable attention. One recent example is the study by Xu (2000) where the effects of pH, peroxide dosage, consistency, and temperature were explored in order to obtain a mechanistic model.

Melzer (1985) have performed many tests to determine the time-dependence of the brightening reaction with groundwood. His experimental results only extend to a reaction time of five minutes, whereas the available retention time in an industrial process is in the order of 45 minutes to an

hour. He investigates the effect of temperature, mixer speed, initial hydrosulfite content, pH and consistencies. Of particular interest for the development of a control strategy are the effects of hydrosulfite and consistency, whereas the other parameters are normally controlled in the plant. Higher brightness increments can be reached by increasing hydrosulfite dosage and pulp consistency, but it is difficult to determine the effect of these parameters on kinetics from his experimental data. Melzer chose not develop an empirical kinetic equation and he stated that the determination of the order of the reaction is merely of formal significance. However, the knowledge of the order of the reaction can be of use in the elaboration of control strategies.

Hydrosulfite kinetic results are often reported in terms of the retention time required for the industrial implementation of a hydrosulfite brightening process. For example, Crawford (1969) stated that 75% of the total brightness increase occurs within 15 minutes and that a retention time of one hour is adequate. In the same manner, Loras (1980) stated that the reaction rate of hydrosulfite brightening is "high" and that most of the brightening was complete after 10 to 15 minutes, but that slight additional brightening occurred for the next 2 hours. Similar results are reported by Becka (1973) with his study of pine groundwood and 1% hydrosulfite dosage. Rapson *et al.* (1965) reported that most of the brightening had occurred in 5 minutes and that 40 minutes were required to reach the maximum brightness in their study with various species of 4% consistency groundwoods. Tyminski (1967) reports experiments with 2.5% consistency groundwood and showed that brightness increases were complete after 15 minutes. Ellis (1996) reported that most brightening occurred within 15 minutes and that the optimum retention time varied between 30-60 minutes in his study with 4% consistency thermomechanical pulp. Of the above authors, Crawford (1969), Rapson *et al.* (1965), and Ellis (1996) give brightness results as a function of time only in graphical form rather than generalized statements.

There are no kinetic results for pulps that have consistencies equivalent to refiner or medium consistency brightening. McArthur *et al.* (1988) only stated that medium consistency brightening required shorter retention times than lower consistency brightening, such as 10 minutes versus 75 minutes, in their study with black spruce groundwood. There are also no published results on the kinetics of hydrosulfite brightening with several injection points.

Detailed kinetic studies of the decomposition of hydrosulfite in aqueous solutions have been published, but the techniques have not been extended

to track the consumption of hydrosulfite in its reaction with pulp. These results are obtained by polarography, such as those obtained by Cermak and Smutek (1975), and Lem and Wayman (1970). Hosoya *et al.* (1970) have studied the time-dependent changes in light absorbancy when synthetic chromophore groups are contacted with hydrosulfite, but their results are not immediately transferrable to the optimisation and control of pulp brightening.

## 5. LIMITATIONS OF EXISTING MODELS

The main problem with the kinetic models found in the literature is that they are almost always based on an empirical view of the reaction. While one model developed under a certain set of conditions may be useful for one particular application, it is unlikely that it is general enough to be useful for another. With a few exceptions, the proposed models only provide a reaction rate for the main variable of interest, e.g. lignin content, Kappa number, concentration of chromophoric groups. It would be highly desirable to also have description of the chemical consumption, rate of polymeric chain breakages, pH, heat transfer. Some bleaching stages use two chemicals (e.g. C/D, D/C) but with one exception (Teder and Tormund, 1977) all kinetic models express the reaction in terms of only one chemical concentration while it is known for instance in the case of chlorine and chlorine dioxide that their effects are not simply additive (Teder and Tormund (1977); Gendron *et al.* (1993)).

## 6. CONTROL OF BLEACHING PROCESSES

The control of bleaching stages can usually rely on an array of sensors located prior to the injection point of steam or bleaching chemicals: stock flow and consistency of the brown stock may be used to compute a first estimate of the required amount of chemical to apply (Rankin and Bialkowski (1984); Wells (1993)). In chlorination towers there may be a set of sensors located at the base of the reaction tower. One sensor measures pulp brightness accompanied by a measure of chemical residual to provide an estimate of the amount of chemical actually consumed. This pair of sensor may be used to update a model based observer of the reaction and since a good portion of the delignification reaction is taking place at an early stage, the brightness sensor may be used for feedback control. J. Soderberg *et al.* (1988) and Edlund *et al.* (1973) report that for a chlorination stage, a Kappa number sensor may be inserted prior to chlorine injection to provide feedforward control on the application of chlorine. They also report

that post tower sensors may provide additional information. These include a residual chlorine sensor after the chlorination tower and a pH sensor after alkali application. In J. Soderberg *et al.* (1988), the post tower chlorine residual sensor is used to provide additional feedback to cut back on the chemical application. An additional Kappa number sensor is installed after the extraction stage but only to monitor Kappa number variations (no feedback is provided). The paper by Edlund *et al.* (1973) describes the use of a post extraction Kappa number sensor for feedback control in very general terms but does not provide any data. For oxygen delignification, Kubulnieks *et al.* (1991) describe the use of a post tower Kappa number sensor to provide full feedback control of an oxygen bleaching stage. The applications do not yield a lot of details on the exact methodology employed to design and tune the controllers. For instance, it is impossible to determine if multivariable control is used. The empirical model of Olm and Teder (1979) are claimed to be used by Wells (1993) and Kubulnieks *et al.* (1991) while Gendron *et al.* (1993) used multiple simple first order plus time-delay models. All applications claim that process control can achieve substantial savings in the cost of chemicals and produce pulp of more uniform brightness when compared to some former system of operation. This is indicative that an adequate combination of feedforward/feedback control can accomplish substantial savings in this area of the mill. There is no reason to believe however (and actually it is never claimed except maybe in very vague terms) that these applications are optimal. Furthermore, all published data describe the application of control to individual stages. But since the operation of one stage influences subsequent stages, it is also necessary to perform a coordinated optimization of the whole sequence as described in Section 8.

## 7. CONTROL OF BRIGHTENING PROCESSES

The control and optimization of a hydrosulfite brightening process is a problem that incorporates the different kinetics and interactions of each injection point and the long and variable time-delays between the injection of hydrosulfite and the brightness reading at the paper machine. There are other control and optimization considerations as well. For example, the quantity of hydrosulfite used at the refiner should be minimized in order to reduce corrosion. Likewise, the quantity of hydrosulfite used near the paper machine should be minimized so that waster and odour problems associated with inefficient reaction conditions are reduced. Furthermore, there are often no reliable brightness measurements in the pulp

process until the final measurement at the paper machine.

In spite of the industrial popularity of hydrosulfite for the brightening of mechanical pulps, the control of such processes has received very little attention. Most industrial processes are still controlled manually, thus creating unnecessary paper brightness variations and hydrosulfite waste. For example, Sopenlehto and Moilanen (1988) have observed that manual control of hydrosulfite can increase the pulp's brightness variability in comparison to its incoming brightness. These authors implemented a brightness control strategy based on two types of instruments: a pulp brightness sensor and a hydrosulfite residual sensor. Although specifics of the control strategy are not given, they reported a reduction in brightness variability and chemical consumption. Their strategy involves the adjustment of hydrosulfite dosage based on the target brightness, the measured incoming brightness, the measured intermediate brightness, and the predicted brightness development. The residual measurement is used to adjust the level of hydrosulfite at a level where the final brightness can be reached. The authors warn that control based solely on brightness measurement can often lead to unnecessary addition of hydrosulfite beyond the maximal brightness that can be reached by the pulp. However, industrial implementation of such a brightness probe has shown that the residual measurement is directly proportional to the hydrosulfite flowrate, thus not giving any additional information for control.

Gough *et al.* (2002) have implemented a predictive adaptive controller using Laguerre functions to estimate the dead time in a single-injection point hydrosulfite brightening process. The authors reported a significant reduction in paper brightness variability. Their study did not take into consideration the optimal use of hydrosulfite in order to prevent waste past a given hydrosulfite dosage. Mongrain *et al.* (2004) have used a model predictive control strategy to control paper brightness in a two-injection point system. Their control strategy incorporated the use of an optimizer that reduced the use of hydrosulfite near the paper machine. They also reported reductions in paper brightness variability. Although their strategy successfully minimized hydrosulfite waste near the paper machine, it did not consider the optimum of the combination of each injection point and the variability of these optima under different conditions.

Sayda and Taylor (2003) have applied a model predictive control strategy to hydrogen peroxide brightening of mechanical pulp. This controller was successfully applied in advisor mode, where the calculated peroxide dosage was suggested to

the operator. They incorporated a time-delay estimator to deal with the long and variable time delays in the bleaching tower, but it was found that this time-delay estimator could only be used when the time-delay uncertainty was less than 7% which makes it impractical for systems that have channelling problems. Other features of their controller include feedforward techniques to compensate for the incoming brightness. Brightening kinetics are given as pure linear gains, since the pulp mixing and transport processes have longer time constants than the brightening reaction.

Qian and Tessier (1997) and Strand and Edwards (1987) have also developed simulations for control strategies of hydrogen peroxide brightening processes. Qian and Tessier considered an empirical model of kinetics and idealized flow patterns in different process units. The simulation was controlled by a combination of feedforward and feedback control. Their simulation revealed that incoming brightness was the most important disturbance. Strand and Edward also used an empirical kinetic model and applied a combination of feedforward and feedback control. The residence time in the tower was found to have little effect on the final brightness, hence the feedforward part of their control focused mainly on the effects of pH, incoming brightness and temperature.

## 8. OPTIMIZATION OF BLEACHING AND BRIGHTENING PROCESSES

Current research and application work has been focused on the individual members that compose a bleaching sequence but little published work exists concerning the specific application of optimization techniques to the bleaching sequence itself. A recent publication (Dumont *et al.*, 2004) provides an interesting insight into how such a problem may be approached. We provide here an extension of the optimization approach described in their article.

First, let a bleaching sequence be composed of  $N$  stages, each requiring the application of some amount of bleaching agents. Let  $x_i$ ,  $i = 1, \dots, N$  be the amount of bleaching agent applied at each stage. Let the cost of each of these bleaching agents be  $c_i$ ,  $i = 1, \dots, N$ . The optimization objective consists of minimizing the total cost:

$$\min_{x_i, i=1, \dots, N} \sum_{i=1}^N c_i x_i$$

subject to the constraint that by the end of the sequence, the brightness of the pulp be equal to some target  $B$ . If  $f_i(x_i)$ ,  $i = 1, \dots, N$  is the



incremental nonlinear brightness gained at each stage, then the constraint may be expressed as:

$$\bar{B} = B_o + \sum_{i=1}^N f_i(x_i)$$

where  $B_o$  is the brightness of the pulp entering the bleach plant. We may write this as an optimization problem using a Lagrange multiplier, i.e.

$$\min_{x_i, i=1, \dots, N} C = \sum_{i=1}^N c_i x_i + \lambda (\sum_{i=1}^N f_i(x_i) + B_o - \bar{B})$$

Taking the derivatives with respect to the bleaching agents application and setting to zero, we get:

$$\frac{\partial C}{\partial x_n} = c_n + \lambda \frac{\partial f_n}{\partial x_n} = 0$$

which is the central result of Dumont *et al.* (2004) for the particular case where  $c_1 = c_2 = \dots = c_N$ . In this particular case, the derivative of every function must be equal at their application point.

## 9. CONCLUSIONS

The literature related to the kinetics of bleaching and brightening was summarized in relation to the knowledge necessary to design control strategies for these processes. Conventional and advanced control applications were reported to underline the main control problems. In the last part, the optimization of bleaching and brightening sequences was underlined in terms of future research opportunities.

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