

Optimization of Bleach Plant in Paper Industry for Waste Minimization

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Abstract- In the present investigation a steady-state mathematical model is developed for a typical four stage CEHH bleaching sequence in Indian Pulp and Paper Industry. Each stage of the sequence is composed of chemical additions and mixing, reaction in a retention tower, and washing. Unit operation models were formulated using mass balances on liquor, fibres, kappa number, chemicals and COD based on the assumption of perfect mixing and quasi steady-state. The COD is used in this paper as an indication of the amount of effluent coming out from four bleaching stages. COD measurement of bleach plant effluent separately is not a routine mill measurement; so COD is calculated in the basis of correlation of COD given by Anjana (9). In order to validate the model, the simulated results were compared the literature data; a good agreement between the simulation results with the literature data is achieved. In the case studies, the optimization of the process variables are performed in which the effects of operating temperature, residence time, consistency, bleaching liquor flow rate and the wash water flow rates in the washers are studied.

Keywords- Bleaching; COD, Steady-state modeling; CEHH; Process variable optimization; Waste minimization

I. INTRODUCTION

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. The concern about the environmental effects of chlorinated organics in bleach plant effluents is steadily increasing. In the interest of reducing environmental impact of pulp mill effluents, many companies are examining ways to modify effluent composition and reduce effluent volume. In the extreme, reduced effluent volume means a closed effluent-free pulp mill operation. With the chlorine-based bleaching processes currently in use, a closed mill is difficult to attain due to the corrosive behavior of the chlorine compounds when they are processed in recovery systems. Hence

operational analysis and optimization of process variables would enable in reduction of hazardous effluents. The available methods for analyzing the individual components responsible for pollution in other industries are also used for bleaching effluents. The most widely used characteristics include adsorbable organic halogen (AOX), total organic chloride (TOCL), biochemical oxygen demand (BOD), chemical oxygen demand (COD), total organic carbon (TOC), total dissolved solids (TDS), and color (Junna and Ruonala, 11).

Besides the respective proportions of lignin and bleaching chemical, there are a number of variables which affect the bleaching reactions and subsequently the pollution load. Most of these effects are common to all bleaching 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 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.

Temperature It is generally agreed that temperature affects positively the quantity of lignin agent. Temperature generally enters the models as a nonlinear coefficient of a rate equation (Axegard et al. (3)).

pH The alkalinity or acidity of the solution as measured by its pH plays an important role in every bleaching stages (Daneault et al.(7); Axegard and Tormund (4); Gellerstedt et al. (8); Myers and Edwards (13)). 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.

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.

In the present study, a steady-state model of a four stage chlorination-extraction-hypochlorination-hypochlorination 2 (CEHH) bleach plant has been developed for the calculating the pollution load in terms of COD. The model is verified and case studies for optimizing process variables are performed with the aim of decreasing the total amount of COD coming out from the four washers as shown in the Fig 1.

II. 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 sequence of existing bleaching and brightening technologies. In the following study, the bleaching technologies have been loosely divided in "conventional" technologies chlorine, alkaline extraction, hypochlorite (CEHH).

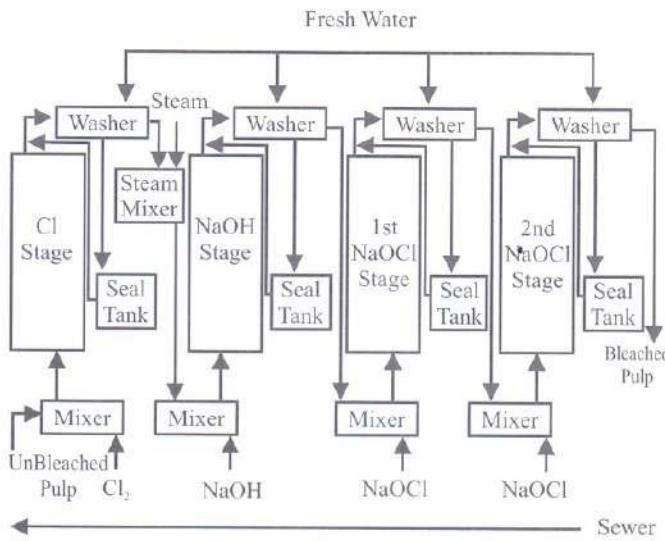


Fig 1. Process flow diagram of a CEHH bleach plant

The flow sheet of CEHH bleaching plant is shown in Fig. 1. Each stage is composed of three unit operations as mixer, retention tower and washer. The process units and variables used to model each stage are shown in Fig. 2.

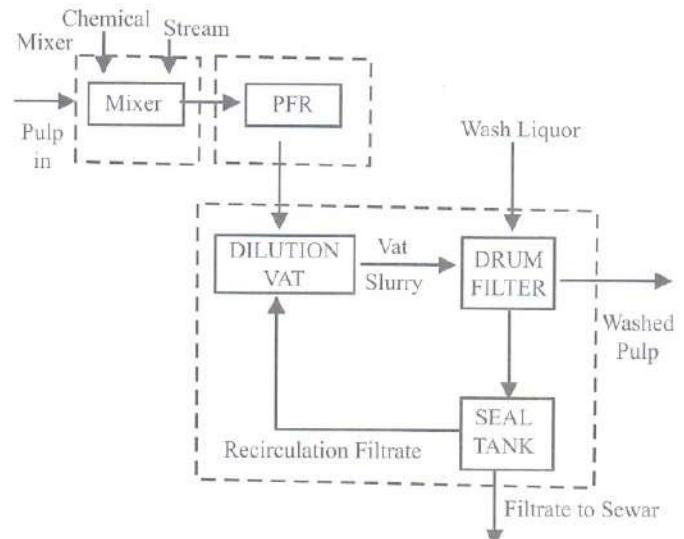


Fig 2 : Process units and variables of the bleach plant.

A. Mathematical Models of Unit Operations

In a bleaching stage, mixers are used to mix the fibre suspensions with bleaching chemicals, steam or recycle streams. The mass balance equations in the mixer for the liquor, fibres, kappa number, and residual chemicals are presented in Table 1 based on the assumption of perfect mixing and quasi-steady-state.

In the retention tower, the reaction between the bleaching chemicals and the fibres takes place. Wang (19), examined different models in order to represent the retention tower. Among the models that are tested, it was found that the sequence of CSTRS-PFR-CSTRS gives the best agreement with the experimental retention time distributions. Several researchers have reported two stage rate mechanisms for various stages of bleaching and if the concentration of reactants is very high in the beginning and fairly low in the end than it may be assumed that all retention towers are acting as single PFR. During further course of study all four towers are considered as PFR. The model equations are given in Table 1.

The modelling of the drum washer is done by dividing it into three parts; as the dilution vat, drum filter and seal tank. In the dilution vat the liquor from the seal tank is mixed with the main pulp stream in order to dilute the pulp to the desired consistency. Therefore, it can be modelled as a steady-state mixer. The drum filter is used to remove the dissolved solids and unreacted chemicals from the pulp with wash water. In order to model the

drum filter, different models can be used. In this study, Norden efficiency approach (Norden, 14; Norden and Pekkanen, 15) is used in order to model the drum filter. In the model, the Norden efficiency factor is taken as 3 for all the washers. In the seal tank the part of the liquor coming from the drum filter is recycled back into the dilution vat and the rest of it is sent to the sewer, which cause different environmental problems. Thus, the seal tank can be modelled as a splitter. The model equations for these unit operations are also given in Table 1.

B. Kinetic Models

The kinetic models discussed in the literature try to describe and/or explain the chemical and physical interactions between the bleaching agent and the pulp fibers. In the modelling of the retention tower the most important parameter is the bleaching reaction kinetics. In the literature different kinetic models are proposed for the bleaching steps Ismail Dogan, et al (10). In the present study the process kinetic models are employed, using Kappa number for the delignification stages (C, E). However, in the brightening stages (H₁, H₂) light absorption coefficient is used.

Most of the chlorination models are based on the kinetic study conducted by Ackert (1). In these equations the effect of consistency introduced through a rate constant multiplier, f_i is equal to 1.0 for individual fibers (low consistency) and 0.52 for fiber mats (high consistencies where fibers contact each other). T is the temperature in the °K. The initial L_{fast} is 0.5 (total initial lignin) and the initial L_{slow} is 0.3 (total initial lignin), with non removable lignin = 0.2 (total initial lignin). These equations kinetically describe chlorine bleaching. The temperature dependence enters through the Arrhenius term for rate constant. The constant f_i is used to adjust the model for the low consistencies used for a conventional bleaching process as opposed to high consistency displacement bleaching. It should be noted that the pH dependence is not included in this model (Sklarewitz et al, 18).

$$R_{fast} = K_1 [Cl_2] [L_{fast}]$$

$$R_{slow} = K_2 [Cl_2] [L_{slow}]$$

$$K_1 = 0.535 f_i \exp\left(\frac{-250}{T}\right)$$

$$K_2 = 0.0107 f_i \exp\left(\frac{-250}{T}\right)$$

where Singh, S.V et al (17) conducted experiment on hardwood and proposed that the course of chlorination was governed by two stage rate equations:

$$R = -\frac{dc}{dt} = K_1 c_f + K_2 c_s \dots (*)$$

where c denote the concentration of available chlorine, c_f the concentration of available chlorine during rapid phase of reaction and c_s the concentration of available chlorine during slow phase of reaction . Singh, S.V et al (17) observed that the first term on right hand side of the equation (*) represented of the kinetics of the initial rapid reaction period up to 1 minute and the second term denoted the slow process and chlorination temperature did not effect the efficiency with which chlorine was consumed .this means that the slight variation in temperature during chlorination stage would not effect the pulp quality and its behavior in subsequent chemical stages of treatment. Chapnerkar (6) concluded the efficiency was also independent of both the initial chlorine concentration and consistency. Therefore, the kappa number after the chlorination as a function of the unbleached kappa number and the degree of chlorination a linear relationship is found in this case, which can be represented by the equation (Axegard, P,2)

$$K_c = K(unbleached) - 82 * \text{consumed Cl}_2$$

* consumed Cl₂ = % of o.d. pulp / kappa number; K_c = kappa number after chlorination

The kappa number after extraction stage could be calculated by using the rate equations given in table 2 (E-stage). Kappa number after the extraction stage (E) should be related with the inlet light absorption coefficient of the hypochlorination stage (H). Wang (19) found a correlation with the light absorption coefficient and the Kappa number of a bleached pulp by fitting the published experimental data. In our study, this correlation is used in order to link the Kappa number after the E stage and the light absorption coefficient entering the H stage as

$$K_{H,i} = 4.69 K_E + .01$$

After the extraction stage, the light absorption coefficient should be converted to brightness, because mills are using brightness for the quality of the bleached pulp. In order to convert the light absorption coefficient to brightness, the Kubelka-Munk equation was used

$$K/S = (1 - R_b)^2 / 2R_b$$

where R_b denotes the brightness, and S is the light scattering coefficient. The value of the scattering coefficient is used as 50 m²/kg (Wang et al, 20). K is the light absorption coefficient of the pulp.

TABLE 1
The Mathematical Models used in a Bleach Plant

	Liquors	Fibers	Kappa Number	Residual Chemical/Dissolved solids
Mixer	$F_0 = F_i + W_i$	$F_0(C_{y0}/1-C_{y0}) = F_i(C_{yi}/1-C_{yi})$	$K_0 = K_i$	$F_0 L_{j0} = F_i L_{ji} + W_i + M_{ji}$
PFR	$F_0 = F_i$	$C_{y0} = C_{yi}$	$dK/dt = -r$	$dL/dt = -(C_{y0}/1-C_{y0})\psi_i r$
Dilution Vat	$F_v = F_i + W_r$		$L_{DS,V} = (F_v L_{DS,I} + W_r M_{DS,I})/F_v$
Drum Filter	$F_0(C_{y0}/1-C_{y0}) = F_i(C_{yi}/1-C_{yi})$		$L_{DS,I} = \{M_{DS,2} + (RW)^E (RW - 1) (F_v - 1)(L_{DS,V} - M_{DS,2})\} \text{ When } RW \neq 1$ $= \{M_{DS,2} + (1/E_N)(L_{DS,V} - M_{DS,2})\} \text{ When } RW = 1$ $M_{DS,d} = (W_d M_{DS,2} + F_v L_{DS,V} - F_i L_{DS,I})/W_d$
Seal Tank	$W_d = W_i + W_r$		$M_{DS,d} = M_{DS,I}$

TABLE 2
The kinetic expressions used in the model

Stage	Reference	Rate Expression	Ψ
C	S.V.Singh et al (1975)	$R = -\frac{dc}{dt} = K_1 c_f + K_2 c_s$ where $K_1 = A \exp\left(\frac{-E}{RT}\right) = 0.85 \text{ (min}^{-1}\text{) at } 30^\circ\text{C}$ $K_2 = B \exp\left(\frac{-E}{RT}\right) = 22.6 \times 10^{-3} \text{ (min \% of } Cl_2)^{-1} \text{ at } 30^\circ\text{C}$ A = constant, B = constant, E = energy of activation, T = temperature in $^\circ\text{K}$	
E	Axegard (1979),	$R_{fast} = K_1 [OH^-]^{0.2} [L_{fast}]$ $R_{slow} = K_2 [OH^-]^{-0.05} [L_{slow}]$ where $K_1 = 1.96 \times 10^6 \left(\frac{-4691}{T}\right)$ $K_2 = 0.0103 \exp\left(\frac{-241}{T}\right)$ $\frac{L_{fast}}{L_{slow}} = 1.75 \times 10^5 \exp\left(\frac{-3368}{T}\right) [OH^-]^{0.25} f_2$ & $f_2 = \text{Chlorine Charge factor}$	
H	Axegard and Tormund (1985)	$R_H = K[OCl]^{0.67} L_H^{1.5}$ where $K = 7.9657 \times 10^{10} \exp\left(\frac{-9622}{T}\right)$	0.688 $L_H^{1.67}$

TABLE 3
The input data for simulation of CEHH bleach plant

C	E	H	H
(1) Mill data Chlorination			
Flow rate (Kg/min)	4.4	-	-
Extraction			
Flow rate (Kg/min)	-	2.2	.25
Concentration (%)	-	8	8
Hypo(1) as Cl_2 basis			
Flow rate (Kg/min)	-	-	2.4
Concentration (%)	-	-	7
Hypo(2) as Cl_2 basis			
Flow rate (Kg/min)	-	-	2.4
Concentration (%)	-	-	7
Steam (Kg/min)	-	12	-
Tower			
Temperature (0C)	25	65	45
Residence Time, t (min)	70	90	190
Washer			
Wash liquor (kg/min)	1700	500	520
Outlet consistency (%)	11	11.3	12
(2) Unbleached pulp			
Pulp flow rate = 111.14 Kg/min	Consistency = 3.0 %		
Kappa number = 25	Dissolved solids = 0.001 %		

The equation for COD taken from Anjana (9) can be used for the mill data. The mill, already having ETP (effluent treatment plant) should consider issue of the impact of effluent load over and above the minimum carry over solids and difference of kappa number from floor kappa value of brown stock. Assuming the value of minimum carry over solids as mentioned in Anjana (9) to be a constant factor for all DF and N values, the Eq. for additional loads can be written as:

$$\text{COD} = 2.4 * \Delta \text{Kappa Number} + 0.61 \text{ (carry over solids)}$$

Where $\Delta \text{ kappa} = K - K_0$, K_0 , being floor kappa number. K_0 depends on the type of fibers (HW, SW etc.) for a

fully washed pulp, pulping condition, control measures, lignin content and type of lignin. Exact value of this parameter is generally not known.

III. MODEL VERIFICATION

The present model is tested by comparing the simulation results with the literature mill data available in the Anjana (9). The input variables are taken from the literature and are tabulated in Table 3. The steady-state Kappa number, the residual chemical and the dissolved solids content of the filtrate from the washer for each stage are calculated and together with the literature data are presented in Table 4. A good agreement between the simulation and the mill data is achieved.

IV. CASE STUDY

The case study performed covers the optimization of the process variables. For each of the towers, the temperature, the concentration, the residence time and the consistency are the parameters investigated and their ranges are shown in Table 5. With in each range the variables were changed at constant intervals and the mid value corresponds to the steady state data of the bleach plant. Only for the chlorine flow rate the minimum allowable value is used.

The results of the case study for the four stages are also given in Table 5. In this table, the first row indicates the steady-state literature data of the total amount of COD that was emerged from the four washers and the brightness at the end of the bleaching sequence. For the pulp to have an acceptable quality the brightness should be greater than 84% ISO. Effect of the flow rate of chlorine in the chlorination stage can not be studied due to lack of data. But it is observed that, in the mill, excess amount of chlorine is used in order to remove as much lignin as possible in the first stage. Therefore, the chlorine flow rate can be decreased without decreasing the brightness at the end of the sequence. Most of the coloring materials are removed in the first two stages. The last two stages of the bleaching plant are mainly used to give the final brightness and to strength the pulp.. Therefore, the dissolved solids content of the filtrate is low in quantity for the last two stages.

The results show that in each case the lowest allowable value of the parameters satisfying the brightness requirement should be used. Although for each stage the change of each parameter has a considerable effect, it has been compensated in the following stages. The pulp and paper industry consumes large volumes of water. An important measure in reducing the pollution charge is the reduction in water consumption and COD.

In order to see the effect of each parameter, the optimized values of the parameters are simulated. The brightness at the end of the fourth stage is decreased to 83.2%, still it is in the allowable range for the pulp quality. The bleaching processes mostly use molecular chlorine as the oxidizing agent to remove residual lignin from the cellulose fibres. The model that is proposed by Singh, S.V et al (17) is used in the simulation. The results obtained showed a 15% decrease in the total amount of dissolved solids with a brightness value of 85.37% ISO.

TABLE 4
Comparison of the mill data with the simulation data

Stage	Kappa no. or brightness		Residual Chemical (%)		COD (%)	
	Simu.	Litera.	Simu.	Report	Simu.	Litre
C	9.2	8-11	0.072	0.07	0.0528	0.0526
E	3.03	2-7	0.261	-	0.1763	0.1518
H1	84.2	75-86	0.249	-	0.1141	0.1170
H2	85.2	75-86	0.249	-	0.1141	0.1170

TABLE 5
Results of process parameter optimization in each stage

Parameter	Optimization		COD	Brightness
	Range	Value	Red. (%)	
Steady state data			-	85.37
<i>Chlorination</i>				
Concentration (%)	6-8	6	1.465	85.37
Temperature (°C)	20-40	20	0.089	85.37
Consistency (%)	3-4	3	0.057	85.37
Resid. time(min)	60-90	60	0.046	85.37
<i>Extraction</i>				
Concentration (%)	7-9	1	0.209	85.37
Temperature (°C)	50-70	50	0.598	85.37
Consistency (%)	10-11.5	10	0.510	85.37
Resid. time(min)	90-110	90	0.423	85.37
<i>Hypochlorination(1)</i>				
Concentration (%)	5-7	5	0.296	84.65
Temperature (°C)	30-40	30	1.197	84.78
Consistency (%)	3-5	3	0.243	84.69
Resid. time(min)	170-210	170	0.176	84.81
<i>Hypochlorination(2)</i>				
Concentration (%)	5-7	5	0.372	84.94
Temperature (°C)	30-40	30	1.141	85.23
Consistency (%)	3-5	3	0.210	85.34
Resid. time(min)	170-210	170	0.148	85.34

V. CONCLUSION

The steady-state model of a CEHH bleach plant was assembled by linking the unit operation models for mixing, reaction and washing. Chemical oxygen demand (COD) is a widely used method for evaluating the bleaching result, but as a collective measurement variable it does not describe the actual compounds that cause the "loss" of bleaching chemicals. Studies have shown that many compounds contribute to COD load but ultimately most of them have no real effect on the bleaching result. The applicability of the CEHH bleach plant model was checked by the literature data. A good agreement between the simulated and the literature data was obtained.

Different case studies were performed in order to decrease the amount of COD coming out from the washers. It was concluded that the effective process parameter for chlorination stage was concentration of bleach liquor where the temperature is more effective in Hypo stages. Extraction stage was the least effected by the above parameters.

Nomenclature

C_c fibre consistency (% on slurry)

E_N Norden efficiency factor (-)

f_1 rate constant multiplier

f_2 chlorine charge factor, % available Cl_2 on o.d.

pulp/unbleached Kappa number

K content of chromophores, expressed as Kappa number in delignification stages (C and E), ml

0.1 N $KMnO_4/g$ fibre or light absorption

coefficient in brightening stage

(H_1, H_2) (m^2/kg) .

k rate constant of bleaching reaction

F flow rate of liquor in pulp stream (kg/min)

R_b brightness (% ISO)

R bleaching reaction rate, i.e. rate of Kappa number decrease in a delignification or rate of light absorption coefficient decrease in a brightening reaction

RW wash liquor ratio (-)

<i>S</i>	light scattering coefficient (m^2/kg)
<i>T</i>	temperature ($^{\circ}K$)
<i>t</i>	mean residence time in PFR (min)
<i>W</i>	flow rate of liquor in non-pulp stream (kg/min)
<i>L</i>	dissolved solids or bleaching chemical content of the pulp stream (%)
<i>M</i>	dissolved solids or bleaching chemical content of the non-pulp stream (%)

Subscripts

<i>C</i>	chlorination stage
<i>d</i>	drum filter stream
DS	dissolved solids
<i>E</i>	extraction stage
<i>H</i>	hypochlorination stage
<i>f</i>	fast
<i>s</i>	slow
<i>i</i>	into process unit
<i>j</i>	bleaching liquor components Cl ₂ , H, OH- and DS
<i>o</i>	off process unit
<i>r</i>	recycle stream
<i>v</i>	dilution vat stream

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