MODELLING OF PEROXIDE-BLEACHING OF PULP USING GAUSSIAN PROCESSES

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Abstract: Recycling of paper is typically performed in a chain of processes. An important process is the peroxide bleaching implementing the means to achieve a high brightness quality. As this process step requires consumption of various chemicals, it is quite cost intensive. This paper presents means to optimize the bleaching stage to achieve an (adaptable) trade-off between cost and brightness: First, the time-varying deadtime of the bleaching stage is estimated using a Kalman filter. Second, the bleaching stage is modelled by Gaussian processes aiming to be used for optimization of the bleaching stage by application of a model predictive controller.

Keywords: Paper Industry, Process Models, Nonlinear Models, Gaussian Processes, Optimization, Quadratic Programming.

1. INTRODUCTION

Recycling of paper to provide pulp for papermaking is performed in a continuous process including different process steps. In the beginning recovered paper is disintegrated to an aqueous suspension typically within a pulper or a drum. Followed by cleaning stages including sorters and centrifugal units, coarse impurities (plastics, CDs...) are removed from the pulp suspension. The next process steps – two-stage flotation, disperger, oxidative and reductive bleaching - are aiming to remove the ink particles from the fibers and to increase the brightness of the pulp to be used for papermaking.

In the flotation process air and chemicals are introduced to enable the removal of ink from the fibers. In principle the ink particles are prepared to stick to the air bubbles such that the rising bubbles move ink particles to the surface. The resulting fluffy dirty foam at the surface is removed and forwarded to the reject handling system. The brighter pulp suspension from the 1st flotation is thickened and fed into the disperger (figure 1). Within this stage remaining ink particles and stickies are dispersed to a size below visibility and fibers are treated mechanically. As a result the brightness of the pulp suspension is decreasing. In most cases the bleaching agents for subsequent oxidative bleaching stage are introduced and mixed already in the disperger. Within the bleaching tower the fibers react with the chemicals resulting in an increase of pulp brightness.

The following process stages – 2nd flotation and reductive bleaching stage – are increasing pulp brightness to the desired level by the above described mechanisms. The final pulp (DIP - de-inked pulp) is typically fed into a storage tower and from that

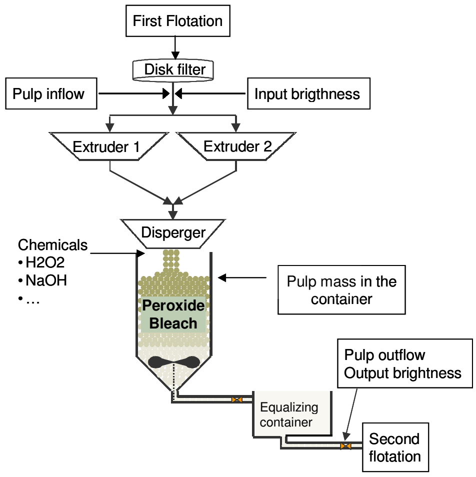

Figure 1: Bleaching process and measurements.

continuously used for papermaking. The process with its single stages to produce DIP can vary regarding the desired requirements. Typical paper grades with use of recycled pulp are newsprint, magazine and hygienic papers.

This paper is focusing on the oxidative disperger bleaching stage (figure 1) using peroxide and sodium hydroxide as bleaching agents. The so called peroxide bleaching is one of the most common bleaching methods for pulps and was originally introduced within the processes for chemical pulp and mechanical pulp (Posio et al., 1998). The chemistry of peroxide bleaching of recycled pulp is nowadays well understood (Göttsching and Pakarinen, 2000). The bleaching effect uses the dissociation of (hydrogen) peroxide (H_2O_2) in water to form a hydronium (H_3O^+) and a perhydroxyl anion (HO_2^-). To achieve a high bleaching effect, increasing the concentration of perhydroxyl anions is necessary. This is possible by raising the peroxide concentration and by adding sodium hydroxide (NaOH) activating the peroxide.

In laboratory studies a non-linear relation between peroxide and sodium hydroxide was determined. Within the process operation of bleaching stage this means to manage the ratio of the two bleaching agents properly to achieve a maximum brigthness increase (Göttsching and Pakarinen, 2000). For example at low NaOH dosage, the peroxide (H_2O_2) does not have a sufficient activation such that the maximum bleaching result is not achieved. After exceeding the optimum, a loss of brightness occurs with additional NaOH, due to the yellowing reaction of excessive hydroxide ions.

The study presented here was carried out in the de-inking plant of the paper mill Gebrüder Lang GmbH Papierfabrik. In the daily operation of the oxidative bleaching stage the dosage of the two chemicals is based on experience, while the ratio of these two chemicals is set to be constant. Accordingly, there is a potential for optimization especially when taking the high costs for peroxide into account. The target derived from the actual process operation was set to determine the concrete possibilities for variation in chemical dosage to achieve a higher brightness with the same costs or an equal brightness with lower costs.

Modern pulp&paper mills use automation to increase production, to improve quality and to decrease production costs and environmental effects (Posio et al. 2002; Runkler et al., 2001 and 2003; Tessier and Qian, 1998). Within peroxid bleaching different approaches for optimization based on different requirements are studied and implemented since 1990. Within mechanical pulp (Posio et al., 1998) the use of oxygen measurements has been suggested. Another approach addresses PH- and temperature control (Meyrant and Dodson, 1990). These approaches for mechanical pulp and chemical pulp with their considerations in regards to control and optimization have to be transformed to be used for recycled pulp.

The objective of this paper is to develop a process model describing the brightness dependence on the chemical agent dosage in the bleaching stage. In a subsequent step the developed model will be used as foundation for the optimization of the bleaching stage.

Variables required for the modelling are incoming (B_{in}) and outgoing pulp brightness (B_{out}) as well as the concentrations of the chemicals (H_2O_2, NaOH) used during the bleach. Besides, the duration spent by the pulp in the tower has to be estimated, in order to enable an accurate relation of incoming to outgoing brightness (see equation 1, where ΔB is the brightness gain and f is a non-linear modelling function). The duration spent by the pulp in the tower is called the deadtime (DT). It is longer than the time required (nearly twenty minutes) by the peroxide reaction during the bleaching process. We assume that pulp is thick enough to absorb chemicals so that they cannot infiltrate in the other layers in the bleaching tower (cf. fig. 1).

$$\Delta B = B_{out}(k + DT) - B_{in}(k) = f(H_2O_2, NaOH) \quad (1)$$

The remainder of the paper is organized as follows: Chapter 2 depicts the approach for deadtime estimation including its smoothing by a Kalman filter. Chapter 3 describes the modelling of the brightness gain by a Gaussian process. The experiments run are sketched, also. Chapter 4 evaluates the optimization potential and summarizes the paper.

2. DEADTIME AND INFLOW ESTIMATION

2.1 Approaches for Deadtime Estimation

For the bleaching process the filling height can vary dynamically as well as the outflow[1]. Hence, the

[1] The inflow monitored by the process control system differs from the inflow to the bleaching tower due to losses in the extruders (see fig. 1).

deadtime is not constant, too. However the curve of deadtimes over time should still be smooth. A wood fiber in the bleaching tower cannot overtake a previously delivered wood fiber, i.e. the deadtime cannot change in large steps. In general, the deadtime can be estimated online by three methods:

- Stationary estimation by calculating the quotient
- Dynamic estimation by integrating the mass flow
- Dynamic estimation by correlation analysis

The first approach estimates the deadtime based on measurements of one time instant of the process. This approach is sensitive with respect to perturbations in the measurement of the outflow, such that jumps in the deadtime are likely. Due to the missing robustness this approach has not been investigated further.

In general, the second and the third approach allow for a dynamic estimation of the deadtime. Since none of the two approaches can be preferred per se, both approaches have been analyzed and evaluated based on process data. Finally, approach two has been chosen for the deadtime estimation due to the following reasoning: Perturbations in the outflow show only a small influence on the resulting deadtime. Robust results are delivered for static and dynamic operation and by moderate computational efforts. On the other hand, the correlation analysis (approach three) would require structured signals, i.e. signals with varying values on the input and output side. This cannot be guaranteed, in general. The reason for this is at hand: the outflow of the bleached pulp should show almost constant brightness, in order to enable a constant quality of the paper to be produced.

2.2 Smoothing of Deadtime Estimation Using a Kalman Filter

As has been sketched in the previous section, the dynamic estimation of the deadtime by integration of the mass outflow results in a quite smooth distribution of the deadtime along the discrete time. However, a detailed analysis has shown that there are further perturbations, which have to be reduced for an accurate modelling and optimization. For smoothing tasks filters can be applied, in general. In particular, Kalman filters (Kalman, 1960) offer the opportunity that free parameters (noise-covariance matrix) can be tuned according to the quality of the measurements. Besides, time discrete Kalman filters can be formulated in an iterative form, which can be applied for online operation.

2.2.1. Model- and estimation equations. For an estimation of the deadtime of the bleaching process the filling mass and the outflow have to be considered. A new filling mass $M_{Bleach}(k+1)$ results from the old filling mass $M_{Bleach}(k)$ and the difference in inflow ($i(k)$) and outflow ($o(k)$):

$$M_{Bleach}(k+1) = M_{Bleach}(k) + (i(k) - o(k)) \cdot \tau \quad (2),$$

with the time step $\tau = 60 Sec$. Using this equation, a lower bound for the new filling mass can be given by setting the inflow to zero:

$$\begin{aligned} M_{Bleach,MIN}(k+1) &= M_{Bleach}(k) - o(k) \cdot \tau \\ &\le M_{Bleach}(k+1) \end{aligned} \quad (3).$$

This limit value $M_{Bleach,MIN}(k+1)$ is used to bound outliers. Additionally a too fast reduction of the deadtime is disabled, in particular the inequality

$$T(k+1) - T(k) \ge -\tau \quad (4)$$

is guaranteed. The inequality takes the FIFO-behavior of the bleaching tower into account, i.e. a wood fiber in the tower cannot overtake a previously delivered wood fiber.

When equation (3) is iterated using the limit value, the fastest way to empty the tower is considered. The time instant, when this process terminates corresponds to the deadtime of the initial filling mass. Here, the deadtime will be estimated using the mass outflow $M_{Out}(k)$ integrated for the deadtime.

Due to the sampling of the system, this mass is determined for an integer deadtime $\tilde{T}(k)$ and by solving the inequality:

$$\tau \cdot \sum_{i=1}^{\tilde{T}_k - 1} o(k+i-1) < M_{Bleach}(k) \le \tau \cdot \sum_{i=1}^{\tilde{T}_k} o(k+i-1) \quad (5)$$

The integrated mass outflow can be determined by:

$$M_{Out}(k) = \tau \cdot \sum_{i=1}^{\tilde{T}(k)} o(k+i-1) \quad (6).$$

With the values $M_{Bleach}(k)$, $M_{Out}(k)$, $o(k+\tilde{T}_k - 1)$ and $\tilde{T}(k)$ the continuous deadtime could be determined by interpolation in the last time step.

$$T(k) = \tilde{T}(k) - \frac{M_{Out}(k) - M_{Bleach}(k)}{o(k+\tilde{T}_k - 1)} \quad (7)$$

Before the presentation of the applied Kalman Filter, the general equations for prediction (8) and update (9) are given. In these equations, **x** is the state vector, **u** is the input vector, **y** is the output vector, $\mathbf{y}_M$ is the measurement vector, **A** is the system matrix, **B** is the input matrix, **C** is the output matrix, **P** is the covariance matrix for the error of the state estimation, **Q** is the covariance matrix for the state noise, **R** is the covariance matrix for the measurement noise, **K** is the Kalman gain matrix and **I** is a unitary matrix. Estimation values are given by „^“, the discrete time *k* is given as index.

$$\begin{cases} \hat{\mathbf{x}}_{k+1} = \mathbf{A} \cdot \mathbf{x}_k + \mathbf{B} \cdot \mathbf{u}_k \\ \hat{\mathbf{P}}_{k+1} = \mathbf{A} \cdot \mathbf{P}_k \cdot \mathbf{A}^T + \mathbf{Q} \\ \mathbf{y}_k = \mathbf{C} \cdot \hat{\mathbf{x}}_k \end{cases} \quad (8)$$

$$\begin{cases} \mathbf{K}_k = \hat{\mathbf{P}}_k \mathbf{C}^T \cdot [\mathbf{C}\hat{\mathbf{P}}_k \mathbf{C}^T + \mathbf{R}]^{-1} \\ \mathbf{x}_k = \hat{\mathbf{x}}_k + \mathbf{K}_k [\mathbf{y}_{M,k} - \mathbf{C} \cdot \hat{\mathbf{x}}_k] \\ \mathbf{P}_k = [\mathbf{I} - \mathbf{K}_k \mathbf{C}] \cdot \hat{\mathbf{P}}_k \end{cases} \quad (9)$$

2.2.2. Estimation of filling height and inflow. The Kalman filter is used to smooth the measured filling mass before it is used to determine the deadtime. Additionally, the inflow to the tower, which is not measured, is estimated. Hence, we have: $\hat{\mathbf{x}} = \begin{bmatrix} \hat{M}_{Bleach} \\ \hat{i} \end{bmatrix}$, $u = o$, $\mathbf{A} = \begin{bmatrix} 1 & \tau \\ 0 & 1 \end{bmatrix}$, $\mathbf{B} = \begin{bmatrix} -\tau \\ 0 \end{bmatrix}$ and $\mathbf{C} = \begin{bmatrix} 1 & 0 \end{bmatrix}$. Besides $\mathbf{P}_0 = \mathbf{Q} = \begin{bmatrix} 1 \cdot kg^2 & 0 \\ 0 & 1 \cdot kg^2/\min^2 \end{bmatrix}$ is used for the prediction. Matrix $\mathbf{R}$ is determined by matching theoretical and empirical covariances (Escamilla and Mort, 2001; Mehra, 1972). The empirical covariance is determined using a window. For $\sqrt{\mathbf{R}} = 37000kg$ and for a window as long as the considered time interval, the theoretical and the empirical covariances match identically.

By the Kalman Filter the time series of the filling mass has been smoothed, fast changes or even jumps have been suppressed[2]. Even more significant are the improvements for the estimation of the inflow. For comparison purposes the inflow has been determined by inverting equation (3). The imprecise measurement of the filling height in the bleaching tower leads to a very noisy variable, compared to the estimated inflow. Accurate knowledge of the inflow is required to model the bleaching behaviour depending on the concentration of chemicals at the beginning of the process. This concentration is defined by the ratio of bleaching chemicals and inflow.

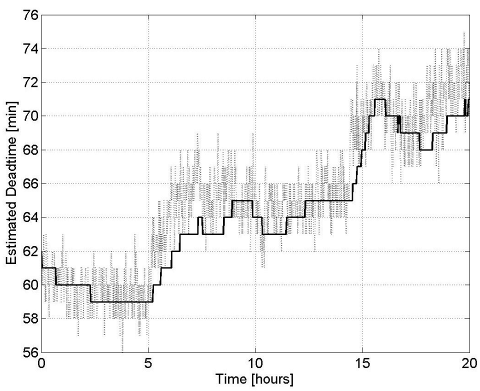

Figure 2: Determination of the integer deadtime (the resolution of one minute corresponds to the time step of the system) by integration of the mass flow - directly (grey line) and after smoothing of the filling mass (dark line). Measurement noise is reduced and a smooth deadtime estimation is achieved.

[2] An analysis based on an approach that directly uses a Kalman filter to smooth the deadtime has shown an undesirable delay in the estimation.

Afterwards, the deadtime is determined based on the estimated filling mass by integration of the mass flow according to equations (5) to (7). Corresponding results are shown in figure 2. Due to the given approach a smooth estimation of the deadtime has been achieved, fast changes or even jumps have been suppressed. Altogether robust results with moderate efforts are achieved. The estimated inflow will be used to determine the initial concentrations of the bleaching chemicals.

3. GAUSSIAN PROCESS MODEL

3.1 Bleaching Experiments

In order to determine the influence of the chemicals on the brightness more than 20 bleaching experiments have been performed on site. The experimental planning resulted in those experiments, with which a reasonable distribution around the currently applied levels is achieved. Further parameters of interest were used to extend this basic experimental plan, e.g. low chemical dosage for minimal costs.

3.2. Brightness Gain.

The important effect of the bleaching process, which can be determined by sensors, is the change of brightness between input and output (cf. equation (1)). After deadtime estimation and data pre-processing the gain in brightness due to the bleaching process can be obtained with a residual method. The brightness gain of the pulp suspension is the sum of a decreased brightness during dispersing and an increased brightness during bleaching. The decrease of brightness in the disperger is almost constant due to the applied control strategy but can be larger than the increase by bleaching.

Using the fact that input and output brightness are strongly linked, when the chemicals' concentrations remain constant, and considering a period, where deadtime, pulp, and chemical inflow stay almost constant, a linear regression is performed in order to find out a linear modelling:

$$B_{out}^{modelled}(k + DT) = a \times B_{in}^{normal}(k) + b \quad (10)$$

In the selected period a very good matching between model and data is given (see figure 3), thereby justifying its further use to calculate the output brightness with the input brightness. The difference or residual error r between this model and real measurements is then calculated by:

$$r = B_{out}^{measured}(k) - B_{out}^{modelled}(k) = \\ = B_{out}^{measured}(k) - \left\{a \times B_{in}^{measured}(k - DT) + b\right\} \quad (11)$$

After smoothing by a median filter to suppress noise, residual variations allow to reveal even very small variations in output brightness (see figure 4) and constitute a good evaluation of the gain in brightness (cf. equation (1)). The gain in brightness is not achieved in one time step, but with a steep continuous slope indicating that there is slow back-mixing in upper layers or slow infiltration in lower layers of the bleach tower.

Using the described approach on all experiments training data, i.e. a table containing the chemical concentrations of the experiments plus the obtained brightness gain, for the modelling of the bleaching process are found. Taking into account the non-linear behaviour of the bleaching process, a Gaussian process has been selected for modelling.

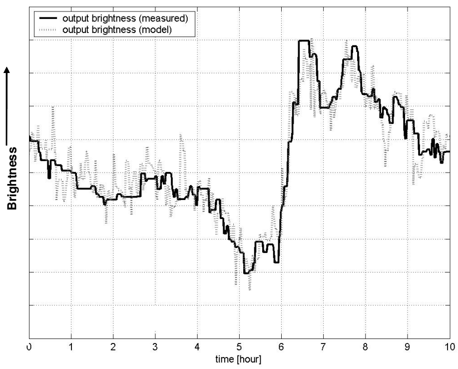

Figure 3: Performance of the regression model for residual error analysis. In the selected period the measured and the modeled output brightness match very precisely.

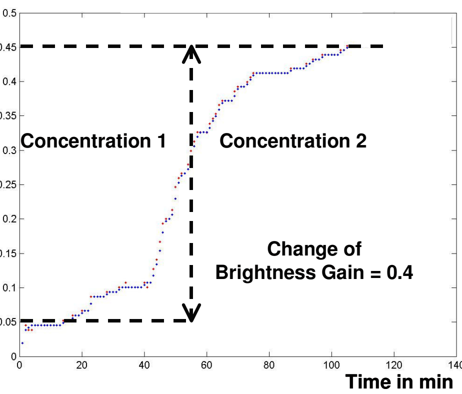

Figure 4: Change of the brightness gain on online measurements for different concentrations.

3.3 Gaussian Processes

A Gaussian process is a stochastic process $\{Y(\mathbf{x})|\mathbf{x} \in X\}$ where any finite set $\{Y(\mathbf{x}_1), \ldots, Y(\mathbf{x}_n)\}$ has a joint multivariate Gaussian distribution, defined by the mean function $\mu(\mathbf{x})=E[Y(\mathbf{x})]$ and the covariance function $C(\mathbf{x},\mathbf{x}')=E[(Y(\mathbf{x})-\mu(\mathbf{x}))\ (Y(\mathbf{x}')-\mu(\mathbf{x}'))^T]$ (Williams and Rasmussen, 1995). In the sequel, Gaussian processes with $\mu(\mathbf{x})=0$ are considered.

Given n pairs of input and output (target) data, $\{(\mathbf{x}_i,t_i),\ i=1,\ldots,n\}$, the output for the test case $\mathbf{x}$ is defined by the n+1 dimensional joint Gaussian distribution for the outputs of the n training cases and the test case. The estimator is thus given by the conditional distribution, i.e. by the distribution of $Y(\mathbf{x})$, conditioned on the observed data. This distribution is again a Gaussian distribution, defined by mean and variance. An elementary calculation reveals that the mean is given by $\mathbf{k}^T(\mathbf{x})\mathbf{K}^{-1}\mathbf{t}$ and the variance by $C(\mathbf{x},\mathbf{x})-\mathbf{k}^T(\mathbf{x})\mathbf{K}^{-1}\mathbf{k}(\mathbf{x})$, where $\mathbf{k}(\mathbf{x})=(C(\mathbf{x},\mathbf{x}_1),\ldots,C(\mathbf{x},\mathbf{x}_n))^T$, $\mathbf{K}$ is the covariance matrix for the training cases with $K_{ij}= C(\mathbf{x}_i,\mathbf{x}_j)$, and $\mathbf{t}=(t_1,\ldots,t_n)^T$.

A reasonable choice for the covariance function is given by $C(\mathbf{x}_i,\mathbf{x}_j) = v_0 \exp(-1/2\ (\mathbf{x}_i-\mathbf{x}_j)^T \mathrm{diag}(w_1,\ldots,w_d)\ (\mathbf{x}_i-\mathbf{x}_j)) + a_0 + a_1\ \mathbf{x}_i^T\mathbf{x}_j + v_1\ \delta(i,j)$, where d denotes the dimension of $\mathbf{x}$.

A main advantage of Gaussian processes compared to other nonlinear regression techniques is that both the estimate and the associated uncertainty are given by an analytic expression. Note that $\mathbf{K}^{-1}$ and $\mathbf{K}^{-1}\mathbf{t}$ do not depend on $\mathbf{x}$, and thus can be calculated in advance when the training cases are given, which further simplifies the required online calculations.

3.4 Gaussian Process Model for Brightness Gain

Finally the bleaching process is modelled by the described Gaussian process. The obtained model is depicted in figure 5. For the choice of the covariance function a local approach has been chosen, in order to reflect the number of about twenty experiments. Accordingly, the global linear parameters a_0 and a_1 are set to zero. The parameters of the nonlinear local part are as follows: $v_0=1$ and constant weighting of the chemical concentrations, i.e. $w_1=w_2=0.35*0.35=0.1225$. The noise level corresponding to the minimum uncertainty has been set to $v_1=0.0225$.

3.5 Evaluation of the Bleaching Modell

The modelling of the bleaching experiments (see figure 5) shows that there are differences in the brightness gain obtained with varying chemical concentrations. However there is an even more interesting fact on this issue: an equal brightness gain can be achieved by different concentration combinations of the two chemicals. On the other

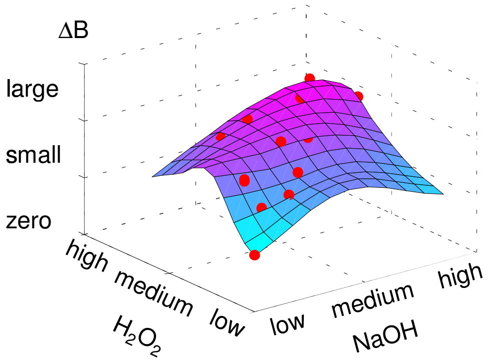

Figure 5: Gaussian process model for the brightness gain ΔB of the bleaching stage.

hand the costs corresponding to the concentrations increase linearly with increasing amount of the corresponding chemicals. Consequently, there is freedom for optimization - either to reduce the costs by achieving the same brightness gain or to identify the best chemical concentrations to obtain a maximum brightness gain with equal costs. A typical approach to solve these types of optimization tasks is to apply sequential quadratic programming [Gill et al., 1981]. This approach is suggested to optimize the amount of chemicals.

4. CONCLUSION

In the chain of processes required for the recycling of paper, peroxide bleaching is an important step to achieve a high brightness quality. This paper has presented a foundation to online optimize the bleaching process to achieve an (adaptable) trade-off between costs and brightness.

The main contributions are the following: reliable estimation of the time-varying deadtime of the bleaching process by a Kalman filter, setup of bleaching experiments, and modelling of the bleaching process by a Gaussian process.

Figure 6 gives an insight into the on-line operating model predictive controller. The diagonal corresponds to a constant ratio of H_2O_2 and NaOH. The model predictive controller enables with its variations in chemical concentrations a new flexibility for cost-efficient operation of the peroxide bleaching stage.

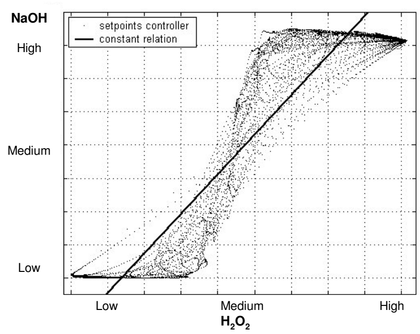

Figure 6: Online variations of the chemical concentrations used to optimize the bleaching process. The dots correspond to measurements taken with a time step of one minute.

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