Study of bleaching of old newsprint recycled paper: reproduction of newspaper material

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Abstract
With the utilization of used newspaper into the paper making industry, bleaching has been an area of concern to obtain a balance between desired brightness and the amount of treatment it requires. Motivation of this study was to optimize bleaching of recycled newsprint fibers in terms of five variables namely hydrogen peroxide charge, amount of sodium hydroxide employed, pulp consistency, bleaching temperature, and bleaching time. Experimental work was conducted in the laboratory to simulate bleaching plant and it was found that higher peroxide charge is better for improvement in brightness, but it is uneconomical to increase peroxide charge indefinitely. The amount of hydrogen peroxide is added so that the residual peroxide at the end of the reaction is not less than 10% of the initially added peroxide. Sodium hydroxide provides alkalinity which is very important for making the fibers flexible and softening of ink particles but it also contributes to pulp yellowing if residual hydrogen peroxide contents are lesser than the minimum required value. So, for 16 kg t\(^{-1}\) of hydrogen peroxide, 6 kg t\(^{-1}\) of sodium hydroxide prevented the brightness reversion in this study. Higher pulp consistency is better but unavailability of very high consistency pulp mixer in the industry is a limiting factor for higher than 30% pulp consistency. Temperature and time are quite interrelated and, with the given bleaching conditions, a temperature of 95 \(^{\circ}\)C with 10 min residence time was found to be optimum for this study. It was also observed that multiple pre-washings increase brightness and save bleaching chemicals.

1. Introduction
‘Secondary fiber is defined as any fibrous material that has undergone a manufacturing process and is being recycled as the raw material for another manufactured product’ [1]. Secondary fiber utilization rate is consistently increasing over the last two decades mainly because of environmental and economic reasons [2]. In Europe, voluntary industry initiative on paper recycling in CEPI countries raised their recycling rate from 49% in 2000 to 54.6% in 2005. New European Declaration, after putting more restrictions to paper industries, raised recycling rate to 66% by 2012 [3].

On the economic side, there is substantial gap between the cost of virgin pulp and that of the pulp which is produced by deinking of recycled paper. Substantial enough to motivate the mills to invest on complex deinking line and subsequent washing stages [4, 5].

Recently China has started collecting recovered paper from all over Europe which has raised prices of recovered paper and is a serious concern for the paper mills around the world [6]. Goal is to obtain reasonable
brightness level if not the same, as of mixed office waste (MOW), using cheaper raw material i.e. old newsprint (ONP). Optimization of the bleaching conditions is pivotal for the realization of this goal and hence is the focus of this study.

Bleaching of deinked pulp (DIP) is a vital step in the processing of recovered paper. Besides color stripping, compensation of any yellowing effect during the natural aging influenced by light, heat and alkaline pulping conditions is the major objective of bleaching wood containing DIP [7]. Recycled fibers are different from virgin fibers in many aspects. They are the mixtures of already bleached chemical and mechanical fibers and are contaminated with residual ink and other impurities. Their morphological conditions are altogether different from that of virgin fibers. These impurities and morphological changes influence the bleaching reaction and virgin fibers bleaching mechanism cannot be applied without reservations to the bleaching of recycled fibers [8].

Hydrogen peroxide is frequently used oxidizing bleaching agent for recycled paper grade. It dissociates in water to form hydronium ion (H$_3$O$^+$) and a perhydroxyl ion (HOO$^-$):

\[
\text{H}_2\text{O}_2 + \text{H}_2\text{O} \leftrightarrow \text{OOH}^- + \text{H}_3\text{O}^+
\]

This perhydroxyl anion created acts as a bleaching agent and to achieve a high bleaching affect, its concentration must be increased. This can be done by increasing peroxide charge, by increasing alkalinity and/or by increasing temperature.

\[
\text{H}_2\text{O}_2 + \text{OH}^- \leftrightarrow \text{OOH}^- + \text{H}_2\text{O}
\]

There is a non-linear relationship between peroxide concentration and optimum sodium hydroxide concentration and an exact ratio of peroxide to sodium hydroxide must be found. At low alkalinity, peroxide is not activated sufficiently and at high alkalinity yellowing effect occurs [9, 10]. Optimum peroxide charge is indicated by the residual peroxide content at the end of bleaching process. Residual peroxide must not be less than 10% of the original peroxide charge in order to prevent yellowing effect and it represents the optimum peroxide charge at a given temperature, time and pulp consistency. Bleaching should occur at optimum H$_2$O$_2$/NaOH conditions not only for economic reasons and for higher brightness but also it is important to keep alkali amount to a minimum level because of the development of the COD load of the process water with increasing alkalinity [8]. Consumption of bleaching chemical not only depends on the initial charge but also on the temperature and time of bleaching. Higher temperature consumes chemicals faster hence shorter bleaching time is required and vice versa [11].

Chemistry plays an important role in many steps of the bleaching process for example fiber swelling, ink removal, anti-redeposition, peroxide stabilization etc [10, 12]. Some chemicals have more than one action during the process: not all of them are desirable. For example, caustic soda other than making the fibers flexible and activating peroxide also causes alkaline darkening of the pulp [13]. Sodium silicate which is used as peroxide stabilizer also prevents the separated ink to redeposit on the fibers. It is important to note that sodium silicate does not stabilize peroxide itself, but it provides the environment in which peroxide works. Sodium silicate also works as a source of alkalinity which must be kept in mind while adjusting the pH of overall bleaching process. The amount of each chemical to be added is also dependant on the amounts of other chemicals and furnish [14, 15].

Pulp brightness increases with high peroxide charge and high alkalinity, but this increase in brightness is very rapid as the process starts then gradually slows down and the final few units increase in pulp brightness consumes the major portion of hydrogen peroxide charge. Further research work needs to be done to understand this phenomenon properly. But this is a well-known fact that bleaching reaction consists of two parts: first is the chromophore removal and other is peroxide decomposition. It is believed that in the early stage of bleaching reaction, easily degradable chromophores are removed causing a rapid increase in brightness. As the bleaching reaction progresses, the remaining chromophores become more resistant, and the second part of bleaching reaction dominates i.e. peroxide decomposition. So, at this stage more peroxide consumes for a given increase in brightness [16].

This study focuses on the five parameters that are critical to bleaching process including hydrogen peroxide charge, pulp pH (directly linked to the amount of sodium hydroxide), pulp consistency, reaction temperature and bleaching time. Each one of these variables has its own significance. For example, increasing peroxide charge increases ISO brightness but the greatest increase occurs with peroxide charge of about 1.5%. Increasing peroxide charge beyond this point increases the brightness but gradient of brightness increase considerably decreases. So peroxide application of more than 2% is not recommended for economic reasons [8]. Sodium hydroxide is primarily considered to be responsible for the pH during bleaching but sodium silicate to some extent also adds to the total alkalinity of bleaching process. Sodium silicate enters the equation to stabilize hydrogen peroxide. Heavy metal ions in the recycled pulp can cause decomposition of peroxide. This undesirable reaction usually occurs slowly but can get accelerated with increased temperature and high alkalinity. This is not only un-economical but also does oxidative damage to fibers due to radicals formed at the
time of decomposition and hence to stop this decomposition of peroxide, sodium silicate is used. The amount of alkali required to reach the optimum pH increases with increasing hydrogen peroxide, however overall pH is established by considering the amount of both sodium hydroxide and sodium silicate \[13\]. Similarly, pulp consistency significantly affects mixing of bleaching chemical with pulp and the dewatering process. Both mixing and dewatering processes become difficult with higher consistency. If mixing is done efficiently, higher consistency improves bleaching results for peroxide bleaching because of the two reasons; first is higher concentration of the bleaching liquor that comes in contact with the pulp and the other is smaller amount of detrimental substance which are dissolved in water \[8, 13\]. In the end, the effect of bleaching temperature and time have been discussed in detail. It is rather easy to understand that both time and temperature are interdependent on each other as increase in temperature allows for a shorter retention time and vice versa, however, temperature is so adjusted that added amount of peroxide is desirably consumed.

This study presents a unique combination of reaction parameters where one parameter is dependent on the other and the impact of their mutual variation on pulp brightness has been reported. For example, the dependency of hydrogen peroxide activation on sodium hydroxide addition (which alters alkalinity) has been explained in detail. Additionally, to the best of authors’ knowledge, the interdependency of temperature and time was not discussed as mutually dependent parameters in the previous studies.

2. Materials and methods

2.1. Variables involved

The pathway adopted to perform this study was divided into three parts.

I. To find the most suitable conditions of variable parameters.
II. To find the optimum values using these suitable conditions.
III. To vary one parameter at a time to find the individual optimum of each parameter.

Variables involved in this study and their values for the first part of study are given in table 1.

Amount of sodium silicate added was in accordance with the amount of hydrogen peroxide and in given in the table S1 (available online at stacks.iop.org/MRX/8/085305/mmedia). Pulp used for this study, old newsprint grade, was obtained from Metsä Tissue, a paper mill at Meriested, Sweden, after the first deinking stage.

In the first part of this study, finding the most suitable parameters and their values in a limited time awarded was a bit tricky. So, factor design technique was used to conduct these experiments in which more than one variable was changed at a time. Sequence of these experiments has been provided in section 3, table 2.

After we got the most influencing values of these parameters, three experiments were conducted in the second part of this study. In the first experiment, most suitable values from the first set of experiments were applied and in the second experiment these values were further extended from the upper and lower limits to see if the results could further be improved. And in the third experiment same extended values were used but pulp was well washed before bleaching to observe the washing effect on brightness. pH measurement and residual alkali measurement were also included in these experiments. Values of this set of experiments are given in tables 3 and 4 respectively in section 3.

In the third and last part of the study, three set of experiments were conducted where values of the three most critical parameters (NaOH charge, temperature, and time) were changed one by one in each set. Values of this set of experiments are given in tables S2–S4.

The general procedure adopted for bleaching, sheet forming, and ISO brightness measurements are given below.
2.2. Bleaching process

30 g oven dry pulp (equivalent wet amount) was taken into plastic bags (which could sustain bleaching temperature). These bags were sealed after adding bleaching liquor. Pulp was well shaken with hands to get it properly mixed with bleaching liquor. These sealed plastic bags were then put into autoclaves which were filled with hot water (about 40 °C) more than its half. Purpose of the hot water was to even out the heating of pulp.

Table 2. Results from initial set of experiments. Four readings are taken for the measurement of brightness of each experiment. These error limits indicate standard deviation.

| Exp No | Run order | H2O2 | Temperature (°C) | P.C. (%) | NaOH | Time (min) | ISO       |
|--------|-----------|------|-----------------|----------|------|------------|-----------|
| 1      | 13        | 10   | 105             | 20       | 0    | 20         | 55 ± 0.1  |
| 2      | 2         | 16   | 105             | 20       | 0    | 5          | 59 ± 0    |
| 3      | 10        | 10   | 125             | 20       | 0    | 5          | 54 ± 0.1  |
| 4      | 3         | 16   | 125             | 20       | 0    | 20         | 55 ± 0.1  |
| 5      | 1         | 10   | 105             | 30       | 0    | 5          | 58 ± 0.1  |
| 6      | 12        | 16   | 105             | 30       | 0    | 20         | 60 ± 0.1  |
| 7      | 8         | 10   | 125             | 30       | 0    | 20         | 50 ± 0.6  |
| 8      | 18        | 16   | 125             | 30       | 0    | 5          | 57 ± 0.1  |
| 9      | 5         | 10   | 105             | 20       | 3    | 5          | 57 ± 0    |
| 10     | 16        | 16   | 105             | 20       | 3    | 20         | 53 ± 0.2  |
| 11     | 19        | 10   | 125             | 20       | 3    | 20         | 53 ± 0    |
| 12     | 6         | 16   | 125             | 20       | 3    | 5          | 59 ± 0    |
| 13     | 15        | 10   | 105             | 30       | 3    | 20         | 58 ± 0.1  |
| 14     | 14        | 16   | 105             | 30       | 3    | 5          | 60 ± 0.2  |
| 15     | 11        | 10   | 125             | 30       | 3    | 5          | 55 ± 0.1  |
| 16     | 17        | 16   | 125             | 30       | 3    | 20         | 55 ± 0.1  |
| 17     | 7         | 13   | 115             | 25       | 1.5  | 12.5       | 54 ± 0.1  |
| 18     | 4         | 13   | 115             | 25       | 1.5  | 12.5       | 54 ± 0.1  |
| 19     | 9         | 13   | 115             | 25       | 1.5  | 12.5       | 54 ± 0.2  |

*a Amount added as kg t⁻¹ of 90% pulp.

Table 3. Optimum conditions derived from first set of experiments. Four readings were taken for brightness measurement and three for each pH and residual peroxide measurement. These error limits indicated standard deviation.

| Temperature (°C) | H2O2 charge (kg t⁻¹ of 90% pulp) | Pulp consistency (%) | NaOH (kg t⁻¹ of 90% pulp) | Time (min) | ISO (%)       | pH         | Residual peroxide (%) |
|------------------|----------------------------------|----------------------|-----------------------------|------------|---------------|------------|-----------------------|
| 105              | 16                               | 30                   | 0                           | 20         | 58 ± 0.1      | 9.86 ± 0.2 | 21.59 ± 2.7           |

Table 4. Extended values of parameters beyond initial and final limits. Results from the washed pulp are also added. Four measurements of the brightness were performed. These error limits indicate standard deviation.

| Temperature (°C) | H2O2 charge (kg t⁻¹ of 90% pulp) | Pulp consistency (%) | NaOH (kg t⁻¹ of 90% pulp) | Time (min) | ISO (%)       | pH         | Residual peroxide (%) |
|------------------|----------------------------------|----------------------|-----------------------------|------------|---------------|------------|-----------------------|
| 95               | 25                               | 30                   | 3                           | 35         | 62 ± 0.1      | 10.2       | 19.9                  |
| 95               | 25                               | 30                   | 3                           | 35         | 63 ± 0.1      | 9.5        | 40.7                  |
Then autoclaves were put in the polyethylene glycol (PEG) bath (Clariant PEG-8, Polyglykol 400) for required time and temperature. Time required to reach the required temperature was about 10 min for every sample.

Care should be taken while preparing bleaching liquor. First of all sodium silicate is added in water. Required amount of sodium hydroxide is then added in that silicate solution. Hydrogen peroxide is added in the last. This sequential addition of the bleaching chemicals is not only efficient for bleaching, but also it leaves greater amount of residual peroxide at the end of the reaction [17].

After required time, autoclaves were taken out of the PEG bath, quenched in cold water, opened and plastic bags were taken out immediately and put into cold water. All of this should be done as quickly as possible to stop bleaching reaction completely. This bleaching process was adopted to simulate industrial bleaching.

2.3. Measurement of the residual peroxide and pH
Pulp was taken out of the plastic bags. There was not enough liquor in the pulp to be squeezed out for the measurement of residual alkali and pH, hence this pulp was diluted and well washed in one liter of distilled water. Liquid was squeezed out of the pulp and pH measurement and residual peroxide measurement were performed with this liquor. Residual peroxide was measured using titration and pH was measured using laboratory pH measuring rod (Digital pH meter, PCSIR) as mentioned in supplementary data (A1, A2 respectively).

2.4. Sheet forming
Sheet forming for the testing of ISO brightness was done according to the standard ISO 3688:1999(E) except for the following few changes.

- Six sheets were prepared for the brightness measurement instead of ten.
- pH measurement of the pulp was not measured before sheet forming.
- Sheets were not dried on drum dryer but in the climate room for about 18 h for free drying.

2.5. Brightness measurement
Brightness measurement was performed using laboratory spectrophotometer CM-3630 of Minolta. Four measurements were performed for average value of each sample.

3. Results and discussion

3.1. Part 1
The interpretation of the results for the first set of experiments was a bit complex as this set of experiments were designed using factor design technique and it is sometimes difficult to interpret these results. But it was possible to see which factors were more influencing on bleaching results as shown in table 2.

The best results are for experiment number 6, 10 and for 14. And based on these results, it can be inferred that higher peroxide charge, lower temperature, higher consistency and longer time are beneficial for improved brightness as has been reported earlier [8, 9, 13]. Due to large number of experiments in this part of study, it was not possible to measure pH and residual peroxide contents. So, it could not be said with certainty if some brightness reversion occurred in any of these experiments but the conditions for three best results were taken and analyzed further.

3.2. Part 2
Based on the best results in part 1 of the study, a new set of three experiments was designed. In the first experiment suitable conditions from first part of the experiments were used which are presented in table 3; Brightness obtained in this case is lower than expected as the conditions used were the most suitable ones and brightness should have been much improved. The reason for this low brightness could be in-efficient activation of peroxide (linked with the absence of sodium hydroxide) which is also indicated by the higher residual peroxide contents. pH value is surprisingly higher (since no alkali was added) however, possible explanation for this could be the addition of sodium silicate and the presence of alkaline earth metals in the DIP, both of which contribute to the overall alkalinity [14, 15].

This set of suitable conditions was based on the provided lower and upper values of the parameters where the experiments were conducted within these limits. However, in order to see the effect of stretching these limits values beyond their starting and ending values, new set of experiment was designed as shown in table 4. An additional experiment was also conducted with the same extended values but pulp was well washed before bleaching.
These results indicate that washing is very beneficial because in case of washed pulp, not only brightness was higher but also more than double the amount of peroxide was conserved which is in accordance with the previous studies [18]. Washing removes some ink prior to bleaching as well as soften the remaining one and hence lesser amount of bleaching chemical is consumed and higher brightness can be obtained.

Much better results were found in these experiments which could be an indication of optimum values of these parameters. Residual peroxide is higher than required even though more than sufficient time was provided for bleaching which indicates that initial peroxide charge can be decreased if the reaction time is lowered. So, 16 kg t\(^{-1}\) of peroxide charge was sufficient for this reaction. Pulp consistency could not be further enhanced because of practical difficulties of pulp handling associated with higher consistencies in the industry. The optimum values of three important parameters i.e. temperature, time, and sodium hydroxide charge were still not reached and hence further experiments were conducted in which the values of these three parameters were changed one by one to find their optimum.

3.3. Part 3
This part consisted of three stages; first stage was to find optimum sodium hydroxide charge, second stage was to find optimum temperature and in the third stage optimum time for bleaching reaction was sought.

3.3.1. Sodium hydroxide charge
In order to find the optimum amount of sodium hydroxide, four experiments were conducted in which following conditions were used:

- Hydrogen peroxide: 16 kg t\(^{-1}\) of 90% pulp
- Pulp consistency: 30%
- Temperature: 105 °C
- Time: 20 min.
- Sodium hydroxide: 0 kg t\(^{-1}\), 3 kg t\(^{-1}\), 6 kg t\(^{-1}\) and 10 kg t\(^{-1}\) in four experiments respectively.

Results obtained in these experiments are given in figure 1.

Pulp brightness is increasing with adding more sodium hydroxide charge (figure 1) which means that more peroxide is being activated which takes part in the bleaching reaction. Higher activation of hydrogen peroxide with increasing sodium hydroxide charge is also indicated by decreasing residual peroxide contents in figure 2. Mechanism of bleaching of recycled fibers is different from that of virgin fibers. Residual impurities in recycled fibers play a vital role in mechanizing the bleaching process. For example, the presence of heavy metals in recycled fibers decompose hydrogen peroxide which is the primary reagent of the bleaching processes. This decomposition of peroxide is enhanced at higher alkalinity and hence the amount of alkalinity added to the system (both in terms of sodium hydroxide and sodium silicate) must be carefully adjusted [13]. Excessive decomposition of peroxide may cause brightness reversion if the residual peroxide content is lower than required as mentioned above [9, 14]. Although brightness is higher with 10 kg t\(^{-1}\) of sodium hydroxide charge, but risk of brightness reversion is higher with longer bleaching time as residual peroxide contents are lower than the required amount as shown in figure 2. Hence optimum amount of sodium hydroxide was taken as 3 kg t\(^{-1}\) or 6 kg t\(^{-1}\) of pulp. It was further analyzed in the next stage.

![Figure 1. Change in ISO brightness with increasing sodium hydroxide charge. Four readings were taken for each measurement. These error bars indicate standard deviation.](image-url)
3.3.2. Optimum temperature

Three experiments were conducted in order to find the optimum value for the temperature and the conditions used were:

- Hydrogen peroxide: 16 kg t\(^{-1}\) of 90% pulp
- Pulp consistency: 30%
- Time: 20 min.
- Sodium hydroxide: Both 3 kg t\(^{-1}\) and 6 kg t\(^{-1}\) for each value of the temperature.
- Temperature: 95 °C, 105 °C and 115 °C. With each value of this temperature, both values 3 kg t\(^{-1}\) and 6 kg t\(^{-1}\) were used.

Results obtained in these experiments are shown in figures 3–5.

Figure 3 shows that for 3 kg t\(^{-1}\) of sodium hydroxide charge increase in temperature will decrease brightness which means that increasing temperature will accelerate the bleaching reaction and once residual amount of peroxide is lesser than required (which is also indicated in figure 4) pulp starts getting yellow in alkaline conditions [9]. Similar results are observed in case of 6 kg t\(^{-1}\) of sodium hydroxide that ISO brightness decrease with the increase in temperature at a certain amount of hydroxide charge.

Effect of bleaching chemicals (hydrogen peroxide and sodium hydroxide) and bleaching temperature on pulp brightness are collectively exhibited figures 3–5 which represents their interdependency. For hydrogen peroxide dose of 16 kg t\(^{-1}\) of 90% pulp, a temperature of 95 °C produced maximum brightness of 61% both at 3 and 6 kg t\(^{-1}\) of sodium hydroxide. Further increase is temperature up to 115 °C decreased the brightness level to 59%. Acquired pulp brightness depends on the activity of hydrogen peroxide. Perhydroxyl anion (OOH\(^-\)) generated as a result of peroxide decomposition acts as bleaching agent and lesser than desired production of this
active bleaching agent does not perform bleaching action effectively [9–11]. Peroxide decomposition can be enhanced by adding alkalinity (by sodium hydroxide supported by sodium silicate in this case) as well as by increasing temperature of the reaction system. Hence a high temperature of 95 °C in combination with the alkalinity provided by the added chemicals produced desired brightness during the reaction. However, higher alkalinity and higher temperature than desired lead to greater decomposition of peroxide which results in brightness reversion as has been observed in this study.

Brightness results were almost similar both for 3 kg t⁻¹ and 6 kg t⁻¹ of sodium hydroxide but more residual peroxide was obtained for 3 kg t⁻¹ of sodium hydroxide and pH level is also more appropriate for 3 kg t⁻¹ of sodium hydroxide (figure 5). Hence optimum conditions of temperature and hydrogen peroxide charge obtained are 95 °C and 3 kg t⁻¹ respectively.

3.3.3. Optimum time
To find optimum time, three experiments were conducted at this stage with following conditions;
- Hydrogen peroxide: 16 kg t⁻¹ of 90% pulp
- Pulp consistency: 30%
- Temperature: 95 °C
- Sodium hydroxide: 3 kg t⁻¹
- Time: 10 min, 20 min and 30 min in three experiments respectively.

Figures 6 and 7 show that brightness decreases with increasing bleaching time from 10 min to 30 min. Bleaching time is another key parameters to reach optimum brightness level which can be defined on the basis of optimum peroxide decomposition. Optimum bleaching time is the one where peroxide decomposition is just enough to maintain the residual peroxide concentration above 10% of the initially added hydrogen peroxide.
Lesser than optimum bleaching time does not produce sufficient amount of perhydroxyl anion which is the active agent in bleaching process and hence required bleaching affect is not obtained. On the other hand, greater than optimum bleaching time causes extensive peroxide decomposition which reverts the acquired bleaching level because of high alkalinity together with high temperature of the reaction system [13, 19].

4. Conclusion

This study reveals that all the parameters discussed above are of individual significant importance and most of them depend on the overall conditions of the bleaching reaction. Higher peroxide charge is better for improvement in brightness but for economic reasons, its amount cannot be increased indefinitely. A minimum amount of 10% residual peroxide concentration at the end of reaction keeps the bleaching process in the forward direction. Sodium hydroxide which provides alkalinity, together with other additives like sodium silicate, and is very important for the activation of peroxide works in combination with the peroxide charge and hence a great care should be taken while adding sodium hydroxide because it also contributes to the pulp yellowing if residual peroxide contents are lesser than the minimum value. So, the optimum value of hydrogen peroxide and sodium hydroxide in the study were 16 kg t⁻¹ and 3 kg t⁻¹ of 90% pulp respectively. Higher pulp consistency is better but unavailability of very high consistency mixer in the industry is the limiting factor for further higher consistency than 30%. Similarly, temperature and time are dependent on each other as higher temperature requires lower time and vice versa (with the given bleaching conditions). A temperature of 95 °C for 10 min of bleaching reaction provided the best results in this study in terms of ISO brightness. This study further suggests that prior washing may increase brightness for a given set of bleaching conditions which eventually results in saving bleaching chemicals. It has also been observed that bleaching process was very rapid at the beginning but slowed down with time and the improvement in brightness towards the end of bleaching reaction was very slow.
Hence further work is suggested to understand the mechanism of decrease in gradient of brightness achievement towards the end of bleaching reaction.

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**Data availability statement**

All data that support the findings of this study are included within the article (and any supplementary files).

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