

HEDP USED AS CHELATING AGENT DURING O_pQP BLEACHING SEQUENCE OF ACACIA MANGIUM KRAFT PULP

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In an effort to resolve problems related to the difficulty of degradation of the chelating agents EDTA and DTPA, this study used a kind of water treatment agent, 1-hydroxy ethylidene-1,1-diphosphonic acid (HEDP), as the chelating agent in the O_pQP bleaching sequence of *Acacia mangium* kraft pulp. The Q stage was optimized with the use of response surface methodology (RSM) based on the Box-Behnken design (BBD). The results showed that the optimum chelating conditions for subsequent hydrogen peroxide bleaching were the following: Temperature of 54 °C, hold time 32 min, and dosage of agents 0.4%. These conditions resulted in the highest brightness value of 80.12% ISO and the selectivity coefficient of 0.46. Under these optimum chelating conditions, the results showed that the effect of HEDP in bleaching is better than that of EDTA with regard to viscosity of pulp, and that HEDP is similar to EDTA and DTPA in other properties.

Keywords: 1-hydroxy ethylidene-1,1-diphosphonic acid; O_pQP bleaching sequence; Box-Behnken design; Response surface methodology

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INTRODUCTION

The effectiveness of hydrogen peroxide as a bleaching agent is limited by its poor selectivity, which is reflected in the severe loss of viscosity. It is generally accepted that the radicals generated during the decomposition of peroxide are responsible for this cellulose degradation (Gierer 1993). According to the Fenton reaction, the formation of these active radicals, namely HO• radicals, is catalyzed by transition metal ions such as iron, copper, and manganese (Cardona-Barrau *et al.* 2001; Duarte and Lachenal 2002; Cardona-Barrau *et al.* 2003; Abrantes *et al.* 2007).

The removal of manganese and iron from the pulp by way of chelation with DTPA (diethylene triamine pentaacetic acid) or EDTA (ethylene diamine tetraacetic acid) or by acid washing results in better selectivity during the subsequent hydrogen peroxide bleaching. These methods seem to be effective for the reduction of both peroxide decomposition and the cellulose degradation that results from it (de la Rosa *et al.* 2002; Duarte and Lachenal 2002; Lapierre *et al.* 2003).

Acid washing is what is mostly responsible for the viscosity decrease during pretreatment, as the chelation stage has very little effect on the cellulose. This is because acid hydrolysis becomes appreciable at pH values below 3 and at temperatures above 80 or 90 °C. Acid washing equipment requires special treatment to reduce acid corrosion, but in this case the equipment expenses will increase.

DTPA and EDTA are the most common compounds used in the chelating stage, and they gradually have become accepted by researchers. But they are likely to be poorly degraded in sewage treatment plants, as their synthetic compounds are commonly

discharged in large amounts to surface water via runoffs of the sewage treatment plants without adequate degradation. This is due to the poor efficiency of wastewater treatment plants in removing various classes of organic pollutants such as antibiotics and other pharmaceuticals (Zhang *et al.* 2008; Loganathan *et al.* 2009; Matamoros *et al.* 2009). Although EDTA-chelates are normally non-toxic (Sillanpää *et al.* 2003), their presence could also affect the underlying groundwater by enhancing the migration of trace metals and radionuclides from contaminated soils or disposal sites to the groundwater (Schmidt and Brauch 2005). Therefore, in order to promote the best environmental practices, a broad range of environmental technologies including biological processes and physico-chemical treatments have been explored in the interest of removing chelating agents from contaminated water (Rahman *et al.* 2009; Al-kdasi *et al.* 2004; Yang *et al.* 2005; Kurniawan *et al.* 2006a). Regarding degradation of HEDP chelation agents, Appelman and his assistants used a combination of O₃ and H₂O₂ to attain an almost complete removal of HEDPA at an initial concentration of 100 mM at pH 9 (Appelman *et al.* 1996). In this regard, the ozonation of HEDPA in basic conditions results in the formation of phosphorus-containing intermediate by-products, while EDTA is not degradable in acidic environment (Sillanpää *et al.* 2011).

1-Hydroxy Ethylidene-1,1-Diphosphonic Acid (HEDP for short), which was used in this work, is widely used in water treatment for the chelation of Fe²⁺, Cu²⁺, Zn²⁺, and Ca²⁺. It has been applied in the chelating stage as well. Response surface methodology (RSM) is a statistical technique for the modeling and optimization of multiple variables that determines the optimum process conditions by combining experimental designs with interpolation via first- or second-order polynomial equations in a sequential testing procedure. The optimization of chelating stage using the response surface methodology allowed a study on the influence of variables (chelated temperature, retention time, and dosage of chelator). The optimum results can be in comparison with the effect of the chelators including EDTA and DTPA.

EXPERIMENTAL

Materials

The *Acacia mangium* kraft pulp was obtained from APRIL SSYMB, located in the Shandong province, China. The pulp, as received, had a kappa number of 27.17 and an intrinsic viscosity of 1016 mL/g. The pulp was thoroughly washed (slit width 0.25 µm) before being used in the bleaching trials.

The unbleached pulp was subjected to extended delignification (oxygen delignification reinforced with hydrogen peroxide) followed by a short bleaching sequence, O_pQP, as described below.

Hydrogen Peroxide Reinforced Oxygen Delignification (O_p stage)

Oxygen bleaching was performed using a CRS 1030 bleaching reactor. The pulp consistency and oxygen pressure during oxygen bleaching remained constant at 10% and 0.5 MPa. The pulp was mixed by hand with the bleaching chemicals and then placed into the reactor. The reaction temperature, alkali concentration, reaction time, and dosage of H₂O₂ were varied in order to obtain the optimal conditions (100 °C, 3.0% NaOH on o.d. pulp, 90 min, and 1%).

Q Stage Experimental Design and Hydrogen Peroxide Bleaching

The pulp was diluted to a 10% consistency using distilled water. The chelating stage (Q) was performed in sealed plastic bags in a thermostated water bath. Table 1 illustrates the reaction temperature (T), retention temperature (t), and the dosage of the agent (D), as these were the most important variables to influence the properties of the pulp during the subsequent peroxide bleaching. The end pH value ranged from 4 to 5 for the Q stage. After this time had elapsed, the pulp was washed thoroughly with distilled water and then dewatered.

The hydrogen peroxide bleaching stage was performed in a water bath at 90 °C. Pulp at a 10% consistency was sealed in a plastic bag with 4% H₂O₂, 2% NaOH, 2% sodium silicate, and 0.3% MgSO₄ (based on o.d. weight). The bag was then immersed underwater for 180 min.

Table 1. Independent Variables Used in the BBD and Actual Factor Levels Corresponding to Coded Factor Levels

| Independent Variable | Code | Actual factor level at coded factor levels | | |
|------------------------|------|--|-----|-----|
| | | -1 | 0 | +1 |
| Temperature (°C) | T | 40 | 60 | 80 |
| Retention time (min) | t | 30 | 60 | 90 |
| Dosage of chelator (%) | D | 0.2 | 0.4 | 0.6 |

Determined Pulp

The kappa number, viscosity, and brightness of all pulp samples were determined according to TAPPI standards. The HexA content of the pulp samples was determined according to the HUT method. The metal analysis was performed by Atomic Absorption Spectrometry (AAS), and the fiber analysis was performed on a Fiber Tester 912 following the procedure described in TAPPI test methods T261 pm-91. Handsheets were made from pulp following the procedure in TAPPI Test Method T205 sp-95, and the handsheets were tested for tear index, tensile index, burst index according to the tests in TAPPI Test methods T220 sp-96.

The pulp selectivity, given by the selectivity coefficient (α), relates the reduction in the molecular weight of cellulose to the reduction in the lignin content of the pulp as measured by the kappa number (Zou 2002).

$$\alpha = \frac{d\left[\frac{1}{DP} - \frac{1}{DP_0}\right]}{dK} \approx \frac{\Delta\left[\frac{1}{DP} - \frac{1}{DP_0}\right]}{\Delta K} \quad (1)$$

RESULTS AND DISCUSSION

The Properties of Kraft Pulp and O_p Pulp

Test results can be viewed in Table 3. The application of a 3.0% NaOH charge in o.d. pulp at 90 °C produced reductions of 67.84% and 37.60% in kappa number and viscosity, respectively, and a final pH of 12.42 for the reaction time of 90 min. The brightness increased from 28.76% ISO to 57.78% ISO.

Table 2. Experimental Matrix and the Results of BBD

| No. | Process Condition of Q stage | | | Pulp Performance After Hydrogen Peroxide Bleaching | | | | |
|-----|------------------------------|---------|-------|--|------------------|----------|-------------------|--|
| | T (°C) | t (min) | D (%) | Kappa number | Viscosity (mL/g) | α | Brightness (%ISO) | Residual H ₂ O ₂ (g/L) |
| 1 | 40 | 60 | 0.6 | 6.19 | 719 | 0.35 | 79.25 | 1.18 |
| 2 | 60 | 60 | 0.4 | 6.11 | 675 | 0.50 | 79.92 | 1.30 |
| 3 | 60 | 30 | 0.6 | 6.00 | 687 | 0.44 | 79.54 | 1.64 |
| 4 | 40 | 30 | 0.4 | 6.31 | 690 | 0.48 | 80.22 | 1.49 |
| 5 | 60 | 30 | 0.2 | 5.97 | 710 | 0.35 | 78.27 | 0.44 |
| 6 | 80 | 60 | 0.2 | 6.04 | 677 | 0.48 | 76.93 | 0.20 |
| 7 | 60 | 90 | 0.2 | 6.16 | 694 | 0.44 | 77.70 | 0.27 |
| 8 | 60 | 60 | 0.4 | 6.11 | 676 | 0.50 | 79.93 | 1.27 |
| 9 | 40 | 60 | 0.2 | 6.16 | 687 | 0.47 | 78.76 | 0.63 |
| 10 | 60 | 90 | 0.6 | 6.23 | 672 | 0.54 | 79.42 | 0.61 |
| 11 | 80 | 60 | 0.6 | 5.95 | 656 | 0.54 | 78.53 | 0.33 |
| 12 | 60 | 60 | 0.4 | 6.12 | 677 | 0.50 | 79.93 | 1.27 |
| 13 | 60 | 60 | 0.4 | 6.11 | 676 | 0.50 | 79.92 | 1.28 |
| 14 | 80 | 90 | 0.4 | 6.92 | 668 | 0.76 | 76.81 | 0.05 |
| 15 | 40 | 90 | 0.4 | 6.35 | 654 | 0.64 | 80.82 | 2.02 |
| 16 | 60 | 60 | 0.4 | 6.11 | 674 | 0.50 | 79.92 | 1.23 |
| 17 | 80 | 30 | 0.4 | 6.20 | 682 | 0.49 | 78.40 | 0.35 |

The HexA content of the kraft pulp and the O_P pulp were 11.86 mmol/kg and 4.72 mmol/kg, respectively. In accordance with previous studies, the influence of HexA on the kappa number was calculated based on the assumption that 10 mmol/kg pulp of HexA is equivalent to one kappa number unit (Li and Gellerstedt 1997; Chai *et al.* 2001; Pedrosa and Carvalho 2003; Petit-Breuilh *et al.* 2004; Colodette *et al.* 2007). The corrected kappa numbers for the kraft pulp and the O_P pulp were 25.98 and 8.28, respectively.

Optimization of the Selected Variables

Response surface methodology (RSM) based on the BBD was applied to determine the optimum levels of the three selected variables that affected the selectivity coefficient and the brightness of the pulp after hydrogen peroxide bleaching. The experimental results are presented in Table 2. The residual lack of fit p-values were below 0.00010 for both selectivity and brightness, indicating a significant lack of fit. The ANOVA for the selectivity coefficient and brightness response models is represented in Tables 4 and 5, respectively.

Table 3. Determination of Performance Parameters of Pulp with Various Bleaching Options

| Bleach Option | Kappa Number | Viscosity (mL/g) | Brightness (%ISO) | Field (%) | Final pH value | HexA (mmol/kg) |
|----------------------|--------------|------------------|-------------------|------------|----------------|----------------|
| Kraft pulp | 27.17±0.31 | 1016±7 | 28.76±0.07 | — | — | 11.86±0.01 |
| O _p stage | 8.75±0.13 | 825±5 | 57.78±0.03 | 96.80±0.11 | 12.42±0.02 | 4.72±0.00 |

Table 4. Analysis of Variance (ANOVA) for the Quadratic Polynomial Model for Optimization of the Selectivity Coefficient (α)

| Source | SS | df | MS | F-value | P-value |
|--------------------|------------------------|----|------------------------|----------|----------|
| Model | 0.13 | 7 | 0.016 | 8.58 | 0.0032 |
| A-Temperature | 0.014 | 1 | 0.014 | 7.61 | 0.0247 |
| B-Time | 0.047 | 1 | 0.047 | 24.83 | 0.0011 |
| C-Dosage | 1.985*10 ⁻³ | 1 | 1.985*10 ⁻³ | 1.05 | 0.3364 |
| AB | 3.025*10 ⁻³ | 1 | 3.025*10 ⁻³ | 1.59 | 0.2423 |
| AC | 7.744*10 ⁻³ | 1 | 7.744*10 ⁻³ | 4.08 | 0.0780 |
| A ² | 0.013 | 1 | 0.013 | 7.11 | 0.0285 |
| B ² | 6.603*10 ⁻³ | 1 | 6.603*10 ⁻³ | 3.48 | 0.0991 |
| C ² | 0.039 | 1 | 0.039 | 20.41 | 0.0020 |
| Residual | 0.015 | 9 | 0.015 | | |
| Lack of fit | 0.015 | 5 | 3.795*10 ⁻³ | 18975.00 | < 0.0001 |
| Pure Error | 8.00*10 ⁻⁷ | 4 | 2.0*10 ⁻⁷ | | |
| Cor Total | 0.15 | 16 | | | |
| R ² | 0.8956 | | | | |
| Adj-R ² | 0.7612 | | | | |

Table 5. Analysis of Variance (ANOVA) for the Quadratic Polynomial Model for Optimization of the Brightness

| Source | SS | df | MS | F | p |
|--------------------|--------|----|-------|---------|----------|
| Model | 19.36 | 6 | 3.23 | 15.64 | 0.0001 |
| A-temperature | 8.77 | 1 | 8.78 | 42.33 | < 0.0001 |
| B-time | 0.35 | 1 | 0.35 | 1.70 | 0.2190 |
| C-dosage | 3.23 | 1 | 3.23 | 15.56 | 0.0027 |
| AB | 1.19 | 1 | 1.20 | 5.78 | 0.0370 |
| AC | 0.31 | 1 | 0.31 | 1.45 | 0.2637 |
| BC | 0.051 | 1 | 0.051 | 0.24 | 0.6391 |
| A ² | 1.66 | 1 | 1.65 | 7.96 | 0.0176 |
| C ² | 3.86 | 1 | 3.85 | 18.58 | 0.0015 |
| Residual | 2.06 | 10 | 0.21 | | |
| Lack of fit | 2.06 | 6 | 0.34 | 4936.55 | < 0.0001 |
| Pure error | 0.000 | 4 | 0.000 | | |
| Cor total | 21.42 | 16 | | | |
| R ² | 0.9037 | | | | |
| Adj R ² | 0.8459 | | | | |

The F-values, 8.58 and 15.64, of the models imply that the models are significant. There is only a 0.01% chance that a “Model F-value” this large could occur due to noise (P-values < 0.0001).

The value of “Pro>F” less than 0.0500 indicates that the model terms are significant. In this case, A, B, A², and C² are significant model terms. Reducing the model and leaving only the significant model terms could improve the model. However, term C was not excluded because it is significant in selectivity (Table 4). Thus, A, B, C, AB, AC, A², B², and C² are the significant model terms.

The coefficients of determination (R²) were 0.8956 and 0.9037 for the selectivity coefficient and the brightness of pulp, respectively. This is an estimate of the fraction of overall variation in the data that can be accounted for by the model, and thus the models are capable of explaining 89.56% and 90.37% of the variation in the response. The adjusted R² values are 0.7612 and 0.8459, indicating that the models are reliable.

The regression equations (Eq. 2 and Eq. 3) describe the correlations that the significant variables obtained after analysis of variance (ANOVA), when using the reducing models.

$$\alpha = 0.50 + 0.043A + 0.077B + 0.016C + 0.028AB + 0.044AC + 0.056A^2 + 0.039B^2 - 0.096C^2 \quad (2)$$

$$\text{Brightness} = 79.82 - 1.05A - 0.21B + 0.64C - 0.55AB + 0.28AC + 0.11BC - 0.63A^2 - 0.96C^2 \quad (3)$$

Mutual Effect of Reaction Parameters

The response surface graph (Fig. 1) for selectivity indicates that the selectivity increased rapidly with an increase in temperature from 40 to 60 °C. However, the effect of holding time on the α value was as significant as the effect of temperature. The effectiveness of the addition of the HEDP is demonstrated in Fig. 1(b), which shows that the selectivity coefficient first increased and then decreased with the increase in charge.

As shown in Figure 2(a, b), within the temperature range investigated (40 to 80 °C), the brightness of the pulp decreased with increasing temperature, reaching a maximum at 40 °C when the holding time was 90 min and the charge 0.4%. It is also clear from Fig. 2(a) that the brightness did not appreciably change over the holding time. Furthermore, the greater the addition of HEDP, the greater the brightness of the pulp following bleaching with hydrogen peroxide. This effect may be explained by the fact that the untreated pulp sample already had a fairly high level of transition metals. Therefore, addition of HEDP has an effect on reducing the metals content of the pulp. The more chelated time, the less content of transition metals in sample. So the selectivity coefficient and brightness of pulp increase with dosage and time. But the effect of chelated temperature effect on the selectivity coefficient and brightness of pulp is not very significant, the reason need to be further researched.

Optimization of the Experimental Conditions

In order to obtain the optimum conditions for the selectivity coefficient and the brightness of pulp, Design-Expert version 8.0.5 software was used, and the desired goals for each variable and response were chosen as summarized in Table 6. Numerical optimization was carried out for this study.

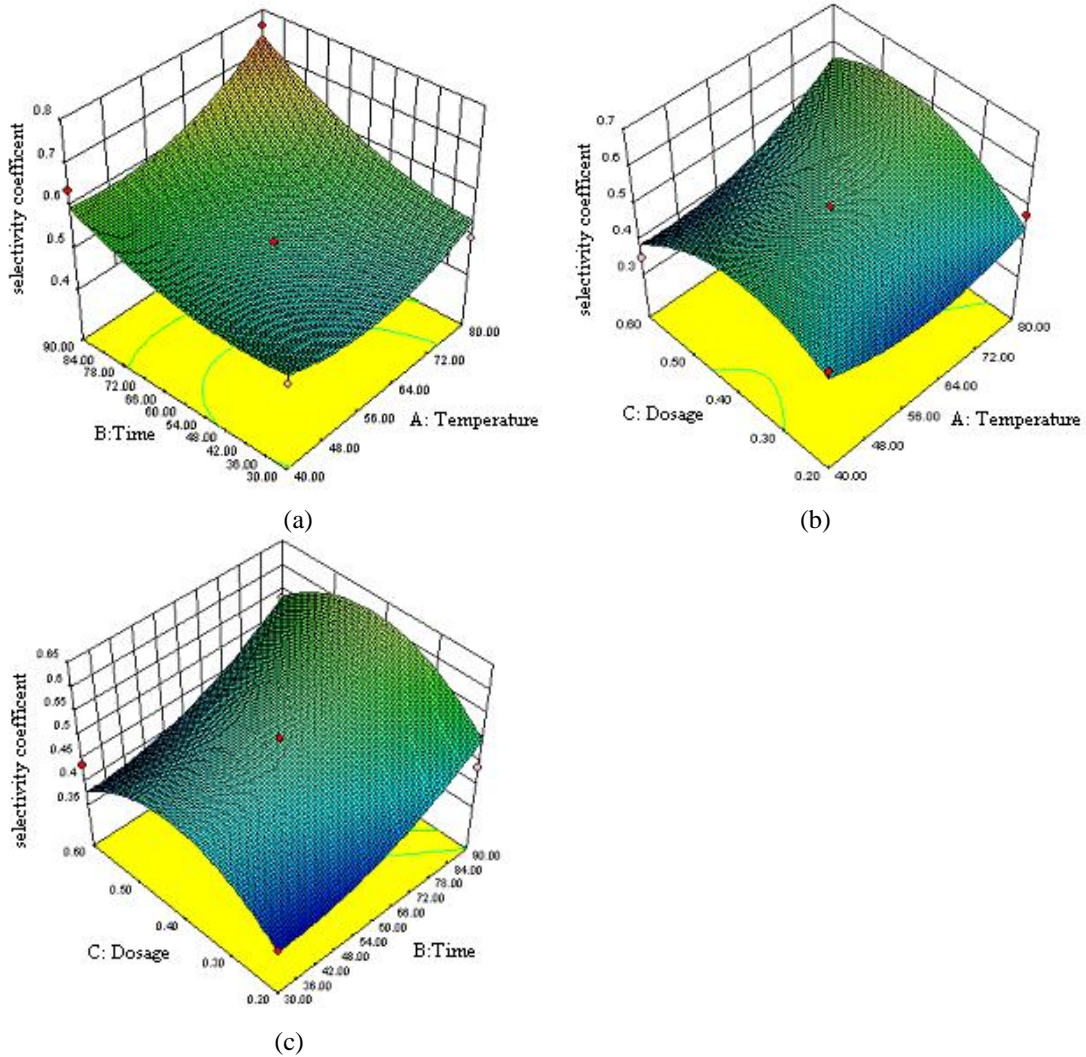


Fig. 1. Response surface plots of Box-Behnken design for the optimization of selectivity coefficient

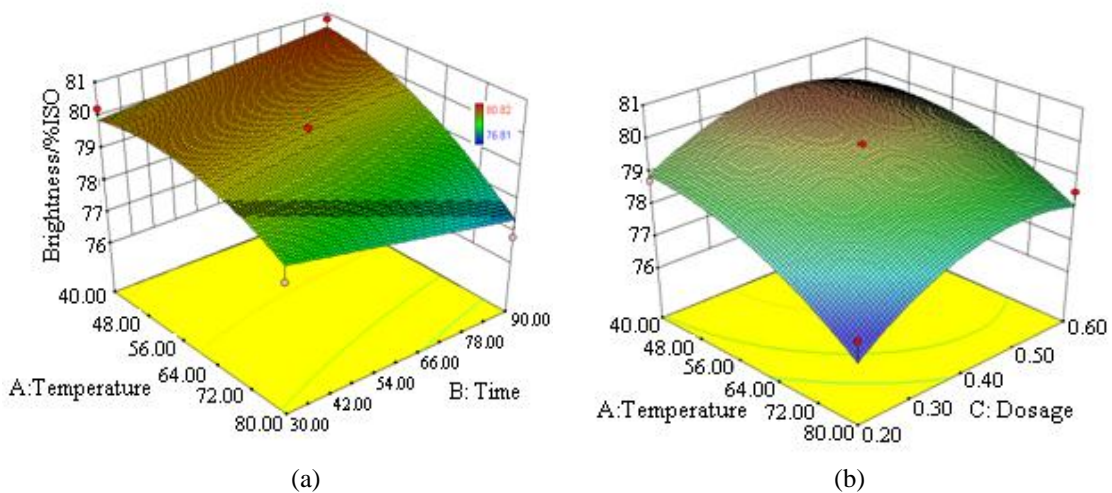


Fig. 2. Response surface plots of Box-Behnken design for the optimization of brightness

Table 6 shows the software-generated optimum conditions for the independent variables along with the predicted values of the responses. These optimum conditions were chelation temperature 54 °C, hold time 32 min, and dosage of agents 0.4%, and they were expected to produce a highest brightness of 80.12% ISO and an α -value of 0.46. This was followed by a confirmatory test.

Table 6. The Optimization of Results for Brightness and Selectivity

| γ | Temperature (°C) | Time (min) | Dosage (%) | Brightness (% ISO) | α | Desirability (%) |
|-----------------|------------------|------------|------------|--------------------|-----------|------------------|
| Predicted value | 54.2 | 32 | 0.40 | 80.12 | 0.46 | 77.6 |
| True value | 54 | 32 | 0.40 | 79.98±0.02 | 0.49±0.03 | — |

Comparative Trial Using Other Chelating Agents

The results of the O_P pulp treated with HEDP were quite similar to the results for the EDTA or DTPA treatments, EDTA and DTPA being the most common compounds used in peroxide bleaching. The kappa number of pulp with O_P QP bleaching sequence was not corrected, since the content of HexA was small enough to be neglected. The results can be viewed in Table 7. These preliminary results showed that the strengths of handsheets were equivalent to each other, following treatments with the above-mentioned chemicals. In particular, HEDP was found to be capable of producing a certain extent of improvements in burst index and tear index.

Table 7. The Comparative Trial Using DTPA and EDTA in QP Stage

| Chelator species | Kappa number | Viscosity (mL/g) | Residual H_2O_2 (g/L) | Brightness (% ISO) | HexA (mmol/kg) |
|------------------|----------------------------------|--------------------|-------------------------------------|-----------------------------------|-----------------------|
| HEDP | 6.55±0.19 | 698±4 | 1.30±0.01 | 79.98±0.01 | 2.04±0.02 |
| DTPA | 6.06±0.10 | 716±6 | 1.42±0.01 | 80.26±0.07 | 1.56±0.01 |
| EDTA | 6.18±0.17 | 686±4 | 0.86±0.00 | 80.14±0.04 | 1.86±0.01 |
| Chelator species | Basis weight (g/m ²) | Folding Resistance | Burst Index (kPa·m ² /g) | Tear Index (mN·m ² /g) | Tensile Index (N·m/g) |
| HEDP | 62.6±0.7 | <5 | 1.30±0.01 | 3.96±0.13 | 26.7±0.1 |
| DTPA | 62.8±1.1 | <5 | 1.26±0.06 | 3.64±0.16 | 26.3±0.1 |
| EDTA | 61.5±0.8 | <5 | 1.24±0.05 | 3.62±0.12 | 25.9±0.0 |

Note: Optimum conditions for HEDP: temperature 54 °C, time 32 min, dosage 0.4%. Conditions for EDTA and DTPA: temperature 60°C, time 60 min, dosage 0.4%. Both are followed by bleaching with hydrogen peroxide. Also, all of the bleached pulp samples were not beaten.

Table 8. The Properties of Pulp with Different Chelators and Stages

| Bleaching Option | Mean Length (mm) | Mean Width (µm) | Transition Metal (µg/kg) | | | |
|------------------------|------------------|-----------------|--------------------------|------------------|------------------|------------------|
| | | | Fe ²⁺ | Cu ²⁺ | Mn ²⁺ | Ca ²⁺ |
| Kraft pulp | 0.779 | 20.1 | 11.19 | 0.575 | 1.557 | 5041 |
| O_P stage | 0.754 | 19.0 | 9.59 | 0.500 | 0.556 | 4864 |
| Q ₁ (HEDP)P | 0.746 | 18.8 | 8.23 | 0.189 | 0.258 | 711 |
| Q ₂ (EDTA)P | 0.739 | 18.8 | 5.75 | 0.048 | 0.243 | 2232 |
| Q ₃ (DTPA)P | 0.740 | 18.8 | 5.53 | 0.056 | 0.236 | 3221 |

HEDP was found to be extremely effective at removing a large percentage of Ca^{2+} , though at the same time its removal of Mn^{2+} was equal to that of the other chelators, as illustrated in Table 8. However, the impact of HEDP on the removal of Cu^{2+} and Fe^{2+} was lower than that of EDTA or DTPA. Moreover, with an addition of 0.4% HEDP, the iron, copper, manganese and calcium content were reduced by 14.18%, 62.2%, 53.6% and 85.38%, respectively. The analysis of the metal content helped to explain some of the results seen previously in Table 4. Due to the fact that HEDP is a chelator, upon its addition to the Q stage it chelated and removed a large percentage of the metals during washing, thereby helping to produce a higher brightness and viscosity at a given degree of dosage. The results of the mean length and mean width of the fiber did not show any significant changes. Further work is needed to research the degradation of the HEDP chelate complex.

CONCLUSIONS

1. The pulp brightness was found to be 80.12% ISO, and the selectivity coefficient was found to be 0.46 (kappa number 6.55, viscosity 698 mL/g) under the experimental conditions of 54 °C and 0.4% HEDP at 32 min. The validation of this model confirmed the agreement between the experimental results and the predicted response.
2. Under the optimum experimental conditions, the results from the O_p pulp chelated with HEDP are close to the results from the pulp chelated with DTPA or EDTA. As a water treatment agent, HEDP can remove large amounts of Ca ion scale formation and at the same time can remove Fe^{2+} , Cu^{2+} , and Mn^{2+} , which protect the hydrogen peroxide. Furthermore, HEDP has the advantage of low cost over DTPA and EDTA.

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