Journal of Korea TAPPI
Vol.37.No.5, 2005
Printed in Korea

Application of Naturally Antioxidant for Inhibition of Brightness Reversion of Paper Made from High Yield Bagasse Pulp

Mohamed El-Sakhawy†, Ahmed El-Ghorab, Ahmed El-Gendy, Mona A. El-Sabour and Essam S. A. El-Sayed

(Received on December 6, 2004: Accepted on November 15, 2005)

ABSTRACT

Different types of antioxidants compounds were prepared economically from natural sources. Application of these compounds towards photo stabilization of high yield bagasse pulp has been investigated. It was found that potato peel antioxidant followed by clove volatile oil provides lower brightness reversion and better photo stabilization effect for paper made from high yield bagasse pulp after exposure to UV lights in comparison with butyl hydroxy anisol or chamomile volatile oil.

Keywords : yellowing, antioxidant, brightness, bagasse, pulp

1. Introduction

To Satisfy increasing pulp demands and to conserve wood resources, there is a great interest in substitution of chemical pulps by high yield mechanical pulps in a wide variety of paper grades. High yield pulps require lower capital cost of the paper mills and have low environmental impact, as no or little chemicals are used. Additionally, the high yield results in roughly twice the pulp produced per ton of raw material as compared to chemical pulps without affecting the positive optical properties (1).

For the bleaching of the high yield pulp the sodium dithionite and hydrogen peroxide are the main processes used alone or in combination in single or two-stage process to obtain a final brightness of 50 - 70 % (2, 3). Sodium dithionite, also known as sodium hydrosulphite, will react with the chromophores present in the lignin to make less colored compounds and consequently, to give a brighter aspect to pulp (4).

The major obstacle to use the mechanical pulp in high grades paper products is their well-known photo reversion properties when exposed to light or heat. This yellowing

• National Research Center, Cellulose & Paper Department, El-Tahrir St., Dokki, Cairo, P O 12622, EGYPT
† Corresponding author : E-mail ; elsakhawy@yahoo.com

phenomenon, also known as a brightness reversion occurs primarily for high yield pulps as a result of exposure to UV or sunlight and is attributed to complex photo oxidation and photo degradation reactions of lignin in the pulp (5).

Diguaiacylstilbenes and other lignin functional groups include coniferyl alcohol, hydroquinones, substituted biphenyl units, and quinines were photo reactive and rapidly yellowed under brightness reversion conditions. It is generally accepted that the yellowing reactions are initiated by ultraviolet radiation in the wavelength range 290-400 nm that is absorbed by lignin chromophores (5). The absorption of UV light induced the degradation of lignin and the photooxidation of CH_2- or CH(OH)- groups (6). The key radical formed as important intermediate in the photoyellowing process are phenoxyl radicals and ketyl radicals, which are oxidized in the presence of oxygen into chromophoric structures of the quinonoid type (7,8).

Although chemical modification of high yield pulp (e.g. methylation, acetylation, propionation (9) reduction or hydrogenation do alter some lignin structures and retard the photo yellowing process, the delay lasts only a short time. Furthermore, massive chemical modification of pulp is expensive and may result in other undesirable changes in properties. Adding photo stabilizers (inhibitors) to paper during papermaking or finishing is an alternative prevention of discoloration. The development of inhibitors to stabilize the brightness reversion of high yield pulp is receiving great attention. Two classes of additives that have shown promising results are UV absorbers and free radical scavengers, since they effectively screen out harmful UV or near UV light (10). To date the most effective UV absorbers for mechanical pulp are benzophenone and triazole-based structures. Free-radical scavengers that have been shown to be effective photostabilization agents include thiols, thioethers, ascorbic acid, and formats (11). The main drawbacks of these inhibitors are that the UV-absorbers are too expensive for paper products, ascorbic acid is readily oxidized in the paper into yellow product during dark storage, and many thiols are not suitable because of their odor.

New and effective inhibitors are needed if stable high-grade paper products are to be made from high-yield pulp. Antioxidant (trapping intermediate radicals) or quenchers (inactivate excited molecules) (12) provided a very effective means of retarding the photoaging process of mechanical pulps.

Unsaturated cyclic and acyclic structures in natural antioxidants could stabilize mechanical pulp towards brightness reversion. In principle olefinic additives could act as radical traps for hydroxy and phenoxy radicals. Furthermore, conjugated dienes can act as triplet quenchers for the exited state of lignin like structures (13).

The successful development of a photo-stabilizing reagent for mechanical pulps will require that the additive exhibit several important properties, including outstanding light stabilizing abilities, cost-effectiveness, imparts no color, and excellent long term thermal aging properties (14).

The objective of this study is to evaluate the effectiveness of some natural antioxidant additives for retarding the brightness reversion of high-yield pulps. Specifically, the antioxidant effects of potato peel extract; clove and chamomile volatile oils were examined when applied to high yield chemimechanical pulp. The effects of these additives were evaluated by monitoring the rates of photo reversion over an extended time period.

2. Experimental

2.1 Raw material

Depithed bagasse, supplied by Edfo mill, Egypt was used in this study. Bagasse is an indefinite material and the approximate composition of dry bagasse including the ash is taken as 46.5 % carbon, 6.5% hydrogen and 46% oxygen. The bagasse analyses are compiled in Table 1.

2.2 Chemimechanical bagasse pulping

The depithed bagasse fibers were first pretreated with 10 % sodium hydroxide (based on bagasse) at liquor ratio 10 : 1, at the room temperature overnight. After chemical treatment, bagasse fibrillation and refining was carried in a laboratory Valley beater at 2 % consistency for one hour. The pulp samples were disintegrated according to SCAN-M 2:64, and screened.

2.3 Bleaching condition

First hydrogen peroxide step (P): 12-15 % consistency, 4 % H_2O_2, 4 % NaOH, 0.3 % DTPA, 0.3 % $MgSO_4$, 3 % Na_2SiO_3. Bleaching time is 2 h at 70℃.

Second hydrogen peroxide step (P): 12-15 % consistency, 2 % H_2O_2, 2 % NaOH, 0.3 % DTPA, 0.3 % $MgSO_4$, 3 % Na_2SiO_3. Bleaching time is 2 h at 70℃.

Dithionite, hydrosulfite, step (D): 5 % consistency, 2% $Na_2S_2O_4$, 0.2% DTPA. Bleaching time is 90 minutes at 75℃. Before bleaching pulp was washed with 0.01 N H_2SO_4 then pH was adjusted to 6 by using Na_2CO_3 solution.

2.4 Sheet Formation

The paper sheets were prepared according to the S.C.A. Standard using the model S.C.A. sheet former (AB Lorentzen and Wettre). In this apparatus a sheet of 165 mm diameter (214 cm^2 surface area) was formed. The weight of oven - dry pulp used for every sheet was about 1.43 g. After sheet formation, the sheet was pressed for 4 minutes (at 5 kg /cm^2) using a hydraulic press. Drying of the test sheets was made with the help of a rotating cylinder drum at 120℃. The sheets were then placed for conditioning at 65% relative humidity and temperature ranging from 18 - 20℃. After conditioning, in accordance to SCAN - M 8:76 standard methods, the tear factor, tensile strength and the basis weights of the sheets were measured.

Two types of paper was used in this study, one is rolling paper from Rakta Co, Egypt, and the other is that of high yield bagasse pulp. Table 1 shows the chemical analysis of bagasse pulp, bleached bagasse pulp and Rakta paper sheets.

2.5 Extraction of antioxidant

Table 1. The chemical analysis of bagasse, bagasse pulp, bleached bagasse pulp and Rakta paper sheets

Sample	Whole bagasse	Bagasse pulp	Bleached bagasse pulp	Rakta Paper sheets	Method
Lignin %	20.1	13.7	4.64	2.52	Tappi T 222 om-88
α- Cellulose	43.3	54.5	81.0	73.3	Tappi T 203 om-88
Pentosan	28	-	-	-	Tappi T 223 om-84
Ash %	2.21	1.27	1.23	1.77	Tappi T 211 om-86

2.5.1 Isolation of the volatile oil from the Clove and Chamomile

The volatile oils from one kg crushed Clove (A) and Chamomile (C) were hydrodistilled separately in Clevenger-like apparatus for 3h (15).

2.5.2 Solvent extraction

Accurate weight (600 g) of potato peel (B) lyophilized material was extracted with I L ethanol for 24 h at room temperature in dark. The solvent was evaporated at 50℃ under vacuum.

2.5.3 Scavenging effect on 1,1-diphenyl-2-picrylhydrazyl (DPPH) radical

The effects of different extracts were estimated according to the standard methods. The extract (4 mg) in 4 ml methanol was added to a 1 ml solution of DPPH radical in methanol (final concentration of DPPH adjusted to 0.2 mM). The mixture was shaken vigorously and allowed to stand for 30 min; then, the absorbance was measured at 517 nm using spectrophotometer (UV-160 Ipc) all tests were run in three replicate and averaged (16).

2.6 Paper treatment with antioxidant

Paper sheets were immersed in the different concentration (1 - 8 %) from the desired antioxidant solution for a 30 second. After immersion, the paper sheets were pressed between two filter paper sheets to remove the excess solution, and then dried on oven at 60 ℃ for 1h. Weight differences between wetted and dried paper sheets illustrate the weight of retained antioxidant solution.

2.7 Paper aging

Exposure of the paper sheets were assessed alongside with a Standard Blue Scale in accordance with ASTM:G23-1990 using Tera Light Fastness Tester (17). The emitted ligth was in the wavelength 280 to 420 nm with the maximum intensity at ~ 350 nm, the temperature was controlled at 35℃.

2.8 Brightness measurements and evaluation

The brightness of all examined paper sheets samples was measured at a wavelength of 457 nm with a Hunterlab instrument using MgO as standard white.

Post-Color number (PCN) was calculated for aged paper according to the Giertz equations (18):

$$PCN = 100\ [(k/s)\ aged - (k/s)\ initial]$$

$$\text{Where } k/s = [(1 - R\#)^2/(2R\#)]$$

$$R\# = brightness/100$$

% Photostabilization effect (19) =

$$100 \times \frac{\Delta \text{in Tappi brightness of control} - \Delta \text{in Tappi brightness of sample}}{\Delta \text{in Tappi brightness of control}}$$

3. Results and Discussion

The profile scavenging of all extracts on DPPH is shown in Fig. 1. The ethanol extract of potato peel exhibited 77 %. Clove and

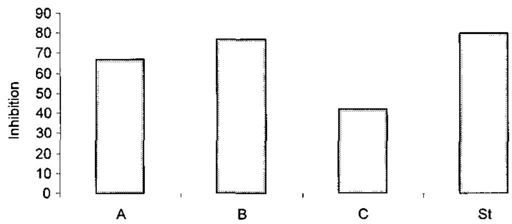

Fig. 1. Scavenging effect of A, B and C on DPPH radical comparing with St (butyl hydroxy anisol).

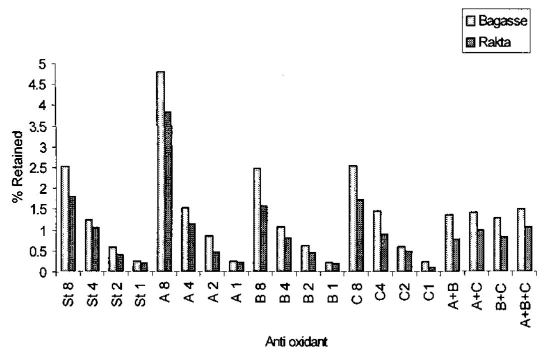

Fig. 2. Retained % for different anti oxidant.

chamomile volatile oils exhibited 67 and 42 % respectively. The remarkable antioxidant activity of potato peel extract might be due to the phenloic nature of it.

Fig. 2 shows the retained % from different antioxidant at bagasse and Rakta paper. It could be noticed that bagasse papers retains higher antioxidant than Rakta paper by about 30 ± 10 % because it have not any additives or fillers. For both papers as the antioxidant concentration increased the retained %

Table 2. Post color number (PCN) and % photo stabilization effect (P S) of antioxidant on bagasse papers after different radiation time

Time, h	20		42		85		165		250	
	PCN	P S %	PCN	P S %	PCN	P S %	PCN	P S %	PCN	P S %
Blank	10.6	--	17.9	--	20.2	--	26.5	--	33.5	--
St 8	11.3	- ve	17.9	0	18.4	6.78	24.4	5.59	28.4	10.2
St 4	13.1	- ve	21.2	- ve	24.6	- ve	27.6	- ve	29.9	7.19
St 2	15.4	- ve	22.4	- ve	25.7	- ve	28.4	- ve	31.0	5.39
St 1	16.2	- ve	23.9	- ve	26.8	- ve	29.9	- ve	33.2	1.06
A 8	18.7	- ve	23.9	- ve	26.5	- ve	29.0	- ve	30.0	5.99
A 4	17.7	- ve	18.9	- ve	20.4	- ve	21.6	13.3	24.1	19.8
A 2	15.2	- ve	16.0	8.33	18.1	7.63	19.9	18.2	24.1	19.8
A 1	10.4	1.40	14.8	13.9	16.6	13.6	19.5	19.6	22.4	24.0
B 8	8.83	14.1	11.0	33.4	13.2	28.0	19.5	19.6	21.6	25.7
B 4	7.67	23.9	12.4	25.0	15.2	19.5	19.5	19.6	21.9	25.1
B 2	7.34	26.8	13.2	21.3	16.0	16.1	20.4	16.8	22.1	24.6
B 1	9.52	8.45	13.8	18.5	16.6	13.6	21.4	14.0	24.6	18.6
C 8	19.1	- ve	21.2	- ve	21.2	- ve	22.6	10.5	27.0	13.2
C 4	17.5	- ve	20.9	- ve	22.9	- ve	23.6	7.69	27.3	12.6
C 2	12.4	- ve	18.3	- ve	23.5	- ve	27.3	- ve	28.1	10.8
C 1	13.6	- ve	18.1	- ve	24.9	- ve	27.6	- ve	31.0	4.79
A+B	11.5	- ve	17.7	0.93	17.5	10.2	21.2	14.7	24.4	19.2
A+C	14.8	- ve	19.7	- ve	20.9	- ve	24.6	4.90	25.7	16.2
B+C	17.9	- ve	20.4	- ve	22.6	- ve	22.6	10.5	24.6	18.6
A+B+C	17.1	- ve	17.1	3.70	17.9	8.47	18.9	21.7	23.1	22.2

St = Standard BHA (Butyl hydroxy anisol)
A= Clove volatile oil
B= Potato peel
C= Chamomile volatile oil
St 8 ... St 1 = Paper immersed in a 8 ... 1 % St solution
A+B Paper immersed in equal volume of 4 % A and B solutions

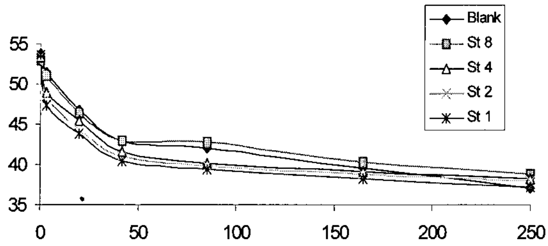

Fig. 3. Effect of St on stabilization of bagasse paper.

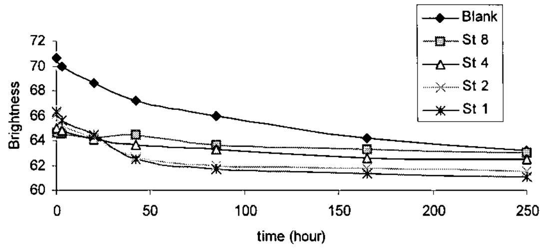

Fig. 4. Effect of St on stabilization of Rakta paper.

increased. Also it could be noticed that papers absorbs higher clove volatile oil (A) % than other antioxidant. The retained antioxidant % on bagasse and Rakta papers was about 0.25 & 0.21 at 1 % antioxidant and 0.60 & 0.5 at 2 % antioxidant and 1.3 & 1 at 4 % antioxidant and 2.5 & 1.7 at 8 % antioxidant (except for clove volatile oil where it reaches to 4.8 & 3.8) respectively.

3.1 Effect of BHA (Butyl hydroxy anisol) (St)

Butyl hydroxy anisol was used as standard antioxidant (St) for comparison. BHA has a reddish yellow color so, as its concentration increased on paper the brightness decreased. After radiation the brightness is higher for paper treated with higher BHA concentration, but it still lower than that of blank for both bagasse and Rakta paper (Figs. 3 & 4). Some improvement could be noticed for bagasse paper (Fig. 3) after radiation for 85 hours or more when using 8 % BHA. Other BHAconcentration stabilizes bagasse paper after 250 h radiation by different extent decreased with decreasing BHAconcentration. Table 2 confirms that PCN is higher for treated bagasse paper and photo stabilization effect is negative till 250 h radiation where photo stabilization effect was observed but it decreased, as BHA concentration decreased, from 10 % for 8 % BHA to 1 % for 1 % BHA. It could be noticed that as the BHA concentration increased the PCN decreased and the photo stabilization effect % increased. In creases in radiation time cause an increase in both of PCN and in photo stabilization effect %. For Rakta paper Table 3 shows that PCN is higher for treated paper and photo stabilization effect is negative for all studied BHA concentration at all given radiation time.

3.2 Effect of clove volatile oil (A)

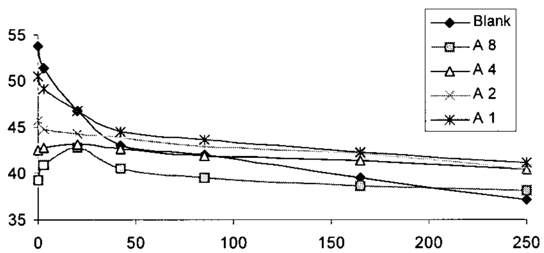

Fig. 5. Effect of A on stabilization of bagasse paper.

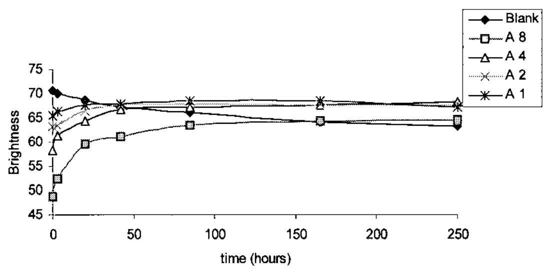

Fig. 6. Effect of A on stabilization of Rakta paper.

Clove volatile oil(A) has a distinct greenish brown color that imparts the paper a color and decreases the brightness as A concentration increase, so brightness is higher for paper treated with lower A concentration at all given radiation time. As the radiation time increase the blank brightness decreased while the brightness increased for the treated paper due to both the discoloration of A color by radiation and due to the antioxidant stabilization effect. Fig. 5 shows that stabilization of treated bagasse paper against photoyellowing could be noticed after 40 h radiation for paper treated with A1 or A2. A4 stabilize paper at radiation time longer than 85 h while A8 stabilization effect began at 210 h radiation. For Rakta paper Fig 6 shows that stabilization effect could be noticed for treated paper after 40 h radiation for all used a concentration except A8 which began to stabilize paper after 165 h radiation. It could be concluded that lower A concentration is enough for paper stabilization under the studied

Table 3. Post color number (PCN) and % photo stabilization effect (P S) of antioxidant on Rakta papers after different radiation time

Time, h	20		42		85		165		250	
	PCN	P S %	PCN	P S %	PCN	P S %	PCN	P S %	PCN	P S %
Blank	1.12	--	1.93	--	2.69	--	3.91	--	4.64	--
St 8	3.91	- ve	3.77	- ve	4.35	- ve	4.57	- ve	4.80	- ve
St 4	3.98	- ve	4.35	- ve	4.57	- ve	5.10	- ve	5.18	- ve
St 2	3.91	- ve	5.02	- ve	5.58	- ve	5.74	- ve	5.98	- ve
St 1	3.77	- ve	5.18	- ve	5.82	- ve	6.15	- ve	6.31	- ve
A 8	7.62	- ve	6.40	- ve	4.42	- ve	3.84	1.54	3.70	17.3
A 4	3.84	- ve	2.18	- ve	2.00	23.4	1.69	52.3	1.34	66.7
A 2	2.37	- ve	1.64	14.3	1.52	40.4	1.64	53.8	1.69	58.7
A 1	1.69	- ve	1.58	17.1	1.17	53.2	1.23	64.6	2.00	52.0
B 8	1.12	0	1.17	37.1	1.23	51.1	1.23	64.6	1.34	66.7
B 4	1.12	0	1.12	40.0	1.23	51.1	1.34	61.5	1.40	65.3
B 2	1.12	0	1.12	40.0	1.29	48.9	1.34	61.5	1.46	64.0
B 1	1.34	- ve	1.06	42.9	1.29	48.9	1.46	58.5	1.64	60.0
C 8	12.1	- ve	10.9	- ve	9.04	- ve	7.17	- ve	6.82	- ve
C 4	6.06	- ve	6.15	- ve	5.74	- ve	4.72	- ve	3.91	13.3
C 2	5.98	- ve	5.66	- ve	4.72	- ve	3.77	3.08	2.95	32.0
C 1	4.42	- ve	4.13	- ve	1.81	29.8	1.58	55.4	1.46	64.0
A+B	3.70	- ve	3.42	- ve	2.95	- ve	1.81	49.2	1.52	62.7
A+C	6.48	- ve	5.98	- ve	4.72	- ve	3.49	9.23	2.24	46.7
B+C	6.06	- ve	5.26	- ve	3.08	- ve	2.49	32.3	2.06	50.7
A+B+C	4.35	- ve	3.28	- ve	2.69	0	1.64	53.8	1.29	68.0

Footnotes as in table 2

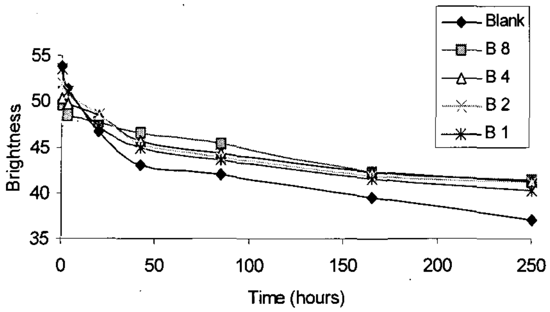

Fig. 7. Effect of B on stabilization of bagasse paper.

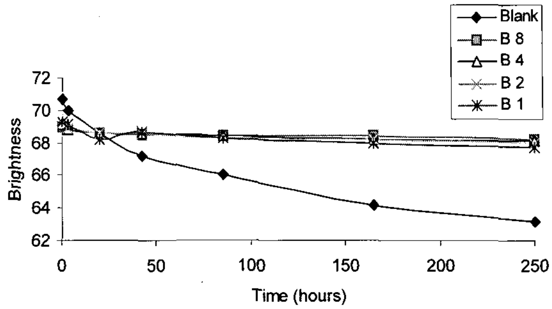

Fig. 8. Effect of B on stabilization of Rakta paper.

level of radiation time. PCN and photo stabilization effect % in Table 2 confirms that for bagasse paper lower A concentration provides lower PCN and higher photo stabilization effect % over all studied radiation time. Photo stabilization effect % increased as the radiation time increase. The same trend was observed for Rakta paper, Table 3, but with maximum photo stabilization for A1 at 165 h radiation.

3.3 Effect of potato peel (B)

Although potato peel (B) is a white colloid it causes a slight decrease on paper brightness after treatment. Figs. 7 and 8 show that after 20 h radiation B stabilizes paper by an observable percent. All used B concentration obviously stabilize paper against radiation. Higher B concentration cause more stabilization, but lower B concentration is more economic and stabilizes paper to a good extent. For bagasse paper Table 2 shows that PCN increased by increasing radiation time and it is higher for paper treated with lower B concentration. Photo stabilization effect % is higher for paper treated with higher B concentration with maximum values at 42 h radiation. Table 3 for Rakta paper shows that PCN is higher for paper treated with lower B concentration and for paper subjected to longer radiation time. High % photo stabilization effect was attained for paper radiated for 42 or longer and this % increased as the radiation time increased.

3.4 Effect of chamomile volatile oil (C)

Although chamomile volatile oil (C) color is light blue it decrease the paper brightness after

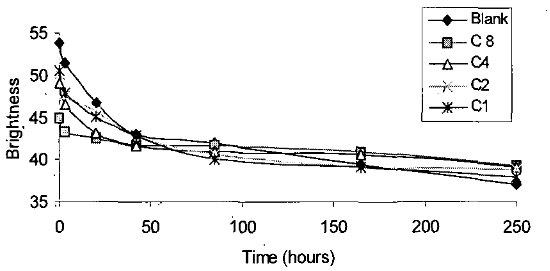

Fig. 9. Effect of C on stabilization of bagasse paper.

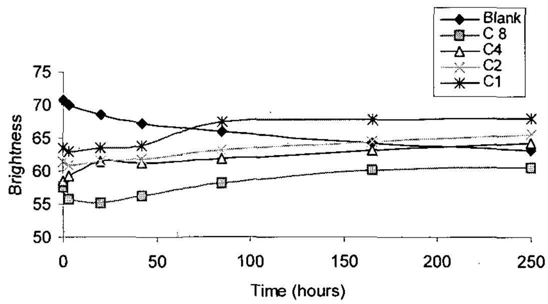

Fig. 10. Effect of C on stabilization of Rakta paper.

treatment by an elevated level in direct relation with the C concentration. Fig. 9 shows that the brightness of treated bagasse paper decreased as the radiation increased and the brightness is lower compared with the blank till 165 h radiation time after which stabilization could be noticed. For treated Rakta paper, Fig. 10, the brightness increases by radiation as a result of color removal. Positive effect is noticed for paper treated with lower C concentration and at longer radiation time. Table 2 shows that for bagasse paper PCN is higher for treated paper than the control except at 165 h radiation for C4 and C8 and at 250 h radiation for all samples. Positive photo stabilization effect % was observed at 165 h radiation for C4 and C8 and at 250 h radiation for all samples and it is higher for paper treated with higher C concentration. For Rakta paper Table 3 shows that lower PCN and higher % photo stabilization effect was noticed for paper treated with lower C conc. This may be due to that Rakta paper has higher brightness and C color affected it more than bagasse paper.

3.5 Effect of mixtures

Equal volumes from 4 % solution of A, B and/or C was used for paper treatment. Fig. 11 shows that for treated bagasse paper ABC stabilize paper subjected to radiation for 42 h or longer. Also AB have the same trend but at lower extent. AC and BC have positive stabilization effect after at least 165 h radiation. For Rakta paper, Fig. 12 shows that positive stabilization effect by all used mixtures was noticed for treated paper after 165 hours radiation or longer. ABC followed by AB provides a higher stabilization. Table 2 shows that for treated bagasse paper although PCN increased by increasing radiation time the % photo stabilization effect increased. For treated Rakta paper PCN decreased by increasing radiation time and photo stabilization effect % obviously increased. For all treated paper ABC mixture followed by AB provides higher stabilization for paper.

4. Conclusion

It is clear that paper treated with potato peel (B) antioxidant followed by clove volatile oil (A) exhibits lower brightness reversion and better photo stabilization effect after exposure to UV lights in comparison with Butyl hydroxy anisol (St) or chamomile volatile oil (C).

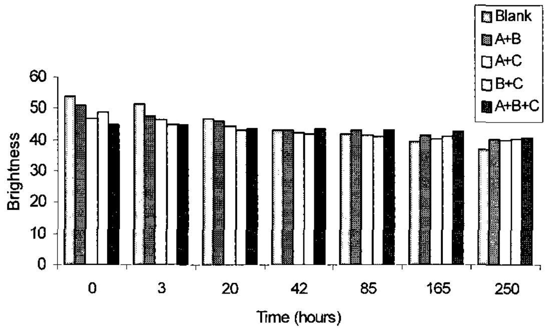

Fig. 11. Effect of mixtures on stabilization of bagasse paper.

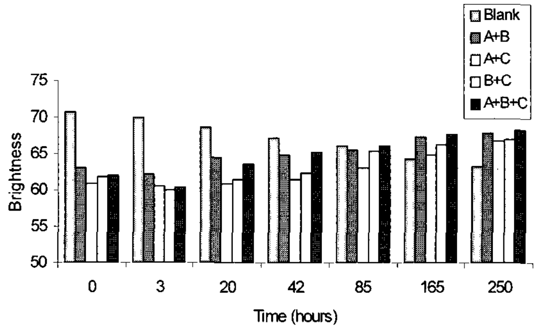

Fig. 12. Effect of mixtures on stabilization of Rakta paper.

Literature Cited

1. T. G. Jones, J. D. Richardson, Appita J. 54 (3):265-271 (2001).
2. J. Ramos, F. Davalos, F. Navarro, 97 Nanjing International Symposium on High Yield Pulping, Nanjing, P. R. China, 21-23 Nov. 152-186 (1997).
3. F. Peng, T.Friberg, Appita J. 45 (4):243-245, 259 (1992).
4. E. Svensson, H. Lennholm, T. Iversen, J. Pulp Paper Sci., 24 (8):254-260 (1998).
5. C. M.Cook, X.Pan, A. J.Ragauskas, J. Wood Chem. Technol. 16 (3):327-345 (1996).
6. Uwe Muller, M.Ratzsch, M.Schwanninger, M. Steiner, H.Zobl, J. Photochem. Photobiology B: Biology 69: 97-105 (2003).
7. M.Paulsson, R.Simonson, U.Westermark, Nordic Pulp Pap. Res. J.10 (1): 62-67 (1995).
8. M.Paulsson, R.Simonson, U.Westermark, Nordic Pulp Pap. Res. J.11 (4):234-238 (1996).
9. M. Paulsson, J.Parkas, J. Wood Chem.Technol., 20 (2):205224 (2000).
10. C.Li, C. M.Cook, A. J.Ragauskas, J. Wood Chem.Technol., 19(1-2):2741 (1999).
11. L. C.Harvey, J. Wood Chem.Technol., 17 (1&2):2739 (1997).
12. M.Paulsson, R.Simonson, U.Westermark, Nordic Pulp Pap. Res. J. 9 (4):232-236 (1994).
13. A. J. Ragauskas, Tappi J 76 (12): 153-157 (1993).
14. A. J. Ragauskas, Cellulose Chem. Technol. 28: 265-272 (1994).
15. J.F.Clevenger, J. Am. Pharm. Assoc. 17: 346 349 (1928).
16. Y. Gow-Chin, W. She-Ching, D.Pin-Der, J. Agric Food Chem. 44:1687-1690 (1996).
17. F. M.Tera, Tera Light Fastness Tester, Egypt Patent 15182 (1981).
18. H. W. Giertz, Svensk Papperstidn 48 (13) : 317(1945).
19. A. J. Ragauskas, Tappi J., 76(12):153-157 (1993).