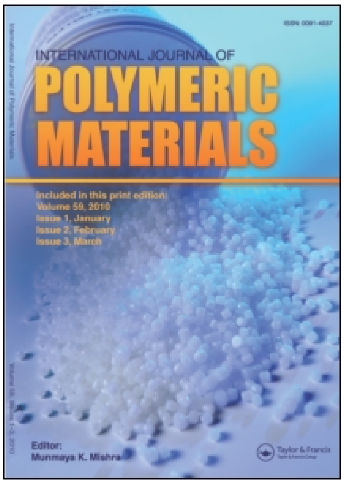

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The Stabilization of Paper Cellulose and Cellulose Diacetate by the Natural Compounds

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Attempts to stabilize paper cellulose and cellulose diacetate through use of natural plant lightstabilizers are discussed.

KEY WORDS paper cellulose, cellulose diacetate, stabilization, natural compounds

INTRODUCTION

The light is one of the more important factors of the ageing of cellulose and its derivatives, in particular, paper and cellulose diacetate (CDA) films. For photochemical degradation studies UV-irradiation of wavelength 250–395 nm is used. Often, though not always, it is adequate to simulate real ageing in natural conditions.

Light ageing of CDA and paper cellulose (PC) leads to deterioration of their physical/mechanical properties.

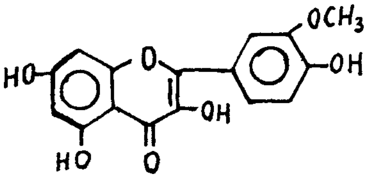
To increase the lightfastness of these polymers the use of lightstabilizers (among them UV-absorbers and phenolic antioxidants) has been proposed. The dyeing of cellulose and CDA by some synthetic dyes increases the lightfastness of these items.[1] However, lightstabilizers are sometimes toxic products and their use is connected with ecological problems. In this connection the light-stabilization of CDA by natural compounds is very real from the viewpoint of environmental protection and the waste of natural materials.

EXPERIMENTAL

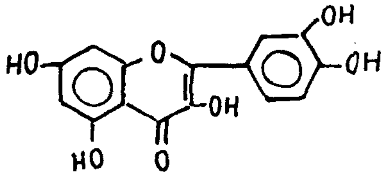
Studies were carried out on CDA films obtained from 5% CDA solutions in acetone, the paper subparchment type A and the micalent paper.

As plant lightstabilizers flavonoids, isoramnetin, quercetin and the extracts of walnut nut peel, staining henna and summa of dyeing substances of mulberry-tree polypore (SDS) have been used.

The structure of isoramnetin and quercetin are presented below

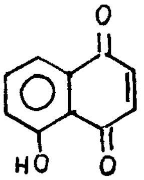


isoramnetin

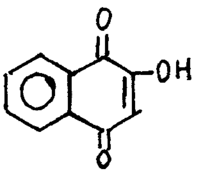


quercetin

The extracts from nut skin and henna contain a substance with quinone and phenole groups: juglone and lavsone accordingly:



juglone



lavsone

The structure of the SDS substances are unknown.

The lightstabilizers are introduced into CDA solutions in acetone. UV-irradiation is accomplished by a middle pressure mercury-quartz lamp PRK-2 at a distance of 35 cm from the sample. The sample strength, average degree of polymerization, reflection coefficient, relative viscosity and COOH- and CHO-groups contained were measured by known methods.

RESULTS AND DISCUSSION

From Table I it is shown that UV-irradiation of micalent paper leads to a large extent the decreasing of the degree of polymerization and an increasing of carboxy- and aldehyde concentration.

The treatment of paper by plant stabilizers retards the decreasing of the degree of polymerization and carboxyl and aldehyde group formation. Quercetin is the best stabilizer.

TABLE I

The effect of UV-irradiation on paper lightfastness

Stabilizer, its concentration (%) or dilution in extracts	COOH-groups, %		CHO-groups, %		Average degree of polymerization	
	Before irradiation	After irradiation	Before irradiation	After irradiation	Before irradiation	After irradiation
No stabilizers	0.048	0.123	0.061	0.028	970	390
Quercetin 1:1000	0.096	0.167	0.012	0.230	990	415
Nut 1:10	0.051	0.118	0.065	0.230	990	420
Henna	0.056	0.117	0.064	0.215	985	425
SDS 1.5%	0.049	0.119	0.063	0.226	995	420

Figure 1 shows that the index of break resistance (lg N) of the mica paper basis after 10 hours of UV-irradiation sharply decreased and paper lost its strength completely. However, the initial paper deteriorates more than stabilized paper. At the same time the reflection coefficient of stabilized samples are maintaned at relatively high levels, whereas for unstabilized ones the value decreases very sharply as a consequence of the strong yellowing of mica paper basis (Figure 2).

The daylight ageing is accomplished in more mild conditions than under UV-irradiation, however at the same time the same increasing of the reflection coefficient occurs (Table II) as a consequence of the partial discoloration of the stabilized samples. The mechanical strength and pH of the paper both under UV-irradiation and daylight are considerably decreased.

To determine the light stabilizing effect, CDA was used as a paper model since it dissolves in organic solvents permitting one to obtain optically transparent films.

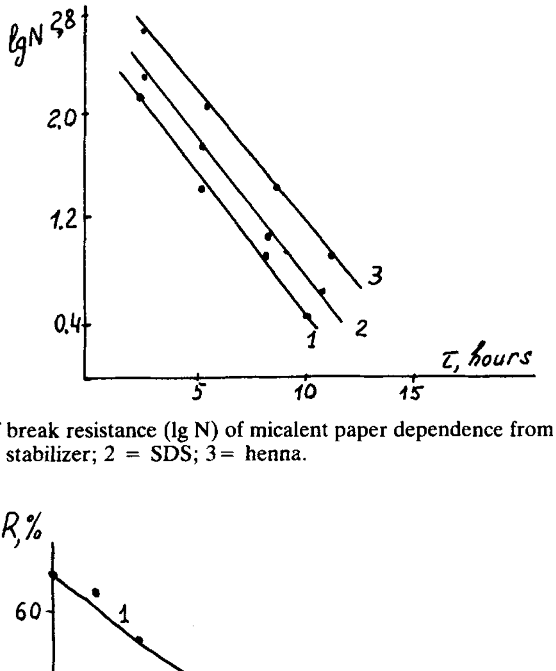


FIGURE 1 Index of break resistance (lg N) of micalent paper dependence from the duration of UV-irradiation (τ) 1 = no stabilizer; 2 = SDS; 3 = henna.

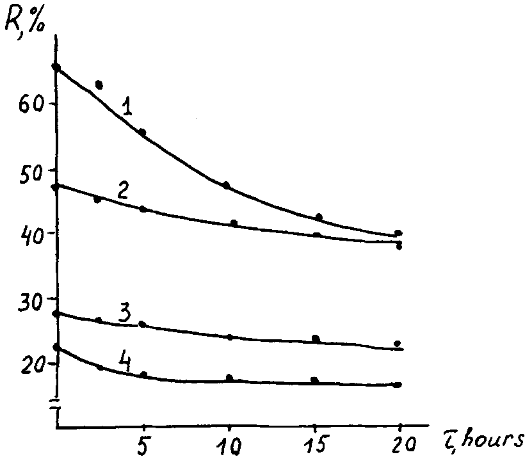


FIGURE 2 The reflection coefficient (R) of micalent paper dependence from the duration of UV-irradiation 1 = no stabilizer; 2 = quercetin; 3 = henna; 4 = nut.

Based on the data presented in Table III, introduction of plant stabilizers into CDA solution exerts practically no influence on the relative viscosity of the initial polymer, observed before irradiation. The high stabilizing activity of the additives was evaluated by the degree of retention by the polymer of its viscosity after irradiation (Table III).

Analysis of the data presented in Table II shows that initial CDA is light unstable and preserves only 7.1% of the initial viscosity after irradiation. Introduction of plant stabilizers in CDA in all cases results in a significant increase of the light stability index. The best stabilizer is isoramnetin at a concentration of 3 weight%. Its use provided viscosity retention of 78%. Under UV-irradiation the discoloration of CDA films, containing isoramnetin occurred by isoramnetin loss. The isoramnetin loss constant was determined from the initial section of the kinetic curve for the change in the optical density. As this relationship in logarithmic coordinates expresses a straight line (Figure 3, curve 1), the loss constant was calculated by an equation of 1st order reaction.[1]

As follows from Figure 3 (curve 2), the isoramnetin loss rate is inversely pro-

TABLE II

The reflection coefficient (R) of initial and stabilized subparchment paper type A during daylight ageing

Stabilizer, its concentration (%) or dilution in extracts	The ageing time, days				
	0	3	6	9	12
No stabilizer	83.30	72.86	69.52	68.13	65.01
Isoramnetin 1:10	45.18	45.32	45.43	45.56	45.62
Quercetin 1:1000	53.51	53.76	53.81	53.88	54.16
Henna	32.43	32.48	32.59	32.71	33.16
Nut 1:10	31.27	31.77	31.25	30.91	31.46
Nut 1:20	33.59	33.68	33.93	33.82	34.09
SDS 0.1%	53.14	53.05	53.46	53.62	53.98
SDS 0.5%	25.94	26.06	26.21	26.63	27.56
SDS 1.0%	22.83	23.15	23.49	23.76	23.84
SDS 1.5%	19.20	19.28	20.12	20.36	20.37

TABLE III

Relative viscosity change of stabilized CDA solutions during ultraviolet irradiation for 24 hour period

Stabilized CDA solution	Concentration, %	Relative viscosity		Viscosity retaining, %
		Before ir.	After ir.	
No stabilizer	—	1.12	0.08	7.1
Isiramnetin	1	1.16	0.56	48.2
Isoramnetin	3	1.30	1.02	78.0
Quercetin	3	1.04	0.74	71.5
Nut	3	1.14	0.74	64.6
Henna	3	1.17	0.73	61.9
SDS	1	1.02	0.59	58.8

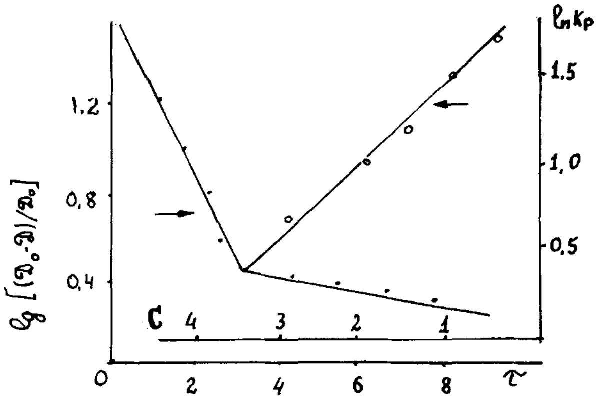


FIGURE 3 Kinetics of isoramnetin loss (1) and its rate under UV-irradiation (2). K = isoramnetin loss constant; τ = duration; ΔD = optical density changing; C = concentration.

portional to the additive concentration. At isoramnetin concentration 3 weight% the rate of its loss was minimal.

The data are in accordance with known information.[2] The additive molecules at low concentration distribute in the polymer with formation of a monolayer. On increasing the isoramnetin concentration the interaction forces between additive and polymer are overcome and all the isoramnetin molecules are converted to aggregates which fade more slowly by a concentration quenching effect.[3]

As the plant stabilizers absorb in the UV-region, one of the possible mechanisms for their lightprotective action is screening.

In view of the phenole OH-groups present in the structures of the plant stabilizers, it can be proposed that an additional mechanism of lightprotection is inhibition.

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