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The Light Stability of Azo Dyes and Azo Dyeings II. Perspiration–Light Stability of Dyeings with Reactive and Non-Reactive Derivatives, Respectively, of Two Selected Azochromophores

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ABSTRACT

The perspiration–light stability of RC-dyed (conventional reactive dyeing) and N-dyed (substantive dyed without subsequent rinsing) cotton fabric dyeings produced with the members of 'A'-dye and 'B'-dye groups, respectively, have been studied. Three main factors influence the perspiration-light stability of the studied dyeings: the nature of the dye–fibre bond, the light stability of the released hydrolysed dye molecules and the action of the salts in perspiration on the latter. The preferred MCT-covalent bonds in the 'A'-group dyeings lead to higher proportion of released dye molecules in acidic perspiration under exposure, than the preferred, and to a lesser extent to acid sensitive VS-type bonds in the 'B'-group dyeings. Covalently fixed dyes in dyeings result in much better perspiration-light stability than dyeings with released hydrolysed dye molecules, and the difference in light stability between hydrolysed dye molecules of the two dye groups, as well as their different salt sensitivity, result in improved perspiration–light stability compared to light stability in the 'B'-group dyeings, and lower perspiration–light stability compared to the light stability in the 'A'- group dyeings. © 1998 Elsevier Science Ltd

Keywords: azodyeing, perspiration–light stability, additive effect, dye–fibre bond.

INTRODUCTION

The perspiration–light stability of numerous azoreactive dyeings is rather poor. The mechanism of photofading under simultaneous exposure to perspiration and light is usually considerably different to that of light induced photofading [1]. The presence of moisture plays an important role in the former process, usually increasing the rate of photofading [2].

Datyner *et al.* [3] observed accelerated photofading in the case of two azo reactive cellophane dyeings out of four studied ones. In one case it was found that the major primary photochemical reaction was photoejection of electrons enhanced in the aqueous environment, while in the second case a back reaction between hydroxyl radicals and dye molecules was facilitated by the aqueous environment.

The light stability of VS and MCT type wet azo reactive cellulose dyeings, respectively, was studied by Okada *et al.* [4–9] under different atmospheric conditions. The VS dyeings studied underwent photofading by an oxidative mechanism caused by singlet oxygen [4]. Dyeings with easily reducible azo reactive dyes facilitate the reductive mechanism in photofading. The governing reaction— oxidation or reduction—depends not only on the sensitivity of the dyeing towards oxidation or reduction but primarily on the concentration of oxygen in the exposured system [5]. Similar conclusions have also been made on MCT type azo reactive dyeings [6].

The pH dependence of the photofading of the VS type azo reactive dyeing in the range from 2.5–9 showed that the highest rate occurred at both ends of the studied range [7]. Three studied cellulosic MCT type azo reactive dyeings showed no fading on exposure in deaerated water and oxidative fading in aerated water, whereas they underwent reductive fading in the presence of DL-mandelate in a nitrogen atmosphere [8].

Imada *et al.* [1] observed photoreduction under joint exposure to perspiration and light in the photofading of azo reactive dyeings, whereas photooxidation occurs in the same systems under exposure to light only. With regard to the influence of the chemical structure of dye, they concluded that reactive dyes containing an H- or K-acid azo chromophore performed better in the conventional light fastness test and that reactive dyes of the J- or γ-acid type exhibited a higher fastness in the perspiration–light test.

Photofading of VS type azo reactive dyeings on cotton cellulose under exposure to perspiration and light was studied by Okada *et al.*, [10]. The observed mechanism was either photooxidation or photoreduction, depending on the structure of the dye and on the conditions of exposure.

EXPERIMENTAL

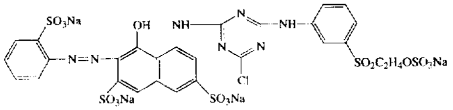
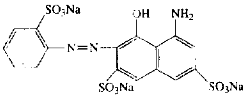
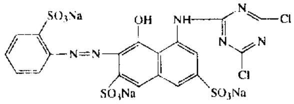
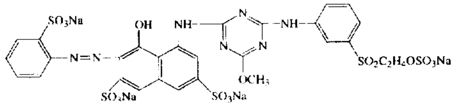
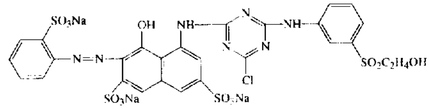
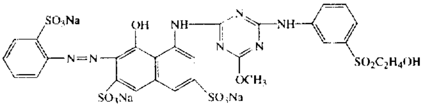
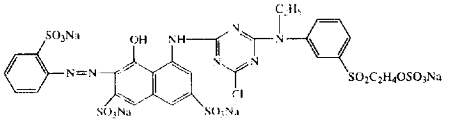
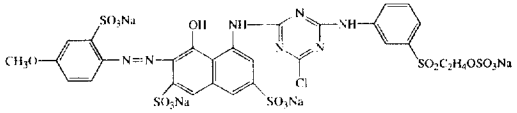
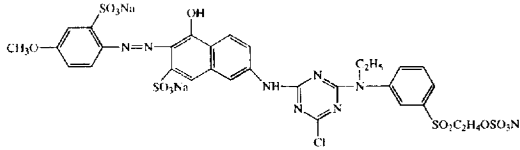
Materials

Dyes

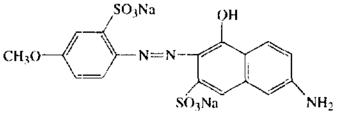
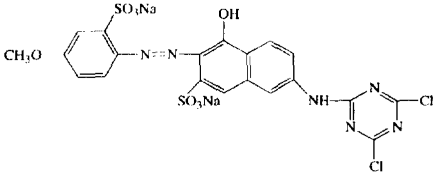
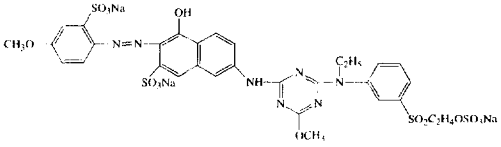
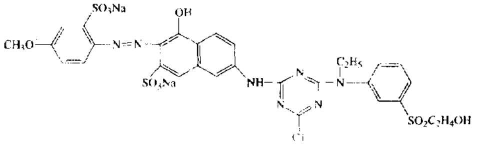
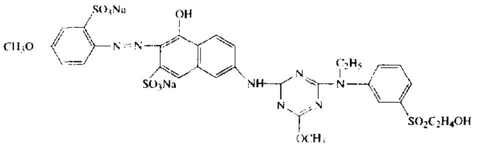
The studied dyes produced by Sumitomo Chemical Co. Ltd., Japan, are shown in Table 1.

TABLE 1

Characteristics of Dyes and Respective RC-Dyeings

Code	*Structure*	*Molar mass*	*Dye concentration in RC-dyeing (10^{-5}mol g^{-1} substrate)*	λ_{max} *(nm)*
A(MCT-VS)		984	1.95	510
A(Chr)		569	-	526
A(DCT)		717	2.31	512
A(MHT-VS)		981	1.89	534
A(MCT-VH)		882	1.92	534
A(MHT-VH)		878	-	534
A(MCT-VS)$_{Et}$		1012	2.08	512
A(MCT-VS)$_{Met}$		1014	1.90	534
B(MCT-VS)		940	2.05	502

continued

Code	*Structure*	*Molar mass*	*Dye concentration in RC-dyeing (10^{-5}mol g^{-1} substrate)*	λ_{max} *(nm)*
B(CHR)		497	–	494
B(DCT)		645	2.28	502
B(MHT-VS)		936	1.96	502
B(MCT-VH)		838	1.89	502
B(MHT-VH)		834	–	502

Cotton fabric

Plain woven scoured, bleached and mercerised cotton fabric:

- yarn density: warp: 288/10 cm; weft: 256/10 cm
- surface density: 108 g m^{-2}
- tensile strength: warp: 265 [N/5 cm]; weft: 235 [N/5 cm]
- elongation at break: warp: 8.25%; weft: 13.91 [%]
- yarn fineness: warp 20 [tex]; weft 20 [tex]
- equilibrium moisture regain at 65% r.h.: 7.33 [%]
- water retention: 44.74 [%]
- degree of polymerisation: 1495 ± 50

The composition of the artificial perspiration (JIS L 0848 Japanese Standard)

- 5 g l^{-1} lactic acid
- 5 g l^{-1} NaCl
- 5 g l^{-1} $Na_2HPO_4*12\ H_2O$
- 0.5 g l^{-1} L-histidine monohydrochloride monohydrate
- 0.5 g l^{-1} DL-aspartic acid
- 5 g l^{-1} sodium D-pantothenate
- 5 g l^{-1} glucose anhydrous
- acetic acid to adjust pH to 3.5
- distilled water

Methods

Dyeing procedures
These were effected as described in Part I of this investigation.

Treatment of dyeings with artificial perspiration
The dyeings were treated with artificial perspiration at ambient temperature for 30 min using a 1:50 fabric to liquor ratio. The samples were then squeezed to a wet pick up 120%.

Exposure to light
Dyeings were exposed to light for 264 h in a Xenotest 450 apparatus. Within this period, samples were removed at intervals to study the kinetics of the photofading.

Colour measurement
Colour coordinates of dyeings before and after the exposure for different periods of time were measured on a ICS Texicon colour measuring instrument. The data thus obtained enabled calculation of colour difference (ΔE^*_{ab}) between the samples before and after the exposure to be made.

Testing light stability in closed systems
The appropriate dyeings were placed in a quartz cuvette (Original Hanau No. 001 451). Three different dyeings were placed in the covette, dry, wetted with distilled water (120% pick up) or with artificial perspiration (120% pick up), respectively. Three different atmospheres were used at room temperature, viz., air, oxygen and nitrogen, respectively.

RESULTS AND DISCUSSION

Comparative observations with dyeings with dyes of the 'A'-group

The three hetero-bifunctional members show a distinct lowering in light stability in P–L testing compared with the corresponding data for L testing (Table 2). (P–L pertains to the measurement of light stability in the presence of artificial perspiration and L to the light stability measurement in the absence of artificial perspiration). Among P–L stabilities, that of the A(MCT-VS)$_{\mathrm{RC}}$ dyeing was the best, followed closely by the A(MCT-VS)$_{\mathrm{Et,RC}}$ dyeing; the A(MCT-VS)$_{\mathrm{Met,RC}}$ dyeing showed a more definite drop in P–L stability (Fig. 1).

No significant difference was observed within the P–L stabilities of the monofunctional A(DCT)$_{\mathrm{RC}}$, A(MCT-VH)$_{\mathrm{RC}}$ and A(MHT-VS)$_{\mathrm{RC}}$ dyeings (Fig. 2). P–L stability of all the three monofunctional dyeings was lower than the corresponding L stabilities (Table 2). The least drop in light stability caused by perspiration occurred on the A(MHT-VS)$_{\mathrm{RC}}$ dyeing. The P–L stability of the hetero-bifunctional dyeings generally exceeded that of the monofunctional dyeings, but the difference in the stability of the best monofunctional and worst hetero-bifunctional dyeing was negligible.

The P–L stability of the otherwise very light sensitive A(Chr)$_{\mathrm{N}}$ dyeing, significantly exceeded that of the A(MHT-VH)$_{\mathrm{N}}$ dyeing (Fig. 3). The P–L

TABLE 2

Comparison Between the Light and Perspiration–Light Stability of the Dyed Systems after Exposure to Light for 165 h

The code of dyed systems	ΔE^*_{ab}	
	Light stability	*Perspiration–light stability*
A(MCT-VS)$_{\mathrm{RC}}$	5.3	9.2
A(MCT-VS)$_{\mathrm{N}}$	5.7	22.2
A(MCT-VS)$_{\mathrm{Et,RC}}$	6.5	10.0
A(MCT-VS)$_{\mathrm{Met,RC}}$	8.0	11.2
A(DCT)$_{\mathrm{RC}}$	6.8	10.5
A(DCT)$_{\mathrm{N}}$	4.8	17.3
A(MCT-VH)$_{\mathrm{RC}}$	7.7	12.6
A(MHT-VS)$_{\mathrm{RC}}$	8.2	11.3
A(Chr)$_{\mathrm{N}}$	16.7	17.1
A(MHT-VH)$_{\mathrm{N}}$	16.6	32.5
B(MCT-VS)$_{\mathrm{RC}}$	9.1	8.3
B(MCT-VS)$_{\mathrm{N}}$	8.6	16.3
B(DCT)$_{\mathrm{RC}}$	10.4	8.8
B(DCT)$_{\mathrm{N}}$	9.3	9.4
B(MCT-VH)$_{\mathrm{RC}}$	8.7	8.2
B(MHT-VS)$_{\mathrm{RC}}$	8.7	6.8
B(Chr)$_{\mathrm{N}}$	23.5	21.9
B(MHT-VH)$_{\mathrm{N}}$	7.8	13.8

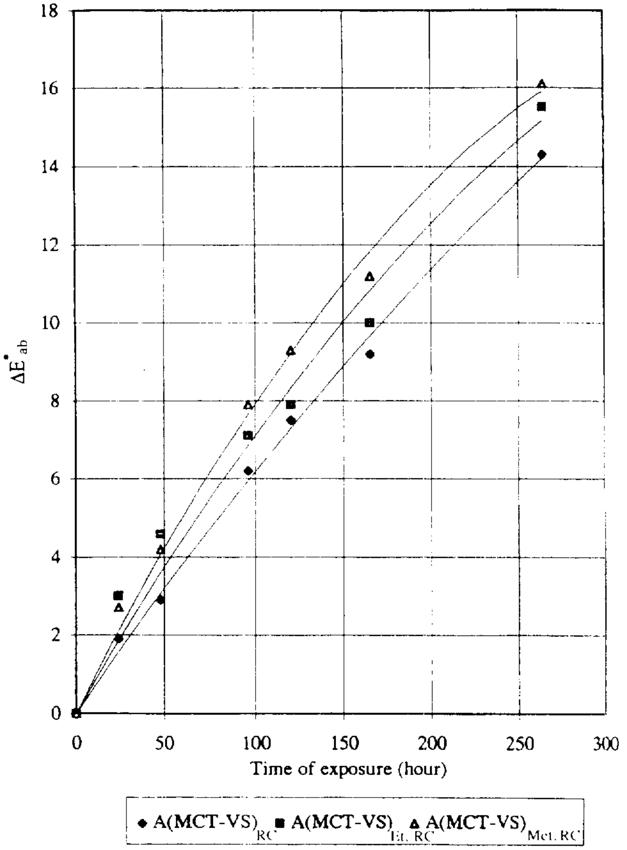


Fig. 1. Kinetics of photofading of the hetero-bifunctional reactive dyeings in the 'A'-dye group in the presence of artificial perspiration.

stability of the A(Chr)$_N$ dyeing under exposure of less than 120 h was better than the corresponding L stability. Both stabilities were identical when the exposure was for 165 h or longer. It is worthwhile to mention that the P–L stability of A(MHT-VH)$_N$ dyeing was much less than its L stability throughout the whole range of light exposure.

All the RC dyeings were significantly more stable to P–L exposure than the studied N-dyeings (Fig. 4). Comparing the P–L stability data obtained after exposure to light for 264 h, within the 'A'-group the value for the A(MCT-VS)$_{RC}$ dyeing was the best, whereas that of the A(MHT-VH)$_N$ dyeing was the worst.

Comparative observations with dyeings with dyes of the 'B'-group

The stability of the B(MHT-VS)$_{RC}$ dyeing to P–L exposure is higher than that of the B(MCT-VS)$_{RC}$, B(DCT)$_{RC}$ and B(MCT-VH)$_{RC}$ dyeings (Fig. 5).

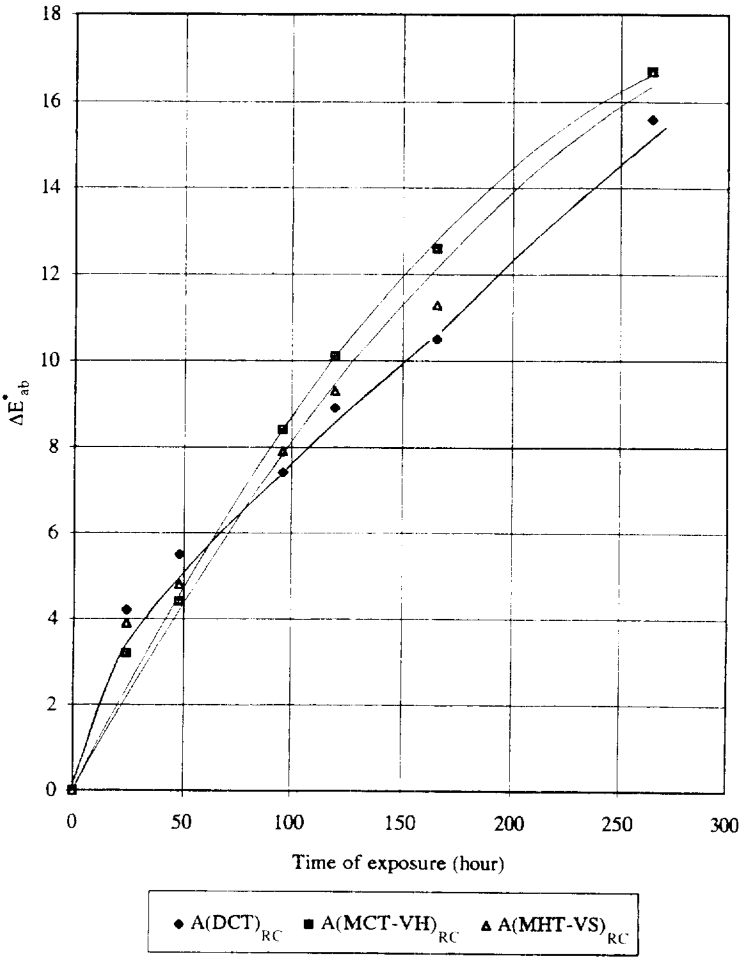


Fig. 2. Kinetics of photofading of the monofunctional reactive dyeings in the 'A'-dye group in the presence of artificial perspiration.

The P–L stability of the B(MCT-VS)$_{RC}$, B(DCT)$_{RC}$ and B(MHT-VS)$_{RC}$ dyeings was significantly higher than the L stability of the same systems. No significant difference was found between the P–L and L stability of the B(MCT-VH)$_{RC}$ dyeings.

The P–L stability of the B(MHT-VH)$_{N}$ dyeing was markedly better than that of the B(Chr)$_{N}$ dyeing (Fig. 6). However, in the case of the B(MHT-VH)$_{N}$ dyeing, the P–L stability was much less than the L stability, whereas in the case of the B(Chr)$_{N}$ dyeing the P–L stability was better than the L stability (Table 2).

The rate of P–L photofading of B(MCT-VS)$_{RC}$ dyeing was markedly slower than that of the B(MCT-VS)$_{N}$ dyeing (Fig. 7). Similar, but less well

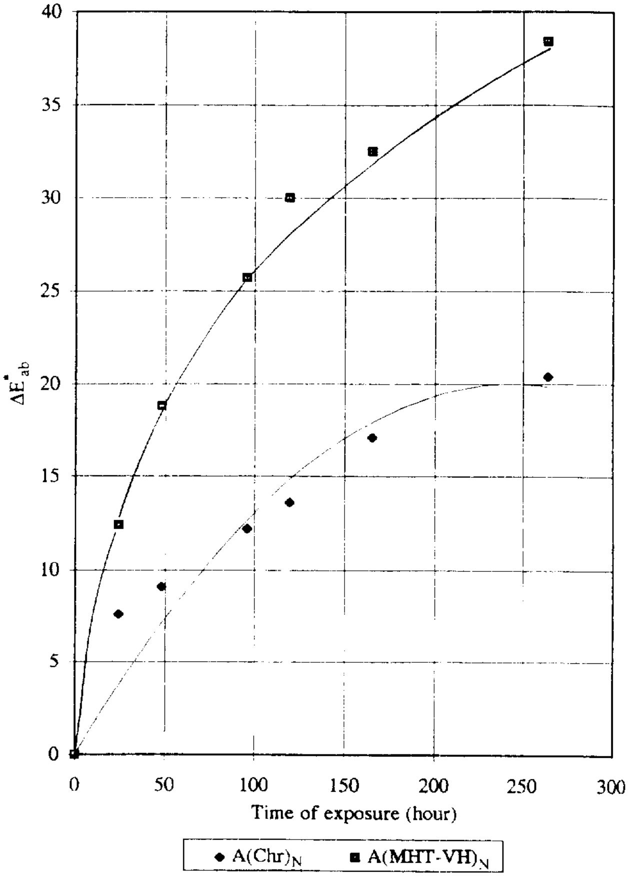


Fig. 3. Kinetics of photofading in presence of artificial perspiration of N-dyeings with 'A'-group members not containing reactive groups.

defined, was the difference in the P–L stability between the $B(DCT)_{RC}$ and $B(DCT)_N$ dyeings. The L stability exceeded the P–L stability of the $B(MCT\text{-}VS)_N$ and $B(DCT)_N$ dyeings, (Table 2).

Within the 'B'-group, the P–L stability of the $B(MHT\text{-}VS)_{RC}$ dyeing was the best and that of the $B(Chr)_N$ dyeing the worst.

Comparison between P–L stability of dyeings produced with 'A'- and 'B'- dye group members

Among the P–L stability values of the four hetero-bifunctional RC dyeings; $B(MCT\text{-}VS)_{RC}$ was the best and $A(MCT\text{-}VS)_{Met,RC}$ the worst. The P–L stability value of the three monofunctional 'B'-dyeings was better after exposure for 264 h than that of the three monofunctional 'A'-dyeings.

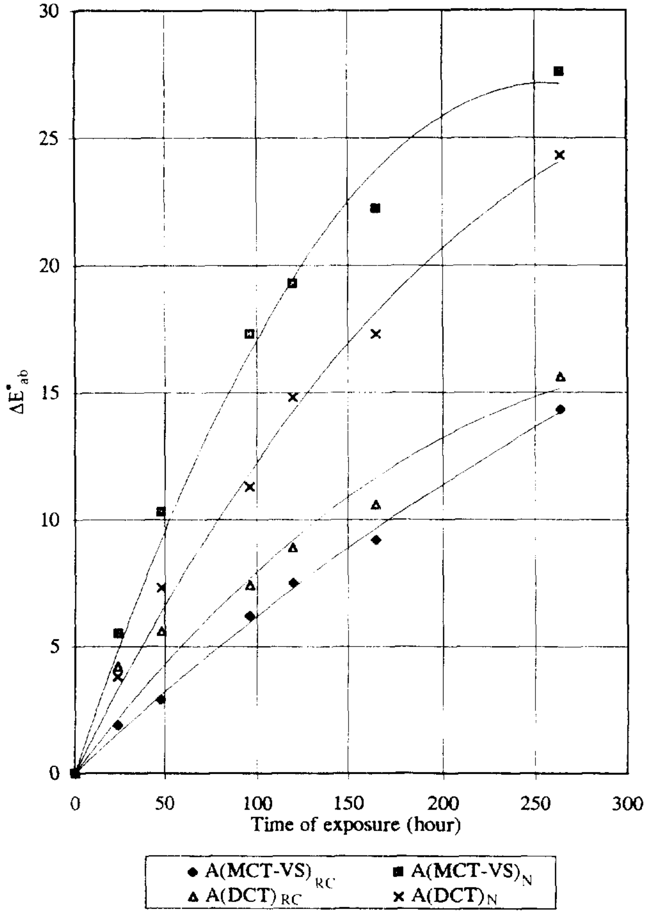


Fig. 4. Comparative kinetics of photofading between RC- and N-dyeings with A(MCT-VS) and A(DCT) dyes, respectively, in presence of artificial perspiration.

The P–L stability of the A(Chr)$_N$ dyeing was better than that of the B(Chr)$_N$ dyeing. Between the poor P–L stability values, that of the B(MHT-VH)$_N$ dyeing was markedly better than that observed on the A(MHT-VH)$_N$ dyeing. N-dyeings produced with A(MCT-VS) or A(DCT) dyes respectively, were less stable to P–L than those produced with B(MCT-VS) and B(DCT) dyes, respectively.

The effect of the presence of the components of perspiration on the light stability of A(MCT-VS)$_{RC}$ and B(MCT-VS)$_{RC}$

The effect of the components of perspiration separately, as well as the action of perspiration with one component not included was studied on the

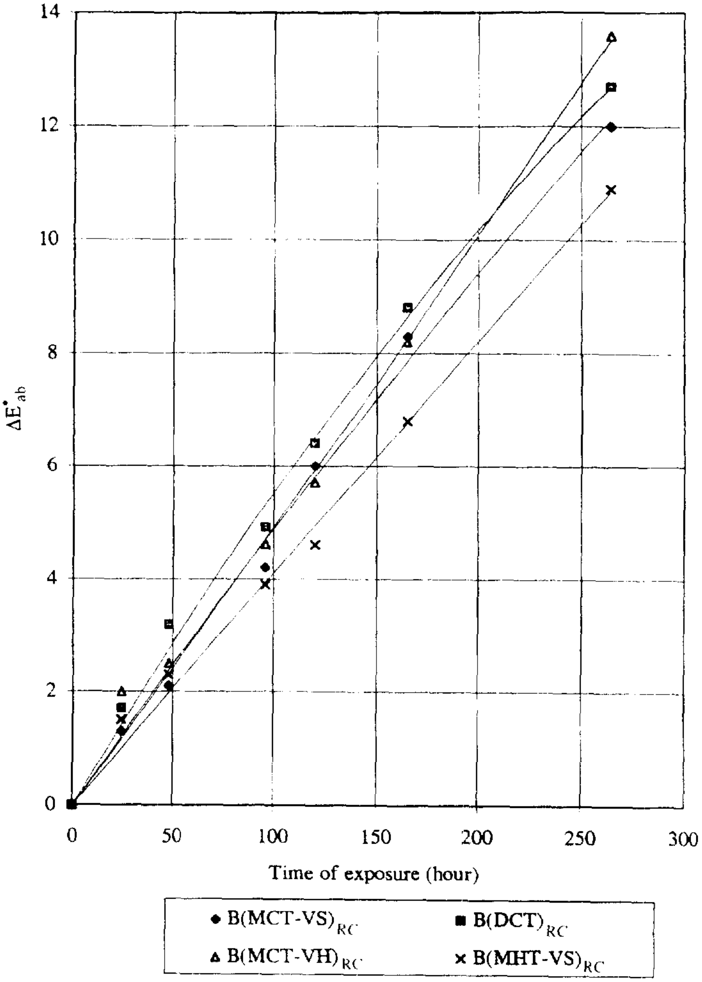


Fig. 5. Kinetics of photofading of one hetero-bifunctional and three monofunctional reactive dyeings in the 'B'-dye group in presence of perspiration.

A(MCT-VS)$_{\mathrm{RC}}$ and B(MCT-VS)$_{\mathrm{RC}}$ dyeings and the results are given in Tables 3 and 4.

A decelerating effect in the rate of photofading was caused by NaCl, Na_2HPO_4*12 H_2O and Na-pantothenate. The effect was more pronounced on the B(MCT-VS)$_{\mathrm{RC}}$ dyeing than on the A(MCT-VS)$_{\mathrm{RC}}$ dyeing. Both dyeings were most sensitive to light in the presence of lactic acid and acetic acid, whereas the light stability deteriorating effect of aspartic acid was only observed on the B(MCT-VS)$_{\mathrm{RC}}$ dyeing. The absence of NaCl, Na_2HPO_4*12 H_2O or Na-pantothenate from the perspiration increased the rate of photofading, whereas the absence of one of the acidic components decreased it.

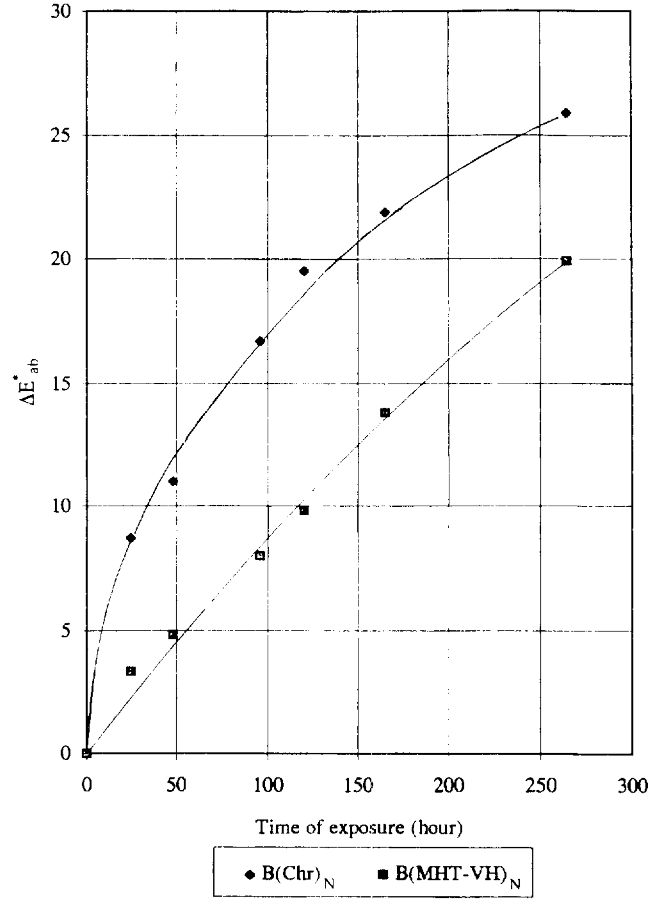


Fig. 6. Kinetics of photofading in presence of artificial perspiration of N-dyeings with 'B'-group members not containing reactive groups.

Generally, it was observed that with the above noted additives the lower the pH of the system the more the rate of photofading increased. A much slower photofading occurred in the pH range between 6–8.

Data for the photofading in open and closed systems

The photofading of the dyeings significantly depends on the presence of humidity, the conditions of exposure and the environmental components. Photofading of dyeings exposed to light in a closed system markedly differed from that obtained in an open system (Table 4).

With respect to the light stability of the $A(MCT\text{-}VS)_{RC}$ dyeing in the closed system, an important role is played by the average moisture content of

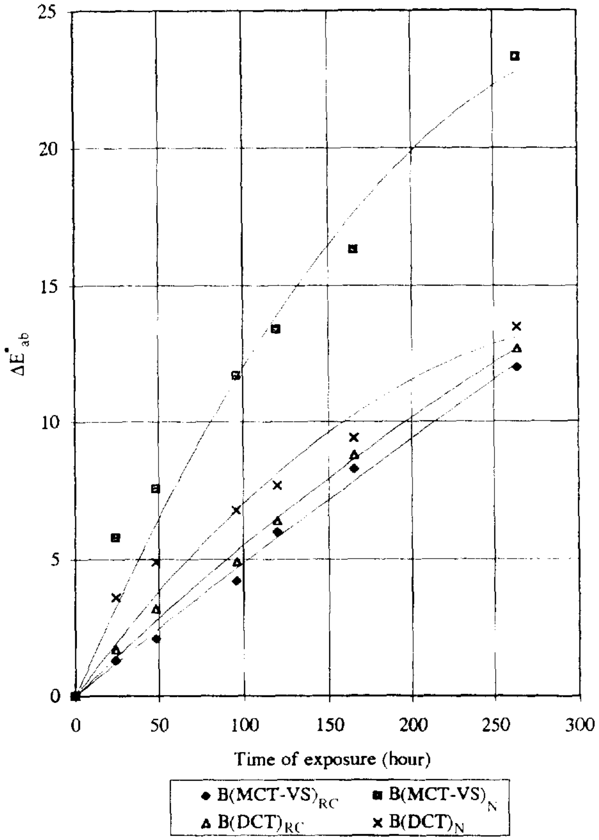


Fig. 7. Comparative kinetics of photofading between RC- and N-dyeings with B(MT-VS) and B(DCT) dyes, respectively, in presence of artificial perspiration.

the dyeing throughout the whole time of the exposure, as well as by the reducing capacity of the substrate and some components of the perspiration, respectively. The oxidative mechanism of fading has also to be taken into consideration, depending on the excess of oxygen in the fading environment. The initially wet dyeing dried out within the first 10 min of exposure in the open system, whereas its moisture content remained constant for the whole period in the closed system. The higher the moisture content of the dyeing, the poorer was its L stability in the closed system.

The role of moisture in the open system is limited to roughly 10 min and the reducing effect of the perspiration components is also markedly reduced by the evaporation of the moisture and by the oxidation of the reducing reagents by the continuously renewing oxygen content of the atmosphere. Photofading after the evaporation of moisture and the consumption of the reducing agents is most probably caused by an oxidative mechanism only.

TABLE 3
Effect of the Perspiration Components and Their Absence from the Complete Perspiration, Respectively, on the Photofading of A(MCT-VS)$_{RC}$ and B(MCT-VS)$_{RC}$ Dyed Systems after Exposure to Light for 165 h.

The component of perspiration	*A(MCT-VS)$_{RC}$*		*B(MCT-VS)$_{RC}$*		*pH*	
	ΔE^*_{ab}				*The component alone*	*Absent from persp.*
	Alone	*Absent from persp.*	*Alone*	*Absent from persp.*		
Lactic acid	7.1	7.7	13.7	6.2	2.48	4.83
NaCl	5.0	11.3	5.8	8.1	6.11	3.73
$Na_2HPO_4*12\ H_2O$	4.7	9.7	5.7	7.5	8.12	3.18
Histidine	6.1	12.5	7.6	7.2	4.18	3.63
Aspartic acid	5.8	9.0	11.5	6.9	3.18	3.75
Panthotenate	5.3	8.9	6.0	7.2	6.45	3.13
Glucose	6.4	10.6	10.9	7.3	5.64	3.71
Acetic acid	7.0	14.6	12.4	7.3	3.50	3.80
Water	6.0	7.5	10.2	6.3	6.55	–

The colour differences of A(MCT-VS)$_{RC}$ dyeing: $\Delta E^*_{ab,\ L} = 5.3$; $\Delta E^*_{ab,\ P\text{–}L} = 9.2$. The colour differences of B(MCT-VS)$_{RC}$ dyeing: $\Delta E^*_{ab,L} = 9.1$; $\Delta E^*_{ab,P\text{–}L} = 8.3$

TABLE 4
The Colour Differences (ΔE^*_{ab}) of A(MCT-VS)$_{RC}$ and B(MCT-VS)$_{RC}$ Dyed Systems after Exposure to Light for 96 h in Closed Air, Oxygen and Nitrogen, Respectively, in Presence and Absence of Artificial Perspiration

Dyed sample	*Control (open air)*		*Closed air*		*Closed oxygen*		*Closed nitrogen*		*Closed air (the sample paded in dist. water)*
	L	P–L	L	P–L	L	P–L	L	P–L	L
A(MCT-VS)$_{RC}$	4.7	7.4	5.8	9.2	10.4	13.3	4.9	14.1	6.3
B(MCT-VS)$_{RC}$	8.9	6.2	9.7	9.8	17.7	17.0	9.7	10.7	9.6

The stoichiometric balance between reducing components and oxygen in the closed atmosphere is decisive with respect to photofading by a reductive or oxidative mechanism, whereas the effect of moisture is continuously separate from the mentioned mechanism.

In the closed system under an oxygen atmosphere, oxygen and moisture are responsible for photofading, whereas in a nitrogen atmosphere reducing impurities play a role besides moisture. The P–L stability of the A(MCT-VS)$_{RC}$ dyeing was markedly lower than the corresponding L stability in all the studied systems.

The improved P–L stability of the B(MCT-VS)$_{RC}$ dyeing against its L stability observed in the open system disappears in a closed moisture and air containing system. Both stabilities deteriorated if the system was closed.

The photofading under both L and P–L exposure was very rapid if the atmosphere was oxygen in the closed system. This observation was true for P–L and L stability of B(MCT-VS)$_{RC}$ and for the L stability of the A(MCT-VS)$_{RC}$ dyeing, generating the idea that the oxidative mechanism is the controlling one if the conditions allow it. As far as P–L stability is concerned, however, the probability of oxidative or reductive mechanism of fading is equal for the A(MCT-VS)$_{RC}$ dyeing.

A previous investigation [11] showed that the existence of the hydrazone tautomeric form of both studied chromophores is preferred against the azo form; this observation is in accordance with the higher sensitivity of the studied dyeings towards oxidation as compared with reductive changes.

CONCLUSIONS

The P–L stability of the dyeing is significantly influenced by the nature of the dye–fibre bonding. The P-L stability of the dye–fibre bond containing dyeings significantly exceeded that of the dyeings in which only secondary forces connected dye molecules with the fibre. Consequently the P–L stability of the reactive dyeing is defined by the sensitivity of the dye–fibre bond towards pH and the particular components of perspiration.

If appropriate reactive groups were present, MCT-type dye–fibre bonds were preferentially formed in 'A'-chromophore containing dyeings, whereas the formation of VS-type covalent bonds were preferred in 'B'-chromophore containing dyeings.

As the MCT-type dye–fibre bonds are easily broken under the effect of acidic pH of perspiration, it is quite understandable that significantly more hydrolysed dye molecules are released under P–L exposure from 'A'-chromophore containing dyeings than from the 'B'-chromophore containing ones, where acid stable VS-type bonds are in majority. This explains why the P–L stability of 'A'-chromophore containing dyeings is significantly poorer than the corresponding L stability, whereas generally the opposite is true for the 'B'-chromophore containing dyeings.

The fact that the P–L stability of B(MHT-VH)$_N$ dyeing is very much better than that of A(MHT-VH)$_N$ dyeing has also to be taken into consideration in explaining the opposite tendency in correlation between the P–L and L stability of the two dye-group systems. The released dye molecules under P–L exposure are generally in the form of A(MHT-VH) and B(MHT-VH), respectively. The significantly smaller amount of released dye molecules in 'B'-group dyeings, together with the less sensitivity of B(MHT-VH) to P–L exposure, are satisfactory explanations for the significant difference in P–L stability between 'A'- and 'B'-type dyeings.

The significant drop in L stability was the consequence of the action of lactic acid in all studied dyeings. The effect of acetic acid approached that of lactic acid in certain cases. Hydrolysis of the dye-fibre bond and simultaneous reductive fading can be attributed to lactic acid. A similar effect can be caused by the acetic acid–glucose binary system.

Salts (NaCl, $Na_2HPO_4*12\ H_2O$ and Na-pantothenate) in perspiration improve L stability in the absence of other components. This effect might be attributed to the aggregation of the released hydrolysed dye molecules, as well as to the reduction of the distance between the released dye molecule and the active site of the fibre. This action is much more effective in 'B'-type dyeings than in 'A'-type ones.

Considering simultaneously the different sensitivities of MCT and VS-type dye–fibre bonds and the different P–L stability of A(MHT-VH) and B(MHT-VH) dye molecules, as well as the different action of salts in perspiration on A(MHT-VH) and B(MHT-VH) molecules, the significant difference between the P–L and L stabilities of 'A'- and 'B-type dyeings can be understood.

The major sensitivity to photooxidation of all the studied 'A'- and 'B'-type dyeings can be attributed to the higher probability of the existence in hydrazone tautomeric form of the dye molecules. In the case of 'B'-type dyeings, equivalent sensitivity toward photoreduction could also be observed.

As long as moisture is present in the dyeings under exposure, hydrolytic processes have also to be taken into consideration, in addition to photooxidation and photoreduction processes.

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