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Synthesis and Properties of Naphthalimide Acid Dyes

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ABSTRACT

*A series of monoazo dyes has been obtained using 4-aminonaphthalimide and 4-amino-*N*-methylnaphthalimide and their 3-sulpho derivatives as diazo components. Derivatives of 1- and 2-naphthol were used as coupling components. It was found that in the electronic spectra of these dyes, additional bathochromically shifted bands of different intensity appear. This effect is caused by the occurrence of the dyes in equilibrium of azo and hydrazone forms.*

The sulpho group in the ortho(3-) position to the azo group influences the dulling of the colour by weakening coupling between the two parts of the dyes. At the same time, it gives better solubility and increases exhaustion and fastness on wool and polyamide.

1 INTRODUCTION

Recent investigations[1,2] have shown that the use of 4-aminonaphthalimides as diazo components in azo dyes has a beneficial influence on the properties of the resultant disperse azo dyes. It was also shown that the Hammett σ value of the azonaphthalimide residue is comparable with that of the 2,4-dinitrophenylazo residue. It is thus possible to obtain dyes of deep and intense colour with these dyes. While evaluating possible methods for the reduction of 4-nitronaphthalimides, using cheaper and more easily accessible reagents like those described in the literature, e.g. stannous chloride,[3] 4-amino-3-sulphonaphthalimide and its *N*-methyl derivatives were obtained in a facile and economical procedure. The sulphonated 4-aminonaphthalimide and 4-amino-*N*-methylnaphthalimide were used in the synthesis of acid dyes and monoazo dyes were also obtained using 4-aminonaphthalimide and 4-amino-*N*-methylnaphthalimide and their 3-sulpho derivatives as diazo components. The following were used as coupling components: 2-naphthol for the synthesis of model dyes and 2-naphthol-6-sulphonic acid (Schaeffer's acid), 2-naphthol-3,6-disulphonic acid (R acid), 1-naphthol-6-amino-3-sulphonic acid (I acid), 1-naphthol-7-amino-3-sulphonic acid (γ acid), and *N*-phenyl-1-naphthylamine-8-sulphonic acid (phenyl-peri acid).

Dyes not containing sulphonic acid groups (**1**, **3**–**A**) were also synthesised in order to obtain comparative models for evaluation of the influence of possible steric hindrance due to the sulpho group on the colour of the dyes.

Dyes were obtained by conventional diazotization of the amines (**1**, **3**–**A**–**F** by direct method) and of sulphonated amines (**2**, **4**–**A**–**F**) by the reverse method.

Coupling compound	*Diazo compound*			
	I	*II*	*III*	*IV*
2-Naphthol	**1A**	**2A**	**3A**	**4A**
2-Naphthol-6-sulphonic acid	**1B**	**2B**	**3B**	**4B**
2-Naphthol-3,6-disulphonic acid	**1C**	**2C**	**3C**	**4C**
1-Naphthol-6-amino-3-sulphonic acid	**1D**	**2D**	**3D**	**4D**
1-Naphthol-7-amino-3-sulphonic acid	**1E**	**2E**	**3E**	**4E**
N-Phenyl-1-naphthylamine-8-sulphonic acid	**1F**	**2F**	**3F**	**4F**

Here **I** is 4-aminonaphthalimide; **II** is 3-sulpho-4-aminonaphthalimide; **III** is 4-amino-*N*-methylnaphthalimide; and **IV** is 3-sulpho-4-amino-*N*-methylnaphthalimide.

2 EXPERIMENTAL

2.1 Typical syntheses

*2.1.1 The direct method (dye **3C**)*[4,5]

4-Amino-*N*-methylnaphthalimide **3** (4·5 g, 0·02 m) was added at 10°C to nitrosylsulphuric acid prepared from sodium nitrite (3 g) and concentrated sulphuric acid (30 ml), followed by dilution with glacial acetic acid (20 ml). Diazotisation was continued for 2 h at 10–15°C and the diazo liquor was added slowly to a solution of **C** (3·3 g, 0·022 m) in 50 ml of water with the addition of 30% NaOH. After stirring for 2 h ($\mathrm{pH} > 7$), the mixture was poured into cold water (400 ml) and the dye was salted out (15% NaCl by volume) and filtered, to give dye **3C** (5·3 g, 89·5% purity).

*2.1.2 The reverse method (dye **2D**)*[6]

4-Amino-3-sulphonaphthalimide **2** (8·2 g, 76·9%) was dissolved in 7·5–10 ml of 30% NaOH and 8–9 ml of 4 M $NaNO_2$. The mixture was cooled to 5°C and poured into a mixture of 7·5–8 ml of 30% HCl and 200 ml of water. After stirring for 2 h at 5–10°C, the diazo liquor was filtered and used in the coupling reaction with **D**, viz: 6·4 g of **D** was dissolved in 6 ml of 30% NaOH and 100 ml of water; the temperature was lowered to 5°C and the diazo liquor was added portionwise. The dye was salted with 15% NaCl by volume, filtered and purified by prolonged washing with 5% NaCl. Dye **2D** was obtained in a yield of 57·7% (pure product).

2.2 Electronic spectra, dyeing and fastness properties

2.2.1 Electronic spectra

Electronic spectra were run in buffer solution at pH 7, at concentration 2×10^{-4}–2×10^{-5} m/dm^3 in DMF for dyes insoluble in water or in 50% DMF/water for dyes weakly soluble in water at concentration 2×10^{-5} m/dm^3, using a Specord M40 (Carl Zeiss-Jena).

2.2.2 Dyeing procedure

Wool and polyamide samples were dyed in a Roaches Engineering machine; samples were dyed for 1 h at 98°C, in 95 ml water, 5 ml buffer solution at pH 5, 2·5 g fabrics and 0·025 g of dye.

2.2.3 Dyeing fastness

The fastness of dyeings were examined according to Polish Standard; the method of these determinations are much the same as those used in other European countries. Light fastness was evaluated on a Xenotest 150S (Hanau).

3 RESULTS AND DISCUSSION

A series of dyes of general formula **1–4-A–F**, containing a sulphonic acid in the diazo or coupling component was synthesised. The dyes had red, claret, violet or blue hues (Table 1).

Literature data[7–12] mention the role of the position of the sulpho group in acid dyes. It has been observed[9,10,13–16] that this can affect the colour of the dye, i.e. position of absorption maximum (λ_{max}), and brightness of shade.

TABLE 1

Purity of Dyes and Colour of Samples of Fabrics Dyed With the Acid Naphthalimide Dyes 1–4 W, wool; P, polyamide)

	1			***2***			***3***			***4***		
	MW	*W*	*P*	*MW*	*W*	*P*	*MW*	*W*	*P*	*MW*	*W*	*P*
A	367 (92·8)[a]	—	PY	469 (87·7)	BrR	VB	381 (51·2)[a]	—	YB	484 (99·0)	BrP	PG
B	469 (67·3)	D	DV	571 (72·9)	DG	V	483 (52·0)	YB	YB	585 (98·0)	BrR	D
C	571 (53·0)	PG	PG	673 (74·9)	BrR	V	585 (89·5)	D	PR	687 (86·2)	PG	PG
D	484 (49·2)	VR	V	586 (57·7)	BR	RB	498 (86·6)	Br	BrR	600 (68·9)	BrR	RBr
E	484 (54·3)	VG	NG	586 (88·6)	GBr	GBr	498 (81·9)	V	N	600 (68·8)	VG	VB
F	544 (59·8)	G	GB	646 (92·5)	Gr	Gr	558 (80·7)	N	BG	660 (77·5)	GrG	GrG

MW, Mass weight. In parentheses are cited purity (in %). All dyes were isolated as Na salts.

Br, Brown; Y, yellow; V, violet; G, Grey; Gr, green; B, blue; N, navy blue; P, pink; R, red; D, deep red.

[a] Dyes separated by adding acid.

TABLE 2

Spectrophotometric Data for the Acid Naphthalimide Dyes and Half-bandwidths of the Absorption Bands

	1 λ_{max}	*2* λ_{max}	*3* λ_{max}	*4* λ_{max}	*1* $\Delta\nu_{1/2} \times 10^{-3}$ $(\Delta\lambda_{1/2})$	*2* $\Delta\nu_{1/2} \times 10^{-3}$ $(\Delta\lambda_{1/2})$	*3* $\Delta\nu_{1/2} \times 10^{-3}$ $(\Delta\nu_{1/2})$	*4* $\Delta\nu_{1/2} \times 10^{-3}$ $(\Delta\lambda_{1/2})$
A	441 (524)[a]	514 (430)	433[a]	524 (359, 450)	7·3 (166)	4·5 (114)	3·9 (71)	3·0 (81)
B	441 (526)[b]	510 (428)	441	511 (377)	7·6 (167)	W	4·0 (75)	W
C	439 (532)[b]	513 (433)	519 (573, 451)	517	4·3 (81)	5·1 (124)	3·4 (94)	4·1 (104)
D	509 (415)	495	517 (413)	495	5·1 (129)	4·4 (104)	12·6 (256)	7·2 (161)
E	535 (430)	527 (454)	535 (407)	528 (460)	W	W	7·0 (185)	W
F	599 (442)[b]	369	357 (599)	392 (437, 519)	9·9 (266)	W	5·3 (187)	W

[a] Spectra run in DMF.

[b] Spectra run in 50% DMF; the remaining spectra run in water at pH 7. Inflexions given in brackets.

W, Wide band with high absorption in the short-wave range of the spectrum, higher than the values $1/2 \times A$ at $\Delta\lambda_{1/2}$.

During these investigations dyes not containing sulphonic groups (**1**, **3**-**A**) were also synthesised. They were treated as models for evaluation of the influence of a sulphonic group in the 3-position on the colour of these compounds; 2-naphthol (**A**) was used as a coupling component.

A range of dyes containing naphthalene residues was thus obtained. Purity of the dyes varied from 49 to 99% (Table 1), as determined by potentiometric titration of chloride ions. Table 2 shows λ_{max} and half-bandwidths $\Delta\lambda_{1/2}$ of the dyes.

The λ_{max} values show the position of the most intense absorption band, which depends on the possibility of the dye existing in the azo or hydrazone forms. In aqueous solutions of pH 7, the naphthol derivatives usually occur in the azo form. The absorption of the hydrazone form is very strongly shifted bathochromically compared to the azo form, by as much as 100 nm, depending on the degree of conjugation in the dye. Half-band widths $\Delta\lambda_{1/2}$ supply further information concerning the structure of the dyes. Dyes **1** and **3**, which do not contain sulpho groups in their diazo compound, usually have sharp maxima; on the other hand, introducing the sulpho group in a position *ortho* to the azo group causes significant widening of the absorption bands, and dulling of the shade. This is due to steric hindrance from the sulphonic group, a factor which has been observed in other groups of azo dyes.[9,10,13,17,18]

In some cases this effect is so strong that a band of intensity comparable to that of the main band appears in the range of 350–360 nm. This probably originates from a naphthalene-azo-naphthalene chromophore, (K band),[16,19–21] similar to the bands of the benzene-azo-naphthalene chromophore.[20–24]

4 SPECTRAL CHARACTERISTICS OF DYES

Spectra were run in DMF for the dyes insoluble in water (**1A**, **3A**), and for derivatives sulphonated in the naphthalene ring (**2A**, **4A**) in water at pH 7 or in 50% DMF, and also in water at pH 10. These spectra enable the position of the bands of the azo and hydrazone forms to be evaluated,[14,22,25–28] the hydrazone bands being particularly apparent in DMF. In the 2-naphthol derivatives, the azo and hydrazone forms exist in equilibrium (dye **1A**).

The relationships considered here are illustrated in Fig. 1. The presence of the *N*-methyl group in the imido system (N—CH_3) of 2-naphthol derivatives results in an increase in the contribution of the azo form. Introducing the sulpho group into the ring of the diazo compound results, as in the derivatives **2A**, **4A**, in an almost exclusive hydrazone form (Fig. 1).

It was observed that, in dyes **1** and **3**, in which R-salt (C) was the coupling component, the hydrazone form prevails; the absorption of this is at

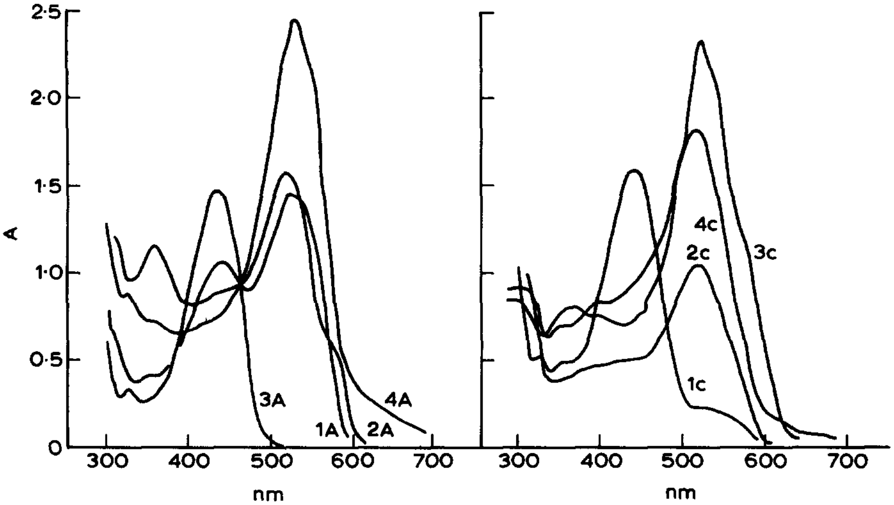
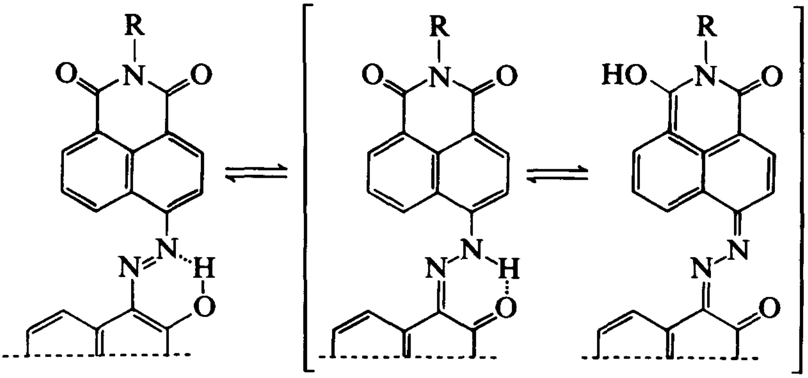


Fig. 1. Typical spectra of dyes **1A** and **3A** in *N*,*N*-dimethylformamide and **2A** and **4A** in water (pH 7) and of dyes **1C**, **2C**, **3C**, **4C** in water (pH 7).

c. 520 nm, and its intensity is lower than that of the azo form in the range 408–443 nm. This is due to the presence of the sulpho group *ortho* to the hydroxyl group in the coupler. In an alkaline environment (pH 10, Table 3), the relative increase of the intensity of the bands at 340–370 nm is characteristic. In some cases this effect is so strong that this band has an intensity comparable to that of the main band. Probably ionisation of the OH and SO_3H groups prevents them from coupling with the azo system by electrostatic interaction, and the system of coupled bonds of naphthalene-azo-naphthalene,[16,19-21] similar to the bonds of the benzene-azo-naphthalene chromophore,[20-24] has the greatest importance.

The dyes which contain the sulpho group in the naphthalimide skeleton (**2**, **4**) possess an additional band in the range of 510–535 nm, which may be caused by the occurrence of the dye in the hydrazone form, e.g. **IIb**, in the



I **IIa** **IIb**

TABLE 3
Spectrophotometric Data for the Acid Naphthalimide Dyes (pH 10)

	1	*2*	*3*	*4*
	$\lambda_{1\,\mathrm{max}}$	$\lambda_{1\,\mathrm{max}}$	$\lambda_{1\ \mathrm{ax}}$	$\lambda_{1\,\mathrm{max}}$
A	—	509 (~413, ~640)	—	356 (427)
B	493	513	357 (440)	358 (413, 514)
C	351 (410, ~515)	442 (502)	352 (452)	513 (~450)
D	526	505 (345, ~644)	536 (410)	512 (~438, ~654)
E	542	340 (557)	535 (407)	346 (570)
F	350 (515, ~640)	358	360 (440, 512, 635)	352 (~396, ~520, 684)

~ Inflexion point. Side bands of intensity lower than that of the main absorption band are given in brackets.

TABLE 4
The Contribution of thd Azo/hydrazone-**II** Form to the Azo-hydrazone Equilibrium of Dyes in Aqueous Solution and in Dyeings on Polyamide (PA) and Wool (W) (in %)

	1			*2*			*3*			*4*		
	H_2O	*PA*	*W*	H_2O	*PA*	*W*	H_2O	*PA*	*W*	H_2O	*PA*	*W*
A	41/0^a	—	—	28/8	33/26	34/26	$_a$100/0^a	—	—	26/0^b	—	—
B	58/0	35/14	32/29	26/0	31/24	30/25	88/0^b	45/20	45/19	43/10	34/27	33/28
C	87/0^b	45/8	34/30	29/5	34/22	31/22	17/29	30/32	29/34	34/0	37/27	29/34
D	25/12	30/33	27/32	28/0	34/12	34/14	27/20	29/35	30/30	37/7	29/29	29/27
E	33/12	33/30	32/28	44/0	37/25	36/24	27/20	26/36	26/31	45/0	29/35	31/31
F	36/9^b	30/33	31/30	91/3	52/21	52/20	34/27	31/34	29/35	77/3	43/27	43/25

[a] DMF solutions.
[b] 50% DMF/H_2O solutions.

formation of which the imido system of the naphthalimide moiety takes part. The form **IIa**, typical for 2-naphthol derivatives, prevails in dyes **2** and **4**, and the azo form **I** begins to prevail in alkaline solutions.

As noted above, the dyes can occur in two different hydrazone forms, viz. either with the imido system (amido-iminol tautomerism **IIb**) or with the azo system (azo-hydrazone tautomerism **IIa**).[1,5]

Change in the position of the azo group from the 1-position into the 2-position creates a bathochromic effect of *c.* 20 nm. Moreover, these dyes (e.g. from coupling components **D** and **E**) occur more often in the hydrazone form than in the azo form. Changes in pH cause a shift of the absorption maxima, without changing the character of the spectra. In a few cases the azo form appears as an inflection point in the shorter wavelength spectral range (e.g. **4D**).

In alkaline environment (pH 10) shift of the absorption bands in relation to those ascribed to the azo form in the neutral environment is mainly the result of ionisation of the hydroxyl group.

For the dyes studied, an approximate determination of the mutual relation between the hydrazone form and the azo one was carried out, in solution and on wool and polyamide fibre (Table 4), according to the formula:[27,29]

$$100 \times \frac{A_x}{A_{IIa} + A_{IIb} + A_I} = \% \text{ quinone hydrazone}$$

where A_x is molar absorbance of any tautomer; A_I is molar absorbance of the azo tautomer; and A_{IIa}, A_{IIb} are the molar absorbances of the respective hydrazone tautomers.

The spectra of some dyes least soluble in water were run in 50% DMF/water. This solvent stabilises the azo form. In an aqueous environment, most of the dyes (1- and 2-naphthol derivatives) occur in the hydrazone form **IIa**, particularly the dyes without the sulpho group in the diazo component (**1**, **3**). Probably the presence of this group makes a planar configuration difficult, as a result of 'blocking' the possibility of free rotation. The weak absorption of the proposed hydrazone form **IIb** can appear in the range *c.* 550–650 nm and its contribution significantly increases on dyed wool and polyamide. Moreover, on fibres, the dyes usually occur as the equilibrium of three forms, viz. azo (**I**) and two hydrazone (**IIa**, **IIb**). In water or 50% DMF solution, the proportion of the **IIb** form is in most cases insignificant. This phenomenon highly affects the purity of the dyeings; the dyeings had dull shades and low chromaticity. The relationships considered here are illustrated in Table 2 and Fig. 2.

The dyes were exposed to standard utility tests, such as wet fastness, fastness to acid and alkaline perspiration, dry and wet rubbing, washing

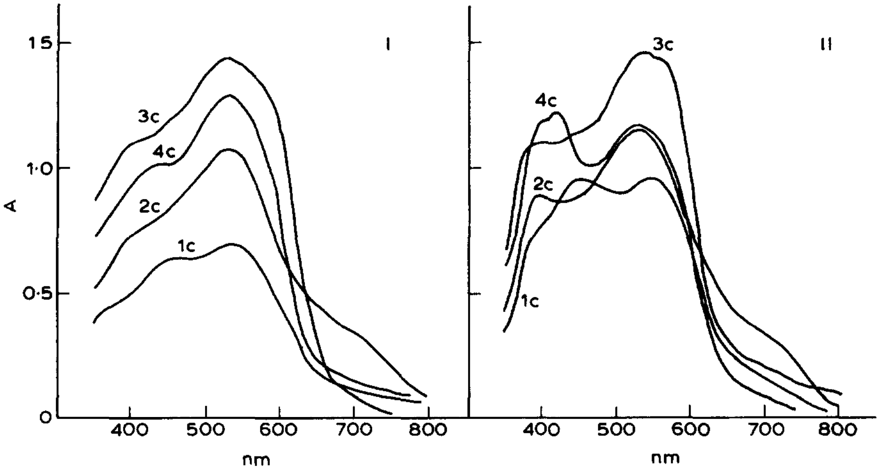


Fig. 2. Reflectance spectra of dyeings of wool (I) and polyamide (II) for dyes **1–4-C**.

(Table 5) and light fastness (Table 6). On the basis of these tests some dependencies characteristic for the particular group of dyes can be observed.

The sulpho derivatives **2** and **4** show better fastness than the dyes synthesised using non-sulphonated naphthalimides, differences being about 0·5–1 units.

Similarly, the type of the dyed fibre has its effect on the changes of fastness. Usually dyeings on polyamide have higher fastness that those on wool.

The ability of dyes to occur in the azo form, i.e. that in which the hydroxyl and the sulpho groups can occur in the anionic form, has its influence on the fastness to alkaline perspiration and washing. It causes lowering of fastness and change in colour of the accompanying sample. This can be confirmed by comparison of the fastness of the dyeings.

Interesting differences can be noticed for the sulpho derivatives (**2**, **4**) and the non-sulphonated (**1**, **3**) naphthalimides, the sulpho derivatives having the higher fastness. This is probably caused by the stronger binding of these dyes with the fibre, resulting from the larger number of group undergoing ionisation and hence the possibility of forming more ionic bonds.

In standard dyeing conditions, dyes **2** and **4** usually give more intense dyeings than the corresponding dyes **1** and **3**, although the sulpho groups could also result in lower dyebath exhaustion due to increase in the solubility of the dyes.

Differences in fastness were also apparent between the imido (**1**) and *N*-methylimido (**3**) derivatives. This may be due to the ability of the imido system to undergo keto-imido tautomerisation. This phenomenon has no equivalent in pigments and in disperse dyes, where the ability of the

TABLE 5
Wet Fastness of Acid Naphthalimide Dyes (Wool/Polyamide)

	1						*2*						*3*						*4*					
	a	*b*	*c*	*d*	*e*	*f*	*a*	*b*	*c*	*d*	*e*	*f*	*a*	*b*	*c*	*d*	*e*	*f*	*a*	*b*	*c*	*d*	*e*	*f*
A							3–4/2	3–4/2	3–4/2	3–4/5	2–3/5	3–4/5												
B	3/2–3	2/2–3	3/2–3	4/4–5	3–4/3–4	4/2–3	4/3–4	4/3	4/3–4	4–5/5	4/4–5	4/5	3/2	3/2	3/2	3/3–4	2–3/3	3/5	3–4/3	3–4/3	3–4/3	3–4/5	2/5	3–4/4–5
C	3–4/3–4	3–4/3–4	3–4/3–4	5/5	4–5/4	4/4	4/4–5	4/4–5	4/4–5	4–5/5	4/4–5	4/5	3–4/3–4	3–4/3	3–4/4	5/5	5/4–5	4–5/5	3–4/4–5	3–4/4	3–4/4–5	4–5/5	4/4–5	3–4/5
D	3–4/4–5	3–4/4–5	3–4/4–5	4–5/5	5/5	4/4	4/4	3–4/2	4/2–3	4–5/5	3–4/5	4–5/5	4/4–5	4/4–5	4/4–5	4/5	3/4–5	4–5/4–5	3–4/3–4	3–4/3	3–4/3–4	3–4/5	3–4/4	3–4/5
E	3–4/4–5	3–4/4	3–4/4	5/4	4/3	3–4/4	3–4/4–5	3–4/4–5	3–4/4–5	4–5/5	4/5	3–4/5	3–4/3	3–4/4	3–4/3	4–5/4–5	2–3/3–4	3–4/3	3–4/4–5	3–4/4–5	3–4/4–5	4–5/4–5	2–3/4	3–4/5
F	3–4/3	3–4/3	3–4/3	4/3	2–3/2	3–4/2–3	4–5/4–5	4/5	4/5	5/5	4–5/5	4/5	3/3–4	3/3	3/3	5/5	5/5	2/2–3	4/4	4/4	4/4–5	5/5	5/5	4–5/4–5

Fastness to dyeings to: a, water; b, alkaline perspiration; c, acid perspiration; d, dry rubbing; e, wet rubbing; f, washing at 40°C.

TABLE 6
Light Fastness of the Acid Naphthalimide Dyes (Wool/Polyamide)

	1	*2*	*3*	*4*
A	—/—	3/2	—/—	3/3
B	3/1–2	2/1–2	2–3/3	3/2
C	3/1–2	3/2	3–4/2	3/2
D	3/2	3/2–3	3–4/2–3	3/2
E	3/2	3/2	3–4/3–4	3/1–2
F	3/3–4	3/2	4/2	3/2

molecules of the dyes to self-associate through hydrogen bonds[30,31] plays an important part.

The sulpho groups in dyes **2** and **4** result in a significant increase in the wet fastness, this being greater for the dyes of series **2** than those of series **4**. These differences are quite considerable. This indicates the stronger bonding of the dye with the fibre and, indirectly, its significant ionisation favoured by the presence of the sulpho group. This conclusion is of a qualitative character only.

Light fastness data of dyeings of dyes **1**–**4** leads to similar conclusions. The fastness are relatively low (**2**–**3**, and in a few cases **4**), although for dyed wool they are 1–2 units higher. This, to a large extent, precludes any wide practical application of the dyes.

5 CONCLUSION

The application of 4-aminonaphthalimide and 4-amino-*N*-methylnaphthalimide derivatives and their 3-sulpho derivatives as diazo components makes it possible to obtain monoazo dyes whose absorption bands are strongly shifted bathochromically. The bathochromic effect is caused by the nature of the diazo component whose Hammett σ constant = 1·07[1] is comparable to that of nitroanilines. It was found that in the spectra of these dyes, additional bathochromically shifted bands of different intensity appear. This effect is caused by the occurrence of the dyes in the hydrazone forms. A number of dyes have their maxima broadened or have the maxima in the form of inflexion points in the longer wavelengths range. Dyes containing sulpho groups (**2** and **4**) show strong absorptions in the shorter wavelength range and have large half-band width because the coupling of the two parts of the dyes is weakened. The scale of this effect depends on the type of the azo compound, but it always adversely affects the shade of the dyeings.

A sulpho group in the *ortho*-position to the azo bond, when in the coupling component, particularly influences the dulling of the colour. Most of the dyes coloured wool and polyamide in shades from red to dark blue and violet. It was also found that some of the dyes showed affinity for cotton to different extents (**1D,E,F**; **2A,D**; **3D**; **4B,D,E**), an observation which might indicate that they would occur, on the substrate, in an associated planar configuration.

Introducing the sulpho group into the 3-position of the diazo component results in better solubility of the dyes and increased exhaustion and fastness on wool and polyamide; this group also causes steric hindrance, and influences the azo-hydrazone equilibrium of the dyes.

REFERENCES

1. Wojciechowski, K., *Dyes & Pigments*, **12** (1990) 273.
2. Wojciechowski, K. & Szadowski, J., *J. Therm. Anal.*, **31** (1986) 297.
3. Niewiadomski, Z., Szadowski, J. & Wojciechowski, K., *Przem. Chem.*, **4** (1990) 161.
4. Yugopolskij, L. M. & Gandelsman, L. M., *Zurn. Obsc. Khim.*, **35** (1965) 1252.
5. Peters, A. T. & Bide, M. J., *Dyes & Pigments*, **7** (1986) 237.
6. Kraska, J. & Blus, K., *Dyes & Pigments*, **5** (1984) 415.
7. Venkataraman, K. (ed.), *The Chemistry of Synt. Dyes, Vol. 1*. Academic Press, New York, 1952, p. 1469.
8. Schirm, E., *J. Pract. Chem.*, **144** (1935) 60.
9. Venkataraman, K. (ed.), *The Chemistry of Synt. Dyes, Vol. 1*. Academic Press, New York, 1952, pp. 427, 515.
10. Brode, W. R. & Ebergarton, H., *J. Org. Chem.*, **5** (1934) 162.
11. Spikmen, A. & Cleg, J. R., *JSDC*, **50** (1934) 318.
12. Lemin, D. R. & Rattee, I. D., *JSDC*, **65** (1949) 217.
13. Wiliamson, B. & Rodebush, W. H., *J. Am. Chem. Soc.*, **63** (1941) 3018.
14. Greenwood, D., Hutchings, M. G. & Lamble, B., *J. Chem. Soc. Perkin, Trans. II*, (1986) 1107.
15. Prejmereanu, I. Culic, C. V. & Ivan, F., *Revista de Chemie*, **34** (1983) 403.
16. Boguslawskaja, M. A., Boguslawskij, B. M. & Jaworskij, B. M., *Zurn. Obsc. Khim.*, **30** (1960) 112.
17. Brode, W. R. & Morris, R. J., *J. Am. Chem. Soc.*, **70** (1948) 2485; *J. Org. Chem.*, **13** (1948) 207.
18. Blout, E. R. & Gofstein, R. M., *J. Am. Chem. Soc.*, **67** (1945) 15.
19. Badger, G. M. & Buttery, R. G., *J. Chem. Soc.* (1953) 2156.
20. Skulski, L. & Urbanski, T., *Bull. Acad. Polon. Sci., Ser. Sci. Chim.*, **10** (1960) 561.
21. Skulski, L. & Urbanski, T., *Rocz. Chem.*, **36** (1962) 801.
22. Skulski, L. & Kleps, J., *Pol. J. Chem.*, **55** (1981) 1809.
23. Pierekalin, N. W. & Sawostianowa, M. W., *Zurn. Org. Khim.*, **21** (1951) 1329.
24. Wojciechowski, K., *Dyes & Pigments*, **9** (1988) 401.
25. Stiepanow, B. I., Trawien, W. F., Wojnowa, S. E. & Czalych, G. P., *Zurn. Org. Khim.*, **17** (1981) 1291.

26. Ball, P. & Nicholls, C. H., *Dyes & Pigments*, **3** (1982) 5.
27. Fabian, J. & Hartmann, H., In *Light Absorption of Organic Colorants*. Springer Verlag, Berlin, 1980, pp. 68–73.
28. Kelemen, J., *Dyes & Pigments*, **2** (1981) 73.
29. Griffiths, J., Roozpeikar, B. & Thomasson, J., *J. Chem. Research (S)*, (1981) 302.
30. Szadowski, J., *Zesz. Nauk. PL.*, (1981) 11.
31. Szadowski, J., Malinowski, W. & Wojciechowski, K., *Przem. Chem.*, **57** (1978) 70.