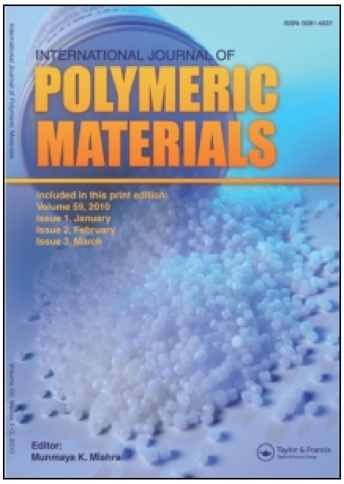

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Synthesis and Characterization of Monomeric and Polymeric Disperse Dyes for Hydrophobic Fibres

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Some monomeric disperse dyes based on 2-amino-5-mercapto-1,3,4-thiadiazole have been synthesized by coupling with derivatives of *N*-phenylacrylamide. Polymeric dyes have been prepared by free radical polymerization of monomeric dyes and were characterized by IR spectra, Intrinsic viscosity, molecular weight by Vapour Pressure Osmometry, thermogravimetry analysis, bleeding test and elemental analysis. The monomeric dyes have been also characterized by IR spectral studies and elemental analysis. The purity of dyes was checked by thin layer chromatography. All the dyes were applied as disperse dyes on nylon and polyester fibres. These dyes have been found to give a wide range of colour shades with very good depth and levelness on each fibre. The percentage dye bath exhaustion on different fibres have been found to be good to very good. The dyed fibres showed good to excellent fastness to light on nylon and polyester fibres. The washing, rubbing, perspiration and sublimation fastness properties of the dyed fibres has been found to be very good to excellent.

Keywords: 2-amino-5-mercapto-1,3,4-thiadiazole; Disperse dyes; Thermogravimetric analysis (TGA); Vapour Pressure Osmometry (VPO); Infra-red (IR) spectra; Intrinsic viscosity; Monomeric; Polymeric; Free radial polymerization; Nylon fibre; Polyester fibre

INTRODUCTION

Heterocyclic diazo components, especially the derivatives of 2-aminothiazole, 2-amino-thiophene and 2-amino-1,3,4-thiadiazole have

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[*] Corresponding author.

played a significant role in the technology of disperse dyes. They have been studied owing to the marked bathochromic effect when compared with the corresponding benzenoid compounds [1]. In recent years much interest has been shown in the disperse dyes derived from 2-amino-1,3,4-thiadiazole [2–4]. These disperse dyes are of relatively small molecular size with very low water solubility and possess substantivity for hydrophobic fibres. The small molecular size leads to better dyeability. These dyes show excellent brightness, intensity and fastness properties compared to their benzenoid counterparts and are commercially competitive with anthraquinone dyes.

Monomeric dye is a polymerizable dye which contains a polymerizable group, normally ethylene type double bond present in the dye molecule. Polymeric dyes may be defined through their applications as polymers and dyes, which are markedly dependent on their relative high molar mass. As such, they are soluble in the medium in which they are applied or in the substrate with which they form a composite. Thus, polymeric dyes are useful polymers or oligomers as well as useful dyes of unique importance of their high tinctorial power [5]. Their utilization has been possible because of certain essential dye properties such as fast leveling on fibres, fastness to light and wet treatment, low sublimation rate and very good thermal and chemical resistance.

In the field of dyestuff chemistry, there has been a constant research for new classes of dyes which have good fastness properties. The colour fastness of the coloured textile is related to the chemical structure, molecular size of dye molecule and physical characteristics of the fibre itself. Giles [6] has given some very useful information on the relationships between dye structure, fibre structure and external conditions and behaviour of the coloured fibres on exposure to light, heat, gaseous impurities and wash. Another important point is that most of the organic dyes are carcinogenic and more or less toxic. Increasing the size of the dye molecules [7] may improve the fastness properties as well as reduce the toxicity. The molecular size of the dye molecule can be increased by polymerization reaction. This reaction has been made use of in the synthesis of polymeric dyes.

A literature survey reveals that in almost all the reported polymerizable monomeric and polymeric dyes, the polymerizable group used are the vinyl type like acrylamide ($-NHCOCH{=}CH_2$) or methacrylamide ($-NHCOC(CH_3){=}CH_2$) [8–10]. The wide use of these groups in the synthesis may be due to their ease of preparation and polymerization. This type of the dyes are mostly based on anthraquinone derivatives and its copolymerization with various monomers also been reported [11, 12]. It seems that no report is available on the use of 2-amino-1,3,4-thiadiazole in the synthesis of disperse azo dyes by coupling with various *N*-phenylacrylamides. Hence, it was thought worthwhile to synthesize some monomeric dyes from 2-amino-5-mercapto-1,3,4-thiadiazole using various substituted *N*-phenylacrylamides as coupling components. The resulting monomeric dyes were polymerized by free radical addition polymerization and their dyeing performance on nylon and polyester fibres has been studied.

EXPERIMENTAL

Materials

All the chemicals used were of commercial grade. They were further purified by crystallization.

Procedures

Synthesis of 2-amino-5-mercapto-1,3,4-thiadiazole (I)

The title compound was synthesized by the method reported in literature [13].

Synthesis of Various N-phenylacrylamides (III_{a-f})

These compounds were synthesized by the method reported in literature [14, 15].

Preparation of 2-(p-N-phenylacrylamido)-azo-5-mercapto-1,3,4-thiadiazole (IV_a)

Synthesis of title compound comprise two steps

(a) Diazotization of 2-amino-5-mercapto-1,3,4-thiadiazole.
98% H_2SO_4 (1.0 g, 0.01 mol) was cooled to 0°C and then dry sodium nitrite (0.69 g, 0.01 mol) was added to it at 5°C. The

reaction mixture was heated to 45°C for 15 minutes and then the temperature was raised to 60–65°C and maintained at this temperature for 1 hour. The resulting solution was cooled to 0–5°C and then treated dropwise at 5–20°C with a mixture of propionic acid and acetic acid (20 ml, 3 : 17). The resulting nitrosyl sulfuric acid mixture was cooled to 0°C and compound **I** (1.33 g, 0.01 mol) was added portiowise and stirred for 2 hours at 0–5°C. The excess nitrous acid (the presence tested by starch-iodide paper) was decomposed with the required amount of urea. The clear diazonium salt solution (**II**) thus obtained was used for subsequent coupling reaction.

(b) Coupling of diazonium salt solution with *N*-phenylacrylamide (**III$_a$**).

N-phenylacrylamide (**III$_a$**; 1.47 g, 0.01 mol) was dissolved in acetic acid (10 ml) and cooled at 0°C. Freshly prepared diazonium salt solution (**II**) was added dropwise to the well-stirred solution over a period of 2 hours, keeping the temperature below 5°C. The reaction mixture was stirred for 45 minutes at 0–5°C, maintaining the pH at 4.0–4.5 by the help of 10% sodium acetate solution. The reaction mixture was then diluted with water (100 ml), the solids filtered, washed with water and dried in an oven at 50°C. The product (**IV$_a$**) thus obtained was crystallized from DMF, yield 80%, m.p. 174°C. The purity of the product was checked by TLC using ethylacetate–benzene (1 : 4) as solvent system. The same procedure was used for the preparation of compounds **IV$_{b-f}$** using various substituted *N*-phenylacrylamides (**III$_{b-f}$**). The whole process for synthesis is drawn in the Scheme I. The characterization data of **IV$_{a-f}$** are given in Table I.

Free Radical Addition Polymerization

2-(*p*-*N*-phenylacrylamido)-azo-5-mercapto-1,3,4-thiadiazole (**IV$_a$**, 0.291 g, 0.001 mol) and 0.006 g (2%) benzoyl peroxide were dissolved in dry DMF (50 ml). The reaction mixture was heated in an oil bath at reflux temperature for 6 hours with occasional stirring. After this period, the reaction mixture was concentrated and the product was isolated by precipitation using petroleum ether (60–80°C) as the non-solvent. The solid product **V$_a$** was separated and filtered and

TABLE I Characterization data and percentage exhaustion of monomeric dyes (IV_{a-f})

Dye No. IV	Coupling component (R)	Yield (%)	m.p.[a] (°C)	Molecular formula	Mol. Wt.	λ max[b] (nm)	log ε	R_f value[c]	Elemental analysis (%) Found (Calc.) C	H	N	Exhaustion (%) Nylon	Polyester
a	*N*-Phenylacrylamide	80	174	$C_{11}H_9N_5OS_2$	291	445	4.65	0.83	45.10 (45.36)	2.92 (3.09)	23.92 (24.05)	84	80
b	*N*-*m*-Nitrophenylacrylamide	85	209	$C_{11}H_8N_6O_3S_2$	336	430	4.42	0.91	39.00 (39.28)	2.18 (2.38)	24.86 (25.00)	84	76
c	*N*-*m*-Chlorophenylacrylamide	75	232	$C_{11}H_8N_5OS_2Cl$	325.5	450	4.44	0.90	40.15 (40.55)	2.30 (2.45)	21.36 (21.50)	82	80
d	*N*-*m*-Tolylacrylamide	79	249	$C_{12}H_{11}N_5OS_2$	305	435	4.58	0.81	47.10 (47.21)	3.41 (3.60)	22.79 (22.95)	82	76
e	*N*-*o*-Tolylacrylamide	68	240	$C_{12}H_{11}N_5OS_2$	305	440	4.65	0.85	47.06 (47.21)	3.46 (3.60)	22.85 (22.95)	86	74
f	*o*-*N*-Acryloylaminbenzoic acid	65	248	$C_{12}H_9N_5O_3S_2$	335	430	4.35	0.86	42.68 (42.98)	2.46 (2.68)	20.60 (20.89)	78	–

[a] Melting points carried out by open capillary method and are uncorrected.
[b] Determine in DMF.
[c] Measure in ethylacetate – benzene (1 : 4).

TABLE II Characterization data and percentage exhaustion of polymeric dyes (V_{a-f})

Dye No. V	*Various poly-N-phenylacrylamide* (R')	*Yield* (%)	*Mol. Wt.*[a]	*Intrinsic viscosity*[b] $[\eta]$, (*dL/g*)	λ *max* (*nm*)	*log* ε	*Elemental analysis* (%) *Found* (*Calc.*) *C*	*H*	*N*	*Exhaustion* (%) *Nylon*	*Polyester*
a′	Poly-*N*-phenylacrylamide	82	5.6×10^3	0.082	440	3.89	45.10 (45.20)	2.95 (3.08)	23.80 (23.97)	76	72
b′	Poly-*N*-*m*-nitrophenylmaleimide	75	5.0×10^3	0.036	425	3.62	39.00 (39.16)	2.25 (2.37)	24.82 (24.92)	80	74
c′	Poly-*N*-*m*-chlorophenylacrylamide	80	5.9×10^3	0.058	422	3.96	40.18 (40.42)	2.32 (2.45)	21.00 (21.43)	75	75
d′	Poly-*N*-*m*-tolylacrylamide	85	4.3×10^3	0.038	420	3.38	46.91 (47.05)	3.46 (3.59)	22.60 (22.87)	76	70
e′	Poly-*o*-*N*-tolylacrylamide	87	4.5×10^3	0.039	425	3.42	46.89 (47.05)	3.50 (3.59)	22.65 (22.87)	78	71
f′	Poly-*o*-*N*-acryloylaminobenzoic acid	85	4.7×10^3	0.065	420	3.78	42.71 (42.85)	2.59 (2.67)	20.70 (20.83)	75	–

[a] Determine in dioxane.

[b] Determine in DMF.

then washed several times with petroleum ether and dried in vaccum at 70°C. The yield was 82%. The same procedure was used for the preparation of compounds $V_{b'-f'}$. The whole process for synthesis is drawn in the Scheme II. The characterization data of $V_{a'-f'}$ are given in Table II.

MEASUREMENTS

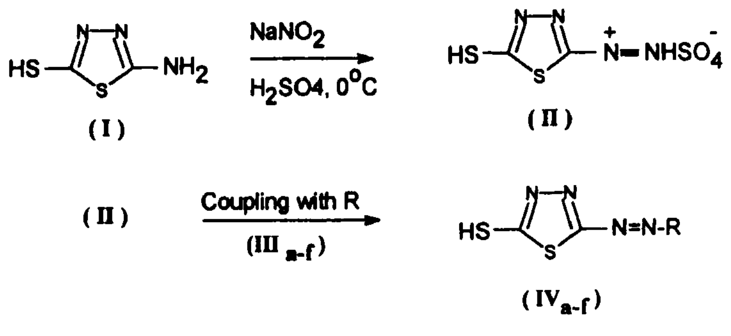
Visible spectra were recorded on CARLZEISS UV/VIS SPECORD Spectrophotometer. A Perkin Elmer model 983 recording infrared spectrophotometer was used for infrared spectra of the compounds in KBr pellets. The C, H, N contents were estimated by means of Carlo ErBa C, H, N, S, P elemental analyzer (Italy). All the polymeric dyes ($V_{a'-f'}$) were characterized by intrinsic viscosity measurement in DMF at 35 ± 0.05°C with a Ubbelohde Viscometer. The molecular weight determination was made in dioxane at 50°C using vapour pressure osmometer, Hewlett Packard model 3028, thermogravimetry analysis (TGA) has been carried out using Du Pont model 950 thermogravimetric analyzer in air at a heating rate of $10\,\mathrm{Kmin}^{-1}$. The fastness to light, sublimation and perspiration were assessed in accordance with BS: 1006-1978. The rubbing fastness test was carried out using crockmeter (Atlas) in accordance with AATCC-1961 and the wash fastness test in accordance with IS: 765-1979.

DYEING METHOD

The dyeing of nylon and polyester fibres was carried out by the method reported in our earlier communication [16].

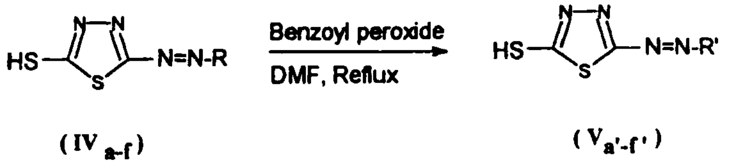
RESULTS AND DISCUSSION

The reaction Schemes I and II represent the synthesis of monomeric and polymeric dyes respectively.



Where R is N-phenylacrylamide(III_{a-f}) coupling components

SCHEME I



Where R' is poly-N-phenylacrylamide moiety

SCHEME II

The typical IR spectra and TG curves of selected dyes are shown in Figures 1 and 2 respectively.

The IR spectra of dyes IV_{a-f} showed characteristic bands around 2960-3010, 1580-1610 and 1500 cm^{-1}. This is due to the aromatic stretching vibrations of phenyl ring. The bands at $600-660_s$ cm^{-1} and $675-700_s$ cm^{-1} (—C—S—H stretching of mercapto group), 1070_w (—C—N stretching), 1200_s (—NHCOCH$=$CH$_2$ group) and 1435 cm^{-1} (—S—C stretching). The bands at 800-870 cm^{-1} (para disubstituted phenyl ring), 960-990 cm^{-1}, $870-900_s$ cm^{-1} are due to the out-of-plane C—H bending of vinyl group. These bands disappear in $V_{a'-f'}$. The azo group (—N$=$N) is confirmed at 1540-1560 cm^{-1}. Two strong bands appear around 1650-1690 cm^{-1} due to the C$=$O group. The amide grouping N—H stretching vibration observe around 3275-3350 cm^{-1}

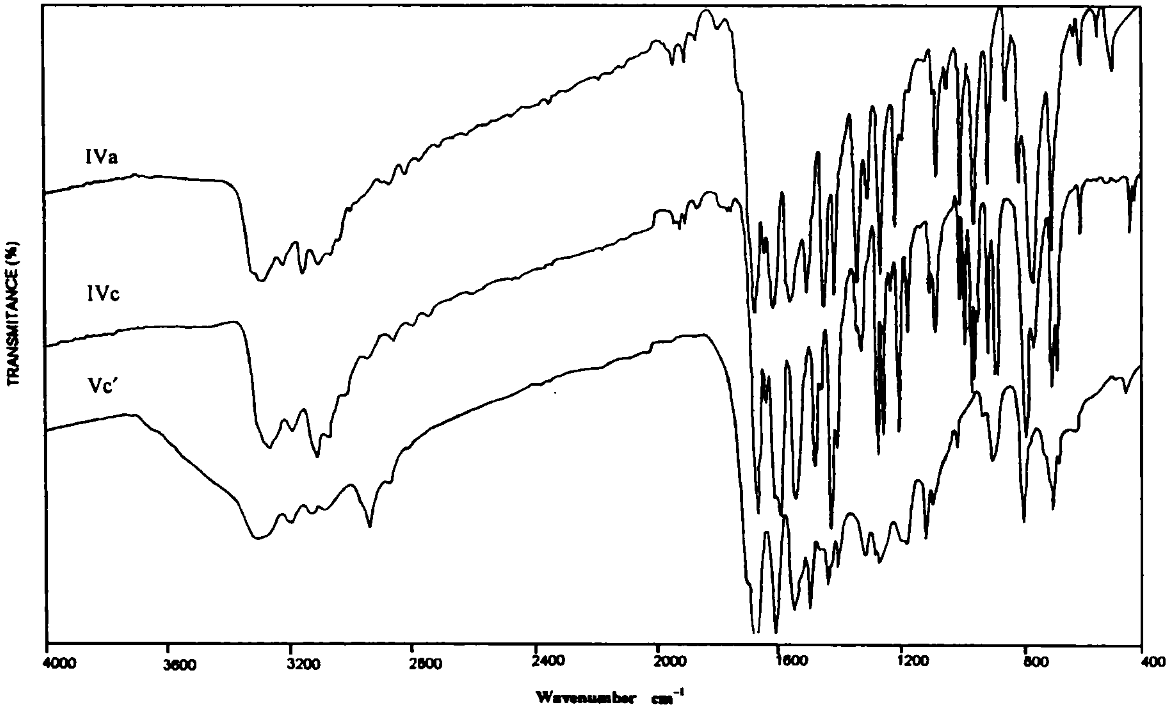


FIGURE 1 IR spectra of dyes IV_a, IV_c, $V_{c'}$.

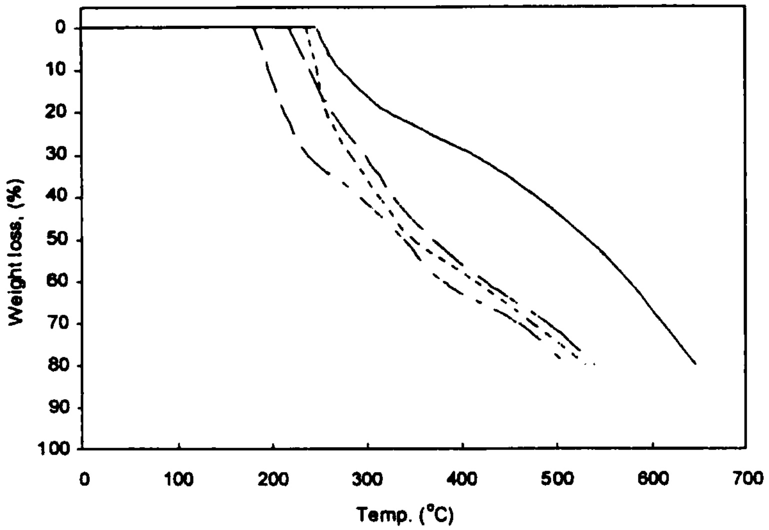


FIGURE 2 TG thermogram of polymeric dyes: $V_{a'}$ (— —), $V_{b'}$ (———), $V_{c'}$ (- - - -), $V_{d'}$ (—— —).

The IR spectra of some of the dyes also showed the following characteristic bands:

IV_b, $V_{b'}$: 1320–1350 cm^{-1} (symmetric stretching due to —NO_2 group)
IV_c, $V_{c'}$: 620–685 cm^{-1} (C—Cl group)
IV_d, IV_e, $V_{d'}$, $V_{e'}$: 1450 and 1385 cm^{-1} (C—CH_3 group)
IV_f, $V_{f'}$: broad peak around 3100–3400 cm^{-1} (inter- and intramolecular H-bonding due to —COOH and —NH group).

Tables I and II show that the exhaustion of the monomeric dyes IV_{a-f} on nylon and polyester fibres is good to very good while its corresponding polymeric dyes $V_{a'-f'}$ show a little less exhaustion on fibres. This may be due to larger molecular size and enhanced interaction taking place between dye molecules and fibre. Hence the rate of diffusion of the dye molecules into fibre decreases, which affect the exhaustion value. The higher exhaustion on the nylon fibre may be expected due to the relatively open structure of nylon fibre. The data also reveal that the change in absorption value in the monomeric and polymeric dyes is due to both the nature and the position of substituent present in the acrylamide ring system. The polymeric dyes show a hypsochromic shift, compared to their corresponding monomeric dyes. This may be due to elimination of the double bond of acrylamide group.

The data of intrinsic viscosity and number average molecular weight of polymeric dyes are listed in Table II. The data indicate that these polymeric dyes are of low molecular weight ranging from 4.3×10^3 to 5.9×10^3. The R_f values of monomeric dyes $\mathbf{IV_{a-f}}$ were measured in ethylacetate-benzene (1:4) (see Tab. I). The high R_f values indicate that there were strong interactions between solvent and dye molecules. Thermogravimetric investigations were carried out in the range from 25°C to 650°C, at heating rate of $10\,\mathrm{K\,min^{-1}}$ in a static air atmosphere. In order to determine the thermal stability trend, TG parameters such as T_0 (temperature of onset of decomposition), T_{10} (temperature for 10% weight loss) and T_{80} (temperature for 80% weight loss) were calculated and shown in Table V. T_0 and T_{10} are the two main criteria for the thermal stability of the polymeric dyes. The higher the values of T_0 and T_{10}, the higher will be the thermal stability [17]. The data given in Table V indicate that the polymeric dyes have good thermal stability. Dye $\mathbf{V_{b'}}$ has better thermal stability compared to other dyes.

All the dyes were applied as disperse dyes on nylon and polyester fibre at 2% shade. The dyed fibres gave attractive colour shade varying from golden yellow to maroom with good levelness, brightness and depth on both the fibres (Tabs. III and IV). The variation in the shades of dyed fibres are due to the nature and position of the various substituent present on the acrylamide ring. The fastness properties of the dyed fibres are shown in Tables III and IV. The light fastness of monomeric dyes $\mathbf{IV_{a-f}}$ ranges from good to very good for nylon and polyester fibres, while its corresponding polymeric dyes $\mathbf{V_{a'-f'}}$ in Table IV show excellent light, washing, rubbing, perspiration and sublimation fastness properties.

Table VI shows the results of the bleeding test. The dyed fibres of polymeric dyes $\mathbf{V_{a'-f'}}$ have excellent fastness to water, very good to excellent fastness to ethanol, acetone and *n*-butanol-xylene (1:9), and fair to good fastness to cellosolve solvent.

Dyes $\mathbf{IV_f}$ and $\mathbf{V_{f'}}$ contain acid groups. Hence these dyes are not exhausted on polyester fibre.

TABLE III Dyeing properties of monomeric dyes (IV_{a-f})

			Fastness to													
							Rubbing				Perspiration					
Dye No.	Shade on		Light		Washing		N		P		N		P		Sublimation	
IV	N	P	N	P	N	P	Dry	Wet	Dry	Wet	Acid	Alkaline	Acid	Alkaline	N	P
a	Dark brown	Light orange	4–5	4–5	5	5	5	5–4	5–4	5	5	5	5	5	5	5
b	Deep brown	Brown	5–4	3–4	5	5	5	5	5	5–4	5	5	5	5	5	5
c	Brown	Reddish pink	3–4	4–3	5	5	5	5–4	5	5	5	5	4–5	5	5	4–5
d	Dark brown	Bright reddish pink	3–4	4–3	5	5	5	5	5	5	5	5	4–5	5	5	5
e	Brown	Reddish brown	4–5	3–4	5	4–5	5	5	5	5	5	5	5–4	5	5	5
f	Reddish brown	–	3–4	–	5	–	5–4	5	–	–	5	5	–	–	5	–

P: Polyester and N: Nylon.

TABLE IV Dyeing properties of polymeric dyes ($V_{a'-f'}$)

Dye No.	*Shade on*		*Fastness to*													
			Light		*Washing*		*Rubbing*				*Perspiration*				*Sublimation*	
							N		*P*		*N*		*P*			
V	*N*	*P*	*N*	*P*	*N*	*P*	*Dry*	*Wet*	*Dry*	*Wet*	*Acid*	*Alkaline*	*Acid*	*Alkaline*	*N*	*P*
a′	Deep orange	Light orange	5	5	5	5	5	5	5	5	5	5	5	5	5	5
b′	Golden yellow	Light pink	5	5	5	5	5	5	5	5	5	5	5	5	5	5
c′	Yellowish orange	Light orange	5	5	5	5	5	5	5	5	5	5	5	5	5	5
d′	Brown	Reddish pink	5	5	5	5	5	5	5	5	5	5	5	5	5	5
e′	Dark brown	Maroom	5	5	5	5	5	5	5	5	5	5	5	5	5	5
f′	Deep brown	–	5	–	5	–	5	5	–	–	5	5	–	–	5	–

P: Polyester and N: Nylon.

TABLE V Temperature characteristics for thermal decomposition of polymeric dyes ($V_{a'-f'}$)% weight loss at °C from TGA

Dye No. V	T_0 °C	T_{10} °C	T_{20} °C	T_{30} °C	T_{40} °C	T_{50} °C	T_{60} °C	T_{70} °C	T_{80} °C
a'	220	242	265	300	330	370	425	490	545
b'	250	245	325	415	480	533	575	610	645
c'	240	220	245	270	315	350	415	475	530
d'	185	198	215	240	295	340	380	460	510
e'	180	200	228	255	298	335	374	464	508
f'	210	220	232	247	282	325	360	458	500

T_0 – Temperature (°C) of onset decomposition.
T_{10} – Temperature (°C) for 10% weight loss.
T_{80} – Temperature (°C) for 80% weight loss.

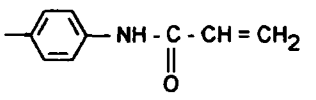
TABLE VI Bleeding properties of polymeric dyes ($V_{a'-f'}$)

Dye No. V	*Water*	*Ethanol*	*Acetone*	*n-butanol-xylene* (1 : 9)	*Cellosolve*
a'	5	5–4	4–5	5	3–2
b'	5	4–5	4–5	5	3–2
c'	5	5	5	5	3–4
d'	5	4–5	5–4	5–4	3–2
e'	5	4–5	4–5	5–4	3–2
f'	5	4	4–5	4–5	2–3

TABLE : A

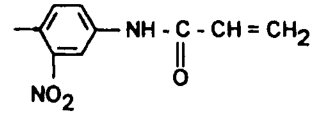
R

a



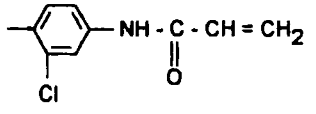
N-Phenylacrylamide

b



N-m-Nitro phenylacrylamide

c

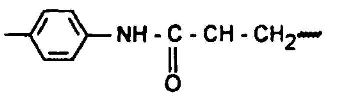


N-m-Chloro phenylacrylamide

TABLE : B

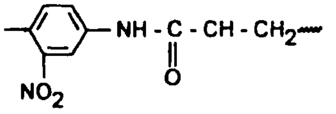
R'

a'



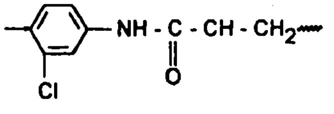
Poly-N-phenylacrylamide

b'



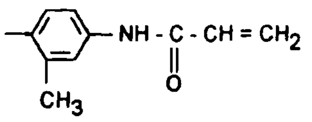
Poly-N-m-nitro phenylacrylamide

c'



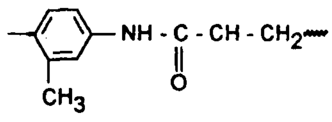
Poly-N-m-Chloro phenylacrylamide

d



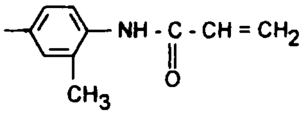
N-m-Tolyl acrylamide

d'



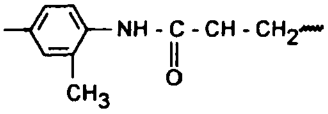
Poly-N-m-Tolyl acrylamide

e



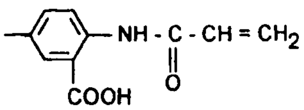
N-o-Tolyl acrylamide

e'



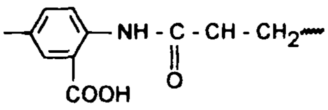
Poly-N-o-Tolyl acrylamide

f



o-N-Acryloyl amino benzoic acid

f'



Poly-o-N-Acryloyl amino benzoic acid

CONCLUSIONS

All the monomeric and polymeric dyes were obtained in good yield. The polymeric dyes having low molecular weight containing *N*-phenylacrylamide moiety. It can be concluded from the above studies that the improvement of the fastness properties with increasing the molecular size of the dye molecule by polymerization reaction leads to brilliancy of shade and excellent fastness properties for both the fibres.

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