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SYNTHESIS, CHARACTERISATION AND DYEING PROPERTIES OF NEW BIFUNCTIONAL DICHLORO-S-TRIAZINYL (DCT) AZO REACTIVE DYES BASED ON 1, 4-DIPHENYLENEDIAMINE

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ABSTRACT

Five novel homo bifunctional dichloro-s-triazinyl (DCT) azo reactive dyes containing 1,4-diphenylenediamine as a tetrazotised component was prepared by coupling to 4-nitroanilino cyanurated acids (H-acid, J-acid, Laurent acid, Tobias acid and Gamma acid). The structures of the synthesised dyes were characterised and confirmed by melting point, UV-visible spectroscopy and FT-IR spectroscopy. The synthesised dyes were applied onto cotton and nylon 6,6 fabrics under typical exhaustion processes and their dyeing properties were evaluated. The percentage exhaustion and percentage fixation of the synthesised dyes on cotton and nylon 6, 6 fabrics were very good. All the synthesised dyes gave good form to excellent properties to washing, light and perspiration, respectively.

Keywords: *bifunctional azo reactive dyes, cotton fabrics, Dyeing, Reactive dyes, Nylon 6,6 fabrics*

INTRODUCTION

Cid *et al*; (2007), Ali *et al*; (2014) and Blanco *et al*; (2016) reported that reactive dyes are textile organic colourants having one or two reactive groups capable of reacting covalently between carbon and phosphorus atom of the dyes and oxygen, nitrogen or sulphur atoms of the fibre. Alan *et al*; (2000), Patel *et al*; (2010) and Almasian *et al*; (2015) opined that reactive dyeing systems have become the most important discovery in the colouration industry in the last century. Ever since they were discovered, diminishing era for the demand of other dye classes set in (Al-Degs *et al*; 2008, Christie, 2001; Bravo-Diaz, 2010). Reactive dyes constitute a very important class for dyeing cellulosic and polyamide fibres and a very high rate of growth is predicted in future (Ahmed, 2005, Broadbent, 2001, Clark, 2011, Patel and Kesshav, 2012).

The earlier dyes synthesised were however, fixed onto the cellulose by an alkaline treatment at times at higher temperatures. Hence, as a result of this the earlier dyes form esters and ethers with cellulose (Anderson and Whitcomb, 1994, Alen *et al*; 1997, Clark, 2002). These dyes were found to have the following setbacks:-lots of dyes wasted due to hydrolysis, could not be applied to cotton/polyester blends as the disperse dye meant for the polyester component decomposes at high temperature of application and due to the high alkalinity required some of the dyes could not be applied onto the substrate (Patel *et al*; 2002, Khosravi *et al*; 2005, Konstantinova and Petrova, 2002, Patel *et al*; 2011).

Patel, *et al*; (2011), Patel, *et al*; (2013) reported that it is necessary to increase the fixation properties of the reactive dyes by building bifunctionality into the dye as an efficient method of improving the application properties of the dye. Klaus *et al*; (2005), Clark, (2011) opined that it is implicit that dyes with two reactive groups provide a higher fixation yield than dyes with one reactive group because if one of the two dye-fibre bonds is hydrolysed, one reactive group is still left for fixation (Patel, *et al*; (2013). The bifunctional azo reactive dyes are more valuable than the mono azo reactive dyes, as they are more tinctorally stable and potentially more economic than mono azo reactive dyes (Patel, *et al*; 2011).

Patel, *et al*; (2014) reported that most interest has recently been shown to the dyes containing DCT reactive groups, which give deep colours on various textile fabrics than the dyes containing only one reactive group. The possibility of forming covalent bond between dyes and fibres had long been attractive to the dye chemist since attachment by physical adsorption and by mechanical obstruction had the disadvantage of either low wash fastness on high cost Mehta et al; (2004). This was anticipated since the attachment by covalent bonding of the dye molecules and fibre would result to high fastness properties due to the covalent attachment between molecules (Patel et al, (2004)

However, s-triazine chemical based have been applied majorly in polymers, dyes, drugs, explosives, pesticides and commodity manufacturing Patel et al, (2001) and had widely been carried out as a consequence, theoretical and experimental studies. Hence, s-triazine ring is now considered as an important conjugate heterocyclic with the result that the electronic properties are expected to show suitable differences from those of benzene due to the alternate replacement of the –CH- group by nitrogen atoms (Khazaei *et al*; 2013)

The purpose of the present investigation is to synthesize, characterise and determine the dyeing properties of five new novel homo bifunctional dichloro-s-triazinyl (DCT) azo reactive dyes containing 1,4-diphenylenediamine as a tetrazotized component prepared by coupling to 4-nitroanilino cyanurated acids (H-acid, J-acid, Laurent acid, Tobias acid and Gamma acid). The synthesised dyes were characterised using FT-IR, UV-visible absorption spectra, and the percentage exhaustion, percentage fixation and the fastness properties (washing, light and perspiration) were assessed.

MATERIALS AND METHODS

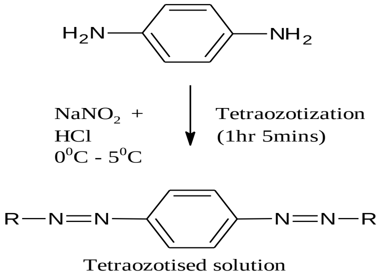
The chemicals used were purchased from Weifang Senya Chemical Company Limited, China. 1,4- phenylenediamine , 4,4-diaminodiphenylsulphone, 2,2-benzidinedisulphonic acid, 4,4'-diaminodiphenylmethane, Cyanuric chloride, H-acid, J-acid, Laurent acids, Tobias acid, Gamma acid. The melting points were determined by open capillary method. The IR and UV-visible absorption spectra were recorded using FT-IR (Perkin-Elmer Spectrum RXIFT-IR Spectrometer and Perkin-Elmer Lambda 25 UV-visible spectrophotometer (at the wavelength of maximum absorption (λmax) at the Kharazmi University, Bureau of International Scientific Cooperations, Department of Organic Chemistry, Tehran, Iran. Applications and evaluation of fastness properties were carried out according to the standard methods in Chemical Processing laboratory, Department of Polymer and Textile Engineering at the Ahmadu Bello University, Zaria.

METHODOLOGY

The synthesis of the bifunctional azo reactive dyes involves four (4) steps which are cyanuration of the acids, condensation of the cyanurated acids, tetrazotisation of the intermediates and the coupling of the tetrazotised intermediates with the condensed cyanurated acids (Patel *et al*; 2011).

Procedure for Tetrazotization of Intermediates

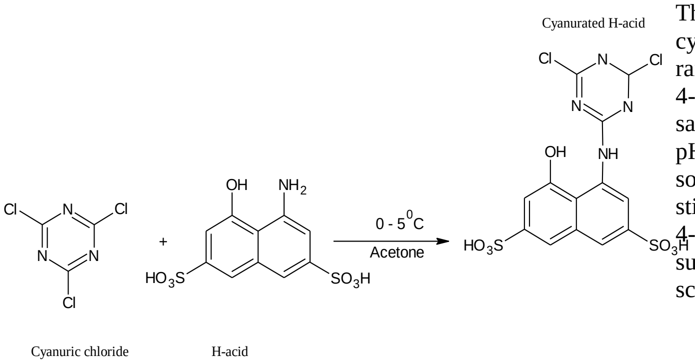
1,4-phenylenediamine (0.54g, 0.005mol) was suspended in distilled water (60ml) and hydrochloric acid (0.36g) was added dropwise to the well stirred suspension. The mixture was gradually heated up to 70°C, till clear solution was obtained. The formed solution was gradually cooled to below 5°C in an iced bath, then already cooled (0-5°C) $NaNO_2$ (0.6g in 4ml H_2O) was added over a period of 30mins with continuous stirring. The stirring was continued for one (1) hour, maintaining the temperature of 0°C-5°C with positive test for nitrous acid with starch iodide paper. After completely destroying the excess of nitrous acid with the required amount of sulphamic acid, the clear tetrazonium solution at 0°C-5°C obtained was used for next coupling reaction. The reaction as shown in the reaction Scheme 1 below:



Scheme 1: Tetrazotization of 1,4- phenylenediamine

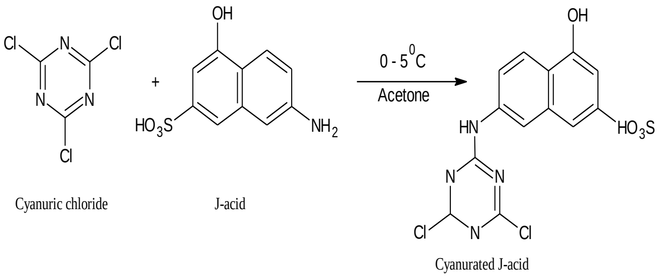
Procedure for cyanuration of the acids

Cyanuric chloride (1.85g, 0.01 mole) was stirred in acetone (25 ml) at a temperature below 5°C for a period of an hour. A neutral solution of H-acid (3.19g, 0.01mole) in aqueous sodium carbonate solution (10% w/v) was then added in small lots for about an hour. The pH was maintained neutral by simultaneous addition of sodium carbonate solution (1% w/v). The reaction mass was then stirred at 0-5°C for further 4 hours. The cyanurated H-acid solution was used for subsequent coupling reaction as shown in scheme 5 below:

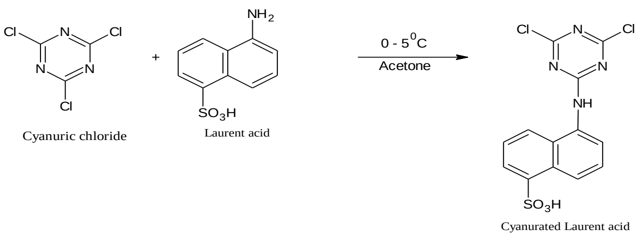


Scheme 2a: Cyanuration of H-acid

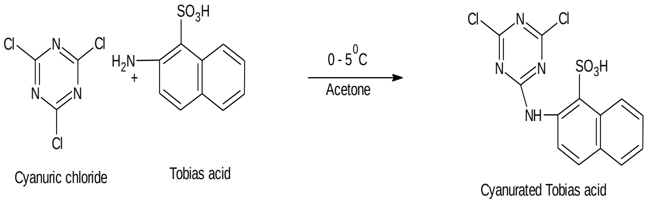
The same procedure (method) was followed for cyanurate J-acid, Laurent-acid, Tobias-acid and Gamma-acid in Scheme 2b, 2c, 2d and 2e respectively as shown below.



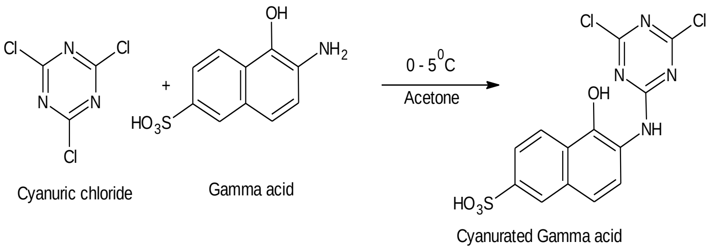
Scheme 2b: Cyanuration of J-acid



Scheme 2c: Cyanuration of Laurent acid



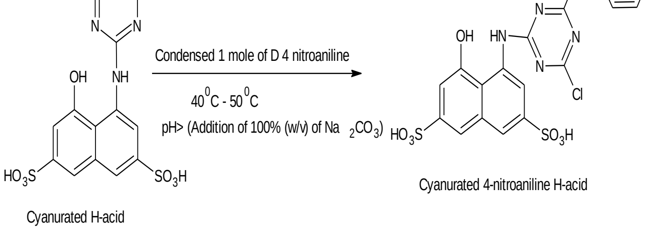
Scheme 2d: Cyanuration of Tobias acid



Scheme 2e: Cyanuration of Gamma-acid

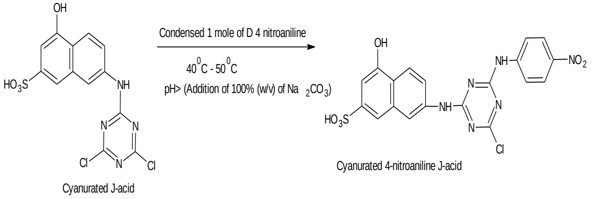
Procedure for condensation of the cyanurated acids

The temperature of ice-cooled well stirred solution of cyanurated H-acid, (4.67 g, 0.01 moles) was gradually raised to 45°C for half an hour. To this cyanurated H-acid, 4-nitro aniline (1.39 g, 0.01 moles) was added slowly at same temperature, for a period of 30 min, maintaining the pH neutral by simultaneous addition of sodium bicarbonate solution (1%w/v). After the addition was completed, stirring was continued for further 3 hours. The cyanurated 4- nitroanilino H-acid solution thus obtained was subsequently used for further coupling reaction as shown in scheme 1

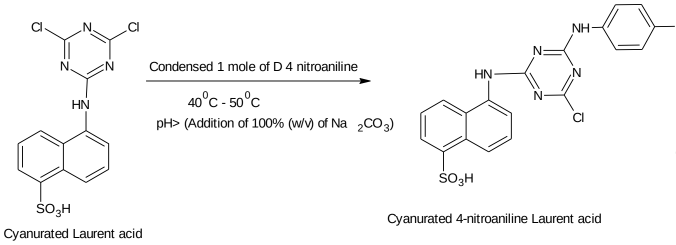


Scheme 3a: Condensation of cyanurated H-acid

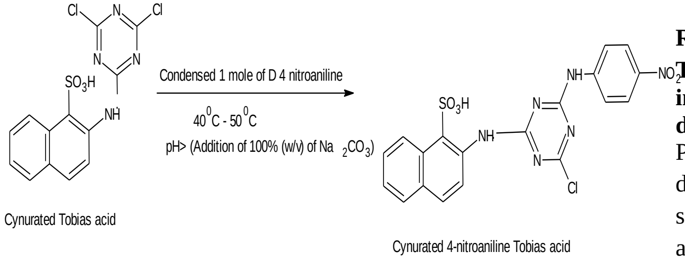
The same procedure (method) was followed for the condensation of cyanurated J-acid, Laurent-acid, Tobias-acid and Gamma-acid in scheme 3b, 3c, 3d and 3e respectively as shown below.



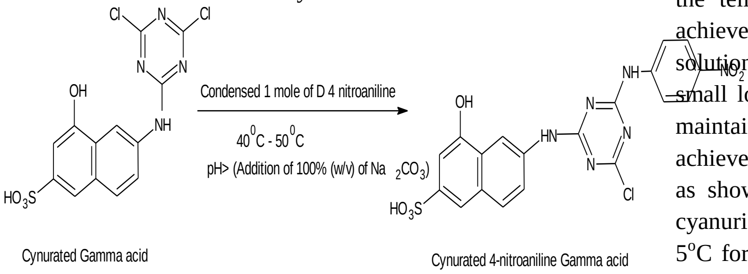
Scheme 3b: Condensation of cyanurated J-acid



Scheme 3c: Condensation of cyanurated Laurent-acid



Scheme 3d: Condensation of cyanurated Tobias-acid

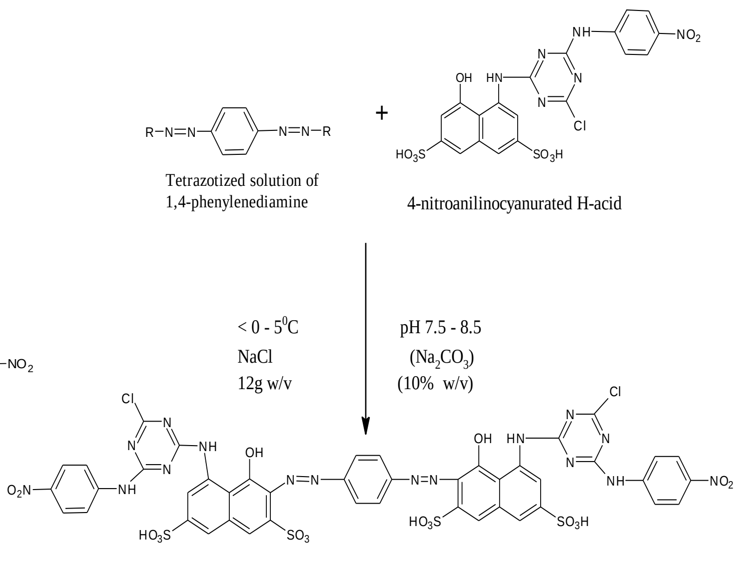


Scheme 3e: Condensation of cyanurated Gamma-acid

Procedure for Synthesis of dye DA_1

The synthesis of DA_1 was done by adding a freshly prepared solution of tetrazotized solution of 1,4-phenylenediamine dropwise over a period of 10 – 20 minutes to the ice-cold and well stirred solution of 4-nitroanilino cyanurated H-acid. The pH 7.5 – 8.5 was maintained by simultaneous addition of sodium carbonate solutions (Na_2CO_3) (10% w/v) where a purple solution was obtained. The stirring was continued to for 3 – 4h at a constant temperature below 0 – 5^0C for another 1h. A solid violet dye precipitated out and was filtered, washed with little amount of acetone and dried at room temperature with a yield of 87% as shown in the Scheme below:

Following the procedure above the other bifunctional dichloro-s-triazinyl (DCT) azo reactive dyes DA_2 (3b), DA_3 (3c), DA_4 (3d) and DA_5 (3e) were synthesised using the various cyanurated coupling components such as J-acid, Laurent acid, Tobias acid and Gamma acid respectively as shown in Table 1.



Scheme 4a: Synthesis of dye DA_1

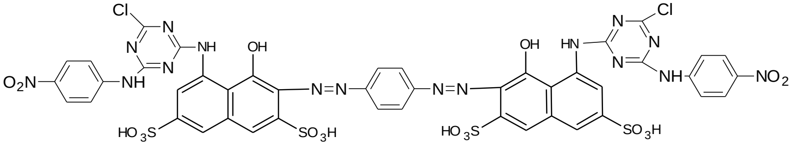
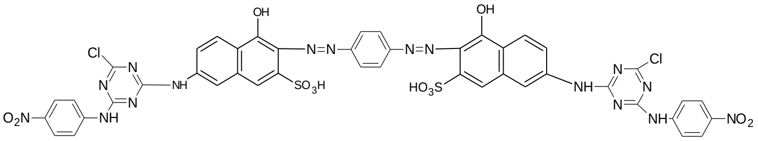
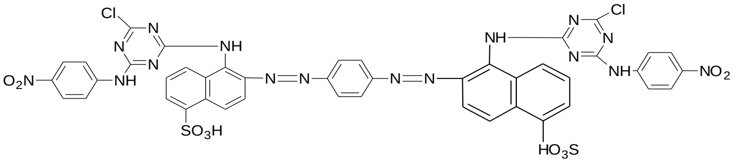
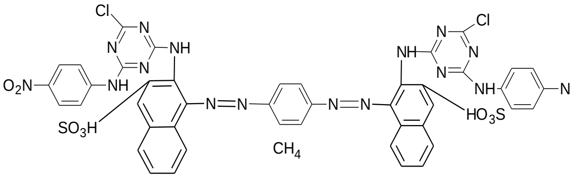
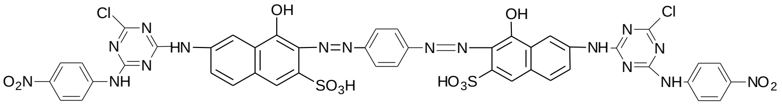
RESULTS AND DISSCUSSION

Tetrazotisation, cyanuration and condensation of the intermediates and synthesis of the bifunctional dichloro-s-triazinyl (DCT) Dyes

Preparation of Tetrazotised solution of 1,4-diphenylenediamine (0.54g, 0.005mol) was done by suspending 1,4-diphenylenediamine (0.54g, 0.005mol) and adding HCl (0.36g) dropwise with continuous stirring and the temperature raised to 70°C till a clear solution is achieved. The temperature lowed to below O°C-5°C and a solution of ice cold $NaNO_2$ (0.6g in 4ml H_2O) was added in small lots over 5 minutes with continuous stirring for 1h maintaining same temperature until a clear solution was achieved as shown in Scheme 1. Cyanuration of the acids as shown in Schemes 2a-2e was carried out by stirring cyanuric chloride in acetone at a temperature below 0°C-5°C for an hr, then an alkaline solution of H-acid (3.19g, 0.01mole) in aqueous solution of Na_2CO_3 (10% w/v) was added in small lots over an hr while maintaining a constant pH (7.0 – 8.5) by adding 1% w/v $NaCO_3$ at a temperature below 0°C-5°C with continuous stirring for 4h until a clear solution was obtained. In Schemes 3a-3e the condensation of the cyanurated acids was carried out by raising the temperature of the ice-cold well stirred cyanurated acids [(H-acid(3a), J-acid(3b), Laurent acid(3c), Tobias acid(3d) and Gamma acid(3e)] gradually to 50°C over 30 minutes, 4-nitroaniline was added in small lots over 30 minutes while maintaining a constant pH (7.0 – 8.5) by adding 1% w/v $NaHCO_3$ with continuous stirring for 5h to obtain the 4-nitroanilino cyanurated acids.

The dye DA_1 was synthesised according the route shown in Scheme 4a by adding freshly prepared solution of tetrazotised solution of 1,4-diphenylenediamine dropwise to an ice-cold well stirred of 4-nitroanilino cyanurated H-acid over a period of 10-15 minutes by maintaining pH at 7.5-8.5 by adding $NaCO_3$ (10%w/v) solution at the same time with continuous stirring for 3-4h at a temperature below 0°C-5°C, after which the dye was precipitated by adding 12g NaCl with continuous stirring for further 1h. The dye filtered, washed and dried at room temperature.

Table 1: Synthesized bifunctional dichloro-s-triazinyl (DCT) azo reactive Dyes Structures with IUPAC names

Dye No.	IUPAC Name	Structures of the Synthesized Bi-functional azo reactive Dyes
DA$_1$	3,3ˈ-(1,4-phenylenebis(diazene-2-1-diyl)bis(5-((4-chloro-6-((4-nitrophenyl)amino)-1,3,5-triazin-2-yl)amino)-4hydroxynapthelene-2,7-disulfonic acid)	
DA$_2$	3,3ˈ-(1,4-phenylenebis(diazene-2-1-diyl)bis(5-((4-chloro-6-((4-nitrophenyl)amino)-1,3,5-triazin-2-yl)amino)-4hydroxynapthelene-2-sulfonic acid)	
DA$_3$	3,3ˈ-(1,4-phenylenebis(diazene-2-1-diyl)bis(5-((4-chloro-6-((4-nitrophenyl)amino)-1,3,5-triazin-2-yl)amino)napthelene-1-sulfonic acid)	
DA$_4$	7,7ˈ-(1,4-phenylenebis(diazene-2-1-diyl)bis(2-((4-chloro-6-((4-nitrophenyl)amino)-1,3,5-triazin-2-yl)amino) napthelene-1-sulfonic acid)	
DA$_5$	3,3ˈ-(1,4-phenylenebis(diazene-2-1-diyl)bis(7-((4-chloro-6-((4-nitrophenyl)amino)-1,3,5-triazin-2-yl)amino)-4-hydroxynapthelene-2-sulfonic acid)	

Physical and Spectral Properties of the Synthesised Dyes

The shift in λmax of the synthesised bifunctional azo reactive dyes in different solvents (solvatochromism) is as a result of the solvatochromic effects emanating from changes in dielectric constant of the solvent. The measuremrnt of λmax in DMF and water indicated that λmax of the intermolecular charge transfer bonds exhibit a remarkable red shift on transfer from polar to non-polar solvent. This behaviour can be attributed to the polar excited states of the synthesised bifunctional azo reactive dyes which are stabilized by polarization interactions forces as the polarity of the solvent increases (Abd El- Aal and Koraierm, 2002).

Table 2: Physical Characteristics of the bifunctional dichloro-s-triazinyl (DCT) azo reactive dyes

Dye No.	**Calculated Mol.Wt (g/mol)**	**Mt.Pt (^{o}C)**	**Yield (%)**	**DMF (λ_{max})**	**Water (λ_{max})**	**ε_{max}inDMF x$10^4$1Mol^{-1}cm^{-1}**
DA$_1$	1204.05	310-312	51	530	529	2.07
DA$_2$	1106.09	338-340	81	510	510	3.18
DA$_3$	1074.10	320-321	50	415	403	8.53
DA$_4$	1074.10	315-318	70	413	413	1.69
DA$_5$	1106.09	300-302	62	526	516	2.30

The visible absorption maxima of the synthesised bifunctional azo reactive dyes as recorded in Table 2 fell within the visible region (400-700nm) of the electromagnetic spectrum. The values of the molar extinction coefficient (ε) that were determined by Beer-Lambert's law are in the range of 16904.14-85316.01mol^{-1}cm^{-1} which is an indication of high absorption intensity of the synthesised bifunctional azo reactive dyes in DMF respectively. However, the synthesised bifunctional azo reactive dyes have different chromophoric functionalities, but same bridging groups and it is also apparent that the value of λmax depends on the coupling components used. The presence of electron donating or electron attracting (withdrawing) groups at the suitable position of the coupler ring affects the absorption characteristics of the synthesised bifunctional azo reactive dyes (Patel *et al*; 2014). Coupling of 1,4 phenylenediamine with cyanurated 4-nitroanilino H-acid gave DA$_1$ which absorbed at 530nm in DMF, but when cyanurated 4-nitroanilino H-acid was replaced with cyanurated 4-nitroanilino J-acid dye DA$_2$ was synthesised which absorbed at 510nm which gave hypsochromic shift of 20nm when compared to DA$_1$. Replacing cyanurated 4-nitroanilino J-acid with Laurent acid dye DA$_3$ was synthesised which absorbed at 415nm in DMF and gave a hypsochromic shift of 115nm when compared to DA$_1$ and hypsochromic shift of 95nm when compared with DA$_2$. Meanwhile, replacing cyanurated 4-nitroanilino Laurent acid with cyanurated 4-nitroanilino Tobias acid, dye DA$_4$ was synthesised which absorbed at 413nm in DMF and gave a hypsochromic shift of 117nm when compared to DA$_1$, hypsochromic shift of 97nm when compared to DA$_2$, and hypsochromic shift of 2nm when compared to DA$_3$ respectively. However, replacing cyanurated 4-nitroanilino Tobias acid with cyanurated 4-nitroanilino Gamma acid, dye DA_5 was synthesised which absorbed at 506nm in DMF and gave a hypsochromic shift of 24nm when compared to DA_1, hypsochromic shift of 4nm when compared to DA_2, bathochromic shift of 91nm when compared to DA_3, and bathochromic shift of 93nm when compared to DA_4 respectively. These observations may be attributed to fact that although the dyes have the same chromophoric functionalities but the positions of the bridging groups are different, hence for dyes that shited bathochromically (higher value) they are more place for groups such as SO_3H which cause faster electron oscillation and hence neutralisation of electron faster than dyes with hypsochromic shift (lower value).

FT-IR spectra characteristics

In general the infra-red spectral of all the dyes DA_1, - DA_5 revealed that O-H and N-H showed stretching vibration at 3405cm-^{1}, C=H showed stretching vibration at 2900cm-^{1}, C-N showed stretching vibration at 1574cm-^{1}, 1305cm-^{1}, 1464cm-^{1}, N=N showed stretching vibration at 1643cm-^{1}, S=O showed stretching vibration at 1165cm-^{1}, 1060cm-^{1} (asym. And sym), N=O showed stretching vibration at 1595cm-^{1}, 1374cm-^{1}, 1335cm-^{1}, C-Cl showed stretching vibration at 784cm-^{1} and SO_3H showed stretching vibration at 841cm-^{1} respectively as showed in Table 3

Table 3: IR of the bifunctional dichloro-s-triazinyl (DCT) azo reactive

Dye no	IR(KBr) cm^{-1}
DA_1	3405 (O-H), 3365 (N-H), 2900 (C-H), 1374, 1469 (C-N), 1643 (N=N), 1165, 1060 (S=O), 1595 (N=O), 784 (C-Cl), 841 (SO_3H)
DA_2	3410 (O-H), 3054 (N-H), 2915 (C-H), 1329 (C-N), 1619 (N=N), 1175 (S=O), 1499 (N=O), 765 (C-Cl), 800 (SO_3H)
DA_3	3451 (O-H), 3136 (N-H), 3055 (C-H), 1597 (C-N), 1650 (N=N), 1153, 1072 (S=O), 1515 (N=O), 779 (C-Cl), 840 (SO_3H)
DA_4	3446 (O-H), 3142 (O-H), 3059 (C-H), 1515 (C-N), 1651 (N=N), 1153 (S=O), 1361 (N=O), 779 (C-Cl), 956 (SO_3H)
DA_5	3416 (O-H), 3051 (N-H), 2995 (C-H), 1563 (C-N), 1659 (N=N), 1150 (S=O), 1512 (N=O), 780 (C-Cl), 841 (SO_3H)

Dyeing procedure

The synthesized dyes DA_1 – DA_5 were applied on cotton and nylon 6,6 fabrics using exhaustion method (Patel *et al.*, 2011). The depth of shade 2% was used. A mild alkaline condition pH 8.0-9.0 (Na_2CO_3, 0.4ml of 10% w/v) was used for the dyeing of cotton fabrics while a weakly acidic conditions pH4.5-5(acetic acid, 1.5ml of 10% w/v. NaCl, 4ml of 10% w/v, H_2O, 14.4ml) was used for the dyeing of nylon fabrics at 1:50 liquor ratio for the dyeing of both fabrics (Patel *et al*; 2014).

Exhaustion and fixation study

The data for the % exhaustion and fixation were calculated using the method by Patel *et al.* (2011) as shown in Table 4. From the result, the percentage exhaustion dyeing on the cotton fabric range from 63-78% where dye DA_4 showed maximum exhaustion of 78% and dye DA_2 showed minimum exhaustion of 63%. Meanwhile, the percentage exhaustion dyeing on the nylon 6,6 fabric range from 68-81% where dye DA_4 showed maximum exhaustion of 81% and dye DA_5 showed minimum exhaustion of 68%. The percentage fixation of 2% dyeing on cotton range from 79-92% where dye DA_1 showed maximum fixation of 92% and dye DA_5 showed minimum fixation of 79%. Meanwhile, the percentage exhaustion dyeing on the nylon 6, 6 fabric ranges from 86-96% where dye DA_1 showed maximum fixation of 96% and dye DA_4 showed minimum fixation of 86% respectively. Good exhaustion and fixation values of dyes are in accordance with structure of dyes bearing polar groups which establishes physical and chemical interaction with the fibre.

Table 4: Exhaustion and fixation study of the bifunctional dichloro-s-triazinyl (DCT) azo reactive dyes on cotton and Nylon 6,6 fabric

			Exhaustion (%)		Fixation (%)	
Dye No	**Shade on cotton**	**Shade on nylon 6,6**	**Cotton**	**Nylon 6,6**	**Cotton**	**Nylon 6,6**
DA_1	Light brown	Pink	69	75	92	96
DA_2	Light orange	Light orange	63	69	87	92
DA_3	Yellow	Yellow	68	71	85	92
DA_4	Light orange	Light orange	78	81	89	86
DA_5	Yellow	Yellow	71	68	79	91

Fastness properties

The data for the fastness properties to washing (IS 765-1979 (Patel *et al*, 2011), fastness properties to light (BS: 1006-1978 (Patel *et al*, 2011) and fastness properties to perspiration – alkaline and acidic (Kkaeaei *et al*, 2013) are recorded in Table 4. From the result the fastness properties to washing, fastness properties to light and fastness properties to perspiration – alkaline and acidic showed very good performance.

Table 5: Fastness properties of the bifunctional dichloro-s-triazinyl (DCT) azo reactive dyes on cotton and Nylon 6, 6 fabrics

					Perspiration fastness			
Dye No	Wash fastness		Light fastness		Alkaline		Acidic	
	Cotton	Nylon 6,6	Cotton	Nylon 6,6	Cotton	Nylon 6,6	Cotton	Nylon 6,6
	Cc	cc	cc	Cc	Cc	cc	cc	cc
DA_1	4	4-5	6	6	4	3-4	4	3-4
DA_2	5	4	5-6	4	3-4	4	4	4
DA_3	4-5	4	4	4	4	3-4	4	3-4
DA_4	4	4	4	5	4	3	4	4
DA_5	4	4	5	5	4	4	3	3-4

cc=colour change

Fastness properties to washing: 1-poor, 2-moderate, 3-fair, 4-very good, 5-excellent

Fastness properties to light: 1-poor, 2-slight, 3- moderate, 4- fair, 5- good, 6- very good, 7-high, 8- very high

Fastness properties to perspiration – alkaline and acidic: 1-poor, 2-fair 3-good, 4-very good

CONCLUSION

New bifunctional dichloro-s-triazinyl (DCT) azo reactive dyes based on 1, 4-diphenylenediamine has been successfully synthesised. 1,4-diphenylenediamine was tetrazotized and coupled with cyanurated 4-nitroanilino R acids (H-acid, J-acid, Laurent acid, Tobias acid and Gamma acid) coupling components to give the corresponding bifunctional dichloro-s-triazinyl (DCT) azo reactive dyes (DA_1-DA_5). The synthesised dyes were applied on cotton and nylon 6, 6 fabrics by exhaustion dyeing methods. These dyes gave pink, orange, brown and yellow shade on cotton and nylon 6, 6 fabrics. The hues of the dyed fabrics (cotton and nylon 6,6) wre varied due to the nature and positions of the substituent's on the coupler ring. The dyes gave better light fastness and seem to have good dyeing performance on cotton and nylon 6, 6 fabrics. The exhaustion and fixation performance of these dyes are very satisfactory. The remarkable degree of levelness after washing indicates the good penetration and affinity of these dyes to cotton and nylon 6, 6 fabrics.

REFERENCES

Abd El- Aal, R. M. and Koraierm, A.I (2000). Synthesis, Absorption Spectra Studies and Biological Activity of some Novel onjugated Dyes. *J. Chin. Chem. Soc.* 47: 389-395.

Ali, S.; Khatri, Z., Khatri, A. and Tanwari, A. (2014). Integrated desizing-bleaching-reactive dyeing process for cotton towel using glucose oxidase enzyme. *J. Clean. Prod*., 66: 562–567.

Alam, M.S., G. A., Khan, S. A., Razzaque, M. J., Hossain, M., Minhaz-ul-Haque and Zebsyn, S. (2000). Dyeing of cotton fabrics with reactive dyes and their physico-chemical properties. *Indian Journal of Fibre & Textile Research*, 33: 58-65.

Al-Degs, Y. S., El-Barghouthi, M. I., El-Sheikh, A. H. and Walker, G.M. (2008). Effect of solution pH, ionic strength, and temperature on adsorption behavior of reactive dyes on activated carbon. *Dyes Pigment*., 77: 16–23.

Allen, N., Thomas, J., Edge, M., Follows, G. and Heinz, R. (1997). Influence of dyeing parameters and fibre setting on the light fastness properties of acid dyed nylon 6, 6 fibres. *Dyes and Pigments*, 34(3): 169-180.

Almasian, A., Olya, M. E. and Mahmoodi, N. M. (2015). Synthesis of polyacrylonitrile/polyamidoamine compositenanofibers using electrospinning technique and their dye removal capacity, *J. Taiwan Inst. Chem. Eng*., 49: 119–128.

Ahmed, N. S. E. (2005). The use of sodium edate in the dyeing of cotton with reactive dyes. *Dyes Pigment*., 65: 221–225.

Anderson, M. J. and Whitcomb, P. J. (1994). Technical Information on Continuous Dyeing of Cellulosics, CH-3, *Society of Dyers and Colourists,*

Blanco, S.P.D.M., Scheufele, F. B., Módenes, A.N., Espinoza-Quiñones, F. R., Marin, P., Kroumov, A. D. and Borba, C.E. (2016). Kinetic, equilibrium and thermodynamic phenomenological modeling of reactive dye adsorption onto polymeric adsorbent. *Chem. Eng. J*., 307: 466–475.

Bravo-Díaz, D. C. (2010). Diazohydroxides, Diazoethers and Related Species. In: Rappoport, Zvi (Ed.), *PATai's Chemistry of Functional Groups,* John Wiley & Sons, Ltd., doi:10.1002/9780470682531.pat0511 https://doi.org/10.1002%2F9780470682531

Broadbent, A. D. (2001). *Basic principles of textile coloration*. Bradford Society of Dyers and Colorists West Yorkshire, 579 pp.

Christie, R. M. (2001). Colour Chemistry. The Royal Society of Chemistry, U.K., pp. 135-147.

Christie, R. M. (2014). Colour chemistry. Royal Society of Chemistry. 372 pp. DOI: 10.1039/C3CP55336A. 54.

Cid, M. V. F., Spronsen, J. V., Kraan, M. V. D., Veugeler, W. J. T., Woerlee, G. F. and Witkamp, G. J. (2007). A significant approach to dye cotton in supercritical carbon dioxide with fluorotriazine reactive dyes. *J. Supercrit. Fluid*. 40: 477–484.

Clark, M. (2011). *Fundamental principles of dyeing, Handbook of Textile and Industrial Dyeing: Principles, Processes and Types of Dyes*, 1. Woodhead Publishing Series in Textiles.

Clark, J. (2011). Chemguide http://www.chemguide.co.uk/organicprops/aniline/propsdiazo.html

Clark M. (2002). The interaction of reactive dyes with modified cellulose and polyamide substrates, Ph.D. Thesis, University of Leeds.

Khazaei, A, Kazem-Rostami, M, Zare, A, Moosavi-Zare, A. R., Sadeghpour, M. and Afkhami, A. (2013). Synthesis, characterization, and application of a triazene-based polysulphone as a dye adsorbent", *Journal of Applied Polymer Science,* 129(6): 3439–3446. doi:10.1002/app.39069

Khosravi, A., Moradiam, S., Garanjig, K. and Taromi, F. A. (2005). Synthesis and characterization of some monoazo disperse dyestuffs based on Naphthalimide Derivatives for dyeing polyester, *Journal of the Chinese Chemical Society*, 52,495-502.

Klaus, H., Peter, M. and Wolfgang, R. (2005). Azo Dyes. In: Ullmann's Encyclopedia of Industrial Chemistry, Wiley-VCH, Weinheim. doi:10.1002/14356007.a03_245

Konstantinova, T. and P. Petrova (2002). On the synthesis of some bifunctional reactive triazine dyes. *Dyes and Pigments*, 52(2): 115-120.

Patel, R., Patel, P., Patel, K. and Patel, S. (2002). Synthesis of non-benzidine reactive dyes and their application on various fibres. *Asian Journal of Chemistry*, 14(1): 420-426.

Patel, D. R. and Patel, K. C. (2010). Synthesis, characterization and application of quinazolinone based reactive dyes for various fibers. *Fibers and Polymers*, 11(4): 537-544.

Patel, D. R and Patel K. C. (2012). Synthesis, characterization and in vitro antimicrobial screening of some new MCT reactive dyes bearing nitro quinazolinone moiety *Journal of Saudi Chemical Society*, doi: org/10.1016/j.jscs.2012.02.002

Patel, D. R., Patel, A. L. and Patel, K. C. (2011). Colorimetric studies of some newly synthesized bisazo reactive dyes. *Arabian Journal of Chemistry.* 4(4): 279-285. 145.

Patel, D. R. and Patel, K. C. (2011). Synthesis and characterization of reactive dyes based on 2-phenyl-3-[4′-(4'-aminophenylsulphonamido)] phenyl-4(3h)-quinazolinone-6-sulphonic acid. *Arabian Journal of Chemistry*, 4(3): 279-285.

Patel, D. M., Patel, T. S. and Dixi, B. C. (2013). Synthesis, characterization and dyeing performance of new bisazo–bisazomethine disperse dyes. *Journal of Saudi Chemical Society*, 17(2): 203-209.

Patel, J. R., Patel, M. H. Pranav, S. and Mallika, S. S. (2013). Effect of domestic laundering on color fastness of three newly synthesized reactive dyes on 100% cotton fabric. *Indian Journal of Science*, 4(11): 68-75.

Patel, J. R., Patel, M. H., Shrivastav, P. S. and Sanyal, M. (2014). Standardization of variables for dyeing cotton fabric with five synthesized reactive dyes, *J. Phy. Chem. Sci*, 1: 1-7.