

Quinoline incorporating pyrimidine heterocyclic azo dye derivatives: Synthesis, characterization and applications

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In the present study, novel quinoline substituted pyrimidine heterocyclic azo dye derivatives (**IVa-x**) have been synthesized. All the prepared compounds have been characterized by several spectroscopic analyses and evaluated for *in vitro* antimicrobial activity. Furthermore, the dyeing performances of all the compounds have been estimated on cotton and silk fabrics. Colour and rubbing fastness properties of all the dyed fabrics have been examined by referencing to standard gray scale. These newly synthesized azo dyes exhibit various hues with good to excellent fastness properties. Out of all the phenol derivatives, Naphthol substituted compounds have come out as remarkably active antimicrobial scaffolds with excellent dye characteristics.

Keywords: Quinoline, Pyrimidine, Heterocyclic Azo-Dye, Antimicrobial activity, Fastness properties

Heterocyclic compounds are leading assembly in innumerable organic compounds and also most periodic natural products because of their application across various areas^{1,2}. Compounds with multiple nitrogen containing heterocyclic rings are considered as a most active class of compound with numerous applications³⁻⁷. Clubbed N-heterocycles have an immense range of medicinal applications such as antimicrobial^{8,9}, anticancer¹⁰, analgesic¹¹, antimalarial¹², antitumor¹³, anti-inflammatory¹⁴, antiviral¹⁵ and antioxidant¹⁶. Other than their medicinal importance they are also used as agrochemicals¹⁷ and as veterinary¹⁸ products. They also find applications as sanitizers¹⁹, developers, as corrosion inhibitors²⁰, as copolymers²¹ and dye stuff²².

Azo dyes are the supreme class of dyes considering more than 50% of all commercial dyes or colourants²³. The importance of azo dyes containing heterocyclic rings is due to their superior properties like chromophoric strength and brilliant hues as colorants. Such dyes have higher intensity dyeing properties and excellent fastness properties²⁴. The application of heterocyclic azo dye lies in the field of textiles, papers, leather, additives, food stuff, cosmetics, holographic data storage materials, xerography laser materials, laser printing, materials for organic solar cells chemo-sensors, harmonic nonlinear optical material and optical switching

devices²⁵⁻²⁸. Numerous heterocyclic azo dyes and their derivatives are utilized as intermediates in dye industry²⁹.

According to literature survey and our previous work in this area suggests that more potent novel chemical entities can be envisaged by uniting two or more heterocyclic systems into a single molecule³⁰⁻³³. Such wide range of applications of clubbed N-heterocycles lead us to the synthesis of quinoline based pyrimidine heterocyclic azo dyes and perceive various possible applications. Results of the research on the preparation of these derivatives and an exploration of their several properties are detailed in this article.

Experimental Details

All chemicals used were of AnalaR grade.¹H NMR and ¹³C NMR spectra were recorded on Bruker spectrometer (400 MHz) using CDCl₃ as solvent and tetramethylsilane (TMS) as an internal standard. The Infrared spectra (ν , cm⁻¹) of all the compounds were recorded on a Perkin-Elmer FT-IR spectrophotometer as KBr pellets. Mass spectra were obtained on Shimadzu LC-MS spectrometer. Elemental analyses were performed on an ECS 4010 Elemental Combustion System and the results were within the accepted range (± 0.40) of the calculated values. Melting points were determined on an electro thermal

melting point apparatus and are uncorrected. The completion of reaction and homogeneity of compounds were checked on aluminium backed silica gel coated TLC plates 60 F245 (E. Merck) using methanol: chloroform (0.5:9.5 V/V) as mobile phase and visualized under ultraviolet (UV) light or in an iodine chamber.

Procedure for the synthesis of 2-chloroquinoline-3-carbaldehyde, I

2-Chloroquinoline-3-carbaldehyde was synthesized by known literature method³⁴.

Procedure for the synthesis of ethyl 4-(2-chloroquinolin-3-yl)-6-methyl-2-(oxo/thio)-1,2-dihydropyrimidine-5-carboxylate, II

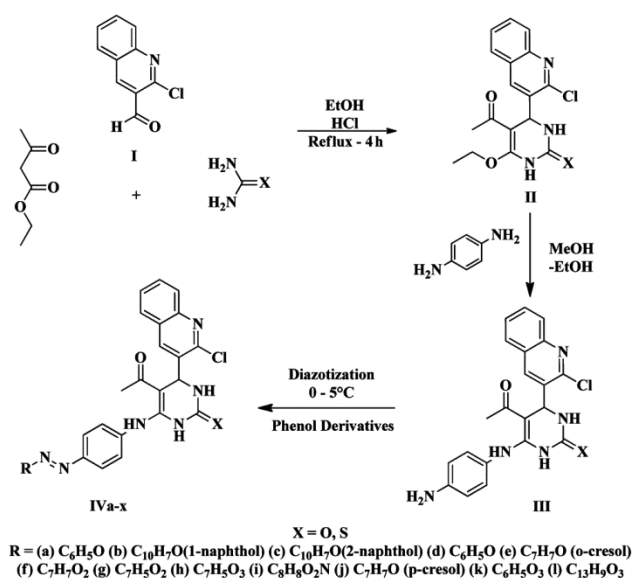
A mixture of 2-chloroquinoline-3-carbaldehyde I (0.01 mol), ethyl acetoacetate (0.01 mol) and urea/thiourea (0.03 mol) in ethanol (25 mL) containing conc. HCl (0.01 mol) were refluxed for 4 h. The completion of reaction was monitored by TLC. The reaction mixture was poured into ice water. The separated products were filtered out, washed thoroughly with water, dried and purified by recrystallization from ethanol to afford the desired compound.

Procedure for the synthesis of *N*-(4-aminophenyl)-4-(2-chloroquinolin-3-yl)-6-methyl-2-(oxo/thio)-1,2-dihydropyrimidine-5-carboxamide, III

A mixture of 4-(2-chloroquinolin-3-yl)-6-methyl-2-(oxo/thio)-1,2-dihydropyrimidine-5-carboxylate II (0.01 mol) and benzene-1,4-diamine (0.01 mol) were dissolved in toluene and refluxed for 1 h. The completion of reaction was monitored by TLC. Products was filtered out, washed thoroughly with water, dried and purified by recrystallization from ethanol to afford the desired compound.

General procedure for the synthesis of (4-(2-chloroquinolin-3-yl)-6-methyl-2-(oxo/thio)-1,2-dihydropyrimidin-5-yl)(4-((2-hydroxyphenyl) diazenyl)phenyl)methanone, IVa-x

A solution of *N*-(4-aminophenyl)-4-(2-chloroquinolin-3-yl)-6-methyl-2-(oxo/thio)-1,2-dihydropyrimidine-5-carboxamide III (0.01 mol) was prepared in minimum amount of hydrochloric acid (2 mL). A cold solution of sodium nitrite below 5°C was then added to it with continuous stirring. A solution of phenol derivative (0.01 mol) was prepared by dissolving in sodium hydroxide solution and then cooled to 0°C. The cold diazonium salt was added to solution of phenol derivative in sodium hydroxide kept at 0°C. The solid dye was filtered out, dried and purified by recrystallization from ethanol (Scheme 1).



Scheme 1 — Synthetic path of compounds IVa-x

4-(2-Chloroquinolin-3-yl)-N-((2-hydroxyphenyl)-diazenyl)phenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVa: Yield 82%. m.p.190–191°C. FT-IR (KBr): 3776.32 (s, O-H), 3210.26 (b, O-H), 689.34 (m, Ar-Cl), 1445.62 (m, C=C), 2840.15 (m, C-H), 1625.69 (s, C=N), 1332.17 (s, C-N), 3612.39 (m, 2° N-H), 1025.12 (s, C-O), 1345.92 cm⁻¹ (m, C-C in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.32 (s, CH₃), 7.30-7.95 (m, 4H phenol ring), 7.55–8.01 (m, 4H quinoline ring), 9.53 (s, C₅-H quinoline ring), 9.70 (s, Ar-NH pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 31.08 (1C, CH₃), 120.87–149.20 (5C, phenol ring), 141.27 (1C, C-N, quinoline ring), 149.28 (1C, C=N, quinoline ring), 151.12 (1C, C=O, pyrimidine ring); LCMS: *m/z* 512.14 [M⁺]. Anal. Calcd for C₂₇H₂₁ClN₆O₃: C, 63.22; H, 4.13; N, 16.38. Found: C, 63.20; H, 4.11; N, 16.33%.

4-(2-Chloroquinolin-3-yl)-N-((1-hydroxynaphthalen-2-yl)diazenyl)phenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVb: Yield 81%. m.p.201–202°C. FT-IR (KBr): 3802.54 (s, O-H), 3212.22 (b, O-H), 618.27 (m, Ar-Cl), 1436.10 (m, C=C), 2846.26 (m, C-H), 1623.54 (s, C=N), 1339.50 (s, C-N), 3609.18 (m, 2° N-H), 1032.78 (s, C-O), 1338.79 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.57 (s, CH₃), 7.46-8.05 (m, 6H, naphthol ring), 7.30–8.40 (m, 4H, quinoline ring), 9.98 (s, C₅-H,quinoline ring), 9.42 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 31.92 (1C, CH₃), 120.22–149.84 (5C, naphthol ring), 141.27 (1C, C-N,

quinoline ring), 149.28 (1C, C=N, quinoline ring), 153.18 (1C, C=O, pyrimidine ring); LCMS: m/z 562.15 [M⁺]. Anal. Calcd for C₃₁H₂₃ClN₆O₃: C, 66.13; H, 4.12; N, 14.93. Found: C, 66.17; H, 4.13; N, 14.93%.

4-(2-Chloroquinolin-3-yl)-N-(4-((2-hydroxynaphthalen-1-yl)diazanyl)phenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVc: Yield 81%. m.p.207–208°C. FT-IR (KBr): 3800.02 (s, O-H), 3214.22 (b, O-H), 648.30 (m, Ar-Cl), 1436.26 (m, C=C), 2846.32 (m, C-H), 1625.50 (s, C=N), 1339.58 (s, C-N), 3649.15 (m, 2° N-H), 1032.54 (s, C-O), 1238.70 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.55 (s, CH₃), 7.45-7.99 (m, 6H, naphthol ring), 7.32–8.06 (m, 4H, quinoline ring), 8.15 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 16.30 (1C, CH₃), 123.50–131.44 (5C, naphthol ring), 145.19 (1C, C-N, quinoline ring), 149.80 (1C, C=N, quinoline ring), 154.70 (1C, C=O, pyrimidine ring); LCMS: m/z 562.15 [M⁺]. Anal. Calcd for C₃₁H₂₃ClN₆O₃: C, 66.13; H, 4.12; N, 14.93. Found: C, 66.17; H, 4.11; N, 14.95%.

4-(2-Chloroquinolin-3-yl)-N-(4-((2,6-dihydroxyphenyl)diazanyl)phenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVd: Yield 80%. m.p.219–220°C. FT-IR (KBr): 3779.36 (s, O-H), 3204.20 (b, O-H), 618.24 (m, Ar-Cl), 1439.51 (m, C=C), 2848.36 (m, C-H), 1627.10 (s, C=N), 1332.21 (s, C-N), 3602.08 (m, 2° N-H), 1025.41 (s, C-O), 1333.54 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.13 (s, CH₃), 7.38-7.90 (m, 3H, resorcinol ring), 7.42–8.11 (m, 4H, quinoline ring), 9.50 (s, C₅-H, quinoline ring), 9.88 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 31.22 (1C, CH₃), 120.56–149.41 (3C, resorcinol ring), 143.22 (1C, C-N, quinoline ring), 149.21 (1C, C=N of quinoline ring), 151.28 (1C, C=O, pyrimidine ring); LCMS: m/z 528.13 [M⁺]. Anal. Calcd for C₂₇H₂₁ClN₆O₄: C, 61.31; H, 4.00; N, 15.89. Found: C, 61.33; H, 4.03; N, 15.91%.

4-(2-Chloroquinolin-3-yl)-N-(4-((2-hydroxy-3-methylphenyl)diazanyl)phenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVe: Yield 78%. m.p.213–214°C. FT-IR (KBr): 3773.42 (s, O-H), 3201.80 (b, O-H), 618.02 (m, Ar-Cl), 1431.25 (m, C=C), 2840.12 (m, C-H), 1622.33 (s, C=N), 1335.64 (s, C-N), 3604.98 (m, 2° N-H), 1021.05 (s, C-O), 1335.28 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400 MHz): δ 2.21 (s, CH₃), 7.22-7.98 (m, 3H,

cresol ring), 7.21–8.87 (m, 4H, quinoline ring), 9.22 (s, C₅-H, quinoline ring), 9.84 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 31.19 (1C, CH₃), 121.22–149.87 (3C, cresol ring), 141.42 (1C, C-N, quinoline ring), 149.33 (1C, C=N, quinoline ring), 151.22 (1C, C=O, pyrimidine ring); LCMS: m/z 526.15 [M⁺]. Anal. Calcd for C₂₈H₂₃ClN₆O₃: C, 63.82; H, 4.40; N, 15.95. Found: C, 63.84; H, 4.38; N, 15.91%.

4-(2-Chloroquinolin-3-yl)-N-(4-((2,6-dihydroxy-4-methylphenyl)diazanyl)phenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVf: Yield 79%. m.p.215–216°C. FT-IR (KBr): 3771.81 (s, O-H), 3209.47 (b, O-H), 622.05 (m, Ar-Cl), 1431.28 (m, C=C), 2845.20 (m, C-H), 1624.89 (s, C=N), 1332.42 (s, C-N), 3601.08 (m, 2° N-H), 1025.42 (s, C-O), 1322.51 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.11 (s, CH₃), 7.37-8.02 (m, 2H, orcinol ring), 7.50–8.22 (m, 4H, quinoline ring), 9.43 (s, C₅-H, quinoline ring), 9.52 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 31.54 (1C, CH₃), 120.14–149.16 (2C, orcinol ring), 141.88 (1C, C-N, quinoline ring), 149.36 (1C, C=N, quinoline ring), 151.25 (1C, C=O, pyrimidine ring); LCMS: m/z 542.15 [M⁺]. Anal. Calcd for C₂₈H₂₃ClN₆O₄: C, 63.70; H, 4.20; N, 13.26. Found: C, 63.73; H, 4.19; N, 13.22%.

4-(2-Chloroquinolin-3-yl)-N-(4-((3-formyl-2-hydroxyphenyl)diazanyl)phenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVg: Yield 78%. m.p.220–221°C. FT-IR (KBr): 3756.32 (s, O-H), 3210.28 (b, O-H), 618.32 (m, Ar-Cl), 1422.17 (m, C=C), 2815.87 (m, C-H), 1628.92 (s, C=N), 1315.29 (s, C-N), 3616.40 (m, 2° N-H), 1024.83 (s, C-O), 1317.30 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.15 (s, CH₃), 7.31-7.89 (m, 3H, salicylaldehyde ring), 7.21–8.15 (m, 4H, quinoline ring), 9.42 (s, C₅-H quinoline ring), 9.80 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 31.12 (1C, CH₃), 121.59–149.85 (3C, salicylaldehyde ring), 141.26 (1C, C-N, quinoline ring), 145.36 (1C, C=N, quinoline ring), 151.25 (1C, C=O, pyrimidine ring); LCMS: m/z 525.12 [M⁺]. Anal. Calcd for C₂₈H₂₁ClN₆O₄: C, 62.17; H, 3.91; N, 15.54. Found: C, 62.19; H, 3.93; N, 15.51%.

3-((4-(2-Chloroquinolin-3-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxamido)phenyl)diazanyl)-2-hydroxybenzoic acid, IVh: Yield 77%. m.p.229–230°C. FT-IR (KBr): 3901.10 (s, O-H), 3210.36 (b, O-H), 618.22 (m, Ar-Cl), 1395.09 (m, C=C), 2842.21 (m, C-

H), 1630.69 (s, C=N), 1239.42 (s, C=N), 3602.33 (m, 2° N-H), 1040.26 (s, C-O), 1322.20 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.31 (s, CH₃), 7.30-7.95 (m, 3H, salicylic acid ring), 7.85-8.42 (m, 4H, quinoline ring), 9.28 (s, C₅-H quinoline ring), 10.61 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 29.20 (1C, CH₃), 120.21-142.36 (3C, salicylic acid ring), 142.52 (1C, C-N, quinoline ring), 144.89 (1C, C=N, quinoline ring), 161.26 (1C, C=O, pyrimidine ring); LCMS: *m/z* 556.13 [M⁺]. Anal. Calcd for C₂₈H₂₀ClN₅O₅: C, 60.38; H, 3.80; N, 15.09. Found: C, 60.35; H, 3.79; N, 15.10%.

N-(4-((5-Acetamido-2-hydroxyphenyl)diazanyl) phenyl)-4-(2-chloroquinolin-3-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVi: Yield 75%. m.p.243-244°C. FT-IR (KBr): 3721.06 (s, O-H), 3210.58 (b, O-H), 619.64 (m, Ar-Cl), 1439.62 (m, C=C), 2822.36 (m, C-H), 1616.41 (s, C=N), 1333.34 (s, C=N), 3609.14 (m, 2° N-H), 1020.17 (s, C-O), 1335.89 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.21 (s, CH₃), 7.21-7.83 (m, 1H, paracetamol ring), 7.41-8.16 (m, 4H, quinoline ring), 9.71 (s, C₅-H, quinoline ring), 9.83 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 31.22 (1C, CH₃), 121.20-150.23 (3C, paracetamol ring), 141.80 (1C, C-N, quinoline ring), 149.22 (1C, C=N, quinoline ring), 151.16 (1C, C=O, pyrimidine ring); LCMS: *m/z* 569.16 [M⁺]. Anal. Calcd for C₂₉H₂₄ClN₇O₄: C, 61.11; H, 4.24; N, 17.20. Found: C, 61.14; H, 4.22; N, 17.21%.

4-(2-Chloroquinolin-3-yl)-N-(4-((2-hydroxy-5-methylphenyl)diazanyl)phenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVj: Yield 76%. m.p.240-241°C. FT-IR (KBr): 3787.41 (s, O-H), 3210.28 (b, O-H), 619.12 (m, Ar-Cl), 1436.18 (m, C=C), 2841.12 (m, C-H), 1628.78 (s, C=N), 1335.23 (s, C=N), 3601.25 (m, 2° N-H), 1029.63 (s, C-O), 1340.03 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.16 (s, CH₃), 7.21-7.84 (m, 3H, cresol ring), 7.89-8.99 (m, 4H, quinoline ring), 9.02 (s, C₅-H, quinoline ring), 9.88 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 31.10 (1C, CH₃), 120.21-149.56 (3C, cresol ring), 141.88 (1C, C-N, quinoline ring), 149.11 (1C, C=N, quinoline ring), 151.24 (1C, C=O, pyrimidine ring); LCMS: *m/z* 526.15 [M⁺]. Anal. Calcd for C₂₈H₂₃ClN₆O₃: C, 63.82; H, 4.40; N, 15.95. Found: C, 63.80; H, 4.43; N, 15.94%.

4-(2-Chloroquinolin-3-yl)-6-methyl-2-oxo-N-(4-((2,3,4-trihydroxyphenyl)diazanyl)phenyl)-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVk: Yield 73%. m.p.235-236°C. FT-IR (KBr): 3774.74 (s, O-H), 3220.47 (b, O-H), 619.10 (m, Ar-Cl), 1430.28 (m, C=C), 2841.45 (m, C-H), 1621.59 (s, C=N), 1336.32 (s, C=N), 3610.25 (m, 2° N-H), 1027.80 (s, C-O), 1378.78 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.12 (s, CH₃), 7.14-8.23 (m, 4H, quinoline ring), 9.63 (s, C₅-H quinoline ring), 9.77 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 31.22 (1C, CH₃), 120.81-149.42 (4C, pyrogallol ring), 142.23 (1C, C-N, quinoline ring), 152.14 (1C, C=N, quinoline ring), 151.90 (1C, C=O, pyrimidine ring); LCMS: *m/z* 544.13 [M⁺]. Anal. Calcd for C₂₇H₂₁ClN₆O₅: C, 59.51; H, 3.88; N, 15.42. Found: C, 59.48; H, 3.82; N, 15.44%.

Phenyl-3-((4-(2-chloroquinolin-3-yl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxamido)phenyl)-diazanyl)-2-hydroxybenzoate, IVl: Yield 70%. m.p.245-246°C. FT-IR (KBr): 3742.15 (s, O-H), 3202.26 (b, O-H), 616.25 (m, Ar-Cl), 1433.69 (m, C=C), 2841.11 (m, C-H), 1621.45 (s, C=N), 1336.77 (s, C=N), 3605.21 (m, 2° N-H), 1027.41 (s, C-O), 1322.52 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.21 (s, CH₃), 6.88-7.06 (m, 8H, salol ring), 7.10-7.44 (m, 4H, quinoline ring), 8.56 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 16.30 (1C, CH₃), 120.81-130.09 (8C, salol ring), 145.19 (1C, C-N, quinoline ring), 149.36 (1C, C=N, quinoline ring), 154.78 (1C, C=O, pyrimidine ring); LCMS: *m/z* 632.16 [M⁺]. Anal. Calcd for C₃₄H₂₅ClN₆O₅: C, 64.51; H, 3.98; N, 13.28. Found: C, 64.53; H, 3.96; N, 13.31%.

4-(2-Chloroquinolin-3-yl)-N-(4-((2-hydroxyphenyl)diazanyl)phenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVm: Yield 75%. m.p.215-216°C. FT-IR (KBr): 3742.12 (s, O-H), 3798.23 (b, O-H), 759.22 (m, Ar-Cl), 1527.21 (m, C=C), 2998.64 (m, C-H), 1664.21 (s, C=N), 1307.47 (s, C=N), 3568.25 (m, 2° N-H), 1064.23 (s, C-O), 1438.21 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 1.73 (s, CH₃), 6.95-7.80 (m, 4H, phenol ring), 7.10-7.90 (m, 4H, quinoline ring), 9.12 (s, C₅-H, quinoline ring), 11.05 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 32.13 (1C, CH₃), 119.37-145.77 (5C, phenol ring), 143.22 (1C, C-N, quinoline ring), 147.57 (C=N, 1C, quinoline ring), 174.52 (C=S, 1C, pyrimidine ring); LCMS: *m/z* 528.11 [M⁺]. Anal.

Calcd for $C_{27}H_{21}ClN_6O_2S$: C, 61.30; H, 4.00; N, 15.89. Found: C, 61.31; H, 3.98; N, 15.91%.

4-(2-Chloroquinolin-3-yl)-N-(4-((1-hydroxynaphthalen-2-yl)diazenyl)phenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVn: Yield 72%. m.p.218–219°C. FT-IR (KBr): 3730.45 (s, O-H), 3394.83 (b, O-H), 759.98 (m, Ar-Cl), 1527.67 (m, C=C), 2998.62 (m, C-H), 1666.55 (s, C=N), 1307.78 (s, C=N), 3568.43 (m, 2° N-H), 1064.74 (s, C-O), 1438.94 cm^{-1} (m, C-H in-ring); 1H NMR ($CDCl_3$, 400MHz): δ 2.21 (s, CH_3), 6.71-7.92 (m, 6H, naphthol ring), 7.51-8.02 (m, 4H, quinoline ring), 9.77 (s, C_5-H quinoline ring), 13.22 (s, Ar-NH, pyrimidine ring); ^{13}C NMR ($CDCl_3$, 100 MHz): δ 29.91 (1C, CH_3), 123.19–137.81 (6C, naphthol ring), 146.10 (1C, C-N, quinoline ring), 149.43 (C=N, 1C, quinoline ring), 163.32 (C=S, 1C, pyrimidine ring); LCMS: m/z 578.13 [M+]. Anal. Calcd for $C_{31}H_{23}ClN_6O_2S$: C, 64.30; H, 4.00; N, 14.51. Found: C, 64.33; H, 4.04; N, 14.53%.

4-(2-Chloroquinolin-3-yl)-N-(4-((3-hydroxynaphthalen-2-yl)diazenyl)phenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVo: Yield 72%. m.p.217–218°C. FT-IR (KBr): 3731.11 (s, O-H), 3794.45 (b, O-H), 759.47 (m, Ar-Cl), 1527.98 (m, C=C), 2998.62 (m, C-H), 1666.46 (s, C=N), 1307.71 (s, C=N), 3568.50 (m, 2° N-H), 1064.13 (s, C-O), 1438.22 cm^{-1} (m, C-H in-ring); 1H NMR ($CDCl_3$, 400MHz): δ 2.24 (s, CH_3), 6.69-7.90 (m, 6H, naphthol ring), 7.50-8.10 (m, 4-H, quinoline ring), 8.41 (s, C_5-H , quinoline ring), 13.27 (s, Ar-NH, pyrimidine ring); ^{13}C NMR ($CDCl_3$, 100 MHz): δ 29.89 (1C, CH_3), 123.10–137.79 (6C, naphthol ring), 146.08 (1C, C-N, quinoline ring), 149.55 (C=N, 1C, quinoline ring), 163.31 (C=S, 1C of pyrimidine ring); LCMS: m/z 578.13 [M+]. Anal. Calcd for $C_{31}H_{23}ClN_6O_2S$: C, 64.30; H, 4.00; N, 14.51. Found: C, 66.32; H, 4.05; N, 14.53%.

4-(2-Chloroquinolin-3-yl)-N-(4-((2,6-dihydroxyphenyl)diazenyl)phenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVp: $C_{27}H_{19}ClN_6O_3S$, Yield 71%. m.p.227–228°C. FT-IR (KBr): 3731.42 (s, O-H), 3794.62 (b, O-H), 759.19 (m, Ar-Cl), 1527.25 (m, C=C), 2998.53 (m, C-H), 1666.78 (s, C=N), 1307.93 (s, C=N), 3568.08 (m, 2° N-H), 1064.54 (s, C-O), 1438.31 cm^{-1} (m, C-H in-ring); 1H NMR ($CDCl_3$, 400MHz): δ 2.18 (s, CH_3), 6.31-7.89 (m, 3H, resorcinol ring), 7.16-7.94 (m, 4H, quinoline ring), 9.44 (s, C_5-H , quinoline ring), 11.78 (s, Ar-NH, pyrimidine ring); ^{13}C NMR ($CDCl_3$, 100 MHz): δ 32.17 (1C, CH_3), 119.24–145.38 (3C, resorcinol ring), 143.09 (1C, C-N, quinoline ring), 147.21 (C=N,

1C, quinoline ring), 174.61 (C=S, 1C, pyrimidine ring); LCMS: m/z 544.11 [M+]. Anal. Calcd for $C_{27}H_{21}ClN_6O_2S$: C, 59.50; H, 3.88; N, 15.42. Found: C, 59.51; H, 3.86; N, 15.44%.

4-(2-Chloroquinolin-3-yl)-N-(4-((2-hydroxy-3-methylphenyl)diazenyl)phenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVq: Yield 70%. m.p.225–226°C. FT-IR (KBr): 3730.68 (s, O-H), 3794.92 (b, O-H), 759.18 (m, Ar-Cl), 1527.95 (m, C=C), 2998.32 (m, C-H), 1667.16 (s, C=N), 1307.11 (s, C=N), 3568.64 (m, 2° N-H), 1064.33 (s, C-O), 1438.10 cm^{-1} (m, C-H in-ring); 1H NMR ($CDCl_3$, 400MHz): δ 2.13 (s, CH_3), 6.06-7.88 (m, 3H, cresol ring), 7.16-7.97 (m, 4H, quinoline ring), 9.17 (s, C_5-H , quinoline ring), 11.02 (s, Ar-NH, pyrimidine ring); ^{13}C NMR ($CDCl_3$, 100 MHz): δ 32.38 (1C, CH_3), 119.24–145.78 (3C, cresol ring), 143.93 (1C, C-N, quinoline ring), 147.47 (C=N, 1C, quinoline ring), 174.19 (C=S, 1C, pyrimidine ring); LCMS: m/z 542.13 [M+]. Anal. Calcd for $C_{28}H_{23}ClN_6O_2S$: C, 61.93; H, 4.27; N, 15.48. Found: C, 61.96; H, 4.29; N, 15.49%.

4-(2-Chloroquinolin-3-yl)-N-(4-((2,6-dihydroxy-4-methylphenyl)diazenyl)phenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVr: Yield 70%. m.p.230–231°C. FT-IR (KBr): 3730.56 (s, O-H), 3840.95 (b, O-H), 715.25 (m, Ar-Cl), 1472.92 (m, C=C), 2902.56 (m, C-H), 1600.29 (s, C=N), 1290.63 (s, C=N), 3607.82 (m, 2° N-H), 1027.14 (s, C-O), 1467.72 cm^{-1} (m, C-H in-ring); 1H NMR ($CDCl_3$, 400MHz): δ 2.14 (s, CH_3), 6.75 (s, 2H, orcinol ring), 7.12-7.88 (m, 4H, quinoline ring), 8.36 (s, C_5-H , quinoline ring), 10.69 (s, Ar-NH, pyrimidine ring); ^{13}C NMR ($CDCl_3$, 100 MHz): δ 29.22 (1C, CH_3), 119.69–145.85 (2C, orcinol ring), 143.73 (1C, C-N, quinoline ring), 149.48 (C=N, 1C, quinoline ring), 184.23 (C=S, 1C, pyrimidine ring); LCMS: m/z 558.12 [M+]. Anal. Calcd for $C_{28}H_{23}ClN_6O_3S$: C, 60.16; H, 4.15; N, 15.03. Found: C, 60.19; H, 4.12; N, 15.04%.

4-(2-Chloroquinolin-3-yl)-N-(4-((3-formyl-2-hydroxyphenyl)diazenyl)phenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVs: Yield 69%. m.p.235–236°C. FT-IR (KBr): 3730.12 (s, O-H), 3794.67 (b, O-H), 759.91 (m, Ar-Cl), 1527.23 (m, C=C), 2998.12 (m, C-H), 1666.61 (s, C=N), 1307.54 (s, C=N), 3568.71 (m, 2° N-H), 1064.16 (s, C-O), 1438.80 cm^{-1} (m, C-H in-ring); 1H NMR ($CDCl_3$, 400MHz): δ 2.22 (s, CH_3), 6.78-7.66 (m, 3H, salicylaldehyde ring), 7.26-7.89 (m, 4-H, quinoline ring), 9.55 (s, C_5-H , quinoline ring), 11.10 (s, Ar-NH, pyrimidine ring); ^{13}C NMR ($CDCl_3$,

100 MHz): δ 32.01 (1C, CH₃), 119.25–145.14 (3C, salicylaldehyde ring), 143.82 (1C, C-N, quinoline ring), 147.78 (C=N, 1C, quinoline ring), 174.79 (C=S, 1C, pyrimidine ring); LCMS: m/z 556.11 [M⁺]. Anal. Calcd for C₂₈H₂₁ClN₆O₃S: C, 60.38; H, 3.80; N, 15.09. Found: C, 60.35; H, 3.79; N, 15.10%.

3-((4-(4-(2-Chloroquinolin-3-yl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxamido)phenyl)diazanyl)-2-hydroxybenzoic acid, IVt: Yield 70%. m.p.237–238°C. FT-IR (KBr): 3731.85 (s, O-H), 3794.22 (b, O-H), 759.70 (m, Ar-Cl), 1527.13 (m, C=C), 2998.74 (m, C-H), 1666.26 (s, C=N), 1307.66 (s, C=N), 3568.12 (m, 2° N-H), 1064.44 (s, C-O), 1438.56 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.21 (s, CH₃), 6.90–7.89 (m, 3H, salicylic acid ring), 7.12–7.88 (m, 4H, quinoline ring), 9.46 (s, C₅-H, quinoline ring), 11.30 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 32.02 (1C, CH₃), 119.30–145.11 (3C, salicylic acid ring), 143.66 (1C, C-N, quinoline ring), 147.61 (C=N, 1C, quinoline ring), 174.12 (C=S, 1C, pyrimidine ring); LCMS: m/z 572.10 [M⁺]. Anal. Calcd for C₂₈H₂₁ClN₆O₄S: C, 58.69; H, 3.69; N, 14.67. Found: C, 58.71; H, 3.70; N, 14.69%.

N-(4-((5-Acetamido-2-hydroxyphenyl)diazanyl) phenyl)-4-(2-chloroquinolin-3-yl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVu: Yield 69%. m.p.245–246°C. FT-IR (KBr): 3730.38 (s, O-H), 3794.53 (b, O-H), 759.61 (m, Ar-Cl), 1527.10 (m, C=C), 2998.80 (m, C-H), 1666.11 (s, C=N), 1307.24 (s, C=N), 3568.20 (m, 2° N-H), 1064.79 (s, C-O), 1438.90 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.20 (s, CH₃), 6.56 (s, 1H, paracetamol ring), 7.24–7.61 (m, 4H, quinoline ring), 9.10 (s, C₅-H, quinoline ring), 11.03 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 32.23 (1C, CH₃), 119.92–145.12 (3C, paracetamol ring), 143.01 (1C, C-N, quinoline ring), 147.40 (C=N, 1C, quinoline ring), 174.14 (C=S, 1C, pyrimidine ring); LCMS: m/z 585.13 [M⁺]. Anal. Calcd for C₂₉H₂₄ClN₇O₃S: C, 59.43; H, 4.13; N, 16.73. Found: C, 59.41; H, 4.12; N, 16.70%.

4-(2-Chloroquinolin-3-yl)-N-(4-((2-hydroxy-5-methylphenyl)diazanyl)phenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVv: Yield 69%. m.p.240–241°C. FT-IR (KBr): 3730.21 (s, O-H), 3794.10 (b, O-H), 759.19 (m, Ar-Cl), 1527.87 (m, C=C), 2998.21 (m, C-H), 1666.18 (s, C=N), 1307.12 (s, C=N), 3568.97 (m, 2° N-H), 1064.91 (s, C-O), 1438.08 cm⁻¹

(m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.26 (s, CH₃), 6.40–7.70 (m, 3H, cresol ring), 7.10–7.81 (m, 4H, quinoline ring), 9.52 (s, C₅-H, quinoline ring), 11.01(s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 32.30 (1C, CH₃), 119.44–145.13 (3C, cresol ring), 143.23 (1C, C-N, quinoline ring), 147.78 (C=N, 1C, quinoline ring), 174.54 (C=S, 1C, pyrimidine ring); LCMS: m/z 542.13 [M⁺]. Anal. Calcd for C₂₈H₂₃ClN₆O₂S: C, 61.93; H, 4.27; N, 15.48. Found: C, 61.96; H, 4.24; N, 15.46%.

4-(2-Chloroquinolin-3-yl)-6-methyl-2-thioxo-N-(4-((2,3,4-trihydroxyphenyl)diazanyl)phenyl)-1,2,3,4-tetrahydropyrimidine-5-carboxamide, IVw: Yield 68%. m.p.250–251°C. FT-IR (KBr): 3731.13 (s, O-H), 3894.15 (b, O-H), 723.27 (m, Ar-Cl), 1510. (m, C=C), 2998.47 (m, C-H), 1689.98 (s, C=N), 1308.42 (s, C=N), 3268.11 (m, 2° N-H), 1064.02 (s, C-O), 1438.05 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400MHz): δ 2.29 (s, CH₃), 7.08–7.60 (m, 4H, quinoline ring), 9.21 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 16.30 (1C, CH₃), 126.24–133.03 (4C, pyrogallol ring), 147.65 (1C, C-N, quinoline ring), 152.29 (C=N, 1C, quinoline ring), 180.75 (C=S, 1C, pyrimidine ring); LCMS: m/z 560.10 [M⁺]. Anal. Calcd for C₂₇H₂₁ClN₆O₄S: C, 57.81; H, 3.77; N, 14.89. Found: C, 57.85; H, 3.74; N, 14.85%.

Phenyl -3-((4-(4-(2-chloroquinolin-3-yl)-6 methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxamido)phenyl)diazanyl)-2-hydroxybenzoate, IVx: Yield 68%. m.p.256–257°C. FT-IR (KBr): 3730.77 (s, O-H), 3794.41 (b, O-H), 759.56 (m, Ar-Cl), 1527.81 (m, C=C), 2998.97 (m, C-H), 1666.10 (s, C=N), 1307.62 (s, C=N), 3568.71 (m, 2° N-H), 1064.33 (s, C-O), 1438.13 cm⁻¹ (m, C-H in-ring); ¹H NMR (CDCl₃, 400 MHz): δ 2.24 (s, CH₃), 6.80–7.91 (m, 8H, salol ring), 7.02–7.71 (m, 4H, quinoline ring), 9.10 (s, C₅-H, quinoline ring), 10.80 (s, Ar-NH, pyrimidine ring); ¹³C NMR (CDCl₃, 100 MHz): δ 32.33 (1C, CH₃), 119.39–145.80 (8C, salol ring), 143.11 (1C, C-N, quinoline ring), 147.90 (C=N, 1C, quinoline ring), 174.22 (C=S, 1C, pyrimidine ring); LCMS: m/z 648.13 [M⁺]. Anal. Calcd for C₃₄H₂₅ClN₆O₄S, 62.91; H, 3.88; N, 12.95. Found: C, 62.94; H, 3.84; N, 12.97%.

Evaluation of antimicrobial activity

The antibacterial and antifungal activities were examined by agar well diffusion method³⁵ using Mueller-Hinton agar media and sterilized potato

dextrose agar media, respectively. The antimicrobial activity of synthesized compounds was tested against several microorganisms: Gram (+) ve bacteria (*Staphylococcus aureus* and *Bacillus subtilis*), Gram (-) ve bacteria (*Escherichia coli*) and Fungi (*Candida albicans* and *Aspergillus niger*). All the tested compounds were dissolved in chloroform and diluted to get final concentration of 50 µg/mL. Under identical conditions, ampicillin (50 µg/mL), chloramphenicol (50 µg/mL) and fluconazole (50 µg/mL) were used as standard drug. Chloroform was used as a control. Antimicrobial activity was determined by measuring the zone of inhibition.

Upon assessing the antimicrobial activity data (Table 1), it has been observed that the newly synthesized compounds displayed weak to excellent activity against all the tested bacterial and fungal strains. It has been discovered that structural

differences, modifications and functional group/s existing on phenol ring attached to azo group could possibly be the cause for the variety in antimicrobial activity. Compounds **IVo** and **IVc** bearing 2-naphthol attached to azo group revealed excellent and very good antifungal activity respectively against *C. Albicans*. Compounds **IVb**, **IVd**, **IVn** and **IVp** showed good antifungal activity against *C. Albicans*. The compound **IVb** also exhibited good antibacterial activity against *S. aureus* and *B. subtilis*. **IVb** and **IVc** showed good antifungal activity against *A. niger*. Every other synthesized compound revealed mild to moderate activity against the tested microorganisms.

Dyeing assay

Dyeing on cotton fabric

The fabric was dyed with 2% dye (calculated on the weight of fabric), 1% NaCl (as dispersing agent),



















Table 1 — Antimicrobial activity of the synthesized compounds

Compd	Conc (µg/mL) in chloroform	Zone of inhibition in mm*				
		Antibacterial activity			Antifungal activity	
		<i>S. aureus</i>	<i>E. coli</i>	<i>B. subtilis</i>	<i>A. niger</i>	<i>C. albicans</i>
IVa	50	8	10	10	7	7
IVb	50	19	6	17	21	19
IVc	50	12	8	12	22	25
IVd	50	7	6	8	20	19
IVe	50	11	6	10	7	8
IVf	50	12	7	7	7	7
IVg	50	10	7	7	8	7
IVh	50	8	7	8	7	8
IVi	50	7	8	10	7	8
IVj	50	7	8	9	9	9
IVk	50	8	10	9	8	11
IVl	50	9	10	7	7	13
IVm	50	7	7	7	10	8
IVn	50	11	15	13	11	20
IVo	50	12	14	15	12	27
IVp	50	7	8	7	7	21
IVq	50	7	9	9	7	8
IVr	50	7	8	7	7	8
IVs	50	8	8	7	8	7
IVt	50	8	7	7	7	8
IVu	50	7	7	8	7	7
IVv	50	7	8	9	7	7
IVw	50	9	7	9	8	7
IVx	50	7	7	7	7	8
Ampicillin	50	26	25	25	—	—
Chloramphenicol	50	20	21	24	—	—
Fluconazole	50	—	—	—	26	25
Control (Chloroform)	50	5	5	6	6	6

*Diameter of well (bore size) - 6 mm

kept at a liquor ratio of 20:1. The process was started at 60°C and the temperature was then raised to 130°C for 30 min and maintained for 1 h. After cooling, the fabric was taken out and treated with a solution of 2%

sodium bisulphite, 2% NaOH and 0.1% NaCl at 70°C for 30 min. Finally, the fabric was rinsed and dried at 60°C³⁶. Table 2 shows the dyeing assay of cotton fabric.

Compd	Dye applied cotton fabric	Colour Fastness	Rubbing Fastness
IVa			
IVb			
IVc			
IVd			
IVe			
IVf			
IVg			
IVh			
IVi			
IVj			
IVk			
IVl			
IVm			
IVn			

























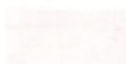








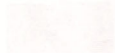
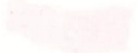







(Contd.)

Compd	Table 2 — Dyeing assay of cotton fabric (Contd.)		
	Dye applied cotton fabric	Colour Fastness	Rubbing Fastness
Ivo			
IVp			
IVq			
IVr			
IVs			
IVt			
IVu			
IVv			
IVw			
IVx			
	Reference Cotton Cloth		































Dyeing on silk fabric

The fabric was dyed with 2% dye (calculated on the weight of fabric), 1% Na_2CO_3 (as dispersing agent), kept at a liquor ratio of 20:1. The process was started at 60°C and the temperature was then raised to 130°C for

30 min and maintained for 1 h. After cooling, the fabric was taken out and treated with a solution of 2% urea, 2% NaOH and 0.1% NaCl at 70°C for 30 min. Finally, the fabric was rinsed and dried at 60°C. Table 3 shows the dyeing assay of silk fabric.

Compd	Table 3 — Dyeing assay of silk fabric		
	Dye applied Silk fabric	Colour Fastness	Rubbing Fastness
IVa			
IVb			
IVc			
IVd			
IVe			
IVf			
IVg			
IVh			
IVi			
IVj			
IVk			
IVl			
IVm			
IVn			

(Contd.)

Compd	Table 3 — Dyeing assay of silk fabric (<i>Contd.</i>)		
	Dye applied Silk fabric	Colour Fastness	Rubbing Fastness
Ivo			
IVp			
IVq			
IVr			
IVs			
IVt			
IVu			
IVv			
IVw			
IVx			

Reference Silk Cloth

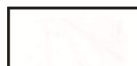


Table 4 — Fastness properties of cotton and silk fabrics

Compd	Cotton		Silk	
	Colour fastness	Rubbing fastness	Colour fastness	Rubbing fastness
IVa	4	5	4	4
IVb	5	4.5	5	5
IVc	5	5	2	3
IVd	3	3	5	5
IVe	5	5	5	5
IVf	5	4	5	5
IVg	3	5	5	5
IVh	4	3.5	2	3
IVi	4	4	3	3
IVj	4	5	5	5
IVk	3	5	4	4
IVl	2	3	3	4
IVm	4	4.5	5	5
IVn	4.5	5	3.5	3.5
IVo	3.5	4.5	5	5
IVp	5	4.5	4	4
IVq	4.5	5	5	4
IVr	4	4	5	5
IVs	4.5	4.5	4	4.5
IVt	4	4	5	5
IVu	4	3.5	4	4
IVv	4	4	4	4
IVw	5	5	3.5	4
IVx	3.5	4	5	4

Table 5 — Compounds with excellent fastness property

Cotton		Silk	
Colour fastness	Rubbing fastness	Colour fastness	Rubbing fastness
IVb, IVc, IVe, IVf, IVe, IVw	IVa, IVc, IVe, IVg, IVj, IVk, IVn, IVq, IVw	IVb, IVd, IVe, IVf, IVg, IVj, IVm, IVo, IVq, IVr, IVt, IVx	IVb, IVd, IVe, IVf, IVg, IVj, IVn, IVo, IVr, IVt

Fastness properties

Rubbing fastness

The dyed cotton fabric was placed on the base of crockmeter, so that it rested flatly on the abrasive cloth with its long dimension in the direction of rubbing. A square of white testing cloth was allowed to slide on the tested fabric back and forth 20 times by making 10 complete turns of the crank. For the wet rubbing test, the testing squares were thoroughly immersed in distilled water. The rest of the procedure was the same as in the dry test. The staining on the white testing cloth was assessed according to the gray scale³⁶ (Table 4).

Colour fastness

A specimen of dyed cotton fabric was stitched between two pieces of undyed cotton fabric all of equal length and then washed at 50°C for 30 min. After soaping treatment, the specimen was removed, rinsed twice in cold water and then in cold running tap water. It was then squeezed and dried in air at a temperature not exceeding 60°C. The staining on the undyed adjacent fabric was assessed according to the gray scale³⁶ (Table 4).

The remarkable degree of levelness after dyeing indicated penetration into an affinity of the newly synthesized azo dyes for the fabric matrix. The dyeing of cotton and silk fabrics indicated various hues with very good intensity in depth. All dyed fabrics showed good to excellent water and rubbing fastness properties. Compounds with excellent fastness properties on fabrics may lead to their large-scale use in textile industry for dyeing clothes in future as well as some unique identification application to detect microbes using colouring properties. Newly synthesized azo dyes with excellent fastness properties are mentioned in Table 5.

Conclusion

It can be indisputably concluded from the results of Table 1 that structural and functional group varieties on different positions of the targeted compounds influenced their antimicrobial activity. All newly synthesized compounds were active against all the tested microbes. On account of the results of biological activity we can state that among all the phenol derivatives, 1-naphthol and 2-naphthol

attached to azo group are most distinctive derivatives because their pre-eminent antimicrobial activity, especially antifungal activity. Concluding remarks from the dyeing results of Table 4 and Table 5 determine that the majority of the synthesized compounds revealed excellent dyeing performance as well as fastness properties due to better dispersibility and intrinsic conjugation of N-heterocycles present in the structure. Naphthol derivatives are found to be most promising motifs with excellent microbial as well as fastness properties on fabrics which may lead to their large-scale use in textile and pharmaceuticals industries in the future.

Supplementary Information

Supplementary information is available in the website <http://nopr.niscares.in/handle/123456789/58776>.

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References

- Chaudhuri S, Ghosh A & Chattopadhyay S K, in *Green Synthetic Approaches for Biologically Relevant Heterocycles*, Vol. 2 2nd Edn, edited by Brahmachari G (Elsevier, Amsterdam), p. 617 (2021).
- Martins M A P, Frizzo C P, Moreira D N, Zanatta N & Bonacorso H G, *Chem Rev*, 108 (2008) 2015.
- Kerru N, Gummidi L, Maddila S, Gangu K K & Jonnalagadda S B, *Molecules*, 25 (2020) 1909.
- Shah P J, *Indian J Chem*, 60B (2021) 1223.
- Desai N C, Harsora J P, Patel B Y & Jadeja K A, *Indian J Chem*, 56B (2017) 976.
- Desai N C, Patel B Y & Dave B P, *Med Chem Res*, 26 (2017) 109.
- Desai N C, Bhatt N, Dodiya A, Karkar T, Patel B Y & Bhatt M, *Res Chem Intermed*, 42 (2016) 3039.
- Desai N C, Patel B Y, Jadeja K A & Dave B P, *NAPDD*, 1 (2017) 555.
- Trivedi H D, Joshi V B & Patel B Y, *Anal Chem*, 12 (2022) 1.
- Akhtar J, Khan A A, Ali Z, Haider R & Yar M S, *Eur J Med*, 125 (2017) 143.
- Saini M S, Kumar A, Dwivedi J & Singh R, *Int J Pharm Sci Res*, 4 (2013) 66.
- Kalaria P N, Karad S C & Raval D K, *Eur J Med*, 158 (2018) 917.
- Garuti L, Roberti M & Pizzirani D, *Mini Rev Med Chem*, 7 (2007) 481.
- Sharma S, Kumar D, Singh G, Monga V & Kumar B, *Eur J Med*, 200 (2020) 112438.
- Mermer A, Keles T & Sirin Y, *Bioorg Chem*, 114 (2021) 105076.
- Chand K, Rajeshwari, Hiremath A, Singh M, Santos M A & Kerri R S, *Pharmacol Rep*, 69 (2017) 281.
- Sanemitsu Y & Kawamura S, *Pestic Sci*, 33 (2008) 175.
- Balasubramanian M & Keay J G, in *Comprehensive Heterocyclic Chemistry II*, Vol. 5, edited by Katritzky A R, Rees C W & Scriven E F V (Pergamon, Oxford), p.245 (1996).
- Üstün E, Özdemir N & Şahin N, *J Coord Chem*, 74 (2021) 3109.
- Obot I B, Onyechu B I & Ahmad M, *US Patent* 0318243, Oct 8, 2020.
- Plajer A J & Williams C K, *Angew Chem Int Ed*, 61 (2022) 1.
- Khattab T & Rehan M, *Egypt J Chem*, 61 (2018) 989.
- Navarro A & Sanz F, *Dyes Pigm*, 40 (1999) 131.
- Georgiadou K L & Tsatsaroni E G, *Dyes Pigm*, 50 (2001) 93.
- Mikroyannidis J A, Tsagkournos D V, Sharma S S, Kumar A, Vijay Y K & Sharma G D, *Sol Energy Mater Sol Cells*, 94 (2010) 2318.
- Zhang D, Zhang M, Liu Z, Yu M, Li F, Yi T & Huang C, *Tetrahedron Lett*, 47 (2006) 7093.
- Russev M M & Hecht S, *Adv Mater*, 22 (2010) 3348.
- Li X, Wu Y, Gu D & Gan F, *Dyes Pigm*, 86 (2010) 182.
- Bhatti H S & Seshadri S, *Color Technol*, 120 (2004) 151.
- Desai N C, Vaghani H V, Patel B Y & Karkar T J, *Indian J Pharm Sci*, 80 (2018) 242.
- Desai N C, Vaghani H V, Karkar T J, Patel B Y & Jadeja K A, *Indian J Chem*, 56B (2017) 438.
- Patel B Y, Karkar T J & Bhatt M J, *Eur Chem Bull*, 10 (2021) 13.
- Desai N C, Patel B Y & Dave B P, *Int Lett Chem Phys Astron*, 69 (2016) 87.
- Meth-Cohn O & Taylor D L, *Tetrahedron Lett*, 34 (1993) 3629.
- Shah P J, *Heterocycl Lett*, 6 (1) (2016) 111.
- Vidya V G & Sadasivan V, *Asian J Chem*, 30 (2018) 2049.