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# One-Pot Synthesis, Characterization, and Antibacterial Activity of Mono Azo-Disperse Dyes bearing Bis-α-Aminophosphonates moiety, and their application in Polyester Printing

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**Abstract**

The synthesis of isatin hydrazide have been done in high yields at room temperature starting from indoledione (Isatin) and hydrazine hydrate. A one –pot three component Kabachnik Fields reaction was used to synthesize mono and di-α-aminophosphonate system using mono and dialdehydes containing isatin hydrazide and triethyl phosphite in methylene chloride⁄ DMF mixture using Cupper triflate as Lewis acid catalyst. The structure of those aminophosphonate derivatives has been confirmed by elemental analysis and by FT-IR, NMR spectral analysis. Mono- α-aminophosphonates azo dyes have additionally been efficaciously applied to make printing pastes for polyester fabric. The color strength and fastness properties of the printed fabrics- to washing, perspiration, and light- demonstrated moderate to excellent results. Those derivatives as well as their printed samples showed high antibacterial activities against Staphylococcus aureus (Gram +ve) and Pseudomonas auroginosa (Gram -ve) and Candida albicans (yeast) at low concentrations (2.5–10 mg /mL).

Key words: Isatin hydrazide; ; Cupper triflate ; mono and bis aminophosphonates; Textile printing ; Azo disperse dyes ; antibacterial activity

## 1. Introduction

Technology development and revolution in the field of health care and environmental protection are the most important issues for the human prosperity in 21st. century. For this purpose, energy-saving and environmentally friendly technologies and starting methods should be developed. More directional synthetic organic chemistry, a type of green chemistry, plays an important role in overcoming these challenges. The traditional applications of azo compounds are as dyes in many fields such as textile coloration, leather and other materials coloration , and in biomedical and organic synthesis application.

Despite few articles reported that azo compounds having antimicrobial activities. The structural motif of 2-indoline is important ring system in heterocyclic chemistry due to is presence in many natural products and biologically active compounds [1-3]. Phosphonates and their related derivatives considered potential bioisosteres of the corresponding carboxylic acids [4]. Therefore, the incorporation of phosphonyl groups into heterocyclic systems has resulted in an important class of organophosphorus compounds that have attracted the attention of both industrial, and medical chemists [5-12].

Recently, α-Aminophosphonates gained great important motif interest among the organophosphorus compounds in medicinal chemistry due to their biological activities, and due to its structural similarity to the corresponding α-amino acid [13, 14]. The biological activity of α-aminophosphonates described in the literature as anti-cancer agents [15, 16] and antibacterial agents [17]. In addition, these compounds used in a broad application as antifungal, anticancer, antiviral agents [18, 19] and herbicides [20, 21].

One of the most well known or most useful synthetic approaches to α-aminophosphonate is the Kabachnik-Fields method. This is a one-pot, three-component condensation of primary or secondary amines, aldehydes or ketones and diethyl phosphite in the presence of acidic or basic catalysts. In some cases, Lewis acid catalysts are inefficient or weak in reaction mixtures due to their low activity[22-24].

Isatin (1-H-indole-2,3-dione) and its subsidiaries, have a wide extend of pharmacological properties. Isatin-based hydrazone has been distinguished as an

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inhibitor of the protein tyrosine phosphatase, which plays a part in cell signaling, cell expansion, separation and movement [24]. Enhan cement and synergistic impacts of the isatin- hydrazide moiety by side chain alteration (C3), has moreover been already detailed to be related with motion) and / or other basic themes [25, 26]. Adding α-aminophosphate group as example to isatin moiety has been appeared to have a critical synergistic impact on organic action.

Based on above-information, the aim of the work displayed is to synthesize a few modern powerful and naturally dynamic hetero-cycles. We planned to design and development of a few unused derivatived of different substituted isatin hydrazide cross breed with α-aminophosphonates bearing azo chromophore and assessed their antibacterial movement for both Gram-positive and Gram-negative microscopic organisms.

## 2. Experimental

*2.1. Instrumentation & Methodology:*

$^1$HNMR measurements were performed in DMSO-$d_6$ with and without $D_2O$ relative to the standard tetramethyl silane (TMS) at ECA-500 II MHz spectrotometer. Infrared (I.R) spectral analysis have been recorded using Nicolet iS10 Thermo Fisher FTIR spectrometer. Melting points have been measured using Start melting point instrument. All synthesized products have been tested by thin layer chromatography using TLC silica gel $F_{254}$ plates [27].

*2.2. Solvents, Chemicals, and Auxiliaries*

Starting materials, glutaraldehyde, isatin, hydrazine hydrate, and triethy phosphite from Sigma Aldrich chemicals. Solvents such as, dichloromethane, hexane, dimethylformamide and diethyl ether were utilized without purification. Hydrochloric acid (HCl, 35.4%), sodium nitrite ($NaNO_2$, 98%), sodium dihydrogen phosphate (98.0%) . Sodium lignosulphonate (Anionic dispersing agent) (99.5 %) were from LOBA Chemie. Sodium acetate (AcONa, 99%) was from East-Chem. Thickener (commercial engineered thickener) acrylate copolymers and Lyprint (sodium salt of nitrobenzene sulfonic acid) provided by BASF Company.

*2.3. Fabric*

Polyester fabric(150 g/m$^2$) were provided by Misr Spinning and Weaving Company, El- Mahalla, Egypt.

*2.4. Synthesis of Isatin Hydrazide [4]*

The reaction product 4 was prepared according to previous literature methods [27]. Equi-molar amounts (10 mmol) of isatin and 85 percent hydrazine hydrate were enthusiastically mixed for 3 hours at room temperature in anhydrous ethanol (10 mL). The precipitate was separated off, dried, and recrystallized from ethanol [27].

*2.5. Synthesis of bis-α-amino phosphonate derivatives [4a-b]*

Glutaraldehyde (0.12 mL, 1 mmol) (or Erphthalaldehyde135 mg, 1 mmol), 2 mmol of Isatine Hydrazide, Triethylphosphite 2 mmol dissolved in 6ml methylene chloride/DMF 1:, Lewis acid catalytic copper(II) Triflat 3.6 mg 10% mol in argon atmosphere was added at the appropriate time. The reaction was monitored using TLC. The reaction mixture was diluted with dichloromethane (10 ml). Then, the combined mixture was treated with water (210 ml) and the organic layer was separated and dried over anhydrous $Na_2SO_4$. The organic layer was then concentrated using a rotary evaporator to give a crude product. The product purified by column chromatography on silica gel (nHexane/EtOAc).

*Tetraethy(1,5-bis((z)-2-(2-oxoindolin-3-yiedene)hydrazinyl)pentane)bis-phosponate) [4a]*

Yield 60% viscous oily brown, mp=(130-132°C); IR (cm$^{-1}$) = 3257, 3144, 2935, 2367, 1655, 1283. $^1$H NMR (DMSO-$d_6$, ppm): δ = 7.90-7.83 (m, 4 H aromatic), 7.40 (q, J = 6.17 Hz, 2 H aromatic), 7.22 (t, J = 6.50 Hz, 2H aromatic), 6.85 (s,1H, -NH), 3.98 (q, J = 7.25 Hz, 8 H, $CH_2$) 3.56 (t, J = 4.3 Hz , 2H), 1.84 (T, J = 4.19 Hz, 4 H, aliphatic), 1.68-1.55 m, 2H, $CH_2$), 1.20 (t, J = 6.3 Hz, 12 H). Analysis of $C_{29}H_{40}N_6O_8P_2$: C, 52.57; H 6.08; N, 12.68; P (calculated) , 9.35 C, 52.80; H, 6.33; N, 12.15; P 9.12 (found).

*Tetraethyl((z)-1,4-phenylene-bis-2-(2-oxoindolin-3-ylidene)hydrazinyl) methylene)) bis -phosphonate [4b] .*

Yield 83 % oily viscous brown, m.p. = (160-162ºC); IR (cm$^{-1}$) = 3257,3144, 2935, 2367,1655,1240. $^1$HNMR (DMSO-d6, ppm): δ = 9.96 (s,1H, -NH), 8.10 (s,1H, -NH), 7.89 ( d, J = 4.3 Hz, 4H aromatic), 7.63 (q, J = 6.25 Hz,2H aromatic), 7.48 (q, J = 4.3 Hz, 2H aromatic), 7.30 (s, 4H aromatic), 4.71(s, 2H), 3.98 ( q, J = 3.25 Hz, 8H, $CH_2$) , 1.19 ( t, J = 6.3 Hz, 12H). Analysis calculated for $C_{32}H_{38}N_6O_8P_2$: C, 55.17; H, 5.50; N, 12.06; P, 8.89 found; C, 55.11; H, 5.89; N, 12.70; P, 8.12.

*(Z)Diethyl(2-hydroxyphenyl)-5-Phenyldiazenyl(2-(2oxoindolinylidene) hydrazinyl) Methylphosphonate [5a ]*

Brown solid; (64%); mp (154-156°C); IR (cm$^{-1}$) = 3199.11, 2975.14, 2666.12, 2333.14, 1715.22, 1514). $^1$HNMR (DMSO-$d_6$, ppm): δ = 9.91 (s, 1H, indole NH); 8.06 (s, 1H, NH), 7.94 (m, 3H, aromatic), 7.73 (t, J = 8 Hz 1H, Aromatic), 7.60 (d, J = 6.7 Hz, 2H, aromatic), 7.33 (d, J = 6.7 Hz, 2H, aromatic), 7.21 (d, J = 8.7 Hz, 2H), 7.096.96 ( m, 2H), 5, 06 (s, 1H, OH), 4.40 (q, J = 7.13Hz, 4H). ), 3.95 (s, 1H), 1.30 (t, J = 7.11 Hz, 6H). $^{13}$C-NMR ($CDCl_3$) δ: 18.44(2C),

58.59, 59.11(2C), 114.60, 117.22, 119.77(2C), 122.32(2C), 123.31(2C), 123,90, 128.57, 129.37(2C), 131.42(2C), 133.15, 141.33, 152.33, 154.21, 157.18, 169.61. Analysis calculated for $C_{25}H_{26}N_5O_5P$: C, 59.17; H, 5.16; N, 13.80; found; C, 59.77; H, 5.06; N, 13.45.

*(Z)-Diethyl(2-hydroxyphenyl)-5((E)-4-methoxyphenyldiazenyl(2-(2-oxoindolin-ylidene)hydrazinyl)methylphosphonate [5b]*

Yellow -solid; ( 70 %); m.p (188-190 ºC); IR ($cm^{-1}$) = 3150.17, 2814.17 , 2801.18, 2390.23, 1755.21,1507.33). 1H-NMR (DMSO-d6, ppm): δ = 10.11 (s, 1H, indole NH); 9.23 (s, 1H, -NH), 7.90 (m, 1H, aromatic), 7.73 (t, J= 8 Hz 2H, aromatic),7.70 (d, J= 6.7 Hz , 2H,aromatic),7.38 (d, J= 6.7 Hz , 2H,aromatic), 7.33 (d, J=8 .7Hz 2H,), 7.04-6.99 (m, 2H), 4.43 ( q, J = 7.13 Hz, 4H), 3.98 ( s, 1H), 3.77 ( s, 1H), 1.30 ( t, J = 7.11 Hz, 6H). Analysic for $C_{26}H_{28}N_5O_6P$: (calculated) C, 58.10; H, 5.25; N, 13.03 ; (found) C, 58.71; H, 5.55; N, 13.51.

*(Z)-Diethyl(2-hydroxyphenyl)-5((E)-4-chlorophenyldiazenyl(2-(2-oxoindolin-ylidene)hydrazinyl)methylphosphonate [5c]*

Yellow -solid; (68%); m.p (172-175ºC); IR ($cm^{-1}$) = 3050.12, 2714.23 , 2790.58, 2299.30, 1743.01,1494.83). 1H-NMR (DMSO-d6, ppm): δ = 10.02 (s, 1H, -NH of indole); 8.97 (s, 1H, -NH), 7.91 (m, 1H, aromatic), 7.71 (t, J= 8 Hz 2H, aromatic), 7.68 (d, J= 6.7 Hz , 2H,aromatic),7.34 (d, J= 6.7 Hz , 2H,aromatic), 7.30 (d, J= 8 .7Hz 2H,), 7.08-7.02 (m, 2H), 4.30 ( q, J = 7.13 Hz, 4H), 3.88 ( s, 1H), 1.27 ( t, J = 7.11 Hz, 6H). Analysis for $C_{25}H_{25}N_5O_6PCl$: Calculated; C, 55.41; H, 4.65; N, 12.92; found; C, 55.71; H, 4.55; N, 12.51.

*(Z)-Diethyl(1,5-dimethyl -3-oxo-2-phenylpyrazolidin-4-yl) 2-oxoindolin-3-ylidene hydrazinyl methyl phosphonate [5d]*

Yellow semi-solid; (,70 %); m.p (188-190ºC); IR ($cm^{-1}$) = 3350,3144, 2985, 2663,1658,1233. 1HNMR (DMSO-d6, ppm): δ = 9.60 (s,1H, -NH), 7.96-7.91 (m,2H aromatic), 7.51-7.45 (m, 1H, aromatic), 7.32-7.27 (m. 5H, aromatic), 6.80 ( t, J = 7.13 Hz, 1H), 3.97 ( q, J = 6.25 Hz, 4H) 3.19 ( s, 3H), 3.11 ( s, 3H,$CH_3$) 1.79 ( t, J = 5.19Hz, 6H), 1.20 ( s,3H, $CH_3$). Analysis for $C_{24}H_{30}N_5O_5P$: Calculated; C, 57.51; H, 6.05; N, 14.02; found; C, 57.80; H, 5.98; N, 14.55.

*2.6. Fastness Properties*

For evaluation of the fastness properties of the printed samples to rubbing, washing, perspiration, light and sublimation ; wash fastness test assessed according with the standard method ISO 105-C06 B2S [28] (4.0 g/L of ECE detergent, 1 g/L of sodium perborate, 25 metallic balls) at 50 °C for 30 min and at a liquor ratio of 50:1; the rubbing fastness test according of ISO 105X12 [29] and fastness to perspiration according with ISO 105-E04 [30]; the sublimation fastness test done using of a fixo-meter at 180 and 210 °C according with ISO 105P01 [31]; the light fastness test assessed using a xenon arc lamp according to ISO 105-B02 [32].

*2.7. Determination of antibacterial activity*

The prepared dye samples separately used to check its antibacterial pastime in opposition to human pathogens Gram-positive bacteria particularly Staphylococcus aureus ATCC 6598, Bacillus subtilis ATCC6633 and Streptococcus faecalis and Gram-negative bacteria particularly Escherichia coli ATCC8739, Pseudomonas aeruginosa ATCC9027 and Candida albicans.

Each bacterial strain was inoculated in to L. B. broth media and incubated at 35±2ºC for 24h., after that, 100 µL of every bacterial strain (1x108 CFU/mL) was seeded onto Muller Hinton agar media in aseptic circumstance and poured into petri dishes. Final concentration of 2.5–10 mg mcg/disc had been aseptically prepared and placed on to the surface of seeded Muller Hinton agar plate and saved with inside the fridge for 2 h. prior to incubation at 35±2ºC for 24h. After incubation, the plates had been tested and the diameters of inhibition zones (mm) had been recorded. The antibacterial behaviors of loaded compounds had been analyzed after 3 cycles for the subsequent treatments; (A) popular disc of ciprofloxacin (Antibacterial agent) as positive control, (B) the synthesized compounds symbolized **4a-5c** [33].

*2.8. Antimicrobial test for the printed fabrics*

The antimicrobial activity of the printed fabrics was tested for gram negative and positive bacteria according to AATCC Test Method 1001999 to test printed fabrics to demonstrate bacteriostatic and antibacterial activities of the diffusible antimicrobial agents on treated textile materials against two different types of bacterial stains. Following this procedure, swatches (ca. 36 mg) of the printed fabrics coded (5a-c) were tested with 36 µL of bacterial inoculum (105 CFU of bacteria (Gram-positive or Gram-negative)). The inoculated swatches were then placed in 10 ml of sterilized water for 5 h as the contact time. After the determined time, 40 L of sterilized water was placed on a nutrient agar and incubated at 37 °C for 18 h. Viable bacterial colonies on the agar plate were counted and the percentage reduction in bacterial count was calculated using the equation:

$$\%R = ((B-A)/B) \times 100$$

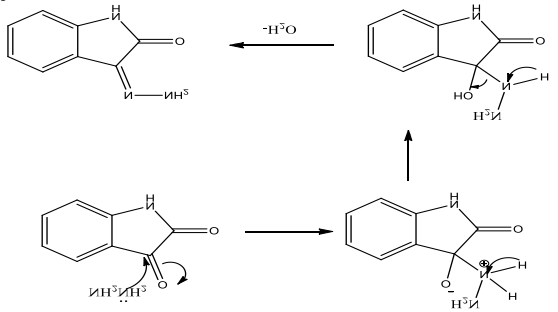
Where R is the percent reduction of the bacterium; B is the count of bacterial colonies from untreated fabric and A is the count of bacterial colonies from treated fabrics [34].

## 3. Results and discussion

In this study, we investigated the synthesis and antibacterial evaluation of the newly synthesized heterocyclic azo- Mono and Bis α-aminophosphonates System. The synthetic approaches

for isatin- α- aminophosphonates conjugates represented in the schemes below (**Scheme1**) . The starting isatin hydrazide 4 was obtained in good yields according to the reaction of indole-2,3-dione (isatin) with hydrazine hydrate in Ethanol, using glacial acetic acid as a catalyst at room temperature (3 hrs.). The structure of compound **4** was concluded based on full mass and $^1$H NMR spectral data. Accordingly, an optimization design of the aminophosphonate structure was performed to refine and optimize the properties of catalytic materials and the optimal operating conditions to react compound **4** with mono- and dibenzaldehyde and triethyl phosphite.

Aminophosphonate derivatives (**4,5**) have been obtained via the Kabachnik-Fields technique by means of reacting 3-hydrazonoindolin-2-one (1), substituted monoaldehydes consisting of azosaldehyde and 4-antipyrine carboxaldehyde, and dialdehydes consisting of gluotraldehyde and terphthaldehyde with triethyl phosphite in a one-pot One-pot synthesis step in the presence of copper triflate as a catalyst. The catalyst turns on imine formation, facilitating the addition of phosphite to provide a phosphonium intermediate. This phosphonium intermediate reacts with water to produce the compounds (**4, 5a-c**). The synthesis mechanism is shown in **Scheme 3**.



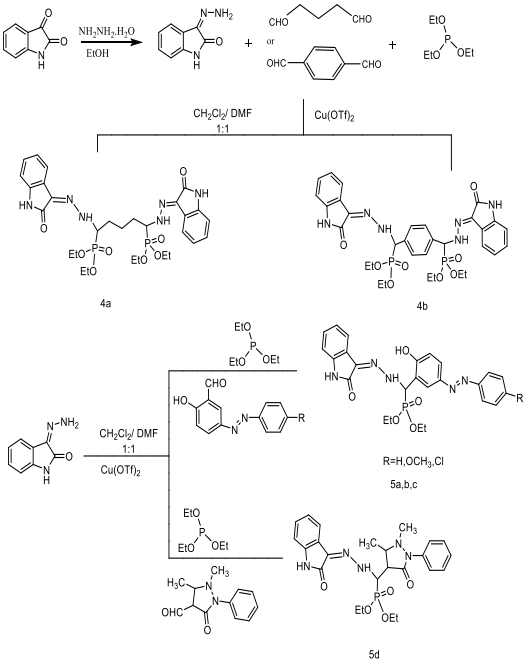
Scheme 1: Suggested mechanism of isatin hydrazide **4**.

*Textile printing paste preparation*

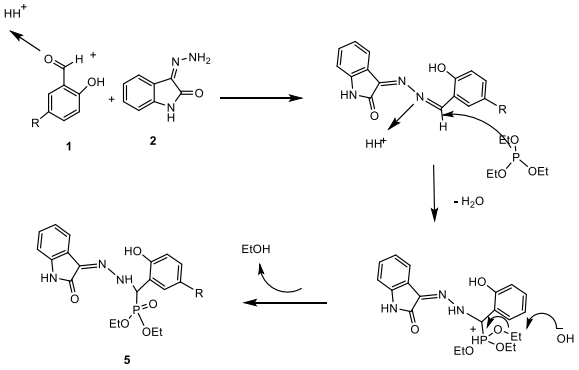
Printing pastes were prepared according to the following recipe:

| | |
|---|---|
| Dye | 2g. |
| Thickener | 75g |
| Lyoprint | 3g. |
| Sodium dihydrogen phosphate | 5g. |
| Sodium Lignosulphate | 1g. |
| Water | X g. |
| Total | 100g. |

The printing pastes discussed above were applied to polyester fabrics using a traditional screen-printing procedure. The prints were then dried at room temperature before being thermo fixed for 4 minutes at 180°C. The fixing temperature is important to promote the mobility of the dye molecules and increase the rate of dye transfer from the printed film to the fabric. The prints were rinsed in cold water, hot water and washed with 2 g/l nonionic detergent at 60°C for 30 minutes



Scheme 2: Synthesis of mono and di-aminophosphonate.



Scheme 3; Mechanism of Synthesis of compound **5**

*3.1. Color intensity (K / S) and fastness properties [35]*

*3.1.1. Color strength measurement*

Table 1 shows the colorimetric intensity data of the printed polyester fabric sample after soaping using light reflectance technology and the Hunter Lab Ultra Scan PRO spectrophotometer. The (K/S) value is proportional to the dye concentration on the fiber under the printing conditions used, at least at the dye concentration used (2.0 g for L100 g paste). Color intensity expressed as (K/S value) using Equation 1 of Kubelka Munk:

$$K/S = \frac{(1-R)^2}{2R} \quad (1)$$

where R is the fractional part of the reflectance of the printed fabrics, K is the absorption coefficient, and S is the scattering coefficient.

Table (1): Color evaluation of printed azo disperse dye samples

| Dye | Shade | $\lambda_{max.}$ (nm) | K/S | $L^*$ | $a^*$ | $b^*$ | $c^*$ | h |
|---|---|---|---|---|---|---|---|---|
| **5a** | Orange Yellow | 403 | 13.14 | 28.09 | 41.60 | 53.88 | 69.33 | 38.65 |
| **5b** | Orange yellow | 412 | 14.64 | 26.75 | 44.93 | 61.44 | 67.32 | 36.08 |
| **5c** | Yellow | 390 | 8.80 | 12.66 | 32.46 | 67.98 | 82.23 | 25.05 |

where; Lightness (L*), redness (+ve) and greenness (-ve) (a*), yellowness (+ve) and blueness (-ve) (b*), saturation (c*), hue ( h) and strength of the printed faric color (K/S).

Tables (1) and (2) illustrate the color tools or parameters of screen-printed polyester fabrics. The results showed that the color strength of the printed polyester fabric (expressed as K/S) is dependent on the chemical structure and nature of the substituents in combination with the benzene-azo dye moiety of the dye produced to produce higher quality prints.

Dye 5a-c had a higher colour strength and better outcomes, which could be owing to the higher conjugation of the benzene coupling component of dyes 5a-c, as well as its higher absorption, which increases dye dispersion and build-up on the fiber.

The azo compounds 5c have a lower lightness (L*) than 5a,b. On colored fabric, the lower the lightness (L*), the darker the colour [37]. The hue angle (h°) and chroma (saturation) (c*) were determined using the following equations [38, 39]:

$$c^* = \sqrt{(a^*)^2 + (b^*)^2} \quad (2)$$

$$h = \tan^{-1}\left(\frac{a*}{b*}\right) \quad (3)$$

The values of $a^*$, $b^*$ are positive which driven the color hue of dyes 5a-c at the polyester material closer to the reddish– yellowish direction. Color became barely better with dye 5b than with others. The PE fabric were silk-screen-printed using dyes 5a-c (thermo-fixation at 180 °C for four minutes).

*3.1.2. Wash fastness measurements*

Wash fastness refers to ability of the printed fabric to retain the color after being washed with detergents and soaps. The printed samples tested for washing fastness by sandwiching a piece of the printed fabric between uncolored cotton fabric pieces and washing it at 60°C for 30 minutes. The washing fastness then evaluated using a grey scale. (Table 2)

All of samples show good to excellent results, which could be attributable to the dyes' lack of solubilizing groups, which renders solubility.

*3.1.3. Color fastness to Perspiration*

The data of perspiration fastness is shown in Table (2). (Alkali and acidic solutions). Table 2 shows that the dye molecular weight and the high binding between the azo disperse dye and the polyester fabric have an impact on dye removal from the surface of the printed fabric. The colour variations were assessed using the grey scale [48]. When compared to other printed dyes that generated moderate to excellent (2-4) outcomes, while samples printed using the dye 5a-c show very good to excellent (4-5).

*3.1.4. Rubbing Fastness*

Table 2 lists the properties of. This test is used to determine and indicate the amount of color that was removed or transferred from the polyester fabric's surface to another surface by rubbing. The rubbing fastness qualities of a dye increase as the molecular weight of the dye increases. The rubbing fastness of most dyes ranges from good to excellent. Fastness properties of the samples are good. This could be attributed to increased dye fixation on the fibre and the efficacy of high physical Van der Waals forces in dye-fibre interactions.

According to the blue scale, the light fastness values of the synthesised azo dyes printed samples gave us good to very good results (Table 2) . The high force of attraction between the fibre and the dye is a sign of good light fastness results. In addition, the type of the substituents group in the coupling component has an impact on them. The greater the colour strength, the greater the light fastness.

*3.1.5. Sublimation fastness*

The sublimation fastness characteristics [31] were expressed and listed in table (2). We used an iron tester (Yasuda No. 138), which is a must-have for anyone working with printed fabrics. Most of the printed samples provided good results according to the international geometric grayscale.

Table (2): The printed samples fastness properties

| Sample dye used | Light fastness | Washing fastness | | Rubbing fastness | | Perspiration fastness | | Sublimation | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | at 120°C. | at 180°C. | Staining on | |
| | | Alt. | St. | Dry | Wet | Alt. | St. | | | Cotton | Polyester |
| **5a** | 6 | 4-5 | 4-5 | 3-4 | 3-4 | 3-4 | 4-5 | 4-5 | 3-4 | 4-5 | 3-4 |
| **5b** | 6 | 3-4 | 4-5 | 3-4 | 4-5 | 4-5 | 2-3 | 4-5 | 4-5 | 4-5 | 3-4 |
| **5c** | 7 | 4 | 3-4 | 3-4 | 4-5 | 3-4 | 3-4 | 4 | 3-4 | 4 | 3-4 |

Alt. : alteration of color , St. staining on the cotton piece.

*3.2. Antibacterial screening*

The biological activities of mono and bis amino phosphonate hybrid compounds containing the isatin moiety are of tremendous interest in the search for new and possibly valuable products. Gram-positive (Staphylococcus aureus ATCC 6598, Bacillus subtilis AATCC6633, and Streptocoucus faecalis), and Gram-negative (Pseudomonas auroginosa ATCC 9027, Escherichia coli ATCC8739, and Yeast) bacteria were tested in vitro for antibacterial activity. The disc diffusion method was used to screen the examined samples. The initial chemical, isatin, exhibited low antibacterial activity against a number of pathogenic bacteria, according to our investigation. Table 3 summarizes the findings, which show that the antibacterial effect of the starting materials is restricted, with inhibition zones ranging from 2.0 to 3.0 mm. Compounds 4a and 4b, which contained mono and bis amino phosphonate, have extremely high antibacterial activity. When compared to the reference medication ciprofloxacin, data analysis revealed that the novel amino phosphonate compounds hybrid analogues comprising indole moiety show different antibacterial activity with a variety of inhibition zones. **4b and 5b** showed the best antibacterial activity against all pathogenic bacteria. Table (3) shows the antibacterial activity of newly synthesised mono and di aminophosphonates against the test pathogens, with the majority of them showing an increase. It's possible that the inhibition capacity of newly produced chemicals to stick to the bacterial cell wall and cytoplasmic membrane is related to a change in selective permeability function, which leads to cellular component leakage and, eventually, bacterial death [22]. The formation of new compounds such as 4b because of introducing amino phosphonate to bacterial cells also has toxic effects [23, 24]. Furthermore, by interacting with phosphorus moieties, the newly produced compounds can block DNA replication. [25].

The treated fabrics' antibacterial efficacy was also tested using the AATC 100-1999 method. Table (4) displays the results. The coloured fabrics demonstrate varied degrees of resistance to the germs tested. The coloured cloth with (5a) exhibits a good inhibition on Bacillus subtilis ATCC6633 (93 %) within 18 hours of contact time, according to antimicrobial data. Surprisingly, among all coloured materials, the fabric dyed with 5b and 5c had the strongest antibacterial action against E. coli, with a reduction of over 80%). The negative control, on the other hand, had no inhibitory impact on any of the bacteria tested. Furthermore, 5a-c dyed fabrics were able to kill around 63 % of Streptococcus faecalis and 58 % of Escherichia coli ATCC8739 within the contact time.

Table (3): Antibacterial screening of newly compounds 4a, b and 5a-d

| Compounds | $G^+$ | | | $G^-$ | | |
|---|---|---|---|---|---|---|
| | ***Staphylococcus aureus* ATCC 6598** | ***Bacillus subtilis* ATCC6633** | ***Streptococcus faecalis*** | ***Candida albicans (yeast)*** | ***Pseudomonas auroginosa* ATCC 9027** | ***Escherichia coli* ATCC8739** |
| **ciprofloxacin** | **35.3±1.6** | **30±0.8** | **29±0.8** | **24±1.6** | **36.6±1.2** | **31±1.2** |
| **4a** | **13.3±.9** | **19.6±1.2** | **16.3±1.2** | **0±0** | **10.6±0.4** | **15±0.8** |
| **4b** | **16.3±1.2** | **20±0.8** | **18.3±1.2** | **16.3±1.2** | **13±0.8** | **22±1.6** |
| **5a** | **13.3±1.2** | **14±0.8** | **15.6±1.2** | **13.3±1.2** | **9.6±0.4** | **21.8±0.8** |
| **5b** | **9.3±0.4** | **24±1.6** | **18±1.6** | **9±0.8** | **21.6±1.2** | **23.3±1.2** |
| **5c** | **11.3±0.7** | **13±1.1** | **20±1.2** | **10±0.9** | **22.6±1.4** | **21.7±1.4** |
| **5d** | **14.3±0.6** | **17.2±1.2** | **21±1.3** | **12±0.9** | **24.6±1.9** | **25.3±1.5** |

Table (4): Activity of printed samples against Gram positive& negative bacteria.

| Printed azo dyes | $G^+$ | | | $G^-$ | | |
|---|---|---|---|---|---|---|
| | ***Staphylococcus aureus* ATCC 6598** | ***Bacillus subtilis* ATCC6633** | ***Streptococcus faecalis*** | ***Candida albicans (yeast)*** | ***Pseudomonas auroginosa* ATCC 9027** | ***Escherichia coli* ATCC8739** |
| **ciprofloxacin** | 35.3±1.6 | 30±0.8 | 29±0.8 | 24±1.6 | 36.6±1.2 | 31±1.2 |
| **Untreated polyester** | 0 | 0 | 0 | 0 | 0 | 0 |
| **Printed fabric 5a** | 16.3±1.2 | 20±0.8 | 18.3±1.2 | 16.3±1.2 | 13±0.8 | 22±1.6 |
| **Printed fabric 5b** | 11.3±0.9 | 10.2±0.7 | 13.6±1.1 | 12.1±1.1 | 8.6±0.5 | 20.8±0.7 |
| **printed fabric 5c** | 8.8±0.3 | 22±1.4 | 16±1.4 | 8.6±0.7 | 20.6±0.9 | 21.1±1.1 |

## 4. Conclusion

In this study, six newly synthesized products prepared and evaluated by classical method achieved via a one–pot three component Kabachnik Fields

reaction of isatin by coupling reaction with mono and di aldehyde, tri ethylphosphite to produce aminophosphonte structures. The IR and $^1$H NMR techniques were used to characterize the newly synthesized dyes. These dyes were used in traditional silkscreen printing on polyester. Furthermore, the printed samples' fastness qualities and color evaluation yielded promising findings. The *in vitro* antibacterial screening of the newly dyes improved applicability and antimicrobial properties. Further investigation of this newly compounds proved to be more antibacterial when compared to reference drug, ciprofloxacin.

**5. Conflicts of interest**

There are no conflicts to declare.

**6. Funding sources**

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