



Recent Advances of Functional Fluorescent Dyes: Textile and Non-Textile Applications

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Abstract

The conventional dyeing technology of textiles using organic dyes is well-known to the majority of customers. However, the applications of organic colorants are increasingly important for high-tech applications. This has led to the emergence of functional dyes, which are used in specialized high-tech applications such as optoelectronics, dye-sensitized solar cells (DSSCs) with high efficiency which record $\eta\% >10$, sensing and fluorescence probes for anions and heavy metal with high sensitivity which can detect ions with concentration < 20 nanomole, data storage, organic light-emitting diodes (OLEDs), photochromic materials, thermochromic printing, and laser applications, among others. Fluorescent dyes, which can absorb and emit electromagnetic radiation through radiative deactivation mechanisms in electronic systems, are particularly interesting. The fluorescence behavior of organic dyes adds value to them and increases their demand in various applications. This review article aims to discuss recent advances in functional fluorescent dyes and their applications in textiles as well as in non-textile areas such as electronics (including lasers, DSSCs, and OLEDs), analytical and sensing technologies, and medical applications.

Keywords: Fluorescent dyes; Textile; Fluorescent probe; Dyeing; Functional Dyes; non-textile applications.

1. Introduction

Organic colorants have been well-known for a long time and are used to introduce colors to various substrates such as textiles, leather, and polymers [1-10]. These colored materials play a crucial role in influencing our emotions and aesthetic preferences. The fluorescent dyes' ability to emit light has now created new opportunities for their use in a wide range of industries. Fluorescence is the process through which ultraviolet (UV) or visible light is absorbed and then released at longer wavelengths. Fluorescent dyes are substances that emit light highly while also absorbing light significantly in the visible spectrum. These substances have the potential to be used because of their extreme fluorescence qualities. Fluorescent dyes application in medical diagnostics and biochemical research have spread significantly [11-19]. Fluorescent functional dyes were applied in different hi-tech fields such as, lasers manufacture [20-23], DSSCs and solar batteries [24-27], fluorescence probe for naked eye detection of different analytes [8, 15, 28-35]. On the other hand, the basic use of textile for clothing purposes evolved alongside the growth and development of society. As a result, protective textile clothing emerged, and research studies seeking solutions for adding functionalities to textiles increased. The use of functional textiles for various application fields has increased dramatically due to its flexibility, breathable nature, and low cost, has gained popularity in almost every field, including aeronautics, defense, aesthetics, and interior design. Thus, fluorescent dyes was used as high brilliant and visible colors which improve the functionality of the dyed fabrics with a significant increase in color brightness, saturation and visibility, which makes the dyed materials more fashionable and easily perceptible [36-38]. This deep perceptibility of the substrates dyed with fluorescent dyes is advantageous in manufacturing of functional fluorescent textile for a variety of purpose such including warning, safety, aesthetics, advertisements, information descriptions, road and traffic signs, firefighter clothing and police officer [39-41]. The investigations about fluorescent disperse dyes used for dyeing polyester fabrics to high-visibility have been presented in recent years [42-45]. Different fluorescent chromophoric system was used for synthesis of different fluorescent dyes as instance coumarin [46-48], 1, 8-naphthalimide [49-54], phenothiazine [16, 55], pyrene [16], anthraquinone [56, 57], hemicyanine-azo [5, 58-60], quinoxaline dyes [61] etc. In this review we present the recent advances for different kind of fluorescent dyes which showing high performance characters for textile and non-textile applications.

1.1. Type of luminescence

There are different type of luminescence depending on the energy influence source which responsible for excitation and electronic transition in the molecule as shown in **Figure 1** and summarized in **Table 1** [62]. The electroluminescence can be obtained by the effect of electrical energy as in optical fiber, while the effect of absorption of photons led to appearance of emission is called photoluminescence. Chemiluminescence formed due to chemical energy and mechanoluminescence can be observed by applying a mechanical stress [63-66]. Bioluminescence is ascribed to the natural process in living organisms, which imparts visible light due to its in vivo biochemical reaction [67, 68]. We will concern with the photoluminescence type

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which concerning with the absorption of light this because the functional fluorescent dyes are materials which showing this kind of luminescence.

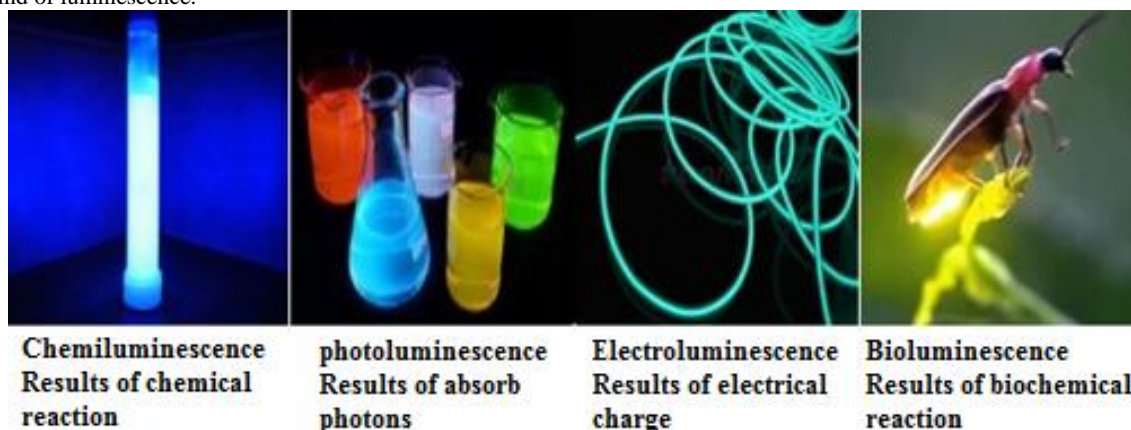


Fig. 1. Types of luminescence depending on the type of energy influence [69].

Table 1: types of luminescence and their application

Type of luminescence	Energy influence source	Application
Photoluminescence	Emission by UV or visible light	Purity and defects in minerals and crystals, forensic science, sensors, imaging
Bioluminescence	Emission ascribed to <i>in vivo</i> biological reaction	Biotechnology, environmental, bioimaging
Thermoluminescence	Emission based on the thermal effect by absorbing light	Environmental and personal dosimeters, archeological pottery, dating, irradiated food identification
Electroluminescence	Excitation due to electric field	LED, lighting, displays, medical applications
Pyroluminescence	Emission from excited atoms, ions, and molecules in flames	Lamps, fireworks, flames
Radioluminescence	Irradiation by high energy photons such as X-rays, γ -rays	Scintillators, X-ray film radiography
Cathodoluminescence	Excitation from electron impact on solids or gases	TV screen, impurity mapping, discharge lamps
Ionoluminescence	Excitation produced from alpha particles emitted by radioactive elements	Early clock dials

To understand the photoluminescence characteristics of fluorescent dyes, we need to introduce some definitions. The light spectrum refers to the range of wavelengths of light. The emission from the dyes occurs after an excitation process where they absorb light in the visible region (approximately 400-700 nm) and then emit the absorbed light. Light is a form of electromagnetic radiation (EMR) that consists of photons. There are two types of EMR: natural sources like sunlight and artificial sources like lamps. This light covers a broad range of EMR wavelengths, as shown in **Figure 2**. Most of colored materials absorb light in visible region while non- colored conjugated material can absorb light in UV region.

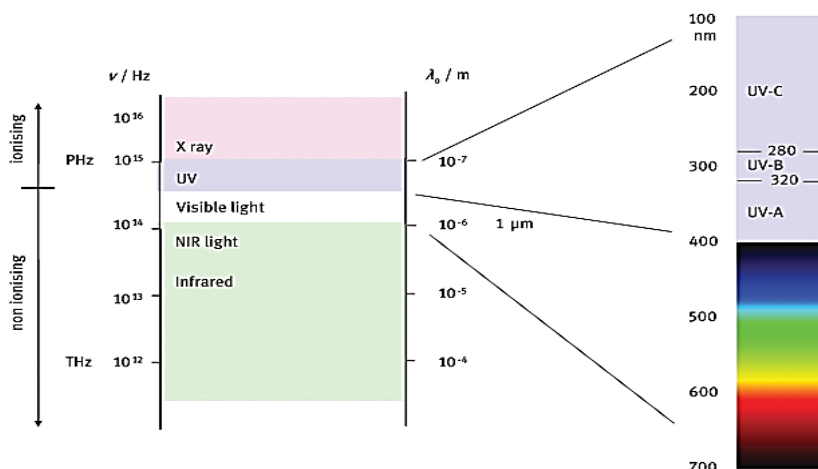


Fig.2. EMR spectrum regain, vacuum wavelengths and frequencies [70]

1.2. Excited state deactivation process in photoluminescence

The absorption of electromagnetic radiation by colored materials can lead to different electronic transitions in the electronic diagram of the substance. The state after absorbing light changes from ground to excited state, to reach the stability the absorbed energy have to lost. Thus, there's two different mechanism of deactivation process as shown in Jablanisky diagram **Figure 3**.

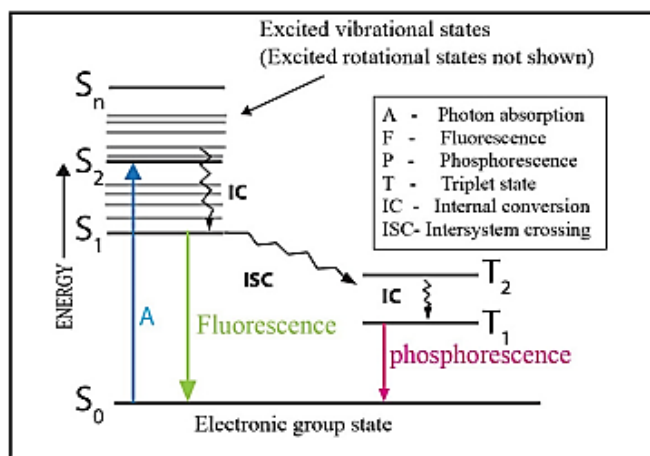


Fig. 3. Jablonsky diagram, photoluminescence process [70]

1.2.1. Non-radiative deactivation process

Absorption of photons causing a vertical transition to a higher excited state results in the creation of a new vibrational level within the excited state S_1 of the molecule. As a result, the molecule loses vibrational energy in order to achieve the excited state's equilibrium configuration (vibrational level 0). In liquid phase, this process of vibrational relaxation is very rapid (10–13–10–12 s) due to frequent collisions with molecules of solvent or other species in the reaction mixture. Given that the lifetime of an excited state S_1 or T_1 is much longer than the period of vibrational relaxation, the lifetime of the equilibrium configuration of the excited state may be considered as the lifetime of this state (including vibrational relaxation). This vibrational relaxation is called internal conversion (IC) which is one of the non-radiative transition. The IC transition occur between an excited state and a state of lower energy of the same multiplicity. While if the transition observed between two energy level with different multiplicity S_1 to T_2 as instance this transition called inter system crossing (ISC).

1.2.2. Radiative deactivation process (luminescence)

After absorbing the visible light, the electron moves from the ground state S_0 to higher energy levels S_1 or S_n . This is known as absorption (A). If the absorbed energy is lost and emission is observed by the transition between two energy levels with the same multiplicity this process is called fluorescence (F). If the electron transfer from a singlet excited state to triplet excited state T_2 by intersystem crossing (ISC) then loss the energy to the ground state, the emission observed by the transition between two energy level with different multiplicity this process called phosphorescence (P). The life time of fluorescence is very short comparing with the life time of phosphorescence and it always in nano-second while life time of phosphorescence usually in μ s.

The emitted radiation energy is smaller than the energy of excitation, and the emission spectra is a mirror image of absorption spectra which shifted to longer wavelength comparing with absorption spectra and the difference between both spectra called (Stocke's shift). While, phosphorescence spectra can be even shifted more than the fluorescence spectra and this can ascribe to the energy of the triplet state is smaller than that of the corresponding excited singlet state as shown in **Figure 4**.

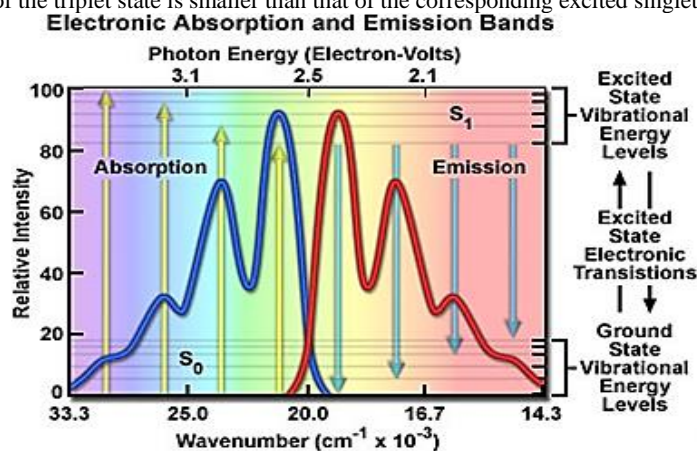


Fig.4. Relation between absorption and emission spectra [70].

The quantum yield of luminescence Φ_L (fluorescence or phosphorescence) can be defined as the ratio between number of photons emitted and number of photon absorbed and can be calculated by using **equation 1**.

$$\Phi_L = \frac{P_L}{P_a} \quad 1$$

2. Fluorescent dyes for textile applications

Due to the desire for high-performance applications, textile solutions with just one characteristic have not met expectations. The necessary functional properties can be achieved through multifunctional fabrics. Consequently, in recent years, a high challenge in scientific research for production of functional and ecological textiles was significantly observed [71, 72]. As a result of the numerous uses that are being found for these materials, smart textiles have undergone rapid growth [73]. The global market for smart textiles was worth \$795 million in 2014, and by 2020, it was anticipated to increase to \$4.72 billion, with a compound annual growth rate of 34%. (Smart textiles market by type, function, industry & geography – Global forecast to 2020). Smart textiles are items that can both receive and respond to external stimuli in order to adapt to changes in their environment. These substances may respond to various physical stimuli, such as light, pH, temperature, solvents with various polarity, chemicals, and electricity, and as a result, they can interact with their surroundings (sense, respond, communicate, and/or adapt) [74]. Three subgroups of smart textiles may be distinguished, Active smart textiles are reactive to stimuli from the environment and integrate an actuator function and a sensing device. Very smart textiles are able to sense, react, and adapt their behavior to the given circumstances. Passive smart textiles are only able to sense the environment/user based on sensors [75, 76]. The incorporation of intelligent textiles into apparel is nothing more than a reflection of modern human lifestyles that include access to things that are technologically advanced, adaptable, and multipurpose. Smart textiles may be used for a variety of purposes, including as power production and storage, personal safety, sports, fashion, communication, and internet of things applications [77, 78]. One of the main objectives of the textile industry is the development of novel smart textiles, which is motivated by consumer and market demands for high-tech materials. The literature has reported on a variety of smart textile kinds, including chromic textiles.

Functional textiles have properties including fluorescence, photochromism, antimicrobial activity, hydrophilicity, super-hydrophobicity, self-cleaning, crease recovery. One such practical and intelligent textile is fluorescent fabric, which may be utilized for a variety of applications in the biomedical and medical industries as well as for warning, safety, aesthetic, and anti-counterfeiting purposes [40, 41]. The usage of luminous textiles has grown recently, along with the development of new application areas like smart textiles [79].

2.1. The chemistry of most common fluorescent dyes

This part will be described the classification of fluorescent dyes according to chemical type, including carbonyl (coumarin, naphthalimide, perylene, benzanthrone, benzoxanthone, rhodamine, methine and benzothioxanthone derivatives) in addition to illustrations of the most significant chemical structures it's very important to present some important rules which basically affect the fluorescent characteristics of dye molecules.

There are several recognized general rules relating the dye structure to their fluorescence intensity as measured by quantum yields [80]. The majority of fluorescent dyes are extended conjugated aromatic systems with fused rings. Structural stiffness is a significant chemical characteristic that is frequently linked to high fluorescence quantum yields. Thermal motion inside a stiff molecule minimizes energy loss from excited states, favoring fluorescent emission over non-radiative energy loss.

The "loose bolt effect" is the term used to describe the decrease in fluorescence intensity in molecules with reduced structural rigidity. Azo dyes are typically non-fluorescent because they have two aromatic systems connected by the azo ($-N=N-$) group and the azo group is quenching the fluorescent which ascribed to the inherently decreasing rigidity than fused polycyclic systems, the chemical structure of azo dyes permits some rotation around the azo bond, similar to this, it is understood that the nitro ($-NO_2$) group, which may rotate rather freely, frequently inhibits fluorescence [9, 81]. However, the reliability of conventional explanations based solely on molecular rigidity has been questioned, at least in part due to the prevalence of exceptions. More sophisticated arguments related to the nature of excited state structures and the associated charge transfer electronic transitions, such as the contribution of non-bonding electrons, have also been put forth. Even though they have planar aromatic ring systems, extremely fluorescent dye molecules frequently also have structural characteristics that give them a significant amount of non-planarity. Planar molecules have a propensity to group together, sometimes taking on pigmentary characteristics, and release excited state energy through relaxation processes involving intermolecular interactions. A famous example of a stiff yet non-planar molecular structure that results in high fluorescence intensity is fluorescein dyes as shown in **Figure 5**. The presence of atoms with a higher atomic number, such as bromine and iodine, is another chemical characteristic known to typically cause lower fluorescence; this phenomenon is known as the heavy atom effect.

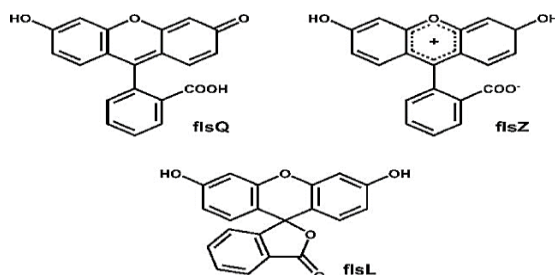


Fig. 5. Fluorescein structure and its tautomeric forms, quinoid (flsQ), zwitterionic (flsZ), and lactoid (flsL).

The substantial impact of environmental variables, such as the composition of the solvent, dye concentration, and temperature, on fluorescence emission spectra, poses a considerable challenge to designing and proving such a strategy. In dyes, strong fluorescence is frequently linked to the presence of certain, recognizable structural units.

2.1.1. Carbonyl based fluorescent dyes

This class of fluorescent dyes is the most important type for textile applications which characterized by high efficient emission and high quantum yield of fluorescence. Many chromophoric systems based on carbonyl dyes such as coumarin, naphthalamide, benzanthrone, perylenes and benzoxanthenes and benzothioxanthenes [82].

a) Coumarin based commercial fluorescent dyes

The primary source of fluorescent industrial dyes is coumarin derivatives. While most products on the market are yellow with a green fluorescence, fluorescent coumarins can absorb and emit light in various visible spectrum regions. Commercially, fluorescent whitening agents (FWAs) based on coumarins are also referred to as fluorescent finishing materials.

In coumarin dyes, the electron-releasing substituent, typically the diethylamino group, is consistently found at the 7-position, while electron-withdrawing substituents are consistently located at the 3-position in the coumarin ring. Thus, the coumarins are common donor-acceptor dyes [83]. The most common types of dyes include a benzimidazolyl, benzoxazolyl, or benzothiazolyl group as the acceptor at the third position as presented in **Figure 6**.

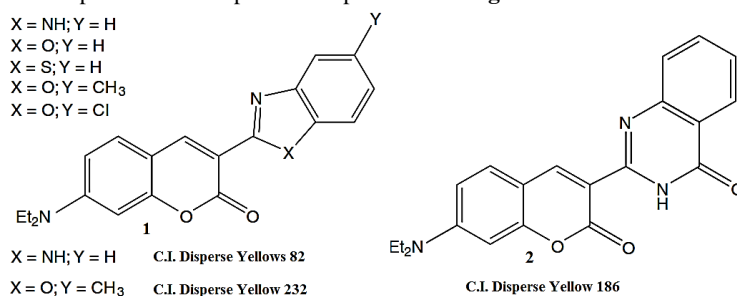


Fig. 6. Structure of some commercial fluorescent disperse dyes based on coumarin derivatives.

This group presents a crucial dispersion dyes that enable polyester as a synthetic fibers to be colored in vivid fluorescent greenish-yellow hues. There are known dyes having different heterocyclic substituents in the 3-position, such as C.I. Disperse Yellow 186 (2) **Figure 6**.

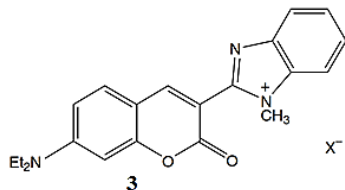


Fig.7. Chemical structure of C.I. Basic yellow 40.

The easy cationization of the benzazolyl-substituted coumarins as presented in **Figure 7** formed a new water soluble derivatives known as C.I. Basic yellow 40 which used for dyeing acrylic fibers with greenish-yellow emission.

Highly conjugated coumarin derivatives were shown in **Figure 8**, which absorb, emitting light at longer wavelength over 600 nm ascribing to the ring-formation, and enhanced electron-withdrawing behavior. This fluorescent greenish-blue considered as red fluorescent dye for dyeing polyester fabrics.

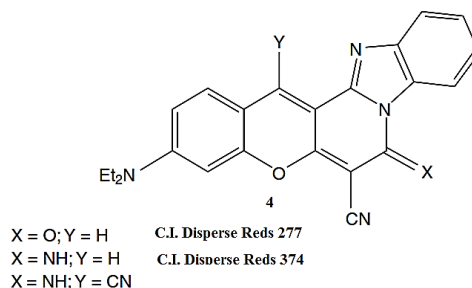


Fig. 8. Chemical structure of highly conjugated coumarin derivatives.

b) Naphthalamide based fluorescent dyes

It is widely known that fluorescent materials based on naphthalamide derivatives [84-87]. When an electron-donating group occupies the 4(5)-position, strong fluorescence is seen. In this situation, the absorption and emission characteristics are linked to charge transfer between the donor group and the electron-withdrawing carbonyl groups. The fluorescent yellow dye C.I. Disperse Yellow 11 (5) **Figure 9**, which has an imide nitrogen and an aryl substituent, is a well-known fluorescent dye despite having only moderate light fastness for polyester dyeing.

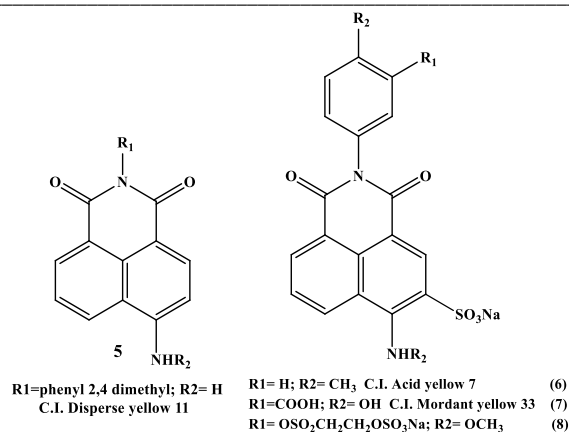


Fig. 9. Chemical structure of amino-naphthalimide fluorescence dyes.

The anionic dyes C.I. Acid Yellow 7 (6) and C.I. Mordant Yellow 33 (7), among others, are water-soluble naphthalimide dyes that have been used for silk dyeing and may also be utilized in non-destructive flaw identification. Reactive fluorescent yellow dyes for cellulosic fibers, such as (8), have been described as shown in **Figure 9**.

Some of disperse dyes based on naphthalimide derivatives such as C.I. Disperse Yellow 199 (9) and Orange 32 (10) are examples of fused heterocyclic derivatives attached with naphthalimide moiety as shown in **Figure 10**.

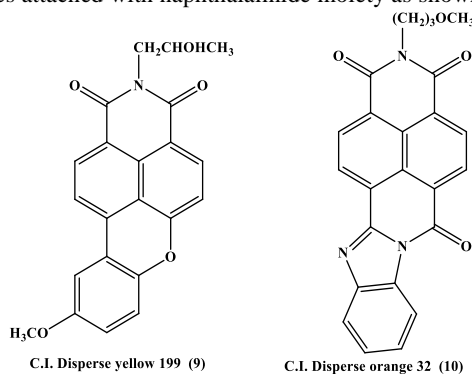


Fig. 10. Fluorescent dyes based on fused heterocyclic derivatives attached with naphthalimide moiety.

c) *Perylene based fluorescent dyes*

Perylene dyes are particularly interesting because they produce highly bright colors with a very high fluorescence efficiency, while also exhibiting exceptional stability against light and heat and chemical assault. For these reasons, they make beautiful thermoplastic coloring materials and are commonly the material of choice for practical applications where durability is crucial, including liquid crystal displays and solar energy gathering [88, 89]. Most of perylene based fluorescent materials have low solubility and are not suitable for textile dyeing and it can be used only in textile printing.

The most significant fluorescent perylene dyes are many based on the imidation of perylene-3,4,9,10-tetracarboxylic acid as shown in **Figure 11** (11), which mainly used in dyeing of polymeric substrates giving a bright orange to red fluorescent colors.

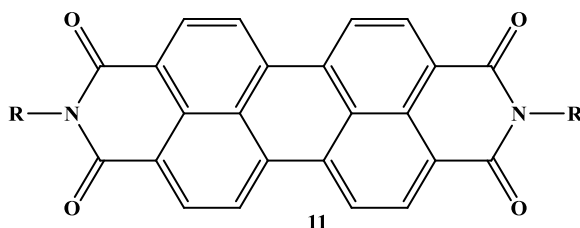


Fig. 11. Alkylated derivatives of perylene fluorescent dyes.

d) *Benzanthrone based fluorescent dyes*

The 3-methoxy derivative, also known as C.I. Disperse Yellow 13 (12) **Figure 12**, is the longest established fluorescence benzanthrone dye derivative. It shows a yellow-green emission. Although this dye is reasonably priced, it has the drawback of having a somewhat poor quantum yield. However, the intermolecular hydrogen bonding between dye and resin molecules is thought to be the cause of the fluorescence intensity increase in the resin systems employed in daylight fluorescence pigments.

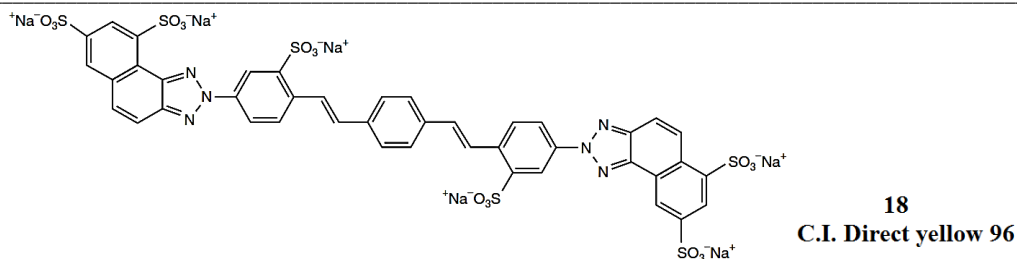


Fig. 15. Structure of methine dye (C.I. direct yellow 96).

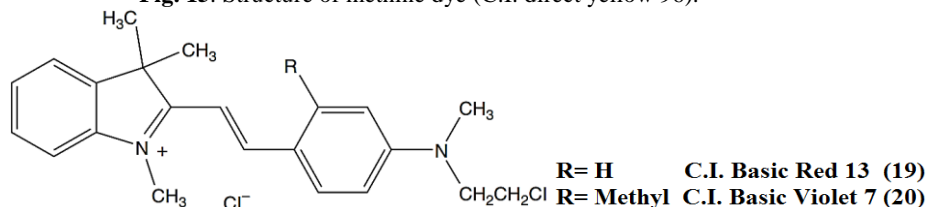


Fig. 16. Structure of cationic methine dyes for acrylic dyeing.

The most common methine dyes are cyanine and hemicyanine derivatives. These type are presented a cationic derivatives of methine dyes for acrylic dyeing applications. The well-known methine dyes for textile applications are C.I. Basic Red 13 (19) and C.I. Basic Violet 7 (20) as shown in **Figure 16**.

2.1.4. Miscellaneous fluorescent dyes

Some of the high conjugated fused heterocyclic compounds are highly fluorescent with superior fastness characters in textile applications such as washing, light and sublimations. This kind of fluorescent dyes showing a bright greenish-yellow shades on polyesters, cellulose acetate and nylon fibers. The most common example of such fluorescent dyes is C.I. Disperse Yellow 139 (21) as presented in **Figure 17**.

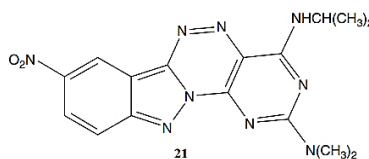


Fig. 17. Chemical structure of C.I. Disperse yellow 139.

3. Recent advances of the fluorescent dyes for textile applications

The most of commercially known fluorescent dyes are used for dyeing of synthetic fibers like polyester, polyamides, and polyacrylonitrile, as well as occasionally in combination with elastane fibers, fluorescent dyes are of far larger value. The most significant fluorescent textile dyes are from the disperse dye class since polyester is the material used in the majority of significant textile applications. On polyamides and cellulose acetates, fluorescent dispersion dyes are additionally utilized, but to a lower extent.

The majority of commercial dyes are yellow, producing the well-known greenish-yellow fluorescent colors that are frequently seen on clothing worn for safety reasons, In recent years, many researchers have been trying to prepare new fluorescent dyes for textile applications.

Chenchen Liu *et al* [94] was prepared five new fluorescent dyes for silk functionalization as aggregation-induced emission luminogens (AIEgens) by metal-free click bioconjugation reaction as presented in **Figure 18**. As tetraphenylethylene (TPE) is known as aggregation induced emission (AIE) unit which ascribed to the restriction of molecular motions. The synthesized dyes showing Blue, green, yellow, orange, and red emission and based on introducing propynone groups (AIE-pyo) **Figure 18**. According to the stated approach, TPE was functionalized with a propynone group to create a blue-emissive AIE-pyo luminogen as TPE is a typical AIEgen that emits blue fluorescence when exposed to UV light. Then, the electron-donating and electron-withdrawing groups, respectively, methoxy and benzothiadiazole groups, were chosen. They were used to create longer-wavelength emissions and D-A structures for MTPEP-pyo, TPEBP-pyo, and MTPEBP-pyo.

The chemically conjugated fluorescent silk fibers demonstrated excellent stability and achieved full-color emissions. Red, green, and blue emissive AIEgens were simultaneously bioconjugated to create a white light-emitting silk. Additionally, real-time and long-term cell tracking using hydrolyzed silk that has been functionalized with red emissive MTPABP has been achieved. The substantial two-photon absorption of MTPABP functionalized silk textiles proved their considerable potential for deep tissue imaging and bioscaffold monitoring.

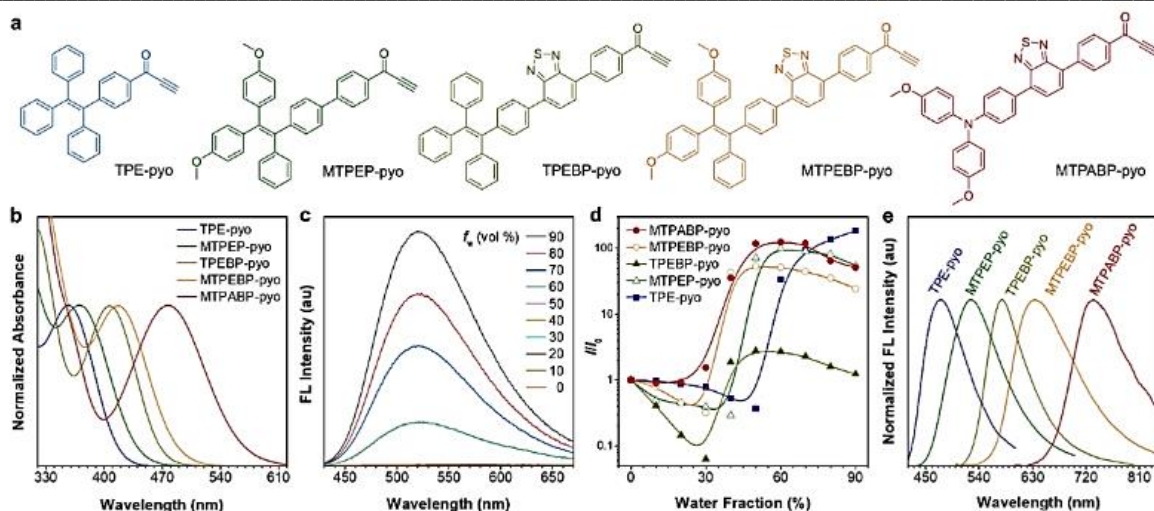


Fig. 18. Synthesized new fluorescent AIE-pyros a), the absorption spectra of dyes b), the emission spectra c), relative fluorescence intensity d), and the emission in solid state e) [94].

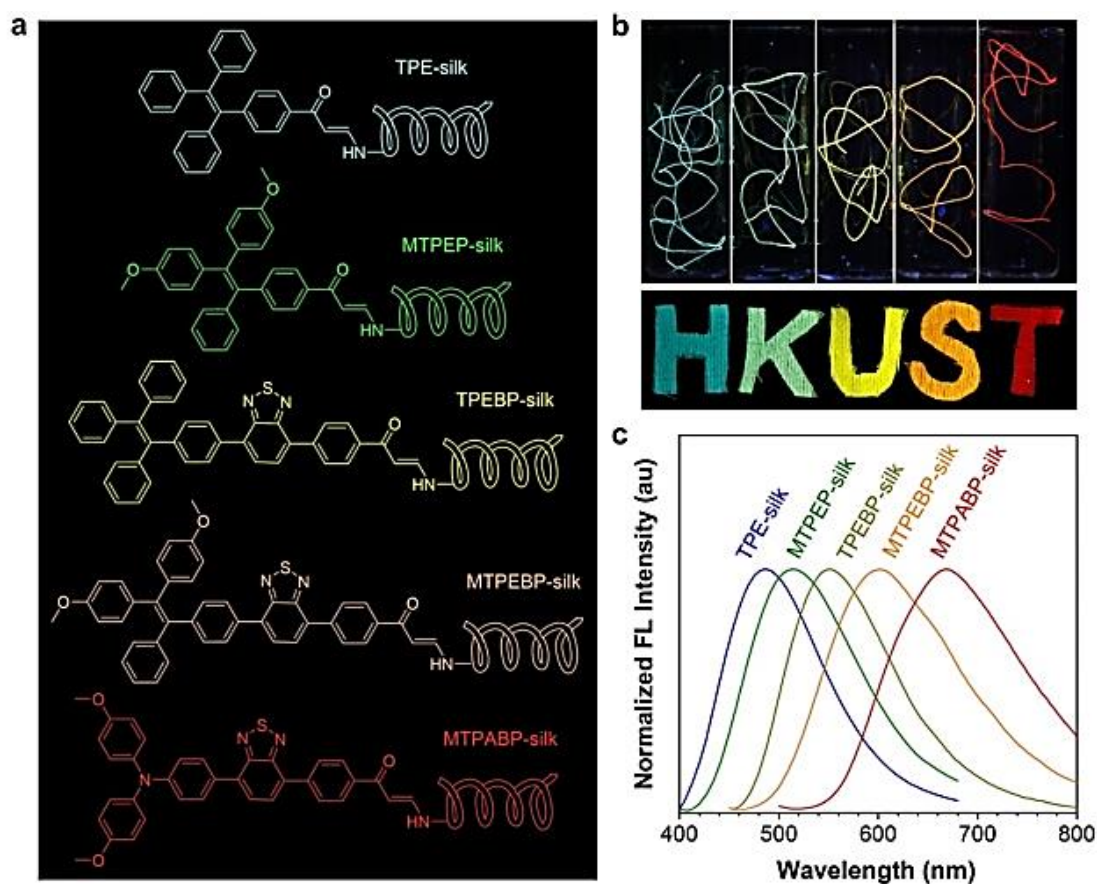


Fig. 19. Emission of AIEgen-silks a), Fluorescent images of AIEgen-silk textiles (lower) and threads (upper) under UV light b), emission on silk fabrics c) [94].

Silk fibers were easily turned into AIEpyro functionalized silks (AIEgen-silks) by soaking them in AIE-pyros solutions overnight at room temperature. The result was AIEgen-silk threads and textiles with consistent fluorescence that covered the full visible light spectrum **Figure 19**.

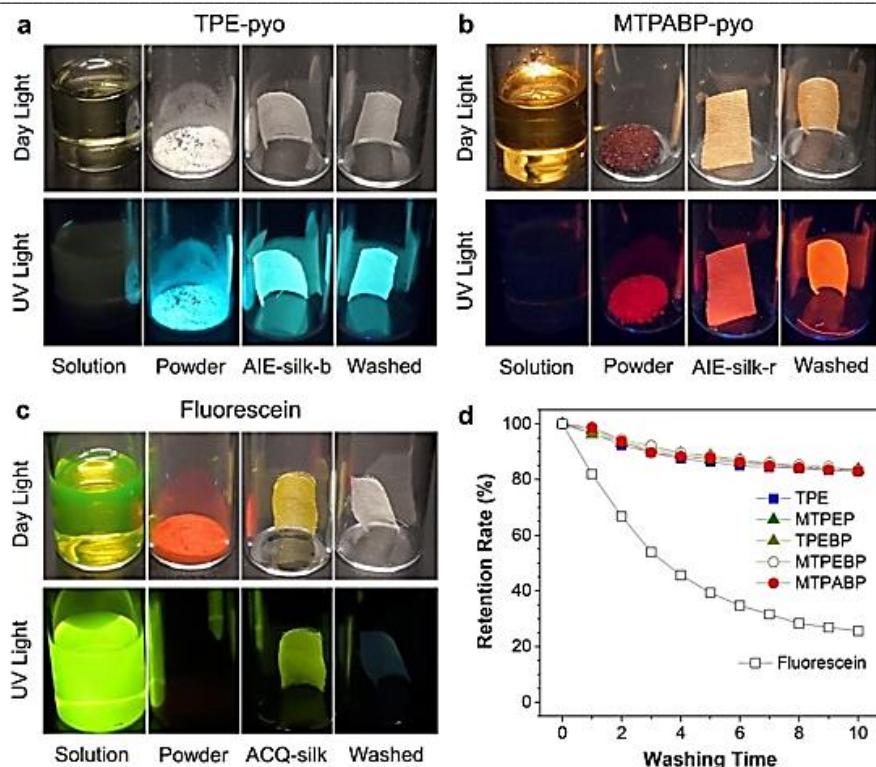


Fig. 20. Emission of TPE-pyo a), MTPABP-pyo b) and fluorescein c) upper image under visible light bottom image under UV, washing time effect in emission d) [94].

For showing the advantage of the prepared AIE-pyo and its emission efficiency on the silk fabrics, the fluorescein is known as aggregation caused quenching (ACQ) which is highly emitting in solution and low emission in powder form, thus after dyeing of silk fabric the emission is significantly decreased due to ACQ while in case of AIE-pyo the emission is very high in solid state and no significant emission can be observed in solution, therefore the AIEgen-silk showing very strong emission as shown in **Figure 20**.

Supercritical carbon dioxide (ScCO_2) is one of the interesting medium for water free dyeing and finishing of textiles as an economical and cleaner dyeing methods for textile industry which still very limited as industrial process [3, 95-97]. Jun Hou *et al.*, was published an interesting work which concerning with the synthesis of fluorescent dyes based on coumarin and naphthamide derivatives using ScCO_2 and studying there application on polyester fabrics as a cleaner production of fluorescent dyes [49].

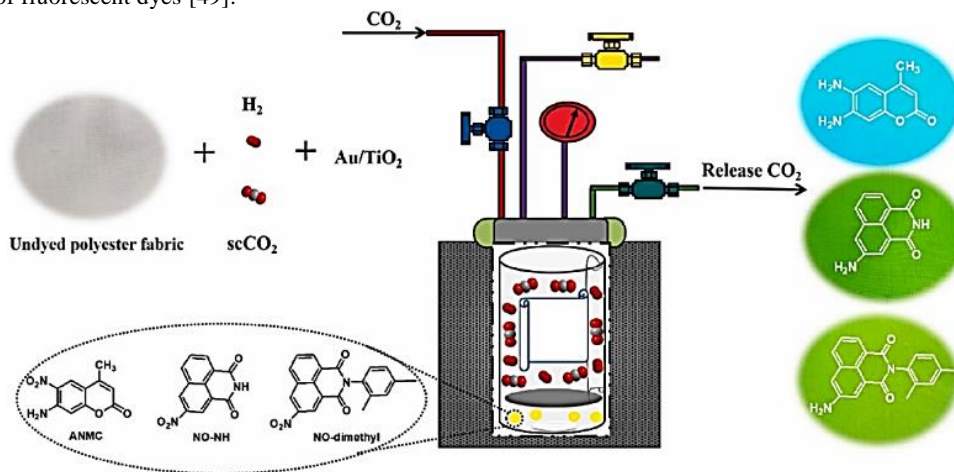


Fig. 21. Schematic diagram of the supercritical CO_2 reductions of the nitro-derivatives [49].

This work was concerning with the cleaner reduction of three precursors 7-amino-6-nitro-4-methylcoumarin (ANMC), 3-nitro-1,8-naphthalimide (NO-NH) and N-(2,4-dimethyl) phenyl-3-nitro-1,8-naphthalimide (NO-dimethyl) using cleaner method (ScCO_2) using of Au/TiO_2 instead of using $\text{SnCl}_2/\text{hydrochloric acid}$. **Figure 21** illustrate schematic diagram of the supercritical CO_2 reductions of the nitro-derivatives forming three fluorescent dyes 6,7-diamino-4-methylcoumarin (DAMC), 3-amino-1,8-naphthalimide (NP-NH), and N-(2,4-dimethyl)phenyl-3-amino-1,8-naphthalimide (NP-dimethyl) as shown in **Figure 22** which showing high emission on polyester fabrics.

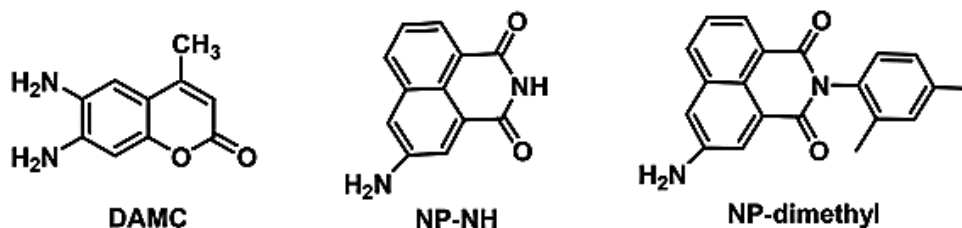


Fig. 22. Chemical structure of the ScCO₂ prepared dyes.

The simultaneous synthesis of the dyes as well as its dyeing application was successful showing excellent photophysical properties on PET fabrics as shown in **Figure 21**.

Previously Xiaoqing Xiong *et al* [37] were studied the supercritical carbon dioxide dyeing of polyester fabrics using a commercial disperse dye (C.I. Yellow 82). This study demonstrated that dyeing variables such dyeing duration, pressure, and temperature had significant effects on the color strength of materials. It was said that the ideal circumstances for supercritical CO₂ dyeing were a dyeing period of 60 minutes, a pressure of 25 MPa, and a temperature of 120 °C. According to the fastness data, the acquired samples under the chosen conditions had satisfactory washing fastness (fading and staining) and rubbing fastness (wet and dry) values of 4-5. Through SEM, supercritical CO₂ did not result in morphological harm to the luminous textiles. **Figure 23** shows the high emissive greenish fluorescent polyester fabric and the using of supercritical CO₂ dyeing method is not showing any significant effect on the emission intensity of the dyed fabric comparing with the conventional dyeing method.

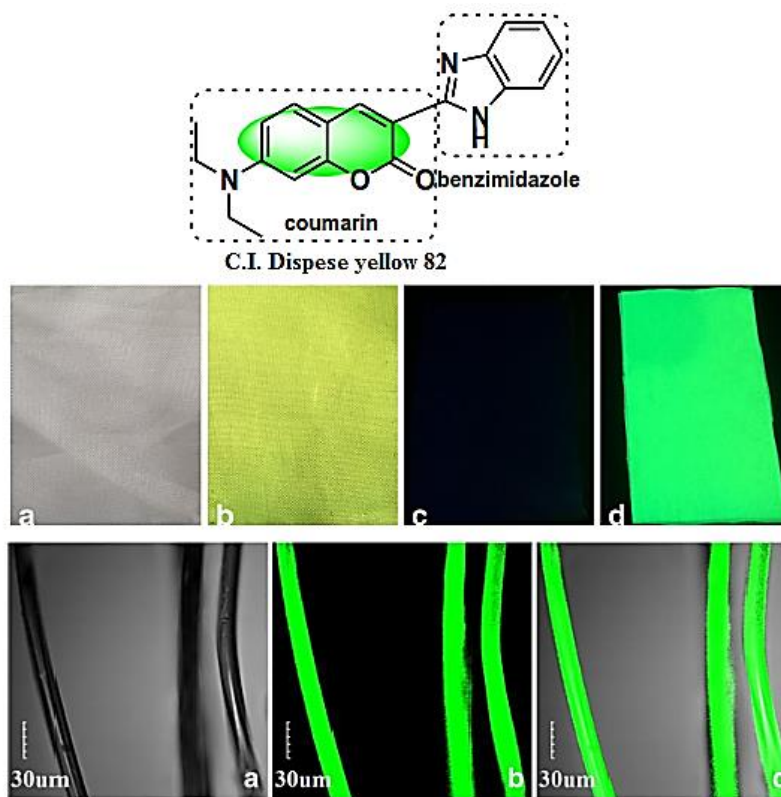
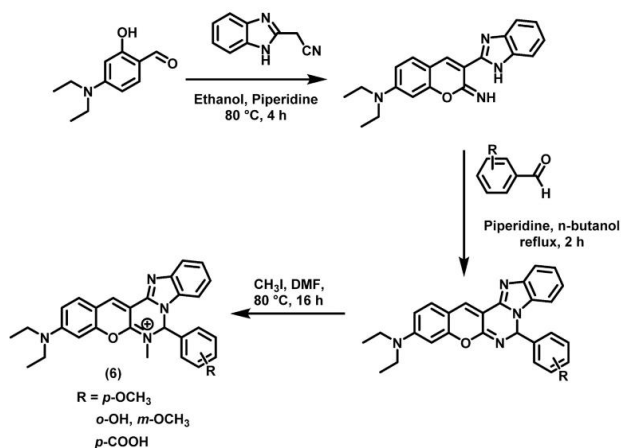


Fig. 23. Control sample and the dyed fabric under visible light a,b) and under UV light c,d) (upper image), the fluorescence confocal image of the dyed fabrics bright filed a), dark filed b) and the merge image c) (bottom image) [37].

Raju Penthala and Young-A Son [98] were prepared three new rigid cationic fluorescent coumarin dyes as shown in Scheme 1. The synthesis was carried out by the condensation reaction of 4-(diethylamino)-2-hydroxybenzaldehyde on the active methylene group in 2-cyano methyl benzimidazole under alkaline condition forming 2-iminocoumarin derivative. The high reactivity of 2-iminocoumarin increase the possibility of the reaction with electrophiles or nucleophiles, thus the 2-iminocoumarin was easily react with aldehyde derivatives followed by cyclization reaction. The quaterization of nitrogen located in pyrimidine ring led to the formation of three cationic fluorescent coumarin dyes presented in scheme 1. The synthetic dyes showing good photostability in when compared to that of a commercial dye (C.I. basic yellow 40). These dyes were used in a dyeing application on modacrylic fibres using a conventional cationic dyeing procedure as shown in **Figure 24**.



Scheme 1. Synthetic procedure of the cationic fluorescent dyes.

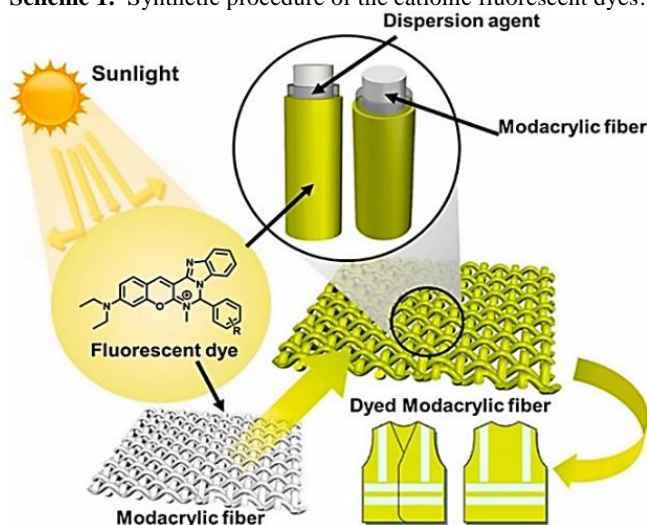


Fig. 24. dyeing application of the cationic fluorescent dye on modacrylic fibers [98].

A novel anti-counterfeiting method that will enable tagging and recognizing authentic objects utilizing cotton fiber and fluorescein was created and described by Khouloud Baatout *et al* [79]. On a cotton yarn, different concentrations of the affordable and environmentally friendly fluorescein fluorophore was applied on cotton yarn both directly and in the presence of the two types of resins using pad dry cure as shown in **Figure 25**. SEM, reflectance, and fluorescence analyses were used to describe the FL-coated fibers, and they showed that fluorescein was present on the cotton surface. When compared to untreated cotton, samples that have been FL-impregnated show a little increase in maximum breaking strength and a slight loss in elongation capacity; adding resin had no impact on the mechanical parameters. The results of the washing experiments demonstrate that the use of resin is necessary for a successful fixation of fluorescein on cotton substrate.

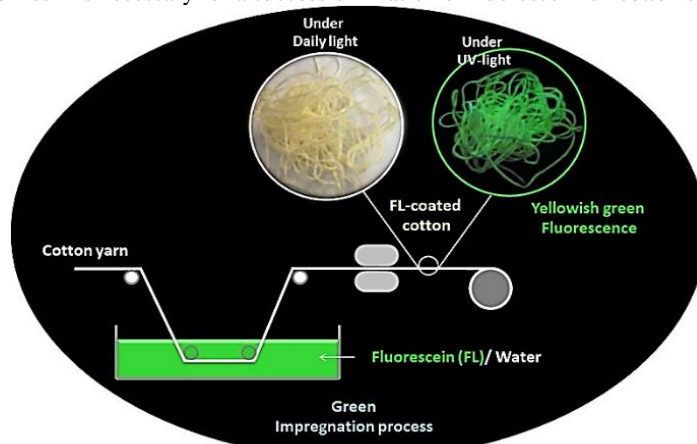


Fig. 25. Fluorescence coated cotton yarn using fluorescein fluorescence dyes fixed by resins [79].

The anti-counterfeit substance was applied to the textile support using the pad dry cure procedure. Samples were given the designations CF1, CF2, CF3, and CF4 for the 7.5 μM , 10 μM , 50 μM and 100 μM concentration respectively. By illuminating the samples at 460 nm, the fluorescence characteristics of FL-coated cotton fibres, CF1-4, were investigated at room temperature, and their optical data were shown in **Figure 26** briefly, this work created and defined a novel anti-counterfeiting method that will allow labelling and recognizing authentic objects utilizing fluorescein and cotton fiber.

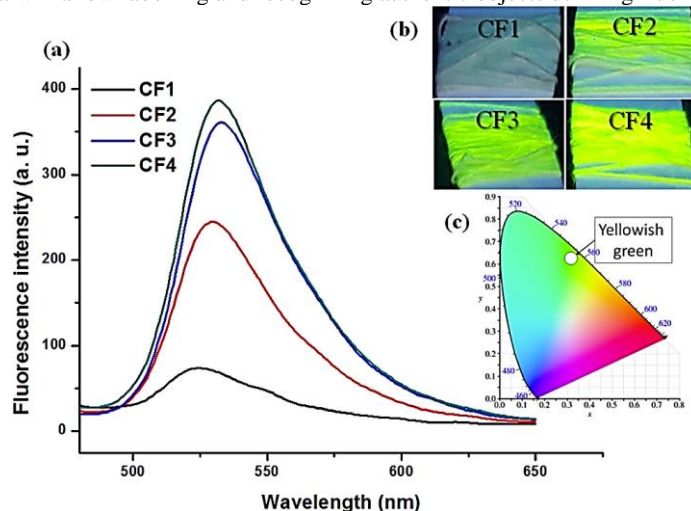


Fig. 26. Emission spectra of the treated cotton yarn a) the photograph of the fluorescent cotton under UV b) and the CIE of the cotton yarn diagram c) [79].

Souhangir *et al* [99], synthesized and characterized new red dyes based on fused iminocoumarin as shown in **Figure 27**. 3(diethylamino)-7-imino-7H-benzo[4,5]imidazo [1,2-a]chromeno [3,2-c] reacted with cyanuric chloride followed by the reaction with nicotinic acid to form a new luminous reactive dye (*E*)-3-carboxy-1-(4-chloro-6-((6-cyano-3-(diethylamino)-7H-benzo[4,5] aminoimidazo [1,2-a] chromeno[3,2-c] pyridin-7-ylidene)amino)-1,3,5-triazin-2-yl) pyridin-1-ium chloride as shown in **Scheme 2**. The dyeing application of the water soluble analogue D2 was investigated in cotton fabrics which showing high exhaustion and fixation results. The quantum yield of fluorescence showing higher value in aqueous medium comparing with organic solvents.

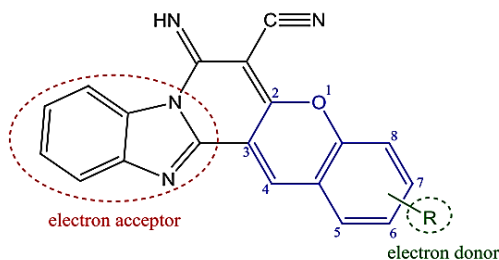
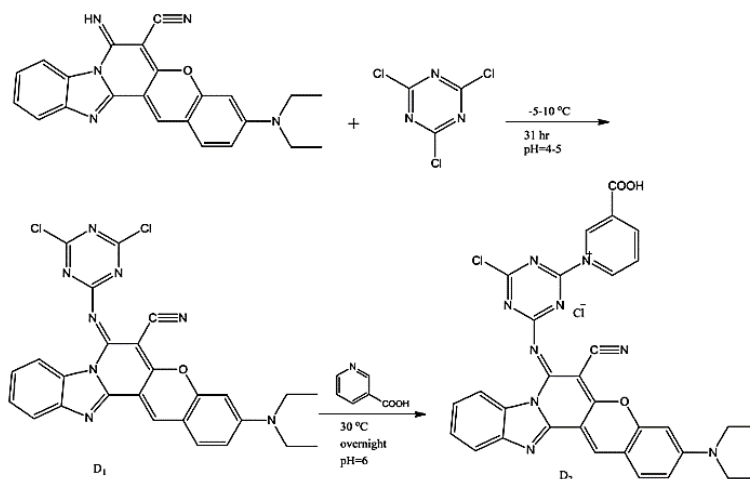


Fig. 27. General structure of fused iminocoumarin dyes.



Scheme 2. Synthetic procedure of the fluorescent reactive dye based on fused iminocoumarin.

The fluorescence intensity on dyed cotton fabric increased as dye concentration increased up to 2% o.w.f while a significant decrease observed at 4% o.w.f. This occurrence showed that dye accumulated at larger quantities on the fibre, reducing the fluorescence of the colored cloth as shown in **Figure 28**.

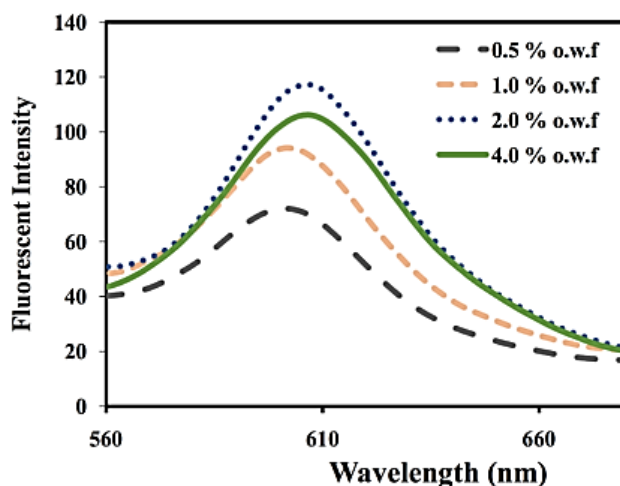
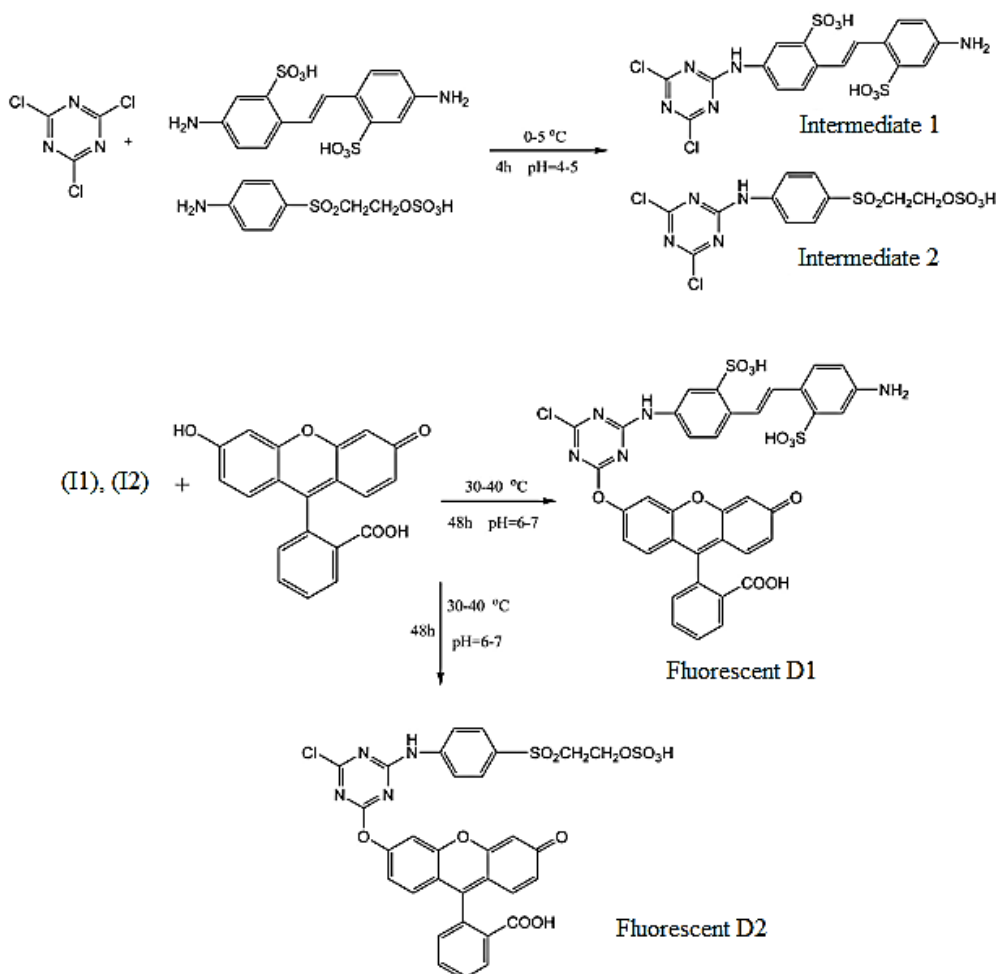


Fig. 28. Emission spectra of the dyed cotton fabric with D2 [99].

Maral Pishgar *et al* [100] report the synthesis and dyeing applications of new fluorescent reactive dyes based on xanthene dye derivatives (fluorescein). The synthesis was based on the preparation of two reactive intermediate bearing the reactive moiety (intermediate 1, 2) which obtained by the reaction of cyanuric chloride with amino-stilbene and with 4-amino sulphatoethylsulphone then followed by nucleophilic substitution reaction with fluorescein forming fluorescent dye 1 and 2 as shown in **Scheme 3**.



Scheme 3. Synthetic procedure of the fluorescent reactive dyes based on xanthene moiety.

The spectral study of the prepared dyes as presented in **Figure 29** showed, D1 have two distinct emission peaks at wavelengths between 300 and 400 nm for stilbene and 500 and 600 nm for fluorescein groups in aqueous medium.

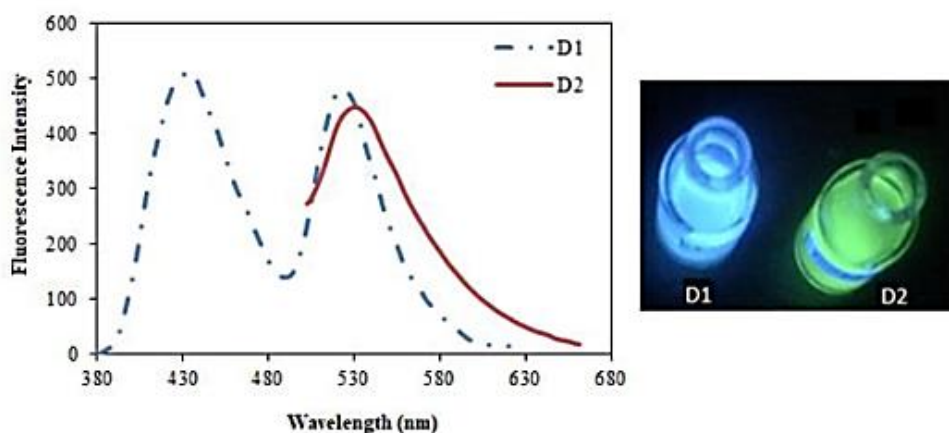
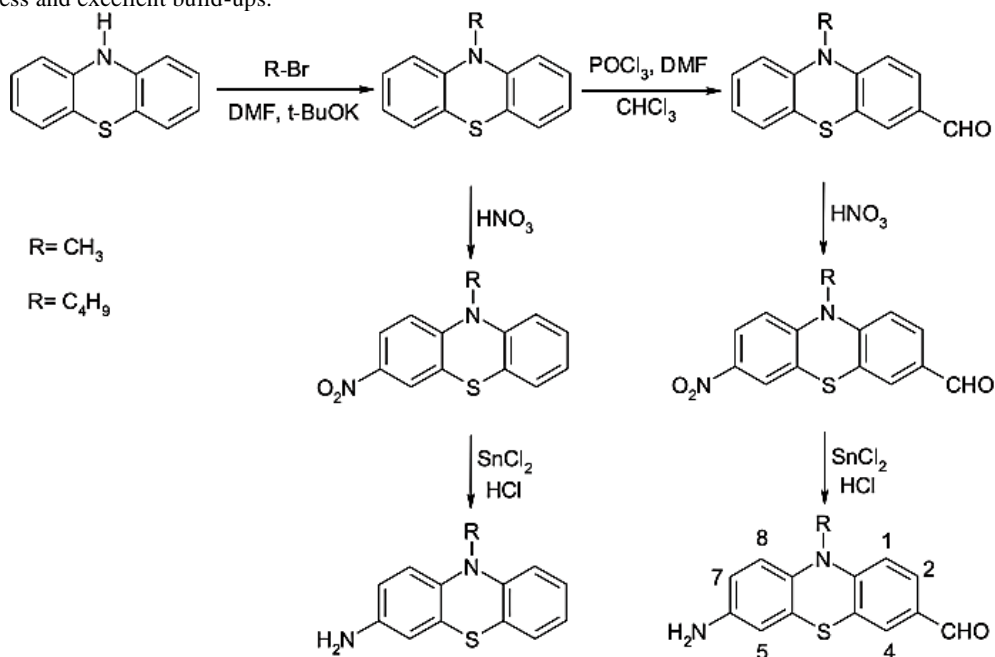


Fig. 29. Emission spectra of the prepared dyes [100].

The prepared fluorescent dyes 1, 2 was applied for dyeing cotton fabric which showing very good affinity to cotton fabric and the dyed fabrics showed a bluish emission in case of D1 while the emission of dyed fabric with D2 showing greenish-yellow emission.

Phenothiazine was used as the starting material to create a number of new fluorescent heterocyclic disperse dyestuffs by conventional reactions which reported by Hosseinezhad *et al*, [55]. The dyes was prepared by the nitration reaction of phenothiazine or phenothiazine carboxaldehyde derivatives as shown in **Scheme 4**, then oxidized, and finally reduced to obtain a series of fluorescent dyes based of pheothizine fluorophore.

The prepared dyes was showing positive solvatochromic properties by increasing the solvent polarity from toluene to DMF. In DMF, the prepared dyes showing good emission in DMF and the hue of fluorescence change from a yellow-green color. The greatest quantum yields were seen in dyestuffs that contained both electron acceptor and electron donor groups. The dyeing capabilities of the synthesized disperse dyestuffs were examined after they were applied to polyester fabrics at high temperatures. On polyester fabrics, the synthetic dyestuffs produced orange color ranges with acceptable colorfastness and excellent build-ups.

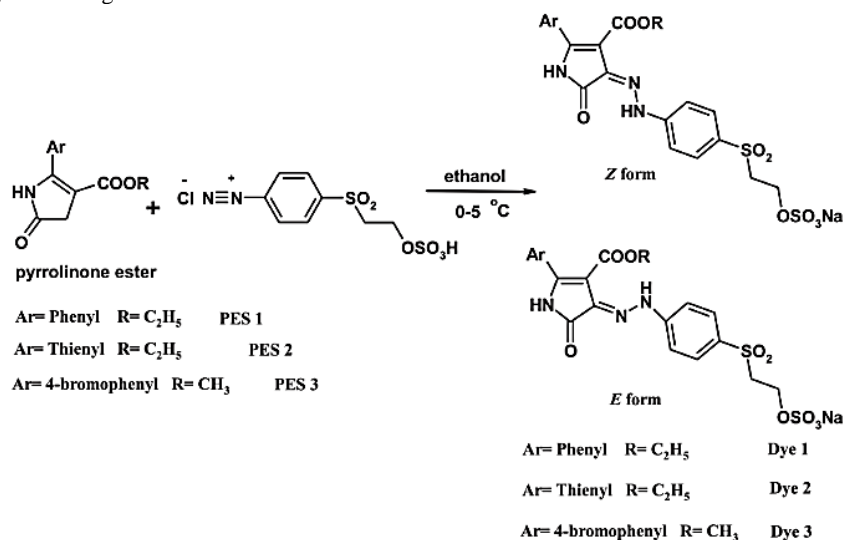


Scheme 4. Synthesis of new fluorescent disperse dyes based on phenothiazine moiety.

Recently Aysha *et al* presented a series of different fluorescent colorants based on pyrrolinone ester for textile applications [8-10]. Interesting reactive disperse dyes based on different analogue of pyrrolinone ester which coupled with a diazonium salt of *p*-aminobenzene sulphatoethylsulphone forming fluorescence reactive disperse dyes as shown in **Scheme 5**. The advantage of the prepared dyes is the easy and simple synthetic procedure and its high color strength observed.

The synthesized dyes exist in its pure hydrazone form as *E*, *Z* isomer as confirmed by NMR data and *Z* isomer the high abundant in the mixture with high fluorescent in polycrystalline phase (powder form).

The prepared dyes was applied for dyeing of polyamide and wool fabric which showing a significant high visible and bright colors and the emission in solutions as well as on the dyed fabrics as shown in Figure 30. These kind of dyes showing very good dyeing performance and color fastness in both polyamide and wool fabrics in additional to its high efficiency against gram⁺ and gram⁻ bacteria.



Scheme 5. The synthetic procedure of the reactive disperse dyes.

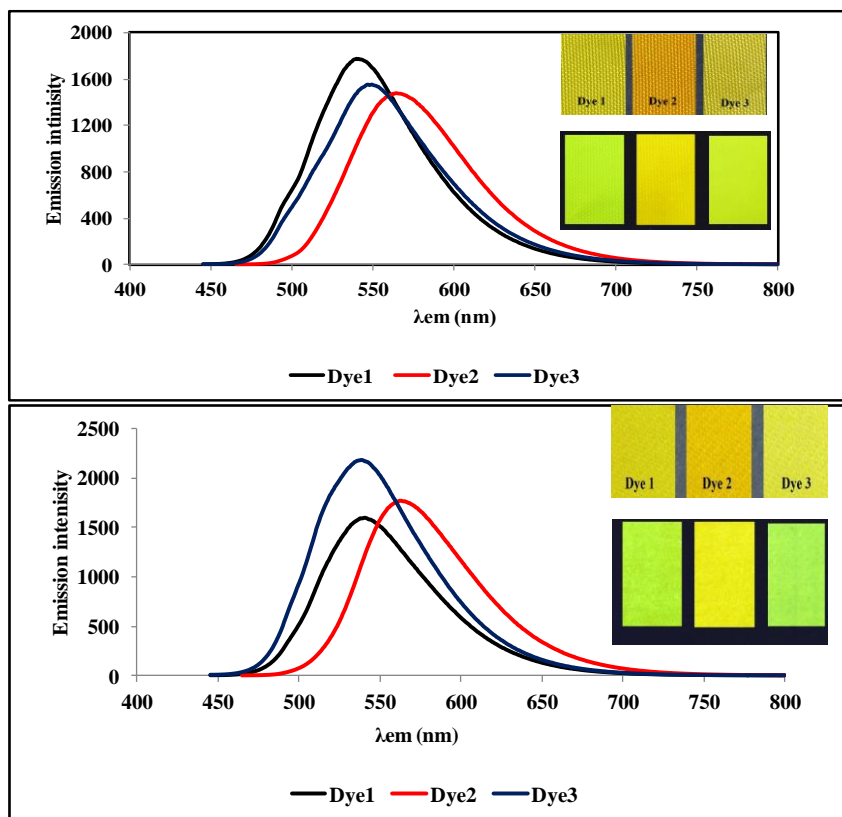
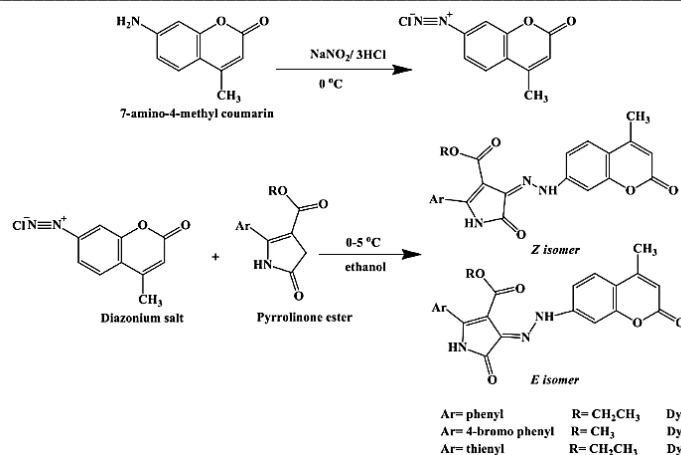


Fig. 30. Emission on the dyed polyamide (up graph) on wool (bottom graph) [10].

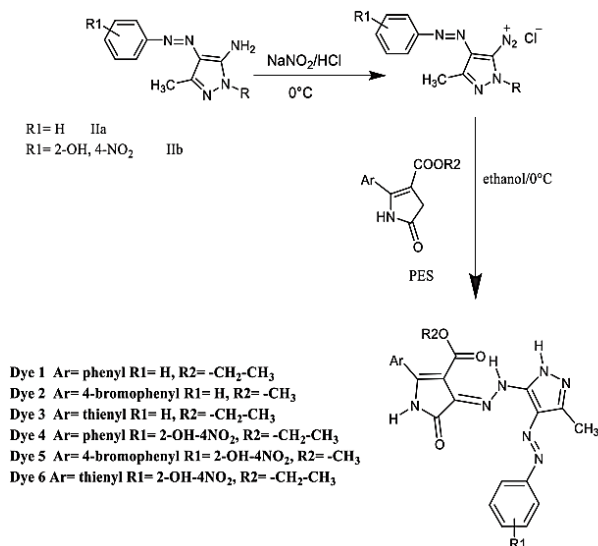
The synthesis of new solid state fluorescent dyes bearing amino-coumarin moiety was also investigated [9]. The synthesis of such fluorescent dyes was achieved by the coupling reaction of diazotized amino coumarin on the active methylene group in pyrrolinone ester as shown in Scheme 6.



Scheme 6. Synthetic procedure of the hydrazone dyes bearing coumarin moiety.

The prepared dyes also confirmed as pure hydrazone as a mixture of *E*, *Z* isomer. The synthesized dyes were used for dyeing polyester fabrics which showing high affinity for dyeing of polyester fabrics. And this dyes were showing good stability even if the dyeing process proceed under neutral or slightly alkaline condition.

One of the recent fluorescent dyes based on pyrrolinone ester was prepared by the same research group is a hybrid structure between azo pyrrazole and pyrrolinone ester hydrazone moiety as shown in **Scheme 7** [8]. A series of 6 dyes was synthesized and its spectral properties was studied which showing high emission on solid state and on dyed polyester fabrics as shown in **Figure 31**. The interesting feature of this series is associated with its alkali stable fluorescent properties for polyester dyeing which showing very good stability and no color change was observed even if the dyeing process continued under alkaline condition.



Scheme 7. Synthetic procedure of the solid state fluorescent azo-pyrrazole/hydrazone pyrrolinone ester dyes.

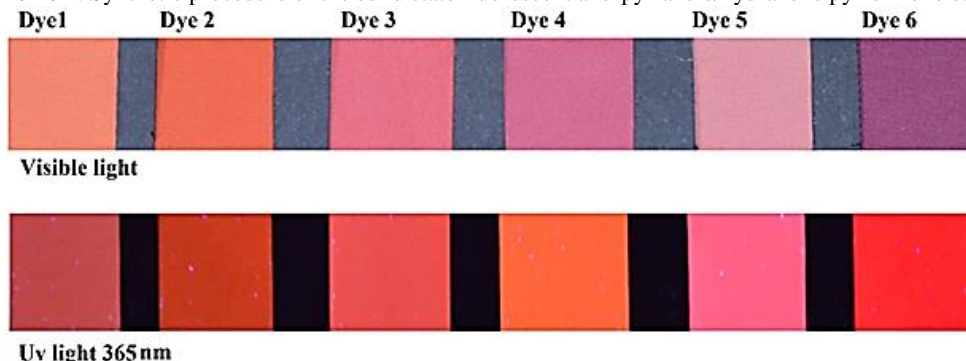


Fig. 31. Emission spectra of the prepared dyes on dyed polyester fabrics under UV light [8].

4. Non-textile applications of fluorescent dyes

Functional fluorescent dyes characterized by its functional application in hi-Tech application rather than it just a color. This state of art provides a brief summary of various uses, not only because of general curiosity but also because it is completely possible that some of these fluorescent dye applications may be used in the future by the developing field of smart and intelligent textiles.

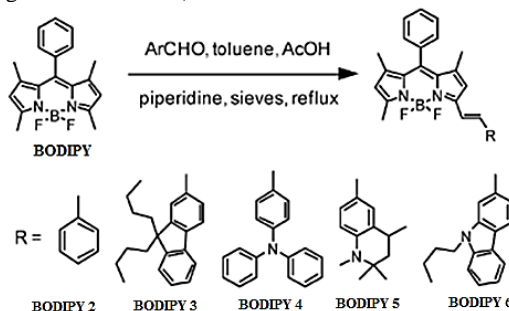
Fluorescent dyes are widely used in a variety of functional applications for instance, laser, OLEDs, DSSCs, fluorescence probes and medical applications in addition to some other applications as crack detection, plastics, synthetic resins, printing inks, non-destructive testing (NDT).

4.1. Electronic applications of Fluorescent dyes

4.1.1. Laser application of fluorescence dyes

Traditional inorganic lasers function in the visible, infrared, and ultraviolet spectrums suffering from disadvantage of emitting only at a few number of wavelengths and in extremely narrow emitting bands. The benefit of tunability across a large wavelength range is provided by fluorescent organic dye lasers, which produce a broad band emission bands in contrast with inorganic lasers. Fluorescent dyes work in dye lasers by absorbing a quantum of light and moving to higher energy sublevels of the first excited state S_1^* from the ground state S_0 . When incoming radiation interacts with the dye molecule when it is excited, fluorescent emission is produced as the molecule transitions to its ground state. This process is known as lasing. The dye molecules must predominately be in the excited state for this lasing effect to occur. By delivering a powerful inorganic laser pulse to the system, the necessary population inversion is accomplished. The stimulated emission from dye lasers is coherent (same phase and polarisation) and of high intensity in contrast to spontaneous emission. Dye lasers still find niche uses in communication technology, microsurgery, spectroscopy, and microanalysis despite significant advancements in solid-state and semi-conducting lasers progressively displacing them from their former dominant position.

Dakui Zhang *et al* [101], have been successfully synthesized new long-wavelength BODIPY dyes and the comparison of extending π -conjugation and tuning Intramolecular Charge Transfer (ICT) effect for the prepared dyes was discussed. The synthetic procedure of BODIPY(4,4-difluoro-4-bora-3a,4a-diaza-s-indacene) was presented in **Scheme 8**, different aryl derivatives of BODIPY was prepared by the reflux of BODIPY with different aromatic aldehyde in presence of a mixture solvents (toluene and glacial acetic acid).



Scheme 8. Synthetic procedure of the fluorescent BODIPY derivatives

The novel chromophores have good optical characteristics in polar or non-polar solvents, including strong fluorescence quantum yields, extraordinarily large molar extinction coefficients, narrow red-emission bands, and moderately significant Stokes shifts. Additionally, the novel dyes display very effective and steady laser emission that is adjustable from the green to NIR spectral range with transverse pumping at 532 nm (570–725 nm) as shown in **Figure 32**. Compounds BODIPY 4 and BODIPY 5, which have longer wavelength emissions and significant Stokes shifts due to the presence of triphenylamino and tetrahydroquinoline groups, respectively, which decrease the fluorescence quantum yield and wide emission bands in polar liquids. These photophysical characteristics allow the novel BODIPY derivatives to be classified as excellent laser dyes in the red-edge spectral region since they improve the laser action of well-known dyes that lase at the same wavelengths when considering lasing efficiency and photostability. Additionally, one of these novel BODIPY compounds (6) exhibits intense intracellular red fluorescence and cell membrane permeability which used in imaging of cancer cell. These positive traits guarantee that these dyes have the potential for biophotonic applications.

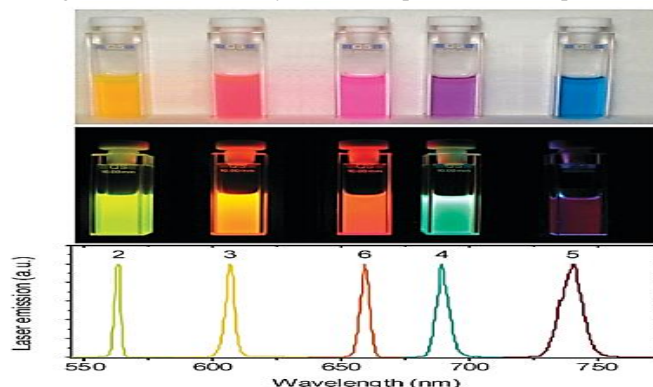
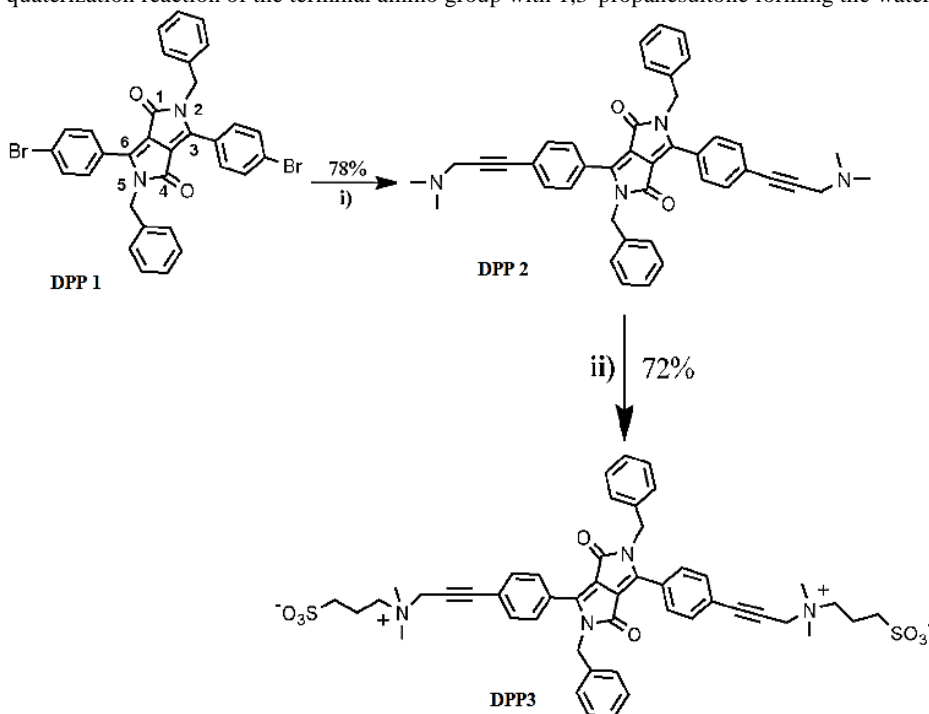


Fig. 32. Laser emission spectra of the BODIPY dye derivatives [101].

Diketopyrrolopyrrol(3,6-diaryl-2,5-dihydro-1,4-diketopyrrolo[3,4-c]pyrrole) based colorants is one of the interesting compounds due to its high fluorescence characters in addition to its high photostability, thus these compounds used in many applications such as inks, paints, DSCCs [102-105]. Soumyaditya Mula *et al.*, has modified new water soluble DPP derivative as shown in **Scheme 9** [106]. The synthesis of the dye DPP 3 was basically depend on the Sonogashira coupling reaction with the bromo DPP1 with dimethylaminopropyne forming DPP2 which followed by the formation of zwitterions by quaterization reaction of the terminal amino group with 1,3-propanesultone forming the water soluble DPP3.



Scheme 9. Synthetic procedure for DPP3 as laser dye.

The lasing measurements for the prepared dye DPP3 showing high lasing efficiency up to 14.6% with maximum narrow band lasing efficiency of 9.5% with a wide tunable range (554 nm to 616 nm) on excitation with a second harmonic of a Q-switched Nd:YAG (532 nm) laser in MeOH/H₂O as shown in **Figure 33**.

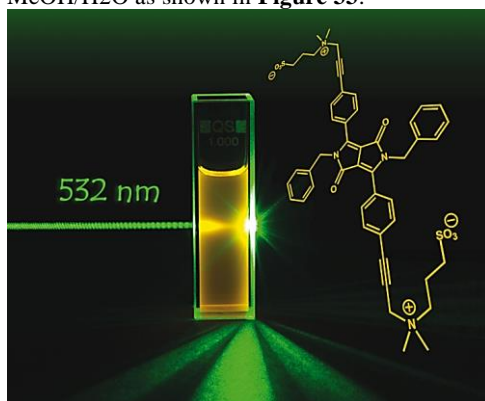
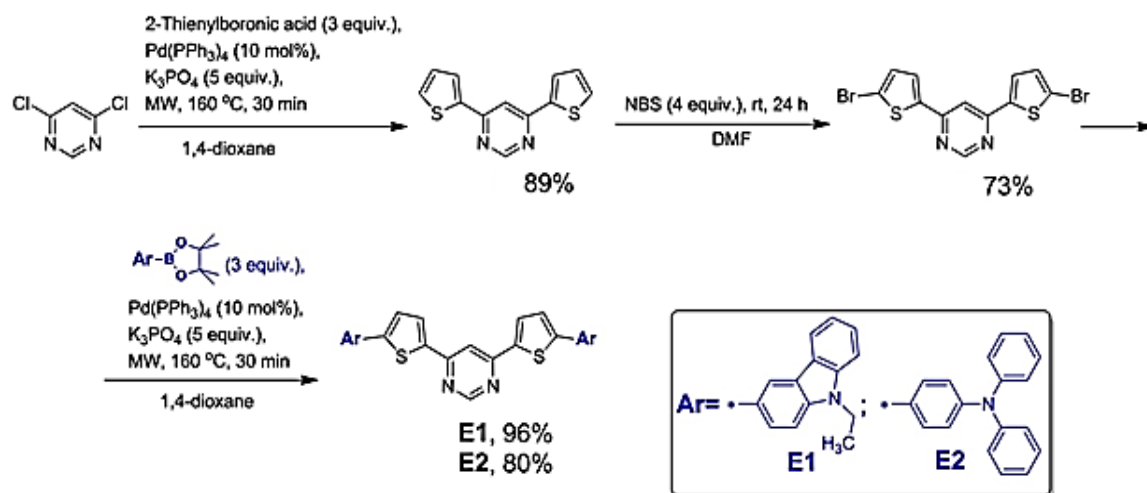


Fig. 33. Lasing performance of the prepared dye DPP3 [106].

Ekaterina M. Dinastiya *et al* [107] have been constructed and studied two new V-shaped push-pull systems (D- π -A- π -D) based on a pyrimidine acceptor bearing carbazole and triphenylamine and thiophenes as the π -linker for OLEDs (organic light emitting diodes) and laser applications as presented in **Scheme 10**. The synthetic procedure of pyrimidine based dyes was basically based on the Suzuki cross-coupling reaction on the halogenated pyrimidine followed by the extinction of the conjugation by further cross-coupling forming two fluorescence dyes E1, E2.



Scheme 10. Synthetic procedure of a push-pull based pyrimidine dyes E1, E2.

The emission bands of the dyes E1, E2 have a charge-transfer nature, as shown by low-temperature observations of the fluorescence and delayed luminescence spectra. OLED devices based on compounds in thermal vacuum deposition (TVD) films have great efficiency, which may be connected to the emission process through delayed fluorescence despite the compounds poor fluorescence quantum yield. It has been discovered that photoproducts produced after fluorophores in chloroform solution are exposed to UV light show laser activity in the red spectrum as shown in **Figure 34**. Push-pull systems have a high quantum yield of fluorescence in solutions but a poor quantum yield in TVD films. However, OLEDs made with these chemicals have very good current efficiency and brightness, which is presumably because delayed fluorescence exists. In addition, UV light swiftly converts chloroform solutions of these compounds into a photoproduct that pumps out laser light at a wavelength of 532 nm.

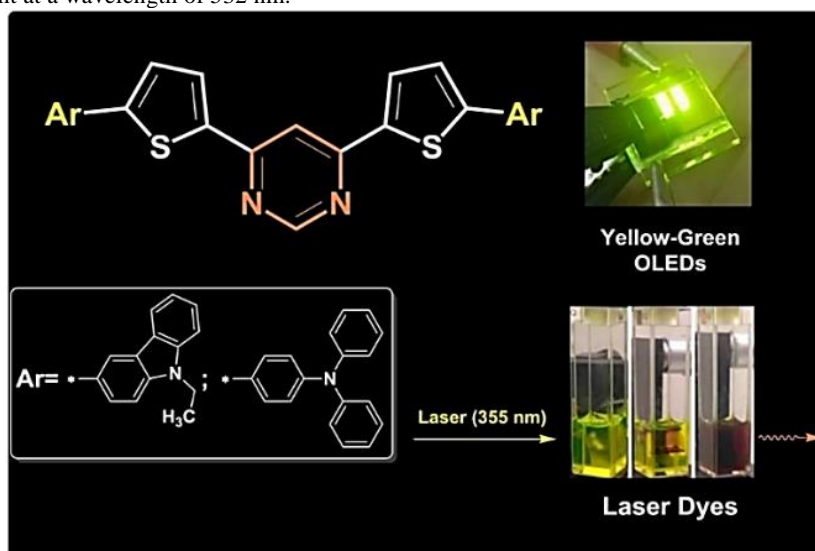


Fig. 34. laser and OLEDs performance of the synthesized dyes E1, E2 [107].

4.1.2. Organic light emitting diodes (OLEDs)

One of the most important applications of fluorescence and highly emissive dyes is OLEDs applications. Because of its tiny structure, low power consumption, high resolution, and self-emissive features, OLED offers numerous unmatched benefits over traditional display technology and is progressively emerging as a new trend in future displays [108-111]. OLED has received a lot of interest from both business and academics since it was developed in 1987 [112] because of the potential commercial uses for its low operating voltage and high brightness. The first flexible OLED device was created by Gustafsson et al. in 1992, and its ground breaking significance has sparked a new round of research [113].

OLEDs have successfully shown their wide variety of applications in displays, lighting, and medical devices owing to their mobility, low power consumption, and mechanical flexibility. The OLEDs have created new opportunities for wearable electronic gadgets due to the biological necessity and ongoing progress of electronic technology. Numerous papers have shown how flexible electrodes, processing technologies, and enhanced manufacturing are enabling OLED devices with good mechanical strain performance [114].

Traditional fluorescent OLEDs had a maximum internal quantum efficiency of 25% and could only utilise singlet excitons. As a result of their ability to exploit all singlet and triplet excitons via heavy atom effects or reverse intersystem crossover (RISC), the new generation of phosphorescence OLEDs type (PhOLEDs) and thermally activated delayed fluorescence (TADF) OLEDs are more well-liked. However, PhOLEDs and TADF OLEDs' light-emitting layers also have difficulties with triplet-triplet annihilation and concentration quenching (TTA). **Figure 35** showing the working mechanism diagram of OLEDs and its historical progress, fluorescent or phosphorescence dyes represent the most important part in the OLEDs components which is responsible for the emitting light and improve the OLEDs efficiency.

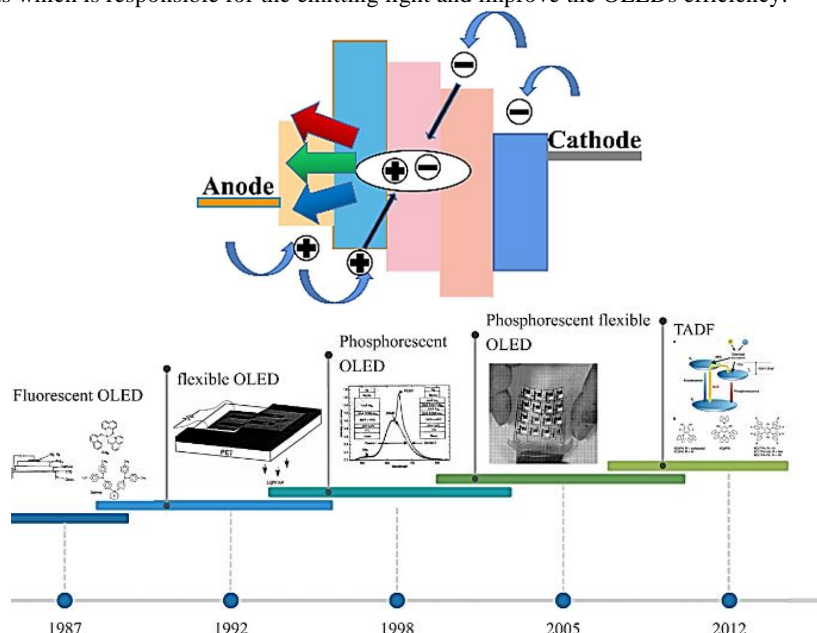


Fig. 35. OLEDs processing mechanism (upper image), present the history of self-emissive materials. Image for 'Fluorescent OLED' [112]. Copyright 1987, American Institute of Physics. Image for 'Fluorescent flex-OLED' (bottom image) [113].

Recently many published articles describe different fluorescent materials for OLEDs application based on varied luminescence system such as Carbazole/triazine, polyfluorene derivatives, Imidazolo pyridine, pyran-containing quinoline fluorophores, triphenylamine/phenanthroimidazole and tetraphenylethene-substituted anthracene and triphenyl amine derivatives [115-120].

4.1.3. Dye-sensitizer solar Cells (DSSCs)

The most effective and adaptable way to turn the cleanest and most numerous energy sources is to use dye-sensitized solar cells (DSSCs). In comparison to conventional solar cells, DSSC are somewhat transparent and more affordable. As a result, it may be a source of power in the future. However, more has to be done before it is deemed to be a marketable product.

Functional fluorescent dyes as sensitizer are essential for DSSCs fabrications because it starts the electrical current in the solar cells and absorbs visible light [121, 122]. The power conversion efficiency (PCE) of metal complex based-ruthenium dyes and porphyrin dyes, which are single sensitizer-based DSSCs, has achieved their maximum as of this writing, while metal-free organic dyes have also demonstrated a PCE of > 14% [123, 124] under normal illumination. For practical use in DSSCs, metal-based sensitizers have a limited number of drawbacks, including scarcity, difficulties in purification, environmental risks, and low molar extinction coefficients [125, 126]. The schematic diagram of the working mechanism of the DSSCs is presented in **Figure 36**, the functional dye is present an important part for the conversion of the solar energy to electricity.

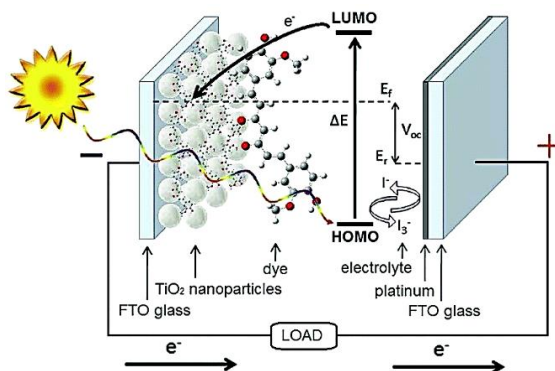


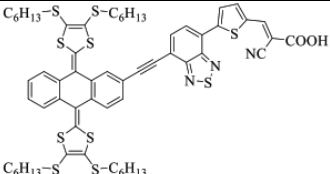
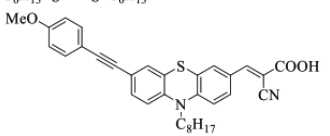
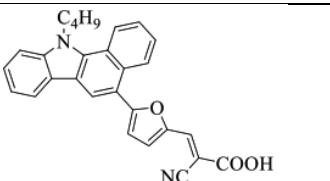
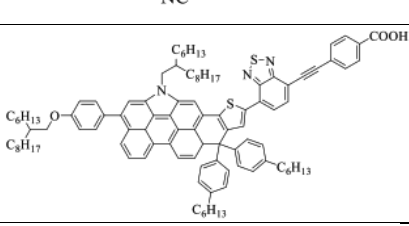
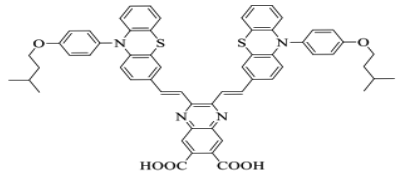
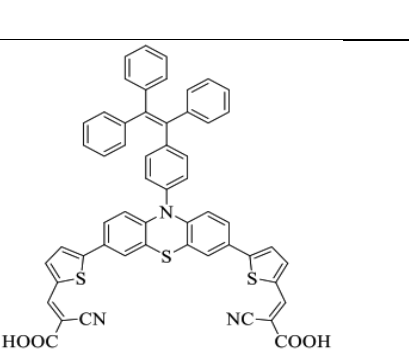
Figure 36: schematic diagram of the working mechanism of the DSSCs [127]

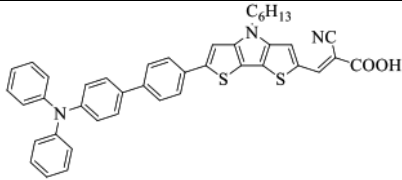
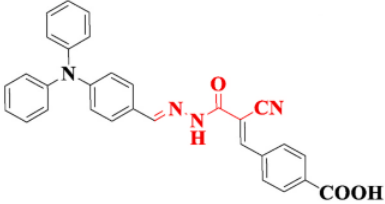
As a result, metal-free organic sensitizers have gained popularity over metal-based organic sensitizers because of their easy synthesis, great structural flexibility, low toxicity, and environmental friendliness [121, 122]. An electron donor (D), a π bridge, and an electron acceptor (A) in the shape of a D- π -A typically make up a metal-free organic sensitizer [128].

As electron donors, many chromophores with different electronic and electrostatic interactions properties are employed, including carbazole (CZ) [129-133], phenothiazine (PTZ) [127, 134], triphenylamine (TPA) [135-137], etc. - bridges are added to increase their capacity for light absorption and to facilitate intramolecular charge transfer. Numerous anchoring units, such as hydroxamate, pyridyl, hydroxyl, catechol, carboxylate, acetylacetonate, phosphonate, salicylate, sulfonate, and 8-hydroxyquinoline, have been successfully identified to attach the chromophore to the semiconductor [138].

There are crucial design requirements that must be met in order to manufacture more powerful sensitizers for DSSCs. To effectively inject electrons into the semiconductors, the sensitizer must first form a strong bond with the semiconductor surface (photoanode). Second, for successful charge injection, the dye's lowest unoccupied molecular orbital (LUMO) should be much higher than the semiconductor CB, and its highest occupied molecular orbital (HOMO) should be lower than the redox electrolyte medium [139-141]. A lot of research has been done over the years to enhance sensitizers in every way possible [142]. **Table 2** shows the chemical structures of the few of those that are provided here.

Table 2. Some example of dye-sensitizer solar cells and its efficiency

Dye structure	$\eta\%$	Ref.
	6	[143]
	6.21	[144]
	7.67	[145, 146]
	10.3	[147]
	4.46	[148]
	7.5	[149]

	6.15	[150]
	5.6	[151]

4.2. Analytical and sensing applications

Sensor materials are recently of great interest as their inclusion in a structure provides it with the "minimum" level of "smartness" by giving a sign about the presence of a stimulant and allows its recognition by easy, fast and low cost method, thus the application of the organic dyes based on its ratiometric fluorescence change (fluorescence probe) by binding with external stimulants such as mechanical, chemical, thermal, optical, magnetic, electric... etc. have been intensively studied in recent years [35, 152].

Fluorescent probes have drawn a lot of interest because of their high sensitivity, simplicity, and quick reaction times for applications in both optical imaging and analytical sensing. Fluorescent probes are often used for sensitive and highly specific quantitative analysis of analytes such as anion, cation and pH changes [29, 153-162].

Fluorescent probes worked in a reversible manner in the early stages of their development since they were predominantly created using molecular recognition and host-guest chemistry [163].

Contrarily, organic reactions have been used in recent years to create fluorescent probes that self-immolate or are reaction-based. However, using fluorescent probes with single emission features to quantify a target analyte can be challenging because various analyte independent factors, such as instrumental parameters, local concentrations of the probe molecules, photobleaching, and microenvironments can impede accurate analysis. The use of ratiometric techniques such as fluorescence probe is the most efficient method for resolving these problems and assuring reliability [164].

There are two main types of fluorescence probe (turn on/ turn off fluorescence probe) based on the optical behaviour change when its bind with external analytes as presented in **Figure 37**.

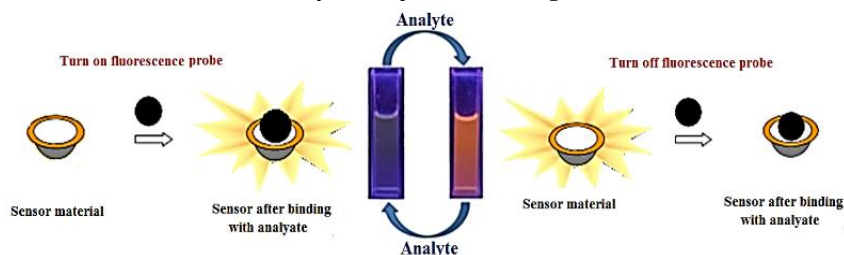


Fig. 37. Suggested mechanism of turn on and turn off fluorescence probe.

Several photophysical features, including as internal charge transfer (ICT), fluorescence resonance energy transfer (FRET), and excited-state intramolecular proton transfer (ESIPT), are often used in the creation of ratiometric fluorescent probes **Figure 38**. Large Stokes shifts are often seen in monomer-excimer production, ICT fluorophores made of conjugated electron-donating/electron withdrawing (donor/acceptor, D/A) moieties **Figure 38 a**. The interaction of one of the donor or acceptor moieties with an analyte causes the excitation/emission wavelengths to change blue or red shift can be observed upon the presence of the analyte. In order for FRET to work, energy must be transferred from the excited state of the donor fluorophore (DF) to its ground state of the acceptor fluorophore (AF) **Figure 38 b**. When the DF's emission spectrum and the AF's absorption spectra overlap well, effective FRET occurs.

The ability to generate excimers is often seen in flat polycyclic aromatic hydrocarbons like anthracene and pyrene **Figure 38 c**. An excimer is an excited state complex that is created when the excited state of one fluorophore of this kind interacts with the ground state of another fluorophore molecule. In this instance, the excimer emits redshifted and widened radiation. Fluorophores from ESIPT exhibit strong emission, light stability, and significant Stokes changes **Figure 38 d**. For instance, ESIPT can convert a favoured enol-form into an excited keto tautomer upon excitation. By reverse proton transfer, the stimulated keto-relaxation form's regenerates the enol form. Importantly, solvent polarity, hydrogen bonding ability, and pH frequently have an impact on the ESIPT process [165].

Cations and anions play an essential roles in various important human life processes [166-168], including osmotic regulation, metabolism, biomineralization, and signalling. The overdose concentration of metal ions are presenting one of the major threats that represent a danger to human life. The biologically important metal ion such as Cu^{2+} , Ca^{2+} , Fe^{3+} and Zn^{2+} should be exist in a suitable range of concentration to control the normal biochemical functions of the human body while the high concentration of Hg^{2+} and Pb^{2+} can be highly toxic. The change of these metal ion concentrations can affect the normal body and physiological functions directly [169]. Thus, the monitoring and quantitative determination of these

metal ions using an easy, fast, low cost even with ultra-low concentration became an important research challenge [170-173].

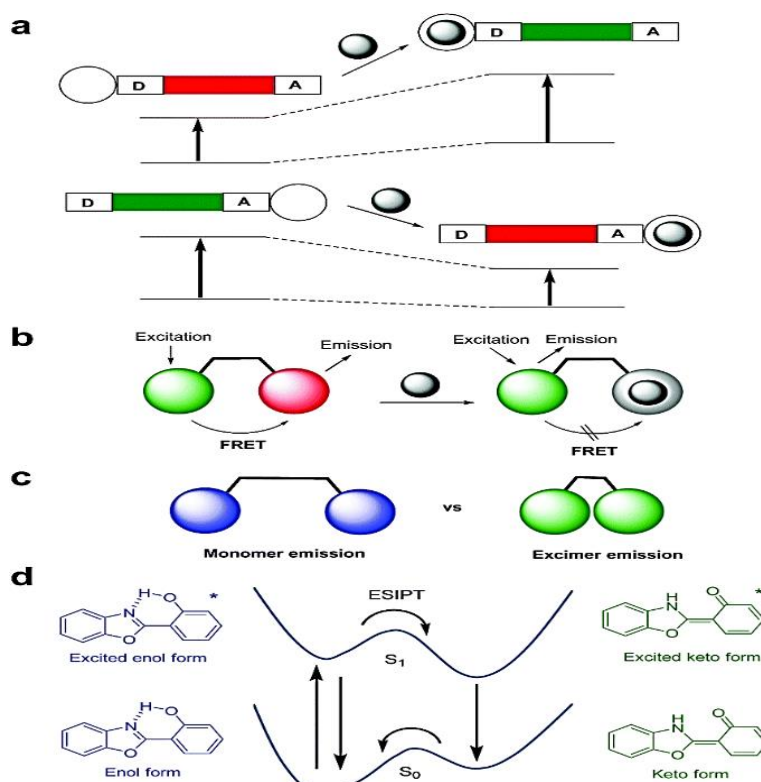


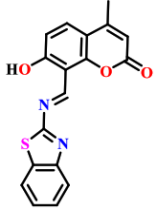
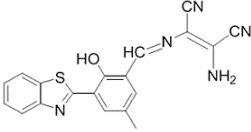
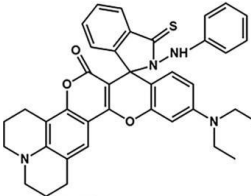
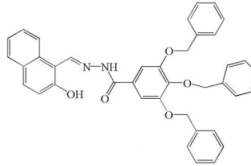
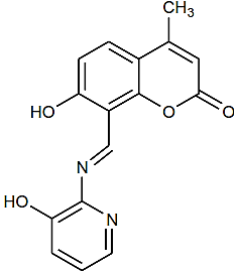
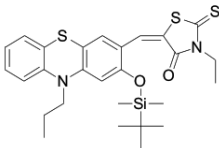
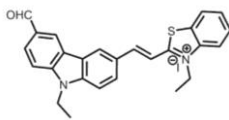
Fig. 38. Photophysical process in ratiometric fluorescence probe, internal charge transfer (ICT) a, fluorescence resonance energy transfer (FRET) b, monomer–excimer formation c and excited state intramolecular proton transfer (ESIPT) d [165].

Furthermore, both exogenous and endogenous anions in living systems alter typical biological processes, the detection of anions has received a lot of interest in recent years [12, 174, 175].

Due to the huge number of publications related to the fluorescence probe for detection of ions and imaging application we will present a few examples for some fluorescence probe for cations and anions in addition to the recorded detection limit was summarized in **Table 3**.

Table 3. Different examples of fluorescence probe for ions detection.

Probe structure	Target analytes	Detection Limit (LOD)	Ref.
	Cu^{2+}	105 nM	[176]
	Cu^{2+}	40.82 μM	[177]
	Cu^{2+}	0.17 μM	[178]

	$\text{Zn}^{2+}, \text{Mg}^{2+}$	17, 18 μM	[179]
	$\text{Al}^{3+}, \text{Zn}^{2+}$	7.06, 2.98 μM	[180]
	Hg^{2+}	0.34 μM	[181]
	$\text{F}^{-}, \text{Al}^{3+}$	0.148, 0.025 μM	[182]
	CN^{-}	0.0278 μM	[12]
	F^{-}	1.29 μM	[183]
	HSO_3^{-}	28 nM	[184]

4.3. Medical applications of fluorescent dyes

The application of fluorescence dyes in health related field is considered as one of the most important functional application and the researchers pay more attention for this topics. Since many applications in vitro or in vivo cannot be seen directly and must be viewed with the use of imaging equipment, thus the fluorescence change of some organic dyes could be very important techniques for this kind of application.

Longer absorption and emission wavelengths, such as those in the near-infrared I and II area, or NIR-I and NIR-II, can effectively reduce background interference from live organisms and enhance signal-to-noise ratio in fluorescence imaging [185]. Other energy conversion pathways of organic dyes, particularly photodynamic and photothermal conversion processes, have received a lot of attention recently in addition to the utilization of their luminous features. In photodynamic therapy (PDT) for skin cancer, breast cancer, and other malignancies, organic photosensitizers with high reactive oxygen species (ROS) or $^1\text{O}_2$ generation efficiency are frequently utilized [186].

For photoacoustic imaging guided treatment of tumor and cardiovascular disease, photothermal conversion would predominate if the energy received by the dye is primarily lost by nonradiative transitions [187].

These optical treatments offer the benefits of being noninvasive, deeply penetrating, having little side effects on healthy tissues, and being better at eliminating tumors. The inadequacies of conventional illness diagnostic and treatment

procedures can be solved, and new approaches can be offered, through the creation of imaging and therapeutic agents based on organic dyes.

Chemical modification procedures, which have made significant progress, are the most effective ways to achieve the functionalization of organic dyes. Two patterns may be seen in the evolution of dye chemistry. On the one hand, it is sought after to manipulate the optical properties of dyes (fluorescence, photothermal, and photodynamic ability) for appropriate application [188, 189].

On the other hand, organic dyes should have properties like water solubility, photostability, targeting ability, and so on for applications that are relevant to health. Additionally, as nanotechnology advances and becomes more sophisticated, nanomaterials based on functional organic dyes also developed [190].

4.3.1. The dyes design for medical application

Functional organic dyes are chemically modified utilizing an application-focused technique that concentrates on the end material's optical characteristics, stability, water solubility, biological safety, and application features.

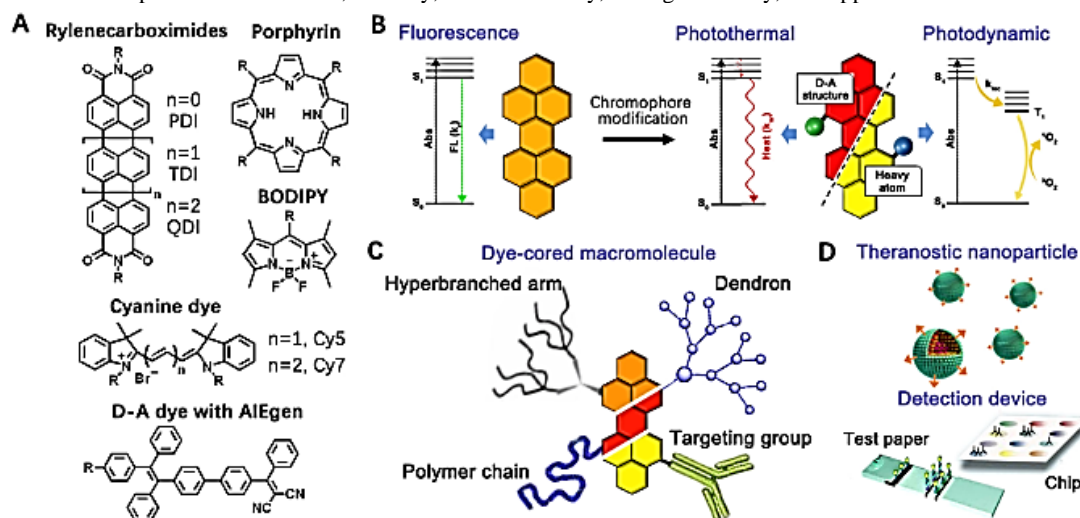


Fig. 39. Some organic dyes structure (A). Fluorescent, photothermal (B), and photodynamic performance control of dye. Abs, absorbance, Non-radiative decay rate, intersystem crossing rate. Scheme of functionalized dye-cored macromolecule (C). Functional organic dye-based materials (D) [190].

A list of several organic dyes chemical compositions may be seen in **Figure 39 A**. The direction in which organic dyes are applied is determined by chemical alteration of the chromophore scaffold. The chromophore undergoes excitation after light absorption, and the energy is released in a variety of ways, including fluorescence, heat, or chemical energy **Figure 39 B**. These dyes' energy conversion pathways can be altered chemically to produce a variety of fluorescent compounds, photothermal molecules, or photosensitizers [190, 191]. The fluorescent bioimaging, for instance, dyes like perylene diimide (PDI), cyanine dye, boron dipyrromethene (BODIPY) dye, and their derivatives need to be created with high fluorescence quantum yields.

The dye's absorption wavelength can be redshifted by adding a donor-acceptor (D-A) structure or lengthening the conjugate, which would considerably improve the performance of $^1\text{O}_2$ or heat generation and have applications in PDT or PTT of malignancies [192]. A photosensitizer with a high $^1\text{O}_2$ quantum yield may be produced for an effective PDT if heavy atoms or metals are added [193].

Additionally, organic dyes with considerably red-shifted emission and absorption spectra were developed and exploited in biomedical applications, particularly in in vivo scenarios, through the development of D-A structures and conjugation elongation [194].

Large conjugated structures give organic dyes their high hydrophobicity and ability to agglomerate. In order to create hydrophilic macromolecules for biological applications, water soluble modification techniques are necessary **Figure 39C**. For modification, conventional techniques employ hydrophilic functional groups (like carboxyl, amino, or zwitterionic groups) or hydrophilic polymers (like dendron, hyperbranched arm, PEG chains, and polyamino acid). For fluorescence imaging, drug delivery, and anticancer research, topological "core-shell" macromolecular structures with organic dyes as the core and hydrophilic molecules as the shell have been designed recently which produced satisfying results [195].

In the field of in vitro detection, organic dyes are very often utilized. Fluorescent molecules can be made into particular fluorescent probes for the detection of gases, ions, disease biomarkers, or antibodies by adding the appropriate stimuli-responsive group. Simple polymer-based fluorescent films or test papers as well as more intricate fluorescent array chips for fluorescence microscope observation are examples of common fluorescent probes **Figure 39D** [196]. The characteristics of the dye molecules in combination with the substrate and the coupling technique are crucial factors to take into account in these applications.

4.3.2. *In vivo* diagnosis, tumor diagnosis

Organic dye-based imaging techniques, such as fluorescence imaging and PA imaging, are commonly used for *in vivo* imaging of damaged organs or tissues. These techniques offer advantages over standard imaging methods for *in vivo* diagnosis as they are real-time, noninvasive, and cost-effective. Urano et al. developed the fluorescent probe HMRef-Gal using an enhanced spirocyclization method for *in vivo* visualization of peritoneal metastatic cancers [197]. The probe can sensitively detect galactosidase, which is substantially overexpressed in early ovarian tumours, and is membrane-permeable **Figure 40A**.

Fluorescence imaging of tiny peritoneal metastases was effectively accomplished in mice models of ovarian cancer by the authors **Figure 40B**, demonstrating the significant potential for early detection of metastases. With better penetration, signal-to-background ratio, and spatial resolution compared to conventional dyes, NIR-II organic dyes with emission wavelengths of 900–1700 nm can be utilized to diagnose deep malignancies such lung and brain tumors [198]. A NIR-II fluorescent probe called CH1055-PEG was successfully created and utilized for brain tumor imaging by Hong and colleagues [199]. The probe has high water solubility and a peak fluorescence emission at around 1055 nm. Additionally, the probe's comparatively modest molecular weight of 8.9 kDa makes it easier for the kidneys to eliminate it (Figure 4C). The probe was employed for noninvasive imaging of a brain tumor at a depth of 4 mm due to its NIR-II fluorescence and acceptable biological safety.

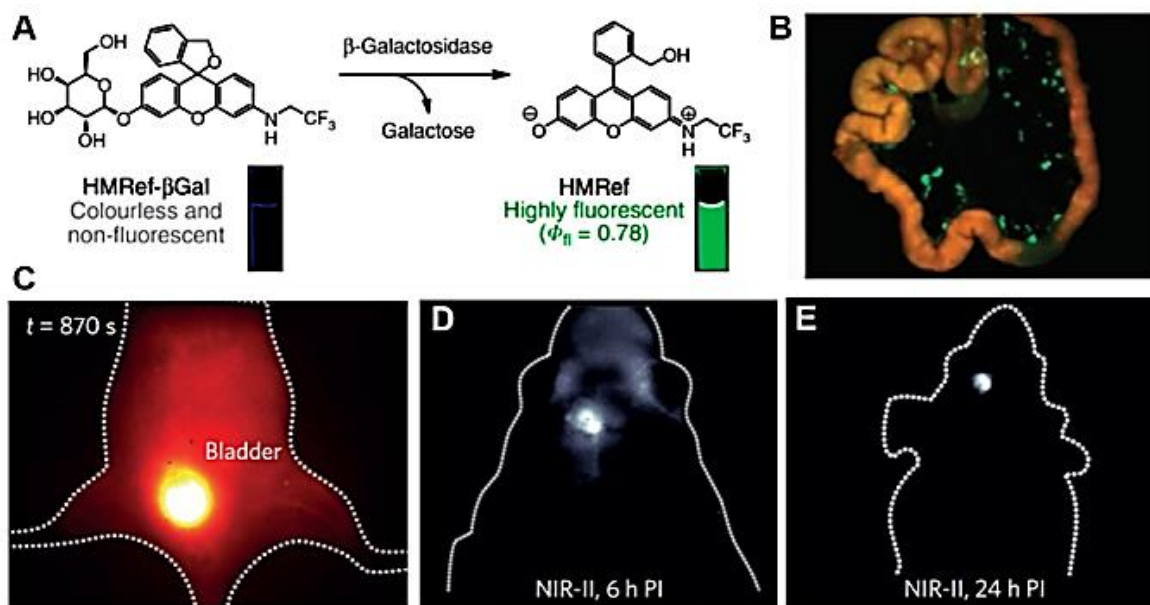


Fig. 40. Chemical structure of HMRef-βGal and activation of HMRef-βGal on enzymatic reaction with β-galactosidase A). Fluorescence imaging of peritoneal SHIN3 metastasis with HMRef-βGal (B). NIR-II imaging of a mouse in the supine position 870 s after intravenous injection of CH1055-PEG (C). Non-invasive imaging of brain tumour at 4 mm depth with CH1055-PEG 6 h postintravenous injection (D). Whole-body NIR-II fluorescence imaging with CH1055-PEG 24 h postintravenous injection (E) [199].

Applying phosphorescent probes with extended fluorescence lifetimes, which may substantially reduce background fluorescence, is another strategy for enhancing the signal-to-background ratio of cancer imaging. Jiang et al., for example, described an iridium-based NIR-I fluorescence sensor that may be initiated by the hypoxic microenvironment associated with cancer. With excellent sensitivity and specificity, the probe may identify lung metastasis of cancer cells through the bloodstream or lymphatics upon intravenous administration.

To summarize, NIR-II fluorescence and phosphorescent imaging are two efficient ways to improve signal-to-background ratio, showing significant promise for cancer diagnosis [200].

Many other application of fluorescence dyes such as bacteria monitoring and imaging [201], drug delivery [202], Cardiovascular diseases diagnosis [203], Imaging-guided phototherapy [204] and chemotherapy have recently played a great research attention for health-related applications.

5. Conclusion

Fluorescent dyes play a unique role in textiles, especially when high visibility is crucial. With the leisurewear and sportswear industries expected to grow, along with increasing safety concerns globally, the demand for protective workwear using fluorescent dyes and pigments is likely to increase. The functional applications of fluorescent dyes, such as in solar-energy conversion, analytical and biomedical fields, and illumination devices, will continue to attract interest from researchers in academia and industry. Recent research reports suggest that functional fluorescent dyes could potentially be used in smart textiles in the future. The light fastness properties of commercial fluorescent dyes used in textiles are often criticized for not meeting the requirements of demanding applications. However, the addition of light stabilizing additives, like UV absorbers, can improve this property. Manufacturers are increasingly incorporating renewable raw materials, such as natural fibers, in response to the growing interest in environmentally friendly solutions. Fluorescent dyes are essential in the textiles industry, providing visibility and standing out in various applications. The use of fluorescent dyes and pigments

in textiles is expected to grow, especially in leisurewear, sportswear, and protective workwear. There is potential for functional fluorescent dyes to be integrated into smart fabrics in the future. Researchers are interested in the applications of fluorescent dyes in solar energy conversion, analytical and biomedical fields, illumination, and display devices. Enhancing fluorescent dyes for high-performance applications, like DSCCs, lasers, and display devices, is a significant research challenge. Improvements are needed for the use of fluorescent dyes in complex detection scenarios for imaging and analytical applications, including probe compatibility with organisms and precision in sensing. Fluorescent probes are expected to advance biology and become a valuable tool in the biomedical field.

6. Conflicts of interest

"There are no conflicts to declare".

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9. References

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