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Molecular switches and their applications in intelligent paper, textile and leather substrates

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ABSTRACT

In most general term a molecular switch is a switch which shows reversible change when exposed to certain type of stimulus (such as, light, pH, temperature, electrical field, magnetic field, pressure). When this change can be controlled and regenerated using specific techniques, it can be used to develop products which have many application potentials ranging from the products in everyday life (for examples, cosmetic, fashion items, optoelectronics, display devices) to mega scale industrial exploitation for many high-tech applications (such as, controlled drug delivery, remote control, security, brand protection, data storage). There are different types of stimuli-responsive molecular switches where some of most frequently used classes include – (a) photochromic, (b) thermochromic, (c) ionochromic, (d) electrochromic and (e) piezochromic switches. In usual consideration, smart surfaces are capable to sense surrounding environment which are mostly based on synthetic polymers. However, relatively less attention has been focused on the use of porous flexible natural polymers (for examples, cellulose materials like paper, cotton and protein substrates like wool, silk and leather). These flexible natural polymers (such as, natural fibre based textiles and leather substrates from animal hides and skins) have the potential to be functionalized using these stimuli-responsive functional materials for value addition and also for aesthetic and advanced high-tech applications (examples include - self-indicating radiation warning systems, controlled detection systems, purpose built localized display systems along with many other application possibilities). This current paper will very briefly discuss some of the core points of these advanced stimuli-responsive materials for their possible applications for producing intelligent paper, textile and leather substrates for advanced applications.

Keywords: *molecular switches, intelligent natural flexible substrates (paper, textile, leather), photochromic, thermochromic, ionochromic, electrochromic and piezochromic.*

1. INTRODUCTION

Molecular switches are the materials which can toggle between two (or more) stable states in order to encode various physical features. When these materials are applied on complex systems (such as, flexible or rigid polymeric systems) they allow complex systems to respond to changes in the physical stimuli of surrounding environment. Some of these types of stimuli include – (a) light, (b) heat, (c) pH, (d) electrical field, (e) pressure, etc. Due to this environmental stimuli-responsive behaviours, these molecules offer many potential advanced applications in sensor technology, biology and information technology as well in other application areas [1-10]. Researchers are continuously working to develop new molecular switches and have already developed many synthetic switches which can complement or artificially imitate many types of biological switches usually found in nature [12-14]. Based on the response to certain stimulus molecular switches can be grouped into different categories some of which include – (a) photochromic (responsive to sunlight or UV light), (b) thermochromics (responsive to heat), (c) ionochromic (responsive to certain ions or pH), (d) electrochromic (responsive to electrical field), (e) piezochromic (responsive to pressure). Selectively some particular molecular switches are very briefly covered in this paper. Sometimes molecular switches are also termed as functional stimuli-responsive materials. Functional stimuli-responsive materials can be applied to natural flexible substrates (such as natural fibre based textiles, paper and leather) to produce environmentally responsive surfaces based on these substrates. When these surfaces react with environmental stimuli to produce observable change (in terms of surface colour) and revert back to original condition without or with negligible change or deformation in original structure (before exposure to the stimulus) can typically be termed as environmentally responsive intelligent surfaces based on paper, textile and leather or more simply intelligent paper, textile or leather substrates. Additionally, within the framework of certain restrictions they can also be termed as smart substrates. Smart flexible materials (such as, paper, textile, leather, etc.) have been in focus for a considerable period for their very high industrial application potentials. For instance, smart textiles have been successful to draw very high levels of active current research interests for their very high application potentials in terms of conventional and advanced applications [14-45]. Additionally, the wide range of application potentials of intelligent and smart flexible materials made them attractive to a number of associated interdisciplinary research areas. There are various ways for producing intelligent paper, textile and leather substrates; one of the methods is to incorporate environmentally stimuli responsive switches by using digital dyeing technique where digitally dyed surface can sense the surrounding environment. These types of stimuli-responsive smart paper, textile and leather have many potential applications some of which include – (a) safety, (b) security, (c) comfort, (d) fashion, (e) health care, (f) remote sensing, (g) biomedical and intelligent applications. This current paper briefly focuses on different aspects of intelligent paper, textile and leather substrates and a few selected methods in order to produce these material based responsive surfaces using different stimuli-responsive molecular switches (such as, photochromic, thermochromic, ionochromic, electrochromic and piezochromic molecular switches). Additionally, the successful incorporation of these stimuli-responsive switches into the structure of these flexible materials needs certain level of basic understandings on the nature of these molecular switches, as a result a brief discussion has been included along with very selected examples of practical applications of particular switches (photochromic and ionochromic switches) to paper, textile and leather substrates.

2. INTELLIGENT FLEXIBLE SUBSTRATES BASED ON PAPER, TEXTILE AND LEATHER SUBSTRATES

In last two decades, there have been many multi-disciplinary approaches in flexible materials (especially in textile research) to incorporate multi-level of functionalities along with other distinct features usually expected from usual as well as special types of clothing based on smart and intelligent textiles along with other useful features sometimes expected from ordinary textiles. With the advancements in different related technologies (such as, material sciences, nanotechnology, information technology, biotechnology, flexible display techniques, etc.) now there are scopes for new possibilities to enhance textiles with a variety of new functionalities. Some of these possibilities include the production of multifunctional new fibre structures, composite materials and coatings at the nano and micro levels to the visible integration of wearble electronic assemblies into clothing system. Currently, there is a high level of convergence of different sophisticated technologies, for example, biochemistry, polymer chemistry and computer processor based techniques which are miniaturised to produce 'lab-on-a-chip' diagnostics, and new forms of textile sensors, actuators and other components that are now available. As a result, many high quality previous efforts to produce high performance functional and intuitively wearable computing systems based on textiles are slowly becoming a practical reality [14-45]. Similarly, paper based electronic displays and their uses in packaging have been in the fore point for certain specific fields of research on flexible materials. Intelligent paper have many applications some of which include – (a) uses as chemical and biological sensors, (b) biomedical applications, (c) applications in display devices, (d) packaging, (e) security and brand protections. Similarly, intelligent textiles incorporated with different functionalities have many uses in a variety of fields, some of them are widely used in the fields of biomedical or healthcare applications. For more specific information, some of the selected application areas precisely included here are – (a) medical textiles (in general), (b) smart wound-care materials, (c) textile-based drug release systems, (d) phase change and shape memory based smart textiles for different applications, (e) textile based sensors for healthcare, (f) smart medical textiles for particular types of patient (e.g. intelligent garments for pre-hospital emergency care), (g) smart medical textiles in rehabilitation, (h) smart medical textiles for monitoring pregnancy, (i) smart textiles for monitoring children in hospital, (j) wearable textiles for rehabilitation of disabled patients using pneumatic systems, (k) wearable assistants for mobile health monitoring, (l) smart medical textiles for monitoring patients with heart conditions, and (m) textiles in surgical implants, tissue engineering and wound care [12-45]. In this context, intelligent leather substrates and their products may be suitable for some the above stated application areas, however, more distinctive and precise application areas may include the following areas – (a) expressive footwear and highly exotic functional leather products, (b) textile –leather composites for specific applications in automobiles, upholstery and furniture leather and their products, (c) bag leather, (d) fancy book binding leather, (e) special type of patent leather, (f) laminated plastic leather composite surfaces for various applications [8-12].

3. ADVANCED MATERIALS AND TECHNOLOGIES FOR PRODUCING INTELLIGENT PAPER, TEXTILE AND LEATHER SUBSTRATES

Current trends in the manufacture of intelligent paper, textile and leather substrates involve a variety of established and also emerging technologies with different level of adaptations in different areas [15-40]. Thus, currently it is extremely important to have meaningful interdisciplinary collaboration for high quality product development. As for instance, in order to produce textile based electronic products, the electronics industry is concentrating on required level of knowledge on different types of textiles (where electronics are increasingly incorporated into clothing systems to produce specific products) to solve problems in wearable electronics. So, it is important to have fundamental knowledge on standard commercial textile production methods along with the knowledge of wearable electronics for an effective adaptation of new developments in both electronics and in textile to produce high quality textile based electronics for specific applications in different target areas within affordable costs [31-40]. As a recent trend, different materials have been incorporated into the structure of textile or related materials used in the clothing systems to serve different purposes, some other advanced materials and techniques have also been used to improve the performance of smart (to some cases also for biomedical) textiles, some of the most important materials and method are – (i) phase change materials, (ii) photochromic materials, (iii) thermochromic materials, (iv) shape memory alloys and polymers, (v) conductive fibres and yarns – metals, wires and conductive polymers, (vi) quantum tunnelling composites for switching devices, (vii) piezoelectric resistance, (viii) organic or plastic electronics, (ix) biomaterials, (x) light-emitting polymers, (xi) light-emitting diodes, (xii) fibre optics, (xiii) photovoltaic and solar cells, (xiv) photoluminescence, (xv) holography, (xvi) plasma technologies, (xvii) nano technologies for fibre and fabric coating, (xviii) micro encapsulation for therapy delivery, (xix) global positioning and wireless communications, (xx) radio frequency identification (RFID) tags, and (xxi) micro-electronic mechanical systems (MEMS) [10-50]. Similarly, there are many attempts for the applications of paper based products in electronic, display and sensoric applications along with many other distinctive application areas where functional materials are used to produce these advanced products. In the same way, there are few reports of using functional materials for their applications on leather and leather products for conventional and high-tech applications [9, 13]. Very selected examples of different features of intelligent paper, textile and leather substrates produced by using selected molecular switches are precisely included in this current paper.

4. MOLECULAR SWITCHES AND THEIR APPLICATIONS FOR PRODUCING INTELLIGENT PAPER, TEXTILE AND LEATHER SUBSTRATES

A molecular switch is a switch which can exist in more than one (usually two or more) stable states (more specifically metastable states) where it shows significant difference in physical, chemical or biological properties. These properties can be reversibly shifted between these metastable states in response to external stimuli, such as, change in pH, light, temperature, electrical current or potential [1-14, 50-54]. As for instance, pH indicators are well-known examples of synthetic molecular switches which show distinct colours as a function of a change in pH. As a result, a simple molecular switch has a potential (when incorporated in a complex molecular system) to act as a trigger to turn off and on important physical, chemical or biological properties of the system, such as surface wettability, polymer elasticity, host-guest recognition, catalysis, enzyme activity, fluorescence, neural activity and membrane activity for intracellular drug delivery [1-14, 50-54]. However, for this current paper a particular emphasis is given on the nature of colour changes of selective molecular switches when exposed to particular environmental stimuli (such as, light, pH, temperature) with a specific aim to integrate the system in terms of the applications of the stimuli-responsive flexible surfaces for environmental sensing (more specifically producing a notable colour change) for a variety of purposes.

Different types of molecular switches show colour change due to a number of reasons (detail discussion on the mechanism is beyond the scope of this current paper), some of the important reasons include – (a) change in conjugation in molecular structures (for example, spirooxazine, spiropyan, naphthopyran based photochromic molecular switches), (b) vibrations and excitations, (c) transitions between molecular orbitals, (d) transitions between energy bands, (e) change in geometrical and physical properties [47-54]. Molecular switches show their presence in different metastable state when exposed to different environmental stimuli, so in that sense they also act as chromic switches. The nature and other selected aspects of some particular types of molecular switches are briefly stated in Table 1.

Table 1. Different major types of molecular switches.

Materials or switches	Usual responsive stimulus	Usual nature of changes	Example
Photochromic	UV light, sunlight (and heat in some cases)	Colour, geometric shape, dipole moment, refractive index, etc.	Spiropyrans, spirooxazines, diarylethenes, chromenes, fulgides
Thermochromic	Temperature	Colour, geometric shape, refractive index, etc.	Spirolactones, bianthrones, etc.
Ionochromic/Halochromic	pH, ions	Colour, producing different ionic form	Phenol Red, Cresol Red, Bromocresol Green, Thymol Blue
Electrochromic	Electrical field	Colour	Viologens
Piezochromic	Pressure, stress	Shape, colour	tris(4-(4-phenylquinolin-2-yl)phenyl)amine, tris(4-(6-(9H-carbazol-9-yl)-4-phenylquinolin-2-yl)phenyl)amine

5. PHOTOCHROMIC MOLECULAR SWITCHES

A photochromic switch is a material which changes colour reversibly and controllably under the effect of ultra-violet irradiation. Photochromic materials (especially, spirooxazines, spiropyrans, naphthopyrans, fulgides, diarylethenes) have attracted intensive current research interest due to their variety of application potentials including in sun-screening, security printing and optical data storage. Photochromic dyes are commonly applied in a polymer matrix. There are many potentially promising applications of photochromic dyes on paper, textile and leather substrates in order to produce novelty or fashionable colour change based design effects on paper, textile and leather which can serve as a conventional item for fashion and design and can also be used for sensing surrounding environments. Some of many potential applications include – (a) camouflage for military purposes, (b) security, (c) brand protection, (d) authentication, (e) UV warning systems, (f) applications in fashion and designs, (g) applications in novelty items.

6. PHOTOCHROMIC MOLECULAR SWITCHES RELEVANT TO INTELLIGENT PAPER, TEXTILE AND LEATHER SUBSTRATES

Some of the most important photochromic molecular switches which can be relevant to the application in intelligent paper, textile and leather substrates are – (a) spirooxazines, (b) spiropyrans, (c) naphthopyrans, (d) diarylethenes, (e) fulgides, (f) fulgimides, (g) azulenes (dihydroindolyzenes) and (h) some other particular types of photochromic supra-molecular switches.

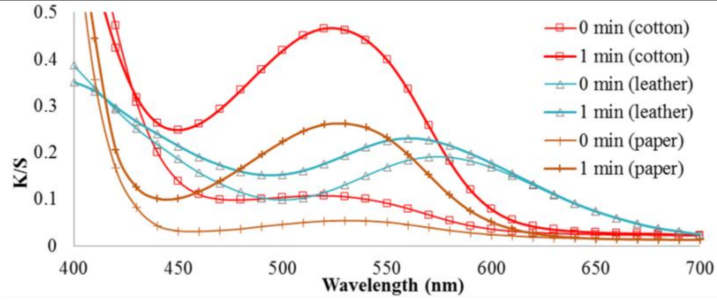

Figure 1. The nature of photochromic colour build up on cotton, paper, leather substrates digitally dyed with a diarylethene based inkjet ink after exposure to UV light for 1 minute.

Figure 1 shows a typical example of photochromic colour build up (where the reflectance is converted to K/S values and plotted against the visible wavelength from 400 nm to 700 nm) on paper, cotton and leather substrates digitally dyed with a photochromic diarylethene based inkjet ink and then exposed to UV irradiation for one minute (where zero minute means without exposure to UV light or before exposure to UV light and 1 minute means after exposure to UV light for one minute). It clearly shows that there is an intense red colour (with a purple tone) build up on digitally dyed cotton after exposure to UV light from a nearly colourless state of the dyed cotton before exposure to UV light. Nearly similar type of behaviour is also observed from digitally dyed paper and leather (white fullchrome crust leather from goatskin, where the wavelength of maximum absorption shows a shift due to the tiny bluish background colour of this specific leather) (Figure 1) and also in other protein substrates (such as, silk and wool) [1, 2, 13].

Detail explanation on the behaviour of spirooxazines, spiropyrans, naphthopyrans and diarylethenes based molecular switches in solid polymer matrices is available from elsewhere (chapter on 'Inkjet printed photo-responsive textiles for conventional and high-tech applications' by the author of this current paper) [1].

7. THERMOCHROMIC MOLECULAR SWITCHES AND THEIR APPLICATIONS

A thermochromic molecular switch (such as, a thermochromic material) change colour as a function of temperature either reversibly or irreversibly. Thermochromic molecular switches which exhibit reversible thermochromic colour change have many potential application in paper, textile and leather substrates. For example, leuco dyes (in appropriate composition with other additives) and cholesteric liquid crystal based thermochromic pigments can be used for a variety of purposes including the display of surrounding temperature when used in appropriate formats to these substrates. A general observation is that a leuco dye based thermochromic switch exhibits a colour change from coloured to colourless or to another colour with an increase in temperature. However, a cholesteric liquid crystal based thermochromic switch shows a vivid range of colour change (may also be termed as a colour play) by passing through the whole spectrum with an increase in temperature. The leuco dye based active thermochromic switch generally composed of three components – (i) a colour former, (ii) a developer and (iii) a solvent in a specific combination where the colour former is a pH sensitive dye (e.g. a spirolactone or a fluoran) and the developer is a proton donor (e.g. an week acid like Bisphenol A). The solvent suitable for this thermochromic composition is usually a low melting hydrophobic, long aliphatic chain fatty acid, amide or alcohol and its melting point is used to associate the system with a specific temperature at which the colour former and developer can easily interact. On the contrary, the liquid crystals with a chiral centre (also known as cholesteric liquid crystals) show colour change in response to a suitable change in temperature in the surrounding environment. In the preliminary stage most liquid crystals were cholesterol derivatives, however, currently, synthetic chiral molecules (also known as chiral nematic liquid crystals) are also available [2, 47-52]. The cholesteric liquid crystals are usually used against a black background for the manipulation of the incident light and also for the reflection of light of selected wavelengths which vary with change in temperature. In this case, the particular reflected wavelengths are dependent on the pitch length of the helix formed by the liquid crystals and this parameter show a change with a change in surrounding temperature. Microencapsulation is a widely used technique for the integration and application of the leuco dye based and liquid crystal based thermochromic pigments. Commercial thermochromic pigments are available since the late 1960s for a variety of applications, including, thermographic recording materials, temperature indicators (e.g. to measure body temperature), recording thermal history (such as, to determine the temperature or history of the food storage when used in a food container). These materials also have other wide ranging applications, some of which are – (a) in medical thermography for diagnosis purposes, (b) in thermal mapping of engineering materials to diagnose faults in product design and in mechanical performance, (c) in the cosmetic industry for moisturizing and as a carrier for vitamins, (d) in memory devices for date storage, (e) in batteries for life indication, (f) in the architecture field for decoration or for its functionality, (g) authentication, (h) brand protection, and also in (i) anti-counterfeit applications. Thermochromic materials are also used as novelty materials in the manufacture of toys, ornaments, kettles, umbrellas, toilet seats, etc. However, in textiles, thermochromic materials are usually applied by printing, coating and extrusion methods. Thermochromic textiles produced from using thermochromic materials have many applications, including in T-shirts, children clothing, jeans, electronic heat profiling circuitry, in man-made cellulose fibres and also in acrylic fibres. Similarly, thermochromic papers can be used for a variety of applications some of which include – (a) temperature indicator, (b) display devices, (c) anti-counterfeit applications, (d) packaging, (e) security, (f) applications in food and biomedical industries. Thermochromic leather have many potential promising applications in various areas where some important uses include – (a) integrated thermal warning systems in leather based garments, gloves and expressive shoes, (b) exotic and novelty leather based items, (c) leather products or similar other items based on leather or leather and textile or other polymer composite (which can be functionalized using appropriate technique(s) to provide more functionality such as displaying temperature responsive colour changes for a variety of applications) [2, 51-54].

8. IONOCHROMIC MOLECULAR SWITCHES AND THEIR APPLICATIONS

Ionochromic molecular switches show ionochromism – a color change phenomenon which is mainly due to the interaction of compounds or materials with ionic species. In this case, some of the main ionic species are the solvated hydrogen ions, metal ions, and onium cations (for examples, tertiary ammonium and phosphonium ions). There are different types of ionochromic materials, some of which include – (a) phthalides, (b) triarylmethanes and (c) fluorans. Most of the commercially available pH sensitive dyes belong to these three classes. Additionally, the organic structures which show the colour change when exposed to ionic interactions are also termed as ionochromes or ionophores [2]. These type of switches exhibit reversible colour change from a colourless state to coloured state or from a coloured state to another coloured state. Besides this, ionochromic can be applied to different flexible substrates (for examples, paper, textile, leather substrates) using different techniques including digital dyeing using inkjet technology. For example, paper, textile and leather substrates were digitally dyed with a range of ionochromic molecular switches for a variety of purposes [2, 9-14, 54]. Figure 2 show the ionochromic behaviour of paper, textile and leather substrates digitally dyed with synthetic anthocyanin (such as, thymol blue) based inkjet ink after exposure to ammonia gas. These type of materials can be used to develop warning systems against toxic gases or other similar type of applications (for details please see, ref. 2).

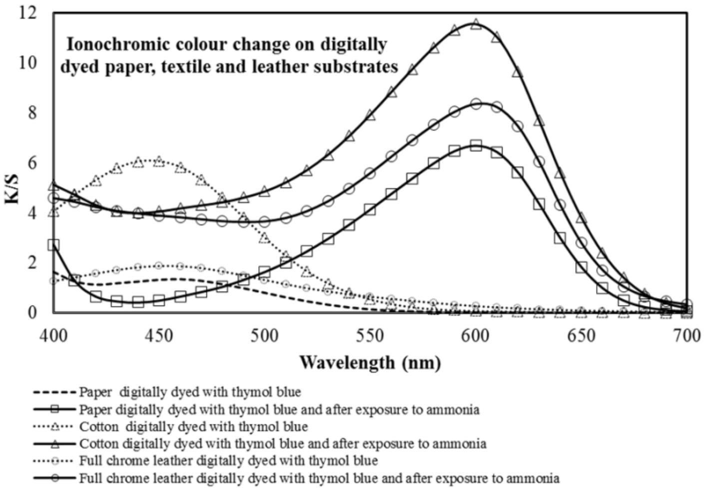

Figure 2. The ionochromic behaviour of paper, textile and leather substrates digitally dyed with synthetic anthocyanin (such as, thymol blue) based inkjet ink after exposure to ammonia gas.

9. ELECTROCHROMIC MOLECULAR SWITCHES AND THEIR APPLICATIONS

Various types of electrochromic molecular switches are practically in use for producing electrochromic displays on flexible substrates (such as, plastic, paper, certain type of textiles) which have many application potentials where some of the most important uses include – (a) wearable display technology, (b) adaptive camouflage, (c) applications in biomimetic systems [2]. Different ways can be used to incorporate electrochromic switches into the structures of flexible materials. One of the method is doping technique – where electrochromic molecular switches can be doped in different fibres production (such as, extrusion, electrospinning, melt spinning, etc). Electrochromic switches show dynamic colour change when exposed to suitable electrical potentials. Some intrinsically conductive polymers (ICPs) and materials (such as, metal oxides, viologens, and particular type of phthalocyanines) are suitable to be doped into fibre structures which are suitable for electrochromic display. Additionally, intrinsically conductive polymers (such as, polythiophene, polyannilne, polypyrrole, etc) are easy to be processed using solution media to print them using screen printing or inkjet printing to produce electrochromic images or circuits on printed substrates. Additionally, there are many other techniques which can be used to incorporate these molecular switches to flexible substrates, some of which are – (a) coating, (b) dipping, (c) dyeing, (d) weaving or knitting with conductive electrochromic threads, (e) graft copolymerisation of conductive polymer based flexible substrates with electrochromic molecular switches. [2] Electrochromic molecular switches are very attractive for their uses in a wide range of areas apart from their main application in display technology, some of which are – (a) chemical and biomedical sensing, (b) remote sensing, (c) security printing and authentication, (d) packaging, (e) glazing control, etc. Certain types of textiles (such as, pre-treated cotton, polyester, nylon-lycra), suitable paper and laminated leather surfaces can also be used for applications when functionalised with electrochromic molecular switches using appropriate techniques [2].

10. PIEZOCHROMIC MOLECULAR SWITCHES AND THEIR APPLICATIONS

Piezochromic molecular switches exhibit a change in shape (and in colour) when exposed to a mechanical stress (or pressure). These materials are of intense current research interest due to their potential application in different areas including in stress detection, especially *in situ* failure monitoring with respect to fracture, corrosion, fatigue, or creep. With the variation on the application of stress piezochromic materials show a change in colour or opacity. There are various types of stress, pressure (compression) and tension where these materials can respond to compressive, tensile, or more complex forms of stress. Piezochromic materials have relative little attention which partly because of challenges associated with synthesis and retention of functional properties as well as other technical performances. Some particular types of polymers and inorganic materials show piezochromic behaviour. Some practical examples include – substituted cyanooligo(p-phenylenevinylenes), thermoplastic polyurethane blends. Piezochromic materials have many application potentials, however, currently they are mostly used in different industries (such as, aerospace, paints, toothbrush, membrane, sports items, sensors, inkjet printhead) for stress sensing and failure detection [47-51, 56].

11. CONCLUSIONS

Molecular switches offer a new horizon for the functionalization of flexible natural substrates using appropriate techniques which have many promising conventional and high-tech application potentials which have been mentioned in different sections of this current paper. However, there are quite a lot of challenges in order to produce high quality and technically performing flexible responsive surfaces using these molecular switches. Rigorous extensive studies are vitally important to address many of these current issues. Future publications of this author will focus on some of these issues.

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