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Review

A narrative review on vital criteria of fungal dyes as dye-sensitized solar cell (DSSC)

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Abstract
The main two reasons for the shift to renewable energy are the depletion and the harmful effects of non-renewable energy, such as burning coal and fossil fuels. As a result, the use of solar cells as renewable energy resources, that prioritise high efficiency while lowering production costs, can provide a promising future for sustaining energy resources. Solar cells have been developed starting with the monocrystalline and polycrystalline solar cells (first generation), non-crystalline and thin film solar cells (second generation) and organic solar cells (third generation). The dye-sensitized solar cell (DSSC) is a new type of solar cell that has gained popularity due to its ability to convert energy at a low cost, ease of fabrication, and non-toxic nature. This paper explores the use of fungal colours as natural dyes for DSSC due to their wide range of colours, large yields, sustainable supply and non-toxic. Natural fungal colours can be extracted using proper extraction conditions and methods. Although fungal dyes have several limitations like instability, non-resistance to harsh conditions, colour loss during processing or storage and limited profitable yield, they still have promising future prospects to overcome the poor interaction of natural sensitizer with semiconductor together with other advantages of fungal colour. DSSC with dye from crude extracts from *Cortinarius* fungi exhibited the highest efficiency of 0.64% among reported DSSC with fungal dye.

Keywords Dye-sensitized solar cell · Fungal colour · Dye · Solar photovoltaic · Mushroom · Natural dye

1 Introduction

Power in the form of electricity is considered an essential resource for the advancement of mankind civilisations along with the amount of electricity used in a society may indicate its standard of living. From the beginning of the industrial revolution, in the eighteenth century to the present time, technological innovation has been the driving force behind societal progress, as well as the cause for the significant increase in energy consumption, which is expected to double by 2100 [1]. As the global primary energy source, coal and natural gas account for more than three-quarters of global energy usage. The natural gas's current mass production is 190,600 m^3 per day, with crude oil accounting for 73,500 m^3 [2]. These sources, however, will eventually run dry. Figure 1 shows the CO_2 emissions data by fuels and industries (coal, oil, gas, cement, flaring and other industries) worldwide from 1900 to 2020. The emissions can be seen increasing

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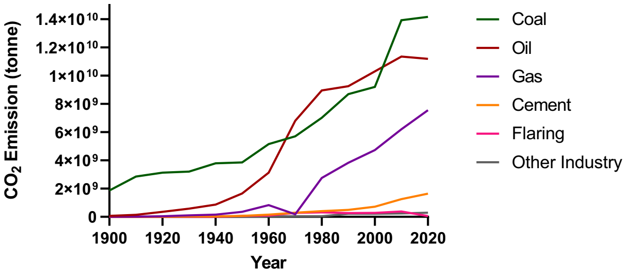
drastically throughout the years [3]. The growth of renewable energy sources may result in some advantages like the advancement of clean, green technologies for a healthy environment. Hydro, geothermal, wind, biomass and solar are the primary renewable energy resources, each having advantages and disadvantages [4]. Meanwhile some of the possible disadvantages include high implementation costs, lack of information, unknown alternative chemical or raw material inputs, unknown alternative process technology, uncertainty regarding performance impacts, and dearth of human resources and skills [5].

The sun provides energy to all living things on Earth through the process of photosynthesis, making it the most fundamental renewable energy source currently accessible. The ease with which solar energy can be used for residential and commercial purposes is a significant advantage [6]. As a result, scientists and engineers are working to create solar-powered systems that can generate electricity directly from the sun [4].

Man-made solar cells use the photovoltaic (PV) effect to convert energy from the sun into electrical power. This approach originated in 1839, but the first developed solar cell was produced in a systematic manner in 1954. The first generation of solar cells consisted of silicon wafers made from single and multicrystalline silicon crystals [7]. The concept of the first generation solar cell is built on monocrystalline and polycrystalline silicon [8]. The term crystalline silicon-based solar PV cells evolved from the method of their manufacturing [9]. This type of cell accounts for the vast majority of cells with increased commercial value in recent years. Monocrystalline solar cells have a high efficiency (27.6%) [10], however, they require less space than other types. They perform better in low-light conditions, making them ideal for cloudy environments [1]. Polycrystalline cells are less expensive than monocrystalline cells but are slightly less efficient, despite having a 23.3% average efficiency [10]. Silicon is the most widely utilised semiconductor material, representing 90% of the present PV market. Despite being relatively expensive, these are the older and most commonly used cells due to their high efficiency [11]. High temperatures, lack of reaction in moderate and overcast weather, the device's fragility, and the heavy weight of the modules [12], reduce efficiency in monocrystalline cells but have a greater negative impact on polycrystalline cells. The aforementioned reduces the polycrystalline cells costs in regions with higher temperatures, which attracts people to those areas [1]. These first-generation solar cells are rather costly compared to other solar cells, making them out of reach for the majority of consumers. However, they have a minor disadvantage when considering other varieties of solar cells. A substantial quantity of industrial waste is produced once silicon gets cut throughout the production process, and the raw material, silicon wafers, is costly and price fluctuations are common due to inadequate supply. Despite the fact that silicon is the second most prevalent element in the planet, it is a necessity for the element to be in low impurities and the purification itself accounts for more than half of the manufacturing cost [13].

Although silicon solar cells have been the dominant contributor in the photovoltaic technology industry for many years, the next generation is focusing more on p-type absorbers and thin film solar cell using chalcogenide semiconductors. Willoughby Smith established selenium's photoconductivity in 1873, while Charles Fritts developed the pioneering solar cells in 1883 employing selenium wafers. Subsequently, numerous chalcogenide semiconductors with chalcogen as the anion, which include S, Se, and Te were investigated as solar absorbers for thin film solar cells like CdTe, CIGS(Se), CIS(Se), and CZTS (Se) [14]. GaAs, CIGS and CdTe based solar cells for example, have currently achieved efficiencies of 25.1%, 23.6%, and 22.6% respectively [10]. Because they are relatively simple to manufacture, they could be more affordable than crystalline solar cells. Conventional solar cells occasionally have higher efficiency in comparison to thin film solar cells, however they have several drawbacks. Because they take up extra space, they are inefficient for domestic use [1]. However, Cd toxicity, element availability concerns, and phase complexity remain the main disadvantages. CuS, $Cu_2Sn(S,Se)_3$, SnS, FeS_2, $Sb_2(S,Se)_3$ and other developing binary and ternary absorbers originating from chalcogen are also being studied.

Fig. 1 CO_2 emission by fuel or industry throughout the world



2 Development of the emerging photovoltaic technology (third generation solar cell)

In terms of global PV penetration, with approximately 767 GW installed globally, PV could generate nearly 1,000 TWh of electricity per year. This represents roughly 4% of total global electricity demand covered by PV [15]. In order to be implemented in building integrated PVs like architectural glass and flexible steel, the forthcoming generation of solar cells needs to exhibit performance close to optimal efficiency within a broad spectrum of light conditions and temperatures, be flexible, durable, have good long-term stability and be appealing. This will allow them to be competitive with crystalline silicon in the current market. In providing electricity to a prospective market in the developing countries, the cost of installation must be reduced [13].

Third generation solar cells are emerging commercial technologies that are less well-known. Solar cells are made of organic materials, which is one of the essential third generation PV technologies. Quantum dot cells, perovskite solar cells, multiple junction cells and dye-sensitized solar cells (DSSCs) are some examples [16]. The following are the key concepts of organic solar cells: Organic semiconductors were discovered in 1977 by Alan G. M. Heeger and H. Shirakawa, which resulted in the discovery of the basic principle behind organic solar cells [1]. Currently, perovskite solar cells, DSSCs, and organic solar cells have the efficiency of 26.1%, 13.0% and 19.2% respectively [10].

DSSCs, which belong to the third generation of PV, offer a unique approach to mimic the key processes of natural photosynthesis. By combining a molecular system with nanoparticles, DSSCs excel in terms of cost-effectiveness, light manipulation, and long-term stability, distinguishing them from traditional inorganic p–n junction solar cells. Moreover, unlike traditional solar cells, DSSCs exhibit consistent high power conversion efficiency (PCE) under various lighting situations. Additionally, DSSCs have the unique advantage of being customisable to be flexible and colourless, or to have a vibrant appearance. These characteristics make them attractive for incorporating into the design of both residential and commercial buildings (referred to as building-integrated photovoltaics: BIPV) as well as portable devices. They have significant potential in the field of IoT, which includes applications in smart homes, healthcare, transportation, and other areas [17].

Figure 2 shows the solar cell development from first, second and third generation.

3 Structure and working principle of DSSC

DSSC uses two conducting glass electrodes made of porous nanocrystalline large band gap metal oxide film based on semiconductors covered in dye-adsorbed nanoparticles including titanium dioxide and zinc oxide. The dye is regenerated by the redox couple-containing electrolyte solution, whereas platinum or carbon incorporates the counter electrode [18]. Because of their large surface area for dye anchoring, nanostructured metal oxide coatings

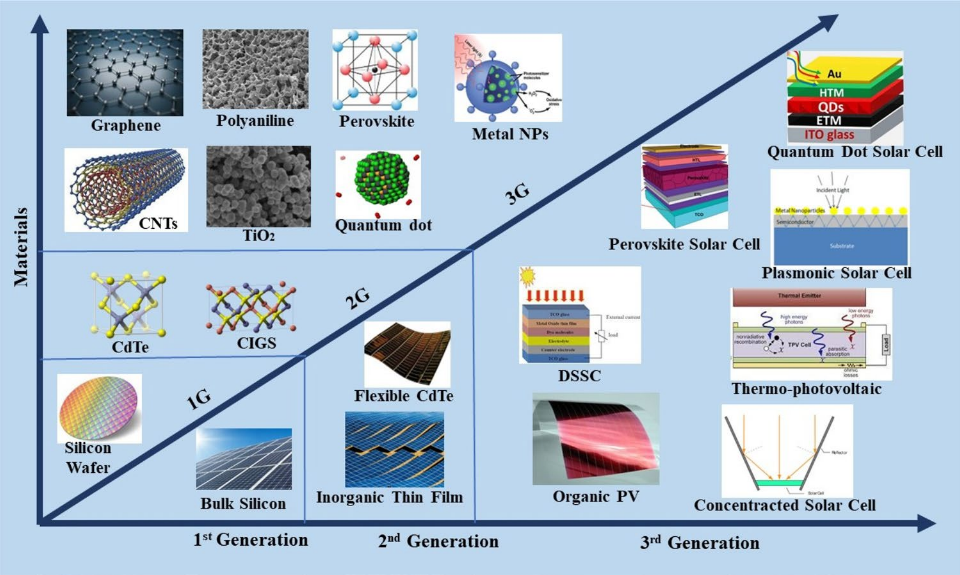


Fig. 2 Solar cell development from first, second and third generation

are notably appealing for DSSC application. The photoelectrode (PE) core of semiconductor component needs to be chemically stable and is not influenced by the types of electrolyte [19].

Figure 3 shows the basic electron transfer process in a DSSC. A DSSC's fundamental electron transport mechanism is depicted in Fig. 3. During the DSSC operation, the sensitising dye molecules are introduced into the n-type TiO_2 photo-anode's conduction band. From there, the dye molecules absorbed solar photon waves to reach the photo-excited state of the nanocrystalline TiO_2. Electrons travel from the conduction band to the valence band when stimulated by light. The iodide or triiodide redox pair, which is present in the electrolyte, passes through by the valence band electrons. In order to complete the external circuit, the tri-iodide/iodide redox reaction mechanism regenerates the dye molecules. Electrons from the external circuit are collected by the counter electrode. To raise the DSSC's efficiency, the dye molecules are altered and deposited on the TiO_2 photoanode. As nanoparticles, various types of nanostructured TiO_2 photo-anodes are utilised for DSSC application [20].

TiO_2, ZnO and SnO_2 are examples of metal oxides with wide bandgaps that may absorb a wide range of wavelengths. Despite extensive research into alternative semiconductors, TiO_2 has proven to be the most efficient in PV devices [21]. Among semiconductor metal oxides, the usage of TiO_2 in photoanodes is well-established. After the first DSSCs were created in 1991, the utilisation of large surface area nanoparticle TiO_2 films sensitised with a dye that injects electrons after photoexcitation increased rapidly, according to a study on the material. TiO_2 is a semiconductor that has favourable characteristics for electron transport, including a wide band gap, high electronic mobility, and good physical properties. With a direct band gap of 3.69 eV, TiO_2 also shows low recombination loss and high exciton binding energy. [19].

Mrinalini et al. [22] indicated that dye solar modules (DSMs) are utilised in the field of building and car integrated photovoltaics. Solaronix Ltd. reported an annual production of DSMs at approximately 20 MWp/year and a normalised manufacturing cost of around €0.97/Wp for an efficiency of approximately 7% [22]. The benefits of DSM were showcased as flexible and transparent, and specifically intended to operate in areas with low light conditions, resulting in increased operation hours. The manufacturing cost for DSMs ranges from \$0.50 to \$1.00/Wp, while the efficiency ranges from 5 to 15% [22]. This makes DSMs comparatively more affordable than first and second generations solar cells. Currently, it is anticipated that the global market for commercialising DSSCs will experience gradual growth, reaching a value of \$290 million by 2023 [22]. The utilisation of DSSCs in many applications, such as chargers, solar bags, calculators, and wireless keyboards, indicates that the cost of DSSCs increased by 2020 due to the excessive output of electricity. Hence, it is imperative to decrease the amount of materials used in the production of each DSSC module in order to avoid strong competition with first and second generation solar cells in the market [22].

4 Photosensitizer properties of DSSC

DSSCs are novel PV cells that imitate photosynthesis in plants. In contrast to conventional solar cells, DSSCs have enhanced performance in low-light conditions and exhibit reduced susceptibility to heat-induced loss of energy. The sensitizer used primarily determines the efficiency of DSSC. As a sensitizer, the dye is crucial in absorbing

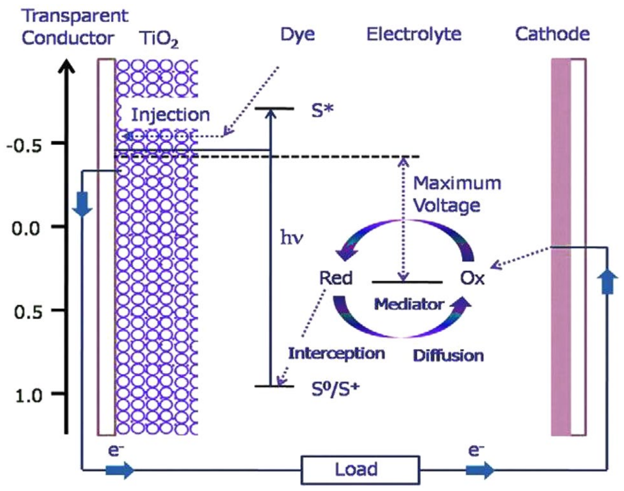


Fig. 3 The basic electron transfer process in a DSSC

sunlight and turning solar energy into electricity [23]. DSSCs use dye molecules to absorb photons and generate electrons to convert sunlight to electricity [24]. It should be offered in nanostructure form to maximise the surface area that is suitable for adsorbing dyes, which thus improving the sun's energy harvesting system efficiency and lattice structure. It must be ideal for bonding of dye and have a conduction band just below the dye's LUMO level for effective electron injection [19].

The dye used as a photosensitizer is a vital component of the DSSC process. The absorption spectrum of the dye and its anchoring level to the semiconductor surface have a significant influence on the efficiency of the cell. The chemical structure of dyes is critical for increasing photon absorption and as a result, improves the electron injection and conversion efficiency [19]. Sensitizers, which are the dye molecules is one of the key elements of the DSSC, encouraging several studies to improve dye efficiency and produce novel dye sensitizer groups. In order to effectively inject electrons from the dyes into the semiconductor's conduction band, an efficient sensitizer for DSSC must: (i) bind thoroughly to the semiconductor with an anchoring group, such as a carboxylic or hydroxyl group; (ii) have a deep absorbance in the visible and near-infrared regions of the solar spectrum, resulting in a high molar absorbance coefficient; and (iii) transfer of electrons from the dye (iv) The lowest unoccupied molecular orbital (LUMO) energy must be greater than the conduction band level and the highest occupied molecular orbital (HOMO) energy level must be lower than the redox pair's in order to permit charge injection into the semiconductor. As a result, oxidizing dye regeneration is possible [25]. Natural dyes, metal complex dyes, and metal-free organic dyes are common types of sensitizers [26].

5 General classes of sensitizers

5.1 Metal complex dye

Due to their high efficiencies achieved in DSSC applications, metal complex dyes are commonly used as sensitizer. Ruthenium bipyridin complex sensitizers (N719, N3, Black dye) are examples of metal complex dye which have the highest efficiency values in DSSCs [26]. N3/N719 Ru-polypyridyl-complexes obtained by the Grätzel group have excellent photoelectrochemical properties, acting as charge-transfer sensitizers by capturing approximately 11% of the solar to electrical energy in the sun's standard global air mass AM 1.5. Ru-based dyes have the ability to introduce charges into semiconductors through the phenomenon of metal–ligand transfer. Nonetheless, the Ru complexes contain heavy metal that is environmentally toxic, and Ru complexes degrade in the presence of water. [25]. Due to their outstanding stability, recent advancements in ruthenium-based dye compounds have focused on molecular engineering. N719, known as red dye with the chemical name of cis-di-(thiocyanato) bis (2.2′-bipyridyl)-4,4′-dicarboxylate) ruthenium (II). It is the ideal light absorber and charge transfer sensitizer compared to other commercial dyes. Over the years, black dye, the newly established, outperforms the performance of the red dye [27].

Ru (II) metal is commonly selected for several reasons: (1) its octahedral geometrical structure allows for the controlled extension of specific ligands; (2) Ru (II) complexes' photophysical, photochemical, and electrochemical properties can be modified; (3) it has stable and accessible oxidation states from I to IV; and (4) it has good solubility in a wide range of solvents. A novel dye named N179 (di-tetrabutyl ammonium cis-bis (isothiocyanato) bis (2,2′-bipyridyl-4,4′- dicarboxylato) ruthenium (II) was published by Nazeeruddin et al. in 2005. It was comparable to N3 with an 11.2% conversion efficiency. The transition metal to ligand charge transfer as well as the HOMO and LUMO molecular orbitals are responsible for the dye's ability to absorb light. The N749 dye, often known as black dye, exhibits radiation absorption up to 860 nm and comparable performance to N3 and N719 dyes. On the contrary, N749 has a lower absorption coefficient than N3 and N719 dyes. In order to adsorb more dye molecules, thicker metal semiconductor oxide electrodes are generally required for lower absorption coefficients. From the perspective of electron transport, there are drawbacks to thickening the metal semiconductor oxide layer. In particular, this may result in lower open circuit voltage and short-circuit current density [28].

5.2 Metal-free organic dye

The demand for metal-free organic dyes has grown recently, and since 2011, their use in DSSC has expanded at a rate that is comparable to that of sensitizers based on ruthenium. The very high molar extinction coefficients, free form of costly and toxic Ru metal, ease of synthesis, easily tunable absorption energies, low cost, stability at elevated temperatures, and/or extended illumination are the benefits of selecting metal-free organic dyes over Ru-based complexes [28]. Utilising well-established synthetic strategies in dye chemistry, metal-free organic dyes have a more simple chemical structure adaptation. This allows for the production of sensitizers with broad spectral absorption, as the chemical properties can be modulated to produce a great deal of variability in the optical properties [29].

The molecular structure of metal-free organic dyes can be broadly classified into three categories: donors, linkers, and acceptors. D-π-A sensitizers, also known as linkers, are typically π-conjugated systems that create bridges between electron-donating (D) and electron-accepting (A) groups. In order to create effective sensitizers, donor groups comprising electron-rich moieties such as phenylamine, aminocoumarin, indoline, (difluorenyl) triphenylamine, triarylamines, and carbazoles should be chosen for metal-free organic dyes, also referred to as D-π-A sensitizers. Benzothiadiazole, polyenes, and thiophenes are good choices for π-conjugated groups [28]. The same group went on to create additional TPA dyes with fluorine-functionalised phenyl groups connected to EDOT groups in a later study. The dye demonstrated good performance as a DSSC photosensitizer, achieving an efficiency of 8.22% [29]. It was discovered that the triphenylamine ring's band gap narrows and the HOMO and LUMO energy levels are tuned when strong electron injection groups are added. This is explained by the fact that the conduction band of TiO_2 prevents charge recombination of electrons to electrolyte [30].

5.3 Natural dyes in DSSC

Concerning the use of biomaterial-derived pigments in DSSC applications, there have been some intriguing natural dye explorations. Several reports have emphasised the importance of researching natural dyes include cyanine, tannins, anthocyanins, cyanidins, carotenoids, betalains, chalcones, as well as chlorophyll and its derivatives for use in DSSCs as a cost-effective, non-time-consuming, lower-energy-required, and environmentally friendly substitute [23].

Dye that is organic has been discovered to replace carcinogenic dyes in an attempt to deliver low-cost, environmentally friendly electronics with considerations of health and environmental impacts. In order to create environmentally friendly DSSCs, a lot of research has been done on the use of natural dyes as a photosensitizer for DSSCs. For instance, it was found that DSSCs employing pomegranate as a dye sensitizer achieved a power-conversion efficiency of 2%. Natural dyes are therefore excellent alternatives. Additionally, using one layer of TiO_2 nanoparticles yields the highest level of efficacy among the natural pigments extracted from Inthanin bok leaves, with an efficiency of 1.138% [19].

Recent years have seen a significant increase in interest in the use of natural dye as a sensitizer in DSSC because of its affordability and ease of manufacturing. Natural dyes can be extracted from various plant parts, including seeds, leaves, fruits, stems, roots, and flowers [21]. Natural pigments consist of chemical compounds that are essential for all living organisms' development. Plant pigments are divided into four primary categories based on their similar structure and biosynthetic basis: chlorophylls, carotenes, flavonoids, and anthocyanins. While synthetic dyes are more stable and efficient than natural dyes, they are more expensive to use, rarer, and often include potentially dangerous substances. On the other hand, natural dyes have several advantages over synthetic dyes, including affordability, availability, biocompatibility and environmental friendliness. *Mitragyna speciosa* is an example of a plant with distinguishing characteristics, such as alkaloids-rich pigments and a dark green colour caused by chlorophyll pigments presence. According to previous studies, the chlorophyll content, the main pigment utilised for photosynthesis, correlates with the shade of green the leaves with three additional pigments serving auxiliary functions [31].

A number of factors influence the efficiency of natural DSSCs, including the choice of the suitable natural dye, quantitatively significant stable electronic anchoring of the dye to the solvent system, photoanode, counter electrodes, and metal oxide nanostructure surfaces. Standardising these crucial elements thus becomes very important when considering to the commercialization of natural DSSCs. Natural dyes are non-toxic to the environment, have high absorption coefficients, excellent light-harvesting efficiency, no resource limits, low cost, and easy manufacturing process. The efficiency produced with these natural dyes are still below what is currently required for large-scale practical applications, but this has not stopped researchers from all over the world from searching for new sources of natural dye. These intriguing investigations on the fabrication of DSSCs utilising dyes derived from bioresources remain ongoing [23].

Humans are now using a greater amount of synthetic colourants in modern society for a variety of useful applications. On the other hand, the overuse of synthetic dye that contain benzidine and carcinogenic azo compounds which are not biodegradable, can have adverse effects on human health as well as the environment. Although natural dyes do not have these side effects, their limited yield and poor capacity for binding draw attention. As a result, further investigation and improvement of natural resource production is highly desirable. There are plenty of plants, animals and minerals around the world that can produce dyes and large quantities can be obtained by cultivating or processing them [32].

However, a basic extraction procedure is used to extract natural dyes from fruits, leaves, flowers, seeds, roots, and peels [33]. Its non-toxicity, full biodegradability, colour variation, low cost, and lack of pollution are its benefits [34]. In comparison to synthetic dyes, the efficiency is lower, however, the majority of the researchers' focus is on the use of natural dyes in the fabrication of DSSC [35].

Table 1 summarises the comparison between complex dye metal-free organic dye and natural dye use in DSSCs.

6 *Fungi* as a source pigment for dye application

Other than natural plants, it is possible to investigate the use of insects and microorganisms in the development of colourants. Additionally, to enhance the colourant performance, the use of natural mordants can also be studied[32]. Due to rising demand, resource constraints, and a number of side effects associated with presently authorised natural pigments—such as seasonal availability limitations, instabilities with respect to light, adverse pH, heat, low water solubility and variability in pigment extraction—more studies into reliable sources of natural colourants are needed. The capacity of fungi to synthesise natural colours with noteworthy light and chemical stability has presently drawn considerable attention, a wide range of colours, large yields, and a sustainable supply. Since the nineteenth century, mycologists have been intrigued by the production of colours by fungal colonies, which can be viewed as a microbial reservoir for the synthesis of food-grade colours. Various colours, such as melanins, anthraquinones, hydroxyanthraquinones, azaphilones, carotenoids, oxopolyene, quinones, and naphthoquinone, are known to be produced by fungi [36]. Figure 4 shows the chemical structure of melanin.

Although not performing photosynthesis, fungi are acknowledged as producers for variety of pigments and secondary metabolites which may increase sensitivity in DSSCs. Secondary metabolites, light-harvesting pigments, and reaction centres (RC) derived from fungi, in addition to light-harvesting complexes, can aid in enhancing the DSSC's small optical absorption window, which has been found to be a major drawback in comparison to first-generation solar cells. The use of a simple deposit approach demonstrated the potential of pigments derived from all of these organisms for DSSC, with efficiencies reaching 4.6% [37].

The oyster mushroom (*Pleurotus sp*.), the world's third-largest cultivated mushroom, is one of the most common types of mushrooms due to its high nutritional and therapeutic value [38]. Secondary metabolites found in oyster mushrooms include polyketides, steroids, diboviquinone, terpenes, phenolic compounds and variegatic acid. Oyster mushrooms have a biological efficiency of about 100% and can be kept for 24 h under normal air conditions. It has low production costs despite having a slightly longer crop cycle (45 days) [39].

Most of the mushrooms are coloured, and various pigments have been discovered in various types of mushrooms, such as carotenoid in Cordyceps militaris and the red pigment lilacinone in *Lactarius lilacinus*. Previous research has shown that melanin is present in mushrooms such as *Auricularia auricula* [40], *Lentinula edodes, Pleurotus cystidiosus*, *Agaricus bisporus* and *Armillaria cepistipes*. Eumelanin, pheomelanin, and allomelanin are groups of high-weight molecules composed of phenolic and/or indolic monomer-based complex heterogeneous polymers. 5,6-dihydroxyindole (DHI) and 5,6-dihydroxyindole-2-carboxylic acid (DHICA), which are the building blocks of eumelanin, are both 8.49% nitrogen and 0.09% sulphur. Pheomelanin consists of benzothiazole and benzothiazine, which have combined sulphur and nitrogen content of 9.36% and 9.78% respectively [41, 42].

Since pheomelanin is yellow-to-reddish in colour and eumelanin and allomelanin are both black, many creatures have been reported having multiple types of melanin. Different shades of melanin, such as black, yellow, or red, are formed in varying amounts and proportions, influencing how colours appear. All three types of melanin can be found in mushrooms, and they are usually mixed together. Moreover, melanin is probably cross-linked to polysaccharides because it has been observed to accumulate in the cell walls of certain mushrooms. Fungi need polysaccharides to live and proliferate. Pigments and polysaccharides combined create important interactions and routes for the transfer of chemical information [41].

The growing demand for dyes ignites an upsurge of research endeavours aimed at investigating filamentous fungi as potential pigment producers. Fungi-derived natural pigments are the most environmentally benign substitute for synthetic pigments, according to scientific research. This is due to their rapid growth, straightforward processing, and crucial functions in transcription and intercellular communication. Fungal pigments exhibit remarkable resistance and

Table 1 Comparison between metal complex dye, metal-free organic dye and natural dye

Criteria	Metal complex dye	Metal-free Organic dye	Natural dye
Source	Metal complex	Electron donor and an electron acceptor linked covalently through a π-conjugated spacer	Natural resources (plants, microorganisms) Synthetic organic
Classification	Polypyridyl complexes. ruthenium and osmium, metal porphyrin, phthalocyanine and inorganic quantum dots	Phenylamine, aminocoumarin, indoline, (difluorenyl) triphenylamine, triarylamines, and carbazoles	Indigoids, betalains, tannins, carotenoids, flavonoids, quinonoids
Example	Ruthenium bipyridin complex sensitizers; (N719 'red dye,' N3, N749 'Black dye')	Triphenylamine (TPA) and derivatives	Anthocyanin from pomegranate, Betalains from beetroot
Functional group present	Phosphonate, carboxylic acid, or boronic acid groups	Azo group, carbonyl group, hydroxyl group, amine group	Hydroxyl group, carboxylic acid, nitrogen-containing functional group
Synthesis process/Production	Chemical synthesis	Chemical synthesis	Natural pigment or chemical synthesis
Efficiency on DSSC	Up to 13.6%	Up to 8.2% [29]	Up to 2%

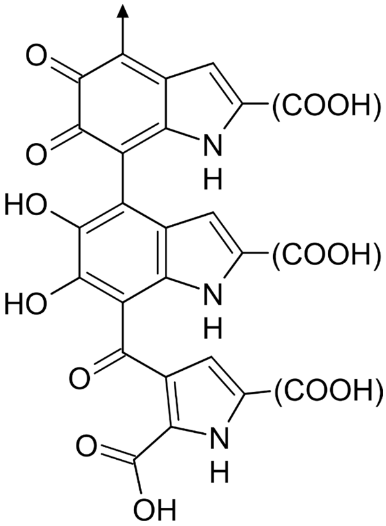


Fig. 4 Chemical structure of melanin

protection against both biotic and abiotic agents, including antagonistic microorganisms and UV radiation. Moreover, they find diverse applications in biology. Potential commercialisation prospects exist for pigmented biomolecules produced by fungi, which find utility in various sectors including food, cosmetics, textile fabrics, and more, owing to their adaptability, structural characteristics, and capacity for extensive cultivation [43].

The phenomenon wherein a dye is absorbed or "exhausted" from a dye bath onto the substrate undergoing dyeing is referred to as "dye exhaustion." During dyeing procedures, specifically textile dyeing, the dye is commonly dissolved or disseminated within a dye bath, which is a liquid medium. Following this, the substrate, which may be fabric or textile, is submerged in the dye bath, which facilitates the transfer of dye molecules from the liquid medium to the substrate. Throughout the process of dyeing, the dye molecules migrate progressively from the dye bath to the substrate until an equilibrium is achieved, at which point no additional dye absorption takes place. This signifies that the substrate has achieved maximum dye absorption within the constraints of the dyeing conditions (temperature, pH, and dye concentration).

To achieve enhanced pigment exhaustion and distribution within the leather, it is necessary to optimise the conditions under which pigments are utilised. The concentrations of all five pigments that were optimised at 6% owl, 1.5 g/L salt, and a 40:1 liquor-to-product ratio were utilised in the research conducted by Velmurugan et al. Seventy percent of the red pigment produced by *M. purpureus* was depleted. In contrast, maximal pigment exhaustion of 40% and 35% was observed at 6% owl of pigment by the fungi *Isaria spp.* (reddish brown pigment), *Emericella spp.* (pink pigment), and *Fusarium spp.* (red pigment), respectively. The findings regarding the impact of pH, temperature, and time duration on pigment exhaustion in the process liquor indicate that pigment exhaustion increases progressively until pH reaches 5.0, after which further pH increases lead to a reduction in pigment exhaustion. The heightened pigment depletion observed at pH 5 may be attributed to various factors. While the acidic pH may induce fibre enlargement in leather samples, the stability and characteristics of pigments remain unaffected at this particular pH. The findings demonstrated that the absorption of colour increases progressively with the passage of time. A maximum of 120 min is necessary for the sample to experience substantial exhaustion in the dye solution. Therefore, a 120-min time interval has been determined to be the optimal duration. The findings pertaining to pigment exhaustion at various temperatures indicate that pigment exhaustion increased in direct proportion to temperature from 30 to 70 °C, but subsequently decreased (80–120 °C) [44].

The process utilises non-toxic natural materials, making it exceptionally well-suited for dyeing fibrous substances like leather, which are crucial for applications that prioritise human health. Previous research has established that *M. purpureus, Emericella spp.*, and *Penicillium spp.* do not exhibit any toxic properties. Furthermore, the pigment generated by these fungi is biodegradable and comprises an extremely minute quantity of phenolic compounds. In contrast to azo dyes, discarded dye liquor therefore does not pose any environmental hazard. Even the spent dye liquor is recyclable and will not produce any undesirable aromas during the subsequent process. Furthermore, the utilisation of fungal pigments in the dyeing process results in substantial enhancements in exhaustion management, serving as an in-plant control mechanism to attain discharge concepts close to zero. Fungal pigment that is acquired can be lyophilised or spray dried to yield a variety of powdered colourants. Additionally, solvent ethanol can be extracted using an appropriate solvent recovery system [44].

The pure extracellular laccase (GLL) obtained from the fermentation broth of the litter degrading fungus *Gymnopus luxurians* exhibited significant enzymatic activity across a wide temperature range of 25–85 °C, with the most favourable temperature range being 55–65 °C. When the laccase activity was measured at 85 °C, about 50% of the original activity was still present. The optimum pH value for GLL was found to be 2.2 when tested against ABTS at a temperature of 37 °C. At pH 3.8, there was a decrease in activity by approximately 50%, while at pH 1.8 or 5.4, there was a significant decrease in activity by 95%. Fungal laccases often exhibit functional activity towards ABTS within a temperature range of 40–70 °C, with an ideal pH range of 2.0–6.0, favouring acidic conditions [45].

Table 2 shows the comparison between fungal dye and synthetic dye.

7 Extraction conditions of fungal dye

Natural colourants are extracted from natural sources using a range of methods. Following the drying and grinding of the plant material, both traditional and modern extraction methods are used to choose an appropriate solvent based on the desired dye [46, 47]. Target isolated compounds are separated for dyeing using spectrum approaches are used to characterise the separated chemicals after they have been isolated using chromatographic or other techniques [32, 48].

Dehydration is one of the most important preservation techniques for extending the shelf life of goods. Using the best drying conditions will improve not only the physical qualities such as shrinkage, dehydration capacity, and colour, but also the antioxidant and health-promoting contents [49]. This is because mushrooms are remarkably temperature sensitive. In terms of colour, size reduction, and rehydration qualities, freeze drying oyster mushrooms is preferred [50]. When vacuum-dried mushrooms were used as the starting material, nevertheless, the extractability of bioactive substances in both simulated digestion fluids and organic solvents was higher, indicating that the physical makeup would also affect the rate at which digestion fluid diffuses into the food matrix [51, 52].

Melanin in different animals is often identified by means of integrative physicochemical tests, which include UV–Vis spectroscopy, nuclear magnetic resonance (NMR) spectroscopy, Fourier transform infrared (FTIR) spectroscopy, and solubility and chemical reaction testing. The colours of oyster mushrooms were first extracted using a variety of solvents, such as water, alkali solution, aqueous acid, and organic reagents (like ethanol and acetone). An acidic solution was discovered to be necessary for extracting and precipitating the colours from oyster mushrooms [41, 53].

8 DSSC performance using fungal dye

The UV–Vis light absorption spectra of oyster mushroom pigments were studied, and it was discovered that the significant UV absorbance and optical density gradually decreased with increasing wavelength, which was consistent with the normal melanin absorption profile. Oyster mushroom pigments have the highest absorbance value at a 235 nm wavelength. The variety of melanin sources may have caused a slight change in the native melanin structure, which could explain the differences in maximum absorbance. The photo-conversion efficiency (PCE) of fabricated DSSCs using natural dyes of *Agaricus bisporus* (button mushroom) as photo-sensitizers was 0.4%. [54].

Samples of crude extracts from *Cortinarius* fungi exhibit two strong adsorption bands with maxima at 555 and 420 nm, in addition to a feeble broad absorption band spanning 650–415 nm. It is widely acknowledged that for dyes to effectively form sensitization bonds with semiconductor surfaces, their molecules must contain anchoring groups such as –COOH, $-PO_3H_2$, $-SO_3H$, –OH, and others. The devices that were presented all demonstrate activity during the

Table 2 Comparison between fungal dye and synthetic dye

Criteria	Fungal dye	Synthetic dye
Stability	Excellent light and chemical stability, a wide range of colours, large yields, and a sustainable supply	High thermal and chemical stability
Range of light absorption	200–750 nm	800–900 nm
Pigment/Colourant	Chlorophyll, anthocyanin, betacyanin, carotene, cyanin, and tannin	Metal complexes
Effect to health	Able to scavenge free radicals and act as antioxidants has positive impacts on health	Carcinogenic
Effect to environment	Non-toxic	Environmentally toxic

conversion of photons to current. The most favourable outcomes were achieved with values of the short circuit current density, J_{sc} of 1.79 mA/cm^2, he open circuit voltage, V_{oc} of 541 mV, the fill factor, FF of 65.9%, and the PCE, η of 0.64%. Possible factors contributing to the suboptimal performance of natural dyes in DSSC employing fungal dye include the following: insufficient energy of the excited state of the dye, rapid recombination within the dye molecule, and/or the presence of compounds in the extracts that facilitate recombination processes within the cells [55].

The red dye producing fungi (Penicillium sp.) while black dye producing fungi (Aspergillus sp.) UV–Vis absorption spectra shows the maximum absorbance of the dye solutions were 523 nm and 416 nm respectively which showed that, both dye solutions can absorb solar light. The I-V measurements for the DSSCs fabricated shown that the V_{oc} and the J_{sc} in Aspergillus sp. dye at 24 h were 463 mV and 7.06×10^{-5} A/cm^{-2} with the FF 0.4427 and η of 0.0144% respectively. At 48 h, the V_{oc} and the J_{sc} were 491 mV and 2.41×10^{-5} A/cm^{-2} with the η of 0.0057% or Penicillium sp. at 24 h, the V_{oc}, the J_{sc}, and the FF were 559 mV, 3.71×10^{-5} A/cm^{-2}, 0.5523 with η of 0.0114%. At 48 h, Penicillium sp. obtained the V_{oc}, the J, the FF_{sc} and the η of 558 mV, 2.937×10^{-5} A/cm^{-2}, 0.5183, and 0.0085% respectively. Although the J_{sc} has been reduced a bit, the V_{oc} has been increased by this dye. This is most likely the result of an undesirable reaction occurring within the cell. The 24-h dipping period appeared to be satisfactory in terms of PCE; however, a significantly extended dipping period for the electrodes drastically decreased the PCE. With the reduction of other factors, even a marginal increase in FF is not gaining dominance. It was observed that the V_{oc} and the J_{sc} values of the two varieties of electrodes coloured red were more stable [56].

In the comparison of the PCE of pigments as sensitizers between algae and microalgae (photosynthetic organisms), bacteria and fungi (non-photosynthetic organisms), Orona-Navar et al. find that the PCE of fungal pigments is intermediate (ranging from 0.26 to 2.3%), in contrast to the significantly higher PCE of photosynthetic pigments (0.001–4.6%) and bacterial pigments (0.004–1.67%). While the obtained values of the V_{oc}, the FF, and the η may not seem particularly high, they do present an opportunity to develop biodegradable and environmentally friendly alternatives for dyes using natural photosensitizers derived from fungi. Although the V_{oc} is relatively low, this value can be increased by connecting the solar cells in series. 2.3% is the greatest reported PCE for dyes derived from fungi. Nevertheless, by refining performance, it is possible to attain greater efficiencies by fully capitalising on the properties of the dye, as has been observed even with the silicon solar cells presently in operation. While the commercialization of these dyes is still a distant goal, natural photosensitizers present a compelling substitute for organic dyes due to their extensive accessibility, economical manufacturing process, module compatibility, and ecological sustainability [57].

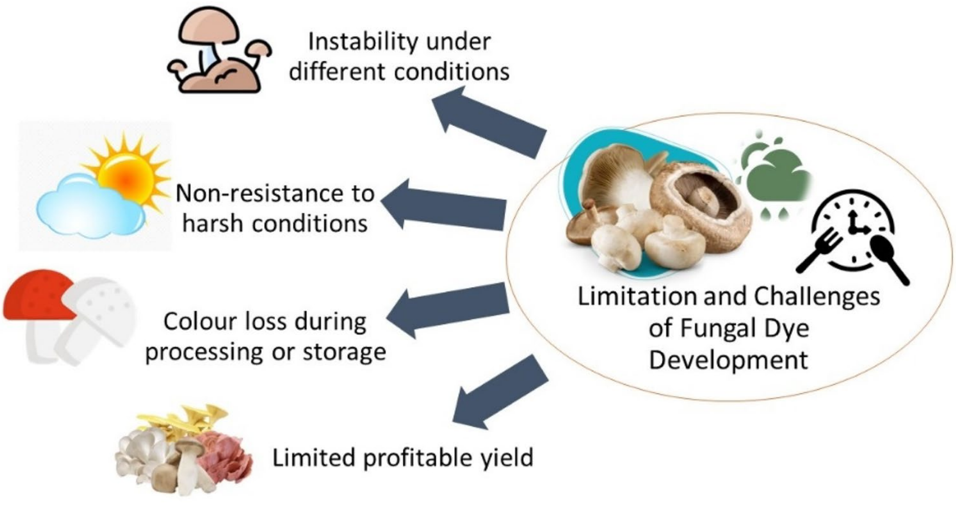
9 Limitation, challenges of fungal dyes and their future prospect in DSSC application

Although there is an abundance of fungal resources that may act as a source of potential pigments, industry faces a number of challenges that impede the commercialisation process of these resources. Figure 5 illustrates the limitations of fungal dye. The continued viability of the fungal-based pigment market being one of the hindrances. The sustainability of fungal pigment is primarily dependent on three important factors: pigment yield, purity and pigment stability [58].

Natural pigment stability in the presence of many environmental elements, including food matrices, pH, light, moisture, and temperature is a significant challenge, with a shorter lifespan as a result of molecular instability under different conditions, which may limit their use as commercial pigments. However, new strategies such as microencapsulation and nanoformulation can be used to address such stability issues. These formulations can aid in the improvement of the physical properties and stability of ingredients used in the food industry, such as milk processing. The technologies of emulsion, coacervation, freeze-drying, and spray-drying utilise shell materials as the basis for pigment microencapsulation. Spray-drying is the most commonly utilised pigment stabilisation method in the food industry, and it significantly improves product shelf life [58].

Carotenoids, for example, are pigments extracted from fungi. Although they are thermally stable, the main sources of instability are acidic degradation and oxygen. Encapsulation techniques, however, limit the degradation caused by oxygen and acidic media exposure. Their restricted application results from their physical stability and low chemical. The lack of process homogeneity is the most significant barrier to using natural colorants. Since these are natural additives, there might be wide variations in the raw ingredients, which makes colour standardisation difficult [59]. There are a lot of potential applications for fungi, but their resistance to harsh conditions has yet to be investigated. When fungi are placed in or exposed to an unfavourable fungal environment, they are more likely to decompose fungal biomass and destroy functional enzymes [58]. Furthermore, colour degradation during handling or preservation may occurr depending on the product. More effective approaches are required to preserve the natural colourants'

Fig. 5 Limitation of fungal dye



chemical and physical durability. These colourants are utilised in a variety of products despite concerns with their chemical and physical stability, however more ways to improve their stability are being discovered especially for the easily accessible naturally coloured dyes in yellow [59], red, and green [60].

The variety of chemical entities and properties limits the fungal pigment's profitable yield [61]. Pigment yield can be increased in a culture by improving biomass growth or intracellular pigment accumulation. Achieving the optimal outcomes requires close monitoring of operational conditions, including temperature, aeration, media composition, agitation and pH [58].

Fungi are gaining popularity as a new, effective and environmentally friendly manufacturing approach based on biological growth to sustain the environment by being highly versatile materials. The scope of impact this technology can accomplish in enhancing the sustainability of the environment globally exemplifies its enormous potential [62]. Fungal usage does have several advantages, but scaling them up to large-scale commercial applications has proven challenging. The change in perspective towards new technology is critical, with important players still failing to fully appreciate the advantages of fungal-based methods, stifling direct application. Interpreting advanced scientific terminology into business expressions is one of the difficulties in making a seamless move from the laboratory to the market. A technological and business-friendly bridging platform is considered essential for introducing fungi as potential biomaterials in the future.

Even though the fact that these fungal pigments are connected to specific biological activities, little is known about the processes controlling their synthesis or physiological function. Understanding the components of pigments, the de-novo routes that lead to its formation, and the genome that drives it are made easier by recent developments in biotechnological and analytical methods that employ computational and molecular methodologies. Other methods for producing these metabolites in large quantities include co-culturing, adjusting culture conditions, and heterologous expression. Obtaining fungi that can produce pigments from a range of environmental ecosystems and habitats needs to be explored as a possible supply of commercial pigments [63].

The main problem with natural sensitizers in DSSCs is that they interact poorly with the semiconductor surface. This can appear as a result of the absence of certain groups or the presence of pigment agglomerates, which prevents strong bonds from forming with the oxide surface. As a result, cells sensitized with natural pigments outperform those sensitized with metal–organic and organic dyes. The functional groups present in the molecular structure are primarily responsible for the stability of natural pigments. These are influenced by environmental factors such as sunlight, oxygen, and air, the acidity of the medium, the extraction solvent and high temperature exposure [64].

Due to the positive health effects of natural colors, society's demand for natural-colored foods has increased. Another noteworthy advancement is the use of molecular biology techniques for pigment production under controlled conditions, but this production is not commercially viable. However, because these pigments are very expensive, numerous tests have conducted to ensure their safety [65].

10 Conclusions

For this kind of developing photovoltaic technology, natural pigments derived from biological sources are useful because they are plentiful and also have a reasonably price, their preparation and extraction processes are readily scalable, and they neither endanger human health nor are regarded as persistent substances in the environment. Fungal pigments have the potential to be a valuable source of natural colorants, but the industry faces challenges in commercializing these resources. The sustainability and development of the industry for fungal-based pigments are hindered by factors such as pigment yield, stability, and purity, as well as the lack of process homogeneity. Stability issues, such as colour loss during processing or storage due to environmental factors, can be addressed through novel approaches such as microencapsulation and nanoformulation. However, more effective approaches are required to keep the natural colourants' chemical and physical longevity. Despite these challenges, fungal pigments used in a wide range of products, and advancements in biotechnological and analytical tools are aiding in deciphering the pigment components and genome accountable of producing it.

One of the challenges in using natural pigments in DSSCs is their inadequate interaction with surface of the semiconductor due to the absence of specific groups in the molecular structure. However, natural pigments have functional groups that are primarily responsible for their stability and positive health effects. Advances in molecular biology techniques for pigment production under controlled conditions have been made, but commercial viability remains a challenge. Despite this, society's demand for natural-coloured foods has increased, and numerous tests have been conducted to ensure the safety of these pigments. The potential of fungi as a sustainable, novel, and smart manufacturing platform with enormous potential for improving global environmental sustainability is gaining popularity, but bridging the language gap between technical science terms and business expressions is seen as critical in developing fungi as biomaterials of the future.

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Declarations

Competing interests The authors declare no competing interests.

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