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Photonic Nanostructures Inspired by Plants

Sherif S. Z. Hindi[1], Uthman M. Dawoud[2]
[1]Department of Arid Land Agriculture, Faculty of Meteorology, Environment and Arid Land Agriculture, King Abdullaziz University (KAU), Jeddah, Saudi Arabia
[2]Department of Chemical and Materials Engineering, Faculty of Engineering, KAU
([1]shindi@kau.edu.sa, [2]udawoud@hotmail.com)

Abstract- An object color is an integrated result of its surface nature and its ability to transmit and/or emit the incident light. The colors can be divided into two categories, namely pigment-based colors and structural colors. The later character is arisen from the physical interaction between the incident light and the nanostructure features of the receptor surface. Investigation of tissue coloration may be achieved by light-scatterometry methods and/or anatomical and morphological studies using SEM, cryo-SEM, and TEM. The photonic structures that can be inspired from plants are flowers, leaves, and fruits. These tissues are differed in their hierarchal nanometric structures. The clear blue color of the fruits of *Pollia condensata* is attributed to presence of cellulose-based helicoidal in the epidermal cell wall similar to those present at the leaves of *Danaea nodosa*. The fruit coloration of *Elaeocarpus angustifolius* is arisen from the presence of so-called iridosomes. These iridosomes are consisted of polysaccharides layers (including cellulose) and are connected directly to the cell membrane. Effect of chemical constituents and micro- and nano-structure of cellulose microfibrils on tissue coloration was discussed. Some attempts in synthesis of bioinspired tissues were summarized.

Keywords- *Tissue Coloration, Pigment-Based Colors, Structural Colors, Iridosomes, Cellulose Microfibrils*

I. INTRODUCTION

A. The Scientific Illustration of Color

The science of color or so-called chromatics is interested in perception of color by the human's eye and brain, the origin of color in materials, and theoretical fundamental of colorimetry in art as well as in physics from the view of electromagnetic radiation within the visible light. It is well known that our eye and brain are able to perceive a range of wavelengths ranged from about 380 nm to 700 nm, so this range is termed as the visible light.

As shown in Figure 1 and Table 1, the electromagnetic spectrum includes six regimes, namely gamma ray, X-ray, ultraviolet, visible light, infrared and radio waves. The visible light can be splitted into seven main divisions with wavelengths ranging from 380 to 740 nm. It is worth mentioning that the wavelength of a spectrum is inversely related to its energy (Figure 1)

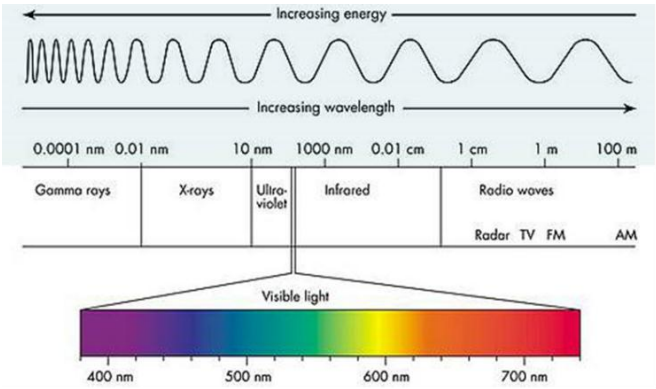

Figure 1. The Electromagnetic Spectra

TABLE I. THE COLORS OF THE VISIBLE LIGHT SPECTRUM (BOHREN, C. F., 2006).

	Color	Wavelength Range (λ) nm	Frequency Range (*ν*) THz
	Red	~ 700–635	~ 430–480
	Orange	~ 635–590	~ 480–510
	Yellow	~ 590–560	~ 510–540
	Green	~ 560–520	~ 540–580
	Cyan	~ 520–490	~ 580–610
	Blue	~ 490–450	~ 610–670
	Violet	~ 450–400	~ 670–750

B. Colors origin

Colors and luminescence of biomaterials occur naturally as a result of either the optical characteristics of their electrons or nanostructures effect (Zela *et al*. 2017).

In general, a color can be observed due to either wavelength-dependent light absorption or diffraction of light at geometrical features (Nassau , 2001).

1) Pigment-based colors

A pigment is a chemical compound that can change the color of reflected or transmitted light according to the wavelength absorbed selectively. This process is different from the luminescence whereby a material emits light. These colors

are caused by well-defined regular structures bearing a pigment (Zela *et al*. 2017).

Bioinspired photonic pigments were synthesized by Goerlitzer *et al.* (2018) using colloidal self- assembly technique. The self- assembly of colloidal particles are acting as wavelength- scale building blocks and can be able to replicate coloration from nature (Goerlitzer *et al*., 2018).

2) Structural colors

They are arisen from the physical interaction between the incident light and the nanostructure features of the receptor surface (Luiggi, 2013). The colors arisen from these structures are termed as structural colors and are brighter than pigment-based colors. (Vignolini *et al*., 2013). These colors of bio-tissues are often more intense and nearly angle-independent (Zela *et al*. 2017).

Recently, structural coloration has been attracting more interest for the applications of bio- inspired functional photonic structures and materials (Fu *et al*., 2016).

a) Structural Color in Plants

Biological tissues are more complicated than most artificial material in their anatomy and composition. This is due to biological materials may be consisted of different types of cells having different morphologies and chemical composition. For instance, the configuration, dimensions and size of epidermal cells affected strongly by light scattering method and visual appearance of a tissue (Vignolini *et al*., 2013).

b) Structural Color in Animals

It was reported by Xiao *et al.* (2015) that structural colors appeared due to interactions between light and nanometric-scale periodic structures have been discovered in many species of taxa. This can produces multiple bio-functions such as sexual signaling, camouflage, and aposematism.

For the animal kingdom (Vukusic and Sambles, 2003), different photonic systems including ordered (Vukusic *et al*., 1999; Michielsen and Stavenga, 2008), quasi-ordered (Heeso *et al*., 2010) and full random morphologies (Vukusic *et al*., 2007) have been investigated in many animal species including butterflies, beetles, jellyfishes and birds.

The optical response of a surface area can be measured using an optical goniometer as shown in Figure 2 (Vignolini *et al*., 2013).

Using the goniometer, the incident light beam illuminates the bio-sample at an angle θ_i which is precisely-controlled by the sample rotation. The reflected and transmitted light are recieved at θ_d which is varied by rotating the detector direction.

For flowers, the spectral range of the incident light ray must be matched with the spectral response of the photoreceptors of the pollinators. Xenon and deuterium lamps are used to determine the spectral characteristics in the UV scale, whereas deuterium and tungsten lamps were integrated to study the spectral properties in the UV to near-infrared regions (Vignolini *et al*., 2012a,b).

In the case of materials with weak scattering ability and/or having multilayer (hierarchal) structures, using polarized microscope can provide clear insight into the anatomy of the sample (Kinoshita *et al*., 2008). At the single cell scale, using confocal microscopy for studying of photonic structures (Vignolini *et al*., 2012).

3) Anatomical and Morphological Study

It is necessary to study the morphology and anatomical plant tissues to understand their spectral response. This is can be done by either of the following techniques:

a) The Scanning Electron Microscopy (SEM)

It is a valuable device to study the tissue surface due to its high resolution, rapid test and requiring easy preparation and pretreatment of the sample (Hindi, 2017). The pretreatment of biological samples is done by dehydration the sample by ethanol followed by chemical fixation using glutaraldehyde stabilize the structure of the tissue. The chemical fixation process is a sensitive technique due to it may destroy or alter the cuticle of the petal surface. Furthermore, sputtering the biological sample by gold nanoparticles using a sputtering coater must be done to prevent charging by the electron beam energy (McCully *et al*., 2009).

b) The Cryo-SEM Technique

It is used to study hydrated bio-tissues without any chemical pretreatments. The green samples are super-cooled using liuid nitrogen before gold sputtering-coated. The samples are imaged below −100°C (McCully *et al*., 2009).

c) Transmission Electron Microscopy (TEM)

It gives higher resolution comparing to the SEM, especially for nanometric photonic structures. In addition, many botanic tissues have hierarchal structures and/or nanocrystals that often need further investigation by TEM. Before imaging, the sample must be dehydrated and chemically cryo-fixed by liquid nitrogen (-196°C) to minimize or prevent structural defects in the sample (Studer *et al*., 2008).

II. Types of Bio-Tissues

There are some photonic structures that can be inspired from plants such as flowers (Whitney *et al*., 2009a,b; Glover and Whitney, 2010; ; Vignolini *et al*., 2012), leaves (Thomas *et al*., 2010) and fruits (Vignolini *et al*., 2012; Kolle *et al*., 2013). These tissues have outer layers with distinct hierarchal nanometric structures. Some of these structures include ordered periodic multilayers and diffraction gratings enhance their optical appearances (Vignolini *et al*., 2013).

A. Photonic Structures in Flowers to Attract Pollen Grain-Dispersers

Flowers are a group of organs that responsible for reproductions (Soltis and Soltis, 2004). It is worth to mention that the pollination process is a migration process of pollens from male anther to female stigma to create an embryo that keeps the botanic species. Many flowers attract pollinators by offering food, odor, temperature, color and/or shape. Studying petals as usually the most attractive parts of a flower is essential to understand the petals role in pollination (Gorton and Vogelmann, 1996; Kevan *et al*., 1996; Kevan and

Backhaus, 1998; Briscoe and Chittka, 2001; Stavenga, 2002; Dyer *et al*., 2007; Whitney *et al*., 2009a). The flower is a photonic structure that naturally specified to produce a strong coloration and iridescence helping pollinators to detect the flowers (Whitney *et al*., 2009a; Whitney *et al*., 2009b).

Flower coloration is arisen from pigments in most angiosperms (Tanaka *et al*., 2008; Brockington *et al*., 2011). Mixing more than one pigment and/or changing their chemical nature or concentration will give a wide color palette (Kay *et al*., 1981).

It was found that the shape of the epidermal cells containing the pigments significantly affects the intensity of the reflected color (Kay *et al*., 1981). For instance, the conical-shaped cells can generally enhance the brightness of the resultant color. This enhancing in the brightness can be attributed to the effect of the cone-shapes as reflectors that focuse the light onto the pigment-rich spots inside the epidermals cell enhancing the scattered light between the adjacent cells (Bone *et al*., 1985, Dyer *et al*., 2007; Kay *et al*. 1981; Vogelmann, 1993).

With the progress of nanometric characterization techniques using SEM, TEM, and X-ray diffraction (XRD), it is well known that structural color is formed essentially from surficial diffractive gratings (Whitney *et al*., 2009a,b). These gratings (Figure 2) can be viewed as arrays that can affect an incident light wave by modulating the phase and/or amplituding periodically of. For instance, in flowers, there are diffractive gratings on the epidermal cells composed of huge number of ordered striations (Kourounioti *et al*., 2013).

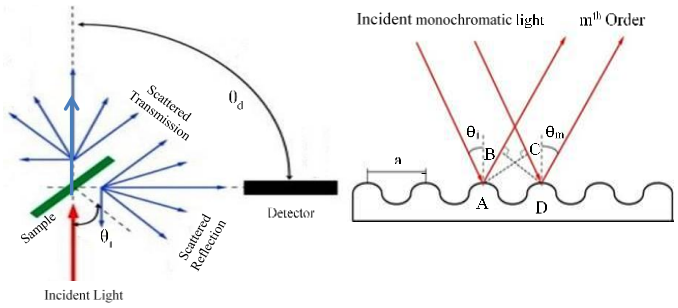

Figure 2. Diffraction gratings in flowers

Diffractive gratings (Figure 2) disperse incident monochromatic ray into different angular directions termed as orders. Light is reflected and scattered in the plane perpendicular to the direction of the striations, based on the following grating formula: $a(\sin\theta_m - \sin\theta_i) = m\lambda$, Where: θ_i is the angle of incidence, θ_m is the angle of the mth scattered order, λ is the light wavelength, and a is the grating periodicity.

The iridescent coloration can be observed in the tulip Queen of the night (Figure 3c). The iridescent effect can be isolated from the purple color arising from the anthocyanin pigment by separating off the transparent epidermal layer by using sharp tweezers and floating it onto a water surface. The epidermis is then transferred onto a planar substrate for optical characterization (Vignolini *et al*., 2013).

It is clear from Figures 3-c,d that the epidermal cells of tulip are flat and uniaxially elongated. Furthermore, the cell dimension is about 80×20 µm2, whereas the distance between striations is about 1µm (Vignolini *et al*., 2013).

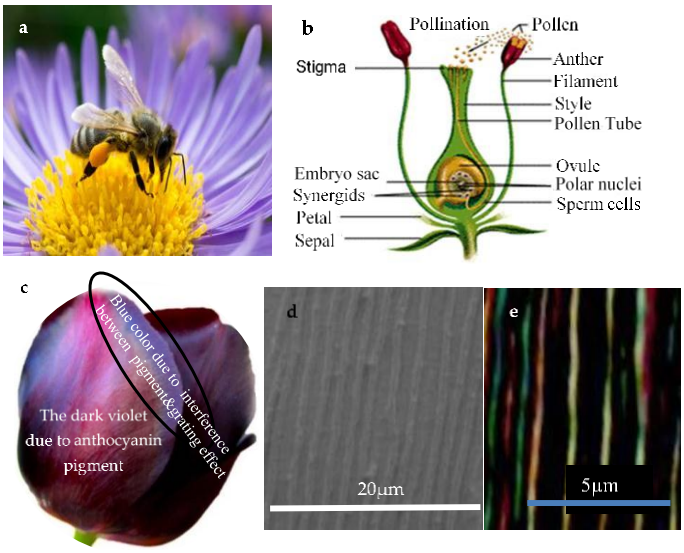

Figure 3. a) Flower pollination, b) Tulip flower, and c, d) Cryo-SEM image of epidermis of tulip petal (adapted from Vignolini et al., 2013)

B. Photonic Structures in Fruits to Attract Seed-Dispersers

Fruits are trying to attract insects or birds as seed dispersers using bright coloration (Vignolini *et al*., 2012; Cazetta *et al*., 2008), featured smell, offering food reward such as the fruit pulp and sugar juice (Galetti, 2002). Some plants trick their seed dispersers by mimicking the appearance of a delicious pulp belongs to other species without offering any true food (Galetti, 2002; Stournaras *et al*., 2013).

The 50-years old sample of African perennial marble berry (*Pollia condensata*) shown in Figures 3-c, 4-a is a good example to illustrate the difference between pigment-based colors and structural colors. It worth to mention that the color of its leaf is arisen due to chemical pigments, whereas the color of its fruit is a structural color. Accordingly, as seen from Figure 4-a that the pigment-derived leaf color has faded, while the fruit color still maintains its intense metallic-blue iridescence.

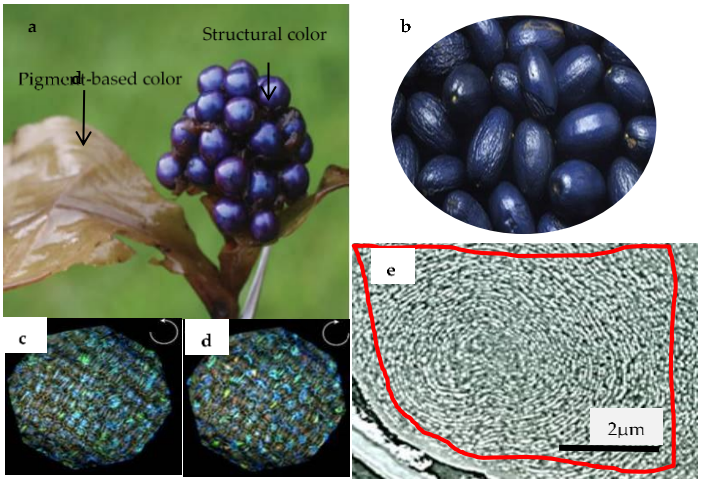

Figure 4. Photograph of *Pollia condensate* sample (adapted from Vignolini *et al*., 2012a, and b-e) *Delarbrea michieana*: b) Photograph of fruits, c,d) Clockwise- and anticlockwise-arranged iridosomes responsible for the fruit coloration, and e) TEM micrograph of a transverse section of the fruit (adapted from Lee, 2007).

In addition, the fruit of Pollia condensata (Figure 4b) is a true example of structural colors (Vignolini *et al*., 2012a). The clear blue color of the fruits is attributed to presence of cellulose-based helicoidal in the epidermal cell wall similar to those present at the leaves of *Danaea nodosa* (Kolle et al., 2013; Cazetta *et al*., 2008). These cellulosic structures constitute the major volume of the cell walls of the epicarp.

Accordingly, the stack micro- and nanostructure as well as the clear coloration were kept intact with the same parent aspects after drying the fruits. Similar cellulosic stack structures are present in fruits of *Margaritaria nobilis*, exhibiting strong iridescence in the blue–green regime of the wavelength scale (Kolle *et al*., 2013; Cazetta *et al*., 2008), so the fruits have a clear metallic aspect.

In addition, the fruit coloration of Elaeocarpus angustifolius was found to be arisen from the presence of so-called iridosomes in mature fruits. These iridosomes are consisted of polysaccharides layers (including cellulose) and are connected directly to the cell membrane. It was expected that the irodosoms are secreted by the cytoplasm of the epidermal cells (Lee, 1991). It worth mentioning that each cell reflects selectively left- or right-handed circularly polarized light that can give us clear insight the photonic structure (Lee, 1991) as indicated in Figure 4c-e.

It was found by Lee *et al*. (2000) the blue-colored fruits of *Delarbrea michieana* (Figure 8) have iridosomes similar to those present in *Pollia condensata* fruit (Figure 4a) that responsiple for their color.

C. Photonic Structures in Leaves

The functional structural color in leaves is not completely understood (Thomas *et al*., 2010; Bone *et al*., 1985.; Lee *et al*., 1990) and still needs an excess of investigations.

There are different mechanisms used by plant leaves to protect themselves from the intense UV-ray by optimizing their light capturing as follow:

- Many tropical rainforest plants have epidermal cells with convex walls that focus the light onto the photosynthetic layers within the leaf (Bone *et al*., 1985).
- In high mountains, plants are suffering from high UV exposure which may harm their tissues. Therefore, to lower the intensity of the UV penetrating to its reproductive organs enveloped by bracts, edelweiss (*Leontopodium nivale*) has a nanostructured-woolly layer around its bracts that is able for multi-scattering the incident UV-ray (Vigneron *et al*., 2005).
- The blue spruce trees (*Picea pungens*) has needles coated with thin waxy layers (45) similar to the leaves of *Dudleya brittonii* (Mulroy, 1979) that is an efficient tool for UV-filtering to a certain extent.
- The epidermal layers in leaves of *Selaginella willdenowii* and *Begonia pavonina* have more complicated structures that converting the incident light wavelengths into the UV and blue spectra. The biological function of this ability is not well understood yet (Gould and Lee, 1996; Bradshaw *et al*., 2010). The expected mechanism arises either from multilayer nature of the epidermal cell walls that exhibit different refractive indices for the incident light (Thomas *et al*., 2010; Lee *et al*., 2000; Hébant and Lee, 1984), or by the presence of cellulosic microfibrils based-stacking layers with different orientations forming a helicoid structure similar to a liquid crystal nematic phase (Gould and Lee, 1996, Graham *et al*., 1993; Neville and Caveney, 1969).

The principle of multilayer effects of light is illustrated in figure 5 (a, b). Incident light is reflected at each interface between two layers with two different refractive indices. Depending on the wavelength and on the angle of incidence, the reflected beams interfere constructively or destructively. The multilayer acts as a color screen reflecting a certain color (else a specific wavelength or a range of wavelengths) while transmitting its complementary color.

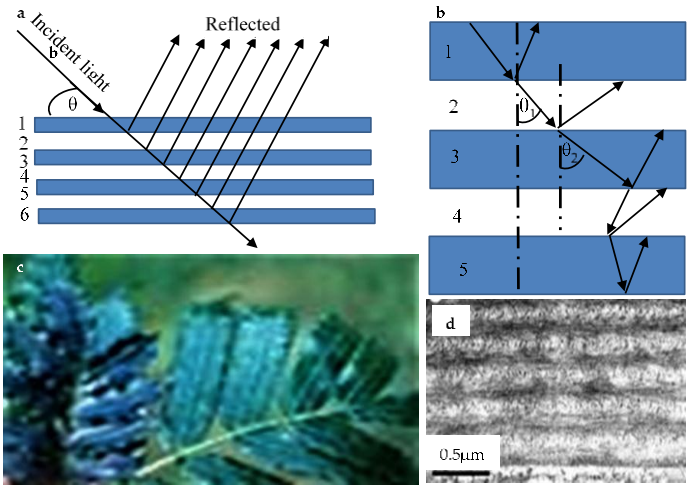

Figure 5. Multilayer interference mechanism. (a) Reflection of incident beam on interfaces between layers of different materials. (b) Close-up image of this light reflection, c) Blue juvenile leaf and green adult leaf of *Danaea nodosa*, and e) TEM transverse section images of outer cell wall of a juvenile leaf of the same species (adapted from Lee, 2007).

Multilayers can be found in plants: a) at the surface of the leaves on top of the epidermis, and b) within special intracellular organelles known as iridoplasts that are located inside upper epidermis cells.

The color of juvenile leaves differs from one species to another. For instance, juvenile leaves of *Danaea nodosa* are displayed a brilliant blue coloration while the adult leaves are colored by green as shown in Figure 5c. (Thomas *et al*., 2010; Lee and Lowry, 1975; Lee, 1997). The blue color of *Danaea nodosa* leaves are arisen due to multilayer helicoidal structure of the upper epidermis (Figure 5d). Contrarily, the blue iridescence in *Begonia pavonina* is arisen from the iridoplasts found within the epidermal cells (Gould and Lee, 1996).

III. Micro- and Nano-Structure of Cellulose Microfibrils

The cell wall of plants is made up of a matrix of polysaccharides (alpha-cellulose and hemicelluloses), pectin in the primary cell wall and lignin secondary cell wall (Roberts *et al*., 2000; Cosgrove, 2005; Hindi and Abouhassan, 2016). Spectroscopic studies showed its differentiation into primary and secondary walls (Li *et al*., 2014). The two classes of walls are different in their chemical constituents and structure that grant the plant kingdom a wide range of biodiversity (Popper, 2008).

A. Chemical Structure of Wood Polymers

1) Cellulose

Cellulose is a straight, uniaxial, and homopolymer constituted of β-D-glucopyranose units conjugated by 1-4-glucosidic linkages (Matthews *et al*., 2010; Brett, 2000; Somerville, 2006).

The conformation of the cellulosic chains and their packing manner into microfibrils is not completely understood (Matthews et al., 2010). Each cellulosic chain is composed of repeated cellobiose units constructing many different architectures (Klemm *et al*., 2005). The degree of polymerization of a single cellulosic chain may be contains of about 10.000 glucopyranose units (Zabel and Morrel, 1992) or more. The longitudinally adjacent cellulosic chains are attached together by hydrogen bonding and/or van der Waals forces according to the distance magnitude between the chains.

2) Hemicelluloses

They are composed from simple sugar monomers, namely glucose, galactose, mannose, xylose, arabinose and glucuronic acid. It has β-(1→4)-linked backbones in an equatorial configuration. These sugars can conjugated to synthes different compounds such as xylans, glucomannans, xyloglucans, mannans and beta-(1→3,1→4)-glucans. The hemicelluloses role is the strengthening the cell wall by interacting the other biopolymers. Contrarily to cellulose, hemicellulose has a random, amorphous structure with weak strength and is easily hydrolyzed by dilute reagent (Hindi and Abouhassan, 2016).

3) Lignin

Lignin is a branched biopolymer found in plant cell wall. It is polymerized from phenylpropane monomers by the cross-linking in different chemical bonds. Although its resistant ability to biochemical effects, white-rot fungi and actinomycetes can secrete enzymes to breakdown it.

4) Pectin

Pectins are heterogeneous polysaccharides consisting of 1→4-α-D-galactosyluronic acid. Three pectin types were isolated from primary cell walls of plants, namely homogalacturonan (HG), rhamnogalacturonan-I and substituted galacturonans. The first type is a linear polymer composed from 1→4-α-D-galactosyluronic units. Some carboxyl groups are methyl esterified or O-acetylated.

B. Microfibrill Organization

Cellulose or so-called glucan chains are aggregated just after they are biosynthesized to form microfibrils aggregates with different cross dimensions. The constructed microfibrils were found to be varied according to the dimensions, hydrogen-bonding and molecular orientation of their parent (French *et al*., 2004; Atalla, 1984; Habibi *et al*., 2010).

The secondary cell wall of plants was found to be differentiated into three distinct layers, namely S_1, S_2, S_3 (Figures 6,7). Within each layer, the cellulose microfibrils are parallel in high organization, while the microfibril orientation looks different from on layer to another (Plomion *et al*., 2001; Barnett *et al*., 2004). The ideal structure of wood cell wall is attributed to either the hierarchal organization of the cellulosic microfibrils or the amazing mixing manner of the principle polymers constituting wood (Chaffey, 1999; Plomion *et al*., 2001; Li *et al*., 2014).

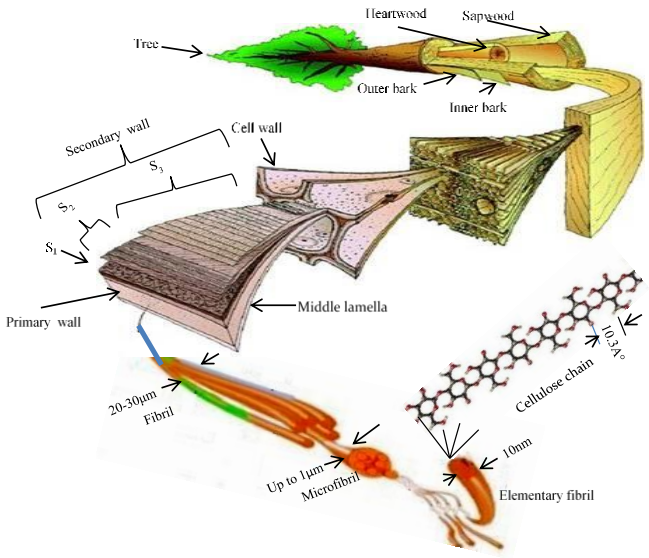

Figure 6. The hierarchical structure of wood (adapted from Teischinger, 2016)

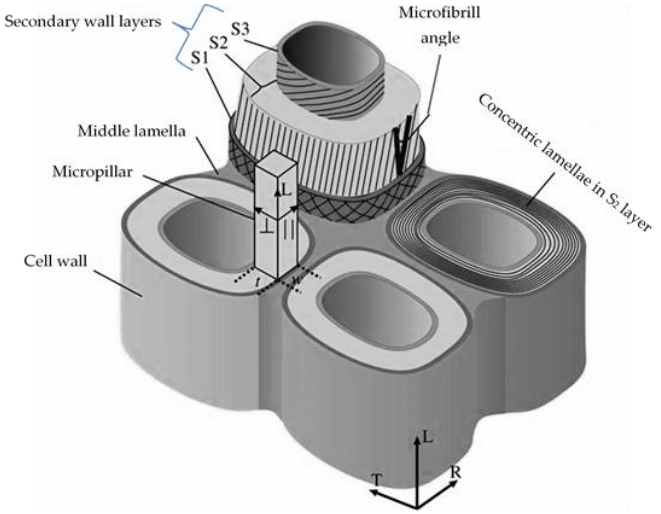

Figure 7. Ultrastructure of perennial cell wall (adapted from Rafsanjani et al., 2014).

C. Photonic Structures in Cellulose Microfibrils

The part of spectrum ranged from ultraviolet up to the blue wavelength (UV–blue) was reported to be generated by cellulose microfibrils which are organized internally in a helicoid construction (Figure 8).

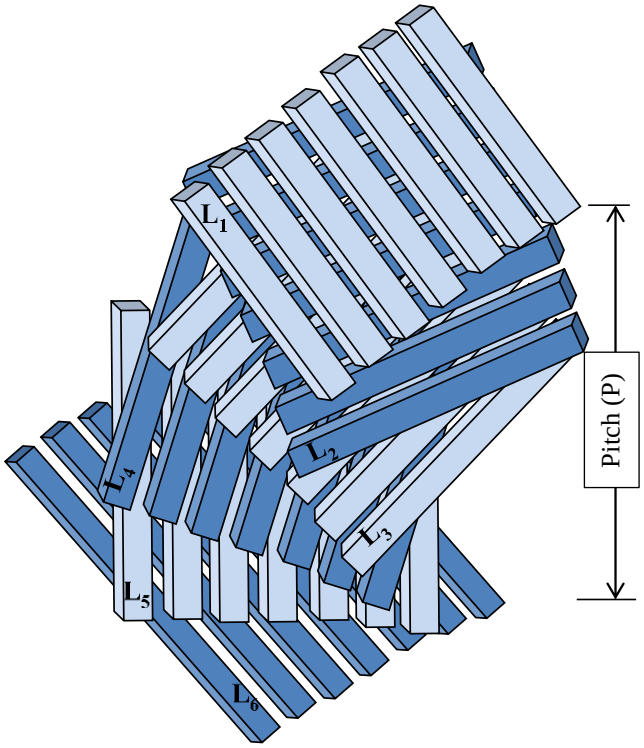

Figure 8. Schematic of a helicoidal-stacking nature of cellulosic chains within microfibrils

Within the cell wall, cellulose microfibrils are deposited parallel to each other in successive layers in which each layer is rotated by a constant angle. Accordingly, each layer has its own refractive index. When the light beam passes from one layer to the subsequent one, it will generate the reflection of circular polarized light with opposite helicoidicity to the rotating stack. The distance (p) between layers of fibrils with the same orientation is an indicator to the reflected wavelength, while the rotation direction is a measure of the circular polarization of the reflected light (De Vries, 1951).

IV. Synthetic Bioinspired Tissues (SBT)

The surface morphology of a SBT can be fabricated by cast method without harming the tissue structure. A double-step pattern copy method has been performing using a quick-setting viscous dental wax for casting onto the sample to produce a negative replica. The dental wax replica is used as a mold to form a positive epoxy replica of the sample (Vignolini *et al.*, 2013).

Zela *et al*. (2017) manufactured a lamellar nanostructure mimicking ridge-shaped of the Morpho butterfly using a 3d-direct laser writing technique. The obtained ultrastructure and its optical characteristics was comparable to the Morpho butterfly features.

In addition, Xiao *et al*. (2015) have inspired the self-assembled melanosomes producing colors in avian feathers using polydopamine-based synthetic melanin nanoparticles.

They get a high refractive index and broad absorption spanning across the UV–visible range analogous to that for natural melanins. This finding provides benefits for structural colors over other polymeric nanoparticles (Xiao *et al*., 2015).

The blue color of a Morpho butterfly wings originates structurally due to the diffraction and interference effects of light due to the wing's microstructures (Sato *et al*., 2009). Furthermore, tunable structural color for damselfish reported by them that changing between green and blue is done reversibly. They these mimicked these phenomenon and prepared functional structural color films using lifting and templating techniques. (Sato *et al*., 2009).

Silicon can be used to fabricate several photonic functionalities useful in many applications. Here, Cao *et al*. (2010) generated wide spectrum of colors by harnessing the strong resonant light scattering properties of Si nanostructures under white light illumination. They achieved this through controlling structure size, dielectric conditions, and illuminations (Cao *et al*., 2010).

Tan *et al*. (2014) assembled a cheap plasmonic material for photorealistic printing with aluminum nanostructures instead of gold and silver using photolithography and nanoimprint lithography. The advantages achieved will create a new generation of cheap and high-resolution plasmonic printing materials for security tagging, cryptography, and data storage.

In addition, An *et al*. (2009) fluorescent nanowires else one dimensional or their higher-dimensional structures such as nanowebs and nanofabrics is expected to open new applications such as nanoscale optoelectronics, sensing, and biological devices.

V. Conclusions

- The colors can be divided into two categories, namely pigment-based colors and structural colors.
- The structural colors are arisen from the physical interaction between the incident light and the nanostructure features of the receptor surface.
- The photonic structures that can be inspired from plants are flowers, leaves, and fruits that differed in their hierarchal nanometric structures.
- The clear blue color of the fruits of *Pollia condensata* is attributed to presence of cellulose-based helicoidal in the epidermal cell wall similar to those present at the leaves of *Danaea nodosa*.
- The fruit coloration of *Elaeocarpus angustifolius* is arisen from the presence of so-called iridosomes which are consisted of polysaccharides layers (including cellulose) and are connected directly to the cell membrane.
- The part of spectrum ranged from ultraviolet up to the blue wavelength is generated in the helicoid construction of cellulose microfibrils.
- Within the cell wall, each of successive layers of cellulose microfibrils has its own refractive index. So, when the light beam passes from one layer to the subsequent one, it will generate the reflection of circular polarized light with opposite helicoidicity to the rotating stack.
- The surface morphology of bioinspired tissues can be fabricated by cast method without harming the tissue structure.

Conflicts of Interest

The authors declare no conflict of interest.

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