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Extraction and Optical Study of Natural Dyes for Dye-Sensitized Solar Cell Application

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Abstract

Extraction and study of the optical properties of natural dyes from several plant parts such as leaves, flowers, tubers, and fruits have been carried out using a simple maceration method and UV-Vis absorption characterization. The samples were divided into several groups in this study, namely the flower group (*Tagetes sp.*, *Paeonia sp.*, *Centaure sp.*, *Chrysanthemum sp.*, *Carthamus tinctorius, Gomphrena globosa, Myosotis sylvatica, Lavandula sp., Clitoria ternatea, Matricaria chamomilla, Hibiscus sabdariffa*, and *Rosa sp.*); the leaf group (*Mangifera indica, Moringa oleifera, Graptophyllum pictum, Spinacia oleracea, Citrus hystrix,* and *Terminalia sp.*); and the tuber and fruit group (*Solanum lycopersicum, Cucurbita moschata, Garcinia mangostana, Curcuma longa, Beta vulgaris*, and *Caessapina sp.*). The extract of natural dyes was successfully produced using a low-cost and simple extraction approach via dehydration, immersion for 2×24 hours in ethanol, and followed by filtration. The optical properties of the dyes in UV and visible light range were observed using a UV-Vis spectrophotometer in the wavelength of 400–700 nm. The absorption behaviour showed dominant peaks at different wavelengths for each group. Potentially, to do a combination of dyes (co-sensitization: using more than a dye with different absorption spectra to achieve a panchromatic response) to widen the wavelength absorption for dye-sensitized solar cell (DSSC) applications.

Keywords: natural dye, optical study, absorption, UV-Vis, maceration

Abstrak

Ekstraksi dan kajian sifat optik zat warna alami dari beberapa bagian tumbuhan seperti daun, bunga, umbi, dan buah telah dilakukan dengan menggunakan metode maserasi sederhana dan karakterisasi serapan UV-Vis. Sampel dibagi menjadi beberapa kelompok pada penelitian ini yaitu kelompok bunga (*Tagetes sp., Paeonia sp., Centaure sp., Chrysanthemum sp., Carthamus tinctorius, Gomphrena globosa, Myosotis sylvatica, Lavandula sp., Clitoria ternatea, Matricaria chamomilla , Hibiscus sabdariffa,* dan *Rosa sp.*); kelompok daun (*Mangifera indica, Moringa oleifera, Graptophyllum pictum, Spinacia oleracea, Citrus hystrix,* dan *Terminalia sp.*); dan kelompok umbi dan buah (*Solanum lycopersicum, Cucurbita moschata, Garcinia mangostana, Curcuma longa, Beta vulgaris,* dan

Caessapina sp.). Ekstrak zat warna alam berhasil diproduksi dengan menggunakan pendekatan ekstraksi yang murah dan sederhana melalui dehidrasi, perendaman dalam etanol selama 2×24 jam, dan dilanjutkan dengan filtrasi. Sifat optik zat warna pada rentang sinar UV dan sinar tampak diamati menggunakan spektrofotometer UV-Vis pada panjang gelombang 400–700 nm. Perilaku penyerapan menunjukkan puncak dominan pada panjang gelombang yang berbeda untuk setiap kelompok. Dari hasil penelitian ini, berpotensi untuk melakukan kombinasi pewarna (ko-sensitisasi: menggunakan lebih dari satu pewarna dengan spektrum serapan berbeda untuk mencapai respons pankromatik) untuk memperluas penyerapan panjang gelombang untuk aplikasi sel surya tersensitisasi pewarna (DSSC).

Kata Kunci: pewarna alami, studi optik, penyerapan, UV-Vis, maserasi

1. Introduction

As the world's energy demand continues to increase, fossil fuels, as the main source of electrical energy, have been explored to provide electric power, affecting critical environmental problems. The development of renewable, clean, and sustainable energy supplies has become an necessary plan to continue the human civilization. Among renewable energies, solar energy is a promising energy source because of its relatively short duration, abundance, pollution-free and safety. Therefore, research on solar cells is a priority in the current strategy for developing renewable energy supplies. DSSC (dye-sensitized solar cell) is a generation of solar cells that utilize photoelectrochemical mechanisms to provide a renewable energy with affordable production costs (Boschloo, 2019). Until now, research is still ongoing to improve its efficiency. Various modification methods that can be used in the development of materials on DSSC components (Benesperi et al., 2018; Błaszczyk, 2018; Boschloo, 2019; Iftikhar et al., 2019; Iqbal and Khan, 2018; Li et al., 2019; Mariotti et al., 2020; Mohiuddin et al., 2018; Mudzakir et al., 2020; Rudra et al., 2019; Sharma et al., 2018; Tański et al., 2019).

Furthermore, since its discovery, most research has involved ruthenium-based dye-based DSSCs (O'Regan and Grätzel, 1991). However, the existence of transition metals in these dyes is related with some dilemmas, namely that transition metal-based dyes are more expensive than metal-free organic ones, toxic, and their availability is limited (Cole et al., 2019). The utilization of natural/organic dyes in DSSC devices is becoming popular because of their ease of production and low cost (Darwis, 2020; Hosseinnezhad et al., 2018). Despite the prospect of replacing ruthenium-based dyes, however, organic dyes have the disadvantage of a narrow and sharp absorption band, such that the consequences will drastically limit the photon-to-electron conversion efficiency of DSSC devices (Chen et al., 2005). When the spectrum of sunlight cannot be completely covered by optical absorption that comes from sensitizing the semiconductor surface with a single dye, one potential solution is co-sensitization, that is, combine multi-dyes with different absorption spectra to reach a panchromatic response (Cole et al., 2019). Before applying that strategy, it is very important to characterize every single dye. A different characteristic of natural dye can be produced from different plant parts.

Indonesia is a tropical country which is rich in natural resources. One of the utilization of natural resources is the manufacture of natural dyes. In almost all areas local plants are widely distributed that can be used as a source of natural dyes, such as butterfly pea flower, secang wood, rosella flower skin, black rice, suji leaves, katuk leaves, grass jelly leaves, purple sweet potato, turmeric, carrots, etc. The types of natural dye compounds contained in plants are chlorophyll (green) in leaves; carotene (orange yellow) in tubers and leaves; lycopene (red) in flowers and fruit; flavones (yellow) in flowers, roots and wood; anthocyanins (reddish yellow, purple red) in fruits and flowers; betalain (red yellow) resembles anthocyanins or flavonoids in red beets; xanthones (yellow) in mangoes (Tranggono, 1990). The existence of these pigments is located in the vacuole cells of the plant itself, so most of them can be found and can be taken from several plant organs, such as flower crowns, leaves, fruits, and seeds.

These natural dyes, extracted from various plant sources, have shown promising potential as viable alternatives to conventional synthetic dyes due to their eco-friendly and renewable nature. The plants

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utilized in this study as natural dyes for solar cell applications include the flower group (*Tagetes sp., Paeonia sp., Centaure sp., Chrysanthemum sp., Carthamus tinctorius, Gomphrena globosa, Myosotis sylvatica, Lavandula sp., Clitoria ternatea, Matricaria chamomilla, Hibiscus sabdariffa,* and *Rosa sp.*); the leaf group (*Mangifera indica, Moringa oleifera, Graptophyllum pictum, Spinacia oleracea, Citrus hystrix,* and *Terminalia sp.*); and the tuber and fruit group (*Solanum lycopersicum, Cucurbita moschata, Garcinia mangostana, Curcuma longa, Beta vulgaris*, and *Caessapina sp.*). The selection of these specific plants was based on several factors, including their abundance, ease of extraction, and their known potential to yield high-quality natural dyes. Furthermore, the decision to use them was also motivated by their established traditional uses as natural dyes in various applications. In this preliminary study with friendly environmental considerations, the extraction using a simple maceration method and optical characterization using UV-Vis spectrophotometer of several natural extracts from plant segments such as leaves, flowers, tubers, and fruits have been reported.

2. Methods

The raw materials were prepared from the part (i.e. flower, leaf, tuber, and fruit) of several plants. The samples were divided into several groups in this study, namely the flower group (*Tagetes sp., Paeonia sp., Centaure sp., Chrysanthemum sp., Carthamus tinctorius, Gomphrena globosa, Myosotis sylvatica, Lavandula sp., Clitoria ternatea, Matricaria chamomilla, Hibiscus sabdariffa,* and *Rosa sp.*); the leaf group (*Mangifera indica, Moringa oleifera, Graptophyllum pictum, Spinacia oleracea, Citrus hystrix,* and *Terminalia sp.*); and the tuber and fruit group (*Solanum lycopersicum, Cucurbita moschata, Garcinia mangostana, Curcuma longa, Beta vulgaris*, and *Caessapina sp.*). The extract of natural dyes was produced using a low-cost and simple extraction approach via maceration method as followed: a dehydration/drying process of the plant part, grinding, immersion of the powder for 2×24 hours in ethanol 99% with ratio of 2 gram : 30 ml, and then followed by filtration. Distillation is carried out to separate the ethanol solvent and natural dye extract by boiling so that evaporation occurs. The results of natural dye extracts were obtained in the form of a colored concentrated liquid without the presence of ethanol solvent. The method steps that have been described are illustrated in Figure 1. The optical properties of the dyes were observed using an Analytic Jena Specord 200 Plus UV-Vis spectrophotometer. The absorption in UV and visible light range were observed in the wavelength of 400–700 nm. The results of the characterization are absorbance vs wavelength graphs which are then merged with the origin software.

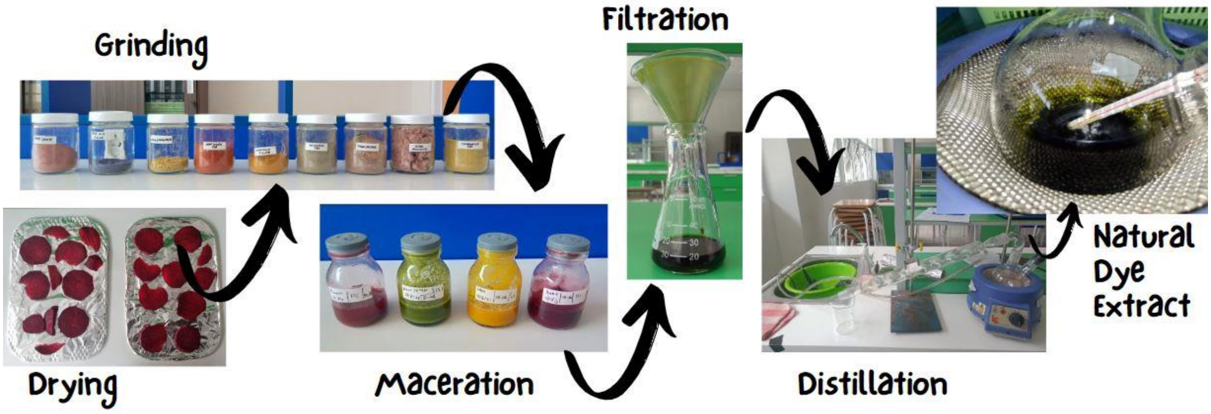

Figure 1: Illustration of the process of extracting natural dyes using a simple method: drying, grinding, maceration, filtration, then distillation.

3. Result and discussion

The absorption of three groups of natural dyes extracted from plant parts in UV and visible light spectrum are shown in Figures 3–4 and Tables 1–3. In general, the absorption band of dye reveal the maxima at points as indicated in the figures. The absorption behavior of some dyes showed similar bands, but the other have unique optical properties at distinctive wavelengths from the other. The absorption bands in this study experienced a shifted wavelength range when compared to the results of other studies using natural dyes (Hosseinnezhad et al., 2018; Iqbal and Khan, 2018; Lin et al., 2021; Sunder Sharma et al., 2020; Tian et al., 2020; Urbani et al., 2019). This is because the use of the extraction method, the type of solvent (related to the level of solvent polarity and solvent concentration), the ratio of the solvent and the mass of the base powder, and the length of the maceration time greatly affect the optical characteristics of natural dyes extracted from natural materials.

Another important thing that greatly affects the absorption characteristics of natural dyes is the pigment content in the dye. Each plant including every part of the plant contains certain pigments that will determine the characteristics of the absorption band against UV-Vis waves. For instance, in the case of leaf-based dyes, one region of absorbance band was found in the blue range at about 420 nm and other in the red range at about 660 nm; these due to the peaks of maximum absorbance of chlorophyll A (Hernández-Martínez et al., 2012). However, appropriate to other bands in those regions, the existence of a mixture of chlorophyll B in leaf-based dyes also should be suggested. The other case, the absorption spectrum peaks of mangosteen pericarp dye, for example, close to the cyanidin-anthocyanin range of 400–500 nm (Agustini et al., 2013). For further observation of the pigment content of these natural dye in this preliminary research, the other examinations like FTIR are needed. However, from the provided UV-Vis results, the absorption characteristics in UV and light spectrum of each natural dyes are sufficient for the basis of selecting a combination of several dyes (co-sensitization) to widen the wavelength absorption in dye-sensitized solar cell (DSSC) applications.

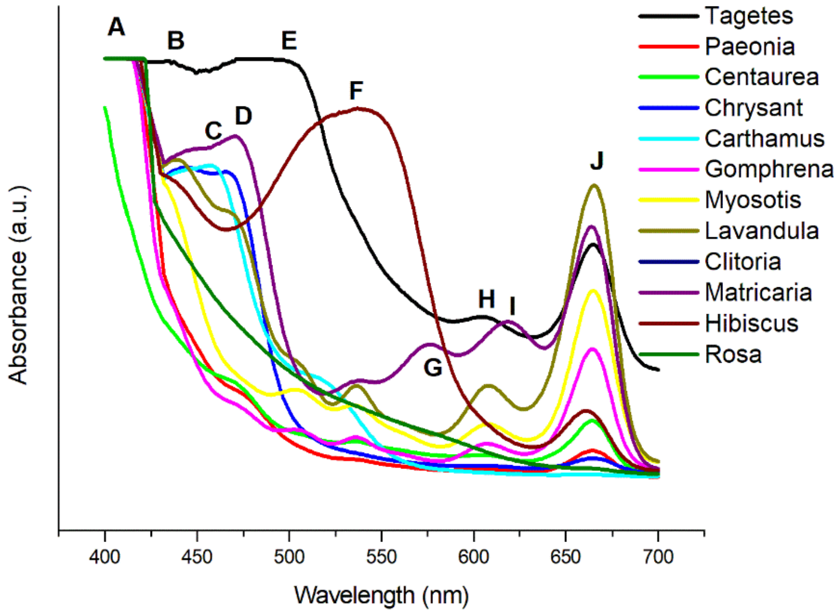

Figure 2: UV-Vis absorption spectra of the natural dyes from the flower group

Table 1: The list of absorption bands of in UV-Vis spectra of the natural dyes from the flower group (referred to Figure 1)

Sample		**Peak**									
		A	**B**	**C**	**D**	**E**	**F**	**G**	**H**	**I**	**J**
Tagetes sp.		√	√	-	-	√	-	-	√	-	√
Paeonia sp.		√	-	-	√	-	-	-	-	-	√

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Centaurea sp.		√	-	-	√	-	-	-	-	-	√
Chrysanthemum sp.		√	√	-	√	-	-	-	-	-	√
Carthamus sp.		√	√	√	-	√	-	-	-	-	-
Gomphrena sp.		√	-	-	√	√	√	-	√	-	√
Myosotis sp.		√	√	-	-	√	-	-	√	-	√
Lavandula sp.		√	√	-	√	√	√	-	√	-	√
Clitoria sp.		√	√	-	√	-	√	√	-	√	√
Matricaria sp.		√	√	-	√	-	√	√	-	√	√
Hibiscus		√	√	-	-	-	√	-	-	-	√
Rosa sp.		√	-	-	-	-	-	-	-	-	-

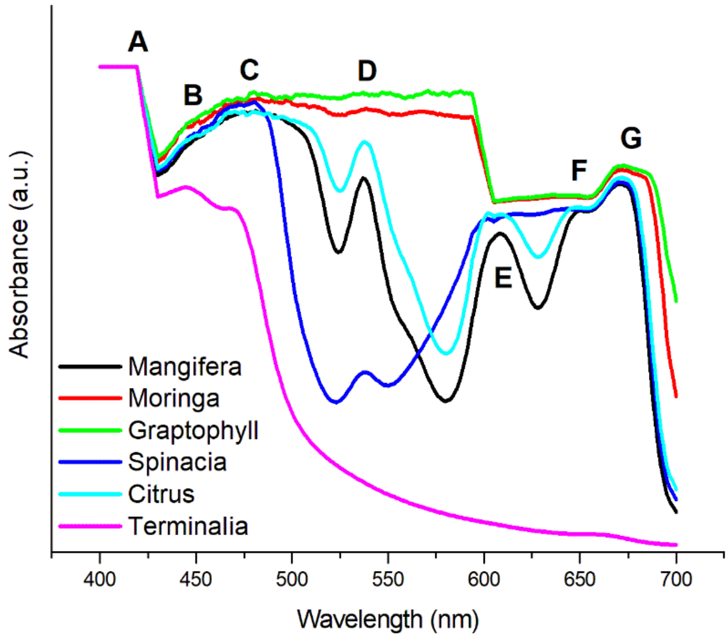

Figure 3: UV-Vis absorption spectra of the natural dyes from the leaf group

Table 2. The list of absorption bands of in UV-Vis spectra of the natural dyes from the leaf group (referred to Figure 2)

Sample		Peak						
		A	B	C	D	E	F	G
Mangifera indica		√	√	√	√	√	√	√

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Moringa oleifera		√	√	√	-	-	-	√
Graptophyllum pictum		√	√	√	-	-	-	√
Spinacia oleracea		√	√	√	√	-	-	√
Citrus hystrix		√	√	√	√	√	√	√
Terminalia catappa		√	√	√	-	-	-	-

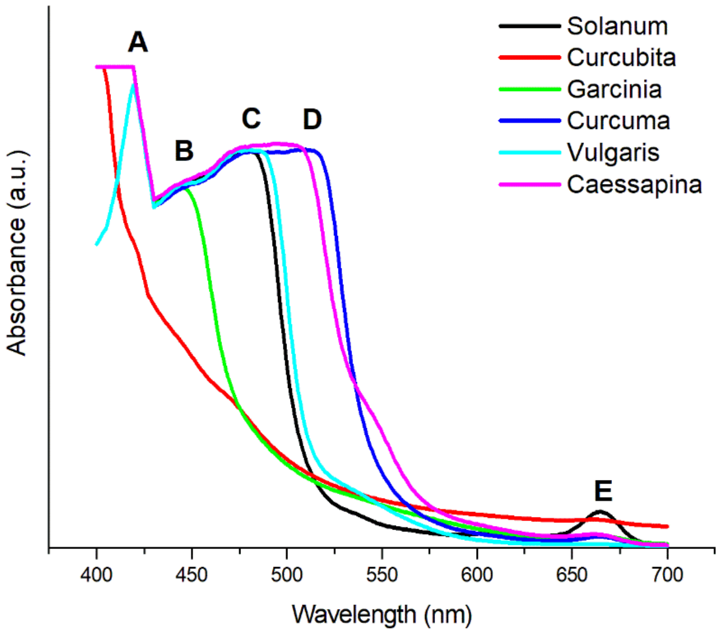

Figure 4: UV-Vis absorption spectra of the natural dyes from the tuber and fruit group

Table 3: The list of absorption bands of in UV-Vis spectra of the natural dyes from the tuber and fruit group (referred to Figure 3)

Sample		**Peak**				
		A	**B**	**C**	**D**	**E**
Solanum sp.		√	√	√	-	√
Curcubita sp.		√	-	-	-	-
Garcinia sp.		√	√	-	-	√
Curcuma sp.		√	√	√	√	√
Beta Vulgaris sp.		√	√	√	-	√
Caessapina lapan		√	√	√	√	√

4. Conclusion

Natural dyes were extracted from several plant parts, i.e. flower, leaf, tuber, and fruit using a simple maceration method. The optical properties of the dyes in UV and visible light range were characterized using a UV-Vis spectrophotometer in the wavelength of 400–700 nm. Their absorption characteristics reveal dominant peaks at different wavelengths. These preliminary results can be the basis for selecting a combination of several dyes (co-sensitization: using more than one dye with different absorption spectra to achieve a panchromatic response) to widen the wavelength absorption for dye-sensitized solar cell (DSSC) applications.

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