A Study on the Optoelectronics Parameters of Natural Dyes Extracted From Beetroot, Cabbage, Walnut and Henna For Potential Applications in Organic Electronics

Peshawa O. Amin

Charmo University

Fahmi F. Muhammadsharif (✉ fahmi982@gmail.com)

Koya University <https://orcid.org/0000-0002-4563-9671>

Salah Raza Saeed

Charmo University

Khaulah Sulaiman

Universiti Malaya

Research Article

Keywords: optoelectronic parameters, natural dyes, beetroot, cabbage, walnut, henna

Posted Date: July 29th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-732502/v1>

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Version of Record: A version of this preprint was published at Journal of Fluorescence on October 25th, 2021. See the published version at <https://doi.org/10.1007/s10895-021-02837-7>.

A study on the optoelectronics parameters of natural dyes extracted from beetroot, cabbage, walnut and henna for potential applications in organic electronics

Peshawa O. Amin¹, Fahmi F. Muhammadsharif^{2,*}, Salah Raza Saeed¹, and Khaulah Sulaiman³

(1) Charo Center for Research, Training and Consultancy, Charo University, 46023 Chamchamal, Kurdistan Region, Iraq

(2) Department of Physics, Faculty of Science and Health, Koya University, Koya KOY45, Kurdistan Region - F.R., Iraq

(3) Low Dimensional Materials Research Centre (LDMRC), Department of Physics, Faculty of Science, Universiti Malaya, 50603 Kuala Lumpur, Malaysia

Corresponding author email: fahmi982@gmail.com

Abstract

In this work, the optoelectronic parameters of natural dyes extracted from beetroot, red cabbage, walnut leaves, and henna are determined and investigated. These include optical energy gap (E_g), extinction coefficient (k), refractive index (n), dielectric constant (ϵ_r) and optical conductivity (σ_r). Results showed that cabbage dye exhibited a maximum values of $n = 6.08$, $\epsilon_r = 37.18$ and $\sigma_r = 13.92 \times 10^{-3} S/cm$, while henna dye presented the lowest values of $n = 1.19$, $\epsilon_r = 1.5$ and $\sigma_r = 0.039 \times 10^{-3} S/cm$ compared to the optoelectronic parameters of the other studied samples. The optical absorption in the dyes were found to obey a direct allowed transition between the molecular energy levels. The results presented here can be of a special importance for some potential applications in organic electronics such as organic photovoltaics and sensors.

Keywords: optoelectronic parameters, natural dyes, beetroot, cabbage, walnut, henna.

1. Introduction

Nowadays, the use of organic dyes has become the focus of interest in both academia and industry owing to the unique features offered by the dyes including cost-effectiveness, flexibility, electronic tuneability and light weight properties [1-3]. Furthermore, organic dyes show higher absorption coefficient and stronger exciton generation compared to those of the inorganic materials [4, 5]. Commercial dyes are commonly used in various applications, but they suffer from high cost, supply shortage, heavy metal toxicity and challenging synthesis, which ultimately limit their broad usage compared to the natural dyes. Alternatively, it is possible to overcome these limitations by utilizing natural dyes [5,6].

Dyes are organic compounds which are capable of well absorbing light and retaining their intrinsic color because of the presence of chromophore [7]. Betalains are vacuolar nitrogenous plant pigments having a core structure known as betalamic acid [8]. The best-known source of betalains can be beetroot and cacti [9]. Anthocyanins was also found in a large group of plants which contain red-blue pigments, which is water soluble and pH sensitive [10]. Red cabbage is one of the main sources of anthocyanins and are chemically glycoside moieties of anthocyanidins derived from the flavylum [11]. In addition to different natural pigments, brown dye and red-orange dye were found in juglone (black Walnuts) and lawsone (henna plant), respectively [12, 13]. A review of literature showed that organic dyes have been successfully utilized in photovoltaic devices [14-20]. Additionally, organic dyes can be of a special interest for the coloration technology and various organic electronics applications. However, before these materials are highly employed for viable applications, a broad investigation on their optoelectronic properties is necessary. Therefore, in the current research work, different natural dyes are extracted through a green method and their optoelectronic parameters are comprehensively investigated. The natural dyes were extracted from

beetroot, red cabbage, walnut leaf, and henna using specified chemical solvents. This can be a unique detailed study which is performed to determine the most important parameters of the dyes including optical energy gap, refractive index, dielectric constant, and optical conductivity. The results are primarily important when these natural dyes are considered for the application of organic electronics.

2. Materials and methods

The Beetroots, Red Cabbages and Henna were purchased in the local markets, while Walnut leaves were collected in a local farm. The raw materials were cut into small pieces and crushed by a grinder followed by drying process to produce very fine powders. To extract the dyes, ethanol, distilled water and a combination of ethanol-HCl were used. Table 1 shows the designation of the extracted samples and the used solvents for this purpose. In a first approach, 100 ml distilled water was put into a specified beaker then boiled on a hotplate for 2 minutes. Later on, the beakers were transferred, and 50 g of each sample was inserted into the boiled water, separately. The prepared solutions of sample B1 and C1 were left for 2 hours and then filtered and stored in the refrigerator for 24 hours. In the second approach, 100 ml of ethanol was mixed with HCl (0.1) with the ratio of 99:1, then 50 g of samples B2 and C2 was put into the beakers and left for 2 hours. Afterwards, filtration process was performed, and the extracted solutions were stored in the refrigerator for 24 hours. In the third approach, 200 g of the mashed materials of sample B3 and sample C3 was separately put into a beaker containing distilled water as a solvent, then the beaker was transferred into a hot bath and agitated at 60°C for 6 hours. After filtration process, the prepared solutions were put into the oven at 50°C until they became a paste. Later, 0.1 g of the paste was diluted using 10 ml of distilled water. Finally, a Soxhlet extractor was used for the extraction of dyes with ethanol

solvent. The same procedure of preparation, filtration and extraction were performed for the other samples.

To extract the dye from walnut leaves, 150 g of its powder was immersed in 500 ml of ethanol and left for 24 hours in a hot bath at 60°C. The extracted samples are shown in Figure 1.

Figure 1: Photo of the extracted dyes.

Table 1: Samples labelling and their extraction description.

Sample	Description
B1	50g Beetroot +100 ml Distilled water
B2	50g Beetroot + 100 ml Ethanol:HCl (0.1) (99:1)
B3	0.1 g Beetroot paste + 10 ml Distilled water (dilution)
B4	0.1 g Beetroot paste + 10 ml Ethanol (dilution) soxhlat method
C1	50 g Cabbage + 100 ml Distilled water
C2	50 g Cabbage + 100 ml Ethanol:HCl (0.1) (99:1)
C3	0.1 g Cabbage paste + 10 ml Distilled water (dilution)
C4	0.1 g Cabbage paste + 10 ml Ethanol (dilution) soxhlat method
E	Sample B1: Sample C1 (1:1)
F	Sample B2: Sample C2 (1:1)
W	150 g Walnut + 500 ml Ethanol
H	100 g Henna + 200 ml Ethanol

Optical absorption studies of the extracted dyes were carried out by using a double beam UV–Vis spectrophotometer (Perkin Elmer, Lambda 25) at room temperature. The extracted samples were inserted into the cuvette for UV-Vis measurement and their absorbance were recorded in the wavelength range from 200 to 1100 nm. The absorption response of the samples was utilized to determine the optical band gap, optical transition types, optical dispersion parameters, optical dielectric constant, loss factor, and optical conductivity.

3. Results and discussion

Figure 2(a-d) shows the absorption coefficient of the dyes in the wavelength range from 200 to 1100 nm which was calculated from the absorbance data using [21]:

$$\alpha = \frac{2.303A}{t} \dots\dots\dots (1)$$

Where t is the thickness of the cuvette (q1 cm) and A is the absorbance. For the beetroot extract, the absorption spectrum indicated the presence of betalain chromophore [10], as showed multiple peaks of absorption around 475, 525 and 575 nm due to the presence of mixed pigments. These pigments are yellow betaxanthins (indicaxanthin), red-purple betacyanin (betanin) and betanidin respectively [22]. It was reported that the batlaian dyes can be affected by temperature and pH [23-25]. Figure 2(a) shows the absorption coefficient spectrum of beetroot dye which was extracted by different methods. Results showed that all samples are strongly absorbent in the ultraviolet region, which can be ascribed to the presence of red-violate betalain group [24]. However, different absorption peaks in the visible region have been observed. Sample B2 showed absorption peaks at 486 and 549 nm, which suggested that the extracts contain both betaxanthins and betanidin, and one peak in B3 sample was observed in the visible region at 417 nm, due to the presence of indicaxanthin. However, no peaks were observed in the visible region of samples B1 and B4.

The absorption spectra of the solutions from red cabbage are shown in Figure 2(b). One can notice that all the samples show a strong absorption in the UV region, which is well matched to the absorption characteristic of anthocyanin dye. This strong absorption in the UV region is due to the conjugated aromatic phenolic double bonds in anthocyanin [26]. Sample C1 and C2 have almost the same absorption peaks around 560 nm, indicating the presence of anthocyanidin [27]. The peaks between 360 nm to 470 nm are belong to a non-acylated pigment [28], which are observed in all samples except C2. Figure 2(c) shows the absorption spectrum of a mixed dyes of B1 with C1 and B2 with C2 by 1:1 (V/V) ratio. Interestingly, both samples presented a broad absorption band spanning along the UV and visible regions.

Figure 2-d shows the UV-vis absorption coefficient for both extracted dyes of Walnut leave and Henna. It can be seen that both dyes exhibited similar spectrum due to their comparable chemical structures. The broad absorption in UV region is referred to the presence of intermolecular hydrogen bonds from benzoquinone and electron delocalization of benzoquinone converts a quinone ring. In addition, the UV absorption points the existence of naphthoquinone in the form of hydrogen bonds with carbonyl [29]. However, the absorption in visible region (blue and red light) are corresponding to the mixture of chlorophyll [30], these peaks were also observed for both extracted dyes.

Figure 2: Absorption coefficient spectra of the dyes extracted from (a) Beetroot, (b) Red Cabbage, (c) mixture of beetroot and red cabbage, and (d) Walnuts leaves and Henna.

The measurement of optical energy gap and the nature of electronic transitions in the organic dyes are very crucial for viable optoelectronics applications. The absorption spectra can be used to determine the type of electronic transitions and optical bandgap. To various extends, Tauc's equation was used by the researchers to deduce the optical band gap and the type of electronic transitions governing the absorption process. We have noticed that the use of Tauc's equation to assign the nature of absorption transitions is highly efficient [31, 32] despite its seldom use by the researchers [33-38]. The Tauc's equation can be represented as follows [39]:

$$\alpha h\nu = \alpha_o (h\nu - E_g)^s \dots \dots \dots (2)$$

where α_o is an energy-independent constant, E_g is the energy gap, and the value of s determine the type and nature of transition [40] $s = \frac{1}{2}$ for direct allowed transition, $\frac{3}{2}$ for direct forbidden transitions, respectively, and this exponent value equals to 2 for the indirect allowed transitions

and 3 is for indirect forbidden transitions, respectively. One can identify the type of transition before estimating the energy gap by taking natural logarithm and derivation of equation 2 to yield:

$$\frac{d \ln(\alpha h\nu)}{d(h\nu)} = \frac{s}{h\nu - E_g} \dots \dots \dots (3)$$

Figure 3 (a-d) shows the plot of $\frac{d \ln(\alpha h\nu)}{d(h\nu)}$ versus $h\nu$ for all samples and the approximate value of $h\nu = E_g$ was taken at the peak of the curve. This is because the denominator of the right-hand side of Equation 3 can become minimum where photon energy is approximately equals to the energy gap. Therefore, the estimate value of E_g was utilized for plotting $\ln(\alpha h\nu)$ versus $\ln(h\nu - E_g)$ and the values of s were determined from the slope of the curves. It was found $s = \frac{1}{2}$, which indicated the presence of a direct allowed transition between the intermolecular energy bands of the dyes. Afterwards, the precise value of the energy gap was determined by Tauc's equation through plotting $(\alpha h\nu)^2$ as a function of $(h\nu)$ and taking the extrapolation of linear portion at $(\alpha h\nu)^2 = 0$. The energy gap location of the samples is depicted in Figure 4 (a-d) and the determined values of E_g are shown in Table 2. Results showed that energy gap for each dyes extracted from beetroot and red cabbage are different because they are pH and temperature sensitive. Noticeably, the dyes extracted from henna and walnut processed a very close value of energy gap due to similarity in their structure.

Figure 3: plot of $d\ln(\alpha h\nu)/d(h\nu)$ versus $h\nu$ for the dyes extracted from (a) Beetroot, (b) Red Cabbage, (c) mixture of beetroot and red cabbage, and (d) Walnuts leaves and Henna

Figure 4: plot of $(ah\nu)^2$ versus E for the dyes extracted from (a) Beetroot, (b) Red Cabbage, (c) mixture of beetroot and red cabbage, and (d) Walnuts leaves and Henna.

Following the absorption coefficient of the samples, refractive index and extinction coefficient are other two optical parameters describe the materials performance. Refractive index tells us how the optical radiation propagates through a medium and it shows that to what extent the speed of light is slowing down inside the medium compared to the speed of light in vacuum. The refractive index is a complex variable, and its imaginary part represents the extinction coefficient, which describes the amount of energy loss in the medium. Both refractive index (n) and extinction coefficient (k) were calculated from the absorbance data using the following equations [21]:

$$n = \frac{-2(R + 1) \pm \sqrt{4k^2R^2 + 16R - 4k^2}}{2(R - 1)} \dots \dots \dots (4)$$

$$k = \frac{\alpha\lambda}{4\pi} \dots \dots \dots (5)$$

Where R is the reflectance and α is the absorption coefficient. They were calculated using the following equations: $R = 1 - T - A$, where A is absorbance and T is transmittance, estimated from $T = 10^{-A}$ and $\alpha = \frac{2.303A}{t}$, where t is the thickness of the cuvette. The refractive index of the samples as a function of wavelength in the range of 190 to 1100 nm is shown in Figure 5 (a-d). The variation of refractive index for each sample with wavelength is related to the polarization of the molecules due to the interaction with electromagnetic wave, where the wide dispersion regions describe the polar nature of the samples [33]. At high wavelength, the refractive index reached a plateau region at which the extrapolation of the curve to y-axis gives the static value of

refractive index, as shown in Table 2. It was observed from Table 2 that the value of n is changed with respect to the method of extraction and the solvent used for extraction. The red cabbage dyes experienced a high value of refractive index (C3), while henna has the lowest value of n . Furthermore, Figure 5 (e-h) shows the extinction coefficient (k) as a function of wavelength for all the dye samples. Based on equation 5, it can be seen that the variation in k is almost similar to the corresponding absorption coefficient, as shown in Figure 2 (a-d). The variation of (k) represent the loss of incident electromagnetic wave because of scattering and absorption per unit thickness in the medium[33]. For most of the studies dyes, the maximum values of (k) was observed to be in the UV region.

Figure 5: Refractive index and extinction coefficient spectra for the dyes extracted from (a & e) Beetroot, (b & f) Red Cabbage, (c & g) mixture of beetroot and red cabbage, and (d & h) Walnuts leaves and Henna.

Figure 6 shows the variation of optical dielectric constant over the wavelength of UV to IR region. The response of electrons in the material upon the incident of light is physically represented by optical dielectric constant (ϵ), which is a frequency dependent parameter. Since dielectric constant is a complex variable, it has real and imaginary parts and can be mathematically represented by [34]:

$$\epsilon = \epsilon_1 + i\epsilon_2 \dots \dots \dots (6)$$

Where ϵ_1 represents the real part and ϵ_2 represents the imaginary part of dielectric constant. The real dielectric constant is further represented by equation 7 and it is a measure of to what extent

the material can be polarized due to the influence of electromagnetic field, while the imaginary part shows the optical loss and is described by equation 8.

Table 2: The optoelectronics parameters estimated for the extracted dyes.

Samples	n_{∞}	ϵ_r	$\sigma_r \left(\frac{S}{cm} \right) \times 10^{-3}$	$E_g (eV)$
B1	1.95	3.87	0.53	2.75
B2	1.65	2.73	0.311	3.72
B3	4.24	18.02	6.1	2.36
B4	3.36	11.52	3.5	3.18
C1	1.54	2.42	0.224	3.18
C2	1.77	3.21	0.448	2.99
C3	6.08	37.18	13.92	2.25
C4	5.41	29.07	10.63	2.67
E	2.25	5	1.05	2.96
F	1.4	2.01	0.128	3.02
W	1.9	3.65	0.57	1.776
H	1.19	1.5	0.039	1.775

The optical loss takes place because of the time lag of $\pi/2$ between the electric field of the incident photon and the component of electronic or atomic polarization in the optical frequency range [41].

$$\epsilon_1 = n^2 - k^2 \dots \dots \dots (7)$$

$$\epsilon_2 = 2nk \dots \dots \dots (8)$$

$$\tan \delta = \frac{\epsilon_2}{\epsilon_1} \dots \dots \dots (9)$$

Noteworthy, the real part of dielectric constant resembles the refractive index due to small value of k . However, the imaginary part is mainly dependent on the absorption coefficient (see Equation 7 and 8). The variation in real dielectric constant for all the extracted dyes is shown in Figure 6(a-d). It was seen that the peak observed in the UV region is belong to B2 sample, while other samples

show their characteristic peaks at different wavelengths. It has been reported that the higher value of real dielectric constant indicates the presence of interaction between electron and incident photon in the extracted dyes[35]. It worth to note that the mixed dye of anthocyanin and betalin dye has a broad peak ranging from 400 to 600 nm (Figure 6-c), whereas walnut leaves and henna has a sharp peak around 500 and 700 nm. The values of real dielectric constants are listed in Table 2, which were determined from the intersection of extrapolation of the plateau region to y-axis. Dyes extracted from red cabbage, especially C3 presented the maximum value of ϵ_1 , while dyes extracted from henna has lowest value. One can notice that the value of ϵ_1 for the extracted dyes with water solvent is greater compared to those extracted with ethanol and HCl solvent. This might be due to the polar nature of water. In addition to dielectric constant, Figure 6 (e-h) shows the imaginary dielectric constant of all the extracted dyes as a function of wavelength. It can be concluded that the variation of imaginary part of dielectric constant is related to the absorption coefficient of the dyes and their mixtures, as shown in Figure 6-g with a broad peak across UV and visible region. Furthermore, the dissipation factor ($\tan \delta$) was calculated from equation 8 by dividing the imaginary part to the real part of dielectric function[31]. The spectra of dissipation factors for all samples were presented in Figure 7(a-d), wherein each dye presented different characteristic peaks regardless of whether extracted from beetroot or red cabbage has different peak value and it's depend the type due to the effect of solvents and extraction methods. The anthocyanin dye, which was extracted from red cabbage with distilled water as a solvent (C3), has maximum peak value in the range of 400 to 500 nm compared to that of betalain dye extracted from beetroot with the same solvent and extraction method. This is ascribed to the rate of absorption of anthocyanin which is greater than that of the betalain dye, whereas their mixture

showed a broad peak in the UV region. However, walnut leave and henna showed a broad peak expanded to 500 nm and a sharp peak near 700 nm.

Figure 6: Real and Imaginary optical dielectric constant spectra for all samples of dyes extracted from (a & e) Beetroot, (b & f) Red Cabbage, (c & g) mixture of beetroot and red cabbage, and (d & h) Walnuts leaves and Henna.

Figure 7: dielectric lost tangent (dissipation factor) spectra for all samples of dyes extracted from (a) Beetroot, (b) Red Cabbage, (c) mixture of beetroot and red cabbage, and (d) Walnuts leaves and Henna.

To determine the electronic characteristics of a material, optical conductivity is a powerful tool to be used, which describes the response of electron to the absorbed photon. Since the optical conductivity is a complex variable, it is derived from optical dielectric constant[36]:

$$\sigma^* = \sigma_r + i\sigma_i \dots \dots \dots (10)$$

$$\sigma_r = \omega \varepsilon_2 \varepsilon_0 \dots \dots \dots (11)$$

$$\sigma_i = \omega \varepsilon_1 \varepsilon_0 \dots \dots \dots (12)$$

Where σ_r and σ_i are the real and imaginary part of complex optical conductivity respectively, ω is the angular frequency, and ε_0 is the permittivity of free space. One can conclude from equation 10 that the real optical conductivity is related to the imaginary optical dielectric constant and hence depends on the absorption coefficient. Thus, increasing the magnitude of real optical conductivity is directly related to the transport response of electrons which are excited by the absorbed photon energy[42]. In contrast, the imaginary optical conductivity is related to the real optical dielectric constant hence represents the interaction between electromagnetic wave and electrons in the form of polarization. The spectra of real and imaginary optical conductivity as shown in Figure 8 and the value of real optical conductivity are listed in Table 2. It was concluded that anthocyanin dye (C3) has the maximum value, which is $\sigma_r = 13.92 \times 10^{-3} S/cm$, while the extracted dye from henna has the lowest value of $\sigma_r = 0.039 \times 10^{-3} S/cm$. In addition, it was seen from the spectra of real conductivity that both betalain and anthocyanin dyes extracted by distilled water as a solvent (B3&C3) have a broad peak over UV and visible region. The henna and walnut leaves have a broad peak starting from UV region until 500 nm in visible region and a sharp peak at near IR region. Figure 8 (e-h) shows the imaginary spectra of the optical conductivity and they are similar to real optical dielectric constant spectra. Noticeably, the combination of two dyes together

has led to a broadened photo-absorption in the UV region, indicating that molecular interactions between their moieties are mostly taken place in the high energy levels.

Figure 8: Real and Imaginary optical conductivity spectra for all samples of dyes extracted from (a & e) Beetroot, (b & f) Red Cabbage, (c & g) mixture of beetroot and red cabbage, and (d & h) Walnuts leaves and Henna.

4. Conclusions

Natural dyes were successfully extracted from beetroot, red cabbage, walnut and henna using a facile approach and were broadly characterized. A broad spectrum of optoelectronics parameters of the samples such as absorption coefficient, optical band gap, dispersion parameters, and optical conductivity was investigated. The UV results showed that dyes extracted from beetroot and red cabbage are btalain and anthocyanin derivatives, respectively. However, dyes in henna and walnut have almost similar spectra due to their similar aromatic conjugation double bonds. The cabbage dye showed maximum value of refractive index, real optical dielectric constant, real optical conductivity, while henna showed the minimum value for these parameters. By taking derivatives of Tauc's equation, the type of electronic transitions was estimated to be direct allowed transition for all the dyes.

Funding statement

This work did not receive a financial support.

Conflicts of interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

Author contributions

Methodology: Peshawa O. Amin; Formal analysis and investigation: Fahmi F. Muhammadsharif, Salah Raza Saeed, and Khaulah Sulaiman; Writing - original draft preparation: Peshawa O. Amin; Writing - review and editing: Fahmi F. Muhammadsharif, Salah Raza Saeed, and Khaulah Sulaiman; Supervision: Fahmi F. Muhammadsharif, Salah Raza Saeed.

Availability of data and material

The data and material are available within the manuscript.

Compliance with ethical standards

Not applicable

Consent to participate

Not applicable

Consent for Publication

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Acknowledgement

The authors gratefully acknowledge the Ministry of Higher Education and Scientific Research-Kurdistan Regional Government-Charmo University—College of Medical and Applied Science —Department of Applied Physics for the financial support. The authors would like to express their deepest appreciation to University of Sulaimani—College of Science—Department of Physics, and Komar University for Science and Technology for their technical support.

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