Physical Properties Investigation on Sunscreens with Colloidal Gold and Moringa oleifera Extract

by Eva Monica

Physical Properties Investigation on Sunscreens with Colloidal Gold and *Moringa oleifera* Extract

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ABSTRACT

Botanicals were known as active ingredients in sunscreens due to their dermal protecting. Gold in colloidal form has a completely unique property for the reason that it may be used as photoaging material. The exposure of UV radiation to skin can cause erythema and skin cancer. The aim of this study was to determine the physical properties of the sunscreens made from Au/ZnO and antioxidant from *Moringa oleifera* extract such as the sun protection factor (SPF), pH, spreadability, and adhesion. The Moringa oleifera were extracted by maceration. Maceration with 70% ethanol of dried leaves exhibited DPPH scavenging activity (EC_{50} 235.01 µg/mL). Diffuse reflectance UV-Vis spectra of ZnO and Au/ZnO revealed absorption at λex of 385 nm which were equivalent to a bandgap energy of 3.22 eV. Au/ZnO posed a localized state at λex of 385 (3.65 eV). The sunscreens with formulations F1, F2, F3, and F4 showed high SPF number of 20.1479, 21.0008, 22.3872, and18.4631. Kruskal-Wallis test showed significant differences between sunscreen formulations.

Keywords: antioxidant, *Moringa oleifera*, Au/ZnO, physical properties of sunscreens

INTRODUCTION

Indonesia is a tropical country with high exposure to sunlight. Indonesian people skin who live in the equatorial region need more protections than other regions. Sunlight spectrum such as ultraviolet light called UVA and UVB has a bad impact on the skin. UVB radiation for example, can cause skin cancer, while UVA radiation can reduce the elasticity of the epidermis layer of the skin so it can cause photoaging. Both of these UV rays work synergistically so the protection is needed to prevent the adverse effects on the skin due to UVA and UVB radiation [1]. Sunscreen is used to prevent skin damage. Inorganic sunscreen such as zinc oxide (ZnO) has been used generally as a sunscreen because of their ability as a barrier to UVA radiation and its protection can be increased by the combination with titanium dioxide (TiO_2) [2]. However, the addition of metal oxide concentrations to increase the effectiveness of sunscreens can cause the product is difficult to spread on the skin and causes the pores and the skin to become blurry white when the sunscreen is applied [3].

Inorganic sunscreens physically work via blocking a few or all of the UV radiation contemplated or scattered far from the surface of the skin. While organic sunscreens work by absorbing a few or all of the UV radiation with the same purpose so that UV radiation does not reach the pores and the skin surface [4]. In this study, the sunscreen was made from zinc oxide and *Moringa oleifera* as an organic antioxidant. *Moringa oleifera* leaves that have been

analyzed in vivo can have different effects on oxidative free radical SOD, CAT, Glutathione-S-Transferase enzymes. *Moringa oleifera* extract can be a natural antioxidant agent because of its health benefits [5]. Organic and inorganic ingredients are formulated and optimized along with other additives needed to make a stable o/w (oil in water) cream sunscreen. Sunscreen products are expected to protect the skin from exposure to UVA and UVB rays, water repellent for use while exercising and on the beach, and filters or filter combinations must be stable to light.

EXPERIMENT
Chemicals and Instrumentation

All reagents, i.e benzoic acid (pharmaceutical grade), stearic acid (pharmaceutical grade), cetyl alcohol (pharmaceutical grade), dimethicon (pharmaceutical grade), 2,2-diphenyl-1-picrylhydrazyl (Merck), ethanol (Merck), glycerin (pharmaceutical grade), isopropyl miristate (pharmaceutical grade), methanol (Merck), liquid paraffin (pharmaceutical grade), perfume, propylene glycol (pharmaceutical grade), spermaceti (pharmaceutical grade), tetrachloro-auric(III) acid trihydrate (Sigma Aldrich, 99.5%), triethanolamine (pharmaceutical grade), tween 80 (pharmaceutical grade), and zinc oxide (Merck) were of analytical grade and used without further purification. Meanwhile, the instrument used for analysis was DRUV-VIS spectrophotometer (JASCO V-730 UV-Vis spectrophotometer).

Extraction Procedure

The dried powdered *Moringa oleifera* leaves (2 kg) were macerated with 1 L of ethanol 70% in a container. The maceration was carried out for 72 h (25 °C) with occasional shaking. The extract was separately filtered. The filtrate was dried using a rotary vacuum evaporator. The crude extract was weighed.

Radical Scavenging Activity using DPPH

The free radical scavenging activity of *Moringa oleifera* extract was determined using DPPH radical scavenging method [6]. A series of volume of exctract (0.1; 0.2; 0.3; 0.5; and 0.7 mL) was added to 3.8 mL of DPPH in methanol (19.6 mg/L). After incubation at 37 °C for 20 min, the absorbance of each solution was determined (λ=517 nm) using UV-vis spectrophotometer.

$$\%RSA = [\frac{A_T - A_E}{A_T}] \times 100$$

where A_T is the absorbance of the control reaction and A_E is the absorbance of the sample. The EC_{50} was calculated from the curve of percent scavenging plotted against the concentration.

Optical Properties Measurement of ZnO and Au/ZnO

Percentage of reflectance (%R) was taken using a diffuse reflectance spectrophotometer (DRS - JASCO). Because a direct-allowed sample transition was used in this experiment, then n equals 1/2 was selected in the measurement. The obtained diffuse reflectance spectra were converted to a Kubelka-Munk function [7]. A vertical axis became converted to amount $F(R\infty)$ value, which turned into proportional to the absorption coefficient. The α in the Tauc equation [8] was substituted with $F(R\infty)$. The value of $(h\upsilon F(R\infty))^2$ was plotted against $h\upsilon$

(described in eV) according to a Kubelka-Munk equation. A tangent line was drawn to the point of inflection of the curve and the hυ. The bandgap energy (Eg) value was derived from a point of intersection between the tangent line and the horizontal line of hυ. Barium sulfate became used as a reflectance coating in the DRS measurement.

Preparation of Emulsion of the Sunscreens

The lipid phase was composed of cetyl alcohol, spermaceti, and stearic acid. It was melted up to 85 °C on the water bath. When the temperature was the same, the aqueous phase was added dropwise to the lipid phase while stirring vigorously. The composition of the emulsion was mention in Table 1.

Table 1. The compositions of the sunscreens

No	Ingredients	Amount of each ingredient (%)				
		F1	F2	F3	F4	F5
1	Moringa extract	0	1	0.5	0	0.5
2	ZnO	0	0	0	1	0.5
3	Au/ZnO	1	0	0.5	0	0
4	Aquadest	31.8	31.8	31.8	31.8	31.8
5	Benzoic acid	0.1	0.1	0.1	0.1	0.1
6	Stearic acid	5	5	5	5	5
7	Dimethicone	1	1	1	1	1
8	Glycerin	5	5	5	5	5
9	Isopropyl miristate	5	5	5	5	5
10	Liquid paraffin	5	5	5	5	5
11	Perfume	0.1	0.1	0.1	0.1	0.1
12	Propylene glycol	10	10	10	10	10
13	Spermaceti	2	2	2	2	2
14	Triethanolamine	5	5	5	5	5
15	Tween 80	5	5	5	5	5

Physical Evaluation of the Sunscreens
Sun protection factor (SPF) determination

The sun protection factor of the sunscreens was determined by measuring the transmittance after passing through a film using a diffuse reflectance spectrophotometer. A rapid and reliable in vitro approach of calculating the SPF is to screen the transmittance of the product between 290-320 nm with an interval of 1 nm. The SPF can be calculated using Mansur equation [9]:

$$SPF = CF \times \sum_{290}^{320} EE\,(\lambda) \times I\,(\lambda) \times Abs\,(\lambda)$$

Spread ability measurement

The spreading diameter of 0.5 g of sample between two horizontal glass plates (10 cm x 20 cm) was measured after one minute. The standard weight applied to the upper plate was 0, 50, 100, 150, 200, 250 and 500 g. Each formula was tested three times [10].

pH values measurement

A 0.25 g of each formulation was dispersed in 10 mL of distilled water and pH was determined using universal pH indicator.

Adhesion measurement

A 0.1 g of sample between two horizontal glass plates (2 cm x 2 cm). A weight of 1 kg was given upper the glass plates for 5 min. Then, the glass plates were tied to the rope with the standard weight of 40 g on both sides. The time when the glass plates separated were measured using stopwatch.

Statistical analysis

The results were reported as mean ± standard deviation. The average contents of adhesion, dispersion, and SPF of sunscreens were statistically investigated using Statistic R 3.5.2. A statistical probability (p value < 0.05) indicated a statistically significant.

RESULT AND DISCUSSION

DPPH Radical Scavenging Activity

The antioxidant activity of *Moringa oleifera* was measured using UV-Vis spectrophotometer. The EC_{50} value is 235.01 μg/mL. This extract showed weak DPPH scavenging activity. Meanwhile, Vongsak et al. [6] reported the EC_{50} value of *Moringa oleifera* extract of 62.94 μg/mL which shows high activity against free radicals. The difference in the EC_{50} value is due to crude and impure extract where there are still other antagonistic compounds that can reduce antioxidant activity compared to pure *Moringa oleifera* extract.
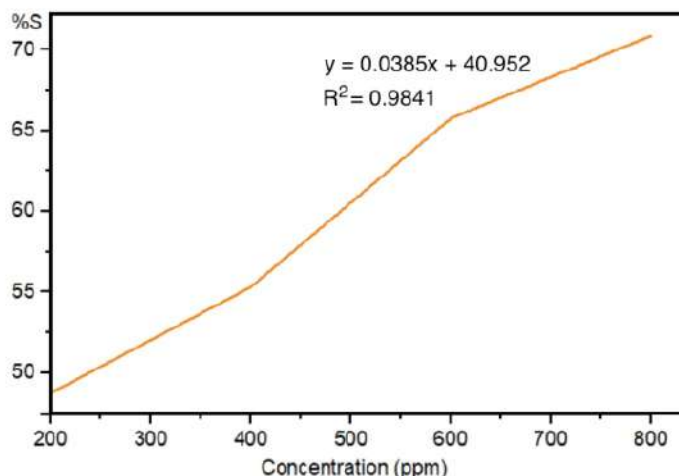

Figure 1. Antioxidant activity of *Moringa oleifera* extract.

Optical Properties of ZnO and Au/ZnO

Optical absorption of ZnO and Au/ZnO were investigated using UV-Vis spectroscopy in diffuse reflectance spectra as shown in Figure 2. The percentage of reflectance (%R) was used to identify the bandgap energy of the ZnO and Au/ZnO using a Kubelka-Munk equation

[7] and Tauc Plot [8]. The main absorption bands of ZnO and Au/ZnO were located in the UV and visible regions. Gold was known to prevent premature aging. Based on the results of the characterization of PXRD, Au which is fed into ZnO has the crystallite size of 68 nm. Zinc oxide has a band gap energy of 3.22 eV which corresponds to absorption at the UV region (385 nm). Addition of Au to zinc oxide-based sunscreen was known to produce a localized state at a wavelength of 340 nm (3.65 eV).
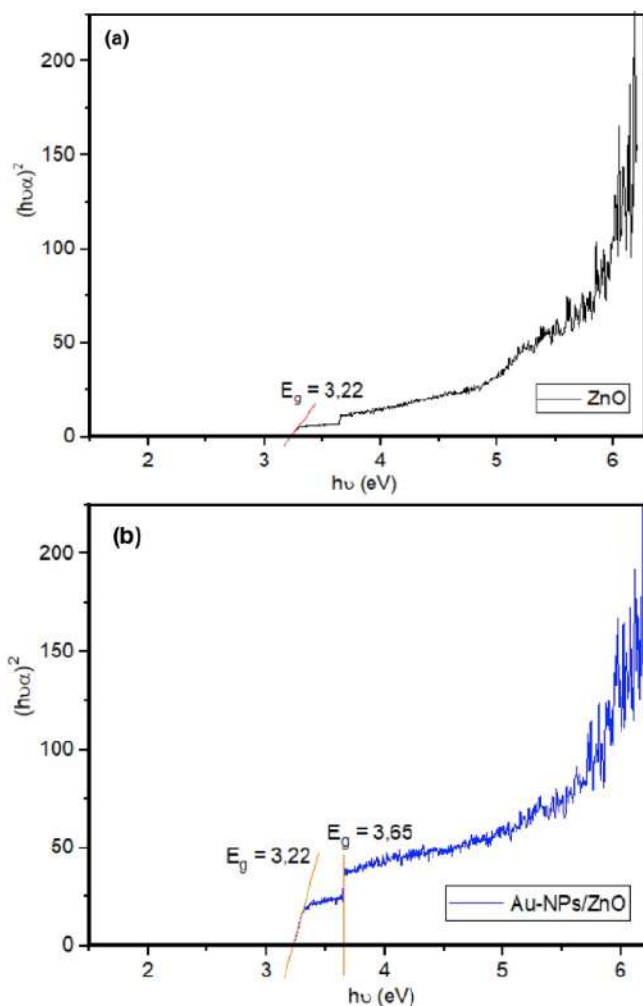

Figure 2. Tauc Plot of (a) ZnO, and (b) Au/ZnO

Physical Evaluation of the Sunscreens

Physical characterization of sunscreens including SPF, adhesion, and spread ability were statistically measured. Adhesion shows how long the sunscreens will attach to the skin. The active ingredients give a great effect as long the sunscreens adhesion. The results of the sunscreen adhesion showed the highest adhesion was F3 (Table 2). The similar consistency of

sunscreens was shown in F2 and F5. The addition of *Moringa oleifera* extract made the emulsion more viscous. While pH value was 7 for each formulation.

Table 2. Physical properties of the sunscreens

Formula	Average value of SPF	Average value of adhesion
F1	20.1479	4.46 s
F2	4.5636	32.75 s
F3	21.0008	104.33 s
F4	22.3872	5.2 s
F5	18.4631	37.33 s

Table 3. Kruskal-Wallis Test of the Sunscreens

Response	*p* value	Conclusion
SPF	0.01	Significant
Adhesion	0.02	Significant

It can be seen that, there are significant differences between sunscreen formulations. This can be proven through Kruskal-Wallis test (p value <0.05) (Table 3). In addition to that, the spread ability is the capacity of a cream or gel to spread on the skin surface. The diameter of each formulation of sunscreens were measured to calculate the spreading vicinity (Figure 3). The values related to how each formulation spread on the skin surface by a small amount of a cream. The results showed F4 and F2 had a greater spread ability that the other formulations.
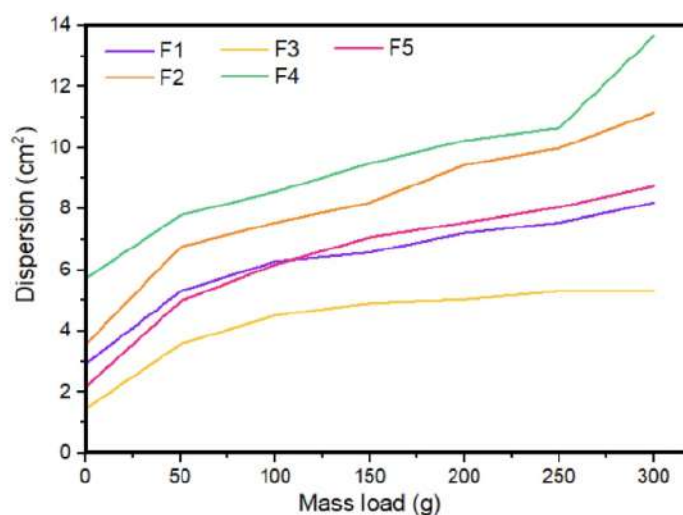

Figure 3. Spread ability values of the sunscreens.

CONCLUSION

The sunscreens were formulated with ZnO, Au/ZnO, and *Moringa oleifera* extract. The physical properties of the sunscreen have been investigated. The highest SPF values were obtained in F4, F3, and F1, the highest adhesion was in F3, F5, and F2. While the best spread ability were obtained in F4 and F2.

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CONFLICT OF INTEREST

Authors declare no competing interests.

REFERENCES

[1]   Balakrishnan KP, Narayanaswamy N., *Int. J. Res. Cosmet Sci.*, **2011**, 1(1), 1–12.
[2]   Gutiérrez-hernández JM, Escalante A, Murillo-vázquez RN, Delgado E, Javier F, Toríz G., *J. Photochem. Photobiol. B*, **2016**,163, 156–61.
[3]   Lusignan PER. United States Patent. US Pat Doc. 1998;54–5.
[4]   Bhunia P, Bengal W, Pramanik A. Patent Application Publication. 2008; (19).
[5]   Jaiswal D, Rai P. K, Mehta S, Chatterji S, Shukla S, Rai D. K., *Asian Pac. J. Trop. Med.*, **2013**, 6(6), 426–32.
[6]   Vongsak B, Sithisarn P, Mangmool S., *Ind. Crop. Prod.*, **2013**, 44, 566–71.
[7]   Viger, R., Ramsey, S., Mayo, T. & Lambrakos, S.G., *J. Electromagnet. Wave Appl.*, **2019**, 33(9), 1163-1171.
[8]   Evstropiev, S.K., Soshnikov, I.P., Kolobkova, E.V., Evstroyev, K.S., Nikonorov, N.V., Khrebtov, A.I., Dukelskii, K.V., Kotlyar, K.P., Oreshkina, K.V. & Nashekin, A.V., *Opt. Mater.*, **2018**, 82, 81-87.
[9]   Malsawmtluangi C, Nath DK, Jamatia I, Zarzoliana E, Pachuau L., *J. Appl. Pharm. Sci.* **2013**, 3(9), 150–1.
[10] Chen, M.X., Alexander, K.S. & Baki, G., *J. Pharm.* (Cairo), **2016**, 2016: 5754349.

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antiferromagnetic Co3O4 with prism like-anchored octahedron morphology: A facile green synthesis using Manihot esculenta Crantz extract", Science and Technology of Materials, 2018
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