

RESEARCH ARTICLE

Ultra Sunscreen and Antiaging Potential of Arabica Coffee (*Coffea arabica L.*) Extract with Oxybenzone and Octyl Methoxycinnamate

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ABSTRACT

Arabica coffee has been known for its high antioxidant polyphenol compounds that can reduce free radicals and reactive oxygen species (ROS). Here, we explore the sunscreen potential and antiaging effect of arabica coffee cream extract combined with UV filters such as oxybenzone and octyl methoxycinnamate. The as-prepared combination cream extract showed good physical stability, including homogeneity, organoleptic, pH, and spreadability. The phytochemical contents by GC-MS showed the presence of stigmaterol, beta-tocopherol, fatty acid, creatindial, and methyl icosanoate. The *in-vitro* test of the as-prepared combination cream at higher concentrations of 5 and 10% showed ultra-protection with SPF values of 17.3, and 22.3, respectively. The antioxidant activity with the DPPH method showed an outstanding value of IC_{50} about 353.72 mcg/mL which was better than vitamin C. The experimental animal design was the post-test-only control group design. The *in-vivo* antiaging tests were examined by anti-irritation, anti-wrinkle effect, and malondialdehyde analyses among 25 wistar rats. Application of combination cream showed no irritation and wrinkles on the soles of the feet of Wistar rats. At cellular levels, combination cream-induced MDA levels were 0.152 $\mu\text{mol/l}$, which was lower than vitamin E (0.160 $\mu\text{mol/l}$) and NA-CMC (0.186 $\mu\text{mol/l}$). This study showed the potential of arabica coffee cream combination extract with oxybenzone and octyl methoxycinnamate for ultra sunscreen and antiaging applications.

Keywords: Sunscreen, Antiaging, Arabica coffee, Oxybenzone, Octyl methoxycinnamate.

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INTRODUCTION

Indonesia is a tropical country that gets longer and higher intensity of sun exposure compared to other countries. Although the sun is a good vitamin D source, excess sun exposure can cause adverse effects¹ such as erythema and skin damage. Excessive UV exposure also causes sunburn, pigmentation, and premature aging of the skin.² Continuous exposure to UV light will deteriorate skin structure and malfunction, accelerating the skin's aging process. This process is called photoaging. Signs of aging can be seen from the presence of fine wrinkles, reduced skin firmness, dry skin, dull and rough skin, and thinning of the skin where this condition is now starting to be felt at an early age and affects skin aesthetics.¹

To prevent skin damage, additional protection is needed, namely by using cosmetics that have a sunscreen effect. Sunscreen is a preparation that contains compounds that can absorb, scatter, or reflect sunlight that hits the skin so that it can protect the function and structure of human skin from damage

caused by sunlight.³ The FDA (Food Drug Administration) requires sunscreen products to be able to provide a "broad-spectrum" effect. The broad-spectrum effect means that the sunscreen must be able to protect the skin from both UV-A and UV-B rays. One of the efforts to obtain a broad-spectrum effect in sunscreen preparations is to combine UV-A and UV-B filters. The combination of oxybenzone (Benzophenone-3) and octyl methoxycinnamate (OMC) has increased sunscreen activity because the mixture of the two provides a good sun protection factor.⁴

Antioxidants are electron-giving compounds that can reduce free radicals and reactive oxygen species (ROS). Antioxidants in sunscreen preparations can increase photoprotective activity and prevent various diseases caused by ultraviolet radiation. Several active antioxidant compounds such as flavonoids, tannins, anthraquinones, cinnamates, vitamin C, vitamin E, and beta-carotene have been reported to have the ability to protect against the light.⁵ Antioxidants in topical form applied to the skin surface can reduce the effect

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of ROS in causing skin damage due to UV exposure. Recently, the use of antioxidants has increased orally and topically to prevent and treat skin aging. Many skin care products use natural ingredients that contain antioxidants found in fruits, leaves, flowers, roots, and other part of the plants.⁶

The coffee plant belongs to the Rubiaceae family and contains high antioxidant polyphenolic compounds derived from phenolic acids such as caffeine, chlorogenic acid, coumarin, ferulic and sinapic acid.³ One of the polyphenolic compounds found in coffee in considerable quantities and believed to be the largest contributor to antioxidant activity is chlorogenic acid.⁷ Phenolic compounds, especially flavonoids found in robusta coffee beans, have the potential as sunscreens due to the presence of chromophore groups that can absorb UV-A or UV-B rays thereby reducing their intensity on the skin.⁸ Chlorogenic acid can protect coffee growth from microorganisms, insects, and UV radiation while the benefits of chlorogenic acid for human health are as an antioxidant, antiviral, hepatoprotective, and play a role in antispasmodic activities. According to the research of Asri Wulandari *et al.*, the tannin content in coffee fruit skin is effective as an antibacterial. In addition, the presence of polyphenolic compounds in coffee beans as much as 0.2% explains the potential of coffee as an antioxidant which is very important for facial skin health.⁹ Furthermore, Ajhar and Meilani, 2020 showed that the antioxidant activity of ethanol extract from arabica coffee beans was very strong with IC₅₀ 12.427 ppm and vitamin C with IC₅₀ 0.273 ppm.¹⁰ According to the results of research conducted by, a combination of sunscreen cream containing 10% corn cob extract and 15% robusta coffee beans showed the IC₅₀ value of 97.34 ppm, showing a strong antioxidant category of SPF value of 37 which indicated an ultra-protection level.¹¹ This result emphasizes that the cream can be developed into a commercialized antiaging cream.

This study aims to observe the effectiveness of arabic coffee extract with oxybenzone and octyl methoxycinnamate for sunscreen and antiaging applications. We assumed that adding and combining oxybenzone and octyl methoxycinnamate might improve the SPF values and reduce the wrinkles on the skin *in-vivo*.

METHODOLOGY

Materials

The tools used in this research are mortar, pestle, analytical balance (Ohaus), water bath, stirring rod, vaporizer cup, volumetric flask, dropper, measuring pipette, suction ball, Erlenmeyer, measuring cup, watch glass, object glass, pH meter (Eutech Instrument), UV-vis spectrophotometer (Shimadzu UV 1800), a, filter cloth, rotary evaporator, pH meter, shaver, gas chromatography-mass spectrometry (GC-MS). The animal experiment used white rats (*Rattus norvegicus*), male, wistar strain, 2 months old, with a body weight of 120 to 200 gm. Rat hair was shaved on the back until clean. To remove fine hair, the feed was used as a hair thresher.

Methods

The experimental research used a posttest-only control group design. This research includes sample preparation, simplistic characterization, extraction phytochemical test with GC-MS method, preparation of arabica coffee bean extract cream in combination with oxybenzone and octyl methoxycinnamate, preparation formulation, evaluation of cream preparation stability (homogeneity, organoleptic, pH, cream spreadability), sun protection factor (SPF) test, DPPH method antioxidant test with vitamin C comparison, anti-irritation test, antiaging effect test (wrinkles count), and malondialdehyde (MDA) test on white rats for 2 weeks.

The following formulation is applied to make cream and its combination (Table 1).

Data Analysis

All data in this study were analyzed with IBM SPSS version 26. Data were analyzed using the Shapiro-Wilk method to determine normality ($p > 0.05$) and a homogeneity test was also conducted. Then continue using the Way ANOVA method to determine the mean difference between groups. If there is a difference ($p < 0.05$), it is continued by using the Post Hoc Turkey HSD test to see the real difference between treatments. But if the data is not normally distributed, the Kruskal-Wallis test is used.

RESULTS AND DISCUSSION

Phytochemistry Analysis

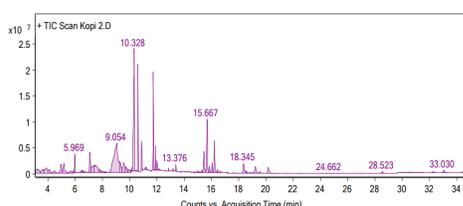
GC-MS analyzed the secondary metabolites in the extract because of the fast, efficient, sensitive, inexpensive analysis and high-resolution performance.¹² GC-MS spectra are shown in Figure 1.

GC-MS spectra have indicated nine metabolite compounds, including 2-methoxy-4-vinylphenol with RT 5,969 and 96,87, tetrahydroxy cyclohexane carboxylic acid with RT 9.054 and score 89.01, caffeine with RT 10.328 and score 99.13, methyl icosanoate with RT 13.376 and score 95.99, creatindial with RT 15.667 and score 77.44, 9,12-octadecadienoic acid (Z, Z)-, 2-hydroxy-1-(hydroxymethyl)ethyl ester with RT 18.345 and score 95.89, octadecanoic acid, 2-hydroxy-1-(hydroxymethyl) ethyl ester with RT 24.662 and score 71.18, beta-tocopherol with RT 28.523 and score 94.66, and stigmasterol with RT 33.030 and score 91.69.

We have found four dominant peaks that have potential bioactive content as antioxidants, namely 2-Methoxy-4-vinylpheno, caffeine, methyl icosanoate, and 9,12-octadecadienoic acid (Z, Z)-, 2-hydroxy-1-(hydroxymethyl)ethyl ester. According to research conducted by Rubab, *et al.*, 2020, *Brassica oleracea* contained the 2-Methoxy-4-vinylpheno compound which has the potential as an antimicrobial, antioxidant, anti-inflammatory, analgesic, and antitermination.¹³ Moreover, J.S.C. Vieira *et al.*, 2020 described that caffeine is a good antioxidant, capable of regenerating adenine products which are hydroxyl radical compounds that play a role in the formation of oxidative stress conditions.¹⁴

Table 1: Cream formulation

| Ingredients | Concentration | | | | | |
|------------------------|-----------------------------|---------------|---------------|---------------|---------------|---------------|
| | Formula 0 (Cream Bases) (%) | Formula 1 (%) | Formula 2 (%) | Formula 3 (%) | Formula 4 (%) | Formula 5 (%) |
| Coffee Bean Extract | - | - | 5 | 1 | 5 | 10 |
| Octyl Methoxycinnamate | - | 5 | - | 5 | 5 | 5 |
| Oxybenzone | - | 2 | - | 2 | 2 | 2 |
| 1,3 butylene glycol | 7 | 7 | 7 | 7 | 7 | 7 |
| Disodium Edetate | 0.05 | 0.05 | 0.05 | 0.05 | 0.05 | 0.05 |
| Triethanolamine | 1 | 1 | 1 | 1 | 1 | 1 |
| Alcohol | 3 | 3 | 3 | 3 | 3 | 3 |
| Stearic Acid | 3 | 3 | 3 | 3 | 3 | 3 |
| Glyceryl Monostearate | 3 | 3 | 3 | 3 | 3 | 3 |
| Sodium Metabisulfite | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 |

**Figure 1:** Gas chromatography spectra of coffee bean extract

Sun Protection Factor

Sun Protection Factor or SPF values were measured at 290 to 320 nm using a UV-vis spectrophotometer. The physical characteristics of cream preparation, as well as the SPF value of each formula, can be seen in Table 2

According to Baskara, *et al.*, 2020, the ideal pH of cream must be around 4.5 to 7.5 which is compatible with the pH of the skin; otherwise, it will cause skin irritation. In this research, the pH value of cream after one cycle has slightly decreased because it is influenced by temperature. However, the pH measured from all cream formulas is still within the ideal range of pH.¹⁵

Homogeneity testing is also carried out to determine whether all the ingredients used can be mixed well, namely the preparation must show a homogeneous composition and no coarse grains are visible. Homogeneity standards are indicated by the absence of coarse grains and an even color in the preparation.¹⁶ The results of the observation show that the six groups of cream preparations are physically homogeneous, this shows that the ingredients used in the cream preparation are perfectly mixed.

Organoleptic testing was carried out through visual observation of the preparation, namely observation of the cream's color, shape, and smell.¹⁷ The results showed that there are differences in aroma, color, shape, and spreadability of preparations between groups F0, F1, F2, F3, F4, and F5. Spreadability is part of psycho-rheology which can be used as a parameter of acceptability.¹⁸ Optimum spreadability will make it easier for the cream to spread when applied on the skin's surface without the need for great pressure. The wider the

spreading area produced by a cream, the better the spreading ability will be when applied.¹¹

The sunscreen protection against UV rays is correlated with the SPF value.¹⁹ The Food and Drug Administration (FDA) classifies the categories of sunscreen ability based on the SPF value, namely 2–4 (Minimal), 4–6 (Medium), 6–8 (Extra), 8–15 (Maximum), and >15 (Ultra).²⁰ Our study showed that the SPF value of the arabica coffee extract which is combined with oxybenzone and octyl methoxycinnamate has a maximum category of UV protection activity. At most, the ultra category of SPF was found at F4 and F5 at 17.3 and 22.3, respectively.

The statistical data analysis of SPF value was initially tested for normality using Shapiro Wilk, the data were normally distributed ($p > 0.05$) and homogeneous ($p > 0.05$). Then the analysis continued by using the ANOVA test, the results can be seen in the Table 3.

Based on the ANOVA similarity test, it is shown that the p -value was 1.00 ($p > 0.05$), which means that the average of each group of arabica coffee bean extract is equally effective in the preparation of a combination of oxybenzone and octyl methoxycinnamate sunscreen. This study's results align with Ajeng *et al.*, (2021) where the results show that Arabica coffee bean extract has the potential as an extract to be made as a sunscreen cream.²¹

Antioxidant Activity

Various concentrations of arabica coffee extract were examined by the DPPH antioxidant test as shown in Table 4.

The classification of a very strong antioxidant whereas the IC₅₀ value < 50 ppm, strong (50–100 ppm), medium (100–150 ppm), and weak (151–200 ppm). The smaller the IC₅₀ value means the higher antioxidant activity.²² The antioxidant activity showed that the extract of arabica coffee had a lower antioxidant activity with an IC₅₀ value of 353.72 mcg/mL than the vitamin C comparator with an IC₅₀ value of 28.22 mcg/mL. According to Rosliuk, 2020 the chlorogenic acid contained in green coffee beans is also susceptible to heat, oxygen, light, and humidity, due to the presence of unsaturated bonds in its molecules which may be the cause of the low antioxidant activity of this green coffee bean macerate.²³

Table 2: Physical characteristics and SPF values of each formula

| | Quality evaluation of cream preparations | | | | | | SPF | | |
|----|--|-------|-----------------|--------------------------------|---------------|-----|-----|---------------------|---------|
| | pH | | Homogeneity (g) | Organoleptic Odor and Color | Spreadability | | | (EE x I x Abs) x CF | |
| | Before | After | | | 0 | 25 | 50 | Repeatability | Average |
| F0 | 6.96 | 6.75 | Homogenous (cm) | No Odor White | 4.6 | 4.9 | 5.2 | - | - |
| F1 | 6.73 | 6.62 | Homogenous (cm) | White Odor | 4.4 | 4.7 | 5.1 | 1 | 8.3312 |
| | | | | | | | | 2 | 8.3528 |
| | | | | | | | | 3 | 8.3321 |
| F2 | 6.65 | 6.53 | Homogenous (cm) | Brown Odor | 4.4 | 4.6 | 5.0 | 1 | 13.7032 |
| | | | | | | | | 2 | 14.4592 |
| | | | | | | | | 3 | 14.7184 |
| F3 | 6.56 | 6.42 | Homogenous (cm) | White Brownish Odor | 4.2 | 4.4 | 4.8 | 1 | 13.7647 |
| | | | | | | | | 2 | 14.4592 |
| | | | | | | | | 3 | 14.7183 |
| F4 | 6.58 | 6.42 | Homogenous (cm) | Brown light brown Odor | 4.1 | 4.4 | 4.6 | 1 | 17.3091 |
| | | | | | | | | 2 | 17.3091 |
| | | | | | | | | 3 | 17.3091 |
| F5 | 6.52 | 6.42 | Homogenous (cm) | Dark Brown | 4.0 | 4.3 | 4.5 | 1 | 23.3850 |
| | | | | | | | | 2 | 21.1261 |
| | | | | | | | | 3 | 22.6491 |

Table 3: ANOVA test of similarity of arabica coffee bean extract averages

| Repeatability Group | Mean ± SD | p-value |
|---------------------|----------------|---------|
| 1 | 0.247 ± 0.2175 | 1.00 |
| 2 | 0.247 ± 0.2182 | |
| 3 | 0.248 ± 0.2176 | |

In this study, we also examined the MDA profile test in the serum of experimental rats. Measurement of MDA levels in the sample group was carried out to see how the profile of MDA levels in the blood of the sample as a marker of cellular damage. The higher the radical level, the higher the MDA level formed.²⁴ From Table 5, the coffee extract slightly inhibits free radicals to bind with antioxidants more than the standard group (vitamin E). This data was also correlated with the antioxidant activity as presented in Table 4. Our study suggests that the arabica coffee bean extract might be the potential as an antioxidant and inhibit cellular damage due to free radicals.²⁴

The MDA results of the various treatments can be seen in the following Table 5.

Table 4: Antioxidant Test with DPPH Method

| Concentration (ppm) | Absorbance | | | % Inhibition | | | %Average Inhibition | IC ₅₀ (mcg/mL) |
|---------------------|------------|--------|--------|--------------|---------|---------|---------------------|---------------------------|
| | 1 | 2 | 3 | 1 | 2 | 3 | | |
| 100 | 0.5028 | 0.4921 | 0.4989 | 43.1800 | 44.4645 | 43.6526 | 43.7657 | 353,72 mcg/mL |
| 200 | 0.4656 | 0.4779 | 0.4673 | 47.3839 | 46.0670 | 47.2216 | 46.9084 | |
| 400 | 0.4329 | 0.4414 | 0.4307 | 51.0792 | 50.1862 | 51.3553 | 50.8739 | |
| 600 | 0.3973 | 0.3869 | 0.3892 | 55.1023 | 56.3368 | 56.0425 | 55.8272 | |
| 800 | 0.3488 | 0.3552 | 0.3464 | 60.5831 | 59.9142 | 60.8764 | 60.4579 | |

Antiaging and Anti-irritation Tests

The antiaging activity of a product is determined by the number of wrinkles caused by UV exposure on the skin of wistar rats. The appearance of more wrinkles indicates the antiaging activity was insufficient. In this study, the irritation test was determined by the patch test method proposed by Remington. The results of the wrinkles and irritation test until 72 hours of treatments can be seen in Table 6.

Based on Table 6 above, it is known that all groups do not react to the administration of arabica coffee bean extract. It indicates that there is no irritation due to the application of arabica coffee bean extract. The anti-irritation effect might be due to the bioactive compounds as well as additional ingredients that have been proven to be safe, inert, non-irritating, and non-toxic orally.²⁵

Table 6 also shows that the wrinkle score in group F0 has slight shallow rough wrinkles at 24 hours in rat 3 and 48 hours in rat 2. Group F1 has slightly shallow rough wrinkles at 72 hours in rats 1 and 4. Group F2 has slightly shallow rough wrinkles at 72 hours in rat 4. Group F3 has no wrinkles

Table 5: MDA levels of experimental rats

| Treatments | Rat group | MDA values | Average of MDA |
|----------------------------|-----------|------------|----------------|
| Control (NA-CMC) | 1 | 0.258 | 0.186 |
| | 2 | 0.169 | |
| | 3 | 0.153 | |
| | 4 | 0.165 | |
| Standard (Vitamin E) | 1 | 0.156 | 0.160 |
| | 2 | 0.147 | |
| | 3 | 0.185 | |
| | 4 | 0.153 | |
| 1% Dose of Coffee Extract | 1 | 0.185 | 0.152 |
| | 2 | 0.139 | |
| | 3 | 0.143 | |
| | 4 | 0.141 | |
| 5% Dose of Coffee Extract | 1 | 0.167 | 0.152 |
| | 2 | 0.139 | |
| | 3 | 0.158 | |
| 10% Dose of Coffee Extract | 1 | 0.156 | 0.152 |
| | 2 | 0.161 | |
| | 3 | 0.141 | |
| | 4 | 0.152 | |

Table 6: Number of wrinkles and irritations in experimental rats

| Treatments | Rat Group | Irritation | | | Wrinkles | | |
|-------------|-----------|------------|----------|----------|----------|----------|----------|
| | | Score | | | Score | | |
| | | 24 hours | 48 hours | 72 hours | 24 hours | 48 hours | 72 hours |
| Vaselin SPF | 1 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 2 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 3 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 4 | 0 | 0 | 0 | 0 | 0 | 0 |
| F0 | 1 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 2 | 0 | 0 | 0 | 0 | 1 | 0 |
| | 3 | 0 | 0 | 0 | 1 | 0 | 0 |
| | 4 | 0 | 0 | 0 | 0 | 1 | 0 |
| F1 | 1 | 0 | 0 | 0 | 0 | 0 | 1 |
| | 2 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 3 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 4 | 0 | 0 | 0 | 0 | 0 | 1 |
| F2 | 1 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 2 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 3 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 4 | 0 | 0 | 0 | 0 | 0 | 1 |
| F3 | 1 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 2 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 3 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 4 | 0 | 0 | 0 | 0 | 0 | 0 |

| | | | | | | | |
|----|---|---|---|---|---|---|---|
| F4 | 1 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 2 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 3 | 0 | 0 | 0 | 0 | 1 | 0 |
| | 4 | 0 | 0 | 0 | 0 | 0 | 0 |
| F5 | 1 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 2 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 3 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 4 | 0 | 0 | 0 | 0 | 0 | 0 |

occurred. Group F4 has slightly shallow rough wrinkles at 48 hours in rat 3, whereas group F5 has no wrinkles occurred. From the results of this study, it can be seen that the majority of samples in each group did not wrinkle after applying arabica coffee bean extract to the samples. The caffeine content in arabica coffee might make the skin smooth and moisturized, making the skin feel smoother and firmer so it is promisingly suitable to be applied as skin cream cosmetics.²⁶

CONCLUSION

When combined with oxybenzone and octyl methoxycinnamate, the arabica coffee bean extract has a maximum category of UV protection activity value of F1, F2, and F3 with SPF values of 8.3, 14.2, and 14.3, respectively. The ultra-category of F4 and F5 with SPF values of 17.3 and 22.3, respectively. In addition, it was found that the cream showed no irritation on the back of rats and no wrinkles on the soles of the feet of rats. Arabica coffee bean extract showed antioxidant activity with an IC₅₀ value of 353.72 mcg/mL when compared with vitamin C with an IC₅₀ value of 28.22 mcg/mL. In this study, the MDA test also indicated that arabica coffee bean extract can reduce MDA levels by 0.152 μmol/l compared with vitamin E of 0.160 μmol/l and the control group (NA-CMC) of 0.186 μmol/l. This research supports that Arabic coffee bean extract could be proposed as a candidate for ultra sunscreen and antiaging cream.

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