Extraction of Coffee Silverskin and the Development of Antioxidant-Rich Products

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Abstract

The coffee roasting industry leaves the silverskin, a thin layer covering the coffee bean, as waste. Recent studies have shown that coffee silverskin is rich in phenolic compounds with antioxidant property. In this work, the optimum conditions for the extraction of coffee silverskin were studied with the purpose of maximizing the phenolic content and antioxidant activity. It was observed that the type of solvent, the extraction temperature, and the extraction time strongly affected the phenolic content of the extract. The phenolic content of silverskin from two varieties of coffee bean, Robusta and Arabica, was also compared. It was found that the phenolic content of Robusta coffee silverskin was higher than that of Arabica coffee silverskin. Further, the coffee silverskin extract was used to develop antioxidant-rich products, including beverage, skin gel and skin lotion. All products added with coffee silverskin extract showed a high antioxidant activity, indicating that coffee silverskin has the potential to be used as a source of antioxidant in various products. A concentration of the coffee silverskin extract of 1% in the skin gel or the skin lotion resulted in the highest antioxidant activity.

Keywords: Coffee silverskin, Extraction, Antioxidant, Beverage, Skin care

Introduction

Indonesia is one of the largest coffee producers in the world. According to the data from the Directorate General of Estate Crops, Ministry of Agriculture of Republic Indonesia, in the year 2015 Indonesia produced 739,005 tons of coffee beans [1], placing Indonesia as the fourth largest coffee producer in the world. Due to large production of coffee beans in Indonesia, large amounts of waste are generated in the coffee industry. One of the coffee industry's waste products is coffee silverskin (CS) that are produced during roasting process of coffee beans. It is actually the outer thin layer of green coffee beans that is usually still intact after the depulping and dehulling process of the coffee cherries. When the green coffee beans come into contact with high temperature due to the roasting process, the coffee beans crack as the result of swelling due to loss of moisture content, and also make the silverskin detached from the beans. Due to its lightness, coffee silverskin usually escapes from the roasting container through the roaster exhaust or cyclone, and is disposed as waste by the coffee roasting industries. However, recent studies showed that coffee silverskin contains phenolic compounds which can be used as a source of antioxidant [2,3]. It is well known that antioxidant is very important for the human body for protection against free radicals. Although the human body has an internal defense system towards free radicals [4], it still requires the intake of antioxidants from the outside such as foods, supplements, beverages or topical medications.

Our previous study showed that coffee silverskin extract solution contains phenolic compounds [5], however a further study on the optimization of the extraction process is very important for the industrial scale extraction process of coffee silverskin. Besides, there is no study of the possible application of coffee silverskin for foods, supplements, beverages or topical medications. In this work, the optimum conditions for the extraction of coffee silverskin were studied with the purpose of maximizing the phenolic content and antioxidant activity of coffee silverskin. The condition of the extraction process such as the extraction temperature and the extraction time was varied to study their effect on the phenolic content and the antioxidant activity of the extract solution. Coffee silverskin extract in form of powder was also prepared. The coffee

silverskin extract was used to develop new products such as antioxidant-rich beverage, skin gel and skin lotion.

Materials and Methods

Materials

Robusta coffee silverskin and Arabica coffee silverskin were obtained from *Panen Raya* coffee roasting company, Bandar Lampung, Indonesia, and from *Morph Coffee* roasting company, Jakarta, Indonesia, respectively. Ethanol 96% was purchased from PT Sumber Abadi, a chemical supplier in Indonesia. Sodium carbonate (BDH, England), Folin-Ciocalteu reagent (Merck, Germany), gallic acid powder (Aktin Chemical, China), aluminum chloride (Merck, Germany), potassium acetate (BDH, England), 2,2-diphenyl-1-picrylhydrazyl (DPPH, Sigma-Aldrich), analytical grade ethanol (Smart Lab, Indonesia) were used. Purified water was used as the solvent. Chemicals to prepare basic skin gel were carbomer 940, propylene glycol, methylparaben, propyl paraben, all imported from China and purchased from PT Intralab Ekatama, while sodium metabisulfite, triethanolamine (TEA), ethylene diamine tetraacetic acid (EDTA) were all purchased from PT Sumber Abadi, Indonesia. Chemicals to prepare basic skin lotion were paraffin liquid, stearic acid, cetyl alcohol, triethanolamine, glycerine, methyl paraben, and perfume, which were obtained from PT Sumber Abadi, Indonesia.

Equipment

Standard laboratory equipment was used in this work, namely hot plate (Cimarec, USA), beaker glasses, Erlenmeyer flasks, magnetic stirrer, thermometer, volumetric glass (Pyrex), water bath shaker (Memmert, Germany), micropipette (Eppendorf, Germany), micropipette tips, cuvette (Brand GMBH, Germany), rotary vacuum evaporator (IKA HB 10, China), spray dryer (BUCHI mini spray dryer B-290, Switzerland), analytical balance (Ohaus PA214, USA), UV-Vis Spectrophotometer (Genesys 10-S, USA), vortex (Vortex-Genie 2, USA), filter instrument, Whatmann

filter paper 1001 125 (GE, UK), desiccator, moisture content analyzer (Sartorius MA35, Germany), mixer (IKA Labortechnik, Germany).

Extraction Procedure

The coffee silverskin sample was first homogenized using a blender to reduce the size. Then, 100 ml of water-ethanol mixture with a weight ratio of 50:50 was poured into a 200 ml beaker and heated using a hot plate. The temperature was varied at 30, 40, 50 and 60˚C. When the temperature was reached, 2 grams of coffee silverskin were poured into the beaker, thus the weight ratio of coffee silverskin and water-ethanol solvent was 1:50. The beaker was covered with aluminum foil to prevent heat loss and evaporation of the solvent. The extraction time was varied at 5, 10, 20, 30, 40, and 60 minutes. The extraction was carried out with agitation using a magnetic stirrer put in the beaker, and the stirring was set at a level of 6 (350 rpm). The extract solution was then filtered using a filter paper and stored in a refrigerator prior to analysis.

Analysis of Total Phenolic Content and Antioxidant Activity

The total phenolic content was analyzed by using Folin-Ciocalteu method with gallic acid as a standard based on the modified method of Costa [3]. Firstly, the Folin-Ciocalteu reagent solution was made by diluting concentrated Folin-Ciocalteu reagent in distilled water at a ratio of 1:10. Sodium carbonate solution (7.5% w/v) was made by diluting 7.5 grams of solid sodium carbonate with 100 mL of distilled water. For the standard curve of gallic acid, a solution of gallic acid was prepared by dissolving 0.1 g of solid gallic acid in 100 ml of distilled water to obtain 1000 mg/L of gallic acid stock solution. Then the standard solutions of gallic acid ware made by diluting the gallic acid solution into concentrations of 10, 20, 50, 70, 100, 250, and 500 mg/L.

Further, the coffee silverskin extract solution was diluted with a weight ratio of 1:10, and 500 µL of the diluted extract solution was mixed with 2.5 ml of the Folin-Ciocalteu reagent solution and 2 ml of the sodium carbonate solution. The mixture was then vortex and incubated in dark at room temperature for 1 hour. After the incubation, the

mixture was poured into cuvette and was directly checked by using a UV-Vis spectrophotometer at 765 nm. The absorbance was then noted and checked to the equation of gallic acid standard calibration curve. The total phenol content (TPC) was then calculated according to the following equation:

$$Total\ Phenolic\ Content\ (mg\ GAE/L) = \frac{Abs\ x\ DF}{m}$$

where Abs is the absorbance (A), m is the gradient of the gallic acid standard curve ($A/(mgL^{-1})$), DF is the dilution factor.

The total phenolic content was expressed in mg of Gallic Acid Equivalent per liter extract solution (mg GAE/L) or mg GAE/g coffee silverskin sample (mg GAE/g CS).

To determine the antioxidant activity of the extract, the extract samples were tested using DPPH Radical Scavenging Assays with DPPH inhibition method. A stock solution of DPPH (250µM) was prepared by diluting 11 mg of DPPH powder in 20 ml ethanol (96% v/v). The stock solution was covered with aluminum foil and stored at a temperature of 4ºC. Next, 100 µL DPPH, 50 µL sample and 850 µL ethanol were mixed in a test tube. For the control, 100µL DPPH, 50µL distilled water and 850µL ethanol were mixed in a test tube. Furthermore, both test tubes were wrapped with aluminum foil and stored in a dark room for 30 minutes. The absorbance reading was performed using a UV-Vis spectrophotometer at a wavelength of 515 nm. The antioxidant activity was expressed as inhibition percentage and calculated according to the following equation:

$$Antioxidant\ Activity\ (\%) = \frac{(Ac - As)}{Ac}\ x\ 100\%$$

where Ac is the absorbance of the control (A) and As is the absorbance of the sample (A).

The antioxidant activity (the radical scavenging activity) obtained by this DPPH inhibition method was also expressed as IC_{50} (in ppm), which means the concentration of the sample needed to inhibit 50% of the free radical (DPPH).

Statistical Analysis

Openstat was the statistical software used to perform the statistical test of the data in this research. Three way ANOVA and Tukey HSD test was performed using Openstat. The significance level throughout the statistical test was 0.05. When the probability (noted as "p") is less than 0.05 then the comparison is concluded to be significant different. In short, there is a 5% probability that the statistical comparison wrongly rejects the null hypothesis.

Procedure for Coffee Silverskin Beverage

The extraction procedure to prepare coffee silverskin beverage is the same as the extraction procedure above, however pure water was used the solvent. The weight ratio between coffee silverskin and water was 1:20, the extraction temperature was set at 50ºC, and the extraction time was 180 min. Affective and descriptive sensory analysis were conducted to give the initial score and responses of the silverskin extract. Then, a formulation of beverage from coffee silverskin extract was carried out to develop a new antioxidant-rich beverage. Affective and descriptive sensory analysis of the new developed beverages were conducted to determine the final formula based on consumers' acceptance.

Procedure for Coffee Silverskin Skin Gel

To produce coffee silverskin extract in the form of powder, the coffee silverskin extract solution was dried using a spray dryer with an inlet temperature of 175ºC and an outlet temperature of 125ºC with a feed flow of 16.7 ml/min. The coffee silverskin extract powder was then analyzed for its total phenolic content and antioxidant activity using the method as previously described. To prepare skin gels, first the basic gel was

prepared using 1% of Carbomer 940 by cold mechanical method described by Smolka [6] with some modification. Required quantity of Carbomer 940 was weighed individually, and sufficient amount of distilled water was mixed in separate beaker. In order to reduce the bubble, the gelling basis need to be kept 24 hours at room temperature. Triethanolamine (TEA) was added to the gel with a ratio of 1:1. Methyl paraben and propyl paraben was dissolved in propylene glycol, natrium bisulfate was also added to the distilled water, Ethylene diamine tetraacetic acid (EDTA) that already dissolved in distilled water was stirred until homogeneous. Then the coffee silverskin extract powder which was already dissolved in water was added and stirred with selected mixer speed (500 rpm, 600 rpm, 700 rpm) for 20 minutes. The concentration of the coffee silverskin extract powder in the skin gel was varied from 0.125%, 0.25%, 0.5% and 1%. The antioxidant activity of the skin gel was analyzed in term of IC_{50} using the method as described previously.

Procedure for Coffee Silverskin Skin Lotion

The basic lotion was prepared according to the method as described in literature [7] with some modification. First the oil phase materials (paraffin liquid, stearic acid, and cetyl alcohol) were put together into the 100 ml beaker glass, mixed and heated at 70ºC. Then the water phase materials (triethanolamine, glycerine, and purified water) were put together into another 250 ml beaker glass, mixed and also heated at 70ºC. When both temperatures have reached 70ºC, the oil phase was poured into the water phase then mixed together using a mixer. The mixing process was carried out using a dispersion type propeller at a speed level of 5 for the first 15 seconds, then the speed was lowered to a speed level of 2 for 3 minutes. Next, the speed was set to speed level 1 until both phases were homogenized and reached a temperature of 40ºC. Finally, methyl paraben, perfume, and the coffee silverskin extract powder were added to the lotion at a temperature of 40ºC and mixed until the mixture become homogeneous. Finally, the lotion was cooled down at room temperature and kept in a storage bottle. The concentration of the coffee silverskin extract powder in the skin lotion was varied from 0.125%, 0.25%, 0.5% and 1%. The antioxidant activity of the skin lotion was analyzed in term of IC_{50} using the method as described previously.

Results and Discussion

Total Phenolic Content and Antioxidant Activity of Coffee Silverskin Extract Solution

Table 1 depicts the total phenolic contents and the antioxidant activity of two different type of coffee silverskin extract solutions which were obtained by the extraction of Arabica and Robusta coffee silverskin at an extraction temperature of 40ºC and an extraction time of 60 min. It was found that the total phenolic content of Robusta coffee silverskin extract was 816.76 ± 63.24 mg GAE/ L, higher than that of Arabica coffee silverskin which had a total phenolic content of 473.51 ± 56.70 mg GAE/ L. This finding is in accordance with the study conducted by Farah [8] who reported that Robusta coffee green bean has a higher amount of chlorogenic acid compared to Arabica coffee green bean. Chlorogenic acid belongs to the phenolic group, therefore contributing to the absorbance reading of phenolic group. Due to the higher phenolic content, Robusta coffee silverskin extract showed a higher antioxidant activity (radical scavenging activity) compared with Arabica coffee silverskin as can be seen in Table 1.

Table 1. Total Phenolic Content and Antioxidant Activity of Robusta and Arabica Coffee Silverskin

	Coffee Silverskin Extract Solution	
	Robusta	**Arabica**
Total Phenolic Content (mg GAE/g CS)	40.84 ± 3.16	23.68 ± 2.84
Antioxidant Activity (%)	54.8 ± 1.2	26.3 ± 1.9

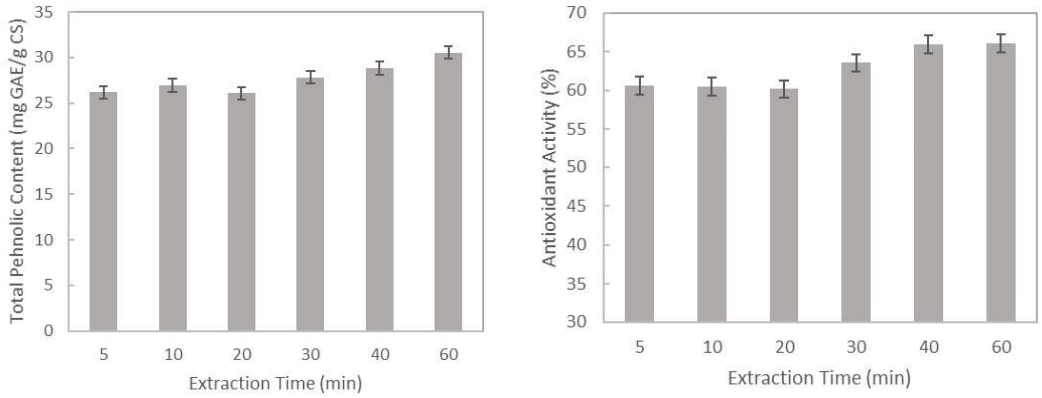

Fig. 1. Effect of extraction time on the total phenolic content and the antioxidant activity of the coffee silverskin extract solution

Figure 1 shows the effect of extraction time on the total phenolic content and the antioxidant activity of the Arabica coffee silverskin extract solution. As can be seen, the longer extraction time resulted in a higher amount of total phenolic content of the coffee silverskin extract solution. As a result, the antioxidant activity increased with increasing extraction time due to the higher phenolic content. According to the statistical analysis using ANNOVA, the extraction time of 60 minutes is the variable which has the highest mean and significant difference in the total phenolic content and the antioxidant activity compared to other extraction times. When the extraction temperature is increased from 30ºC until 60ºC, an increase of the total phenolic content and the antioxidant activity was also observed as shown in Figure 2. The statistical analysis using ANNOVA showed that the total phenolic content and the antioxidant activity at the extraction temperature of 60ºC had a significant difference compared to the other extraction temperatures.

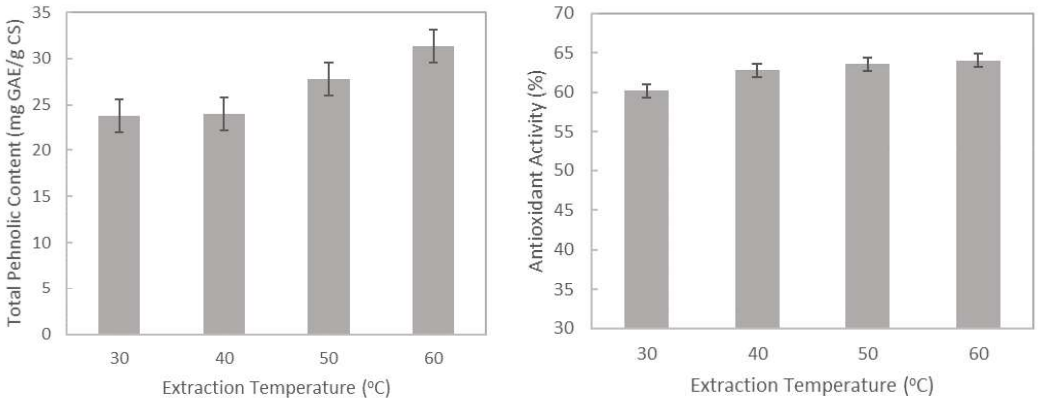

Fig. 2. Effect of extraction temperature on the total phenolic content and the antioxidant activity of the coffee silverskin extract solution

Development of Coffee Silverskin Beverage

Based on the finding that coffee silverskin contains a high phenolic content and antioxidant activity, a new beverage was developed using the coffee silverskin extract with the purpose to obtain an antioxidant-rich beverage product. Sensory analysis was conducted in order to plot the taste profile of the coffee silverskin extract solution. The panel consisted of 30 untrained people who were asked to score the intensity of each flavor in the extract, the taste resemblance, as well as the improvement needed to make the extract become more palatable. The intensity of each taste characteristic was evaluated using 1 – 10 scale, with 1 as the weakest/lowest intensity and 10 as the strongest/highest intensity. The result was plotted against a radar/spider-web chart, as can be seen in Figure 3 below. It seemed that bitter taste was the most dominant taste, followed by astringent, smoky/roast, and cocoa taste. The bitterness and astringency of the extract was suspected due to its high phenolic content, more specifically, its chlorogenic acid content. Chlorogenic acid has been known to contribute bitterness, acidity and astringency of brewed coffee – even is more influential than caffeine [8,9,10]. The roast/smoke note was probably coming from the roasting process, as it was common to be found in roasted coffee bean. This result was useful to decide which ingredients and flavors needed to be added, to improve the overall taste.

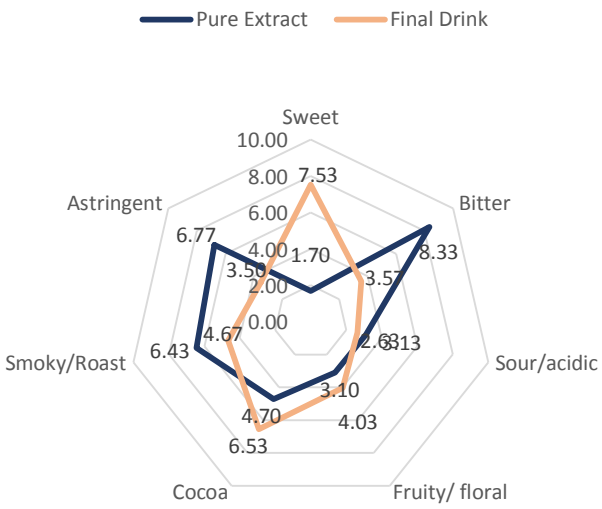

Fig. 3. Comparison of taste characteristic between the pure coffee silverskin extract and the newly developed coffee silverskin beverage

The panelists were also asked about the resemblance of the extract's taste, or whether there was any beverage that had similar taste with the extract. As much as 40% of the panelists agreed that the extract resembled the taste of coffee, whereas 20% of the panelist said it resembled the taste of tea, and the rest of the panelists said it resembled the taste of other beverages such as chocolate or herbal drinks. The panelists were also asked about their suggestions to improve the extract's taste. The majority agreed that the sweetness must be increased while the bitterness should be decreased. Some suggested adding chocolate or fruit flavor to enrich the overall taste, while the astringent and smoky/roast taste should be minimalized. Based on this result, cyclodextrin was added to the coffee silverskin beverage to mask the bitterness, while sugar and flavor were added to increase the sweetness and to improve the taste. The optimization of the formulation was done by the help of Design Expert Software, which generated two optimum formulas that matched the target and later decided based on affective test. It was found that the final formula consisted of 4.36% silverskin, 5.83% sugar, 0.22% chocolate flavor and 1.00% cyclodextrin (w/v). The analysis result showed that the new coffee silverskin drink had 1219.08 mg GAE/L of total phenolic content and 54% of antioxidant activity. Based on sensory analysis, the overall taste of the new drink had

also gone through a significant improvement that led to a high acceptance level as shown in Figure 3.

Development of Coffee Silverskin Skin Gel

The high antioxidant activity of the coffee silverskin extract has a great potential for topical products such as skin gel which can be applied to the human skin to prevent the skin from free radicals such as UV light. To prepare an antioxidant-rich skin gel, the extract powder of Robusta coffee silverskin was added into the basic gel with different concentrations of 0.125%, 0.25%, 0.5% and 1%. Figure 4 shows the antioxidant activity expressed by the IC_{50} value of the skin gel. The IC_{50} value indicates the concentration in ppm of antioxidant which is necessary to inhibit 50% of the free radical (in this case DPPH). Thus, a low value of IC_{50} corresponds to a high antioxidant activity. As can be seen in Figure 4, the addition of more coffee silverskin extract reduces the IC_{50} value, which means it resulted in a skin gel with a higher antioxidant activity. The statistical analysis using ANNOVA showed that there is a significant difference of IC_{50} with the variation of the extract concentration.

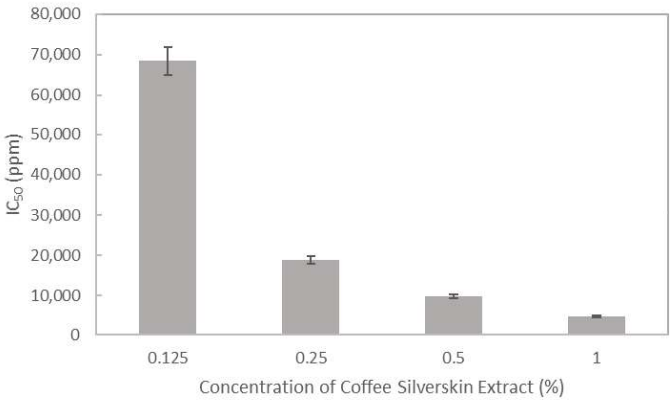

Fig. 4. IC_{50} of skin gels with various concentrations of coffee silverskin extract powder

Development of Coffee Silverskin Skin Lotion

Skin lotion is a water-oil emulsion which is usually used as body lotion. The addition of antioxidant into the lotion is very beneficial for human skin to protect it from free radicals such as UV light. Robusta coffee silverskin extract powder was added into the basic skin lotion with various concentrations of 0.125%, 0.25%, 0.5%, and 1%. Figure5 shows the IC_{50} value of the skin lotion containing the coffee silverskin extract. A similar result with the skin gel above, the addition of coffee silverskin extract into the skin lotion was very effective obtaining a skin lotion with a high antioxidant activity. The statistical analysis using ANNOVA also showed that there is a significant difference of IC_{50} with the variation of the extract concentration in the skin lotion.

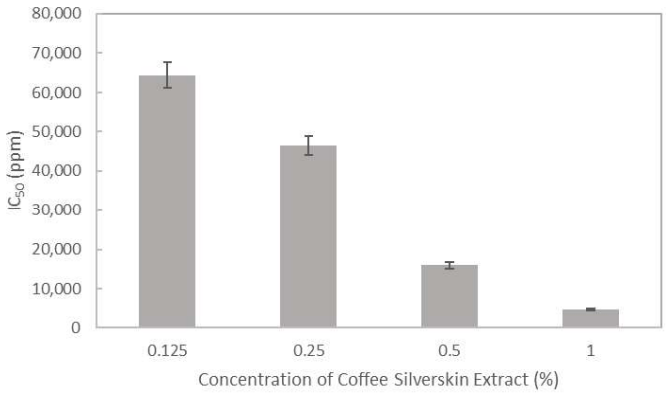

Fig. 5. IC_{50} of skin lotions with various concentrations of coffee silverskin extract

Conclusion

The extraction of coffee silverskin, a waste from coffee roasting industry, resulted in a product having a high antioxidant activity due to the high phenolic content in the coffee silverskin. It was found that Robusta coffee silverskin showed a higher total phenolic content, thus a higher antioxidant activity, compared to the Arabica one. The extraction time and the extraction temperature strongly affected the total phenolic content and the antioxidant activity. The application of the coffee silverskin extract as source of antioxidant are wide. The coffee silverskin extract was used to develop antioxidant-rich

products, in this work including beverage, skin gel and skin lotion. All products added with coffee silverskin extract showed a high antioxidant activity, indicating that coffee silverskin has a great potential to be used as a source of antioxidant in various products.

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