Optimization Of Natural Body Scrub Formulation Based On Oilseed Press Cake Of Nyamplung (*Calophyllum Inophyllum* L) Using D-Optimal Mixture Experimental Design

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Abstract. UV light and air pollution are sources of free radicals that can trigger skin problems. Skin problems can be overcome by using skin treatments that contain antioxidant compounds so that oxidation reactions derived from free radicals can be inhibited (Halliwell and Gutteridge, 2000). Skin care that can overcome skin problems is body scrub preparation. Body scrub cream-based preparations consists of oil and water phase, emulgator, and abrasives. Oilseed press cake or residual product of nyamplung (tamanu) oil (*Calophyllum inophyllum* L.) has characteristics as an abrasiver and reported degree of hydrolysis that can counteract free radicals using the DPPH method through a % RSA value of 39.82%. Oilseed press cake of nyamplung will be developed as a raw material for the formulation of body scrub that have added value. Body scrub formulation was done by optimizing the emulgator material (stearic acid and triethanolamine) using Expert Design software D-Optimal and the optimal emulgator concentration was obtained (stearic acid 3% and triethanolamine 1%) and physical in stability of the optimum formulation is tested in 28 days in 1,2,3 and 4 week at room temperature by looking at the parameters of organoleptic physical properties, pH, viscosity, spreadability, and adhesion test. Based on physically examination, it showed that the formula of oilseed press cake of nyamplung body scrub for 28 days at room temperature is a semi-solid, brown color and smells of apple, pH value is 7.64±0.128; the viscosity value is 3.63±0.20 cm, and adhesiveness is 2.3±0.04 second.

1. INTRODUCTION

UV rays and air pollution are a source of free radicals that can have a harmful effect on the body. The impact is a skin problem[1]. One effort to help the body from free radical attack and provide protection to the skin is the need for antioxidant compounds[2] by inhibiting oxidation reactions caused by free radicals. The human skin is the outer covering of the body so it will be exposed to free radicals directly. Therefore, protection and care for the skin needs to be done.

Oilseed press cake or residual product of nyamplung oil (Calophyllum inophyllum L.) is reported still has a nyamplung oil content of around 4-5% [3]. Said et al (2007) reported that nyamplung oil contains compounds that have the potential to have antioxidant activity[4]. The compound content in nyamplung oil which has the potential to have antioxidant activity is flavonoids, steroids, saponins and triterpenoids [5]. Residual product of nyamplung oil is reported to contain 21.56% protein [6] which results from the hydrolysis of the protein to produce protein hydrolyzate and is reported to have the ability to capture free radicals as proton donors and metal ion binders. The greater the degree of hydrolysis (DH) hydrolyzed protein of residual product of nyamplung oil showed an increase in its ability to capture free radical DPPH through the% RSA value that was equal to 39.82%[7].

Body scrubs are a cream-based preparation that contains abrasives which help to remove dead skin cells and help the skin to exfoliate and nourish skin [8]. In scrub preparation, it requires a cream base component consisting of an oil and water phase containing an emulgator and an abrasive. An emulgator is an active ingredient that can reduce the tension between oil and water phase that droplets are dispersed inside a strong layer which is enhanced by preventing coalescence and changing the dispersed phase [9] which can optimize the emulgator in using cream stability. The D-optimal mixture experimental design was employed to optimize emulgator in body scrub based on Oilseed press cake or residual product of nyamplung oil.
Based on this explanation, the development of cosmetic formulation using Oilseed press cake of nyamplung oil is one of efforts to tackle its waste problem. Waste that originally had no economic value, will be processed into products that have added value as raw materials for natural cosmetics.

2. Experimental

2.1. Materials

Oilseed press cake or residual product of nyamplung oil was obtained from BUMDesa, Bantul Regency, Yogyakarta, Indonesia which is a partner in processing nyamplung seeds into cosmetic raw material. Dried coconut (Vegan Indonesia), phenoxyethanol (Nusae Chemical), Triethanolamine (Nusae Chemical), Cetyl alcohol, stearic acid, Propylene Glycol, glycerin, aquadest, corrigen odoris (Brataco Chemika). All others chemicals and ingredients used were of analytical and cosmetics grade.

2.2. Formulation of body scrub

Oilseed press cake of nyamplung oil is mashed using a blender and sieved using a 30 mesh number sieve. Then roasted for 10 minutes at 60°C. Body scrub was formulated using a mixture of oilseed press cake of nyamplung oil and dried coconut as scrub, prepared at the laboratory scale (Table 1).

| Ingredients                        | Formula 1 (%) | Formula 2 (%) |
|------------------------------------|---------------|---------------|
| Oilseed press cake of nyamplung oil| 5             | 5             |
| Dried coconut                      | 3             | 3             |
| Cetyl alcohol                      | 4             | 4             |
| glycerin                           | 5             | 5             |
| Propylene Glycol                   | 5             | 5             |
| phenoxyethanol                     | 0.5           | 0.5           |
| stearic acid                       | 2             | 3             |
| Triethanolamine                    | 1             | 2             |
| Aquadest                           | ad 100        | ad 100        |
| Corrigen odoris                    | qs            | qs            |

Firstly, the oil phase (A) made from stearic acid and cetyl alcohol is heated at 70°C until the mixtures melted, then phenoxyethanol is added. The aqueous phase (B) made of propylene glycol is dissolved in hot water (70°C), then glycerin, triethanolamine, and phenoxyethanol are added. The mixtures (A and B) were homogenized together using homogenizer, oilseed press cake of nyamplung oil and dried coconut are added, stirring until homogeneous [10 with modifications]. The protective materials are added at the end.

2.3. D-optimal design

The preliminary formulation of body scrub was prepared in order to obtain a suitable range to be added in the mixture design using Design Expert 7.1.5 D-Optimal software. The optimum formulation that was suggested in previous work consists of combination emulgator 2 and 3% of stearic acid, 1 and 2% of triethanolamine. Thus, two preliminary formulations were made according to this previous work in order to obtain the lowest limit and highest limit for the amounts to be added in the D-optimal design.

The response variables which used are two responses namely the value of viscosity (dPas) and pH value. Response targets are adjusted to obtain optimal combinations of stearic acid and triethanolamine emulgator. The results of the optimal formula obtained by the software are then tested for variable response and compared with the predicted value of the variable response from the Design Expert software D-Optimal method. Statistical analysis for formula verification uses one sample t-test
to find out whether the predictions produced by the software produce significantly different data on
the body scrub preparations from experimental results.

2.4 Body scrub evaluation
The Body scrub cream was evaluated for organoleptic [11], viscosity [12], pH [13-14], spreadibility
and adhesion [14]). Evaluation of body scrub cream is carried out at weeks 1, 2, 3, and 4 where each
body scrub is stored at room temperature.

2.5 Data analysis
Analysis of the physical properties stability of the body scrub cream which includes pH, viscosity,
spreadability and adhesion test was carried out statistical analysis with one-way ANOVA test.
Organoleptic test was analyzed descriptively.

3. Results and Discussion
The body scrub formulations are made in the form of cream preparations and contain oilseed
press cake of nyamplung oil that act as abrasives. Oilseed press cake of nyamplung oil which has the
potential for antioxidant activity will be used as an abrasive which can facilitate the process of
removing dead skin cells (exfoliate). Dried coconut was used in this study as an abrasive for having
moisturize and soften to the skin.

Cream preparation is chosen because it was easily applied to the skin. This cream is based on oil
cream in water (o/w). The type of oil in water is easily applied to the skin [15]. The mixture of stearic
acid and triethanolamine will make salt forming which is triethanolamine stearic is anionic and can
stabilize the type of emulsion oil in the air[16].

The mixture of emulgator is chosen because combination is more effective than single emulgator.
The ability of the combined emulgator is able to increase the strength of the interface layer, and can
increase the stability of the emulsion [17]. Cetyl alcohol acts as a stiffening agent. Glycerin functions
as a humectant and emollient, as a material that retains water in preparations and can also increase
skin moisture. Glycerin is hygroscopic that can bind water or reduce the amount of water evaporation
[18-19]. Propylene glycol act as humectants and phenoxyethanol as preservative[19]. The process of
mixing the oil and the water phase is carried out by heating at a temperature of 70°C which aim to
accelerate the reaction of the accumulation of stearic acid by the Triethanolamin base, and reduce the
surface tension between the oil and water phase so that the formation of the emulsion system can
occur perfectly [20]. In cream preparations, an emulgator is a material used to reduce the interface
tension between oil and water surrounding the droplets dispersed in a strong layer. It prevents
coalescence process and separation of the dispersed phase [9].

Therefore, it is necessary to optimize the emulgator materials. In this research, optimization of
materials that act as emulgator is stearic acid and triethanolamine. The two variable response body
scrub preparations using combination two emulgator compositions viscosity and pH value having a
significant model and insignificant lack of fit. Then two response variables can be used as parameters
determining the optimal formula. Determination of the target viscosity and variable response is in
range. The selected desirability value is the highest value which is 1 with the composition of stearic
acid and triethanolamine 3% and 1%. Design Expert prediction showed that viscosity response
variable is 2992.66 dPas, and the pH value is 7.92412. Verification of the optimal formula for the
body scrub using a combination of stearic acid emulgator and triethanolamine was carried out based on the
prediction results which aimed to determine whether the predictive value suggested by the software
matches with the actual experimental results.

Prediction of the optimal formula variable response obtained from the analysis using Design
Expert 7.1.5 software was then compared with the optimal formula variable response obtained in
experiments using IBM SPSS Statistics 2 software with one sample t-test method.

The response value of the viscosity and pH variables of the experimental results showed that the
results were not significantly different. It indicated the optimal formula prediction suggested by
Design Expert 7.1.5 software D-Optimal method could be used to predict the response of the variable
viscosity and pH of the body scrub combination of stearic acid emulgator and triethanolamine.
The verified formula was evaluated based on physical stability of the preparation with three replications. The aim of this evaluation is to determine the stability of the preparation during the storage period by testing the physical properties at weeks 1, 2, 3, and 4 at room temperature. Physical stability tests include organoleptic, pH, viscosity, spreadability, and adhesion tests were evaluated [21].

The body scrub was visually tested organoleptically to observe changes in shape, aroma, and color during week 1 through week 4 at room temperature storage. Organoleptic test results did not show any changes in color, shape and aroma during storage. It can be state that the body scrub is stable with characteristic semi-solid form and brown color and having pleasant odor.

The purpose of pH evaluation was to determine the pH value of the preparation and it can be known whether irritating to the skin. If the pH of the preparation showed alkaline it will affect the elasticity of the skin, but if the preparation is acidic it will cause the skin to be easily irritated [10]. Based on the requirements of SNI 16-4954-1998 regarding the pH of cream preparations, the pH cream requirements are 4.5–8. The results showed that pH value every week during storage decreased which can be seen in Figure 1.

![Figure 1. Graphic of pH body scrub testing during 4 weeks storage](image)

Decreasing pH value during 4 weeks storage is possible due to the influence of anionic soap formed from stearic acid and triethanolamine in the formula that serves to regulate the pH of the preparation. Acid formed in the preparation can not be longer balanced by anionic soap, so the presence of the acid causes the pH to decrease [22]. Test results using One way Anova show p-value data <0.05 seen through the Post Hoc Test with Bonferroni test there are differences in pH values at week 1 to week 3, at week 1 to week 4, at week 2 to week 4, week 2 to week 1, week 3 to week 1, and week 4 to week 2 during storage at room temperature. However, the decrease in pH did not cause a significant effect because it was still within the limits of the pH value for cream preparations (4.5-8) so it does not cause irritation to the skin.

Viscosity test was carried out to determine the thickness of the body scrub preparation. Viscosity in the body scrub preparation is a resistance of a preparation to flow. The greater the resistance, the greater the viscosity. The viscosity of a preparation influences the extent of its spread. According to SNI 16-4399-1966 requirements regarding topical cream preparations, cream preparations have a viscosity between 2000-50000 dPas.

Viscosity value is influenced by thickening agent, surfactant, proportion of dispersed phase and particle size. As the proportion of the dispersed phase increases, the concentration of the emulgator increases and the particle size decreases, then the viscosity of the emulsion will increase [23]. The results of the average viscosity value of body scrub preparations in the first week to the fourth week during storage at room temperature can be seen on Figure 2.
The purpose of the spread ability test is to know the softness of the body scrub mass. Then it can be seen the ease of application of cream to the skin. Good spreadability causes the contact between cream and the skin to become wider, so that absorption of the preparation to the skin takes place quickly [10]. The spreadability requirement for topical cream preparations is 5 - 7 cm [9]. Even though the results of the average spread test of the cream during 4 weeks at room temperature showed a decline (Figure 3) but the difference in viscosity did not affect the requirements (5-7 cm). Except in third week the value did not meet the requirements.

![Figure 2](image2.png)
**Figure 2.** Graphic of pH body scrub testing during 4 weeks storage

Increasing viscosity value can be caused by a decrease in pH value. Low pH will occur isoelectric point which is a condition where the protein in the material will clot so that it will increase the thickness [24]. This is in accordance with the statement of Restiani (2016) [6] which states that oilseed press cake has a high protein content and it can cause high viscosity. One way Anova Test showed that p-value less than 0.05. By using the Post Hoc Test with Bonferroni test, there were differences in the value of viscosity at week 1 to week 3, week 1 to week 4, week 2 to week 4, week 2 to week 1, week 3 to week 1, and week 4 to week 2 during storage at room temperature. However, the difference in viscosity does not affect the requirements (2000-50000 dPas).

The purpose of the adhesive test is to determine the ability of the cream to stick to the surface of the skin when applied. The longer a semisolid preparation can stick to the skin, the absorption of active substances in the skin will be better [24]. The good adhesion of semisolid preparations is more than 1 second [24]. The results of the average test of the adhesive strength of the scrub body in the first week to the fourth week during storage at temperature showed more than 1 second.

![Figure 3](image3.png)
**Figure 3.** Graphic of spreadability test of body scrub during 4 weeks storage

The purpose of the adhesive test is to determine the ability of the cream to stick to the surface of the skin when applied. The longer a semisolid preparation can stick to the skin, the absorption of active substances in the skin will be better [24]. The good adhesion of semisolid preparations is more than 1 second [24]. The results of the average test of the adhesive strength of the scrub body in the first week to the fourth week during storage at temperature showed more than 1 second.
The results showed there was an increase in the value of adhesion in the cream. Increasing the value of adhesion can be caused by increased viscosity values [10]. Test using One way Anova showed p-value less than 0.05. Using the Post Hoc Test with the Bonferroni test, there were differences in the value of adhesion at week 1 to week 3, at week 1 to week 4, at week 2 to week 4, at week 2 to week 1, to week 3 to week 1, and to week 4 to week 2 during storage at room temperature. However, the difference of adhesive power did not affect the stickiness value requirements (more than 1 seconds).

The body scrub formula made from residual waste of nyamplung oil (*Calophyllum inophyllum* L.) fulfills physical stability criteria, with characteristics semi-solid, brown, pleasant odor, pH value of 7.647 ± 0.128; viscosity value of 3,633.33 ± 152.75 dPas; the spread value is 3.87 ± 0.20 cm, and the adhesive power is 2.3 ± 0.04 seconds.

4. Conclusion
The results of the research showed that the residual waste from the production of nyamplung oil can be used as a raw material of cosmetics derived from natural ingredients that have antioxidant activity. By optimizing formulation using d-optimal design, the body scrub made from residual waste of nyamplung oil (*Calophyllum inophyllum* L.) fulfills physical stability criteria. Utilization of this waste is an effort to minimize waste and produce innovative products that have economic value.

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