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Original Article

OPTIMIZATION OF BENTONITE LIQUID DETERGENT FOR CLEANSING OF EXTREME NAJS USING SIMPLEX LATTICE DESIGN

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ABSTRACT

Objective: The objective of this research was to formulate bentonite (clay) liquid detergent (BLD) using bentonite combined with vegetable oils intended for cleansing of extreme najs.

Methods: In this study, five formulas of BLD in combination with vegetable oils (coconut and palm oils) were made. The quality of BLD was evaluated in terms of organoleptic, foam power, foam stability, viscocity, pH, free alkali, and total fatty acids. These parameters were analyzed using simplex lattice design (SLD) in order to get the optimum formula of BLD. The verification of optimum formula was carried out using statistical analysis of one sample t-test at a significance level (P) of 0.05.

Results: The results showed that the optimum concentration of bentonite was 6.4% with the percentage of cocount oil and palm oil of 95.1% and 4.9%, respectively. Based on one sample t-test, there was no significant difference (P>0.05) of foam power, foam stability, and pH between optimum formula BLD predicted using SLD and that actually formulated.

Conclusion: BLD formulated can be used as innovatine means for cleansing extreme najs.

Keywords: Bentonite, Liquid detergent, *Najs mughalladzah*, Simplex lattice design, Islamic cleansing

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INTRODUCTION

Currently, the needs for halal products which includes foods, pharmaceuticals, cosmetics, services, and other products, has increased globally, especially in the countries Muslims. It is predicted that Muslim population is 27% of the global population in 2030 [1]. Over the last few years, the global market of halal products and services has increased exponentially in which food products captured the sales of USD 632 billion, which is equivalent to 16% of whole global food industry [2]. In addition, if food sectors are combined with other Islamic sectors such as Islamic finance and insurance, hoteling, toys, fashion, cosmetics, pharmaceuticals, and entertainment, the global halal industry is estimated to be over $2.1 trillion a year [3].

The development of the halal industry in Indonesia has emerged; as a consequence, Indonesia has stipulated Act on Halal Producst Assurance (Indonesian Act No. 33, 2014) [4]. The survey conducted by Yusuf and Ab. Yazid [5] showed that the awareness of halal products was considered quite high, in which 79% are aware in halal products. Halal products are products which fit to *Syariah* law and free from non-halal components such as pig derivatives and najs. If Muslims is in contact with any types of najs (religiously-prohibited dirty), they should carry out the cleansing of najs before they performed certain religious duties like prayer [6].

There are three categories of najs, namely ligh (mukhaffafah), medium (mutawassithah), and extreme or severe (muhgalladzah). The cleansing methods of najs are dependent to these categories. The objects including part of the human body are considered najs mughalladzah if they are in contact either directly or indirectly with najs of muhgalladzah, i.e. pig (khinzir) and dogs including any liquid coming from both animals such as lard or pig fat [7, 8]. The use of water in combination with detergent did not fulfill the requirement of syariah law during the cleansing of najis muhgalladzah. Islamic law required the use of seven steps of washing, with one of which shall be water mixed with clay or soil [9]. The procedure applied to cleansing najs muhgalladzah is known as samak or dibagh. However, the use of bulk clay mixed in water to clean najs muhgalladzah is not practical; therefore, clay formulated in solid or liquid detergent could be an effective approach to clean najs muhgalladzah. Therefore, this study formulated liquid detergent containing clay (bentonite) intended to clean najs muhgalladzah using an experimental design approach of simplex lattice design.

Table 1: Formula of bentonite liquid detergent used for cleansing of extreme Najs

Components	Formula				
	I	II	III	IV	5
Coconut oil (g)	30	22.5	15	7.5	0
Palm oil (g)	0	7.5	15	22.5	30
Olive oil (g)	5	5	5	5	5
Stearic acid (g)	10	10	10	10	10
Lanolin (g)	5	4	4	4	4
Sodium laurylsulphate (g)	5	5	5	5	5
Butyl hydroxytoluene (g)	0.1	0.1	0.1	0.1	0.1
KOH 20% (mL)	20	20	20	20	20
Citric acid (g)	1	1	1	1	1
Sucrose solution 30% (mL)	10	10	10	10	10
Glycerin (g)	10	10	10	10	10
Coco DEA (g)	2	2	2	2	2
Perfume (g)	0.5	0.5	0.5	0.5	0.5
Bentonite (g)	6.4	6.4	6.4	6.4	6.4

MATERIALS AND METHODS

Materials

Bentonite (pharmaceutical grade), lanolin, glycerin, and Coco DEA were bought from Brataco Chem (Yogyakarta, Indonesia). Perfume, olive oil, coconut oil, and palm oil were purchased from supermarkets around Yogyakarta. Stearic acid, sodium lauryl sulphate, butylhydroxytoluene, and KOH were obtained from E. Merck (Darmstat, Germany). Bentonite was sieved on mesh 100 to make detergent with homogenous particle. To optimize bentonite liquid detergent, five formulas as in table 1 were prepared.

Preparation of bentonite liquid detergent (BLD)

Briefly, BLD was prepared by placing coconut oil, palm oil, olive oil, stearic acid, lanolin, sodium laurylsulfate, and BHT into Beaker glass and heated until 60 °C in the water bath, and let until all components melt. NaOH 20%, previously heated at 40 °C, was added and stirred at 1000 rpm for 3 min. Then, sucrose 30% and citric acid were added into the mixture. Bentonite was added into glycerin and coca DEA, stirred, and added into the mixture. Finally, the perfume was added into the formula.

Evaluation of physico-chemical properties

Organoleptic evaluation was performed by investigating the appearance of BLD in terms of odor and color according to Indonesian National Standard (SNI 06-4085-1996) [10]. Determination of pH was undertaken following the method described in American standard for testing materials (ASTM) D1172-95 [11]. Determination of viscosity was performed using a Brookfield Viscometer DV-II+applying spindle number S25, at 5.0 rpm speed and at a shear rate of 1.10 s^{-1} according to [12]. The foam power and form stability were determined according to Piyali *et al.* [13]. Briefly, 1 gram of BLD was put into the reaction tube and then added with 10 ml aquadest. The mixture was vortexed for 1 min. The height of the foam was measured at 0 min and at min 60. While foam stability was calculated as follow:

$$\text{Foam loss (\%)} = \frac{\text{foam power at t=0 min} - \text{foam power at t=0 min}}{\text{foam power at t=0 min}} \times 100\%$$

Density of BLD was determined based on Indonesian National Standard (SNI 06-4085-1996) [10] at the temperature of 25 °C. Free alkali was determined by weighing 5 gram of BLD in Erlenmeyer and then added with 100 ml neutral alcohol 96%. The mixture was then heated for 30 min, and after cooling, added with 3 drops of phenolftalein as indicator and titrated with HCl 0.1 N until red color disappeared [10]. Determination of active components calculated as total fatty acids was performed using gravimetric analysis as in [10].

Optimization of bentonite liquid detergent (BLD)

Optimization of BLD was performed using software of Design-Expert versi 9.0.3 with Mixture Simplex Lattice method to obtain equations of Simplex Lattice Design. Data were analyzed based on the results (responses) obtained from the evaluation of physico-chemical caharcteristics.

Verification of bentonite liquid detergent (BLD)

Verification of BLD formula was carried by comparing the predicted responses obtained using software Design Expert and responses obtained from actual laboratory prepared BLD. The statistics of one sample t-test was used for this purposes using software SPSS version 22.

RESULTS AND DISCUSSION

In this study, the clay used to facilitate the cleansing on najs *mughalladzah* is bentonite type. Bentonite is a rock formed of highly colloidal and plastic clays composed mainly of montmorillonite, clay mineral of the smectite group, and is produced by in situ of volcanic ash devitrification [12]. The evaluation of physicochemical properties of bentonite liquid detergent (BLD) was intended to know wether BLD fitted the requirement and had good quality.

The optimization of BLD

The results of physicochemical properties evaluated including organoleptic, foam power, foam stability, viscocity, density, pH, free alkali, and active components (total fatty acids) were compiled in table 1. From these results, the optimum formula of BLD was determined using ssimplex lattice design (SLD).

Table 1: The results of physicochemical properties of bentonite liquid detergent

Physico-chemical properties	Form I	Form II	Form III	Form IV	Form V	Requirement
Organoleptic physical form Odor color	**Viscous liquid fragrant brown**	**Viscous liquid fragrant brown**	**Viscous liquid fragrant brown**	**Viscous liquid fragrant brown**	**Viscous liquid fragrant brown**	**Viscous liquid specipic specipic**
Foam power (cm)	6.87	5.47	5.63	5.13	4.83	-
Foam stability (%)	18.94	17.69	10.06	15.86	8.26	-
Viscocity (cPs)	20	31	30.33	43.33	110	-
Density (g/ml)	1.02	0.92	0.95	0.75	0.97	1,01-1,10
pH	10.12	9.89	10.2	9.92	10.45	8-11
Free alkali	0	0	0	0	0	max. 0.1
Active components	4.24	5.42	5.33	2.13	2.98	min. 15

Formula I: coconut oil-palm oil (100%:0%); Formula II: coconut oil-palm oil (75%:25%); Formula III: coconut oil-palm oil (50%:50%); Formula IV: I: coconut oil-palm oil (25%:75%); and Formula V: coconut oil-palm oil (0%:100%).

Fig. 1: Bentonite liquid detergent obtained using formula I, II, III, IV and V. The composition of each formula can be seen in table 1

The organoleptic evaluation revealed that all BLD formulas had brown colour due to the presence of bentonite (fig. 1) with fragrant odor due to the addition of perfumery components. Form power is essential during BLD formulation, therefore the higher the foam power, the better of BLD. The increased level of coconut oil make the levels of foam were increased. The main fatty acid which composed coconut oil is lauric acid (C12:0), which is known as good source to yield foam [13, 14]. Analysis of variance (ANAVA) test resulted in P value>0.05 which indicated that there were no significant differences among BLD formulas evaluated. Analysis using design expert indicated that oil components (coconut oil, palm oil and its combination) contributed significantly to make foam.

The foam stability was needed to make BLD stable over a certain time. The higher the levels of palm oil, the more stable of BLD due to the high level of palmitic and stearic acids present in palm oil. In addition, the presence of oleic acid (nearly 41%) also contributed to foam stability [15]. The other components used in BLD formula which may contribute to foam stability is olive oil with high levels of oleic acid and surfactant (coco diethanolamide and sodium lauryl sulphate). ANAVA test resullted that there were no significant differences of foam stability among formulas evaluated. The equation obtained to describe foam stability using simplex lattice design (SLD) was as in Eq. 1. SLD was used to explore the interior and the boundaries of the simplex. The coefficients of the model equation can be calculated easily [18].

$$Y = 80.39A+91.07B+0.83AB+18.70AB\ (A-B)\quad \text{(Eq. 1)}$$

In which Y was the response of foam stability (%); A was the fraction of coconut oil; B was fraction of palm oil and AB was an interaction between coconut oil and palm oil. From equation 1, it is known that form stability was influenced by components of coconut oil, paln oil and its interactions in which they contobuted to positive effect, i.e. the increased levels of coconut oil, palm oil, and its interaction would increase form stability.

Viscocity evaluation of BLD proved that the higher of palm oil used, the more viscous of BLD obtained. Oleic and palmitic acids present in palm oil could increase the viscosity of BLD. In contrast, coconut oil contained lower oleic and palmitic acids so that the consistency of BLD tend to become liquid [16]. ANAVA statistics test showed that there was significantly different (P<0.05) among the five formulas evaluated. Using LSD, the equation to describe viscosity was as in Eq. 2.

$$Y = 19.79A+109.79B-142.87AB+174.24AB\ (A-B)\quad \text{(Eq.2)}$$

In which Y was the response of foam stability (%); A was the fraction of coconut oil; B was the fraction of palm oil and AB was an interaction between coconut oil and palm oil. Eq. 2 indicated that palm oil and coconut oil affected BLD, in which palm oil (coefficient of 109.79) was more affecting toward viscosity of BLD than coconut oil (coefficient of 19.79). The presence of palmitic acid in palm oil having saturated chain fatty acid contributed to the hardness of BLD [12]. An interaction between coconut oil and palm oil had a negative coefficient, meaning that the interaction between coconut oil and palm oil reduced the viscocity of BLD.

The density of BLD formula I-V ranged in 0.75-1.02 g/ml. According to Indonesian National Standard SNI 4085: 2017, the density of liquid shop was in the range of 1.01-1.10 g/ml [15]. ANAVA test showed no significant difference in density among formulas (I-V) evaluated (P>0.05). Using SLD, Eq. 3 described response of viscosity with variables of the fraction of coconut oil (A), the fraction of palm oil (B) and interaction between coconut oil and palm oil (AB).

$$Y= 1.04A+0.93B-0.48AB\quad \text{(Eq. 3)}$$

Eq. 3 indicated that viscocity of BLD was affected by palm oil, coconut oil, and its interaction. Both oils positivily affected (increased) viscocity, while the interaction of coconut oil and palm oil affected (decreased) viscocity negatively.

pH is one of the important factors in making liquid detergent due to its application in the skin. Human skin typically has pH 4.5-7.0, and according to Indonesian National Standard, pH value of liquid detergent is in the range of 8.0-11.0. Formulas I-V of BLD has pH values in the range of 9.89-10.45. Van der Valk et al. reported no evidence that pH of cleansers could be related with skin irritation. Based on these pH studies, the five formulations should not be problematic in terms of causing skin irritation or poor skin compatibility [17]. ANAVA statistics test showed that there was no significant different (P>0.05) in pH values among the five formulas evaluated. Using LSD, the equation to describe pH value (Y) was as in Eq. 4.

$$Y = 10.11A+10.39B-1.05AB\quad \text{(Eq. 4)}$$

Coconut oil (A) and palm oil (B) contributed pH value positively, but its interaction (AB) affected pH values negatively. Fig. 2 exhibited SLD response of pH.

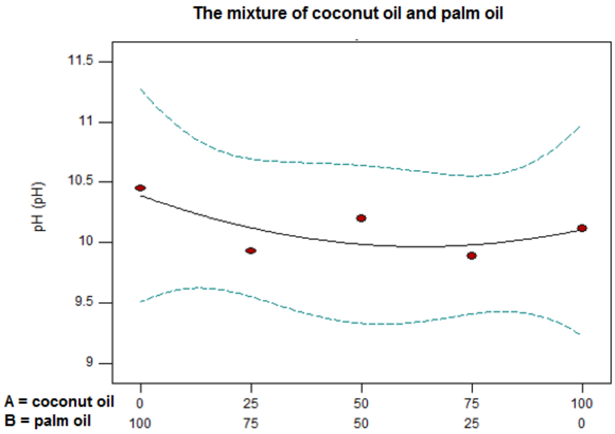

Fig. 2: The pH profiles of bentonite liquid detergent using simplex lattice design. x = proportion of coconut oil and palm oil; y = response of pH

Free alkali calculated as an amount of NaOH is one of the quality requirements in liquid detergent, with a maximum allowable value of 0.1%. All formulas have no free alkali; therefore formula I-V fitted the requirement. Indonesian National Standard SNI 4085: 2017, required that liquid detergent have total active components (total fatty acids) acting as cleansing agent minimal 15%. The high percentage of total fatty acids in liquid detergent indicated good quality which makes liquid detergent stable after being used for certain periods [17]. ANAVA test showed no significant difference in terms of total fatty acids among formulas tested. Based on Eq. 5, the composition of coconut oil (A), palm oil (B) and its interactions (AB) increased the levels of total fatty acids (Y).

$$Y= 4.11A+2.85B+4.33AB+14.21AB\ (A-B)\quad \text{(Eq. 5)}$$

Fig. 3 showed SLD response of total fatty acids using the different proportion of coconut oil and palm oil.

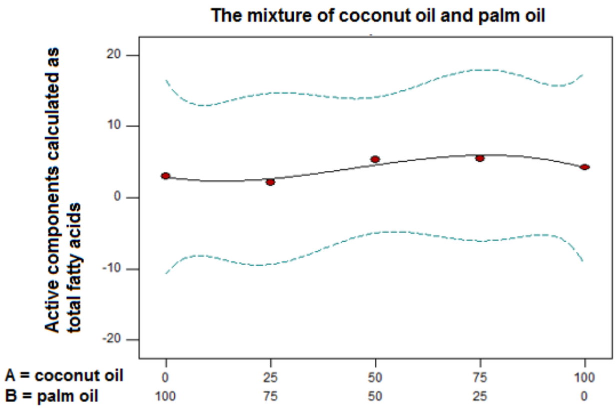

Fig. 3: The active components of bentonite liquid detergent using simplex lattice design. x = proportion of coconut oil and palm oil; y = response of active components

The optimum formula of BLD was obtained from the optimization results using SLD based on physicochemical properties including foam power, foam stability, viscocity, pH and active components as total fatty acids. The varied combination of coconut oil and palm oil affected the physicochemical properties of BLD. All responses wer not significantly different except viscocity. The parameters used for determination of optimum formula could be seen in table 2. Fig. 4 showed the optimization curve of BLD formula indicating the desirability value of 0.826 with the composition of coconut oil 95.1% and palm oil 4.9%. The desired response from optimum formula was BLD with foam power of 6.52 cm, foam stability of 81.74%, the viscosity of 24.87 poise, the density of 1,01 g/ml, pH of 10.07 and active component of 4.84%.

Verification of optimum formula of BLD

Verification of optimum formula of BLD was performed to compare the optimization results obtained using an experimental design of SLD and those obtained using actual laboratory experiment. The statistics test of one simple t-test was used to know whether there is a significant difference in terms of responses of formula using experimental design and actual laboratory-prepared formula. Table 3 compiled the output of one sample t-test which indicated that foam power, foam stability, density and pH are not significantly different (P>0.05), while viscocity and activite components were statistically different (P<0.05).

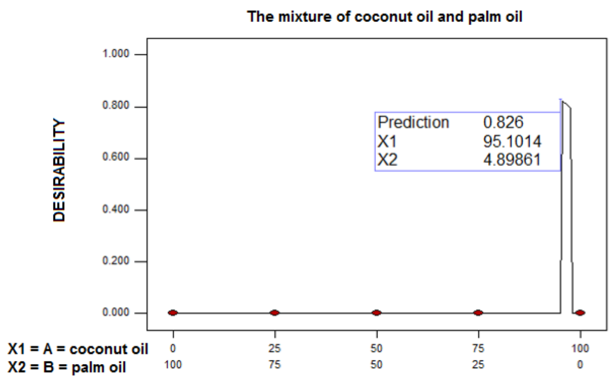

Fig. 4: The optimization curve of bentonite liquid detergent formula with a desirability value of 0.826. X1 and X2 are a composition of coconut oil and palm oil accounting of 95.1% and 4.9%, respectively

Table 2: The values and response weight used for formula optimization of bentonite liquid detergent

Response	Objective	Minimum value	Maximum value	Desirability
Foam power	Maximum	4.83 cm	6.87 cm	5
Foam stability	In range	81.05%	91.73%	3
Viscocity	In range	20 poise	110 poise	3
Density	In range	1.01 g/ml	1.10 g/ml	3
pH	In range	8	11	3
Active components (Total fatty acids)	Maximum	2.13 %	5.42 %	5

Table 3: The output of one sample t-test of physicochemical properties between formula obtained from experimental design and from prepared laboratory

Physico-chemical properties	Prediction by experimental design	The value from prepared laboratory	P value
Foam power	6.52	5.70	0.033
Foam stability	81.74	83.09	0.375
Viscocity	27.87	17.67	0.001
Density	1.01	0.94	0.150
pH	10.07	10.06	0.874
Active components (Total fatty acids)	4.84	2.13	0.005

CONCLUSION

The experimental design using simplex lattice design has been used for optimization of bentonite liquid detergent (BLD). The optimum concentration of bentonite was 6.4% with the percentage of cocount oil and palm oil of 95.1% and 4.9%, respectively. There was no significant difference of foam power, foam stability, and pH between optimum formula BCD predicted using SLD and that actually formulated based on one sample t-tst. BLD can be used as innovative means for cleansing extreme (*mughalladzah*) najs.

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AUTHORS CONTRIBUTIONS

DSW and M performed research activities and prepared manuscript. M and AR designed research, analyzed data, and made critical thinking on the manuscript.

CONFLICTS OF INTERESTS

All authors have none to declare

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