Valorization of Red Ginger Hydrodistillation Wastes as Foot Sanitizers

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Abstract

Red ginger is known as one of the medicinal and aromatic plants that have high value in the form of essential oils. The distillation of red ginger essential oils required large amounts of water, and then most of the water was disposed of as wastewater. This wastewater is known as hydrosol, which still contains a small amount of essential oils and secondary metabolites such as zingiberene and geraniol. These compounds were considered to have the potential to be utilized in cosmetic products such as foot sanitizers. The aim of this study was to evaluate a foot sanitizer product based on red ginger hydrosol that combined with coffee extracts based on its antibacterial activity against Staphylococcus aureus and the physical characteristics of the product. Foot sanitizer was produced in three formulations with different concentration (in percentage) of red ginger hydrosols and coffee extracts consist of F1 (45:25), F2 (35:35), and F3 (25:45). The results showed the F3 formulation provided the largest antibacterial inhibition (2.89 \pm 0.60 mm) compared to other formulations. The F3 formulation has a physical characteristic such as coffee scent aroma, a dark brown color, a pH of 5.5, a density of 960.73 kg/m³ and has a weak antibacterial activity. Based on the comparison results with previous studies, the utilization of coffee extracts in red ginger hydrosols-based foot sanitizer needs to be substituted with other natural extracts, especially having a strong antibacterial activity. It should be carried out to increase the potential value of valorizing red ginger hydrosols as foot sanitizer.

Keywords: Coffee Extracts, Red Ginger Hydrosols, Staphylococcus aureus, Zingiber officinale

INTRODUCTION

Red ginger (*Zingiber officibale*) is known as herbal and traditional medicine by Indonesian communities. The aroma, flavor, and pungency of red ginger provided various benefits and properties for related fields, thereby enhancing its value. According to Chung *et al.* (2021), the main bioactive metabolites of red ginger consist of of *zingiberene* (31.8%), *sesquiphellandrene* (13.0%), and *ar-curcumene* (7.6%). Based on gas chromatography analysis, several monoterpene compounds, including *neral*, *cineol*, *borneol*, *β*- geraniol, and β -linalool have been found on red gingers (Akshitha et al., 2020). These compounds are used as aromatic stimulants by related industries in the form of extracts, oleoresins, and essential oils (Alighiri et al., 2017).

The essential oils of red ginger have been highly valued, and their utility has increased. It occurred because red ginger essential oils (RGEO) are widely used in the cosmetics, food, aromatherapy, and pharmaceuticals industries (Marwati et al., 2021). The value of RGEO is known to be the highest of any form of red ginger derivative. According to Azalia *et al.* (2020), RGEO is valued at 500-600 thousand Rupiah per kg (approximately US\$40), which is higher than red ginger oleoresins, which are 110 thousand Rupiah per kg. Sinkona Indonesia Lestari (SIL) is one of the essential oil companies in Indonesia that is capable of producing RGEO on a large scale, with its capacity reaching 850 kg of red ginger per production batch. A large amount of RGEO production would necessitate a large amount of hydrodistillation water for the process; approximately 1,500 to 2,000 L, which would then be converted to wastewater in the amount of 1,000 L.

The wastewater resulting from the hydrodistillation of red ginger is known as "hydrosol", and it still contains various bioactive metabolites. Shafie et al. (2022) stated the hydrosols can be classified as hydrodistillation byproducts processes because the biomass still allows overflow of water to dissolve essential oils and other bioactive metabolites up to 1 g/L. The current trend shows hydrosols are very popular and attractive to utilize as natural and inexpensive flavoring additives and are widely used as cosmetic ingredients. It suggests that waste red ginger hydrosols (RGHs) from SIL have a high potential for valorization, particularly in cosmetic and personal care products such as foot sanitizer (Siregar, 2020).

Foot sanitizer products were developed in response to problems with sweaty feet. Balfas and Rahmawati (2022) stated the feet sweated as a result of the long time feet were covered; this could be stimulating microbial metabolisms and causing an unpleasant odor. One of the bacteria that causes smelly feet is the Staphylococcus bacterial genus, such as Staphylococcus aureus, which produces isovaleric acids in human skin (Tiran and Nastiti, 2014). By valorizing RGHs, Staphylococcus bacteria could be inhibited in their growth and metabolic activity (Ali et al., 2013). In addition, according to Purnama et al. (2022), the addition of coffee extracts to foot sanitizer products could increase antibacterial activity due to the presence of simple phenolics such as chlorogenic acids and caffeic acids.

Recent studies have shown the development of foot sanitizer products such as antibacterial soaps, powders, antiperspirants, and creams. The availability of these products was limited, they were unreachable by communities, and they had high prices. According to Balfas and Rahmawati (2022), producing foot sanitizer in spray form would be preferred by communities because it is easy to use and dries quickly. In addition, foot sanitizer in spray would reduce production costs and product prices. It could increase the potential for valorizing RGHs as foot sanitizers when combined with coffee extracts. The aims of this study were to evaluate foot sanitizer sprays made from RGHs and coffee extracts based on their antibacterial activities against *Staphylococcus aureus* and the physical characteristics of the products.

METHODS

RGHs, arabican coffee powders, and a bacterial strain of *Staphylococcus aureus* obtained from SIL were the main materials. Chemical reagents used consisted of distilled water and 70% of ethanol (technical grade), Tryptic Soya Agar (TSA), propylene glycol, and phenoxyethanol, which were purcased from Merck, Singapore.

Maceration for Coffee Extraction

The extraction technique was modified slightly from the procedure described by Dewantoro *et al.* (2022). The coffee powder was sieved to obtain particle sizes between 40 and 60 mesh and then soaked in 70% of ethanol with a 1:10 (w/v) ratio. The maceration was conducted for 5 days with periodic agitation every 24 hours. Extraction results were filtrated and separated before evaporation to obtain yields as $6.11\pm0.46\%$ (w/w). The coffee extracts were dissolved in 70% of ethanol to obtain an extracts mixture concentration of 10% (w/v).

Formulation of Foot Sanitizer

Foot sanitizer spray was produced by following the procedure and formulation from Santoso & Riyanta (2019) with a slight modification. The formulation variants and modifications were applied in this study are showed in Table 1.

Physical Characteristics Analysis

The product characteristics of foot sanitizer spray were determined by following procedure of Santoso & Riyanta (2019), which consisted of aroma, form, colors, sensation, pH, and density.

Antibacterial Activity Assay

Antibacterial activity carried out to evaluate foot sanitizer spray followed the procedure of Hossain *et al.* (2019), towards *Staphylococcus aureus*. Following the quantified inhibition zone of antibacterial activity, Dewatisari *et al.* (2021) proposed categories of weak (\leq 5 mm), moderate (5 to 10 mm), strong (10 to 20 mm), and very strong (\geq 20 mm).

Statistical Analysis

The collected data was analyzed using oneway ANOVA, followed by the Duncan Multiple Range Test (DMRT), which was included in IBM® SPPS Statistic 26.

RESULTS AND DISCUSSION

Physical Characteristics of Foot Sanitizer

The addition of coffee extracts to RGHsbased foot sanitizer affected the physical characteristics of the products. Table 2 shows the physical characteristics of products that consist of aroma, form, color, sense, pH, and density. Coffee extracts provided additional aroma and color to products, presenting a coffee scent and brown colors due to their higher concentration than RGHs. Based on Shafie *et al.* (2022), hydrosols have a maximum concentration of bioactive metabolites of 1 g/L, which is equivalent to 0.1% (w/v), which is smaller than the dissolved coffee extracts at 10% (w/v). In addition, a higher percentage of coffee extracts in the products increased the intensity of the brown color that is specific to coffee.

The high level of ethanol in foot sanitizer affected the form, sensation, and density of the products. Because more than half of the products formulations used in this study were aqueous. Moreover, the high ethanol content has proven to reducing the density of each formulation, which had a high content of coffee extracts, from 993.68 to 960.73 kg/m³. The cold sensation caused by the products was caused by the exothermic reaction ethanol, which evaporates when it comes into contact with human skin (Atiku et al., 2014). This phenomenon was observed in the study conducted by Santoso & Riyanta (2019), which produced foot sanitizer from coffee and ginger extracts.

Materials —	Formulation (in %)			F
	F1	F2	F3	Function
RGHs	45	35	25	
Coffee Extracts	25	35	45	
Propylene Glycol	15	15	15	
Phenoxyethanol	1	1	1	
70% of Ethanol	Ad. 50	Ad. 50	Ad. 50	

Table 1. Foot sanitizer formulation

Tabel 2. Physical characteristics of RGHs based foot sanitizer spray with coffee extracts addition

Characteristics —	Formulation				
	F0 ^a	F1	F2	F3	
Aroma	Tidak Berbau	Coffee scent	Coffee scent	Coffee scent	
Form	Aqueous	Aqueous	Aqueous	Aqueous	
Color	Transparant	Brown	Brown	Dark Brown	
Sensaion	Cold	Cold	Cold	Cold	
рН	6.45±0.03	5.18 ± 0.05	5.37±0.02	5.54±0.02	
Density ^b	993.68±1.43	985.50±3.10	976.13±0.29	960.73±4.22	

^aF0 was product formulation without coffee extracts addition; ^bDensity units expressed in kg/m³.

The foot sanitizer that was produced in this study has a pH value in the range of 5.18 to 5.54. According to Riyanta & Febriyanti (2018), the recommended pH range for topical products in accordance with the regulation of the Indonesian Ministry of Health was 5 to 7. The pH of human skin was in the range of 4.5 to 6.5, which indicates the results of this study are safe and appropriate (Balfas & Rahmawati, 2022). Besides that, the pH value of coffee combined formulations (F1, F2, and F3) was decreased compared to the F0 formulation is due to the increased chlorogenic and caffeic acids in the products.

Antibacterial Activities of Foot Sanitizers

The antibacterial activities of RHGs-based foot sanitizers provided weak activity against *Staphylococcus aureus*. The inhibition zone for each formulation is shown in Table 3, which is 2.03 to 2.89 mm. These ranges were higher than the F0 formulation with no addition of coffee extracts. The addition of coffee extract indicated an improvement in the antibacterial activities of products against *Staphylococcus aureus*, the bacterial cause of smelly feet.

Bioactive metabolite compounds in RGHs and coffee extracts have an important role in increasing antibacterial activity. The essential oil content in RGHs was known to disrupt bacterial cell membranes, resulting in an increase in osmotic pressure and cell turgor (Al-Harrasi et al., 2022). This could disrupt the cellular processes of bacterial cells, such as decreasing metabolic activity. Moreover, the addition of ginger extracts to the products has been shown to increase antibacterial activity due to the presence of polyphenolic compounds. According to Baenas et al. (2018), the -OH of the hydroxyl group in polyphenols could be used to neutralize toxins from bacteria and disrupt the main structure of bacteria in the form of proteins and lipids. Based on the antibacterial mechanisms of the two types of bioactive metabolites, it was necessary to add coffee extracts to the foot sanitizer products in this study.

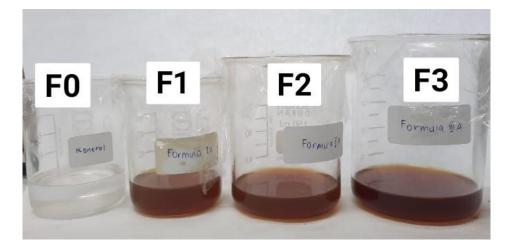


Figure 1. Foot sanitizers formulation results

Table 3. Antibacterial activities of foot sanitizers

Formulations	Inhibtion Zone* (mm)	Categories
FO	0.87±0.10 ^a	Weak
F1	2.03 ± 0.30^{b}	Weak
F2	2.24±0.19 ^{bc}	Weak
F3	$2.89 \pm 0.60^{\circ}$	Weak

*The difference symbols indicated a significant difference based on DMRT test (α = 95%)

Comparison of This Study Results and Recent Relevant Studies

Table 4 shows a comparison between the results of previous studies and the findings in this study. The valorization of RGHs for producing foot sanitizer spray has advantages over previous studies because it utilizes abundant hyrodistilled wastewater. The results of this study showed weak antibacterial activity against the bacteria that cause foot odor because the utilization of coffee as an extratc was considered inappropriate. The studies from Santoso & Riyanta (2019) and Ashfia *et al.* (2019) showed, respectively, that the utilization of coffee extracts provided weak to moderate antibacterial activity against *Staphylococcus epidermidis.* This indicated that the addition of coffee extracts, especially in this study, needs to be substituted with other types of natural extracts in order to enhance the potential of RGHs valorization products.

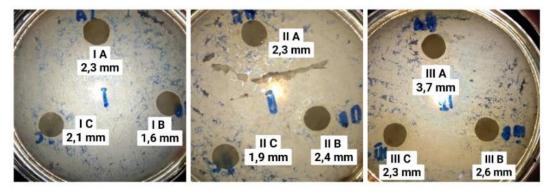


Figure 2. Antibacterial activities based on inhibition zone towards *Staphylococcus aureus*.

Table 4. Comparison with	previous studies towards	sanitizer products
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		Antibacterial Activities				
Bioactive Materials	pH*	Bacterial Strain	Inhibition Zone (mm)	Categories	References	
Rosella Extracts	4.0	Staphylococcus aureus	12.4	Strong	Adiansyah <i>et al.</i> (2018)	
Coffee Extracts and Ginger Extracts	6,0	Staphylococcus epidermidis	1.9	Weak	Santoso & Riyanta (2019)	
Lime Extracts and Coffee Ground Extracts	5.0	Staphylococcus epidermidis	6.0	Moderate	Ashfia <i>et al. (</i> 2019)	
Mangrove Leaves Extracts	n.d.	Staphylococcus aureus	5.7	Moderate	Lestari <i>et al</i> . (2020)	
Papaya Leaves Extracts	6.1	Staphylococcus epidermidis	10.1	Strong	Leny <i>et al</i> . (2020)	
Lime Peels Extracts	5.2	Bacilus subtilis	13.2	Strong	Ulfa <i>et al</i> . (2020)	
<i>Plantago major</i> Leaves Extracts and Betel Extracts	7.0	Staphylococcus epidermidis	5.0	Weak	Sinaga <i>et al</i> . (2020)	
Orange Essential Oils and Green Tea Extracts	n.d.	, Staphylococcus epidermidis	20.0	Very Strong	Veranita <i>et al</i> . (2021)	
Lime Leaves Extracts	5.3	, Staphylococcus epidermidis	14.3	Strong	Marwarni & Dalimunthe (2022)	
RGHs and Coffee Extracts	5.5	Staphylococcus aureus	2.9	Weak	This Study	

*n.d. mean not determined.

The addition of natural extracts that have deodorizing properties to replace coffee extracts was considered important and should be carried out. Based on Table 4, the utilization of orange and lemon extracts could increase the antibacterial activity of sanitizer spray products. The utilization of lemon leaves and peels by Sinaga et al. (2020) and Marwarni & Dalimunthe (2022) gave an inhibition zone of 14.3 mm and 13.2 mm, respectively, which belonged to a strong activity. Veranita et al. (2021) produced a deodorant spray using combination of calamansi orange essential oils and green tea extracts known to provide very strong antibacterial activity against Staphylococcus epidermidis. Thus, the utilization of orange and lemon extracts could be used as an alternative to substituting coffee extracts in the valorization of RGHs which function to eliminate foot odor.

CONCLUSION

The valorization of RGHs combined with coffee extracts affected the physical characteristics and antibacteral activity of foot sanitizer products. The best results were given by the F3 formulation, which has a higher concentration of coffee extracts (45%) than RGHs (25%). This formulation has a pH of 5.54±0.02 and a density of 960.73±4.22 kg/m³ and provided weak antibacterial activity against Staphylococcus aureus. It has a coffee scent aroma, dark brown color, aqueous form, and cold sensation. Recommendations that could be given based on the results of this study were that to enhance the potential of RGHs valroazation products, it was necessary to utilize alternative natural extracts (such as those from oranges and lemons) to subtitute for coffee extracts as a deodorizer and increase the antibacterial activities of the products.

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