The Effect of Clove Leaves Essential Oil Addition on Physicochemical and Sensory Characteristics of Milk Chocolate Bar

Danar Praseptiangga\textsuperscript{a,*}, Agitya Renanda Qomaruzzaman\textsuperscript{a}, Godras Jati Manuhara\textsuperscript{a}

\textsuperscript{a}Department of Food Science and Technology, Faculty of Agriculture, Universitas Sebelas Maret, Surakarta, 57126, Central Java, Indonesia

Corresponding author: \textsuperscript{*}dpraseptiangga@staff.uns.ac.id

Abstract—As one of the most prominent cocoa and clove producers globally, Indonesia needs to develop a chocolate signature with unique characteristics due to the international cravings and high consumption of chocolate compared to other confectionery products. There are three main types of chocolate—based on their compositions: dark chocolate, milk chocolate, and white chocolate. However, milk chocolate has been discovered to be a much-preferred type due to its taste and flavor, but its innovative development is limited despite consumers’ interest. Moreover, clove leaves have been less utilized despite their ability to be further processed to the essential oil, which possesses a potency of health benefits such as antioxidant activity due to its main bioactive compounds and its usefulness a flavor in food. This study was, therefore, aimed to determine the effects of adding several concentrations of clove leaves essential oil at 0.1\% (F1), 0.2\% (F2), and 0.3\% (F3) on the physicochemical and sensory characteristics of milk chocolate bars as well as to define the best formula to achieve the optimum product. The results showed different trends at each concentration, while the best formula was recorded at the addition of 0.2\% (F2) clove leaves essential oil to the milk chocolate bar, and the panelists further confirmed this.

Keywords—Cocoa; milk chocolate bar; clove leaves essential oil; sensory characteristics; physicochemical characteristics.

I. INTRODUCTION

Chocolate is the most famous confectionery product available in the world, and its popularity is associated with the bioactive compounds which potentially provide beneficial health benefits reported to be contained in its main ingredient, cocoa \textit{(Theobroma cacao L.)} \cite{1}, \cite{2}. Moreover, the cocoa processed into chocolate or powder has certain polyphenolic compounds mostly composed of flavonoids reported to contain antioxidants \cite{1}, \cite{2}.

Meanwhile, milk chocolate is one of the favored derived products compared to dark and white products due to its sweet taste, texture, and soft flavor \cite{3}. Consumers prefer the absence of bitter taste but the milk and sugar added could decrease the effectiveness of bioactive compounds contained in the chocolate \cite{4}. Milk chocolate is a complex rheological system made by mixing cocoa, milk powder, and sugar as solid particles that are dispersed in the fat phase (cocoa butter) \cite{5}, \cite{6}, \cite{7}, \cite{8}. It is formulated to be softer than dark chocolate and has a creamier taste, making it a much-preferred type of chocolate, particularly by Indonesian consumers \cite{7}.

There has been an increase in bioactive substances investigations in recent times, mainly due to their use in food processing and their potential health benefits \cite{9}, \cite{10}. Meanwhile, the use of medicinal plants and spices in the formulations of functional food for traditional enhancement of health or healing diseases has been reported to be promising \cite{10}. There is also growing interest in utilizing essential oils that are derived from plants and spices for food and pharmaceutical applications. Essential oils are volatile hydrophobic liquids extracted from plants comprised of various groups of compounds such as phenols, aldehydes, ketones, mono, and sesquiterpenes that are reported to be responsible for various biological activities.

The essential oils extracted from the medicinal plants and/or spices such as cinnamon, clove, basil, oregano, and thyme show significant antioxidant activities due to major constituents’ presence such as cinnamaldehyde, carvacrol, eugenol, or thymol, etc. The antioxidant potential of essential oils mainly depends on their chemical compositions, and phenolic compounds and other secondary metabolites are responsible for the substantial antioxidant activity of essential oils \cite{11}, \cite{12}, \cite{13}.

Clove \textit{(Syzygium aromaticum)}, which is one of the spices native to Maluku islands in east Indonesia, has been applied
The ball mill was set at a speed of 90 rpm and purchased at CV. Orizho Indonesia (Bantul, Indonesia). The essential oil of clove leaves (Syzygium aromaticum) was the main material used in producing milk chocolate bars. Clove leaves were added to milk chocolate bars by using a scraper and a chocolate knife at room temperature of 26–28°C for 5 minutes until the dough temperature was stable at ± 3°C. The dough was then poured into a wooden mold and placed in a refrigerator at 5°C for 2 hours. The dough was then de-molded and packaged in aluminum foil, and stored at room temperature until further analysis.

Eugenol has been reported to be anti-inflammatory and antioxidant as the main bioactive compound [15], [16], [17]. Eugenol and 17,4-dihydroxy-17β-estradiol [20] are considered to be flavor due to its distinctive aroma and a hot and spicy taste [17].

The samples' color attributes were analyzed using a universal testing machine (UTM) [3]. It was used in determining the hue angle and the total color difference (TCD). Moreover, the analysis of the texture was conducted using an Agilent 6890N GC-MS (Gas Chromatography-Mass Spectrometry) with a split ratio of 1/10 and helium, a carrier gas used at 1.5 mL per minute. The GC injection method slightly modified from the one used by [21].

Moreover, the GC-MS injection method slightly modified from the one used by [21]. The samples' active compounds were identified by comparing the mass spectra of each peak with those of authentic samples in a mass spectra library. The result was used in determining the percentage (%) of water in the sample (w/w) [22]. The samples' active compounds were identified by comparing the mass spectra of each peak with those of authentic samples in a mass spectra library. The result was used in determining the percentage (%) of water in the sample (w/w) [22].

The samples were analyzed using a UV-Vis spectrophotometer (Agilent Technologies, USA). Methanol was used as the solvent for the extraction and the method applied for the extraction was vacuum extraction. A 2% NaCl solution was added to the milk chocolate bars and the mixture was heated and stirred for 20 minutes. The extract was then filtered, and the absorbance at λ=750 nm was measured using a UV-Vis spectrophotometer. The absorbance was used to calculate the total phenolic content.

The samples were analyzed using a Karl Fischer method and a Moisture Meter CA-200 Karl Fischer. The samples were analyzed using a Karl Fischer method and a Moisture Meter CA-200 Karl Fischer. The samples were analyzed using a Karl Fischer method and a Moisture Meter CA-200 Karl Fischer. The samples were analyzed using a Karl Fischer method and a Moisture Meter CA-200 Karl Fischer. The samples were analyzed using a Karl Fischer method and a Moisture Meter CA-200 Karl Fischer.
expressed as content (%) based on the area percentage obtained.

**D. Data Analysis**

One-way ANOVA with a significance level of $\alpha = 5\%$ was used in analyzing the data. Meanwhile, Duncan multiple range tests were applied for further analysis in the case of significant differences in the results.

### III. RESULTS AND DISCUSSION

**A. Sensory Characteristics**

Human panelists’ sensory perception related to the thresholds in determining attributes and the variance in individual sensory response experimental design were used to measure the food products’ sensory characteristics and acceptability. Table 1 shows the variation in the clove leaves' concentrations do not affect the panelists’ preference for a particular color.

<table>
<thead>
<tr>
<th>TABLE I</th>
<th>SENSORY CHARACTERISTICS OF MILK CHOCOLATE BAR WITH ADDITION OF CLOVE LEAVES ESSENTIAL OIL</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample</td>
<td>Parameter</td>
</tr>
<tr>
<td>F1 (0.1%)</td>
<td>4.73±0.45</td>
</tr>
<tr>
<td>F2 (0.2%)</td>
<td>4.67±0.48</td>
</tr>
<tr>
<td>F3 (0.3%)</td>
<td>4.63±0.61</td>
</tr>
</tbody>
</table>

This means the color is an essential sensory attribute and one factor influencing any product's appearance. It is also associated with food quality, with its brightness in some foods being a benchmark for quality [25]. The milk chocolate bar has a light brown color compared to the dark brown usually found with dark chocolate due to paste dominance. A dairy component is also attached to the bar's sweetness and creaminess [7]. Furthermore, the interaction between reducing sugars and the amino acids contained in milk causes browning or Maillard reactions, and this also contributes to the formation of the color [26]. The addition of essential oils had no significant effect on the color attribute of the milk chocolate bar [8], [27].

The aroma is also one of the factors to measure food product quality. The aroma of chocolate is due to the very complex chemical reactions including amino acids, sugars, and compounds of flavonoids during roasting as well as the degradation of the Strecker amino acid derived from aldehyde volatiles [3], [7]. There was a decrease in panelist's preference for milk chocolate bars along with the concentration of essential oil added. Table 1 shows that the preference for aroma at 0.1% or F1 with 4.43 ± 0.77 was not significantly different from 0.2% or F2 with 4.30 ± 0.79, which confused their aroma by the panelists. Meanwhile, the 0.3% or F3 sample had the lowest preference with 3.90 ± 0.88 compared to the other two, and this was allegedly due to the stronger pungent flavor of the essential oil. The panelists were not favored, and its anesthetic effect causes numbness on the tongue when consumed too much [28].

Aroma is another element with great influence on the level of consumers’ preference for chocolate. Most of those believed to be good usually have a light brown to dark brown uniform and a shiny or glossy surface [8], [26]. However, the milk chocolate bar’s appearance is closely related to its brightness or glossiness, which is an optical phenomenon associated with the ability of a surface to reflect the light that hits it. The results showed appearance is not an important parameter to determine the quality of the chocolate [29]. This was confirmed by the information on Table 1 where no significant difference was observed with the preference of the panelists based on appearance. Meanwhile, the tempering process is believed to have more influence on the milk chocolate bar’s appearance. Over-tempering caused a significant increase in the product’s hardness, stickiness with reduced gloss, and surface darkening while under-tempering induced fat bloom with consequential quality defects on texture, color, and surface gloss [8], [30]. Therefore, chocolate’s shiny or glossy surface is an indicator of the tempering process’s success, thereby indicating its quality [26].

The overall parameters, including the sensory attributes of color, aroma, taste, and appearance, were also jointly measured, and the panelists accepted all the three formula of milk chocolate bars developed in this study. However, a decrease in their preference for the overall parameters with an increase in the clove concentration leaves essential oil added. This was observed because there was no significant difference with the color and appearance but a decrease in line with the increase in concentration was recorded with aroma and taste. The overall values obtained for F1, F2, and F3 were $4.43 \pm 0.68$, $4.10^{+0.80}$, and $3.87 \pm 0.86$, respectively. The milk chocolate bars were strongly influenced by the two parameters of taste and aroma, which is reflected in the overall results.

**B. Physicochemical Characteristics**

The measurement of color in this study was intended to provide objective information on the difference in milk chocolate bars due to the addition of clove leaves essential oil. The L* value indicates the sample's brightness level,
which ranges from black to white (0-100) [31]. The samples with essential oil were observed to have significantly different lightness from each other, but 0.2% or F2 was not substantially varied from the control treatment. Moreover, the $a^*$ value indicates the chromatic color of red ($a^*$ positive, +60) to green ($a^*$ negative, -60) while $b^*$ value indicates the chromatic color of yellow ($b^*$ positive, +60) to blue ($b^*$ negative, -60) [31]. However, all the samples were observed to have positive $a^*$ and $b^*$, which means the milk chocolate bars color with and without the essential oil ranged between red and yellow. Table 2 shows the hue angle of the samples are between 18 and 54, thereby indicating the product color tends to be red [8]. Furthermore, the total color difference value (TCD) was calculated to determine the color difference between the treatment samples and control as well as the visibility of the variation. The TCD value, based on [31], can be <0.5 (invisible), 0.5-1.5 (slightly noticeable), 1.5-3.0 (noticeable), 3.0-6.0 (well visible), or >6.0 (great difference).

### TABLE II

**COLOUR AND TEXTURE MEASUREMENT OF MILK CHOCOLATE BAR WITH THE ADDITION OF CLOVE LEAVES ESSENTIAL OIL.**

<table>
<thead>
<tr>
<th>Sample</th>
<th>$L^*$</th>
<th>$a^*$</th>
<th>$b^*$</th>
<th>$\delta$Hue</th>
<th>TCD</th>
<th>Effect</th>
<th>Hardness (N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C (0%)</td>
<td>32.46±0.25</td>
<td>15.91±0.06</td>
<td>14.58±0.15</td>
<td>42.51±0.23</td>
<td>-</td>
<td>-</td>
<td>27.71±0.33</td>
</tr>
<tr>
<td>F1 (0.1%)</td>
<td>34.52±0.22</td>
<td>15.53±0.44</td>
<td>13.38±0.70</td>
<td>40.72±0.96</td>
<td>2.41</td>
<td>Noticeable</td>
<td>26.38±0.35</td>
</tr>
<tr>
<td>F2 (0.2%)</td>
<td>32.84±0.44</td>
<td>15.54±0.47</td>
<td>13.59±0.94</td>
<td>41.14±1.07</td>
<td>1.12</td>
<td>Slightly Noticeable</td>
<td>24.07±0.23</td>
</tr>
<tr>
<td>F3 (0.3%)</td>
<td>33.62±0.26</td>
<td>15.45±0.13</td>
<td>13.28±0.49</td>
<td>41.26±0.81</td>
<td>1.80</td>
<td>Noticeable</td>
<td>25.54±0.69</td>
</tr>
</tbody>
</table>

According to the information in Table 2, the addition of clove leaves essential oil presents a noticeable effect on the samples' color difference (F1 and F3) and a slightly noticeable effect for F2. Due to the closeness of the TCD values of the treated samples to the threshold value of 1.5, there was only a small difference that the panelists were unable to find a significant difference in the color during the sensory evaluation.

The texture is another important component in determining the quality of chocolate products. A smaller and better distribution of particles in chocolate usually leads to higher hardness [26], [30]. Table 2, therefore, shows a significant difference regarding hardness was induced by the addition of clove leaves essential oil to the milk chocolate bar and the sample. The highest value was recorded in control with 27.71 ± 0.33N followed by F1 with 26.38 ± 0.35N, and F3 with 25.54 ± 0.69N while a slight decrease was observed in the F2 with 24.07 ± 0.23N allegedly due to an uneven distribution of particles thought to be caused by a non-uniform tempering profile. It is important to note that the chocolate was tempered using hand with the combination of time and temperature found to be the most important part of the process to trigger crystals’ formation. Moreover, there was the influence of the refining, conching, and tempering on the milk chocolate bar. This is observed from several activities such as cooling, heating, and inverting the chocolate to ensure the formation of fat crystal $b_2$ (V form) with resistance to the changes in temperature [26], [30]. It has also been previously reported that under-tempering increases chocolate softness while over-tempering increases its hardness [8]. Meanwhile, an optimized low residence time (<6 minutes) was used in this study, and it is impossible to equal the crystal size and polymorphous size distribution produced with the use of a fully mature stabilized tempering method usually takes 12 minutes - 2 hours. This decline in hardness is related to the increase in the chocolate moisture content due to the addition of the essential oil. This water binds with the sugar to produce a chocolate with a gritty texture, reduced hardness, and high viscosity when melted [3].

Moisture content is one of the critical parameters in chocolate products and is required to be avoided as much as possible due to its ability to reduce quality. It can soften the texture [32] and accelerate the appearance of blooming on the chocolate's surface and these reduce its shelf life [26]. Table 3 shows an increase in the water content of milk chocolate bars with the lowest recorded in the control sample with 2.14 ± 0.03% while F1 had 2.30 ± 0.01%, F2 had 2.48 ± 0.08%, and F3 had 2.51 ± 0.01%.

### TABLE III

**MOISTURE CONTENT, TOTAL PHENOLIC CONTENT, AND ANTIOXIDANT ACTIVITY OF MILK CHOCOLATE BAR WITH ADDITION OF CLOVE LEAVES ESSENTIAL OIL.**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Moisture content (%)</th>
<th>Total phenolic (%)</th>
<th>IC$_{50}$ (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C (0%)</td>
<td>2.14±0.03</td>
<td>1.86±0.26</td>
<td>1434.31±30.20</td>
</tr>
<tr>
<td>F1 (0.1%)</td>
<td>2.30±0.01</td>
<td>2.16±0.27</td>
<td>1422.04±67.49</td>
</tr>
<tr>
<td>F2 (0.2%)</td>
<td>2.48±0.08</td>
<td>2.53±0.76</td>
<td>1157.90±27.83</td>
</tr>
<tr>
<td>F3 (0.3%)</td>
<td>2.51±0.01</td>
<td>2.61±0.66</td>
<td>1029.73±24.37</td>
</tr>
</tbody>
</table>

Table 3 shows that the control and F1 are significantly different from others. In general, the water content increased along with the concentration of the oil added. This was assumed to be due to the hydrophilic properties of the essential oil [33] and the presence of its volatile polar components [34]. An increase in moisture content was also found in the milk chocolate bar with an addition of cinnamon essential oil [8], with the value observed to have been increasing with the concentration, even though the variation was not significant. Moreover, the storage and transfer of chocolate also could contribute to the increase in the moisture content. This is in line with the explanations of Beckett [3] that several factors contribute to this increase and one of them is the transfer of the product from a cold to room temperature with high humidity. The condensation process causes the formation of condensed water on the chocolate bar's surface, which leads to an increment in the moisture content. Furthermore, milk as one of the main ingredients contains lactose crystals which are hygroscopic natural sugar and easily bind with water [26]. This is generally believed to be the reason for the higher moisture content in milk chocolate bars compared to dark bars [3].

Phenolic compounds have great potential in preventing diseases and serving as antioxidants [35]. The total content of phenolic compounds was determined in this study using
the Folin-Ciocalteu method because it is easy, rapid, and most used in cocoa and chocolate studies [1], [21]. Table 3 shows the total phenolic content increases with the concentration of essential oil. The control sample had the lowest at 1.86% ± 0.26 followed by F1 at 2.16% ± 0.27, F2 at 2.53% ± 0.76, and F3 at 2.61% ± 0.66. The control and F1 samples were observed to be significantly different from others and the trend was assumed to be due to the presence of eugenol, which is the main phenolic compound [36] in clove essential oil.

The antioxidant activity test conducted on the milk chocolate bar is presented as IC$_{50}$ value as shown in Table 3 and it shows the quantity of extract concentration required to inhibit 50% oxidation. There was a decrease in IC$_{50}$ as the essential oil was added, indicating the milk chocolate bar's stronger antioxidant activity. As the concentration of the essential oil increased, there was a need for the chocolate concentration to scavenge the 50% free radicals of DPPH. This, therefore, means the control sample has the highest IC$_{50}$ value of 1434.31 ± 30.20 ppm which is not significantly different from the 1422.04 ± 67.49 ppm recorded for F1 and this was followed by 1157.90± 27.83 ppm for F2 while the lowest was 1029.73 ± 24.37 at F3. The active compounds such as eugenol and β-caryophyllene in the clove essential oil which are classified as super-antioxidant and antioxidant, respectively, were observed to have caused the increment [37],[38].

Moreover, flavonoids and flavanols such as catechin and epicatechin are polyphenols in cocoa [3], and the methylxanthines are theobromine and caffeine in cocoa, are also antioxidants [39]. According to Molyneux’s classification [40], a super potent antioxidant has IC$_{50}$<50 ppm, strong is 50-100 ppm, the middle is 10-150 ppm, weak is 150-200 ppm, and super weak is >200 ppm. This, therefore, means all the samples are super weak antioxidants. Meanwhile, the values are not constant and have been observed to be varying based on several factors such as the condition, processing, or assay method of the materials [3],[21]. However, this study's findings are in line with the total phenolic and GC-MS evaluations in previous studies [1], [21], which demonstrated the antioxidant activity to be positively correlated with the total phenolic content.

The active compounds in the milk chocolate bar after the essential oil has been added were determined using GC-MS analysis, and Table 4 shows an increase in the percentage of eugenol in was in line with a higher concentration of the essential oil. Table 4 shows the relative content in F1 was 28.95% while F2 had 53.26% and F3 62.77%. Eugenol is a phenolic compound with several pharmacological activities, roles in the redox status, and applications in food [17]. It has also been reported to have several health benefits by serving as anti-inflammatory, antioxidant, antimicrobial, and anesthetic agents [17],[41]. This means the increase in the concentration of eugenol is expected to improve the milk chocolate bars' functionality for human health.

Further studies are, however, needed to understand its functionality comprehensively, β-Caryophyllene, which is another main compound found in clove essential oil, is a natural bicyclic sesquiterpenoid. It has been demonstrated to possess great potential to be applied for several pathological conditions. In vitro and in vivo (preclinical), many studies suggested the treatment with β-caryophyllene was able to improve the phenotype of animals used in modeling different inflammatory pathologies such as nervous system diseases [42]. The same trend with eugenol was observed to have increased with the concentration of essential oil with the relative content in F1 recorded to be 9.64% while F2 had 18.60%, and F3 25.06%.

<table>
<thead>
<tr>
<th>No.</th>
<th>Retention time</th>
<th>Compound</th>
<th>Content (%)</th>
<th>F1</th>
<th>F2</th>
<th>F3</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>13.95</td>
<td>Eugenol</td>
<td>-</td>
<td>28.95</td>
<td>53.26</td>
<td>62.77</td>
</tr>
<tr>
<td>2</td>
<td>14.54</td>
<td>Vanillin</td>
<td>47.01</td>
<td>16.69</td>
<td>7.87</td>
<td>2.05</td>
</tr>
<tr>
<td>3</td>
<td>14.82</td>
<td>β-caryophyllene</td>
<td>-</td>
<td>9.64</td>
<td>18.60</td>
<td>25.06</td>
</tr>
<tr>
<td>4</td>
<td>19.71</td>
<td>Caffeine</td>
<td>25.36</td>
<td>16.60</td>
<td>8.99</td>
<td>6.66</td>
</tr>
<tr>
<td>5</td>
<td>20.22</td>
<td>Theobromine</td>
<td>Trace</td>
<td>9.74</td>
<td>3.83</td>
<td>Trace</td>
</tr>
<tr>
<td>6</td>
<td>20.48</td>
<td>Pentadecanoic acid, methyl ester</td>
<td>7.36</td>
<td>5.26</td>
<td>1.77</td>
<td>1.20</td>
</tr>
<tr>
<td>7</td>
<td>21.10</td>
<td>Ethyl palmitate</td>
<td>Trace</td>
<td>1.75</td>
<td>0.61</td>
<td>0.51</td>
</tr>
<tr>
<td>8</td>
<td>22.11</td>
<td>10-octadecanoic acid, methyl ester</td>
<td>Trace</td>
<td>-</td>
<td>0.99</td>
<td>Trace</td>
</tr>
<tr>
<td>9</td>
<td>22.34</td>
<td>Heptadecanoic acid, methyl ester</td>
<td>Trace</td>
<td>1.06</td>
<td>Trace</td>
<td>Trace</td>
</tr>
<tr>
<td>10</td>
<td>22.66</td>
<td>Linoleic acid, ethyl ester</td>
<td>Trace</td>
<td>0.919</td>
<td>Trace</td>
<td>Trace</td>
</tr>
<tr>
<td>11</td>
<td>22.71</td>
<td>Ethyl oleate</td>
<td>10.39</td>
<td>2.10</td>
<td>2.00</td>
<td>1.76</td>
</tr>
<tr>
<td>12</td>
<td>29.67</td>
<td>Beta-tocopherol</td>
<td>9.88</td>
<td>6.11</td>
<td>3.06</td>
<td>Trace</td>
</tr>
</tbody>
</table>

| TABLE IV GC-MS DETECTION OF MILK CHOCOLATE BAR WITH ADDITION OF CLOVE LEAVES ESSENTIAL OIL |

Note: “Trace” shows the proportion of compounds is very small due to the dominance of other compounds (Similarity index used at least 80%)
IV. CONCLUSIONS

This study evaluated the effect of adding clove leaves essential oils on milk chocolate bars’ physiochemical properties and sensory characteristics by measuring panelist’s preferences. Sensory analysis showed a decrease in the preference for taste and aroma due to the addition of the essential oil and the overall parameters of the milk chocolate bars were highly influenced by these two parameters. Meanwhile, color, and appearance were observed not to have any significant difference as observed with the small variation noticeable in the color analysis’s TCD values. The physicochemical characterization showed that the decline in hardness is related to the increase in chocolate moisture content due to the essential oil added. Moreover, an increase was also discovered in the total phenolic content and antioxidant activity based on the increment in the active compound concentration derived from clove leaves essential oil. Based on all aspects of the tests, the best formula was found to be F2, which involves adding 0.2% of clove leaves essential oil to the milk chocolate bar to develop an Indonesian chocolate signature.

ACKNOWLEDGMENT

The authors appreciate and express sincere gratitude to the Ministry of Research and Technology (Ristek- BRIN), Republic of Indonesia, for the partially supported research grant scheme of World Class Research and Universitas Sebelas Maret PNB research grant.

REFERENCES


