Optimization of BCR Microwave from Fe Assessment in Sediment Material in The Gulf of Prigi

A R Wijaya¹ ¹*, I Oktaviana¹, S Wonorahardjo¹, Y Utomo¹, M Muntholib¹ H Daupor², Md. S Hossain³, E H Sanjaya⁴

¹Chemistry Department, Faculty of Mathematics and Natural Science, Universitas Negeri Malang (UM), Jln. Semarang No. 5 Malang
²Chemistry Major, Faculty of Science Technology and Agriculture, Yala Rajabhat University, Yala 95000, Thailand
³Department of Aquaculture, Faculty of Fisheries, Bangladesh Agricultural University (BAU), Mymensingh-2202, Bangladesh
⁴Department of Civil and Environmental Engineering, School of Engineering, Tohoku University, 6-6-04, Aramaki Aza Aoba Aoba-ku, Sendai, Miyagi 980-8579, Japan

*Corresponding author’s e-mail: anugrah.ricky.fmipa@um.ac.id

Abstract. BCR methods are known for leaching of heavy metals in sediment material. However, there is no optimization of modification BCR using a microwave in Fe in the Gulf of Prigi sediment. Thus in this paper, we attempted to use the variation of modified microwave power (10%, 25%, 50% watt) followed with contact time ranges (from 1 to 3 minutes). The results showed the fast method of BCR for leached Fe using a microwave with an optimized condition in the fraction 1 and fraction 2 with 50% watt at 2 minutes, fraction 3 (10% watt at 2 minutes) and fraction 4 (temperature used at 185 degrees Celsius at 10 minutes) in sediment materials. These investigated were also supported by characterization using SEM EDAX and AAS measurements of Fe in the filtrate sediment.

Keywords: BCR, leaching, sediment material, Fe, Prigi.

1. Introduction

Prigi is located in Tasikmadu Village, Watulimo District, Trenggalek, East Java. The coastal area of Prigi is often used by the community as settlement, tourism, sea transportation, and fish port. Trenggalek has the largest fish port in the southern coastal area of Java Island. Prigi port possibly becomes one of the sources of heavy metal contamination from its activities and from urbanized areas. Metals are toxic if they exceed quality standards and then affected human health. That metals are found in the contaminated environmental samples such as sediment, water, and corals [1–3]. Many researchers used the residue of marine products such as chitosan and geochemical of sediment fractions to reduce the level of metal contents [4–6].

Sediment can absorb a high level of metal contents comparing with water and marine organisms. Sediment is a recording of global environmental change [7], and then widely used in the analysis of heavy metal contamination in the environment. Considering one of the heavy metals in the sediment of the port area is Fe. The corrosion of marine ship and harbor piles can contribute to the presence of Fe in
seawater. Fe ions in seawater are divided as Fe$^{2+}$ (Ferrous) and Fe$^{3+}$ (Ferric) ions. Fe$^{2+}$ ions are more soluble and absorbed as micronutrients by marine plants, whereas Fe$^{3+}$ is present in sediments in the form of oxide or hydroxide [8]. Small amounts of iron are essential elements for the growth and development of an organism but the high level of Fe in the sediments causes poisoning and damage of the seawater food chain. In addition, the ability of high level of Fe ions to catalyze the formation of radicals can attack cellular macromolecules, increase cell death, damage of tissue and organ failure for human beings [6]. The BCR method is well known as the best method for selection of solvent usage, metallic mobility analysis, and a high recovery percentage. However, this BCR method still has a long time for leaching about 51 hours at 26 minutes. This causes the BCR method has a less efficient, and then many researchers modified the BCR method using ultrasonic waves as well as microwaves. Microwave BCR method possibly give advantages in terms of leaching time. However, the microwave BCR method has a disadvantage in the application of microwave temperature and its energy. This is due to the difference in bonding break energy in each of metals. Fe dissociation energy is also different in each of fraction in sediment. Optimization of energy and temperature is needed to determine how much power and time are used to produce energy proportional with dissociation of Fe in the chemical leaching, and then it is expected to obtain good results with the lowest possible energy usage. This study aims to (1) optimize of microwave power and time in Fe dissociation in the chemical leaching in sediment, (2) to determine the accuracy and precision of microwave BCR method in Fe leaching, (3) to measure of Fe concentration in each of sediment fractions using optimized BCR-Microwaves. Moreover, the advantages of this research are to identify the source and status of Fe contamination in sediment for supporting policies government to protect the environment.

2. Methods

2.1. Methods

2.1.1. Determination of Sampling Location
Geographical determination of sediment sampling was collected in the 3 main locations in the coastal of Prigi Port using GPS and then plotted into the map through Google maps (Table 1).

| Code of Sample | Position of sample | Sampling location |
|----------------|-------------------|------------------|
| S1             | 8°18'42.750"S   111°44'52.876"E | Recreation place of the coastal Prigi |
| S2             | 8°18'40.050"S   111°44'54.280"E | Nearly Residences |
| S3             | 8°18'37.548"S   111°44'54.103"E | Port of Prigi |

2.1.2. Sampling and Sample Preparation
Sediment was collected using grab sediment. Each sample was put into a plastic bag and labeled according to the code of the sampling location. Samples were put into an icebox and treated to the laboratory of chemistry. Sediment samples were separated from stones, sand, and corals. Samples were then dried for 3-4 days using an oven at 60 °C until the sample completely dried. The sample was weighed about 0.25 grams.
2.1.3. XRF analysis of sediment
The XRF (PANanalytical Type Minipal 4) analysis of powder sediment was performed in the Laboratory of Minerals and Advanced Materials at State University of Malang. We used this instrument for preliminary qualitative analysis of metal contents based on their detection and analysis of characteristic X-ray fluorescence in sediment.

2.1.4. Accuracy determination of power (energy) and microwave time on BCR Method
The accuracy study was conducted on JMS-1 referent samples obtained from Geochemical Survey of Japan with the following, as follows: 0.25-gram samples were put in a digestion vial and added 20mL of 0.11 M CH₃COOH, then placed on the microwave with power and time variations, and then centrifuged at 3500 rpm for 10 minutes. The filtrate sample was transferred into a glass vial (Fraction 1). The residue from fraction 1 was washed with distilled water until pH-neutral, then added 20 mL of 0.5M NH₃OH-HCl (pH = 1.5) and placed on the microwave with a variation of power and time, then centrifuged at 3500 rpm for 10 min. The filtrate was transferred into a glass vial (Fraction 2). The residue then was washed with distilled water until pH-neutral. The residue from fraction-2 was added 5 mL of 8.8 M H₂O₂ in a vial and placed on the microwave with a variation of power and time, then 25 mL of 1 M CH₃COONH₄ (pH = 2), put on microwave with a variation of power and time, then centrifuged with 3500 rpm for 10 minutes. The filtrate was then transferred into a glass vial (Fraction 3). The residue from fraction-3 was washed with distilled water until pH-neutral, then placed in digestion vial on the hotplate containing sand and added 10mL of 6 mL conc. HF and aqua regia to digestion vial, and heated at 185°C until dry and then added 10 mL of 1% HNO₃. Filtrate sample was put into plastic vials (Fraction 4). All of the filtrate in each fraction was analyzed using AAS Perkin Elmer A Analyst-700.

2.2. Calculations

2.2.1. Accuracy and Precision
Accuracy and precision analysis of BCR method analysis were calculated as follows:

\[
\% \text{recovery} = \frac{C_{\text{Leaching}}}{C_{\text{reference}}} \times 100\%
\]

\[C_{\text{Leaching}} = \text{Fe concentrations in the BCR method of sediment reference JMS-1}\]
\[C_{\text{reference}} = \text{Database of Fe concentrations in sediment reference}\]

2.2.2. Fractionation analysis of Fe bounding in Sediment
Percentage fractionation analysis of each sediment was measured as follows:

\[
\% Fe_i = \frac{\text{Fraction Fe}_i}{\text{Total Fe - Fractions}} \times 100\%
\]

\[Fe_i = \text{Concentration of Fe in the fraction i (i = 1, 2, 3, 4) in sediment. Total Fe-Fractions = Summations of Fe concentration in the each fraction (Fraction 1 + Fraction 2 + Fraction 3 + Fraction 4) in sediment.}\]

2.2.3. Determination of contamination factor (CF)

\[
\text{CF(Fe)} = \frac{Fe_{\text{sampel}}}{Fe_{\text{reference}}}
\]

The value of CF<1 indicates low contaminant, 1≤CF<3 indicates medium contaminant, 3≤CF≤6 indicates highly contaminant and CF>6 indicates extremely contaminant.
3. Results and Discussion

3.1. XRF analysis in sediment

Table 2 shows the composition (% Fe) in sediment at the location S2> S3> S1. The Fe bond with silica minerals becomes the main point of Fe mobility analysis to the environment. Based on their chemical composition of Fe residences, Fe-silica minerals are divided into two types: ferromagnesian and non-ferromagnesian silicates. The ferromagnesian mineral silicate has a darker color, containing the highest component of Fe element. The non-ferromagnesian mineral silicates have a lighter color and more contain Ca, Al, and K elements than the Fe [6]. Based on Table 1, at the location S1 and S3, sediment samples have a percentage of Ca, Al and K compositions greater than those in samples at the location S2. Sediment samples at location S1 and S3 tended to be non-ferromagnesian silica minerals, suggesting that Fe samples were more bounded to other compounds such as carbonate, Mn oxide, and organic matter. The presence of Fe in sediment at the location S2 was more bounded to silica minerals, suggesting Fe in sediment sample at location S2 was more resistant and not easily detached and potentially contaminated to the environment compared with Fe in sediment samples at location S1 and S3.

| Code of sample | Metal contents (% wt/wt) |
|----------------|--------------------------|
|                | Al | Si  | K   | Ca  | Mn  | Fe   |
| S 1            | 6.90 | 31.5 | 1.40 | 17.9 | 0.60 | 36.6 |
| S 2            | 4.00 | 9.60 | 0.20 | 4.27 | 0.58 | 72.9 |
| S 3            | 6.20 | 27.6 | 0.96 | 16.9 | 0.57 | 41.4 |

3.2. Optimized power and time of leaching microwave BCR Method

Figure 1 shows that the condition optimum of microwave power and time required for leaching Fe in each of sediment fraction. It is affected by attacking solvent to bind of Fe in the carbonate, sulfide, oxide, and organic matters. As shown in Figure 1, the optimum condition of power and time of microwave time is 50% power - 3 minutes for leaching Fe at fractions 1 and 2. At fraction 3, the condition of power and time of microwave time needed 10% power at 2 minutes for maximum leaching of Fe. However, at the fraction 4, we did not use the microwave BCR method due to consider explosive solvent and then sand bath above hotplate at 185°C was used. As listed in Table 3, the accuracy of leached Fe in sediment fractions 1 and 2 about 100%, suggesting this optimized method and selective solvents leached of Fe and successfully attacked organic and Mn oxide bounding in the two sediment fractions. At the sediment fraction 3, the optimized power and time of microwave BCR for leaching of Fe had the lowest recovery of 68.18% (Table 3). This is possibly affected by Fe chelated by strongly organic bounding in sediment. Moreover, in sediment fraction 4, the percent accuracy of leached Fe was about 99.68% indicates that the hotplate methods using aquaregia for dissolving completely of Fe in sediment.
Figure 1. Optimized of power and time of microwave BCR for leaching of Fe in sediment

Table 3. The accuracy of the optimized of power and time of microwave BCR

| Fraction                | Optimized of power and time | 1)Leached of Fe (% w/w) | 2)Reference of Fe (% w/w) | Accuracy (%) |
|-------------------------|----------------------------|-------------------------|---------------------------|--------------|
| Fraction 1              | 50% - 3 minutes             | 0.040                   | 0.034                     | 119          |
| Carbonate binding       |                            |                         |                           |              |
| Fraction 2              | 50% - 3 minutes             | 0.99                    | 0.91                      | 109          |
| Mn-Oxide binding        |                            |                         |                           |              |
| Fraction 3              | 10% - 2 minutes             | 0.30                    | 0.44                      | 68.2         |
| Organic binding         |                            |                         |                           |              |
| Fraction 4              | Hotplate (185°C)            | 3.15                    | 3.16                      | 99.7         |
| Silica-alumina binding  |                            |                         |                           |              |

1) BCR microwave method  2) BCR method

3.3. Test of accuracy and precision of optimized BCR-Microwave

The sample used in this accuracy and precision test was a JMS-1 sediment sample with 3 repetitions. At list in Table 4, the optimized BCR-microwave method has accuracy and precision values of 89.4% and 1.30 %, respectively. This result suggests BCR-microwave with an optimized condition has met the good criteria with good accuracy and precision with RSD <2%. Moreover, this BCR-microwave method is recommended to use the assessment and evaluation of sediment contamination.
3.4. Analysis of Fe binding in sediment fraction

The fractionation analysis of Fe bonds in the three Prigi sediment samples was performed using the optimized power and time of BCR-microwave method with the high accuracy and precision after comparing with the JMS-1 reference sediment. As listed in Table 5, the portion of Fe bounded of sediment sample at S2 tended to be resistant fraction. This is because the sediment is categorized as a type of ferromagnesian silica sediment. Fe bound to this fraction is more difficult to leach into the environment. The chemical reactions of bounded Fe can be written as shown in Equation 1-3.

\[
\begin{align*}
\text{Fe}^{2+} (aq) + 2\text{CH}_3\text{COOH} (aq) & \rightarrow \text{H}_2(g) + (\text{CH}_3\text{COO})_2\text{Fe} (aq) \quad (1) \\
\text{FeCO}_3 (\text{sediment}) + 2\text{CH}_3\text{COOH} (aq) & \rightarrow (\text{CH}_3\text{COO})_2\text{Fe} (aq) + \text{H}_2\text{CO}_3 (aq) \quad (2) \\
\text{Fe}_2(\text{CO}_3)_3 (\text{sediment}) + 6\text{CH}_3\text{COOH} (aq) & \rightarrow 2(\text{CH}_3\text{COO})_2\text{Fe} (aq) + 3\text{H}_2\text{CO}_3 (aq) \quad (3)
\end{align*}
\]

As shown in Figure 2, % Fe in the fraction 1 shows 1.57%, 0.56%, and 0.98% at S1, S2, and S3 locations, respectively. The portion of Fe-fraction 1 in the total concentrations of Fe ranged between from 0.56 to 1.57% indicating the probability of Fe contaminated sediment.

### Table 4. Test of accuracy and precision of optimized BCR Microwave

| Fraction | Fe concentrations (mg/L) | [Fe] (mg/L) | [Fe] (%/w) | SD | %RSD |
|----------|--------------------------|-------------|-------------|----|------|
|          | 1 | 2 | 3 |          |       |       |       |
| 1        | 0.81 | 0.64 | 0.76 | 0.74 | 0.03 | 0.07 | 9.68 |
| 2        | 8.26 | 8.14 | 8.25 | 8.22 | 0.82 | 0.05 | 0.67 |
| 3        | 1.86 | 1.45 | 1.68 | 1.66 | 0.23 | 0.17 | 9.96 |
| 4        | 11.2 | 11.9 | 11.2 | 11.5 | 2.98 | 0.37 | 3.25 |
| **Total Fractions** | 22.2 | 22.2 | 21.8 | 22.1 | 4.06 | 0.29 | 1.30 |

Reference of Fe (%/w) 4.54 Accuracy (%) 89.4

As shown in Figure 2, % Fe in the fraction 1 shows 1.57%, 0.56%, and 0.98% at S1, S2, and S3 locations, respectively. The portion of Fe-fraction 1 in the total concentrations of Fe ranged between from 0.56 to 1.57% indicating the probability of Fe contaminated sediment.

### Table 5. Fractionations of Fe in the samples (S1, S2, and S3)

| Fraction | Fe concentrations (mg/kg) in sediment |
|----------|---------------------------------------|
|          | S1 | Fe bounded (%) | S2 | Fe bounded (%) | S3 | Fe bounded (%) |
| Fraction 1 | 182 | 1.57 | 126 | 0.56 | 125.6 | 0.98 |
| Fraction 2 | 8022 | 69.28 | 2609 | 11.5 | 5563 | 43.4 |
| Fraction 3 | 515.2 | 4.45 | 404.6 | 1.79 | 387.8 | 3.03 |
| Fraction 4 | 2860 | 24.70 | 19468.8 | 86.1 | 6728.8 | 52.6 |
| **Total of Fe (mg/kg)** | 11579 | 11260.4 | 12805.2 |

The Fe concentrations in the fraction 1 revealed Fe bounded in the carbonate and released the environment due to the presence of an acid and changes of salinity. Fraction 1 used a weak acid solvent, CH\text{3}COOH to attack and break the bond of Fe with carbonate. The percentage of Fe in sediment fraction 2 was 69.28%: 11.54%; and 43.44% at S1, S2 and S3 locations, respectively. This indicates that bounded Fe at S1 and S3 sample sediments undergoing reaction of reduction from Fe\text{3}+ to Fe\text{2}+ ions released to the seawaters. The high portion of bounded Fe in fraction 1 at S1 was influenced by the depth of the sediment caused anoxic sediment. Moreover, the level of concentrations of Fe\text{2}+ from anoxic sediment was added with photosynthetic reaction from plants and then caused the high-level concentration of Fe\text{2}+ which contaminated or polluted environment. We used hydroxylamine solvent to leach bounded Fe in...
this reduction fraction. This fraction was dominated by Mn oxide to bind Fe. The Fe at this fraction is also very sensitive to redox potential changes. The dissolution of the Fe-oxide or Mn-oxide minerals by \( \text{NH}_2\text{OH.HCl} \) to release Fe can be written as Equation 4-5.

\[
\begin{align*}
\text{Fe}_2\text{O}_3(s) + 6\text{H}^+(aq) & \rightarrow \text{Fe}^{3+}(aq) + 3\text{H}_2\text{O(l)} \quad (4) \\
4\text{Fe}^{3+}(aq) + 2\text{NH}_2\text{OH.HCl(aq)} & \rightleftharpoons 4\text{Fe}^{2+}(aq) + \text{N}_2\text{O(g)} + \text{H}_2\text{O(l)} + 2\text{HCl(aq)} + 4\text{H}^+(aq) \quad (5) \\
\text{Fe}^{3+}\text{Fe}_2\text{O}_3(\text{sediment}) + \text{NH}_2\text{OH.HCl(aq)} & \rightarrow 4\text{Fe}^{2+}(aq) + \text{N}_2\text{O(g)} + \text{H}_2\text{O(l)} + 2\text{HCl(aq)} + 4\text{H}^+(aq) + \text{Fe}^{3+}(aq) \quad (6)
\end{align*}
\]

![Figure 2. The composition of Fe bounded in sediment samples](image)

Figure 2 shows the portion of leached Fe in sediment fraction 3 with the ranges from 1.79 to 4.45%. All of %Fe in sediment fraction 3 was less than 5%, indicating the solvent could not break Fe in the bonding strength of the organic material and the chelate reaction. As listed in Table 3, the % accuracy was also recorded low about 68.2%. Fraction 3 is well known as the oxidation fraction. Fe is bonded with organic materials as well as sulfide ions. Organic matter in sediment is generally composed of humic acid and fulvic acid. Humic acid has a functional group -COOH, -OH phenolate and -OH alcoholate which can bind to Fe. The presence of H\(_2\)O\(_2\) in seawater is produced by the phytochemical process by organic compounds and then bound with Fe in sediment fraction 2 as Fe residence. We used the H\(_2\)O\(_2\) and CH\(_3\)COONH\(_4\) solvents to oxidize of Fe\(^{2+}\) to Fe\(^{3+}\) ion and prevent the readsorption of the leached Fe to the oxidized substrate. The chemical reactions can be written as follows (Equation 7).

\[
\begin{align*}
\text{AH}(s) - \text{Fe}^{3+}(\text{sediment}) + \text{H}_2\text{O}_2(aq) + 2\text{CH}_3\text{COONH}_4(aq) & \rightarrow \\
\text{AH}(s) + (\text{CH}_3\text{COO})_2\text{Fe(aq)} + 2\text{NH}_4\text{OH(aq)} + \text{H}_2\text{O(l)}
\end{align*}
\]

The percentage of Fe in sediment fraction 4 was 24.70%; 86.11%; and 52.55% at S1, S2 and S3 locations, respectively (Figure 2). This is related to the type of ferromagnesian silicate-alumina in this fraction that has covered of Fe in sediment. In the sediment sample at S1 had a lower percentage comparing with those at S2 and S3 locations, indicating more pollution. Fe in this fraction is strongly bonded to silicates and alumina. The strength of Fe bonds with minerals caused the Fe difficult to dissolve into the seawaters, especially sediment at S2 and S3 (residual fractions). Natural weathering can break Fe in this fraction, due to its contents of Fluoride in natural stones. We applied HF and H\(_2\)O\(_3\) to leach of Fe in sediment fraction 4. The chemical reactions can be written as follows (Equation 8).

\[
\begin{align*}
\text{Fe}^{2+}\text{SiO}_2\text{Al}_2\text{O}_3(s) + 4\text{HF(aq)} + x\text{HNO}_3(aq) & \rightarrow \\
\text{Fe(NO}_3)_x(aq) + 2\text{Al(NO}_3)_3(aq) + \text{SiF}_4(g) + x\text{H}_2\text{O(l)} + x\text{O}_2(g)
\end{align*}
\]
4. Conclusion
The optimized of power and time of BCR-microwave method in sediment collected from Prigi has a recovery percentage of 89.4% and RSD% of 1.30% indicating this method is accurate and precise. The concentration of Fe was in the total fractions at the recreation place of the coastal Prigi location > Port of Prigi location > Nearly Residences location, indicating the vicinity of tourist location has great potentials to contribute Fe to the Gulf of Prigi.

References
[1] MD Hossain Sazzad, Anugrah Ricky Wijaya, Kentaro Tanaka and Shigeru Ohde 2010 Environmental effects on the stable carbon and oxygen isotopic compositions and skeletal density banding pattern of Porites coral from Khang Khao Island, Thailand Afr. J. Biotechnol. 9 5373–82
[2] Wijaya A R, Ouchi A K, Tanaka K, Shinjo R and Ohde S 2012 Metal contents and Pb isotopes in road-side dust and sediment of Japan Journal of Geochemical Exploration 118 68–76
[3] Wijaya A R, Ouchi A K, Tanaka K, Cohen M D, Sirirattanachai S, Shinjo R and Ohde S 2013 Evaluation of heavy metal contents and Pb isotopic compositions in the Chao Phraya River sediments: Implication for anthropogenic inputs from urbanized areas, Bangkok Journal of Geochemical Exploration 126–127 45–54
[4] Wijaya A R, Farida I, Wonorahardjo S, Utomo Y, Daupor H, Hossain M S and Kunisue T 2018 BCR Sequential Leaching for Geochemical Fractions and Assessment of Fe, Ni, and Mn in the Coastal Sediments Sendang Biru Port, East Java, Indonesia Journal of Physics: Conference Series 1093 012002
[5] Ariadi Lusiana R, Putri Protoningtyas W, Ricky Wijaya A, Siswanta D, Mudasir M and Juari Santos S 2017 Chitosan-Triply Phosphate (CS-TPP) Synthesis Through Cross-linking Process: the Effect of Concentration Towards Membrane Mechanical Characteristic and Urea Permeation Oriental Journal of Chemistry 33 2913–9
[6] Wijaya A R, Ohde S, Shinjo R, Ganmanee M and Cohen M D 2016 Geochemical fractions and modeling adsorption of heavy metals into contaminated river sediments in Japan and Thailand determined by sequential leaching technique using ICP-MS Arabian Journal of Chemistry
[7] Wijaya A R, Farida I, Sakbaniah A, Rahmawati A M, Budiasih E, Daupor H, Hossain M S and Kunisue T 2018 Distribution and Assessment of Fe and Mn in the Coastal Sediments of Sendang Biru, East Java, Indonesia Journal of Physics: Conference Series 1093 012013
[8] U.S. Enviromental Protection Agency 2003 Guidance for Developing Ecological Soil Screening Levels
[9] Guven D E and Akinci G 2013 Effect of sediment size on bioleaching of heavy metals from contaminated sediments of Izmir Inner Bay Journal of Environmental Sciences 25 1784–94
[10] Nigerian Institute for Oceanography & Marine Research, Victoria Island, Lagos, Ladigbolu I A and Appia J 2014 Geochemical fractionation of Heavy metals in the sediment at the point of discharge of Iyalaro stream into Lagos Lagoon IOSR Journal of Environmental Science, Toxicology and Food Technology 8 13–6
[11] González-Davila M, Santana-Casiano J M and Millero F J 2005 Oxidation of iron (II) nanomolar with H2O2 in seawater Geochimica et Cosmochimica Acta 69 83–93
[12] Filgueiras A V, Lavilla I and Bendicho C 2002 Chemical sequential extraction for metal partitioning in environmental solid samples. Journal of Environmental Monitoring 4 823–57

Acknowledgements
This research is funded by the PNBP Universitas Negeri Malang grant, 2018.