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Removal of Heavy Metals from a Contaminated Green Mussel [*Perna viridis* (Linneaus, 1758)] using Acetic Acid as Chelating Agents

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Abstract

Dried green mussel [*Perna viridis*, (Linneaus, 1758)] was soaked in acetic acid solution at a concentration of 10 %, 15 %, 20 %, and 25 % for 0 min, 30 min, 60 min, and 90 min. The content of heavy metals such as Pb, Cr, and Cd after soaking were analyzed by *Atomic Absorption Spectrometer* (AAS). The results indicated that after soaking in acetic acid solution at a concentration of 25 % for 90 min reducing heavy metal Pb from 2.879 $\mu g \cdot g^{-1}$ to 1.407 $\mu g \cdot g^{-1}$, Cr from 0.730 $\mu g \cdot g^{-1}$ to 0.362 $\mu g \cdot g^{-1}$ and Cd from 0.710 $\mu g \cdot g^{-1}$ to 0.441 $\mu g \cdot g^{-1}$. The increasing concentration of acetic acid solution and the longer soaking time, the levels of heavy metals (Pb, Cr, Cd) in green mussel will decreasing. Interestingly, acetic acid was able to chelate the studied heavy metals in green mussel.

Keywords: Acetic acid solution; green mussel [*Perna viridis*, (Linneaus, 1758)]; heavy metal.

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1. Introduction

Green mussel [*Perna viridis* (Linneaus, 1758)] is a commercially important bivalve. Bivalves are filter feeder feeding on phytoplankton, zooplankton, and other orgaic materials. Increased levels of heavy metals in seawater will be followed by increased levels of heavy metals in marine biota, one of which is the green mussel. Kahle and Zauke (2002) had reported that some aquatic organisms having the ability to concentrate contaminants in their tissue and organ systems to more than a million times, compared to their concentration in their habitat. Mussel as a filter feeder is accumulates huge amounts of toxic pollutants mainly heavy metals from its habitat. Effluents arising from human activities, infrastructure developments, agricultural activities, tourism, and allied activities are the major source of heavy metal contamination of mussel growing areas.

Heavy metals could be classified as potentially toxic (e.g arsenic, cadmium, lead, mercury), probably essential (e.g copper, zinc, iron, manganese). Toxic elements could be harmful even in low concentration when ingested over a long time period. The essential metals could also produce toxic effect when their intake is excessive (Uluozlu et al., 2007). Furthermore, through the food chain will cause acute and chronic poisoning, even carcinogenic to humans who eat the shellfish. One easy way to do by the consumer of shells to reduce the influx of heavy metals including Pb, Cd, and Cr is a way of purification.

Metal dissolution by organic acids is likely to be more representative of a mobile metal fraction that is available to plants. The chelating organic acids are able to dislodge the exchangeable, carbonate, and reducible fractions of heavy metals via washing procedures (Labanowski et al., 2008). Sodium acetat is able to chelate the heavy metals (As, Pb, Cd, and Ni) in green mussel to levels permissible for human (Azelee et al., 2014).

Purpose of the study is to develop method that could safely remove heavy metals (Pb, Cr, and Cd) from contaminated green mussel by soaking in a solution of acetic acid. This study examines the potential of acetic acid solution for purification of lead (Pb), Chromium (Cr), and Cadmium (Cd) in green mussel.

2. Material and methods

2.1. Sample preparations

Green mussel was purchased from markets in Semarang, Central Java, Indonesia. Samples of green mussel was washed, then boiled to separate their shells and meat. The meat was dried at a temperature of 100 °C to obtain the dry weight. Dried meat of green mussel was soaked in a solution of acetic acid at a concentration of 10 %, 15 %, 20 %, and 25 % for 0 min, 30 min, 60 min, and 90 min.

2.2. Heavy metals analysis

The heavy metals such as Pb, Cr and Cd of green mussel meat were analyzed by *Atomic Absorption Spectrometer* (AAS) in the Laboratory of Analytical Chemistry, Faculty of Science and Mathematics, Diponegoro University. Samples (500 mg) were dissolved and 1 mL of nitric acid and perchloric acid and 2.5 mL distilled water were added. Then put in a microwave and analyzed using AAS. This method worked by comparing the absorbance of the sample solution with standard solution to obtain the sample concentration. AAS absorbances was calibrated with a standard series of unknown concentration. The results of the analysis was a calibration curve.

2.3. Statistical analysis

The differences between the mean values of multiple groups were analyzed by one-way analysis of variance (ANOVA) with Tukey methods range test. ANOVA data with a $P < 0,05$ was classified as statistically significant. SPSS 17 software were used.

3. Results and discussions

3.1. Pb content in green mussel

Acetic acid with varying concentration and soaking time was used to reduce the level of Pb in green mussel. The levels of Pb studied were successfully reduced by acetic acid and are presented in Fig. 1. The obtained results from AAS showed that the Pb content in green mussel before soaking treatment was 2.879 $\mu g \cdot g^{-1}$. This shows that the content of Pb in green mussel samples exceeded the permissible limit. Based on research by Azelee et al. (2014), the permissible levels of Pb set by the Commission Regulation of EU (2006) for human consumption was 1.00 $\mu g \cdot g^{-1}$. Sivapullaiah et al. (2010) also explained that the Pb in fish may be due to the contamination from air pollutant, such as PbO, may dissolve and advance through the water. Then the green mussel absorbed heavy metals from water.

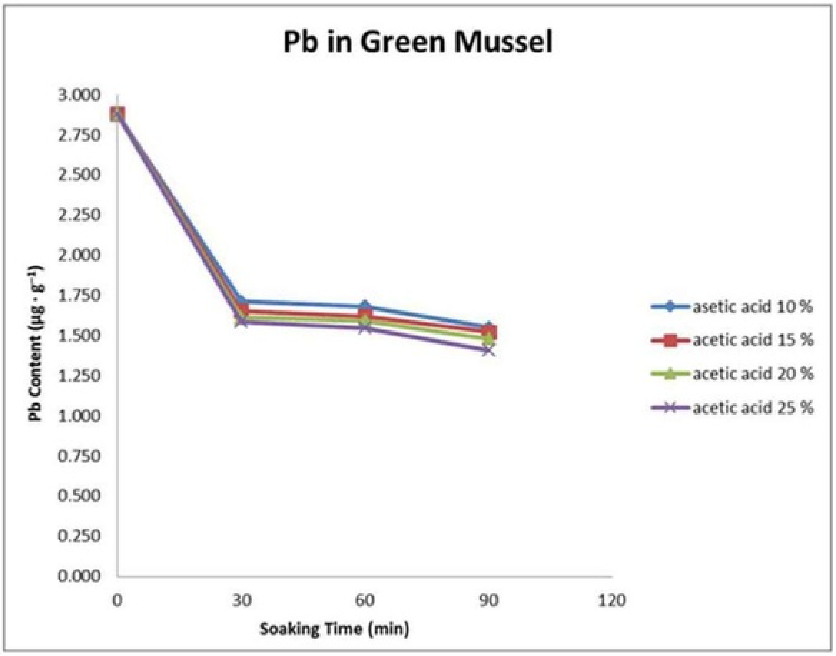

Fig. 1. Effect of acetic acid at varying concentrations and soaking times on Pb content in green mussel.

The Pb content in initial green mussel also showed that the waters had been contaminated with Pb. According Putri et al. (2012), the current level of Hg, Pb, and Cd in Kamal estuary, Jakarta Bay, make it suitable for the propagation of shellfish and other marine biotas. Lead (Pb) in green mussel successfully removed above 50 % after soaked with 25 % of acetic acid for about 90 min, but still not achieve the permissible limit for Pb. Heidari et al. (2015) reported that a slight decrease in pH coincided with a slight increase in the removal of Pb. It was mostly bound to carbonates and under alkaline conditions precipitation of $PbCO_3$ is favoured.

The statistical analysis showed that each treatment was significantly different. The data suggests that the higher concentration of acetic acid and the longer period of treatment time can increase the removal of Pb out from green mussel. It was indicates that an increase of the acid concentration in a corresponding increase of the offered energy for the breakdown of the chemical bonds of the metals in the material and the longer period of treatment time will give enough time for chelating agents to chelate with the heavy metals and excreted out from the *P. viridis* flesh (Stylianou et al., 2007; Azelee et al., 2014; Gzar et al., 2014).

Reduction of heavy metals in green mussel treated with acetic acid could be due to formation of insoluble acetate salts of these metals. Green mussel liquid contained a considerable amount of metals, leaked with liquid by filtration, and thus should be discharded to minimize metal exposure (Elnimr, 2011).

3.2. Cr content in green mussel

Acetic acid with varying concentration and soaking time was also used to reduce the level of Cr in green mussel. The levels of Cr studied were successfully reduced by acetic acid and are presented in Fig. 2. The results showed that the Cr content in green mussel before soaking treatment was 0.7300 $\mu g \cdot g^{-1}$. According to WHO (2006), Cr content in foods that allowed consumed was 0.07 $\mu g \cdot g^{-1}$ per day. Heavy metal exposures assessment have monitored by metal determination in human eye (Haddad, 2012). Too much Cr content in body will cause cancer, because Cr in human's body can inhibit benzopyrene hydroxilase enzyme's operation (Suprapti et al., 2014).

Cr content in green mussel are also affected by the season. Base on the research of Suprapti et al. (2012), the chromium content of mud crab in rainy season is lower than in dry season because of the rain water. Beside that, the difference was also because of the significant difference of physical-chemical factors like temperature, salinity, conductivity, and river flow speed between dry season and rainy season. The Cr content in the crab on dry season and rainy season seems almost the same, with the average value of 5.237 $\mu g \cdot g^{-1}$ for dry season and 4.848 $\mu g \cdot g^{-1}$ for rainy season.

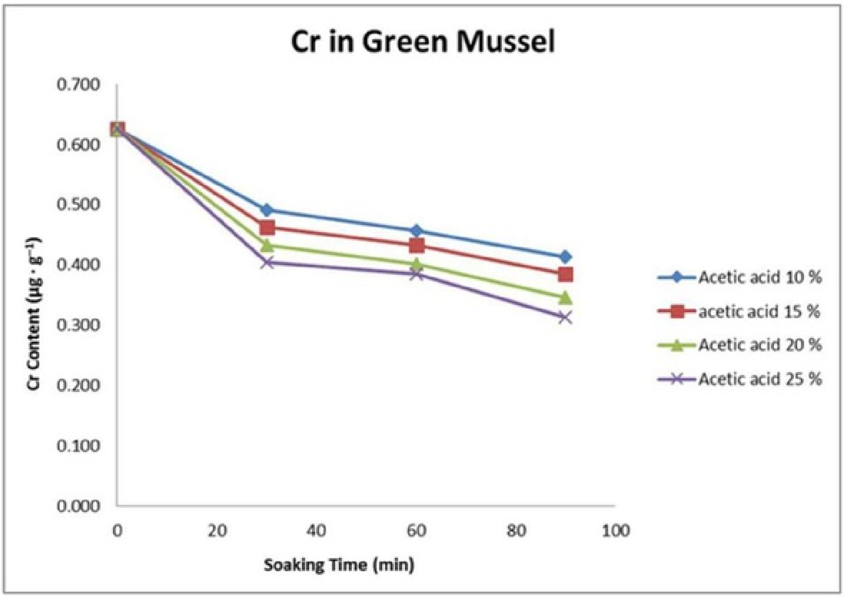

Fig. 2. Effect of acetic acid at varying concentrations and soaking times on Cr content in green mussel.

Chromium spreads widely on the nature. In the water bodies, Cr can enter through two ways, naturally and non-naturally. The natural way of Cr entering the waters is because some physical factors like erosion. The non-natural way of Cr entering the waters is as side effect of human activity of industry like electroplating, tannery, textile industry, paint industry, and domestic waste disposal. Cr entering the body and then undergo a process of biotransformation and bioaccumulation in the living organisms (Suprapti et al., 2014).

Cr in green mussel successfully removed above 50 % after soaked with 25 % of acetic acid for about 90 min. The statistical analysis showed that each treatment was gave significantly different effect to the samples. The data suggests that the higher concentration of acetic acid and the longer period of treatment time, the higher amount of the removal of Cr out from green mussel.

3.3. Cd content in green mussel

Acetic acid with varying concentration and soaking time also was used to reduce the level of Cd in green mussel. The levels of Cd studied were successfully reduced by acetic acid and are presented in Fig. 3. The results from AAS showed that the Cd content in green mussel before soaking treatment was 0.7100 µg · g^{-1}. This shows that the content of Cd in green mussel samples was still below the permissible limit. Based on research Azelee et al. (2014), the permissible levels of Cd set by the Commission Regulation of EU (2006) for human consumption was 1.00 µg · g^{-1}.

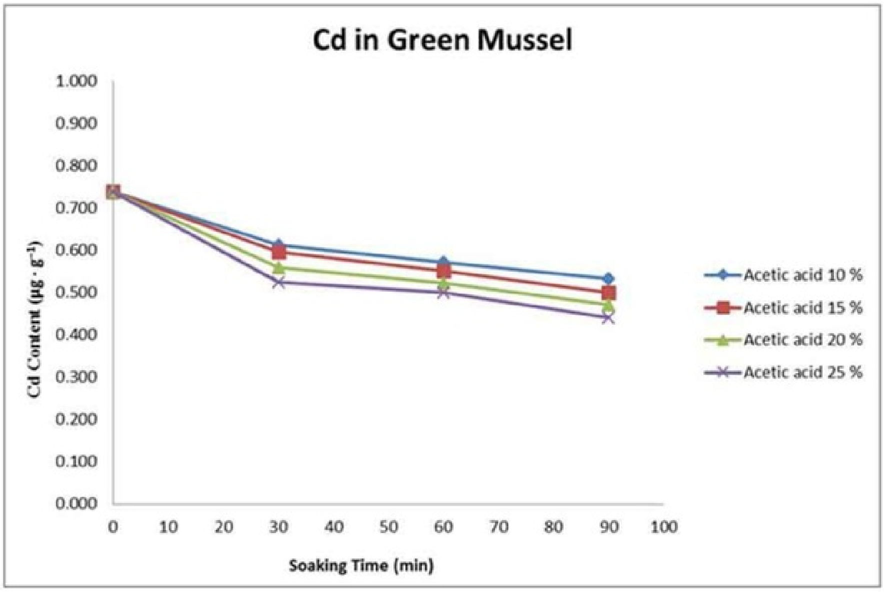

Fig. 3. Effect of acetic acid at varying concentrations and soaking times on Cd content in green mussel.

Although the levels of Cd have not exceeded the permissible limit, the content of Cd in green mussel should be reduced. If the green mussel contaminated Cd for human consumption, can cause Cd accumulates in the body. Because of Cd is not easy to excreted out from body. The adverse toxic effects caused by Cd are widely recognized for their detrimental effects on human health. The Cd also may act as allergens, mutagens, or carcinogens (Wafaa et al., 2003).

The Cd content in initial green mussel also showed that the waters and sediment had been contaminated with Cd. Other study also supported that there were significantly correlation between mussel and concentration of Hg and Cd in sediment at Paria Gulf, Trinidad, and Venezuela (de Astudillo et al., 2005) and for Cd and Pb at West coastal waters of Peninsular Malaysia. Sediments are an important source of metals to filter feeders, among mussels and oysters (Yap et al., 2004).

Cadmium is widely distributed at low level in the environment and most foods have an inherently low level of Cd which has been shown to bind to the protein and accumulate significantly in higher level (FDA, 2001). Ololade et al. (2008) reported that Cd level is almost 10 times higher in shell fishes than in finfishes. This could also be due to fertilizer application in nearby area are transported to the estuaries by leaching and erosion as agriculture is also an important activity of the village folks besides fishing. Bhouri et al. (2010) also explained that the Cd in fish may be due to the contamination from industrial waste of factories in the town. Industrial discharges are considered the major sources of heavy metal pollutants of water. Fish absorbed heavy metals from water through the gills, skin, and digestive tract.

The statistical analysis showed that each treatment was significantly different. The data suggests that the higher concentration of acetic acid and the longer period of treatment time the higher the removal of Cd out from green mussel. Cd in green mussel successfully removed above 37.88 % after soaked with 25 % of acetic acid for about 90 min. Wuana et al. (2010) reported that oganic acid appeared to remove most of the heavy metals associated with exchangeable and reducible fractions. The reaction between the metal binder with metal ions causes Cd loses its ionic properties. On the other hand, a slight increasing concentration of acid was higher removal efficiency of Cd (Oustan et al., 2011).

4. Conclusion

The chelation method was found to be a potential technique for the removal of heavy metals studied in green mussel. Interestingly, acetic acid was able to chelate the studied heavy metals in green mussel. The increasing concentration of acetic acid and the longer soaking, the lower the levels of heavy metals (Pb, Cr, Cd) in green mussel decreasing.

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