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The effect of depuration on lead levels of the cockles *Anadara* sp. by using activated carbon as a filter

A R Firdaus¹, A S Mubarak¹ and W Tjahjaningsih^{1,2}

¹ Faculty of Fisheries and Marine, Universitas Airlangga, Surabaya, Indonesia

² Corresponding author: wahju_fpk@yahoo.com

Abstract. Consumption of cockles with high concentrations of lead metal (Pb) will hurt harm humans from health aspects. The concentration of Pb metal in cockles can be reduced through a depuration process. This study aims to determine the efficient time of retention and the rate of decrease in the concentration of Pb in the cockles (*Anadara* sp.) by using activated carbon as a filter during the depuration process. The treatment of study consists of P0 (no depuration), P1 (depuration 24 Hours), P2 (depuration 48 Hours), and P3 (depuration 72 Hours). The results showed that the process of depuration of the cockles for 24 hours with activated carbon filters was able to reduce the Pb metal content to 0.926 mg/kg and even the depuration process for 72 hours, the efficiency of the depuration process increased to 86.52% so that the Pb content in the cockles became 0.273 mg/kg. Overall, the duration of the process of depuration of the cockles with activated charcoal as a filter can increase the efficiency of Pb metal reduction in the cockles so that it according to SNI 7387: 2009, which is the maximum metal content of Pb in the shellfish 1.5 mg/kg.

1. Introduction

Industrial waste is one source of heavy metal pollution such as Pb. Lead is the most significant poison of heavy metals, and the inorganic form is absorbed through the consumption of food and water, and inhalation [1]. Pb toxicity causes reduced hemoglobin synthesis, impaired renal, joint, reproductive and cardiovascular function and chronic damage to the central and peripheral nervous system [2].

Heavy metals can enter the body of the shellfish through two ways, the process of respiration and digestive mechanisms. Pb heavy metals can be distributed into the body of organisms and some will be accumulated through various intermediaries, one of which is through food contaminated by heavy metals [3]. Pb heavy metals can easily accumulate in organisms in the cockles. Shellfish exposed to heavy metal Pb did not show significant changes [4].

The concentration of Pb heavy metals in the waters of Gesek Sedati Sidoarjo waters is +0.60 mg/L [5]. The concentration of Pb heavy metals in these waters is not in accordance to the Decree of the Minister of Environment No. 51 of 2004 concerning the Pb heavy metal content of marine biota <0.008 mg/L. Marine biota that lives in polluted waters has the potential to be exposed to heavy metals [6], [7], [8]. Pursetyo *et al.* [9] found shells with a relatively high Pb metal concentration of 2.144 mg/L.

Naturally, the cockles can detoxify to reduce levels of heavy metals in the body and excreted through the process of excretion [10]. However, the high concentration of heavy metals contained in the waters has an impact on the ability to remove heavy metals in the body of the shellfish not



comparable to the levels of heavy metals that enter through the process of respiration or the absorption of food by shellfish [11].

Pb metal content in shells can be reduced by doing the depuration process. The principle of the depuration process is the maintenance of shells in seawater media with low pollutants (heavy metals Pb) and under running water conditions [12]. This will make the Pb metal excretion in the body of a cockle occur continuously. Pb metal excretion in the body of the cockles can be increased by the addition of compounds/materials that can bind heavy metals in maintenance media. The addition of these compounds aims to bind Pb metal released by the cockles through the excretion system into the media.

Anacleto *et al.* [13] stated that the process of depuration of shellfish is influenced by the length of time of the depuration process. Sulmartiwi *et al.* [14] added that the filter used also affects the results of the depuration process. Activated carbon is a good adsorbent for the adsorption of gases, liquids, and solutions because it has a high internal porosity, so it has absorption [15]. The process of adsorption on activated carbon starts from the outside of activated carbon absorbs substances, moves to the pores, and then is absorbed into the inner walls of activated carbon [16].

This research was conducted to determine the efficient depuration process time by using active charcoal filters to reduce the concentration of Pb metal in cockles (*Anadara* sp.). Information on the duration of the process of depuration of the shellfish with charcoal filters is beneficial for the shellfish fisherman to be applied in the post-catch handling of the shells before the shell processing stage as an effort to improve the quality of shellfish products.

2. Materials and methods

2.1. Materials

The materials used in the study are seawater, activated charcoal, and cockles (*A. antiquata*) with an average length of 3.6 ± 0.055 cm; average width of 2.1 ± 0.032 cm; average height of 2.5 ± 0.037 cm; and an average weight of 15.37 ± 0.148 g.

2.2. Methods

2.2.1 Preparation of depuration tanks

Depuration tanks that have been cleaned are sterilized using chlorine, then installed with water pump equipment, aerators, baskets, UV lamps, and flow meters. The speed of water flow in this study was 6 liters/minute for 100 liters of water volume. The water flow velocity for the depuration process is 20 liters/minute for a 500 L water volume [12]. Also besides, active charcoal is needed as a filter in the depuration process. Activated charcoal is used with a minimum dose of 6 g/L [17]. Depuration tanks that are ready for use are filled with seawater as much as ± 100 L for each test. The depuration process is carried out with a DO range of 4.66-5.97 mg/L; pH 6.8-7.8 and temperatures from 26.8-28.2°C.

2.2.2 Depuration process of the cockles

Each tank is filled with seawater that has been deposited as much as ± 100 liters and installed with a water pump and filter. Shellfish are placed in a basket and put in a depuration tank. During the depuration process, water in the tank exits through the outlet to the filter tank. Water entering the filter tank will be filtered by charcoal. The filtration water is pumped using a water pump back into the depuration tank through the inlet at the top of the depuration tank. Placement of the filter tank outside the tank depuration and connected to the pipe. Depuration is carried out for the time specified according to treatment. The research treatments consisted of P0: without depuration, P1: 24 hours depuration, P2: 48 hours depuration, and P3: 72 hours depuration.

2.2.3 Measurement of Pb

Pb heavy metal measurements were carried out before and after the depuration of cockles, seawater, and activated charcoal using the AAS method. The main parameter in this study was the efficiency of reduction of heavy metal lead (Pb) in the cockles after depuration. The efficiency of decreasing the concentration of heavy metals uses the overall efficiency equation [15].

2.2.4 Measurement of water quality

Measurement of water quality as a supporting parameter is carried out in the morning, afternoon and evening. The parameters measured include parameters of temperature or water temperature, pH, salinity, and dissolved oxygen (DO).

2.2.5 Analysis of data

The results of this study were then analyzed using Analysis of Variance (ANOVA). Duncan's multiple range test needs to be done next if the treatment given shows a real effect. This test was conducted to determine differences between treatments [18].

3. Result and discussion

3.1. Result

The depuration process causes the concentration of Pb metal in the cockles to decrease, it can be seen from the value of Pb metal reduction efficiency (Table 1). The highest Pb metal reduction efficiency in shells after a 72 hours depuration process was $86.52 \pm 0.57\%$ ($p < 0.05$).

Table 1. Data of Pb concentration, reduction efficiency, and rate of decrease in Pb in the cockles after the depuration process

Treatment	Final Concentration (mg/kg)	The Efficiency of Reduction (%) \pm sd	Rate of decline SD (mg/kg)
P0	2.021	0 ± 0^d	-
P ₁	0.926	54.20 ± 3.08^c	$1,095 \pm 0,05^a$
P ₂	0.365	81.95 ± 1.33^b	$0,561 \pm 0,06^b$
P ₃	0.273	86.52 ± 0.57^a	$0,093 \pm 0,03^c$

Note: P0 (without depuration), P1 (depuration for 24 hours), P2 (depuration for 48 hours) and P3 (depuration for 72 hours). The notation indicated by letters superscript in the same column shows a noticeable difference ($p < 0.05$).

The decrease in Pb metal in the cockles during the depuration process is followed by an increase in Pb metal in water and filters (activated charcoal). Pb metal concentrations in water continue to increase along with the increase in depuration time. Pb metal concentrations in water after 24 hours, 48 hours and 72 hours depuration of 0.351 ± 0.020 mg/L; 0.584 ± 0.027 mg/L; and 0.657 ± 0.013 mg/L, respectively (Figure 1A). The Pb metal concentration in the activated charcoal filter after 24 hours, 48 hours and 72 hours depuration was 0.402 ± 0.044 mg/kg; 0.823 ± 0.095 mg/kg and 0.784 ± 0.71 mg/kg respectively (Figure 1B).

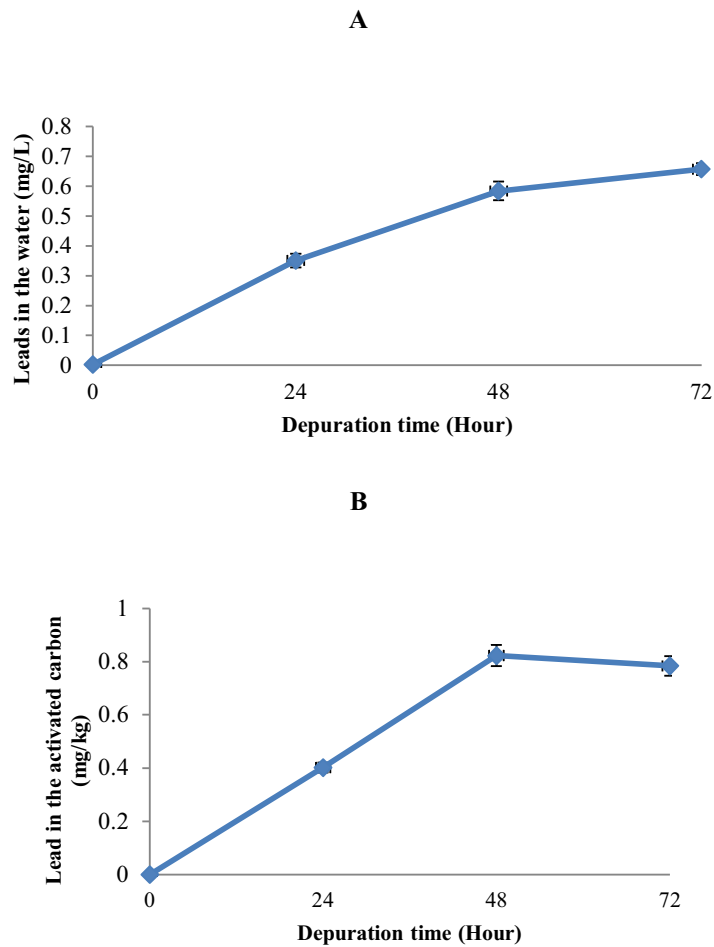


Figure 1. Graphics of lead concentration in the water (mg/L) (A) and activated carbon (mg/kg) (B) during depuration.

Pb metal concentrations in shellfish and water have a correlation of $R^2 = 0.9901$, which follows the equation $y = -2.7136x + 1.9779$ (Figure 2A). The concentration of Pb metal in the cockles and activated charcoal has a correlation of $R^2 = 0.9701$ following the equation $y = -2.0557x + 1.9287$ (Figure 2B).

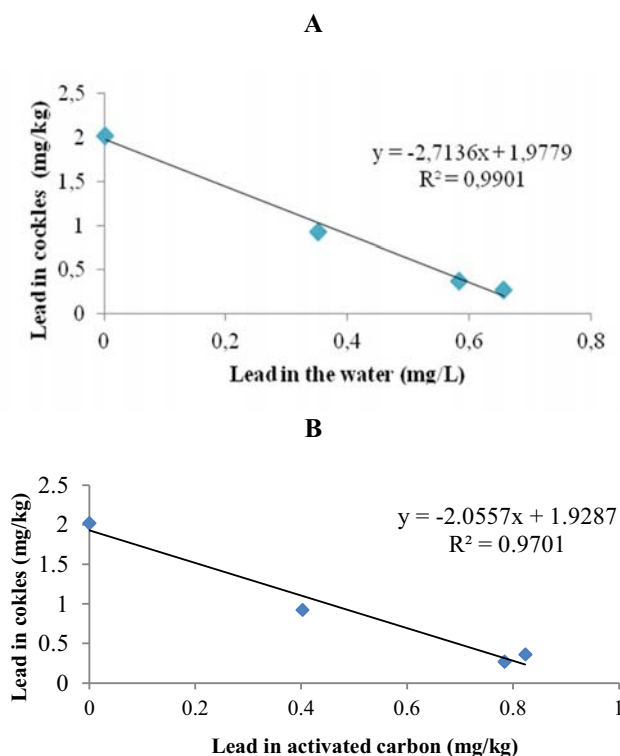


Figure 2. Relationships graphics of lead concentration decreases in cockles inside the water with lead concentrate (A), and water with activated carbon concentrate (B).

The results of measurements of water quality parameters during depuration showed temperatures ranging from 26.8-28.6°C with an average of 27.73°C, DO range from 4.66-5.97 mg/L with an average of 5.49 mg/L, water salinity an average of 30 ppt, and water pH ranges from 6.8-7.8 with an average of 7.08.

3.2. Discussion

The shells used in this study have a uniform size so that the initial Pb concentrations in the shellfish between treatments were not significantly different. Different sizes of shells can result in different Pb metal concentrations in the body of the shellfish [19]. Depuration causes decreased Pb metal concentrations in the cockles followed by an increase in Pb metal concentrations in water and filters (activated charcoal). The impact of the depuration process can reduce Pb metals in shellfish [13, 14, 20, 21]. Pb heavy metal in the cockles during the 24-hour depuration process was able to reduce the Pb metal content to 0.926 mg/kg to meet the SNI 7387: 2009 standard of 1.5 mg/kg. Even with the 72-hour depuration process, the efficiency of the depuration process was 86.52%. Anacleto *et al.* [13] proved that depuration of *Scrobicularia plana* for two days was able to reduce Pb levels to below the threshold level.

Pb metal reduction efficiency in the process of depuration of the cockles has increased by 54.20% - 86.52%. These results prove that the activated charcoal used for the depuration process has an increased concentration of Pb metal. Increasing the concentration of Pb metal in charcoal shows that activated charcoal can bind Pb metal. According to [17] activated charcoal can also bind Cu, Hg, Cd

and Ni metals well. Activated charcoal has functional groups of carbonyl, carboxyl, quinone, phenol, lactone which can bind to heavy metals Pb [22, 23, 25]. The functional groups in activated charcoal (-COOH) cause chemical reactions that occur during the filtration process. The adsorption mechanism of the functional group (-COOH or -OH) in the activated charcoal by releasing H^+ ions so that the functional group is negatively charged (-COO⁻ or -O⁻) and Pb^{2+} ions can be bound. H^+ ions released from activated charcoal will mix with water and cause a decrease in pH from 7.8 to 6.8 [24, 25].

The concentration of Pb metal in activated charcoal after 48 hours depuration is higher than that of activated charcoal after 72 hours depuration. This can be caused because the ability of activated charcoal to bind has decreased. According to [26], if the activated charcoal functional groups have been fulfilled by contaminant materials, the ability of charcoal to bind the contaminants decreases. At this point, the contaminants in the water will pass through the charcoal and return to the waters.

The concentration of Pb metal in activated charcoal at the 72nd hour decreased from the 48th hour. The concentration of Pb metal in activated charcoal after 48 hours depuration was 0.823 mg/kg higher than that of activated charcoal after 72 hours depuration of 0.784 mg/kg. The concentration of Pb metal in the cockles and activated charcoal has a correlation of $R^2 = 0.9701$ following the equation $y = -2.0557x + 1.9287$ (Figure 2B). Activated charcoal functional groups that have been met by contaminants cause the ability of activated charcoal to bind to contaminants has decreased [27]. Active charcoal is saturated because of the increasing time of filtration, high Pb metal concentrations and a small amount of activated charcoal [27, 28, 29]. Active charcoal that experiences saturation can no longer bind Pb metal in water [30, 31, 32]. Absorbency is the ability possessed by activated carbon [15]. The reduced ability of activated carbon is caused by pores on the surface of carbon covered by molecules that have been absorbed [33], so that activated carbon is no longer able to absorb and therefore must be replaced with new activated carbon [15].

The concentration of Pb in water continues to increase with increasing depuration time (Figure 1A). Pb metal concentration in water after depuration increased from 0.002 mg/L to 0.657 mg/L. Pb metal concentrations in shells and water have a correlation of $R^2 = 0.9901$, which follows the equation $y = -2.7136x + 1.9779$ (Figure 2A). The longer the depuration process causes the concentration of Pb metal in water to increase. The Pb metal concentration in water after 72 hours depuration is high so that it affects the ability of the shells to remove Pb metal from the body. This is evident from the rate of decrease in Pb metal in shells after 72 hours depuration of 0.093 mg/kg, lower than the rate of Pb metal reduction after depuration of 24 hours (1.095 mg/kg) and 48 hours (0.561 mg/kg) (Table 1). According to [11, 34, 35] did not rule out the possibility of Pb metal in the water returning to the body of feather shells through the process of respiration. The shellfish that live in waters with high Pb metal concentrations can accumulate Pb metals. Presented by [29, 30, 32] increasing the dose of activated charcoal can increase metal adsorption.

The life of a shellfish during the depuration process is influenced by water quality. Water quality that affects the life of the cockles includes temperature, dissolved oxygen, salinity, and pH [36]. Water temperature has a considerable influence on the metabolism of aquatic organisms. In the temperature range of 18-32°C, shellfish metabolism occurs well [37]. During the depuration process, the temperature in the water media ranges from 26.9-28.6°C according to the optimal temperature range for shellfish life between 25-32°C [38].

DO values in this study ranged from 4.66 to 5.97 mg/L. The use of aerators during the depuration process is an effort to obtain and maintain optimal DO for the metabolism of shellfish. According to [10], respiration with abundant oxygen will make the shellfish metabolic process take place well. Lee *et al.* [12] and [39] also stated that the dissolved oxygen content in water by ± 5 mg/L during the process of depuration of the shells took place could facilitate the metabolic activity of shellfish well.

At the beginning of the depuration process, the pH value is above 7.0 and at the end of the depuration process, the pH value tends to decrease to 6.8. This happens because the filter material used has acidic properties. The functional groups present in the filter material having the largest acid content are the carboxyl group and the phenyl group [25]. The acidic properties of the filter material

make the pH of the water decrease. The pH range of water during the depuration process is still within tolerance for marine life, which is between 6 - 9 (Decree of the Minister of Environment – 02/1988).

Salinity is a parameter of water quality that shows the level of salt in water. Salinity in estuarine waters tends to be easy to change [40]. During the depuration process, the salinity of the water used was 30 ppt. According to [41], water salinity of 30 ppt shellfish has a very high survival rate.

4. Conclusion

The duration of the depuration process affects the efficiency of Pb metal reduction in the cockles. The depuration process for 24 hours with the activated charcoal filter was able to reduce the Pb metal content to 0.926 mg/kg and even the depuration process for 72 hours, the efficiency of the depuration process increased to 86.52% so that the Pb content in the cockles became 0.273 mg/kg.

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