The Impact of ^{242}Pu Speciation on the Bioaccumulation of Plutonium by *Babylonia spirata* from Jakarta Bay

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Abstract: *The research of bioaccumulation Plutonium of* Babylonia spirata *from Jakarta Bay using ^{242}Pu radiotracer has been conducted. The aquaria experiments were applied by two oxidation states of Pu speciation with three replications. The experiment was carried out by 2 steps, such as uptake and depuration. The bioavailability of ^{242}Pu in the (III) and (IV) oxidation states from the sea has been studied for* Babylonia spirata. *Biokinetics parameters, such as concentration factors (CF_{ss}), uptake rate constants (k_u), elimination rate constants (k_e), bioconcentration factors (BCF), and biological half-life ($t_{b1/2}$), were investigated. The dissection is carried out to separate the parts of the gastropod's body so that the target organs for Plutonium contamination can be identified. Each part of the gastropod's body was prepared radiochemically and analyzed using an alpha spectrometer. The results show that Pu^{4+} is potentially accumulated in more significant value than Pu^{3+} by* B. spirata, *in which Pu is more rapidly distributed and retained longer in the proboscis and gastrointestinal tract.*

Keywords: Babylonia spirata*; bioaccumulation; Jakarta Bay; Plutonium; speciation*

■ INTRODUCTION

Plutonium is a transuranic element whose isotope, ^{239}Pu, is one of the three primary fissile isotopes (^{233}U and ^{235}U are the other two) with a half-life of 24,000 years [1]. Pu emission can spread to the environment and be deposited in soil and water. The ocean acts as a dispersant and accumulates radionuclides. The total release of Plutonium in the Pacific Ocean as a result of a nuclear facility accident and world nuclear weapon testing is estimated at 6.3 pBq [2]. The distribution of plutonium in the ocean is affected by particle formation and radioactive decay.

Plutonium entering the waters will interact with sediment and suspended particles. The food chain plays an important role as a tracer factor and a radionuclide cycle in ocean waters because each chain can absorb radionuclide, and radionuclide migration between the chains has a relatively fixed absorption factor [3]. A bioindicator is used to monitor the level of Pu contamination in marine waters through marine biota sampling in a regular time interval and measurement of Pu concentration in the body of the biota.

Anthropogenic radionuclide such as $^{239/240}$Pu has high toxicity and can be ingested into the human body through contaminated water, soil, and sediments, as well as ingestion of contaminated food, inhalation, and in some cases a dermal absorption pathway, can also be considered [4].

The increased number of Pu concentrations shows the ability of marine biota to accumulate Pu by multiplying the factor in the body of marine biota as Pu movement from environment to marine biota through various exposure paths. The accumulated potential of Pu in the body of marine biota is determined as the Bioconcentration Factor (BCF) obtained by comparing the Plutonium concentration in the biota to the Plutonium concentration in seawater. However, the data

are not sufficient to describe the overall dynamic process of bioaccumulation of plutonium that is also influenced by environmental factors and biotic factors. Pu bioaccumulation by marine biota is affected by its bioavailability towards an organism's body tissues, the chemical properties of Pu, and the metabolic system of contaminated biota [5]. The bioaccumulation factor (BAF) is determined as the main parameter that describes Pu's transfer from marine waters to the human body through the food chain. Plutonium in marine waters can enter the human body by consuming food from the sea, such as fish, shellfish, and seaweeds, with varying concentration factors of radionuclide [6].

Tiger snail (*Babylonia spirata*) is one of the gastropods from the mollusk phylum with high economic value and can accumulate various types of heavy metals and radionuclides contained in marine waters. *B. spirata* is utilized from its meat through its shell. Meat used as a source of food (seafood) is popular, operculum for medicinal materials, and shells for the manufacture of ornaments [7]. Habitat *B. spirata* is a sandy or muddy seabed at 5–20 m depth with a relatively settled life and limited mobility. *B. spirata* distribution is abundant and evenly distributed across several Indo-Pacific coastlines, including in some coastal areas of Indonesia. Based on its behavior, spread, and capability to accumulate contaminants, *B. spirata* can be used as a Plutonium bioindicator [1,8].

The study of bioaccumulation of radionuclides and other contaminants that is based on aquaria experiments is still limited. The biokinetic study was reported for the bioaccumulation of ^{137}Cs by *Chanos Chanos* and by *Cyprinus carpio* [9-10]. The study of bioaccumulation of chloramphenicol by *Paneous monodon* and heavy metal by *Perna Viridis* has been reported [11-12]. However, the number of Plutonium bioaccumulation studies by marine biota is still limited. Experiments on the dynamics of accumulation and elimination of plutonium were carried out in marine mollusks and wildlife mammals [13-14].

Nevertheless, these studies were conducted for environmental monitoring and based on large concentrations of biokinetics [15-16]. Furthermore, no bioaccumulation study of plutonium-based on its chemical speciation in *B. spirata* has never been carried out. In this research, the impact of Pu speciation by *B. spirata* will be observed using a concentration corresponding to its concentration in the environment. In addition, the bioaccumulation of Pu by *B. spirata* was quantified through this study using the single-compartment model.

■ EXPERIMENTAL SECTION

Materials

The materials used in this experiment were HCl, $NH_2OH{\cdot}HCl$, $FeCl_3$ and $NaNO_2$, NH_4OH, HNO_3, H_2SO_4 (E. Merck, Germany), radiotracer ^{232}Pu (Eckert & Ziegler Analytics), and stainless steel plate (Canberra, USA).

Instrumentation

The instrumentations used in this experiment included Alpha spectrometer model 7401 Canberra equipped with a vacuum chamber, PIPs detector, HV bias supply, preamp-amplifier, pulser-discriminator, counter, and digital display. This system was connected to a PC that is operated by Genie 2000 basic software.

Procedure

Biota sampling was referred to Johansen et al. [14]. Sampling was conducted in August 2017. Some of *B. spirata* were collected from farming sites in Tanjung Kait, Jakarta Bay.

Acclimatization [14]

A total of 80 *B. spirata* were cleaned and placed into an aquarium containing 250 L of seawater equipped with the filtration and aeration system to maintain the quality of water and the amount of oxygen. In addition, the measurements of physical conditions of seawater, such as pH, temperature, and salinity, were performed to ensure that no effect from all three factors during the bioaccumulation experiment tainted the result. The acclimatization was done by maintaining the *B. spirata* for 7 days in contaminant-free seawater. Seawater replacement was performed every 3 days. Feeding of fish pellets was done in the morning and the evening. The aquarium was given 12 h of light and then was left in the dark for the following 12 h during the acclimatization

process. Experiments can be continued if the number of marine biota during the acclimatization process was more than 80% alive.

Bioaccumulation [14]

Bioaccumulation experiments and measurements of Plutonium content were performed in the aquatic laboratory of PTKMR BATAN using an alpha spectrometer. The bioaccumulation experiment was carried out with various species of $^{242}Pu^{3+}$ and $^{242}Pu^{4+}$ through seawater on *B. spirata* with three repetitions.

Uptake $^{242}Pu^{3+}$ and $^{242}Pu^{4+}$ through seawater [14]

After undergoing acclimatization, *B. spirata* was transferred into a jar containing 2 L of filtered seawater using a 0.2 μm filter at pH 7 (normal seawater) and containing 1 mL of $^{242}Pu^{4+}$ 0.74 Bq mL^{-1}. Each jar contained three *B. spirata* and was equipped with an aerator. A similar process was also performed on *B. spirata* contaminated by $^{242}Pu^{3+}$ (obtained from reducing $^{242}Pu^{4+}$ using $NH_2OH{\cdot}HCl$ with heating). The uptake process was conducted for 18 days. One snail was taken from each jar every day and dissected to obtain: (1) gills and kidneys, (2) proboscis and gastrointestinal tract, and (3) shells and remainders; then, it was weighed.

Furthermore, the samples were prepared and analyzed for Pu content using an alpha spectrometer. Determination of the ability of *B. spirata* to accumulate $^{242}Pu^{3+}$ and $^{242}Pu^{4+}$ through seawater was derived from Eq. (1). Concentration factor (CF) was the ratio between Pu concentration in the biota (Bq g^{-1}) and the Pu concentration in seawater Bq mL^{-1}), while the uptake rate constant (k_u) was obtained as the slope of the concentration factor over time curve [17]

$$CF_t = CF_{ss}(1 + e^{-ku.t}) \quad (1)$$

The depuration of $^{242}Pu^{3+}$ and $^{242}Pu^{4+}$

B. spirata from the uptake process was transferred to an aquaria system containing contaminant-free seawater under flowing conditions (50 L/h debits). Feeding, sampling, dissection, and weighing of biota samples were performed the same as previous treatment for 7 days during the depuration process. Furthermore, the samples were prepared and analyzed for Pu content using an alpha spectrometer. According to Whicker and Schult equation [18], the loss was expressed in terms of the percentage of remaining radioactivity (i.e., radioactivity at time t divided by initial radioactivity (t_0) measured in the whole body biota at the end of the uptake period, multiplied by 100). The losses were described by:

$$A_t = A_0 e^{-ke.t} \quad (2)$$

where A_t and A_0 are the remaining activities (%) at time t (days) and 0 days, respectively. k_e is the biological loss rate constant (day^{-1}). The determination of k_e allows the calculation of the radionuclide biological half-life:

$$t_{1/2b} = \frac{\ln 2}{k_e} \quad (3)$$

The Pu preparation and determination method is based on the IAEA procedure with modifications [19]. The biota samples were cleaned and burned at 550 °C for 7 h using a furnace, then being cooled in the atmosphere for 12 h. Ash was dissolved in HCl (12 M) by heating, and then $NH_2OH{\cdot}HCl$ was added. The hot solution was added with $FeCl_3$ and $NaNO_2$; then, we waited until the vapor was gone. The addition of NH_4OH (1:1) and HCl was performed to adjust the pH of 6–7. It was characterized by the formation of brown granules, which are precipitated. The sample was decanted to separate the precipitate from the solution. The precipitate was diluted with HCl (12 M), then HNO_3 (14.6 M) and followed by evaporation. Furthermore, the sample was added with HNO_3 (8 M) and $NaNO_2$ by heating. The solution was filtered, and the filtrate was evaporated and dissolved in H_2SO_4 5%. As many as 3 drops of the thymol blue indicator and a few drops of NH_4OH were added until the color changed from pink to yellow, and then we added a few drops of H_2SO_4 5% until it changed from yellow to pink. The solution was electrodeposited for 2 h in 1 A of current at pH 2 on a stainless steel planset. The planset was washed with aquades and dried in oven at 100 °C for 5 min, and then counted by using an alpha spectrometer [19].

■ RESULTS AND DISCUSSION

The uptake process of ^{242}Pu as Pu^{3+} and Pu^{4+} by *B. spirata* through the seawater for 18 days is shown in Fig 1. The uptake rate constant (k_u) describes the speed of Pu distribution to various organs of *B. spirata* as the

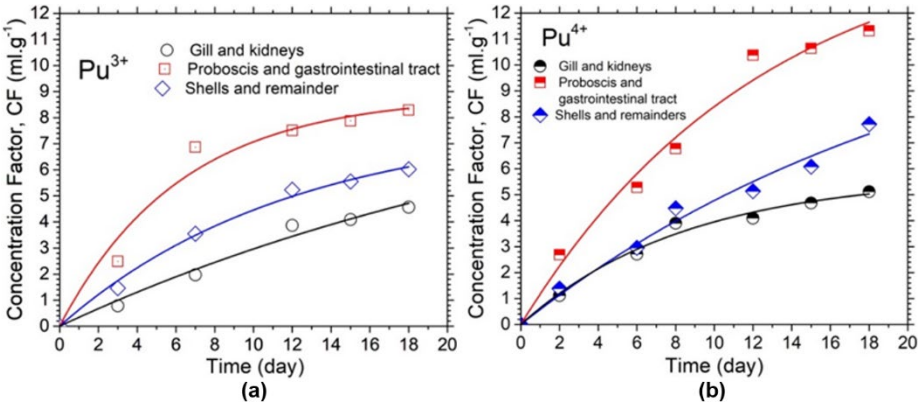

Fig 1. Bioaccumulation of Pu^{3+} and Pu^{4+} by *B. spirata*

interaction of the physiological factor of the biota with Pu. In this research, ^{242}Pu being used is the Pu^{3+} and Pu^{4+} resulting in Pu of the one entering the biota body is a single-form ion. The physiological factor, including the metabolism process of contaminants in the body of the biota, is assumed to be constant.

The experiment reveals that Plutonium in Pu^{3+} and Pu^{4+} forms are distributed more rapidly in the proboscis and the gastrointestinal tract, followed by shells and remainders, and then in gills and kidneys (Table 1). Fig. 1 shows that the ability of Plutonium accumulation for Pu^{3+} and Pu^{4+} is significantly different in the proboscis and gastrointestinal tract and shells and remainders, but not in gills and kidneys. Plutonium in Pu^{4+} form is potentially accumulated in a more significant number compared to Pu^{3+} for all organs.

The elimination process of Plutonium as Pu^{3+} and Pu^{4+} is shown in Fig 2. The results of the depuration experiment show the significantly different elimination rate in gills and kidneys, also in the proboscis and gastrointestinal tract, but almost no difference in shells and remainders. Pu^{3+} can be eliminated faster from gills and kidneys, also from proboscis and gastrointestinal tract by *B. spirata*.

The bioaccumulation capability of Plutonium as Pu^{3+} and Pu^{4+} through seawater can be observed more comprehensively based on BCF values obtained as the ratio between the uptake rate constant (k_u) and the elimination rate constant (k_e). Table 1 shows a higher BCF value for Pu^{4+} than Pu^{3+}. In addition, the BCF value in the proboscis and gastrointestinal tract is much higher than in gills and kidneys, in shells and remainders for both Pu speciation. A component is considered to be bioaccumulative when the BCF value exceeds a specific value threshold. Depending on the regulatory framework, the bioaccumulation threshold value ranges from 500 to 5000 [20]. Table 1 shows that the value of BCF for Pu^{3+} and Pu^{4+} by *B. spirata* was under 500; therefore, *B. spirata*

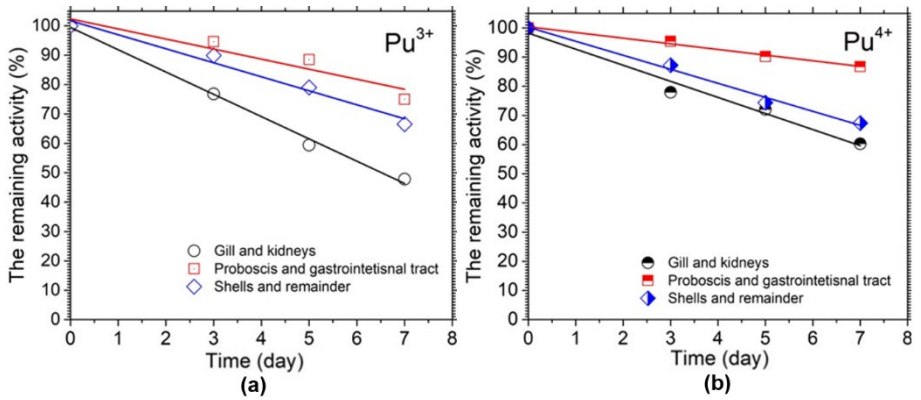

Fig 2. The elimination process of ^{242}Pu by *B. spirata*

Table 1. Summary of biokinetic parameters of Plutonium bioaccumulation (as ^{242}Pu) by *B. spirata* through seawater

Pu Speciation	Body fractions	CF_{ss} (mL g^{-1})	k_u (day^{-1})	k_e (day^{-1})	BCF	$t_{b1/2}$ (day)
Pu^{3+}	Gills and kidneys	4.57	0.268	0.076	3.53	9.1
	Proboscis and gastrointestinal tract	8.29	0.447	0.034	13.06	20.3
	Shells and remainders	6.02	0.339	0.048	7.11	14.5
Pu^{4+}	Gills and kidneys	5.11	0.274	0.055	4.97	12.6
	Proboscis and gastrointestinal tract	11.32	0.636	0.019	32.78	35.7
	Shells and remainders	7.72	0.400	0.048	8.34	14.4

cannot be used for environmental risk assessment [20]. The experimental results show that Pu^{3+} and Pu^{4+} retained the longest in the proboscis and gastrointestinal tract and the shortest in gills and kidneys. While in the shells and remainders, Pu^{3+} and Pu^{4+} have biological half-lives that do not differ significantly. Table 2 shows a comparison of Concentration values of various marine organisms. Base on these data, the Concentration Factor values in various marine organisms are very widely in a reasonably wide range.

Plutonium is an actinide metal with electron configuration of [Rn] $5f^6$ $7s^2$. The core structure of Pu is known clearly along with the development of nuclear physics, and then it is known that Pu is a radioactive element consisting of various isotopes. Twenty radioactive isotopes of plutonium have been characterized. The longest-lived is ^{244}Pu with a half-life of 80.8 million years, ^{242}Pu with a half-life of 373,300 years, and ^{239}Pu with a half-life of 24,110 years. All of the remaining radioactive isotopes have half-lives that are less

Table 2. The value of CF_{ss} from another study of Plutonium bioaccumulation by mollusks through seawater

Biota and Radiotracer	Concentration Factor (mL g^{-1})		Time (days)	Reference
Venerupis decussata				
(^{237}Pu(III)+(IV))	Whole body	74 ± 5	22	[34]
		140	17	
(^{237}Pu(V)+(VI))	Whole body	61 ± 1	22	
Hermione hystrix				
(^{237}Pu(III)+(IV))	Whole body	370 ± 10	22	[34]
		130	20	
(^{237}Pu(V)+(VI))	Whole body	275 ± 11	22	
Mytilus galloprovincialis	Soft tissues	28	21	[35]
(^{239}Pu)	Shell	148		
Tapes decussatus	Soft tissues	18	21	[36]
(^{239}Pu)	Shell	165		
Scrobicularia plana	Soft tissues	38	21	[37]
(^{239}Pu)	Shell	200		
Babylonia spirata				
(^{242}Pu(III))	Gills and kidneys	4.57	18	Present study
	Proboscis and gastrointestinal tract	8.29		
	Shells and remainders	6.02		
(^{242}Pu(III))	Gills and kidneys	5.11	18	
	Proboscis and gastrointestinal tract	11.32		
	Shells and remainders	7.72		

than 7,000 years. This element also has eight metastable states, though all have half-lives less than one second [21].

Plutonium is primarily derived from nuclear testing and waste from fuel elements. ^{239}Pu is one of the three primary fissile isotopes synthesized the most (^{233}U and ^{235}U are the other two) [22]. In a reactor, more than 50% of ^{239}Pu will undergo fission, and a small part of it captures a neutron and produces ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{243}Pu [23]. The concentration of ^{239}Pu in nature is so small that its existence can be ignored. The ^{239}Pu is derived from Uranium ores that capture neutrons as a result of spontaneous fission of Uranium in nature, and also it comes from the decaying of Cm in a tiny amount [24].

The first nuclear experiment was conducted in the New Mexico Desert, the USA, on May 16, 1945, followed by dropping atomic bombs on Hiroshima-Nagasaki on August 6, 1945. The highest frequency of nuclear experiments was conducted by developed countries, such as the USA, USSR, UK, and France, from 1954 to 1964. Initially, nuclear experiments were openly conducted in the atmosphere, but since 1979 nuclear experiments were conducted underground. Nuclear testing in previous decades has resulted in the release of Pu into the atmosphere, which is then distributed by the wind as a fall-out away from the source, especially in the northern hemisphere. Other sources of Pu come from Chernobyl and Fukushima Dai-ichi accidents of the past few years.

For multivalent elements such as plutonium, its behavior and complex stability in the environment are quite varied. Releases from reactors may contain Pu incorporated in fuel particles, Pu-colloids, oxidized forms (Pu(V, VI)), reduced forms (Pu(III, IV), or organic complexes. How the Pu partition itself between possible species in any given environmental system is known to depend upon the chemical form in which it enters the system, redox conditions, pH, ionic strengths, the type and concentration of organic and inorganic complexing agents, and the extent of hydrolytic colloid formation [25].

The ocean is where the radionuclide accumulated from direct releases of nuclear installations, land deposition from the geomorphology process, atmospheric fall-out and cosmic ray exposure from outer space, and nuclear accidents [26]. In seawater, Pu naturally forms cations, such as $Pu(III)^{3+}$, $Pu(IV)^{4+}$, $Pu(V)O_2^+$, $Pu(VI)O_2^{2+}$, and $Pu(VII)O_4^-$ anion [27]. $Pu(VII)O_4^-$ is highly unstable and tends to be reduced to $Pu(VI)O_2^{2+}$, while $Pu(III)^{3+}$ is instantly oxidized to $Pu(IV)^{4+}$, and it causes plutonium in seawater to be dominated by $Pu(IV)^{4+}$, $Pu(V)O_2^+$, and $Pu(VI)O_2^{2+}$ [28].

Pu(III), Pu(IV), Pu(V), and Pu(VI) are present in the waters simultaneously according to the equilibrium reaction:

$$3PuO_2^+ + 4H^+ \rightarrow Pu^{3+} + 2PuO_2^{2+} + 2H_2O \quad (4)$$

$$Pu^{4+} + 2H_2O \rightarrow 2Pu^{3+} + PuO_2^{2+} + 4H^+ \quad (5)$$

Radioisotope Pu^{5+} and Pu^{6+} are insoluble in water and precipitated on marine sediments. However, when the nitrite concentration is high, Pu^{5+} and Pu^{6+} can be reduced to $Pu^{4+,}$ which is more soluble in water. Nitrite in seawater can be produced from the decomposition of dissolved organic compounds [29].

The conversion rate of Pu is varied from one oxidation number to another. The redox reaction between $Pu(III)^{3+} \rightarrow Pu(IV)^4$ and $Pu(V)O_2^+ \rightarrow Pu(VI)O_2^{2+}$ is generally faster because it is only through the electron exchange process. Meanwhile, the reaction between $Pu(IV)^{4+} \rightarrow Pu(V)O_2^+$ and $Pu(IV)^{4+} \rightarrow Pu(VI)O_2^{2+}$ is slower because it undergoes forming and breaking the Pu-O bond process [30]. The behavior of Pu in the solution is affected by its oxidation number. The tendency of Pu to hydrolyze is reduced by order of oxidation numbers IV << VI ≤ III < V. Plutonium is made in $Pu(IV)^{4+}$ form to reduce its solubility. Pu^{3+} and Pu^{4+} are strongly bonded to the particles compared to PuO_2^+ and PuO_2^{2+}. Thus, Pu with various oxidation states can form many form complexes can affect their solubility [31].

Bioaccumulation is a complex and dynamic process but can be explained by the model constructed from experimental results. The single-compartment model is widely used for various aquatic species by providing mathematical explanations on the number of contaminants that are determined by the uptake rate and elimination rate. Biokinetic parameters of the bioaccumulation process consist of the concentration factor (CF_{ss}), uptake rate constant (k_u), elimination rate constant (k_e), bioconcentration factor (BCF), and biological half-life ($t_{b1/2}$).

The ability of *B. spirata* to accumulate Plutonium as Pu^{3+} and Pu^{4+} in the environment is influenced by internal and external factors. Internal factors are the active side of cell membranes belonging to *B. spirata* as metal-binding molecules, while external factors are pH, salinity, particulate content, and dissolved organic matter content [30,32]. The organic particulates are eliminated by filtering the seawater, while other external factors are ignored because the seawater comes from the waters of Jakarta Bay [33].

■ CONCLUSION

Bioaccumulation of Plutonium by *B. spirata* through seawater is affected by its speciation, where Pu^{4+} has higher bioavailability than Pu^{3+}. It is shown by concentration factors (CF_{ss}) and uptake rate constant (k_u) of $Pu^{4+,}$ which is much higher in gills and kidneys, also in the proboscis and gastrointestinal tract, but almost similar in shells and remainders. Plutonium is distributed more rapidly and retained longer in the proboscis and gastrointestinal tract for both Pu speciations.

B. spirata can be used as a bioindicator of plutonium-based on its ability to accumulate heavy metals and radionuclides in the environment. However, *B. spirata* as a bioindicator has a BCF value less than 500; therefore, this biota cannot be used for environmental risk assessment.

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■ AUTHOR CONTRIBUTIONS

BB, MWA, WRP, and HS are all playing roles as the main contribution of this study due to their expertise and knowledge.

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