International Journal of Agricultural Technology 2019 Vol. 15(2): 207-214
Available online http://www.ijat-aatsea.com
ISSN: 2630-0613 (Print) 2630-0192 (Online)

The Application of tannin extract from plants to reduce the concentration of arsenic

Aroonsrimorakot, S.[1*] and Whangchai, N.[2]

[1]Faculty of Environmental and Resource Studies, Mahidol University, Thailand; [2]Faculty of Fisheries Technology and Aquatic Resources, Maejo University, Thailand.

Aroonsrimorakot, S. and Whangchai, N. (2019). The application of tannin extract from plants to reduce the concentration of arsenic. International Journal of Agricultural Technology 15(2): 207-214.

Abstract This research prepared the tannins extract from agricultural wastes i.e. rice straw, tea leaves, longan leaves and banana leaves with fresh water for 2 months. The highest concentration of tannin was banana leaves (6,464.98 mg/L) followed by longan leaves (4,478.99 mg/L), rice straw (4,000.00 mg/L) and tea leaves (1,397.95 mg/L) respectively. Tannins extract were used for arsenic treatment in synthetic waste water. The result showed the efficiency of arsenic removal by 10, 20, 30, 40 and 50 mg/L of tannins concentration from banana leaves were 50.82%, 52.54%, 54.56%, 58.42% and 51.01% respectively, tea leaves were 69.74%, 73.42%, 60.29%, 63.60% and 70.54% respectively, longan leaves were 61.03%, 56.07%, 54.36%, 40.01% and 51.80% respectively and rice straw were 53.68%, 46.70%, 55.23%, 49.22% and 54.11% respectively which indicated that there were not significantly differed to arsenic reducing. In addition, increasing of pH in solution from 2.5 to 5.5, 6.5, 7.5 and 8.5 led to decrease of arsenic removal efficiency.

Keywords: Tannin extract, Agricultural wastes, Arsenic

Introduction

The expansion of community and development of technology has led to increase of the discharge of wastewater, which is leading to a serious environmental problem. Heavy metals in wastewater from plating, paint manufacturing and mining etc. cause the presence of high toxicity to human, animals, plants and environment, that can be accumulated in living organism and transfer in food chain (Mishra, 2014; Wan Ngah and Hanafiah, 2008). Arsenic is one of the most hazardous heavy metal. The toxicity of arsenic can cause acute and chronic health effects such as hyperkeratosis and pigmentation changes, circulatory disorders, diabetes and cancer in the liver, lungs, skin, bladder, kidneys (Bang *et al.*, 2005; Zhao *et al*., 2012).

To date, several techniques for heavy metal removal from wastewater have been established such as chemical precipitation, ion exchange, membrane

[*]**Coressponding Author:** Aroonsrimorakot, S.; **Email:** sayamthai88@gmail.com

filtration, electrochemical, and reverse osmosis (Arief *et al.,* 2008). Even though, these processes have high efficiency to reduce metal ion, they are usually difficult to apply in developing countries due to the limitations of high cost for setting and maintenance system. Biosorption is a good alternative for removal of heavy metals in wastewater because it is inexpensive and highly efficient (Diola and Orozco, 2014). In the recent time, researchers are interested in the application of biosorption process with tannin extract because it can be used as an effective agent in water treatment in developing countries (Mailoa *et al.,* 2013). Tannin is generally defined as polyphenol compounds that can be found in different parts of plants and agricultural waste such as seeds, roots and leaves (Sengil and Ozacar, 2009). The structure of tannin can be divided into two classes as: 1) Hydrolyzed tannins and 2) Condensed tannins (Figure 1) (Huang *et al.,* 2009). Recently, many reports presented the high efficiency of tannins to reduce the concentration of various heavy metals such as Cd^{2+}, Cu^{2+}, Hg^{2+}, Pb^{2+}, Ni^{2+} and Zn^{2+} (Sengil and Ozacar, 2009; Huang *et al.,* 2009; Heredia and Martin, 2009). Therefore, tannins extraction from 4 types of plants as banana leaves, longan leaves, tea leaves and rice straws were studied in this research. The details of each tannin extracted were presented to see its efficiency to reduce the concentration of arsenic (III) ion in synthetic wastewater.

In this study, we aimed to extract tannin from 4 types of plants as banana leaves, longan leaves, tea leaves and rice straws**.** To study the effect of tannin concentration on arsenic reduction. To study the effect of pH in solution on arsenic removal.

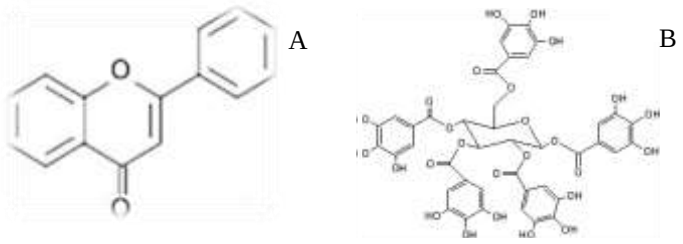

Figure 1. Structures of tannin: A= Hydrolyzed tannins and B = Condensed tannins (Ramkul, 2010)

Materials and methods

Tannin extraction

Maceration extraction is the method for extracting tannins from banana leaves, longan leaves, tea leaves and rice straws. 5.0 Kg of each material were

soaked in 100 L of fresh water for 2 months at room temperature and filtered under vacuum using a Buchner funnel. The polyphenols in tannins extract were reacted by Folin & Ciocalteaus reagent for 95 minutes to form a blue complex (Figure 2) that can be quantified by visible-light spectrophotometry (Blainski *et al.*, 2013).

Figure 2. Tannins analysis by Folin & Ciocalteaus reagent method

Preparation of synthetic wastewater

Stock solution of heavy metal ions was prepared by diluted arsenic standard solution (1000 mg/L) with deionized water (DI) to produce a 10 mg/L concentration for the experiment.

Effect of tannin concentration on arsenic removal

The experiment of arsenic removal was conducted by mixed 10 mg/L of arsenic solutions with 0, 10, 20, 30, 40 and 50 mg/L of tannins extract from banana leaves, longan leaves, tea leaves and rice straw respectively (Figure 3). The mixed solutions were filtered through a 0.45 µm nylon filter after 30 minutes at room temperature and the residual of arsenic concentration in the filtrate was estimated by Atomic Absorption Spectrometer (AAS) (APHA, AWWA and WEF, 1995).

Effect of pH on arsenic reduction

The effect of pH on arsenic removal with tannin extract was studied by varying the pH of arsenic solution from 2.5 to 5.5, 6.5, 7.5 and 8.5 respectively. 10 mg/L of tannin extract from each material were added into arsenic solution and mixed to make it homogeneous. The homogeneous solution was filtered by 0.45 µm nylon filter after 30 minutes and the residual of arsenic was analyzed by AAS.

Statistical analysis

The concentrations of As^{3+} in three replicate were presented as mean value ±SD. Analysis of Variance (ANOVA) was employed for the comparison among groups. Least Significant Difference (LSD) post hoc comparison was tested to show the differences between groups.

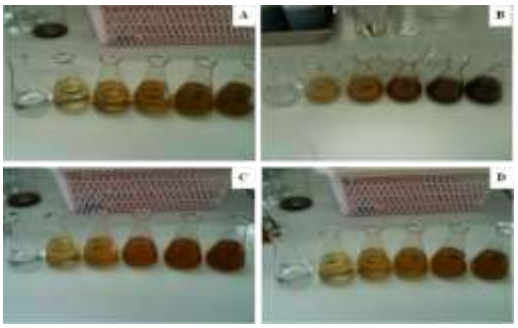

Figure 3. Arsenic treatments with tannins extract from A = banana leaves, B = tea leaves, C= longan leaves and D = rice straws

Results

The quantity of tannins extract

The amounts of extracted tannin from banana leaves, longan leaves, tea leaves and rice straws were extracted by fresh water for 2 months at room temperature. The result showed that banana leaves are having the highest tannin concentration followed by longan leaves, rice straw and tea leaves respectively (Table1).

Effect of tannins extracts concentration

The effect of tannins extract concentration on arsenic removal in synthetic wastewater was shown in Figure 4. The tannins extract from each material were mixed with arsenic solution and analyzed residual of arsenic concentration by AAS. The result showed the efficiency of tannins from banana leaves, tea leaves, longan leaves and rice straws, in reducing arsenic were 50.81%±1.31, 69.74%±5.56, 61.02%±26.83 and 53.68%±23.88 respectively at 10 mg/L of tannin concentration. The ANOVA test was presented in Table 2. It can be seen that there was no significant differences (p-value > 0.05) in removal of arsenic with different ratio of tannin concentration.

Moreover the test of LSD post hoc presented no significant differences on tannins efficiency from tea leaves compared with banana leaves and longan

leaves (p-value > 0.05). However, the efficiency of reducing arsenic by tannins from tea leaves was different with tannins from rice straws due to p-value was less than 0.05.

Table 1. The amounts of extracted tannin from agricultural wastes

Materials	Amounts of tannin (mg/L)
- Banana leaves	6,464.98 ±23.45
- Tea leaves	1,397.95 ±16.98
- Longan leaves	4,478.99 ±59.61
- Rice straws	4,000.00 ±42.77

Table 2. ANOVA test of tannins efficiency from 4 plants

Source	Sum of squares	df	Mean square	F	p-value
Materials	2476.63	3	825.544	2.041	0.042
Concentration	218.186	4	54.546	0.135	0.968

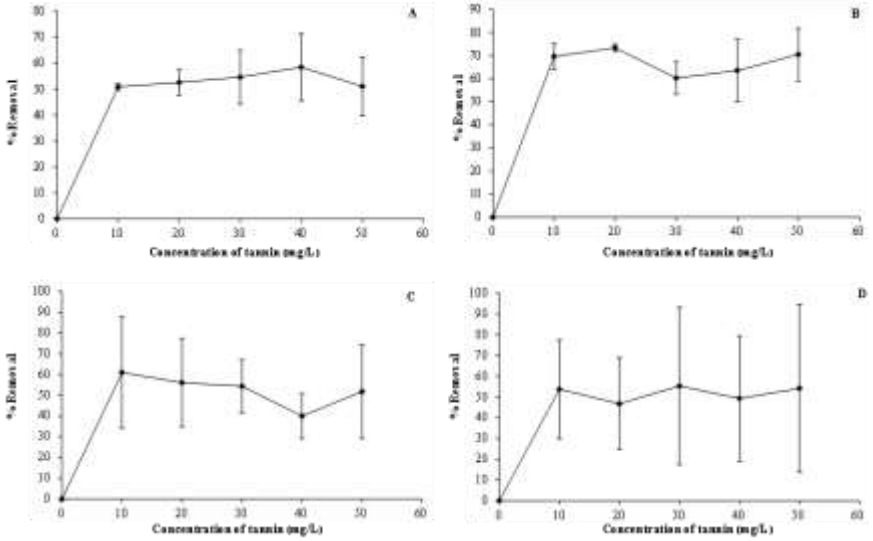

Figure 4. Effect of tannins concentration on arsenic removal:A = banana leaves, B= tea leaves, C= longan leaves and D =rice straws

The reaction mechanism is ion exchanged with hydroxyl functional group of polyphenols structures in tannins that can be seen as shown in equation 1 (R is the structure of polyphenol in tannin extract). That was similar to study of Nakano *et al.* (2001) who found the adsorption process between tannin and chromium ($Cr_2O_7^{2-}$) as ion exchanged with hydroxyl functional group (Nakano *et al.* 2001).

$$R\text{-}OH + H_2AsO_3^- \leftrightarrow R\text{-}H_2AsO_3 + OH^- \quad (1)$$

Effect of pH

The effect of pH in solution on arsenic removal was showed in Figure 5. That result showed the efficiency of arsenic reducing from each tannin extract. Banana leaves tannin able to reduce arsenic at pH 2.5 was 50.81%±1.31 which decreased to 37.10%±1.22, 40.40%±2.35, 38.17%±3.36 and 34.41%±2.42 with increasing pH to 5.5, 6.5, 7.5 and 8.5 respectively, similarly to tea leaves that can be reduced arsenic concentration was 69.74%±5.56, 34.30%±4.64, 37.7%±1.55, 39.89%±0.77 and 34.32%±3.27 at pH 2.5, 5.5, 6.5, 7.5and 8.5 respectively. While, longan leaves tannin at pH 2.5, 5.5, 6.5, 7.5 and 8.5 are able to reduce arsenic was 61.02%±26.83, 37.81%±0.40, 40.29%±3.85, 40.11%±3.60 and 39.29%±2.60 respectively and rice straws was 53.68%±23.88,37.04%±1.08, 39.54%±2.67, 44.11%±2.14 and 38.41%±1.74 respectively.

The statistical analysis is shown in Table 3. The result showed that there was no significant differences on the efficiency of tannin extract concentration on arsenic removal from the 4 types of plants. However, the effect of different pH values on arsenic removal was significant difference because p-value was less than 0.05. In addition, LSD post hoc showed tannin efficiency at pH 2.5 which indicates that there was significant difference with other pH in solution at 95% of confidence level.

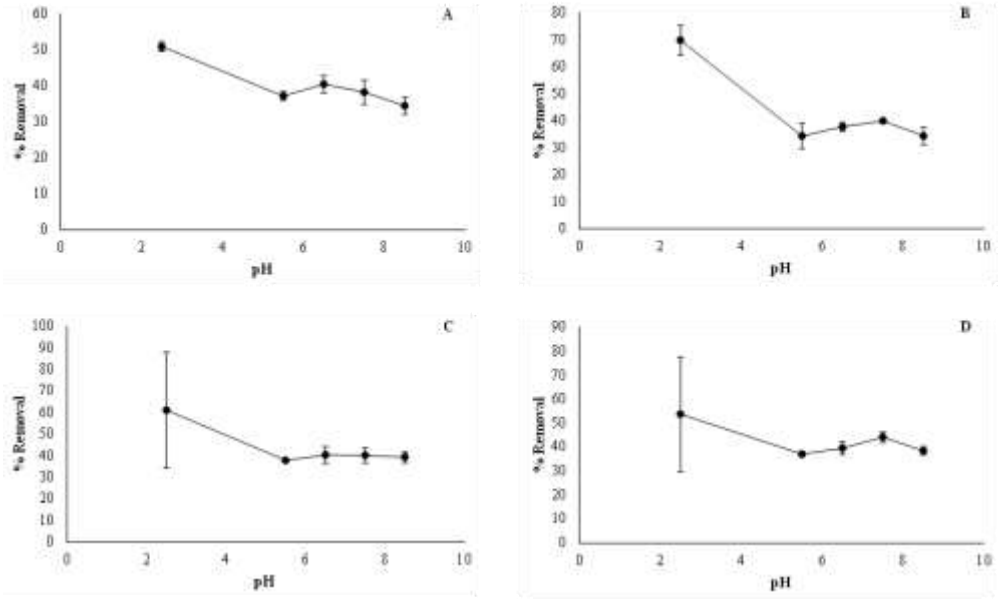

Figure 5. Effect of pH on arsenic removal by tannin extract from A = banana leaves, B= tea leaves, C= longan leaves and D =rice straws

Table 3. ANOVA test of arsenic removal at different pH values

Source	Sum of squares	df	Mean square	F	p-value
Materials	109.421	3	36.474	0.509	0.678
pH	4187.436	4	1046.859	14.607	0.000

Discussion

Extraction is the process to separate different substances in solubility of the two different liquids. The principle of extraction is the polar compounds dissolved in polar solvents and non-polar compounds dissolved in non-polar solvents (Mailoa *et al.,* 2013). The amounts of extracted tannin from agricultural wastes revealed that banana leaves were the highest tannin concentration followed by longan leaves, rice straw and tea leaves respectively. Each plants or different parts of plants had different amount of tannins such as Dwarf Cavendish Banana peels contain more tannins than Cultivated Banana peels and Pisang Maspeels while Black Tea leaves contain more tannins than Oolong Tea leaves and Green Tea leaves etc. (Khasnabis *et al.,* 2015; Surojanamethakul and Hiraga, 1994). In addition, the efficiency of tannins from banana leaves, tea leaves, longan leaves and rice straws was not significant differences in removal of arsenic. In accordance with Huang *et al*. (2009) found that a rise in the concentration of tannin from 1.0 g/L to 2.0, 3.0 and 4.0 g/L was no effect in reducing Hg2+. Moreover, the result of pH variation from each tannin extract, it can be seen that the efficiency of arsenic removal was decreased at basic (alkaline) condition consistent with Sornsathian *et al.* (2016) who found the efficiency of chromium removal by tannin are decreased with increasing pH value in wastewater.

Conclusion

Maceration extraction was used for extracting tannins from 4 types of plants as banana leaves, tea leaves, longan leaves and rice straws. Banana leaves were found with the highest tannin extract concentration followed by longan leaves, rice straws and tea leaves respectively. However, regarding the efficiency of tannin extracted from tea leaves, there was no significant difference with tannins extracted from banana leaves and longan leaves but different with rice straws. Moreover, raising tannin concentration from 10 mg/L to 20, 30, 40, and 50 mg/L does not lead to increase in percentage of arsenic removal because the ratio between arsenic concentration and tannin extracted were decreased. In addition, arsenic removal efficiency was decreased with increasing pH value in solution.

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(Received: 29 December 2018, accepted: 11 March 2019)