Efficiency and Kinetic Rate of Nanoscale and Microscale of Zero Valent Iron and Zinc Oxide particles for Chromium, Cadmium, and Lead Removal from Pak Thong Chai Silk Dyeing Effluent, Thailand

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Abstract: This present study involved the application and comparison of nanoscale particles in remediation for contaminated heavy metal effluent discharge. It aimed to investigate the removal efficiency and kinetic removal rate of the heavy metals including lead, chromium, and cadmium in silk dyeing effluent discharge by using nano and microscale of ZVI and ZnO particles. The contaminated samples were collected and later treated by one of the particles in the laboratory at 25 ºC. The mixtures were incubated for the required period before the heavy metal concentrations were measured. The overall results showed that the optimum dosage of all four particle types for heavy metal treatments was 60 mg/L with an optimal time of 60 minutes. The experiments with nZVI demonstrated the highest removal efficiency for all heavy metal types followed by that of nZnO, ZVI, and ZnO, respectively. The removal efficiency rate by all four type particles for Cr was higher than that of Cd and Pb, respectively. In addition to this, the kinetic removal rate of nZVI was also faster than that of nZnO, ZVI, and ZnO, respectively..

Index Terms: Kinetic rate, Nanoparticles, Heavy metal, nZVI, nZnO, Wastewater treatment.

1. Introduction

Several chemicals are currently used in industrial applications and everyday life. These chemicals are contaminated in environments, water, and soil [1]-[6]. As a consequence, the environments are contaminated with hazardous substances such as heavy metals. These heavy metals are widely used in many applications in both the industrial and agricultural sectors. For instance, lead and cadmium are used in battery manufacturing, metal smelting plant, metal plating factory, dyeing factory, and caustic soda production plant [7]. The textile dyeing industry is regarded as a medium-scale industry that involves a process of adding color to a yarn or fabric. It is later known to be a raw material for varieties of goods including garments. Chemicals and dyeing color reagents are used to add and stabilize the color. Wastewater quality from the textile dyeing industry sometimes fails to meet the effluent quality standard. For example, there are high organic substances, high BOD (100-1,000 mg/L) high COD (500-1,200 mg/L), high TDS, high TSS, and high heavy metal.

Lead and cadmium are known to be one of the major problems as pollutants that have effects on a human being. Industrial effluent discharge with higher lead concentration than effluent standards (0.2 mM per liter) has been previously reported. This wastewater discharge from factories certainly contains many types of heavy metals. Cadmium can be deposited in bones and kidneys. It leads to Itai-Itai Disease. The consumption of lead can result in anemia. It causes muscle weakness and paralysis.

Nanotechnology is involved in research, particularly in sciences and technology [8]. It is used to be an alternative method with higher efficiency [9]-[11] and a minimal time than that of biotechnology. Nanotechnology is well known as the process of creating or analyzing materials or products at the nanometer level (approximately 1-100 nm) [12]. Nanoparticles are increasingly involved in environmental concerns to remedy the contaminated environment. Nano-Zero valent iron (nZVI) is a type of nanoparticles that are used to treat chemicals and hazardous contaminants in soil, water, and air [13]-[17]. Nano Zinc oxide (nZnO) is commonly known as the catalyst and photo-catalyst [18]-[20].

This current study focused on the possibility of nanotechnology (nZVI and nZnO) that can be applied for remediation of the effluent discharge from the silk dyeing process. The objective of this study involved the application and comparison of the efficiency of zero-valent iron (ZVI) and zinc oxide (ZnO) both at a micro (normal size) and nanoscale in the heavy metal removal from the effluent discharge of silk dyeing process. This research aimed to study the removal efficiency and kinetic removal rate of the heavy metals including lead (Pb), chromium (Cr), and cadmium (Cd) in silk dyeing effluent discharge by using nano and microscale of ZVI and ZnO particles.

2. Material and Method

A. The studies samples

The effluent discharge of the silk dyeing process was used as the studied samples contaminated with heavy metals. It was directly collected from one of the nearby textile dyeing factories in Nakhon Ratchasima province, Thailand, and transported to the laboratories for further experiments. The samples were treated by nanoscale zero-valent iron (nZVI), nanoscale zinc oxide (nZnO), microscale zero-valent iron (ZVI), and microscale zinc oxide (ZnO).

B. Chemical and Research Instruments

Nanoscale zero-valent iron (nZVI), nanoscale zinc oxide (ZnO), microscale zero-valent iron (ZVI), and microscale zinc oxide (ZnO) were used for treatments. Homogenizer (IKA: Disperser Model T25 digital Ultra-turrax (C-MAG HS7), pH meter (Index: ID 1000), Shaker (Forma Scientific: Model 4586), Microcentrifuge (Hettich: Model EBA 21), Atomic Absorption Spectrophotometer (Varian: Spectr AA 55B) were used for the experiments.

C. C. Research Methodology

The ability and efficiency of nZVI, nZnO, ZVI, and ZnO in the heavy metal removal from the effluent discharge of silk dyeing process was operated in volumetric flasks at 25 °C. The mixture between the effluent discharge and each type of those four particles was later incubated for a required period on the shaker.

The experiment was divided into 2 parts as described below.

I. Determination of the appropriate amount (Optimum dosages) of nZVI, nZnO, ZVI, and ZnO for heavy metal removal

The optimum dosages of nanoparticles were determined by varying concentrations of nZVI, nZnO, ZVI, and ZnO were prepared at 20, 40, 60, 80, and 100 mg/L. The contaminated samples were later added with one of these four particles at varying concentrations. The mixture was incubated in the volumetric flasks for a period of 60 minutes with gentle shaking while the controls were carried out without any particles. All samples were wrapped in aluminum foil. Then, the amount of residual heavy metals (Pb, Cr, and Cd) after the incubation was measured by Atomic Absorption Spectrophotometer (AAS). The experiments were performed with three replications. The results of the removal efficiency by particles with varying concentrations were calculated and presented using either bar or line graphs.

II.The removal efficiency and the kinetic removal rates of nZVI, nZnO, ZVI, and ZnO for heavy metal removal

The optimum concentration of particles from the previous experiment was used to study the removal efficiency and the kinetic removal rate. The contaminated samples were added with nZVI or nZnO, ZVI, or ZnO and incubated for a period of varying times. The incubation times with an hour interval were varied at 1, 2, 3, 4, and 5 hours. To obtain the optimum time, the experiments with incubation time at 10-minute intervals may be applied where appropriate. The samples were wrapped in aluminum foil. Then, the amount of residual heavy metals (Pb, Cr, and Cd) after the incubation was measured by Atomic Absorption Spectrophotometer (AAS). The experiments were performed with three replications. The results of the removal efficiency and the kinetic removal rates with varying times were calculated and presented using line graphs.

Kinetic Removal Rates were plotted as semi-log to determine the first-order reaction rate of removal degradation refer to (1) and (2):

(1) $$-\ln(Ce/Co) = kt$$

(2) $$k = (-\text{In}(Ce/Co))/T$$

where:

Ce is the concentrations in supernatant samples after reacting with nanoparticles (mg/L)

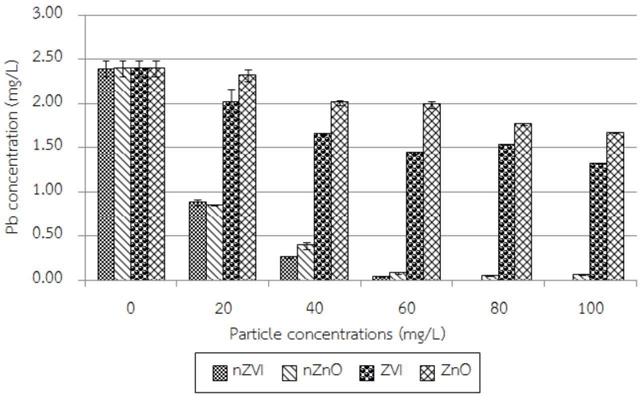
Co is the concentrations in supernatant samples before reacting with nanoparticles (mg/L)

k is the first-order reaction (min-1)

t is reaction time (min).

3. Results

A. Determination of the appropriate amount (Optimum dosages) of nZVI, nZnO, ZVI, and ZnO for heavy metal removal



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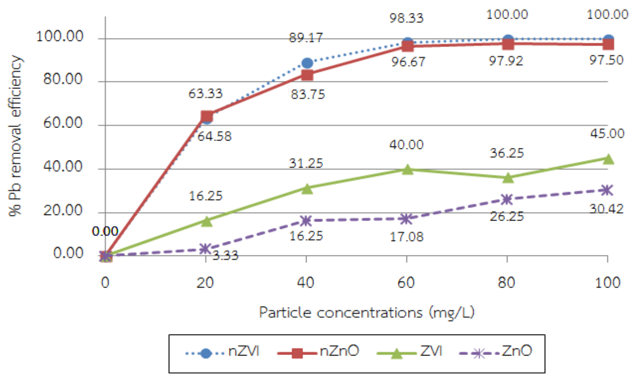
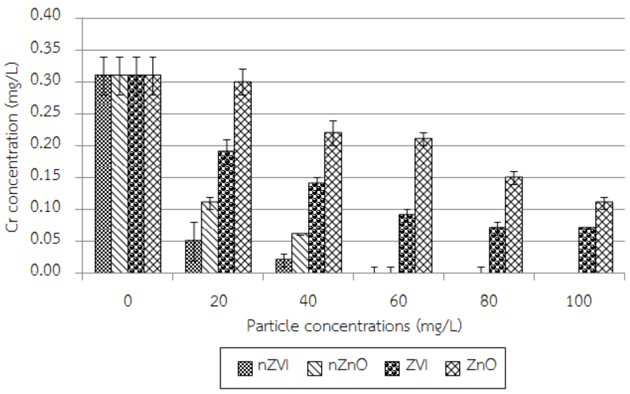


(b)

Fig. 1 Lead (Pb) treatments in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor (a) Concentrations of Pb (b) The efficiency (%) of Pb removal

The results of the removal efficiency of Pb by four types of particles with varying concentrations from 0 to 100 mg/L showed in Fig. 1. Generally, nZVI and nZnO demonstrated much higher efficiency than that of ZVI and ZnO. The overall results also indicated that the efficiency was increased according to particle concentrations. The efficiency by nZVI and nZnO with 60 mg/L concentration was found at 98.33 % and 96.67 %, respectively. Meanwhile, that with 80 mg/L and more showed 100 % removal efficiency. This efficiency was substantially higher than that of ZVI and ZnO. Though, the concentration of 60 mg/L was selected as an optimum dosage for Pb removal from the effluent discharge of the silk dyeing process.



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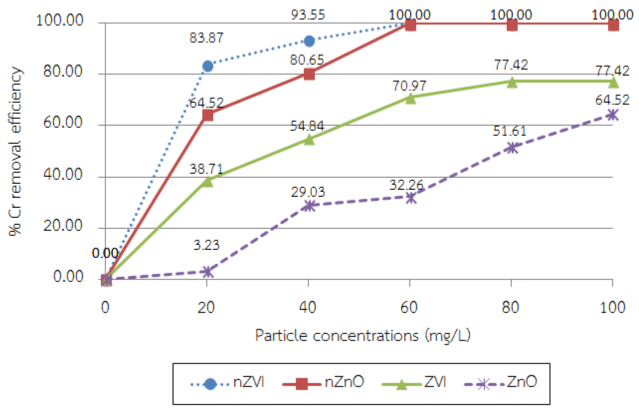
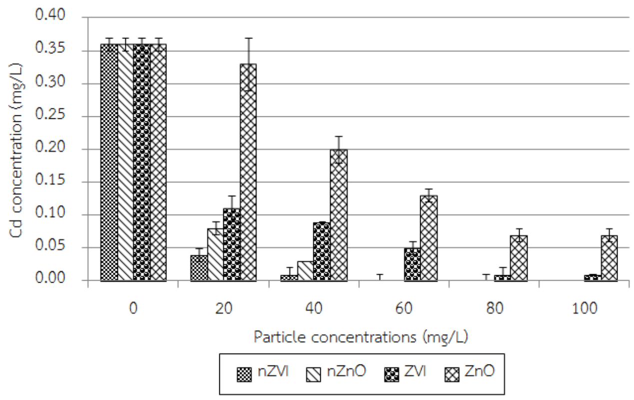


(b)

Fig. 2 Chromium (Cr) treatments in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor (a) Concentrations of Cr (b) The efficiency (%) of Cr removal

The results of the removal efficiency of Cr by four types of particles with varying concentrations from 0 to 100 mg/L showed in Fig. 2. Generally, nZVI and nZnO demonstrated much higher efficiency than that of ZVI and ZnO. The overall results also indicated that the efficiency was increased according to particle concentrations. The efficiency by nZVI and nZnO with 40 mg/L concentration was found at 93.55 % and 80.65 %, respectively. Meanwhile, that with 60 mg/L and more showed 100 % removal efficiency. This efficiency was substantially higher than that of ZVI and ZnO. Though, the concentration of 60 mg/L was selected as an optimum dosage for Cr removal from the effluent discharge of the silk dyeing process.



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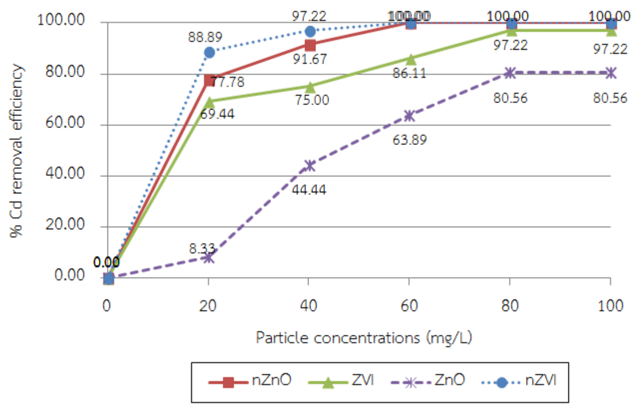
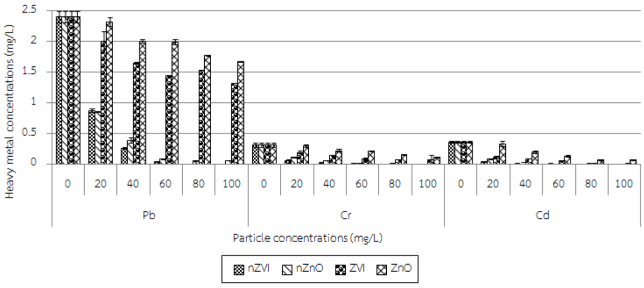


(b)

Fig. 3 Cadmium (Cd) treatments in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor (a) Concentrations of Cd (b) The efficiency (%) of Cd removal

The results of the removal efficiency of Cd by four types of particles with varying concentrations from 0 to 100 mg/L showed in Fig. 3. Generally, nZVI and nZnO demonstrated much higher efficiency than that of ZVI and ZnO. The overall results also indicated that the efficiency was increased according to particle concentrations. The efficiency by nZVI and nZnO with 40 mg/L concentration was found at 97.22 % and 91.67 %, respectively. Meanwhile, that with 60 mg/L and more showed 100 % removal efficiency. This efficiency was substantially higher than that of ZVI and ZnO. Though, the concentration of 60 mg/L was selected as an optimum dosage for Cd removal from the effluent discharge of the silk dyeing process.



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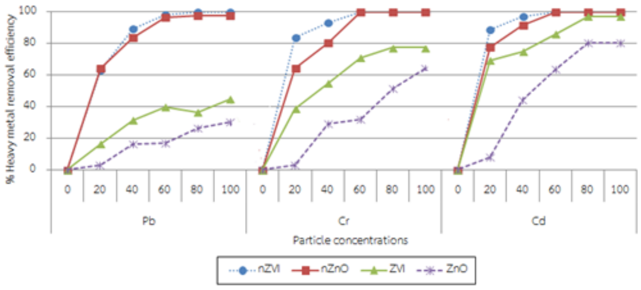


(b)

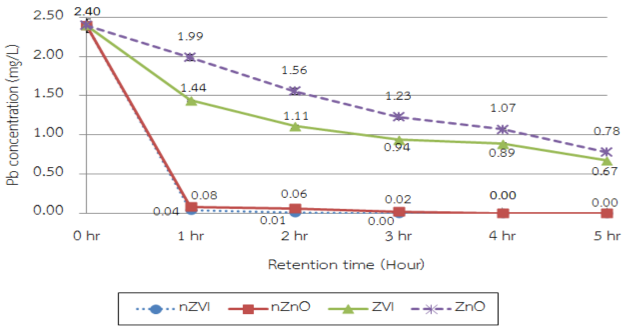
Fig. 4 Heavy metal (Pb, Cr, and Cd) treatments in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor (a) Concentrations of Heavy metal (Pb, Cr, and Cd) (b) The efficiency (%) of Heavy metal (Pb, Cr, and Cd) removal

The comparison in the removal efficiency of three heavy metals (Pb, Cr, and Cd) with varying concentrations from 0 to 100 mg/L showed in Fig. 4. It was found that the initial Pb concentration (controls) in the effluent discharge of the silk dyeing process was highest (2.4 mg/L). The initial Cr and Cd concentrations (controls) were relatively similar and much less than that of Pb. The overall results also indicated that the efficiency was increased according to particle concentrations. The results of three heavy metals in terms of removal efficiency demonstrated the similar pattern. According to the experiments with micro-scale particles (ZVI and ZnO), the highest efficiency was found with Cd followed by Cr and Pb, respectively. The concentration with 60 mg/L was selected as an optimum dosage for heavy metals removal and used for future experiments.

The removal efficiency and the kinetic removal rates of nZVI, nZnO, ZVI , and ZnO for heavy metal removal

The removal efficiency at incubation time with an hour interval

The optimum concentration of 60 mg/L from the previous experiment was used to study the removal efficiency and the kinetic removal rate. The samples were added with nZVI or nZnO, ZVI or ZnO and incubated for a period with an hour interval at 1, 2, 3, 4, and 5 hours.



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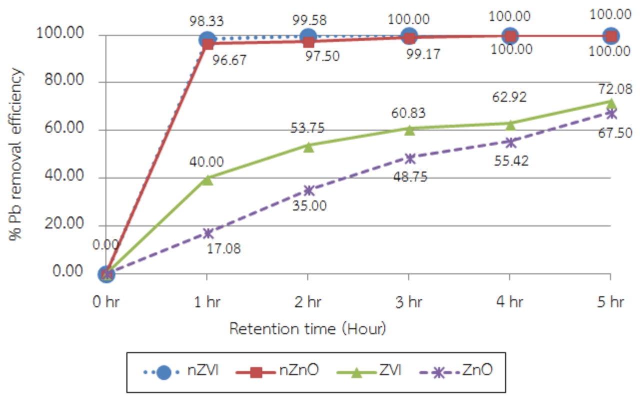
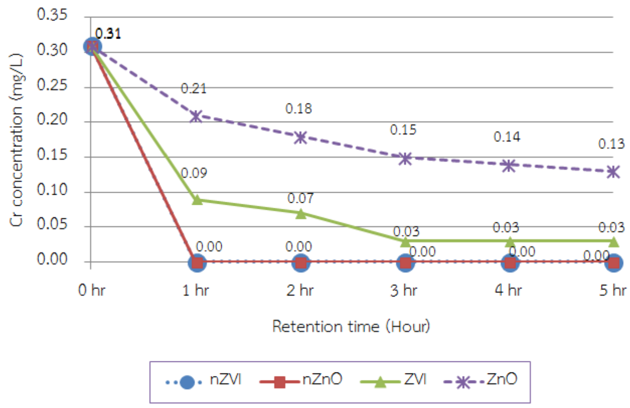


(b)

Fig. 5 Lead (Pb) treatments in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor at incubation time with an hour interval (a) Concentrations of Pb (b) The efficiency (%) of Pb removal

The results of the Pb removal efficiency with varying time showed in Fig. 5. About experiments with nanoparticles, it was found that the efficiency with hour incubation was 98.33 % and 96.67 % for nZVI and nZnO, respectively. It was up to 100 % at 3-hour incubation or more. In contrast, the experiments with micro-scale particles were found that the efficiency gradually increased from 1-hour to 5-hours. The maximum efficiency of ZVI and ZnO was found at 5-hour incubation with 72.08 and 67.50 mg/L, respectively.



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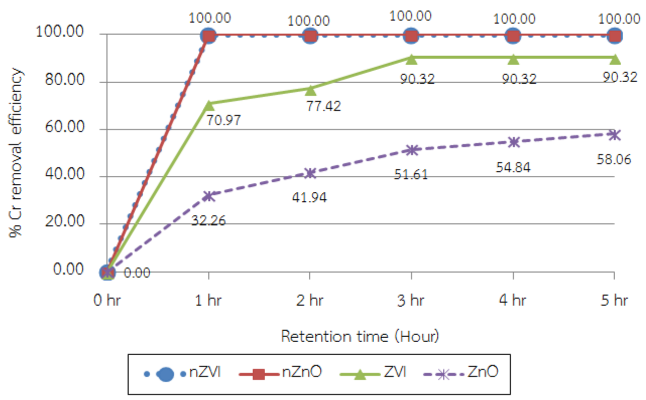
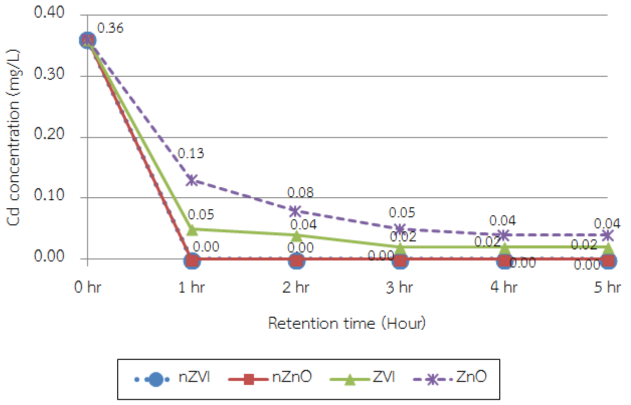


(b)

Fig. 6 Chromium (Cr) treatments in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor at incubation time with an hour interval (a) Concentrations of Cr (b) The efficiency

(%) of Cr removal

The results of the Cr removal efficiency with varying time showed in Fig. 6. Concerning experiments with nanoparticles, it was found that the efficiency was up to 100 % within an hour of incubation for both nZVI and nZnO. In contrast, the experiments with micro scale particles were found that the efficiency gradually increased from 1-hour to 5-hours. The maximum efficiency of ZVI and ZnO was found at 5-hour incubation with 90.32 and 58.06 mg/L, respectively.



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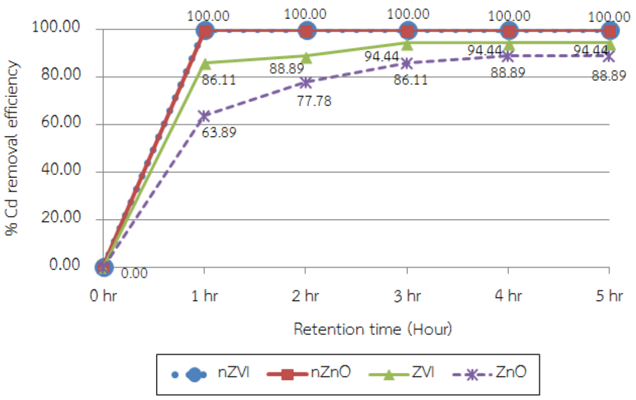
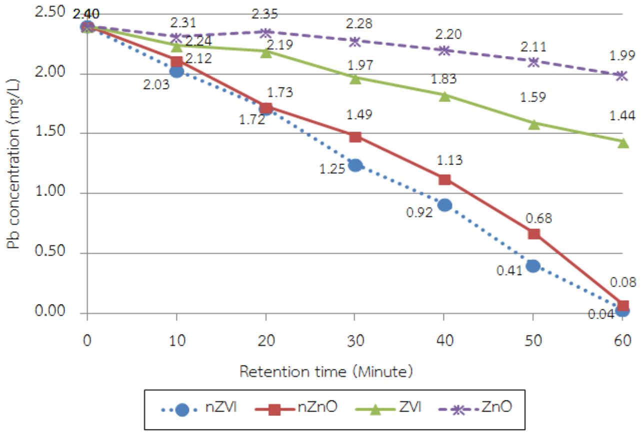


(b)

Fig. 7 Cadmium (Cd) treatments in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor at incubation time with an hour interval (a) Concentrations of Cd (b) The efficiency (%) of Cd removal

The results of the Cd removal efficiency with varying time showed in Fig. 7. Concerning experiments with nanoparticles, it was found that the efficiency was up to 100 % within an hour of incubation for both nZVI and nZnO. The experiments with micro-scale particles were found that the efficiency rapidly increased within the first hour. The maximum efficiency of ZVI and ZnO was found at 5-hour incubation with 94.44 and 88.89 mg/L, respectively.

The removal efficiency at incubation time with a 10-minute interval



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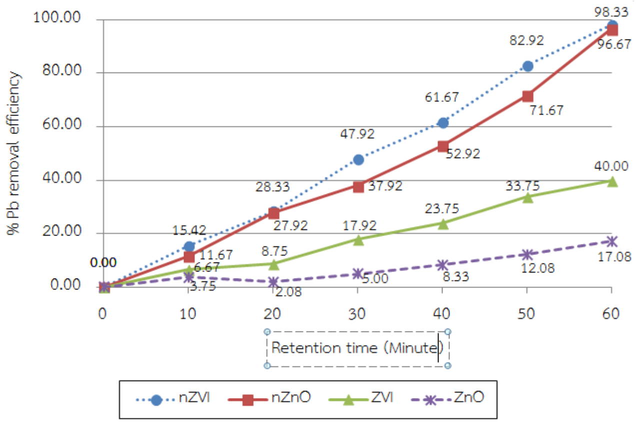

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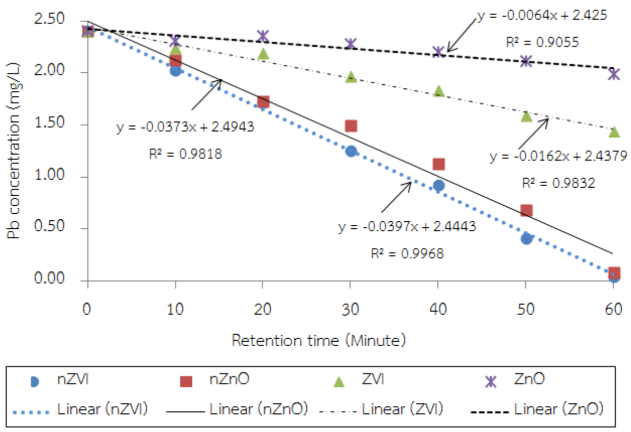
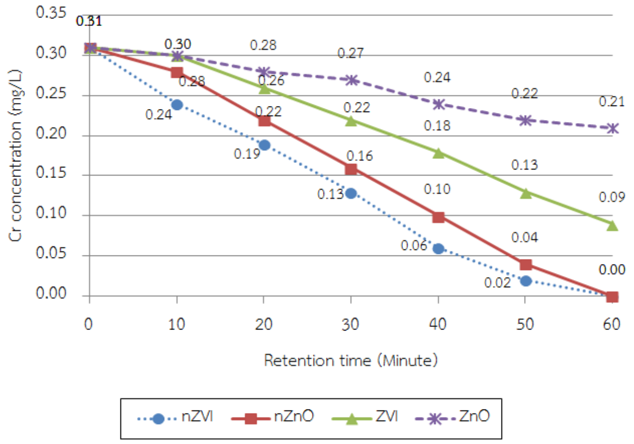


(c)

Fig. 8 Lead (Pb) treatments in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor at incubation time with the minute interval (a) Concentrations of Pb (b) The efficiency (%) of Pb removal (c) Linear equation of Pb removal

The results of the Pb removal efficiency with varying time from 0 to 60 minutes showed in Fig. 8. About experiments with nano particles, it was found that the removal efficiency gradually increased with the maximum of 98.33 % and 96.67 % at 60 minutes for nZVI and nZnO, respectively. The experiments with micro-scale particles were found that the removal efficiency gradually increased with the maximum of 40.00 % and 17.08 % at 60 minutes for ZVI and ZnO, respectively.



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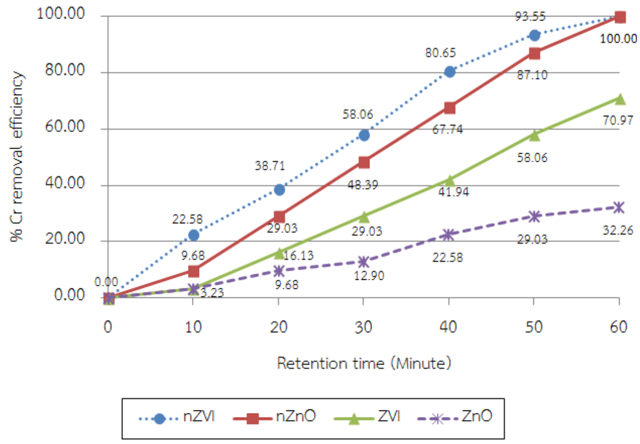

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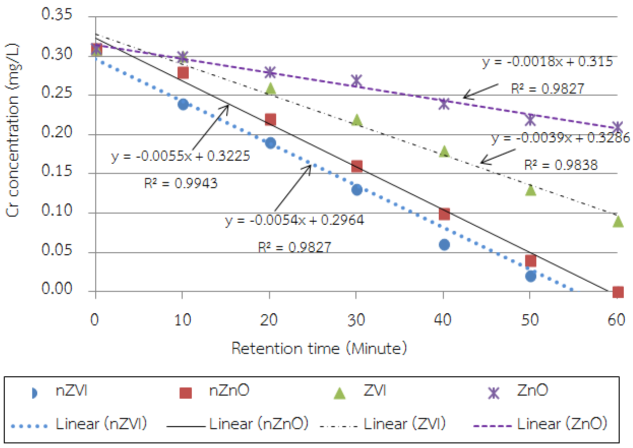
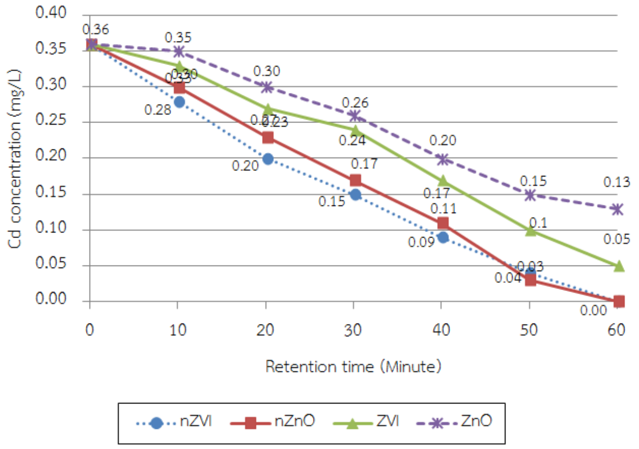


(c)

Fig. 9 Chromium (Cr) treatments in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor at incubation time with the minute interval (a) Concentrations of Cr (b) The efficiency (%) of Cr removal (c) Linear equation of Cr removal

Fig. 9 showed the results of the Cr removal efficiency with varying time from 0 to 60 minutes. Concerning experiments with nanoparticles, it was found that the removal efficiency gradually increased with the maximum of 100 % at 60 minutes for both nZVI and nZnO. The experiments with micro scale particles were found that the removal efficiency gradually increased with the maximum of 70.97 % and 32.26 % at 60 minutes for ZVI and ZnO, respectively.



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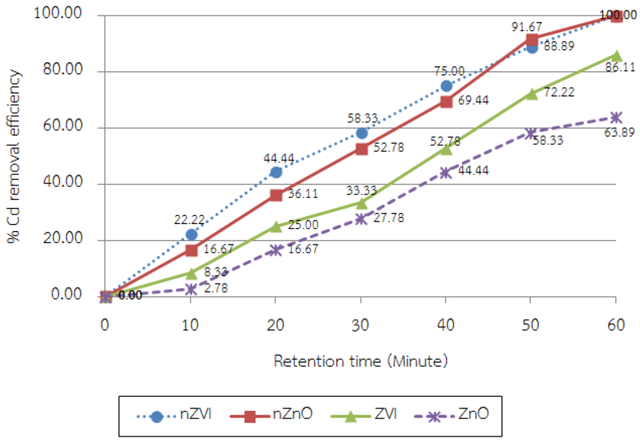

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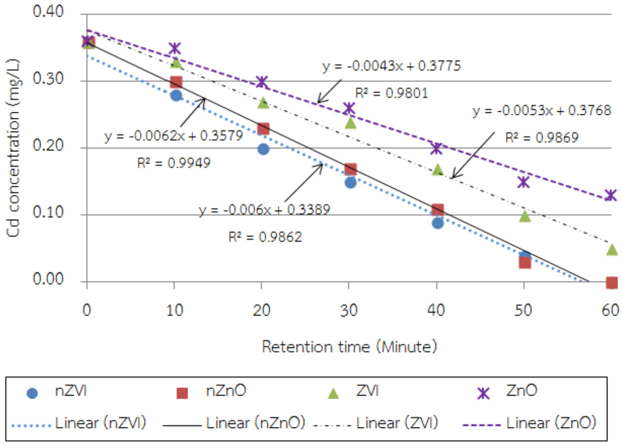

(c)

Fig. 10 Lead (Pb) treatments in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor at incubation time with the minute interval (a) Concentrations of Pb (b) The efficiency (%) of Pb removal (c) Linear equation of Pb removal

The results of the Cd removal efficiency with varying time from 0 to 60 minutes showed in Fig. 10. Concerning experiments with nano particles, it was found that the removal efficiency gradually increased with the maximum of 100 % at 60 minutes for both nZVI and nZnO. The experiments with micro-scale particles were found that the removal efficiency gradually increased with the maximum of 86.11 % and 63.89 % at 60 minutes for ZVI and ZnO, respectively.

The kinetic removal rates

Kinetic removal rates were plotted as semi-log to determine the first-order reaction rate of removal degradation refer to (1) and (2).

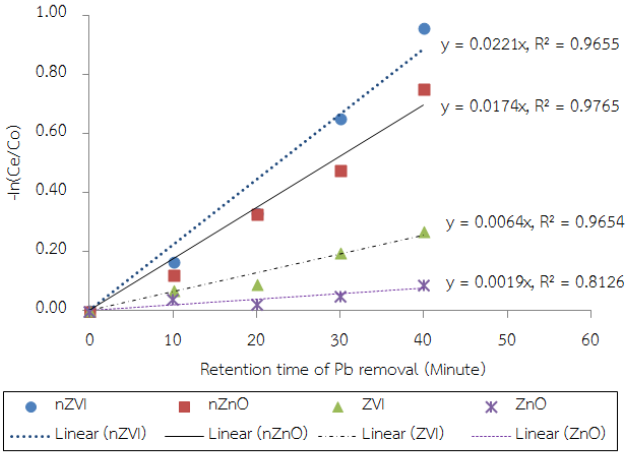

Fig. 11 Kinetic removal rates of Lead (Pb) in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor

The kinetic removal rates of Pb by four particles were showed in Fig. 11. The results demonstrated that the kinetic removal rate by nZVI was highest at 0.0221 min^{-1}. This was followed by nZnO, ZVI, and ZnO at 0.0174, 0.0064, and 0.0019, respectively.

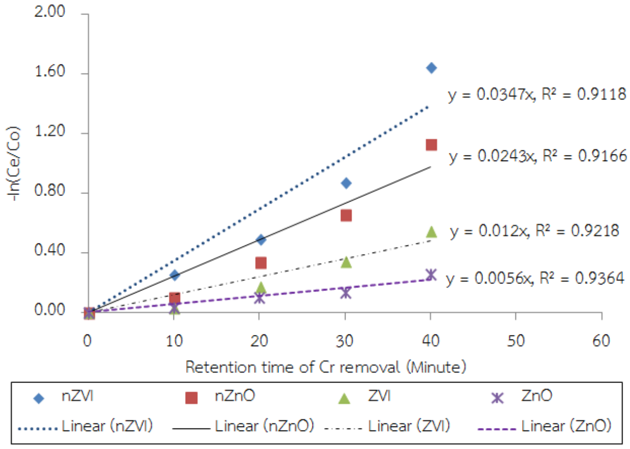

Fig. 12 Kinetic removal rates of Chromium (Cr) in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor

The kinetic removal rates of Cr by four particles were showed in fig. 12. The results demonstrated that the kinetic removal rate by nZVI was highest at 0.0347 min^{-1}. This was followed by nZnO, ZVI, and ZnO at 0.0243, 0.0120, and 0.0056, respectively.

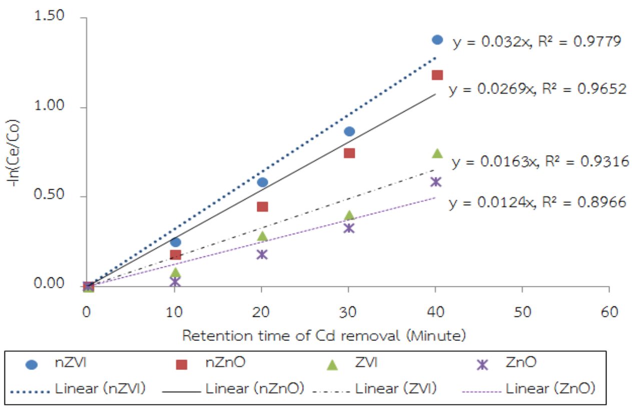

Fig. 13 Kinetic removal rates of Lead (Pb) in the effluent discharge from the silk dyeing process by nZVI, nZnO, ZVI, and ZnO in the wastewater reactor

The kinetic removal rates of Cd by four particles were showed in fig. 13. The results demonstrated that the kinetic removal rate by nZVI was highest at 0.0320 min^{-1}. This was followed by nZnO, ZVI, and ZnO at 0.269, 0.0163, and 0.0124, respectively.

4. Discussion

Generally, nZVI and nZnO demonstrated much higher efficiency than that of ZVI and ZnO. The results also indicated that the efficiency was increased according to particle concentrations. The results of three heavy metals in terms of removal efficiency demonstrated a similar pattern. According to the experiments with micro-scale particles (ZVI and ZnO), the highest efficiency was found with Cd followed by Cr and Pb, respectively. The concentration of 60 mg/L was selected as an optimum dosage for heavy metals removal. According to the results of the removal efficiency with varying time, it was found that the removal efficiency gradually increased. The results showed the optimal time at 60 minutes for all four heavy metals. The kinetic removal rate by nZVI was highest and followed by nZnO, ZVI, and ZnO for all four heavy metals.

5. Conclusion

The optimum dosage was found at 60 mg/L for all types of heavy metals and all types of particles. The highest efficiency and kinetic rates were found with nZVI followed by nZnO, ZVI, and ZnO, respectively.

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