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Study of Nanoparticle Distribution in Water Treated with Combined Filtration-Inductively Coupled Plasma System

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Abstract. Nanoparticles represent a specific type of organic or inorganic matter with at least one dimension less than 100 nm and possess unique physical and chemical properties. The presence of nanoparticles in drinking water may pose a direct human health threat or an indirect risk through ingestion. Thus, the removal of nanoparticles from drinking water is needed. This research was carried out to study the distribution of nanoparticles in water treated with a combined filtration-inductively coupled plasma system by continuous processing. The flow rate was set at 100 and 200 mL/minute. The results showed that after 180 minutes of treatment, the amount and volume of nanoparticles in the treated water produced were different from those contained in raw water. The nanoparticles were within the diameter ranges from 2.70 to 6.50 nm and 0.62 to 712 nm for flow rate at 100 and 150 mL/minute, respectively. The particles within these diameter ranges belong to the group of proteins, small molecules and atoms. The measured zeta potential of the treated water had also decreased, indicating that the stability of the nanoparticles was reduced and removal by the combined filtration-inductively coupled plasma system had occurred. Further research is required to find the optimum flow rate and determine the exact components of the groups of nanoparticles.

1. Introduction

The demand for safe drinking water increases day by day due to the rapid growth of the world population and the insufficient capabilities of conventional treatment methods. Various conventional and contemporary purification techniques are available, such as basic filtration and adsorption as well as advanced technologies, such as membrane filtration and advanced oxidation processes (AOPs). In general, the aim of drinking water treatment is to provide potable water that is free from toxic matters (organic, inorganic and biological) [1]. Among the advanced technologies, AOPs are recognized as highly effective in producing safe drinking water that is free of organics, inorganics and microbes [2].

AOPs are one of the advanced drinking water treatment technologies that involve the production of hydroxyl radicals (OH•) in sufficient quantities to induce water purification at standard temperature and pressure [3]. Hydroxyl radicals are extraordinarily reactive species with an oxidation potential of 2.80 V, which is higher than that of ozone of 2.07 V [4]. This causes the hydroxyl radicals to easily react with and decompose toxins in water by a series of chemical reactions, which ultimately leads to the conversion of organic compounds in solution into carbon dioxide and water along with mineral acids and salts [5]. Thus, AOPs are considered an environmental-friendly drinking water technology.

The aforesaid physical processes leading to hydroxyl radical formation in water occur when plasma is contacted with water. Plasma is generated by introducing electrical energy into a reaction zone, which



leads to various physical and chemical effects, such as the formation of oxidizing species, including radicals ($\text{H}\cdot$, $\text{O}\cdot$, and $\text{OH}\cdot$, etc.) and molecules (H_2O_2 , O_3 , etc.), shockwaves, ultraviolet light, and electrohydraulic cavitation [6]. The formed reactive species can drive decomposition and ultimately destruction of organic contaminants in solution, including microorganisms.

Numerous reports have been published about plasma application in water purification. Sun et al. [7] have reported the using of a pulsed-streamer corona discharge for organic compound removal with 83% of the total organic carbon removed without oxygen bubbling and 86% removed with oxygen bubbling. Desmiarti et al. [8] reported that the removal efficiencies of fecal coliforms and total coliforms by radio frequency plasma treatment were 95% and 93%, respectively. Recent research by Desmiarti et al. [9] showed that the inductively coupled plasma system (ICPS) was able to kill microorganisms in water with removal efficiencies of fecal coliforms and total coliforms at 100% and 74%, respectively. The effect of flow rate on removal of fecal coliforms, total coliforms, other coliforms [10] and Salmonella [11] by means of radio frequency ICPS was also investigated. However, there are very few reports regarding the removal of nanoparticles in drinking water treatment.

Nanoparticles are particles that have at least one dimension at the nanoscale (approximately 1 to 100 nm), a feature that alters their behavior. They are commercially applied in sunscreens, toothpastes, sanitary ware coatings and even food products [12]. Little has been reported regarding the use of engineered nanomaterials for water treatment. Chiavola et al. [13] have reported the application of iron-based nanoparticles as adsorbents for arsenic contamination removal from groundwater. In spite of the benefits of nanomaterials having been widely publicized, their potential health and safety risks pose a concern for drinking water utilities because these systems are designed to deal with microparticles and not with nanoparticles [14]. Consumption of drinking water contain nanoparticles such as normal organic matter and virus caused a direct or indirect risk human health including DNA damage [15].

This research aimed to study the distribution of nanoparticles in water by a combined filtration-inductively coupled plasma system (ICPS). The effect of flow rate was studied and the nanoparticle distribution was considered to study the number and volume distribution. The nanoparticle was measured by Zetasizer Nano. To expect the stability of the nanoparticles in the water, the zeta potential was also investigated.

2. Materials and Methods

2.1. Materials

A water sample was collected from the Kuranji River, a river in Padang City, Indonesia, and used as water source for a drinking water treatment plant. Prior to use, the sample was filtered to remove relatively large organic matters. The initial water properties were analyzed, of which the results are shown in Table 1.

Table 1. Kuranji river water properties

Parameters	Unit	Value
pH		10-10.8
Total dissolved solids (TDS)	mg l^{-1}	49-65
Dissolved organic carbon (DOC)	mg l^{-1}	28.75
Fecal coliforms	CFU/100 ml	3,800
Total coliforms	CFU/100 ml	16,400
Average particle size (diameter)	nm	1.12-2.70

2.2. Experiments

The experimental set-up of the combined filtration-ICPS was adapted from our previous study [9]. The designed system consisted of a plasma reactor and a plasma generator. The plasma reactor was made of 2 mm thick glass, with a length of 30 cm and a diameter of 1 cm, wrapped in 2 mm copper wire. The plasma generator was equipped with a high-voltage probe (Tektronix P6015A, Tektronix Inc., USA), a current probe (Tektronix P6022, Tektronix Inc., USA). To record the applied voltage and discharged currents, a Picoscope 4424 (Picotech, United Kingdom) with four channels was used. The frequency was set at 1.8-1.9 MHz and concurrently the flow rate was controlled at 100 and 150 ml/minute using a peristaltic pump.

2.3. Analysis

All water samples were filtrated using a 0.2 μm cellulose acetate membrane filter (ADVANTEC Corporation) before being measured to a zeta potential meter (Zetasizer Nano, manufactured by Malvern). The data were recorded as number distribution and volume distribution. The zeta potential and the electrophoretic mobility were also measured to expect the stability of the nanoparticles in the treated water.

3. Results and Discussion

3.1 Nanoparticles size distribution

The nanoparticle size distribution is displayed in Figures 1 and 2. From Figures 1 and 2 it can be seen that after 60 minutes of running process, particles in the ranges of 1.74 to 4.85 nm and 0.71 to 6.5 nm categorized the majority of nanoparticles occupying the volume for flow rates of 100 ml/minute and 150 ml/minute, respectively. After 180 minutes of running process, there was a shift in the size distribution. The average size of the nanoparticles found in the treated water produced at a flow rate of 100 ml/minute was increased, as larger sizes of nanoparticles occupying the treated water were identified. This shows that during the treatment removal of nanoparticles from the water sample occurred, resulting in only larger particles remaining in the treated water. After the treatment, the particle sizes ranged from 2.70 to 6.50 nm. The particles within this diameter range belong to the group of proteins, small molecules and atoms [16].

However, this phenomenon was not found in the treated water produced at a flow rate of 150 ml/minute. After 180 minutes of treatment, nanoparticles with smaller sizes still remained in the treated water, as the particle size ranged from 0.62 to 712 nm. This result indicates that the flow rate is a very important parameter. Further research to investigate the optimum flow rate in the combined filtration-ICPS to increase the removal efficiency with respect to nanoparticles can be conducted.

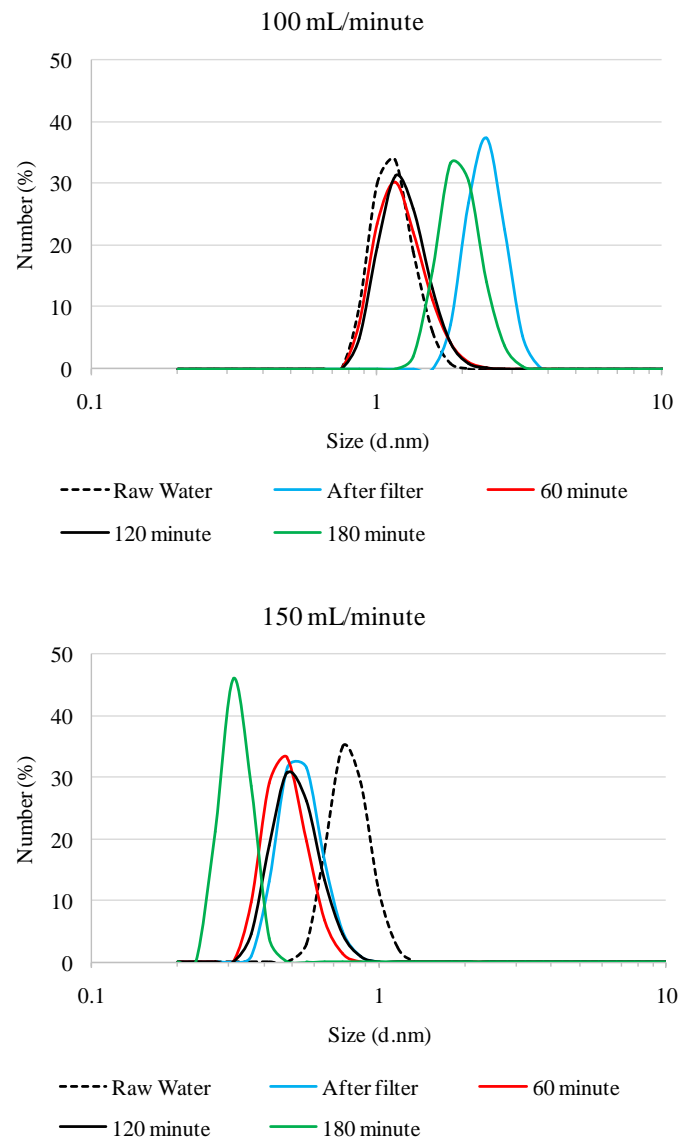


Figure 1. Number size distribution of nanoparticles.

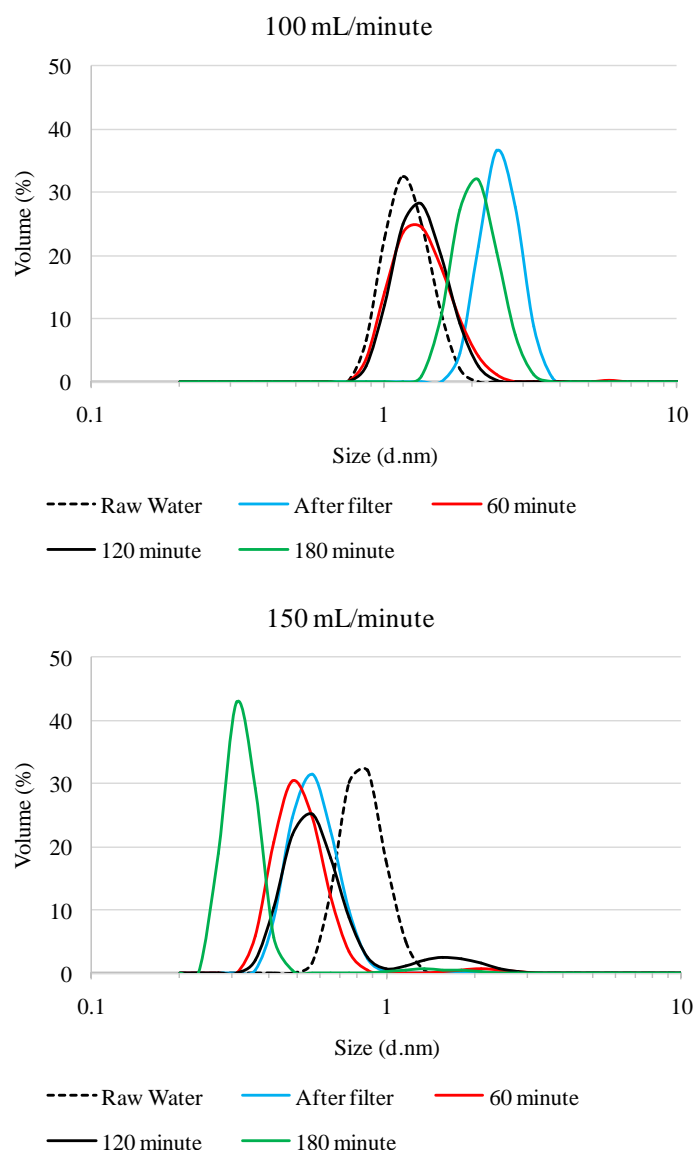


Figure 2. Volume size distribution of nanoparticles.

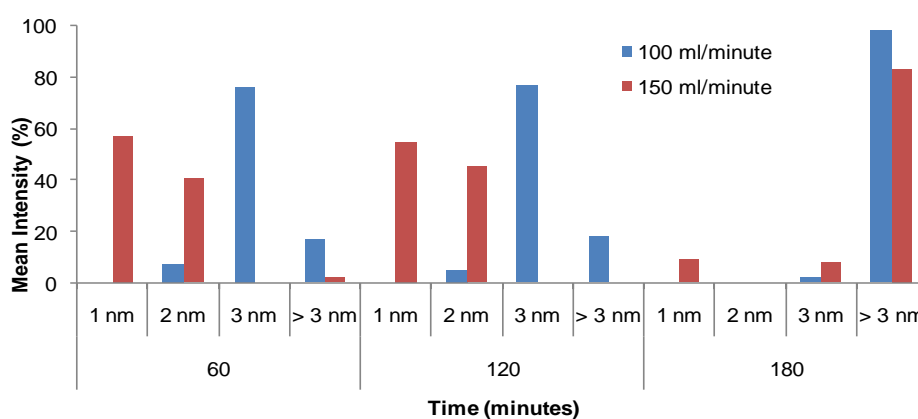
The average size distribution of nanoparticles occupying the treated water during the treatment process is shown in Figure 3. It can be seen that at flow rates of 100 ml/minute and 150 ml/minute a trend of gradual decrease of nanoparticles with smaller size was detected. These results suggest that the combined filtration-ICPS seems to be a feasible and efficient prospective method for nanoparticles removal in drinking water treatment plants.

3.2 Zeta potential and electrophoretic mobility

Table 2 summarizes the zeta potential and electrophoretic mobility values obtained for the raw water and the treated water produced with the combined filtration-inductively coupled plasma system. The table also consist of the conductivity of treated water values.

Table 2. Zeta potential and electrophoretic mobility values of raw and treated water produced by combined filtration-inductively coupled plasma system.

Sample	Flow rate (ml/minute)	Time (minutes)	Zeta potential (mV)	Electrophoretic mobility ($\text{m}^2\text{Vs}^{-1} \times 10^{-8}$)	Conductivity (mS cm^{-1})
Raw water	100		-19.3	-1.5110	0.114
		60	-11.4	-0.8938	0.111
		120	-10.6	-0.8274	0.108
		180	-8.18	-0.6411	0.109
Treated water	150	60	-12.4	-0.9743	0.143
		120	-10.3	-0.8055	0.141
		180	-18.4	-1.4390	0.144

**Figure 3.** Average size distribution of nanoparticles in the treated water.

In the results as shown in Figure 3, the values of measured mobility after 60 minutes of running experiment were increased for each flow rate. After 180 minutes of running process, the mobility values became less negative, i.e. the magnitude of mobility decreased. This shows an increase in the average size of the nanoparticles present in the treated water, resulting in obscuration of light transmission. This result supports the previous result, where the average particle size had increased after treatment. Furthermore, the measured zeta potential values were reduced after 180 minutes of running process for both flow rates. Solutions with low zeta potential tend to have poor stability, suggesting that the removal of nanoparticles by the combined filtration- inductively coupled plasma system had occurred.

4. Conclusion

Nanoparticles are particles with at least one dimension at the nanoscale (approximately 1 to 100 nm) and might pose a direct human health threat or an indirect risk when consumed. Thus far, conventional and advanced treatment processes have been designed for dealing with microparticles and not nanoparticles. The combined performance of the filtration-inductively coupled plasma system with respect to nanoparticles were investigated in this study. The effect of flow rate on nanoparticle size distribution were also studied. The results showed that the size distribution shifted after treatment. The average particle sizes ranged from 2.70 to 6.50 nm and 0.62 to 712 nm at a flow rate of 100 ml/minute and 150 ml/minutes, respectively. The measured mobility values became less negative and the zeta potential was decreased after 180 minutes of treatment, indicating that the removal of nanoparticles by

the combined filtration-inductively coupled plasma system had occurred. These results suggest that the combined filtration-inductively coupled plasma system seems to be a feasible and efficient prospective method for nanoparticle removal in drinking water treatment plants. Further research is required to find the optimum flow rate and determine the exact components of the groups of nanoparticles.

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