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Removal of Pathogenic Bacteria from Water by Radio-Frequency Thermal Plasma Treatment.

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ABSTRACT

Removal of phatogenic bacteria from water by radio-frequency thermal plasma treatment was investigated using three different sizes of reactors and three different applied frequencies in batch experiments. The sources of the water samples were river water, groundwater and mountain water. The results show that the removal efficiency (RE) of total coliforms was 86, 91 and 100% for river water, groundwater and mountain water, respectively, after a 20-minute treatment in a radio-frequency (RF) plasma reactor with a diameter of 2 inch and an applied frequency of 3.7 MHz. Compared to total coliforms, the RE of fecal coliforms was 91, 100 and 100% for river water, ground water and mountain water, respectively. The RE of total coliforms and fecal coliforms increased with an increase of the frequency or with a decrease of the diameter of the reactor. Increasing the frequency or decreasing the diameter of the reactor caused a decline in the death yield of phatogenic bacteria (CFU/kW h). Based on these results, the removal of phatogenic bacteria from water by RF thermal plasma treatment could be a viable alternative process for decontaminating drinking water in the future.

Keywords: Radio-frequency thermal plasma, Microorganisms, Half-life, Death yield, Water treatment.

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INTRODUCTION

The removal of organic pollutants and phatogenic bacteria from river water, groundwater and mountain water is one of the urgent topics in drinking water treatment. Actually, they cannot be removed effectively by conventional methods such as physical, biological and chemical methods [1-6]. Hydroxyl radicals are generated by Advanced oxidation processes (AOP) that have high oxidation potential to complete oxidize organic substances to carbon dioxide and water [7]. Many researchers have studied the generation of hydroxyl radicals for removal of organic carbon. The hydroxyl radical reaction activated by ultraviolet photolysis to be slow[8]. The applicability of corona discharge technology for water and wastewater treatment using a non-thermal plasma system has been investigated by Lukes [9]. This research improved by Lukes [7]and explained that ozone oxidation and found it needs time to form hydroxyl radicals in a neutral solution. Non-thermal plasma systems can have a high removal efficiency for removing organic compounds [3, 10, 11]. The disadvantage of non-thermal plasma systems is corrosion on the electrode surface due to direct contact with water in the reactor.

The thermal plasma is consist of electrons, free radicals, ions, and neutral as partially or fully ionized gas[12]. These gases could be produced with enough energy to consent plasma substances to reach thermal equilibrium. Radio frequency (RF) induction is one of the methods to create thermal plasma. It can produce active compounds (•H, •OH, H_2O_2, O_3, etc.) that have a high oxidation potential, so it can potentially be used for removing pathogenic bacteria or microorganisms from water (fecal coliforms and total coliforms). Because these active species have a high oxidation potential and are capable to kill microorganisms and to degrade organic compounds in water [13]. Another advantage of this system is that the plasma can produce ultraviolet light and shockwaves that can kill microorganisms in the water as well[14].

Some researchers have done investigations into the reduction of microorganisms in the water content using non-thermal plasma systems [15, 16]. The removal efficiency of microorganisms at 100% using a corona discharge to generate plasma for the treatment of drinking water has been examined by Aragi[15]. Another study, the removal of microorganisms in drinking water by using a dielectric barrier discharge plasma system has been studied byHazmi et al. [16]. Their results shown that the removal efficiency of fecal coliforms was in the range 25-100% and total coliforms was in the range of 44-100%, after the water was supplied to a pulsed high voltage of 5-10 kV for 60 minutes. To our knowledge, no studies using a thermal plasma system to remove organic compounds and microorganisms have been done, although such a system can achieve a high quality product and a high removal efficiency. Another advantage of this system is that it does not cause corrosion, because there is no direct contact with the water.

The aim of this research was to study the ability of a radio-frequency thermal plasma system to remove pathogenic bacteria from river water, groundwater and mountain water respectively. Experiments were carried out in a batch system to see the effects of frequency and diameter of reactor on removal efficiency, half life and death yield of pathogenic bacteria.

MATERIAL AND METHODS

Source of Water

Water samples were collected from the Kuranji River, ground water, and mountain water in Padang City, Indonesia, representing high, medium and low pathogenic bacteria content respectively. The characteristics of the water samples are displayed in Table 1.

Experimental set-up

A diagram of the experimental set-up used in this study is shown in Fig. 1. The system included an MI007 passive oscilloscope probe, a P6021A Tektronix current probe, a PicoScope 4424 with four-channel input, a radio-frequency generator and a plasma reactor. Three kinds of glass plasma reactors were used, with a length of 30 cm and diameters of 1, 2, and 3 inch respectively. A high-frequency pulse power supply was used to generate a discharge in the reactor. A copper wire ($\varnothing$ 2 mm) was wrapped around the reactor.

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Experimental procedures

Two series of batch experiments were carried out to investigate the performance of the RF plasma reactors. The first series was carried out in order to investigate the effect of frequency on removal efficiency of pathogenic bacteria for three kinds of water samples at 3, 3.3, and 3.7 MHz respectively, using a reactor with a diameter of 2 inch. The second series studied the effect of diameter on removal efficiency of pathogenic bacteria, using three reactors with diameters of 1, 2, and 3 inch respectively and an applied frequency of 3.7 MHz. Temperature and pH of the solution were quite stable during the experiment, with values of 6.7±0.03 and 29.2±0.14 respectively. The experimental conditions used to investigate the effect of frequency and diameter on removal efficiency of pathogenic bacteria, half-life and death yield are shown in Table 2.

Figure 1: The schematic diagram of RF plasma system

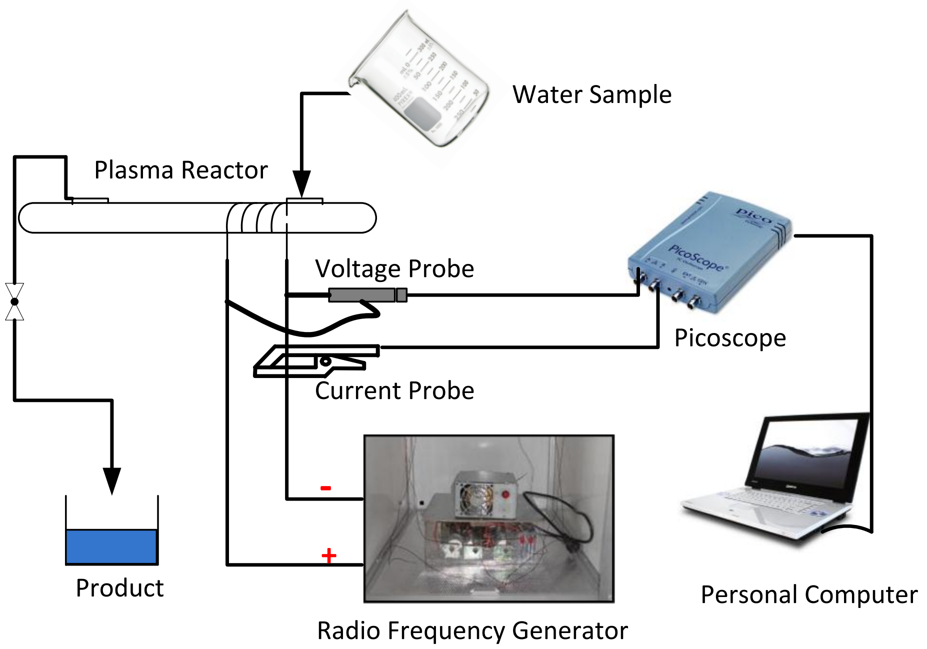

Table 1: Characteristics of water samples

Component	Unit	Source of Water		
		KuranjiRiver	Ground water	Mountain Water
Turbidity	NTU	50.2	28.2	0,207
Phenol	mg/L	3.5	1.84	< 0,1
Electric Conductivity (EC)	µS/cm	1472	4010	684
Total Dissolved Solid (TDS)	ppm	748	2030	398
Temperature	°C	30.9	30.9	29,1
Manganese (Mn)	mg/L	< 0.2	< 0.2	< 0,2
Nitrite (NO_2^-)	mg/L	< 0.01	< 0.01	< 0,01
Nitrate (NO_3^-)	mg/L	< 0.08	< 0.08	< 0,08
Iron (Fe)	mg/L	0.05	0.14	< 0,02
Fecal Coliforms	CFU/mL	7400	600	200
Total Coliforms	CFU/mL	39200	4600	1800

Table 2: The experimental conditions

Series	Parameters and values		
	Source of water	Frequency (MHz)	Diameter of Reactor (inch)
Series 1	Kuranji River	3 3.3 3.7	2
	Ground water	3 3.3 3.7	2
	Mountain water	3 3.3 3.7	2
Series 2	Kuranji River	3.7	1 2 3
	Ground water	3.7	1 2 3
	Mountain water	3.7	1 2 3

Analysis Methods

Fecal coliform and total coliforms were analyzed by colony forming unit (CFU) assay using Brilliance E.coli/coliform Selective Agar (Oxoid, CM1046B). The conductivity, pH, temperature and total dissolved solid were analyzed using a Milwaukee MI 180multiparameter bench meter. Nitrate, nitrite, manganese and iron were analyzed using a LovibondMaxidirect 600 photometer. The turbidity was checked with a Hach 2100 turbidity meter.

Removal Efficiency (RE) and Death Rate Calculation

The RE of total coliforms and fecal coliforms was calculated according to the following equation:

$$RE\ (\%) = \frac{N_o - N}{N_o} x100\% \tag{1}$$

The kinetic death rate of the pathogenic bacteria was calculated using the first order equation, as follows:

$$\frac{dN}{dt} = -kN \tag{2}$$

where N is the number of pathogenic bacteria (CFU/mL), t is time (h), and *k* is death rate constant (h^{-1}). The value of *k* was calculated by plotting the value of ln (N/N_0) versus t using the following equation:

$$ln\left(\frac{N}{N_0}\right) = kt \tag{3}$$

with N_0 as the initial number of microorganisms. On the basis of the data, the value of half-life death time ($t_{1/2}$) was calculated for all runs.

Calculation of death yield

The efficiency of pollutant degradation or pathogenic bacteria removal is illustrated by the degradation yield, defined as the amount of pollutant or number of microorganisms decomposed or deaths per unit of energy consumed in the process [10, 11, 17].The death yield of microorganisms was calculated using the following equation:

$$Y = \frac{N_0 \times V \times RE}{100 \times P \times t} \qquad (4)$$

where Y is death yield (CFU/kWh), V is volume of water in the reactor (L), P is average power (kW), and t is radiation time.

RESULTS AND DISCUSSION

Effect of frequency on removal efficiency (RE) of pathogenic bacteria

The effect of frequency on the RE of total coliforms and fecal coliforms in different water sources is displayed in Figure 2 (a-f). The results show that the RE of total coliforms and fecal coliforms increased with an increase of the frequency in all runs. For example for river water, the RE of total coliforms slightly increased to 75%, 76% and 82% after 5 minutes of treatment at frequencies of 3, 3.3 and 3.7 MHz, respectively. Compared with total coliforms, the RE of fecal coliforms also slowly increased to 81%, 86 and 88% after 5 minutes of treatment. The final RE of total coliforms and fecal coliforms achieved 86-91% and 96-98% after 60 minutes of treatment at frequencies of 3-3.7 MHz, respectively. This can be attributed to the intensity of the discharge electric field, shock waves and ultraviolet light radiation increasing with the increase of the frequency, which increases the amount of reactive species in the gas phase and aqueous solution [11]. This condition is consistent with the theory that a higher frequency will increase the efficiency of microorganism removal (fecal coliforms and total coliforms). Increasing the frequency will also increase the production of active species, such as •OH, •H, O_3 and H_2O_2. These active species will kill microorganisms by means of electroporation, which perforates the cell membrane due to the influence of an electric charge by ions and electrons, resulting in deactivation of the microorganism. In addition, the active species generated will degrade the DNA of the microorganisms so that fecal coliforms and total coliforms will become inactive [14]. Similar results were found for the samples of ground water and mountain water. Figure 2 (a-f) also shows that the RE of total coliforms was generally lower than that of fecal coliform. Compared with the research using a non-thermal system that has been conducted by Hazmi et al. [16], the RE of pathogenic bacteria using thermal plasma was 50% higher and 75% faster.

For a better understanding of the behavior of the microorganisms, the half-lifes of total coliforms and fecal coliforms were calculated based on the experimental data, as shown in Table 3. Generally, Table 3 shows that the half-life increased with an increase of frequency. The half-life of fecal coliforms increased faster than that of total coliforms. This means that the removal of total coliforms was the key for the removal of the microorganisms from the water.

Effect of diameter of RF reactor on removal efficiency (RE) of microorganism

The effect of the RF plasma reactor diameter on the RE of total coliforms and fecal coliforms in different water sources is displayed in Figure 3 (a-f).

Figure 3 displayed that the RE of total coliforms and fecal coliforms decreased with increasing of the diameter of the reactor in all runs. For example for groundwater, the RE of total coliforms decreased to 91%, 87% and 48% after 5 minutes of treatment for reactor diameters of 1, 2, and 3 inch, respectively. Compared with total coliforms, the RE of fecal coliforms also decreased to 100-66% after 5 minutes of treatment. This can be attributed to the intensity of the discharge electric field, shock waves and ultraviolet light radiation decreasing with an increase of the reactor diameter, which increases the distance of the radiation that produces reactive species in the gas phase and aqueous solution. Similar results were found for the water samples of river water and mountain water. Figure 3 (a-f) also shows that the RE of total coliforms was generally lower than that of fecal coliforms. The half-life of fecal coliforms also increased faster than that of total coliforms (Table 4). This means that the removal of total coliforms was the key for the removal of the microorganisms from the water. Similar results was found by Hazmi et al [16].

Figure 2: Total Coliforms and Fecal Coliforms removal percentage versus treatment time at different frequency with diameter of RF reactor at 2 inch

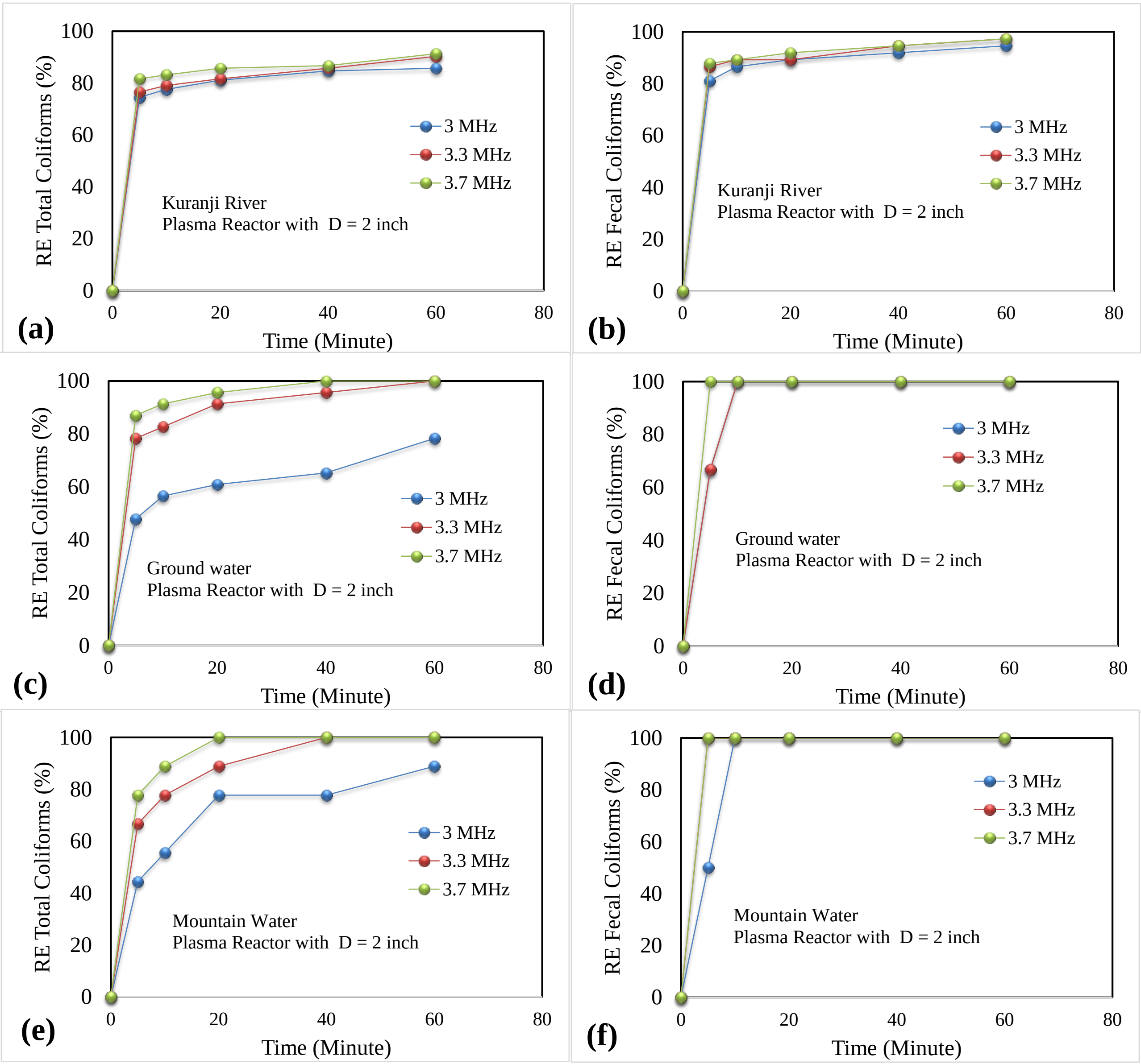

Table 3: The apparent half life ($t_{1/2}$) of microorganisms with Diameter of Reactor at 2 inch

Source of Water	Frequency (MHz)	$t_{1/2}$ (minute)	
		Total Coliforms	Fecal Coliforms
Kuranji River	3	3.36	3.08
	3.3	3.27	2.89
	3.7	3.06	2.85
Groundwater	3	6.25	5.00
	3.3	3.19	3.75
	3.7	2.88	2.50
Mountain Water	3	5.63	5.00
	3.3	3.75	2.50
	3.7	3.21	2.50

Figure 3: Total Coliforms and Fecal Coliforms removal percentage versus treatment time at different diameter of RF reactor with frequency at 3.7 MHz

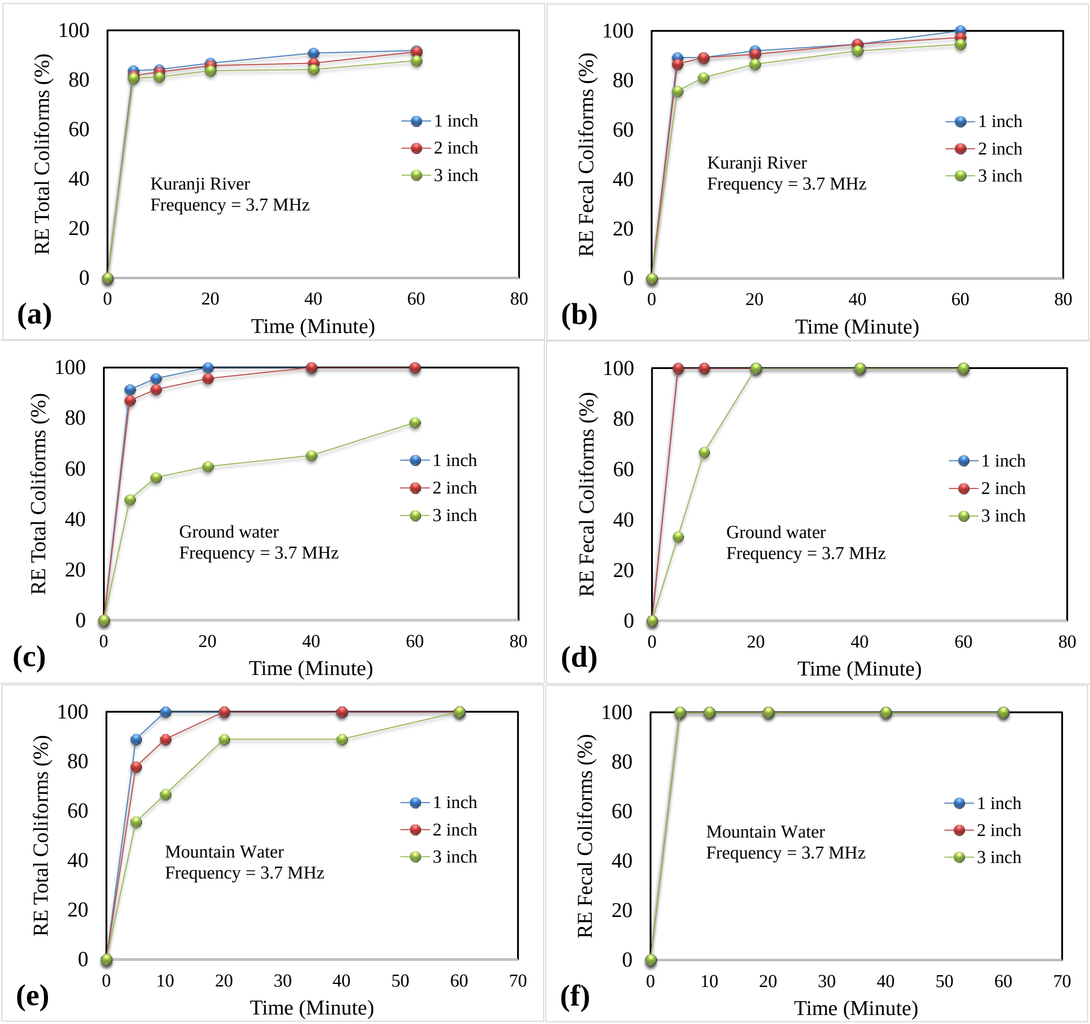

Table4: The apparent half life ($t_{1/2}$) of microorganisms with frequency 3.7 MHz

Source of Water	Diameter of RF Reactor (inch)	$t_{1/2}$ (minute)	
		Total Coliforms	Fecal Coliforms
Kuranji River	1	2.99	2.80
	2	3.06	2.89
	3	3.10	3.30
Groundwater	1	2.74	2.50
	2	2.88	2.50
	3	5.23	7.50
Mountain Water	1	2.81	2.50
	2	3.21	2.50
	3	4.50	2.50

Death yield of microorganisms

The RE of microorganisms is better illustrated by the death yield, defined as the amount of microorganisms killed per unit of energy consumed by the RF plasma reactor, as shown in Eq. (4). The effect of frequency on death yield at a reactor diameter of 2 inch and a treatment time of 20 minutes is displayed in Table 5. The death yield for total coliforms and fecal coliforms decreased with the increase of frequency. The advantage of using plasma is the direct generation of OH radicals in the electrical discharge, in addition to those formed by ozone. Photocatalytic degradation requires radiation sources whose energy consumption is usually high. Similar results can be found in Table 6.

Table 5: Effect of frequency on death yield at Diameter of RF Reactor 2 inch and with treatment time at 20 minutes

Source of Water	Frequensi (MHz)	Total Coliforms		Fecal Coliforms	
		η (%)	Y (10^{10} CFU/kW h)	η (%)	Y (10^{10} CFU/kW h)
Kuranji River	3	81	8103.68	89	1681.90
	3.3	82	4333.51	91	907.33
	3.7	86	2317.50	92	469.02
Groundwater	3	57	662.56	100	152.90
	3.3	91	568.77	100	81.25
	3.7	96	303.48	100	41.38
Mountain water	3	78	356.77	100	50.97
	3.3	89	216.68	100	27.08
	3.7	100	124.15	100	13.79

Table6 Effect of Diameter of RF Reactor on death yield at frequency 3.7 MHzwith treatment time at 20 minutes

Source of Water	Diameter of RF reactor (inch)	Total Coliforms		Fecal Coliforms	
Kuranji River	1	η (%)	Y (10^{10} CFU/kW h)	η (%)	Y (10^{10} CFU/kW h)
	2	86	113.50	91	22.63
	3	84	115.75	86	22.58
Groundwater	1	100	36.69	100	4.79
	2	96	14.86	100	2.03
	3	61	9.88	100	2.12
Mountain water	1	100	14.36	100	1.60
	2	100	6.08	100	0.68
	3	89	5.65	100	0.71

CONCLUSION

The most important conclusions that can be drawn from the present study can be summarized as follows:

- The RE of total coliforms was 86, 91 and 100% after 20 minutes of plasma treatment in an RF reactor with a diameter of 2 inch and an applied frequency of 3.7 MHz for river water, groundwater and mountain water, respectively.
- The RE of fecal coliforms was 91, 100 and 100% after 20 minutes of plasma treatment in an RF reactor with a diameter of 2 inch and an applied frequency of 3.7 MHz for river water, groundwater and mountain water, respectively.

- The RE of total coliforms and fecal coliforms increased with an increase of frequency or with a decrease of diameter of the RF plasma reactor.
- Increasing the frequency or decreasing the diameter of the plasma reactor caused a decline in the death yield of microorganisms (CFU/kW h).

These results suggest that RF thermal plasma treatment may be of interest for the treatment of drinking water in order to reduce microorganisms content.

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