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Removal of $CaCO_3$ scales on a filter membrane using plasma discharge in water

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ABSTRACT

In modern wastewater treatment, filters are routinely used for removing unwanted particles from water. The present study investigated if a pulsed spark discharge in water can be used to remove deposits from the filter membrane for its potential application in drinking and industrial water treatment. The test setup included a circulating water loop and a pulsed power system. The present experiments used artificially hardened water with hardness of 1000 mg/L of $CaCO_3$ made from a mixture of calcium chloride ($CaCl_2$) and sodium carbonate (Na_2CO_3) in order to produce calcium carbonate deposits on the filter membrane. Spark discharge in water was found to produce strong shockwaves in water, and the efficiency of the spark discharge in cleaning filter surface was evaluated by measuring the pressure drop across the filter over time. Results showed that the pressure drop could be reduced to the value corresponding to the initial clean state and after that the filter could be maintained at the initial state almost indefinitely, confirming the validity of the present concept of pulsed spark discharge in water to clean dirty filter.

1. Introduction

Hard water causes a number of problems in both industry and home, such as mineral fouling in water chillers, heat exchangers, boilers, water heaters, shower heads, and dish washers. According to the Water Quality Association of the United States, hard water is defined as water with more than 80–120 mg/L of calcium. In recent years the rising demand for high-quality water has called for the development of more economic methods for treating hard water around the world. Ion-exchangers have been one of the most commonly used means for water treatment. However serious environmental problems can be caused by the discharge of water contaminated by sodium ions. Water treatment methods without the use of chemicals are generally called physical water treatment (PWT) [1,2]. Examples of the PWT include permanent magnets, solenoid coil devices, electrostatic precipitators, sudden pressure drops, catalytic alloys, electrolysis, etc. Some of PWT devices are believed to produce induced electric field in hard water, which produces submicron-size colloidal particles in water [3,4]. Hence, if a PWT device can keep producing suspended particles in water and at the same time a filter can continuously remove suspended particles from water, one can mechanically reduce the hardness without the use of chemicals almost indefinitely. Such a system, if successfully developed, can be considered as a mechanical water softener.

In a cooling-tower application, the cycle of concentration in cooling water is often maintained at 3.5. Hence, if the hardness of makeup water is 100 mg/L, the calcium hardness is approximately 350 mg/L in the circulating cooling water. In order to avoid the performance degradation in heat transfer equipment (i.e., condenser), a part of cooling water is periodically or continuously discharged via blowdown. Thus, if the cycle of concentration can be increased through continuous precipitation and removal of calcium ions using a self-cleaning filter, one can significantly reduce the amount of water that has to be discharged to sewer, resulting in conservation of fresh water. For example, consider a modern 1000-MW fossil-fueled power plant with 40% efficiency which rejects 1500 MW of heat at full load. Such a power plant uses about $1.3 \times 10^5\ m^3/h$ of circulating water based on 10 °C temperature difference in a condenser [5]. As heat is removed via evaporation of pure water at a cooling tower, the need for the makeup water is about $2400\ m^3/h$ for the typical fossil plant, resulting in $57{,}600\ m^3$ a day [5]. With the present concept using both PWT and filtration, one hopes to be able to operate cooling tower at a higher cycle of concentration of 8–10, thus reducing the freshwater consumption by approximately 25%. This means that the makeup water can be reduced by $14{,}400\ m^3$ a day in a 1000-MW fossil-fueled power plant.

Various microfiltration methods are used to remove suspended particles from water. Whenever a filter is used in a water system, the pressure drop across the filter gradually increases with time and/or the flow rate gradually decreases with time. This reduced

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Nomenclature

C	electric capacity
C_0	speed of sound
E	electric field
H	specific enthalpy at the bubble wall
I	current
r	distance from spark source to pressure transducer
r_c	radius of curvature of electrode tip
R	radius of gas bubble in water
P	pressure
t	time
V	voltage

Greek symbols

Δt	electric pulse duration time
ε_0	vacuum permittivity
ε_r	relative permittivity
ρ	density
σ	electric conductivity

Subscripts

0	ambient condition
b	condition on the capacitor bank
c	condition on the electrode

performance of a filter is due to the accumulation of impurities on the filter surface, and the clogged area becomes sites for bacteria growth for further reducing the opening in the filter surface, increasing the pumping cost. Therefore, in order to continuously remove suspended particles from water, the filter must be replaced frequently, a process which is prohibitively expensive in most industrial water applications. To overcome the drawbacks of frequent filter replacement, self-cleaning filters are commonly used in industry. Although there are a number of self-cleaning filter technologies available on the market, most self-cleaning filters use a complicated backwash method, which reverses the direction of flow during the cleaning phase. Furthermore, the water used in the backwash must be clean filtered water, which reduces the filter capacity. Aforementioned drawbacks of the conventional filter technologies motivated us to develop a new self-cleaning filter using spark-generated shock waves.

1.1. Plasma discharge in water

Water is a polar liquid with a relative permittivity of $\varepsilon_r = 80$. The electrical conductivity of water ranges from about 1 μS/cm to several thousand μS/cm, depending on the dissolved ion concentrations. In order to generate spark in water, one needs to use a short pulse of high voltage. Given that a specific water is exposed to an electric pulse with a duration time of Δt, when $\Delta t >> \varepsilon_r\, \varepsilon_0/\sigma$, where ε_0 is vacuum permittivity and σ is the conductivity of water, the aqueous solution behaves as a resistive medium [6]. One of the major results of such a long electric pulse is the electrolysis of water with hydrogen and oxygen production. For much shorter times, i.e., when $\Delta t << \varepsilon_r\, \varepsilon_0/\sigma$, water behaves as a dielectric medium [6], and a high applied voltage will lead to the breakdown of the solution. It was found in experiments that a threshold electric field of the order of 1 MV/cm is necessary to initiate the discharge [7]. If the discharge does not reach the second electrode it is called pulsed corona discharge using analogy with discharges in gases, and branches of such a discharge are called streamers. If a streamer reaches the opposite electrode, it makes a conductive channel between electrodes and consequently a spark is forming. If the current through the spark is very high (above 1 kA), this spark is usually called a pulsed arc. Various electrode geometries have been used for the generation of the plasma discharge in water for the purpose of water treatment.

Two of the simplest geometries are a point-to-plane geometry and a point-to-point geometry [7], as electric discharges in water usually start from sharp electrodes. For a point electrode, the electric field can be estimated as $E \sim U/r_c$, where U is the applied voltage and r_c is the radius of curvature of the needle tip. It is obvious that a relatively small applied voltage is needed for a sharp electrode. The point-to-plane geometry is often used for pulsed corona discharges, whereas the point-to-point geometry is often used for pulsed arc systems [7]. Sunka et al. [8] pointed out that the anode with a sharp tip would be quickly eroded by the discharge and one had to find some compromise between the optimum sharp anode construction and its lifetime for extended operations.

Another concern in the use of plasma discharges in water is the limitation posed by the electrical conductivity of water on the production of such discharges [8,9]. As the electric conductivity of water increases significantly greater than 400 μS/cm as in the cooling water, it becomes more difficult to form a spark discharge as a large portion of energy can be dissipated to a high-conductivity water through electrolysis.

Locke et al. [7] have recently published a comprehensive review on the application of strong electric fields for the treatment of water or organic liquids with 410 references. They explained in detail the types of discharges used for water treatment, physics of the discharge, and chemical reactions involved in the discharge in water. When a high-voltage high-current discharge takes place between two submerged electrodes, a large part of the energy is consumed on the formation of a thermal plasma channel. This channel emits UV radiation and its expansion against the surrounding water generates an intense shock wave [10,11]. The water surrounding the electrodes becomes rapidly heated, producing bubbles, which help the formation of a plasma channel between the two electrodes. The plasma channel may reach a very high temperature of 14,000–50,000 K. The plasma channel consists of a highly ionized, high-pressure and high-temperature gas. Thus, once formed, the plasma channel tends to expand. The energy stored in the plasma channel is dissipated via radiation and conduction to surrounding cool liquid water as well as mechanical work. At the phase boundary, the high-pressure build-up in the plasma is transmitted into the water interface and an intense compression wave (i.e., shock wave) is formed, traveling at a much greater speed than the speed of sound. The energy transferred to the acoustic energy can be calculated as [12]:

$$E_{\text{acoustic}} = \frac{4\pi r^2}{\rho_0 C_0} \int (P(r,t) - P_0)\mathrm{d}t \tag{1}$$

where r is the distance from the spark source to the pressure transducer, ρ_0 is the density of water, C_0 is the speed of sound in water, P_0 is the ambient pressure. One can conclude that the pressure created by the spark discharge is much higher than ambient pressure at positions close to the source. Traditionally, the high-pressure shockwave is studied for high-voltage insulation and rock fragmentation [13], while recently it has found more applications in other areas including extracorporeal lithotripsy [14] and metal recovery from slag waste [15].

In order to validate the present concept to use spark discharge for filter cleaning, an experimental setup was built where discharges could be produced in water and pressure drop across a filter surface was measured over time at various spark frequencies and flow conditions. It is hypothesized that the energy deposited by the spark shock wave onto water-filter interface is enough to remove the contaminants having Van der Waals bonds with filter surface. The objective of the present study was to examine the feasibility of a self-cleaning water filtration concept using spark discharges in water.

2. Methods

An experimental system was designed to test the effectiveness of the self-cleaning filter concept using spark discharges in water under various flow conditions. The system consisted of two parts: a flow loop with a filter to simulate a cooling-tower water system and a pulsed power system to produce spark discharges in water. A schematic diagram of the test loop is shown in Fig. 1. To simulate deposits on filter surfaces, artificially hardened water with hardness of 1000 mg/L of $CaCO_3$ was made by adding calcium chloride ($CaCl_2$) and sodium carbonate (Na_2CO_3) in proper proportions to tap water. To minimize the abrasion of mechanical parts by calcium carbonate particles, a peristaltic pump (Omega FPU259) was used to circulate the hard water in the test loop. The flow rate in the test system was varied from 50 to 400 mL/min using a valve in a flow meter. In all experiments 5% of the untreated water was bypassed for the purpose of the creation of the tangential flow along the filter surface. It is of note that some tangential flow was believed to be necessary for the successful removal of the unwanted deposits from the filter surface using the spark-generated shock waves.

Usually filters have to be cleaned or replaced when excessive amounts of foreign materials are accumulated on the filter surface. The decision to clean or replace a filter is often based on the changes in flow rate or pressure drop across the filter. When the pressure drop increases to a pre-determined value or the flow rate reduces to a pre-determined value, the filter is cleaned or replaced. In the present experiment the pressure drop across the filter with a filter surface area of 25 cm^2 was measured using a differential pressure transducer (Omega PX137-015AV). The analog signal from the pressure transducer was collected and digitized by a data acquisition system (Dataq DI-148U) and processed by a computer.

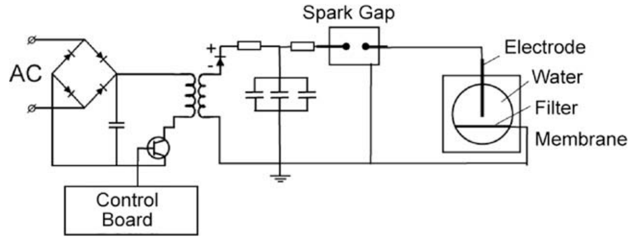

Fig. 2. Schematic diagram of a pulsed power system used in the present study.

A pulsed power system in the present study consisted of three components: a high-voltage power supply with a capacitive energy storage, a spark-gap based switch, and a discharge source immersed in water. A schematic diagram of the pulsed power system is shown in Fig. 2. The high-voltage pulses were provided by a pulsed power supply. The power supply charged an 8.5-nF capacitor bank and the pulse was triggered by an air-filled spark-gap switch. Arc discharge was initiated in the switch from the overvoltage produced by the power supply and capacitor, and the spark gap made use of a very low impedance of arc to transfer high-power energy within nanoseconds. Power deposited into water was analyzed by measuring the current passing through the discharge gap and the voltage drop in the gap. For measurements of the current a magnetic-core Pearson current probe was utilized (1 V/Amp +1/−0% sensitivity, 10 ns usable rise time, and 35 MHz bandwidth). Voltage was measured using a wide bandwidth 1:1000 voltage probe (PVM-4, North Star Research Corp.). Signals from the current and voltage probes were acquired and recorded by a Digital Phosphor Oscilloscope (DPS) (500 MHz bandwidth, 5×10^9 samples/s, TDS5052B, Tektronix). Acquired data were then integrated using a customized MATLAB code.

Typical voltage and current waveforms are shown in Fig. 3. A fast rise time (~8 ns) was obtained with the closure of a spark-gap switch. The peak-to-peak voltage was 29.6 kV. The time before the spark formation was about 12 μs. During this period of time, the energy was mostly consumed by electrolysis and streamer development. After this period, the abrupt increase in current indicated that spark was formed when the streamer reached the other electrode. The Full-Width at Half-Maximum (FWHM) of the major current pulse during spark discharge was 3 μs. The typical peak-to-peak breakdown current was 116 A. It is worth to mention that the energy dissipated in electrolysis can be comparable with, or even higher than the energy deposited in spark, especially at high-conductivity water conditions, because of the conduction current. The pulsed energy stored in the capacitor E_b was about 2.0 J, which was calculated by

$$E_b = 0.5CV_b^2 \tag{2}$$

where C was 8.5 nF, and V_b the capacitor voltage was 21.5 kV. The value was much lower than the peak-to-peak electrode voltage because of the oscillation in electric circuit upon the closing of spark gap. By integrating the voltage and current, the energy deposited into spark discharge was calculated as

$$E_p = \int_{t1}^{t2} V(t)I(t)\mathrm{d}t \tag{3}$$

where $V(t)$ and $I(t)$ is the voltage and current measured by the oscilloscope, respectively, $t1$ and $t2$ is the starting and ending time of the spark. The result was approximately 1.9 J/pulse, showing that

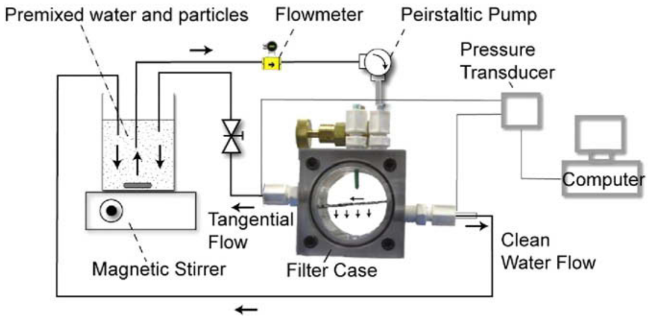

Fig. 1. Schematic diagram of the testing loop.

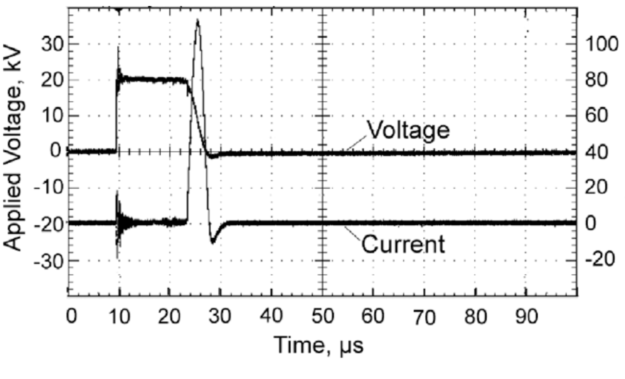

Fig. 3. Typical voltage and current waveforms of a pulsed discharge in water.

most of the energy stored in the capacitor finally went into the spark discharge.

The spark discharge source in water consisted of a stainless steel 316 wire electrode (anode) with a radius of 2 mm and an exposed length of 5 mm, and a stainless steel mesh which acted as both a filter surface and grounded cathode. The tip of the anode electrode was sharpened to 0.2 mm diameter to provide a field enhancement. The distance between the anode electrode and stainless steel mesh was 10 mm. The opening in the stainless steel mesh was 10 μm. The electric conductivity of the tap water (provided by the City of Philadelphia) used in the present experiment was approximately 400 μS/cm. The value was maintained at 1000 μS/cm after the introduction of $CaCl_2$ and Na_2CO_3. No significant change was observed in the conductivity after the application of the spark discharge.

3. Results and discussion

Fig. 4 shows the changes in the pressure drop under various flow rates ranging from 200 to 400 mL/min without spark discharge. The pressure drop for a flow rate of 400 mL/min was approximately 50 Torr at the beginning of the test, which approached to an asymptotic value of about 400 Torr at $t = 3.5$ min, indicating that the filter was fully covered by the particles. In all three cases of different flow rates, the pressure drop slowly increased during the first 30 s. In the following 2–3 min the pressure drop increased rather rapidly, arriving at respective asymptotic values.

Fig. 5 shows the long-time response of the pressure drop across the filter surface after one single spark discharge at three different flow rates of 200, 300 and 400 mL/min. We could visually observe that some particles were dislodged from the filter surface and were pushed away from the filter surface by tangential flow, and a sudden change in the pressure drop immediately following the single spark discharge confirmed the removal of the deposits from the filter surface.

The cleaning effect can be explained by the pressure pulse produced by spark discharge. A number of researchers studied the bubble growth by spark discharge in water [16–19]. One of the most effective models is Kirkwood-Bethe model [12] as given below:

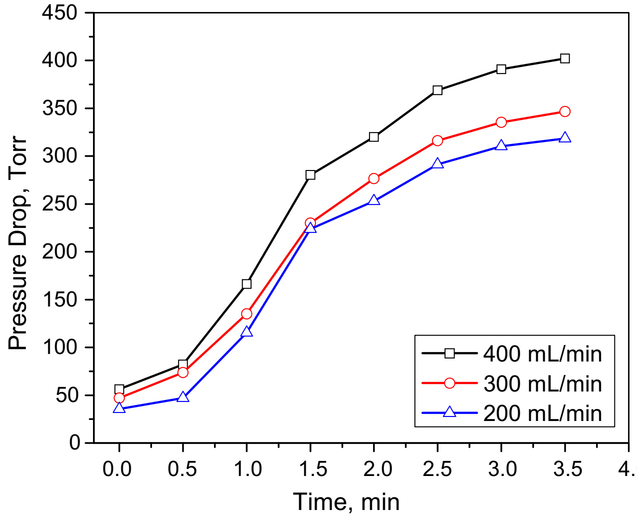

Fig. 4. Changes in pressure drop at three different flow rates with an artificially hardened water.

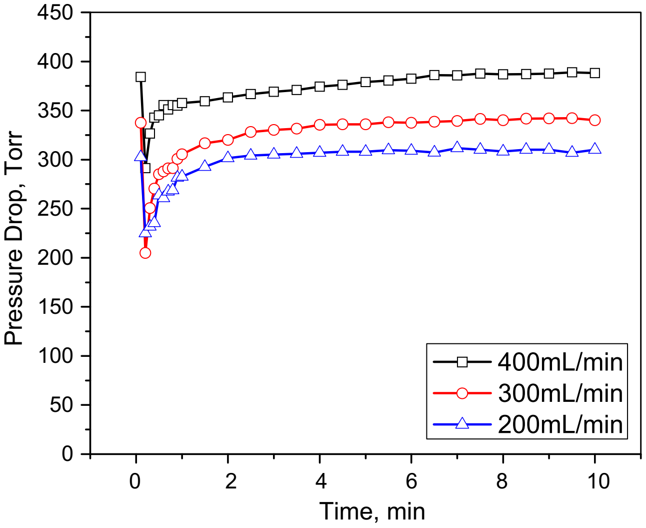

Fig. 5. Variation of pressure drop after one single spark discharge at three different flow rates with an artificially hardened water.

$$\left(1-\frac{\dot{R}}{C}\right)R\ddot{R}+\frac{3}{2}\left(1-\frac{\dot{R}}{3C}\right)\dot{R}^2=\left(1+\frac{\dot{R}}{C}\right)H+\left(1-\frac{\dot{R}}{C}\right)\frac{R}{C}\dot{H} \quad (4)$$

where C and H are the speed of sound of the water and the specific enthalpy at the bubble wall, respectively. R is the radius of the bubble wall. The overdots denote the derivatives with respect to time. By expressing the time derivative of specific enthalpy as a function of derivative of plasma pressure P inside the bubble [12], Lu et al. showed that it was possible to solve P as:

$$P(r,t_r)=A\left[\frac{2}{n+1}+\frac{n-1}{n+1}\left(1+\frac{n+1}{rC^2}G\right)^{1/2}\right]^{(2n/(n-1))}-B \quad (5)$$

where A, B and n are constants ($A = 305.0$ MPa, $B = 304.9$ MPa, $n = 7.15$), r is the distance from the source of the spark to the pressure transducer, and

$$G=R(H+\dot{R}\dot{R}/2), \quad t_r=t+(r-R)/C_0 \quad (6)$$

Using the above equation, Lu et al. simulated that for a spark discharge with energy of 4.1 J/pulse, the maximum pressure at a distance of 0.3 m can be up to 7 atm [12]. The cleaning effect can be easily explained by the rapid pressure change produced by a spark discharge, which is strong enough to remove any contaminants that have Van de Waals bonds with filter surface.

With a single pulse, it took approximately 3 min for the pressure drop to return to its asymptotic value after the application of the single spark discharge. This suggests that one needs to repeatedly apply spark discharges to effectively remove the particles from the filter surface over an extended period.

Fig. 6 shows the changes in the pressure drop over time for three different flow rates. One spark discharge was applied every minute from the supply-water side (i.e., untreated water side) where the accumulation of suspended particles takes place. For the case of 300 mL/min, the pressure drop decreased from the maximum asymptotic value of 350–230 Torr after the first spark discharge. Since water with particles was continuously circulated through the filter surface, the pressure drop began to increase immediately after the completion of the first spark discharge as shown in Fig. 5. The second and third spark discharges further reduced the pressure drop to 170 and 125 Torr, respectively. The pressure drop again began to increase immediately after each spark discharge. The sixth spark discharge brought the pressure

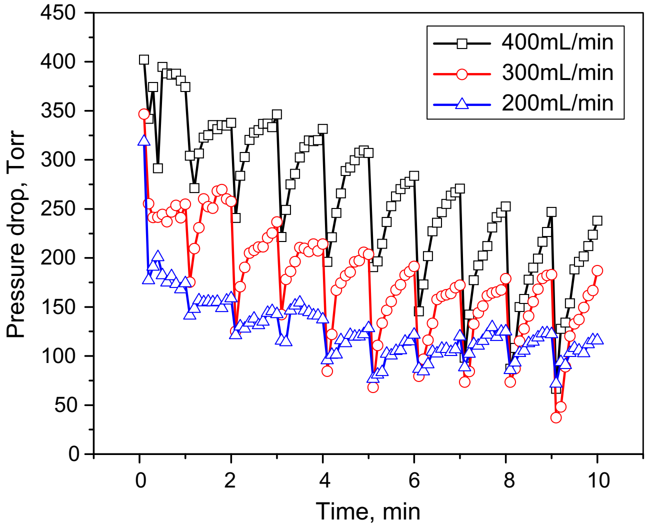

Fig. 6. Changes in pressure drop under repeated pulsed spark discharges with an artificially hardened water.

drop down to a value of approximately 65 Torr, and subsequent spark discharges almost resulted in the minimum value of the pressure drop. For the cases of 200 and 400 mL/min, similar trends of the changes in the pressure drop were observed.

Fig. 7(a and b) shows the changes in the pressure drop under repeated pulsed spark discharges with frequencies of 2 and 4 pulses/min, respectively. Three horizontal arrows indicate the original asymptotic values for three different flow rates, which were the maximum pressure drop due to clogged filter surface by calcium carbonate deposits. First spark discharge significantly reduced the pressure drop in both cases. After that, the rate of the reduction slowed down. The pressure drop oscillations because of the application of spark pulses reached quasi-steady conditions after about 10 pulses for both cases. In these oscillations, the maximum pressure drop decreased to about 45% of its original asymptotic value, while the minimum pressure drop was close to that of the clean filter. These results demonstrate the validity of the present spark discharge method. Note that the present cleaning method using the spark discharge does not require a backwash to remove deposits from the filter surface nor stopping the flow. Furthermore, the present spark discharge method can maintain the pressure drop across the filter at a rather low value (i.e., almost close to the initial clean state), thus providing a means to save not only fresh water but also electrical energy for the operation of pump and required for the backwash in the conventional backwash system.

Fig. 8 shows the changes in the pressure drop over time with the anode electrode placed beneath the filter membrane (i.e., plasma discharge was applied from the treated water side). In this case, the momentum transfer from the shockwave to particles on the filter surface was indirect and had to go through the membrane. Fig. 8 clearly shows that the pressure drop did not improve significantly in this case, indicating that the cleaning effect is negligible comparing with the case when the electrode was placed at the untreated water side. The fact that the momentum transfer from the shockwave to the membrane is weak is actually good news. The low energy transfer rate means that the present spark discharge may not deform the membrane significantly and therefore will not damage the membrane, and has the potential to be applied in the cleaning of more delicate membranes such as in a reverse osmosis as well as solid filters over an extended period of time.

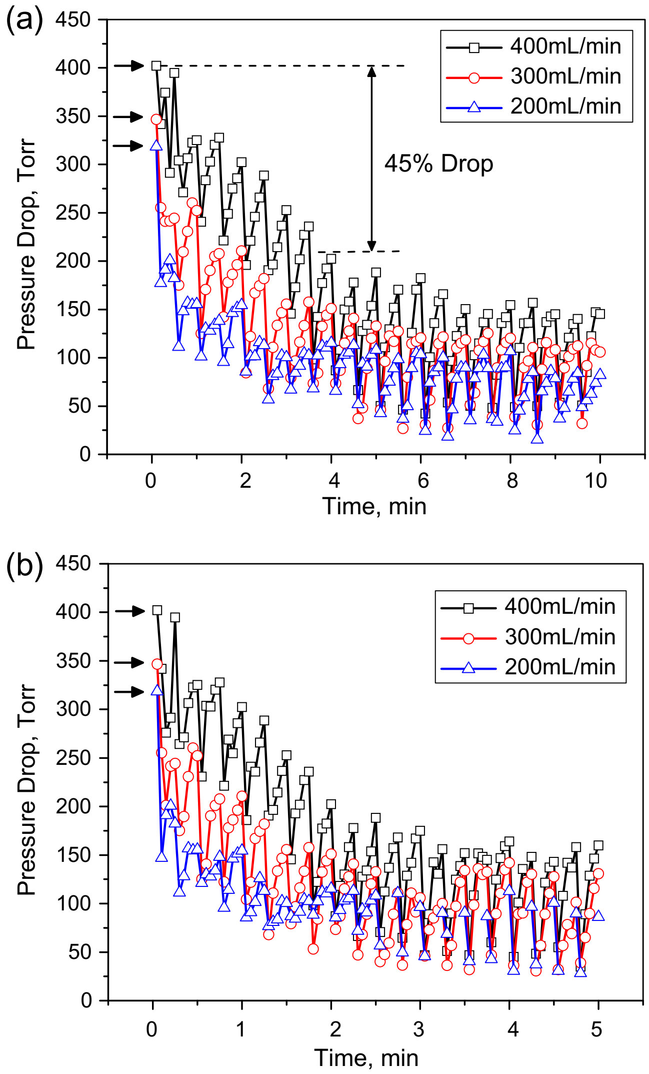

Fig. 7. Changes in pressure drop under repeated pulsed spark discharges with frequency of (a) 2 pulses/min and (b) 4 pulses/min.

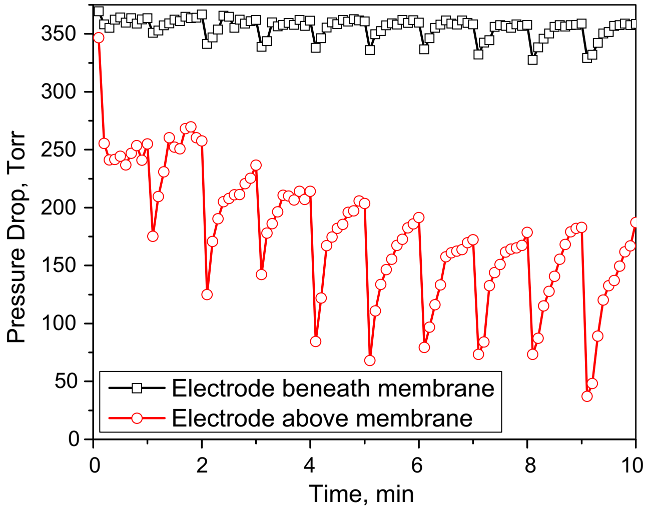

Fig. 8. Comparison of pressure drop across filter membrane under repeated pulsed spark discharges in water: electrode beneath membrane vs. electrode above membrane.

4. Conclusions

The present study investigated the validity of spark discharges in water to remove calcium carbonate deposits from a filter surface. The results obtained in this study demonstrated the benefit of the application of spark discharge for the purpose of keeping filter clean. The spark discharges generated from short electric pulses produced relatively strong shockwaves in water which propagated from a discharge channel in all directions, including the direction toward the filter surface. The momentum transfer from the shock wave produced enough force to dislodge the deposited particles from the filter surface. The dislodged particles were pushed away from the filter by a tangential flow, resulting in a considerable decrease of pressure drop across the filter. The energy consumption is almost negligible, comparing with conventional self-cleaning technologies using backwash flow. The plasma discharge method investigated in the present study can be integrated into a mechanical water softener. The electrical energy required to operate such a system is expected to be on the order of 10–20 W for the flowrate of 10 L/min. In addition to the self-cleaning effect demonstrated in the present study, it was found that the same spark discharge could be used for the deactivation of microorganisms in water, which will prevent biofouling over the filter surface. This will be reported in a separate publication in the future.

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