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Enhanced physical and chemical processes by solid packing in the plasma reactor for the inactivation of *Microcystis aeruginosa*

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ABSTRACT

To investigate the effects of solid packing on the physical and chemical processes and the removal of cyanobacterium (*Microcystis aeruginosa*), the comparisons of properties of the plasma reactors with and without glass pellets and the algal removal efficiency in the plasma reactor were discussed. Experimental results show that more than 27% of the peak value of discharge current and 14.14% of the removal efficiency were increased by the addition of glass pellets. H_2O_2 concentration in the plasma reactors with glass pellets was at least eight times greater than that without them at an air flow rate of 0.75 m^3/h after 40 min of treatment. The algal removal efficiency and H_2O_2 concentration in the system with glass pellets having a diameter of 3–4 mm were higher than that of 7–8 mm. These results implicated that *M. aeruginosa* growth was inhibited by plasma, demonstrating the considerable potential of such an alternative process for water purification.

Keywords: Physical and chemical processes; Cyanobacterium (*M. aeruginosa*); Solid packing; Glass pellets; Plasma reactor

1. Introduction

Recently, periodic and widespread excessive algal growth related to cyanobacteria blooms have occurred in the estuaries and lakes of China, leading to a variety of problems for environmental problems, especially human drinking water safety. The conventional treatment, for the management and reduction excessive algal blooms in the drinking water source, such as chemical coagulation, flocculation and filtration have been proved to be ineffective in removing algae and their toxins [1,2]. Consequently, the economical and free-pollution methods should be studied urgently.

Compared to the traditional water treatment, advanced oxidation processes (AOPs) are attractive alternatives and recently have received considerable attention. Non-thermal discharge plasma techniques, an AOP, have been studied for the gas purification, oxidation of toxic organics, inactivation of microorganisms and treatment of water and wastewater either as a sole means of treatment or in combination with other treatment techniques [3–7]. Non-thermal discharge plasma techniques offer an innovative approach to the cost-effective solution of these problems without secondary pollution.

Breakdown voltage of water is on the order of ~MV as reported in Sunka [8]. Electrical discharges often initiate

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the particles suspended in water or the bubbles, and then initiate in water in the case of the higher electric filed so that the breakdown voltage will be greatly decreased in water with particles or dissolved gases. Air, oxygen, argon, activated carbon particles, Al_2O_3, TiO_2 and ferroelectric pellets were usually used as the artificially created bubbles and particles [9–14]. The plasma reactors with solid packing for water purification have only recently received interests. But the algal removal by plasma in the dielectric barrier discharge (DBD) plasma reactors with solid packing was rarely used in any of the studies.

In this study, the scope of using bipolar pulsed DBD to investigate the feasibility of the algal growth control was assessed using *M. aeruginosa*, as test species. The specific research objectives were to discuss enhanced physical and chemical processes by solid packing in the plasma reactor from the aspects of the electrical discharge characteristics (including the averaged and peak amplitudes of discharge current and current pulse frequency), the removal of *M. aeruginosa* and H_2O_2 concentration. This paper might provide some experimental supports for further research on the removal of *M. aeruginosa* caused by plasma in packed-bed discharge system.

2. Materials and methods

2.1. Experimental setup and apparatus

The parameters of the bipolar pulsed power supply in the literature [15] and the different configuration of the treatment cell is shown in Fig. 1. The height difference ΔH (67.5 mm) is the increased height due to the addition of the glass pellets with

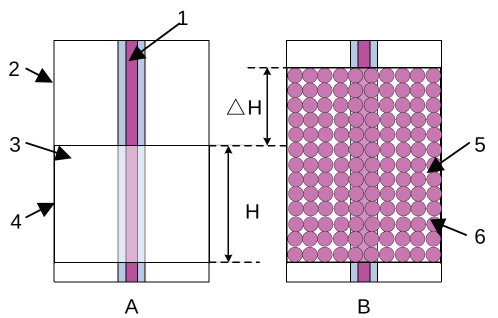

Fig. 1. Different configuration of the treatment cell with and without glass pellets. (1) HV electrode; (2) glass cylinder; (3) algal solution (250 ml); (4) ground electrode; (5) algal solution plus volume of glass pellets (250 ml + Vglass pellet); (6) glass pellet; (7) ΔH, 67.5 mm (the increased height due to the addition of the glass pellets).

the same volume of algal solution at the air flow rate of 0.75 m^3/h.

2.2. Experimental methods

2.2.1. Experimental procedure of M. aeruginosa *removal*

M. aeruginosa cells, provided by the institute of Hydrobiology, Chinese Academy of Sciences, in the exponential growth stage was diluted by the same volume of 0.85% sterilized physiological saline for electrical discharge treatment. A total of 250 ml of algal solution was poured into the treatment cell and treated for 40 min, and then the treated samples were taken out from the reactor for analysis to determine the changes in optical density (OD) of the reaction medium immediately after the interruption of electrical discharge. It was also important to know how electrical discharge process affects the potential of cells to grow in the reaction medium after electrical discharge process stopped. Therefore, the treated samples were subjected to optical light and temperature condition plant, which were 24 ± 1°C temperature under illumination on a 12-h light/12-h dark cycle with 2500 lux light intensity for 1–5 days incubation, to determine the changes in OD during the incubation period [16]. The control sample, which was not treated by plasma, was also exposed to the same conditions as the treated samples.

2.2.2. Determination of the removal efficiency of the algal cells

OD_{680}, as the indirect index of cell viability, is OD of algal culture suspension at 680 nm, which is the maximal absorbance band of *M. aeruginosa* cell suspensions. The removal efficiency of the algal cells by the measurement of OD during 5 days incubation was determined by the following equation:

$$\eta = \frac{CS_0 - TS_t}{CS_0} \tag{1}$$

where η is the removal efficiency; CS_0 is the OD of the control sample and TS_t is the relevant indexes of the treated samples after t min or t day during 5 days incubation after treatment.

2.2.3. Production of hydrogen peroxide in the plasma reactor

Hydrogen peroxide is likely to persist for long period of time among all the physical conditions and chemical species produced by electrical discharge process and the concentration of hydrogen peroxide in de-ionized water (250 ml) was measured by a colorimetric method developed by Joshi et al. [17].

3. Experimental results and discussion

3.1. Effect of solid packing on the electrical discharge characteristics

Fig. 2 shows the typical applied voltage and discharge current waveforms of the plasma reactor with and without glass pellets by digital oscilloscope with envelope mode. The applied voltages generated by bipolar pulsed power supply were the distorted square waveforms. Though time averaged discharge currents were slightly increased with the increasing of the air flow rate for the presence of the larger quantities of the artificially created air bubbles ($\varepsilon_{\text{air bubbling}} \approx 1$) in water, those without glass pellets were weak (as Fig. 2(a) and (b)). Conversely, with glass pellets included, pulse discharge was more intense under the same operational conditions, as shown in Fig. 2(c) and (d).

Time averaged discharge current depends on discharge current peak amplitude and discharge current pulse frequency [18]. For the reactor with glass pellets, discharge current peak amplitude and discharge current pulse frequency were more significant than that without glass pellets, as shown in Fig. 3, e.g., the peak value of discharge current in discharge system without glass pellets obtained at the air flow rate of 0.75 m^3/h was 26 A, but more than 27% of that in the system with glass pellets was reached. Discharge current pulse frequency in the system with glass pellets was higher than that without them, too. Especially, discharge current peak amplitude increased and discharge current pulse frequency decreased with increasing pellet diameter, as shown in Fig. 3(d).

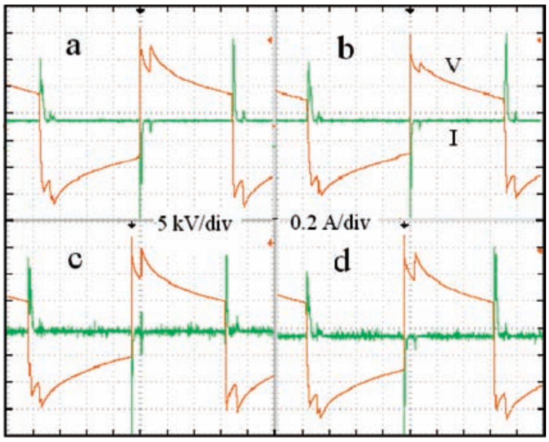

Fig. 2. Typical waveforms of applied voltage and discharge current. Discharge system in the absence of glass pellets with the air flow rate of 0.50 m^3/h (a) and 0.75 m^3/h (b). Three-phase discharge system in the presence of glass pellets having 3–4 mm (c) and 7~8 mm (d) in diameter with the air flow rate of 0.75 m^3/h.

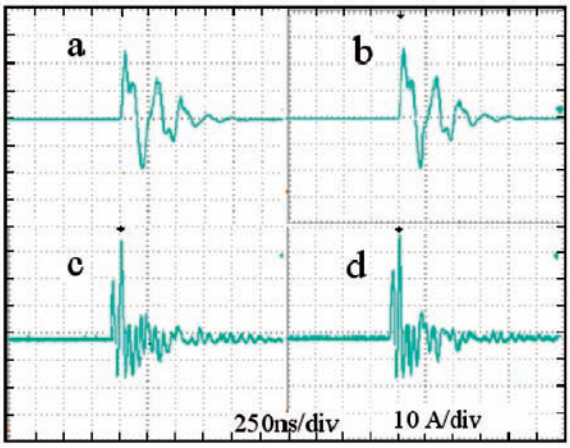

Fig. 3. Temporary waveforms of discharge current (a–d represents the same meaning of Fig. 2).

The breakdown voltage of the system is further reduced when air and particles are simultaneously suspended in water. Meanwhile, the presence of glass pellets makes the existence of small spaces not only around the contact points of the adjacent pellets and but also between the pellets and the electrodes, where a larger number of intermittent micro-discharges occurs in the vicinity of the contact points, modifies the electric field distribution at the contact points [19–21] and then induces current along the surface of the pellets, at least before the discharge takes place. With the higher electric field, discharges also occur on the pellets with no contact with the electrode. The discharges connect the gap between such pellets and the electrode, and then form surface discharges which spread over pellets. The current, flowing from one pellet to another pellet or from the electrode to pellet, concentrates on the immediate vicinity of the contact points and creates large potential differences between the systems with and without glass pellets. Discharge current pulse frequency depends on the surface area/volume of the barrier, glass pellets can be seen as the barrier in some sense. Hence, a reactor with glass pellets shows much higher discharge current pulse frequency than that without glass pellets. The onset voltage tends to decrease with the increasing of pellet diameter, but the larger diameter will also induce the lower electric filed intensity at the pellet surface and the decreasing number of micro-discharges according to the literature [22]. So the current peak amplitude increased and discharge current pulse frequency decreased in the reactor filled with glass pellets having a diameter of 7–8 mm, compared to that of 3–4 mm.

3.2. Effect of solid packing on the removal of M. aeruginosa

The changes in the removal of *M. aeruginosa* in the presence and absence of the glass pellets (3–4 mm in diameter) under the air flow rates of 0.75 m^3/h were investigated (Fig. 4).

The curves show the slight differences of the removal of *M. aeruginosa* between the treated samples and the control sample during two days incubation. But the differences of the curves indicated the removal of *M. aeruginosa* was obvious after two days incubation, indicating a significant residual effect of electrical discharge process on the algal removal in two reaction systems. The removal of *M. aeruginosa* with glass pellets was 11.97% and that without glass pellets was 8.38% at an air flow rate of 0.75 m^3/h as a function of 40 min immediately after electrical discharge, but at the end of the fifth day, more than 87.30% of OD in the plasma reactor with the presence of the glass pellets, within the fifth day, was removed at an air flow rate of 0.75 m^3/h with treatment for 40 min, while the removal efficiency was 73.16% in the case without pellets. As a result, more than 14.14% of the removal efficiency could be increased by the addition of solid packing, glass pellets.

With the same relative dielectric constant, the effect of the diameter of glass pellets on the algal removal was investigated by using glass pellets of 3–4 mm and 7–8 mm in diameter and the results were illustrated in Fig. 5. As these results show, during the incubation period, the removal of *M. aeruginosa* in the system with glass pellets of the diameter of 3–4 mm decreased more obviously than that of 7–8 mm at an air flow rate of 0.75 m^3/h with 40 min of treatment, e.g., at the end of the fifth day, the former removal of *M. aeruginosa* was 87.30%, while the latter was 86.07%.

3.3. Effect of solid packing on the production of hydrogen peroxide

Electrical discharge can produce the chemically active species, including OH•, H•, O•, O_3 and H_2O_2, intense UV radiation and over pressure shock waves. Among them, only H_2O_2, shown to a half-life of 60 h in sterilized surface water, is likely to lead to the residual effects on the algal growth during the storage after electrical discharge due to its persistence for a long period of time [23]. Role of bipolar pulsed DBD on the growth of *M. aeruginosa* in three-phase discharge plasma reactor was discussed in our previous study [15]. So the H_2O_2 concentration in each experimental condition was measured. It can be seen from Fig. 6, under the same operational conditions, that H_2O_2 concentration markedly increased by the addition of glass pellets, e.g., H_2O_2 concentration in the plasma reactor with glass pellets was 38.31 μM after treatment for 40 min, which was at least eight times greater than that in the plasma reactor without them at an air flow rate of 0.75 m^3/h. Meanwhile, the former of the reaction rate constant 3.48 min^{-1}

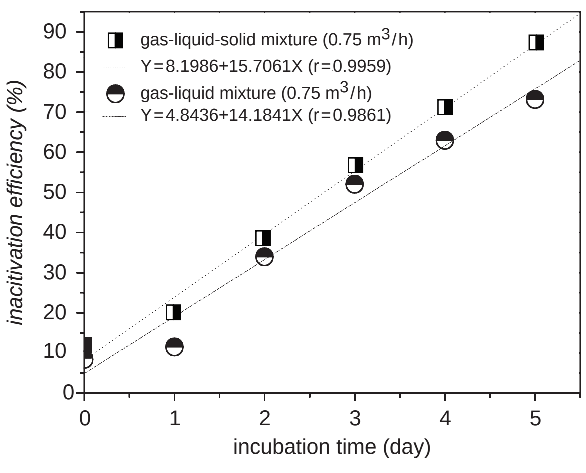

Fig. 4. Effects of plasma reactor with and without glass pellets on the inactivation of cyanobacterium *M. aeruginosa*.

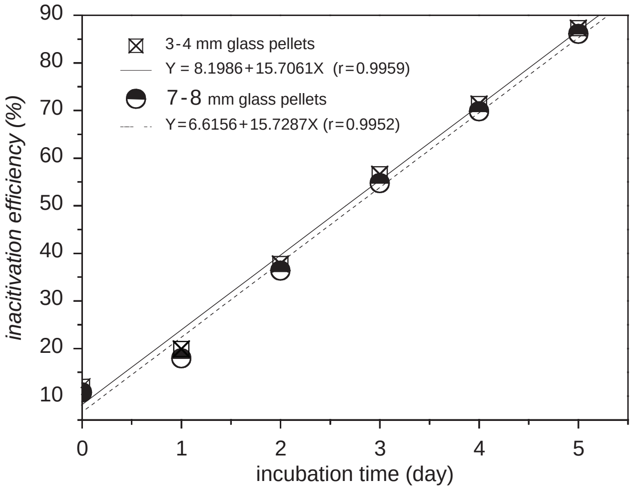

Fig. 5. Effects of the diameter of glass pellets on the inactivation of cyanobacterium *M. aeruginosa*.

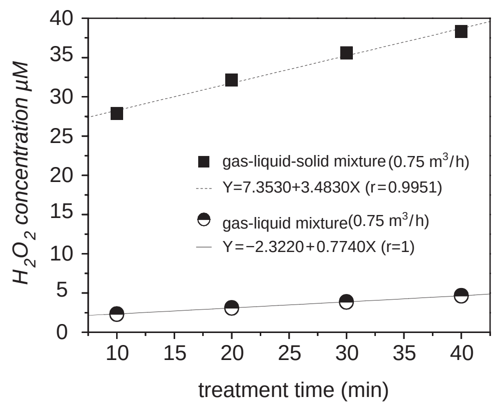

Fig. 6. Effects of plasma reactor with and without glass pellets on concentration of hydrogen peroxide.

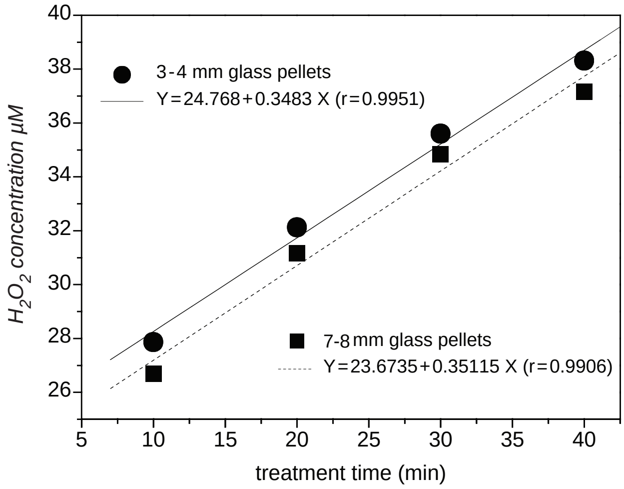

Fig. 7. Effects of the diameter of glass pellets on the concentration of hydrogen peroxide.

was significantly higher than the latter 0.77 min^{-1}. The general trend of H_2O_2 concentration with two different diameters of the pellets was similar to that of the algal removal. The H_2O_2 concentration using a diameter of 3–4 mm glass pellets was higher than that of 7–8 mm with an increase in the treatment time, as shown in Fig. 7.

In addition, the reactor capacitance was increased from 99.8 pF (without glass pellets) – 104.7 pF (with glass pellets) due to the addition of glass pellets so that the amount of power deposited to the discharge increased.

4. Conclusion

In consideration of electrical discharge characteristics, the removal of *M. aeruginosa* and H_2O_2 concentration, the plasma reactor with glass pellets are better than that without them under the same working conditions. The main advantages of plasma reactor with glass pellets are the following:

1. The breakdown voltage of the system decreases and a larger number of intermittent micro-discharges occurs not only around the contact points of the adjacent pellets and but also between the pellets and the electrodes, which enhanced the averaged and peak amplitudes of the discharge current and discharge current pulse frequency.
2. More than 14.14% of the removal efficiency could be increased by the addition of solid packing, glass pellets at an air flow rate of 0.75 m^3/h with treatment for 40 min at the end of fifth day of incubation.
3. Chemical reactions are increased from the aspect of the increase in the concentration of H_2O_2.

Acknowledgements

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