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Diamond & Related Materials 13 (2004) 2016–2019

DIAMOND AND RELATED MATERIALS

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Legionella inactivation with diamond electrodes

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Available online 20 May 2004

Abstract

The DiaCell® technology has been successfully tested against Legionella infection in several water types and under various working conditions. Depending on the water composition, Legionella can be completely inactivated with current densities as small as 50 mA/cm^2 with low contact times (< 5 min). The higher the oxidant concentration in the electrolyzed water, the more rapid is the Legionella inactivation after injection. Bicarbonates in contaminated water were identified as very good supports for electrochemical disinfectants production for Legionella inactivation without high chlorine concentration. At the same time, sulfates in water do not provide any disinfection capacity by DiaCell® electrolysis.

Keywords: Diamond electrodes; Electrochemistry; Water disinfection; Water treatment

Boron doped conductive polycrystalline CVD diamond exhibits good electrochemical stability together with the largest overpotential in known electrodes for water electrolysis. In particular this combination of properties is the reason for increasing efforts [1,2] with the aim of developing highly efficient electrochemical processes with Boron doped diamond electrodes, known also as BDD electrodes. The developments focus on water disinfection, in particular *Legionella pneumophila* inactivation for domestic water treatment purposes or industrial cooling water systems to avoid the propagation of legionaires' disease.

Diamond electrodes are produced by mainly coating conventional electrode materials like thin film conductive diamond deposition on highly doped polycrystalline or monocrystalline silicon substrates. For diamond deposition the CVD-technology with maximum deposition surfaces of 100 and 200 mm in diameter or up to 200 × 300 mm depending on available substrate geometries.

BDD/Si electrodes are applied in typical closed and pressurized electrolyzer especially developed for silicon substrates, called DiaCell®. The DiaCell® technology has been successfully tested against Legionella infection in several water compositions and under various working conditions in vitro.

1. Experimental

1.1. Electrochemical tests

Fig. 1 shows the schematic unit employing DiaCell® to be used for preparing electrolyzed water which are fabricated from tap water or deionized water (approximately 1 μS/cm DI water conductivity, $T = 22–26$ °C). Tap water used for tests without any addition, with NaCl injection (75 ppm chloride) and with NaOCl injection. On the other hand, the DI water was used with different salt dissolutions, i.e., 330 ppm NaCl, 440 ppm Na_2SO_4 or 476 ppm $NaHCO_3$. Several current densities (25, 50, 100 or 150 mA/cm^2) were applied to the DiaCell® with an electrode active surface of 65 cm^2. The hydraulic flux is fixed at 160 l/h (DP = 1.4 bar) and the water temperature was maintained between 22 and 26 °C. Five milliliters of Legionella are injected into each water sample without any treatment (respectively injection) and into samples of electrolyzed water. No test was performed passing Legionella through the DiaCell® electrolyzing.

1.2. Analysis

All analysis of a total disinfectant (oxidant) concentration and a microbiological bacteria counting were conducted at 0, 5, 20 and 60 min after injection of the Legionella. The

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doi:10.1016/j.diamond.2004.03.006

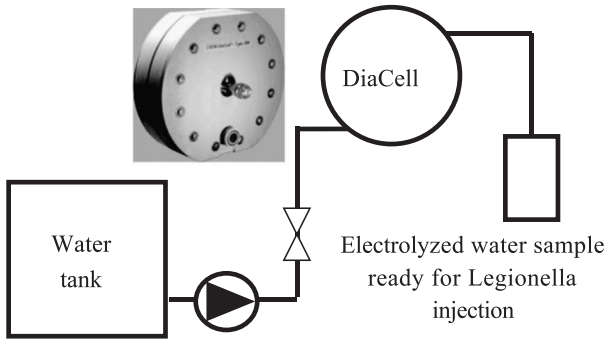

Fig. 1. The schematic unit employing DiaCell® to be used for fabricating electrolyzed water.

total disinfectant concentration was measured by DPD standard method and expressed as total chlorine (ppm), including all the disinfectants in the solution, free or combined chlorine, peroxo-disulfates, peroxo-bicarbonates, hydrogen peroxide, and etc.

Total viable counts of Legionella were performed using two procedures;

1. Plating of 0.1 ml and/or 1 ml directly onto GVPC agar (Biomérieux) and incubating at 37 °C during 5–7days.
2. Concentration of 1 l of water using 0.45 μm membranes (cellulose nitrate, nalgene). Each membrane was placed in 5 ml sterilized tap water, flushed 5 min with sterilized N_2 and agitated for 5 min to detach the bacteria. Part of the suspension (0.1 or 0.2 ml) thus obtained underwent immediate culture examination on GVPC agar (Biomérieux) and was incubated at 37 °C during 3–5 days. The remaining part (4.8 or 4.9 ml) was decontaminated by exposure to a temperature of 50 °C for 30 min (heat treated sample). The heat treated sample was then placed in 50 ml of sterilized 1% yeast extract with added growth and selective GVPC supplements (Biomérieux), and incubated at 37 °C while agitating (150 rpm) during 3–5days. The presence of Legionella was demonstrated by plating 0.1 ml and/or 1ml directly onto GVPC agar (Biomérieux) and incubating at 37 °C during 3–5days.

2. Results and discussion

Before each test of inactivation Legionella was injected into the tap water sample, and it was confirmed that the Legionella viability remains stable in the tap water during test duration of 1 h as shown in Table 1.

Table 1
Legionella viability in tap water over test duration

Time (min)	Legionella (CFU/l)	Elimination (%)	Oxidant (ppm as Cl_2)
<1	44,000,000	0	0
20	42,000,000	0	0
60	46,000,000	0	0

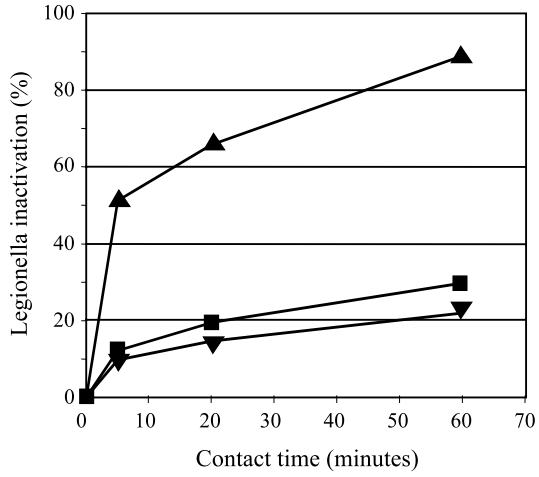

Fig. 2. Legionella inactivation in electrolyzed tap water; (▼) 50 mA/cm^2–0.11 ppm oxidant as Cl_2, (■) 100 mA/cm^2–0.13 ppm oxidant as Cl_2 and (▲) 150 mA/cm^2–0.19 ppm oxidant as Cl_2.

2.1. Legionella inactivation with electrolyzed chloride containing tap water solutions

Tap water containing a very low level of chlorides (3.5 ppm) was electrolyzed. Fig. 2 shows the inactivation of Legionella over contact time and current density. It is interesting that at low current densities respectively at a similar low oxidant level (50 mA/cm^2–0.11 ppm oxidant as Cl_2; 100 mA/cm^2–0.13 ppm oxidant as Cl_2) there is no major difference in Legionella inactivation in comparison with the next current density step (+50%), where a factor three in inactivation can be observed. The major inactivation jump seems to occur at low contact times (<5 min). The highest inactivation (89%) was reached after 60 min of contact time at 150 mA/cm^2, respectively, at a 0.19 ppm oxidants level. Fig. 3 shows the difference in Legionella inactivation by dosing chemical HOCl and electrolyzed tap

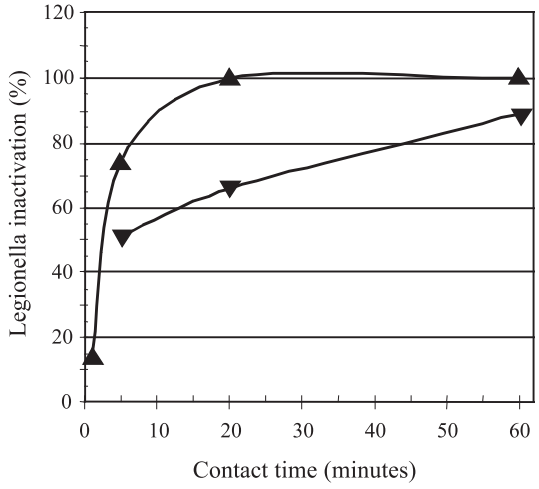

Fig. 3. Legionella inactivation with NaOCl and electrolyzed tap water; (▲) 0.18 ppm oxidant as Cl_2 (tap + NaOCl) and (▼) 0.19 ppm oxidant as Cl_2 (tap—150 mA/cm^2).