



The dosing accuracy of copper and silver ionisation systems: separate high purity copper and silver electrodes versus copper/silver alloys



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ABSTRACT

Copper and silver ionisation is a widely accepted technique to abate and control *Legionella* bacteria in contaminated water. Currently two types of copper and silver ionisation systems are commercially available: (1) systems using separate high purity copper and silver electrodes and (2) systems using copper/silver alloys in various compositions. The aim of this study was to determine the dosing accuracy of these two distinguishable copper and silver ionisation systems. This study showed that both copper and silver can be dosed accurately, according to Faraday's law, with systems using separate high purity copper and silver electrodes as anodes. Systems with new copper/silver alloys (Alloys 70/30 and 90/10) as anodes can only dose copper in predictable quantities. With new copper/silver alloys, silver concentrations were under dosed (<20 µg/l Ag; minimum silver target level at outlets specified to control *Legionella* in water distribution systems). This is most likely caused by a difference in the efficiency of the electrolysis process of copper and silver in alloys. Silver is more oxidation and corrosion resistant than copper (more noble). The dosed silver concentrations with copper/silver alloys remained below the minimum recommended concentration for effective *Legionella* abatement, making alloys unsuitable especially when dosing directly into the water supply.

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1. Introduction

The target organism *Legionella*, usually *L. pneumophila* serogroup 1, is the causative agent of Legionnaires' disease, a severe form of pneumonia, and may cause Pontiac disease, a self-limiting nonpneumonia illness [15,18,1]. Legionnaires' disease can be very serious and can cause death in up to 5–30% of cases [21,11]. *Legionella* outbreaks can, amongst others, occur in water distribution systems including drinking water, process water (e.g., cooling water), fountains and swimming pools. Aerosolized water droplets from these contaminated water systems pose significant health risks to people [9]. Because *Legionella* can cause devastating disease in humans (especially in immuno-suppressed patients), it is important to prevent water systems from becoming contaminated and to control the risk of exposure [10,1]. The control of hazardous pathogens, such as *Legionella* in water distribution systems and cooling towers, is therefore a priority for health authorities worldwide. A well-recognized disinfection method is copper and silver ionisation [19,13,18,26]. The method is based on channelling

the water through a device that applies low potential electricity to copper and silver electrodes. The positively charged copper and silver ions thus released, form electrostatic bonds with negatively charged sites found on bacterial cell walls. These electrostatic bonds create stresses leading to distorted cell wall permeability; coupled with protein denaturation this leads to cell lysis and cell death [25,1]. Importantly, some authors have demonstrated that these ions are able to penetrate the biofilms in which other bacteria, algae, protozoans, and fungi cohabit with *Legionella* species in water pipes [20,8]. The amount of copper and silver dosed must remain within a certain range for efficiency, and at the same time remain below the WHO and other guidelines [3]. Studies carried out in-vitro demonstrated that effective copper and silver concentrations (to inactivate *L. Pneumophila*) are 400 µg/l for copper and 40 µg/l for silver (amongst others [15,16,2]). The target concentrations for *Legionella* eradication are 200–400 µg/l and 20–40 µg/l, respectively [19,16,18]. The maximum permitted levels in drinking water in the Netherlands, and most other European countries, is 2000 µg/l copper [5,7] and 100 µg/l silver [4,30].

Currently there are various copper and silver ionisation systems available on the market. Two authorized types for treatment of drinking water in the Netherlands can be distinguished: (1) systems with separate high purity copper (>99.9 wt.%) and high purity

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silver (>99.99 wt.%) electrodes and (2) systems with a copper/silver alloy (Cu (wt.)/Ag (wt.) 70/30). In other countries various copper/silver alloy compositions are applied (e.g.,: 50/50, 60/40, 70/30 and 90/10). To our knowledge, there are no open source publications available in which the dosing accuracy (the accuracy by which copper and silver can be dosed to drinking water; see Section 2.3 for details) of copper and silver ionisation systems is studied and reported, and in which the dosing accuracy of separate high purity copper and silver electrodes is compared with copper/silver alloys. In this study, we present results of theoretical considerations and practical experiments to establish the dosing accuracy of separate copper and silver electrodes and copper/silver alloys.

2. Methods and materials

2.1. Theoretical considerations

The electrolysis processes in copper and silver ionisation systems are based on Faraday's Laws of electrolysis. According to Faraday's 1st law of electrolysis 'the mass of a substance altered at an electrode during electrolysis is directly proportional to the quantity of electricity transferred at that electrode.' Quantity of electricity refers to the quantity of electrical charge, typically measured in coulomb. Faraday's 2nd law of electrolysis states that, 'for a given quantity of DC electricity (electric charge), the mass of an elemental material altered at an electrode is directly proportional to the element equivalent weight.' Faraday's laws can be summarized by,

$$m = \frac{(Q \times M)}{(F \times z)} \quad (1)$$

where m is the mass of the substance liberated at an electrode in grams, Q is the total electric charge passed through the substance in Coulombs or ampere second, F is the Faraday constant (96485 C/A s for 1 g equivalent), M is the molar mass of the substance in g mol^{-1} and z is the substance's valency. In case of constant-current electrolysis, $Q = I \times t$, and with n (amount of substance in moles) = m/M , Eq. (1) becomes,

$$n = \frac{(I \times t)}{(F \times z)} \quad (2)$$

where I is the electric current in ampere and t is the total time the constant current was applied.

With Faraday's laws the amount of current needed to dose 400 $\mu\text{g/l}$ copper and 40 $\mu\text{g/l}$ silver to water is calculated (see Section 3.1). In addition the theoretical composition of the copper/silver alloys to dose the desired concentrations of copper and silver is calculated (see Section 3.1). However, a complicating factor with dosing copper and silver concentration using copper/silver alloys is the difference in standard potential ($E^\circ_{\text{Cu}} = 0.34$, $E^\circ_{\text{Ag}} = 0.80$, vs. SHE at 25 °C; [6]) of copper and silver within the alloy. According to the galvanic series (e.g. [12]), silver is more noble than copper (more oxidation and corrosion resistant), which implies that copper is preferably released from the alloy upon electrolysis. It is therefore assumed that ionisation with a new copper/silver alloy will lead to a relative enrichment of the more noble silver metal at the electrode surface during the ionisation process. This will result in a continuously changing alloy composition at the electrode surface in which the silver content will continuously increase. The rate of change depends on the electrolyte (e.g., treated drinking water and process water) in which the alloy is ionised and the difference in nobility between copper and silver present in the alloy.

2.2. Laboratory experiments

To determine the dosing accuracy of separate high purity copper (>99.9 wt.% Cu supplied by Staalmarkt) and silver (>99.99 wt.% Ag supplied by Thessco) electrodes and homogeneous copper/silver alloys (Alloys 70/30 and 90/10; standard alloy suppliers), 3 laboratory experiments were performed with new electrodes and alloys. The setup of these experiments is given in Table 1. Experiment 1–3 were performed in accordance to Faraday's law with a power supply with adjustable current regulator. Both copper and silver ionisation systems were connected via a bypass to the same water distribution system. It concerns a water distribution system in an office building in the Netherlands. The average composition of drinking water in the water distribution system – as specified by the water supply company – is pH 7.98, 42 mg/l Cl, 22 mg/l SO_4 , 21.6 mg/l Na, 0.87 mg/l K, 37.3 mg/l Ca, 4.52 mg/l Mg and 5.61 mg/l NO_3 [29]. The systems consist of a control unit with a programmable logic controller (PLC). The separate high purity copper and silver electrodes were placed in separate ionisation chambers (Table 1, experiment 1). Each ionisation chamber contained two electrodes of copper or silver. In most commercial ionisation equipment the polarity is periodically changed to prevent fouling. During polarity change the double layer on the electrode surface will be discharged and recharged. These processes cost a certain amount of Coulomb and this amount of Coulomb depends on the composition of the water and the electrode surface. The higher the ionic strength of the water and/or the higher the electrode surface, the higher the amount of Coulomb necessary to start generation of copper and silver ions by electrolysis. An important issue is that this amount is also not available for the ionisation processes. Polarity change of the separate electrodes (to prevent fouling) has been switched off during the tests because the experiments were conducted within a short period of time (few hours) and fouling did not occur. The copper/silver alloys were placed in a single ionisation chamber (Table 1, experiments 2 and 3). Also here, polarity change was switched off. In both copper and silver ionisation systems, water was channelled through the ionisation chamber(s) and low DC voltage was applied to the separate copper and silver electrodes or copper/silver alloys.

The copper and silver ionisations systems were set to effectively dose the copper and silver concentrations (set target values) as listed in Table 1. The actual dosed Cu and Ag concentrations, according to Faraday's law, were calculated according to Eqs. (3) and (4),

$$\text{Cu}_{\text{cal}} = \left(\frac{(I - 2)/f \times 60}{3.0389} \right) \quad (3)$$

$$\text{Ag}_{\text{cal}} = \left(\frac{(I - 2)/f \times 60}{0.8943} \right) \quad (4)$$

where Cu_{cal} and Ag_{cal} are the calculated dosed Cu and Ag concentrations in $\mu\text{g/l}$, I is the measured DC current in mA over the electrodes or alloy, f is the total flow through the electrode/alloy chamber in l/min, 60 is the conversion factor of seconds to minutes, 3.0389 is the amount of Coulomb (mAs) per μg Cu, 0.8942 is the amount of Coulomb (mAs) per μg Ag and 2 is a correction for the current measured through the electrodes in water in mA. Under the test conditions – temperature of $\sim 8^\circ\text{C}$ and conductivity of 320 $\mu\text{S/cm}$ (at 25 °C) – the electrolysis start at ≥ 2 mA current through the electrodes.

The current measurements ($\pm 2\%$) over the electrodes and alloys is the average current of a PWM (Pulse Width Modulation) current. The set currents over the over the copper and silver electrodes and copper/silver alloys during the experiments are automatically adjusted for variations in water flow. A pair of electromagnetic flow

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