Reduction of Biofilm Formation on Cooling Tower Heat Exchangers using Nano-silica Coating

Environmentally sustainable antifouling coating demonstrated on stainless steel heat exchanger tubes

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Cooling towers are industrial cooling units operating to dissipate heat. As with any surface in contact with aqueous systems, biofilm formation appears on the surface of heat exchangers. Although biofilm formation on plastic tower fill in wet cooling towers has been studied widely, no studies were found regarding biofilm formation on steel heat exchangers in closed-loop systems. In this study, heat exchangers were coated with nano-silica, which is known to reduce the formation of biofilm. Natural biofilm formation was monitored for six months. Biofouling was examined monthly using epifluorescence microscopy by assessing the numbers of live and dead bacteria. It was observed that the biofilm layer formed on the nano-silica coated heat exchanger surfaces was significantly lower than on the control surfaces. 3 log microbial reduction was recorded on coated surfaces in the first month. After six months, total biomass on control surfaces reached 1.28 × 10^{12} cell cm^{-2}, while the biomass on nano-silica coated surfaces was 6.3 × 10^{4} cell cm^{-2}.

1. Introduction

A cooling tower is a heat dissipation unit which cools bulk water in industrial systems. Cooling towers provide cooling by spraying the heated water coming from the system onto a fill material and rejecting the heat to the open atmosphere (1). The cooled water returns to a basin to recirculate again through the system. Common uses of wet cooling towers include air conditioner systems, manufacturing facilities, telecommunication devices and power plants. Such man-made installations provide an ideal environment for bacterial growth similar to an incubator, supported by water temperatures ranging between 24°C and 38°C (2–4). The heated water comes from the source to the heat exchanger that allows the exchange of heat between two liquids at different temperatures by indirect contact inside water jacketed tubes (5).

Wet cooling towers providing cool water for heating, ventilating and air conditioning (HVAC) systems are known to be subject to contamination. Organic and inorganic substances in bulk water are deposited on the water contact surfaces, reducing the heat transfer significantly and threatening the operating stability of the whole system. Established biofilms offer cleaning challenges because they are resistant to most chemical and physical cleaning protocols and they also reduce the heat transfer efficiency (6, 7). HVAC systems are responsible for about half of the energy consumed in modern buildings and industrial facilities. Therefore, biofouling is always a significant issue for heat exchangers and should be taken into account during heat exchanger design and production. As a solution, altering the surface properties could be an effective approach to reduce biofouling in such hard-to-reach articulated systems (2, 8, 9).
A biofilm layer is a community formed by bacterial cells living in a polymeric matrix that they produce; a functional partnership adhered to a living or inanimate surface organised by microorganisms in a dense exopolymer matrix. The capability of microbes to stick to a substratum and to produce a biofilm layer has great significance in a diversity of cooling towers, where fouling can act as a perpetual source of contamination. Biofilm layer must be kept to a minimum in order to prolong the operating life of man-made water systems and facilitate control of pathogens. Disinfectants may be used for this purpose (4, 9).

Industrial cooling towers can be manufactured from different materials. Generally, towers are made of reinforced concrete or fibreglass, stainless steel, wood or reinforced plastic sheets. The fill material is generally made of plastic sheets (polypropylene, polyethylene or polyvinylchloride) where heat dissipation occurs. For corrosion resistance, towers are specially treated, painted and covered with a protective film layer (7). In the case of corrosive water or atmospheric conditions, the use of plastic towers is recommended. But heat exchanger units are made of stainless steel or copper for better thermal conduction (5). The critical issue that affects cooling is the aggregation of deposits over the heat exchanger surfaces which includes biofouling. Conventional steel heat exchangers may have corrosion or deposits may have formed on the heat exchanger tubes. Both of these factors reduce the heat transfer rate (10). To solve this problem, novel anti-fouling coatings are considered. Nano-silica can be used in the form of liquid composites in many matrices as coating materials. Nano-silica is used in the textile and automotive industries because of its self-cleaning, abrasion resistant, hydrophobic and oleophobic features. It is known that nano-silica is able to create low-cost, hard and tough coatings which are resistant to wear and weathering (11).

Although biofilm formation on plastic fill surfaces in wet cooling towers has been studied widely, no studies were found on biofilm formation on steel heat exchangers in cooling towers. As coating of heat exchangers is not common, the aim of the current work was to limit tenacious biofouling on heat exchangers using a nano-silica coating, which will lead to longer material life, better cooling of water and less clogging in closed-loop systems.

Materials and Method

A brand new fabricated closed-loop cooling tower was monitored for six months. A real size, fully working closed-loop cooling tower system was kept in operation by the manufacturer during the experimental period at the factory test laboratory. The system was filled with distributed network water. Regular blowdown was implemented to limit the concentration of dissolved solids. In a circulation rig, hot process water was kept separate from the cooling water in a closed-loop system (Figure 1). For the experiments, a half portion of the stainless steel (316 SS) heat exchanger tubes were coated with nano-silica and the other part was left without coating. Coating was done by coaxial electrospraying before assembly and left to cure in air for 24 hours. Coaxial electrospraying has several implicit advantages such as high encapsulation efficiency and uniform particle distribution. The coating thickness was between 4–6 µm. Before coating, the heat exchanger tubes were sprayed with 96% ethanol to remove any dirt, oil or grime. This application made the bonding of the coating stronger.

Silica in powder form is hydrophilic. To produce hydrophobic nano-silica, the silica particles were transformed by fluorination to confer hydrophobicity. The final particle size was about 40 nm. The aqueous form of the nano-silica coating contains ethanol as solvent to keep it in liquid form before
use. The final nano-silica product was supplied by a local company. After curing, the coating was solid on the surfaces, and no colour change, shedding or weight loss were observed on any of the coated test surfaces after the experimental period. The stability of the coating was tested in a different study by the present author (9) and the mean overall adhesion capability of the coating was recorded as 1.6 using a pull-off adhesion tester, which matches very well with the general rating of adhesion. Water was circulated over the stainless steel (316 SS) heat exchanger tubes, where natural biofilm formation was allowed to occur. Sampling of the biofilm required dismantling the outer shell of the heat exchanger unit every month. The system temperature water was kept constant at 37°C using an electrical heating unit to eliminate temperature fluctuation which might influence biofilm formation over time.

Pipe segments were cut monthly from the heat exchanger using an angle grinder, kept in a container filled with system water and brought quickly to the laboratory for analysis. LIVE/DEAD® BacLight™ Bacterial Viability Kit (Invitrogen™, Thermo Fisher Scientific, USA) dye was added immediately to cover the surfaces completely to stain the actively respiring and dead bacteria. After 15 min, the surfaces were rinsed with sterile bi-distilled water to remove unattached cells, air dried, covered with immersion oil and cover slip, then examined in the dark. This was repeated every month until the study finished at the sixth month. An epifluorescence microscope (Eclipse 80i, Nikon Instruments Inc, Japan) was used to visualise the biofilm cells in situ. The camera enables counting and taking images of bacteria on solid surfaces, with the signals displayed on the computer monitor. Counting and recording were carried out using special software (NIS-Elements, Nikon Instruments Inc, Japan). Signals obtained from 20 randomly selected regions were recorded. Images were saved for later analysis.

The LIVE/DEAD® kit stains dead cells red and live cells green in colour. The LIVE/DEAD® test kit contains two DNA-binding dyes, propidium iodide and SYTO® 9. These dyes differ in their spectral properties and their ability to enter the living bacterial cell. The first dye in the kit is SYTO® 9, which can pass through the membrane of all bacteria and stain the cells green. Propidium iodide only enters into cells with a damaged cell membrane, allowing them to appear red under fluorescent light. The number of viable and dead bacteria on surfaces can be determined in a single step using a dual emission filter cube (Chroma Technology GmbH, Germany).

For both parameters over the six-month duration of the experiment, the difference between the average bacterial numbers were compared by two-way analysis of variance. A follow-up post-hoc analysis was done in order to determine differences. The difference was considered significant when p < 0.05. SPSS® Version 18.0 (IBM Corp, USA) was used for the statistical analyses.

### Results and Discussion

The bacterial numbers from the LIVE/DEAD® test kit were analysed in situ on the surfaces using the manufacturer’s software during the experimental period for six months. The results are given in Table I. The number of signals per cm² were calculated using the magnification factor. Since the raw data were too scattered, the values are given in the logarithmic (log_{10}) base for better

<table>
<thead>
<tr>
<th>Months</th>
<th>Nano-silica coated test surfaces, cell cm⁻²</th>
<th>Uncoated control surfaces, cell cm⁻²</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Dead (log_{10})</td>
<td>Live (log_{10})</td>
</tr>
<tr>
<td>1</td>
<td>3.6 ± 0.07</td>
<td>3.6 ± 0.05</td>
</tr>
<tr>
<td>2</td>
<td>3.3 ± 0.09</td>
<td>3.6 ± 0.10</td>
</tr>
<tr>
<td>3</td>
<td>3.3 ± 0.04</td>
<td>3.7 ± 0.07</td>
</tr>
<tr>
<td>4</td>
<td>3.0 ± 0.02</td>
<td>3.8 ± 0.04</td>
</tr>
<tr>
<td>5</td>
<td>3.1 ± 0.09</td>
<td>3.8 ± 0.10</td>
</tr>
<tr>
<td>6</td>
<td>3.2 ± 0.08</td>
<td>3.9 ± 0.06</td>
</tr>
</tbody>
</table>
The logarithmic reduction was clearly significant starting from the first sampling. The total bacterial numbers on coated tubes were recorded as 49,090 cell cm\(^{-2}\) after the initial month, and 13,016,957 cell cm\(^{-2}\) on uncoated surfaces after the first month. The results distinctly showed that this type of coating reduces biofouling formation on heat exchanger surfaces from the start of the experimental set-up. The numbers of surface associated bacteria on uncoated control tubes gradually increased and reached \(1.28 \times 10^{12}\) cell cm\(^{-2}\) after the sixth month, at which time the biomass on nano-silica coated tubes was \(6.3 \times 10^{4}\) cell cm\(^{-2}\). No significant rise (p < 0.05) of bacterial numbers on nano-coated heat exchanger tubes was recorded during the six-month period in terms of total biofilm counts. This outcome demonstrates that a nano-silica coating can clearly reduce the bacterial biofilm layers on coated heat exchanger surfaces.

As expected, nano-silica coating slowed down the adhesion and colonisation of bacteria on the substrata thanks to its strong hydrophobic properties. The pH, dissolved oxygen, total dissolved matter and temperature values of the water in the system during the six-month test period were recorded and are given in Table II. The values in Table II were important to monitor circulating water due to the blowdown regime.

It is known that even with conventional cleaning and disinfection regimens, there is a problem fighting against biofilm formation and development of microbial resistance (12). Based on previous studies conducted in this field (13, 14), it is impossible to eliminate the formation of biofilm layers on surfaces, but biofilm formation can be reduced (9, 15, 16). For this purpose, it is possible to modify surfaces with different coatings. The nano-hydrophobic coating changes the surface properties of the material and supports less biofilm formation (16–18). Hydrophobic coatings limit the wettability of the surface, making it difficult for organic and inorganic matter or microorganisms to adhere; and even if they do, they can easily be detached from the surface by physical factors such as laminar or turbulent water shear stress (19).

The issue of antimicrobial coatings has been extensively studied (20–24). The problem with these products is development of bacterial resistance against the agent (11, 25). Even antibiotic-containing coatings have been reported to promote biofilm formation (26). Silver compounds combined with silica, silane and titanium coatings in particular gave antimicrobial properties but the problem of toxicity in medical devices was mentioned (27). In industrial use, the resistance of microorganisms is at the top of the list as a disadvantage (28). In addition, silver compounds in water systems will reach the aquatic environment and appear as a separate environmental problem.

It is also emphasised that anti-biofilm coatings are very important for preventing the formation of a biofilm layer at an early stage (29, 30). However, studies conducted to date are mostly aimed at solving clinical problems and have been done \textit{in vitro} with pure cultures (15, 17, 18, 31–33). Using monospecies biofilms is a sterile approach and cannot represent mixed cultures in the natural environment and their interaction with each other. Sol-gel products and superhydrophobic coatings which are more strongly water repellent (31, 34) have also been tried. It was observed that the life of these coatings was not as long as hydrophobic coatings. On the other hand, the high cost of superhydrophobic coatings was a drawback. Contrary to hydrophobic coatings, some hydrophilic coatings were also found to be effective against biofouling. Holberg \textit{et al.} (8) reported that the bacterial numbers on coated tubes were significantly lower than on uncoated surfaces.

<table>
<thead>
<tr>
<th>Months</th>
<th>pH</th>
<th>Dissolved oxygen, mg l(^{-1})</th>
<th>Total dissolved solids, ppm</th>
<th>Temperature, °C</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>7.33</td>
<td>7.40</td>
<td>110</td>
<td>37</td>
</tr>
<tr>
<td>2</td>
<td>7.48</td>
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<td>3</td>
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<td>4</td>
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<td>7.24</td>
<td>108</td>
<td>37</td>
</tr>
<tr>
<td>5</td>
<td>7.30</td>
<td>7.55</td>
<td>107</td>
<td>37</td>
</tr>
<tr>
<td>6</td>
<td>7.36</td>
<td>7.39</td>
<td>110</td>
<td>37</td>
</tr>
</tbody>
</table>

*The numerical data were the average of three consecutive measurements*
biocide-free silicone coatings showed promising real-life performance on fresh water-cooled heat exchangers and also performed well in laboratory tests.

Ding et al. (35) tested an environmentally friendly antifouling coating product, butenolide, which was designed for controlled release from biodegradable polyurethane. The anti-fouling effect was shown by in situ tests. The main target was marine biofouling, especially larval settlement on surfaces. Since the adhesion of fouling organisms relies on a microbial biofilm layer, inhibition of primer settlement is crucial. Hu et al. (36) sprayed bacterial-anti-adhesive modified polystyrene microspheres to construct bacterially-anti-adhesive surfaces. It can be used on any surface thanks to the lotus effect. It was reported as robust and durable on surfaces. Similar surface engineering strategies focus on altering the physicochemical properties of the material surface. In general, reduced efficacy of regular disinfectants leads to progress in development of antimicrobial surfaces and coatings (37, 38).

Conclusions
This is the first report of a nano-silica coating on a stainless steel cooling tower heat exchanger. The study showed that the nano-silica coating significantly reduced bacterial fouling on surfaces. There are many similar surfaces with biofouling problems which have contact with water and require a solution. Nano-silica has proven to be effective at reducing the formation of biofilms on surfaces and can be applied as a cost-effective, effortless, non-toxic, readily available material. Due to growing restraints on environmental release of biocidal agents and the growing restrictions on the use of disinfectants in man-made water systems, as well as demand to decrease the cost of system maintenance, different ways to limit biofilms in man-made water systems hold much expectation.

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