Corrosion Science 80 (2014) 473–481

Contents lists available at ScienceDirect

Corrosion Science

journal homepage: www.elsevier.com/locate/corsci

Experimental evidence for enhanced copper release from domestic copper plumbing under hydrodynamic control

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ARTICLE INFO

Article history:
Received 13 August 2013
Accepted 30 November 2013
Available online 10 December 2013

Keywords:
A. Copper
B. SEM
C. Microbiological corrosion

ABSTRACT

We present experimental results to analyze copper release from biotic and abiotic pipe surfaces and its relation to flow parameters, from Reynolds Number (Re) 1027–11,618.

For abiotic and biotic surfaces, increasing the flow velocity affected the time scale of the copper release but not necessarily the net amount of total copper incorporated into the bulk flow.

For biotic conditions the total copper concentration in the bulk flow was an order of magnitude higher than for abiotic surfaces (3.65 mg/L vs. 0.32 mg/L). Similarly, higher flow velocities enhance the presence of larger size copper nanoparticles in the bulk flow.

1. Introduction

Copper has been extensively used in household drinking water pipeline systems worldwide, because of its durability, relative low cost, and antibacterial properties against some organisms that are harmful to humans (e.g. *Salmonella enterica* [1,2]). However, copper corrosion and its release from the pipe wall to the water by the combined effect of bio-chemical processes and flow hydrodynamics may produce several types of damages in infrastructure [3] and in household pipes. This may result in an increase of copper concentration with important consequences to human health (e.g. liver diseases [4]), and for other species, like fishes, with concentrations under 0.01 mg/L [5].

Although copper is an antibacterial material, biofilm formation is well documented [6–8]. Biofilm in copper could increase slowly in time with corrosion after 120 days [9], depending on environmental conditions. Traditional studies of biocorrosion do not include the flow as a key factor. However, some researchers have studied copper release in the bulk flow under different flow conditions: Lehtola et al. [10] studied the effect of flow velocity on biofilm formation, and showed that rapid changes in water flow could increase the bacterial and copper concentration in the bulk flow for laminar flow Reynolds number (Re) typically under 2200. Calle et al. [11] studied the influence of flow condition in copper pipes and proposed a hydrodynamic model for copper release under unsteady conditions which demonstrated that diffusive transport from the boundary to the bulk flow is not the main copper release mechanism from copper pipes. Aisopou et al. [12] studied the effect of unsteady-state flow on chemical mixing within the pipe and the consequent change on water quality due to hydrodynamics. Indeed, the authors expressed the need for further experimental research to study the effect of hydrodynamics on surface scouring and the detachment of biotic and abiotic features into the water.

During the initial stages of pipe flushing after stagnation, hydrodynamic processes may also influence corrosion and the release of copper into the water, increasing its concentration in the bulk water [8,11,13]. However, very little work has been done to clarify the mechanisms responsible for flow induced copper corrosion in domestic plumbing pipes and the quantification of the additional amount of copper mass that can be released under the action of flowing water. The different types of flow induced corrosion that have been identified in the literature are [14–17]: (i) Mass-transport controlled corrosion, (ii) phase-transport controlled corrosion, (iii) erosion–corrosion, and (iv) cavitation, in which hydrodynamic processes are likely to enhance pipe corrosion. In particular, through a series of experiments performed in sea water copper pipes [15] found that the threshold in shear stresses for erosion–corrosion is over 9.4 N/m^2. Under steady state conditions this value is reached for Reynolds numbers over 30,000, which is almost 3 times higher than typical household pipes. Other researchers suggest that flow could also enhance other corrosion processes, such as pitting, but this would depend on both, flow and water quality, including pH and chlorine concentration [18,19].

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http://dx.doi.org/10.1016/j.corsci.2013.11.064

Although pipe hydrodynamics is a well-known topic, there are many unanswered questions regarding how the flow in the conduit affects pipe corrosion and copper release, in particular at Reynolds numbers corresponding to laminar and transition to turbulent conditions ($\mathrm{Re} < 10{,}000$), which are often found in domestic water installations. The Chilean regulation for water supply systems fixes the pressure head between 147.1 and 490 kPa in range, and in household installations a typical maximum value for conduit flow discharge is 12 L/min, which in turn implies an approximate Reynolds number of ($\mathrm{Re} \approx 12{,}000$) for 3/4 inch pipes. A tap opening process produces a rapid acceleration of the water contained in the pipes thus generating unsteady shear stresses that might be responsible for an increase of copper release. The transition from laminar to turbulent conditions occurs in pipes between $\mathrm{Re} \approx 2100$ and $\mathrm{Re} \approx 4000$ where the exact threshold value would be strongly dependent on initial conditions [20]. The flow in this regime shows complex structures and processes that makes it difficult to analyze even when reaching the steady state [21]. For high Reynolds numbers, near-wall roughness influence is larger on the flow dynamics because the viscous sub-layer is thin enough and turbulent structures have sufficient kinetic energy to affect the corrosion products [22]. It has also been reported that flushing in pipes aged under biotic conditions could result in a higher copper concentration in the bulk flow than the one obtained after stagnation under abiotic conditions [23]. On the other hand, chemical parameters in tap water are also important. It has been reported that hard water could also affect the rate of corrosion by forming a passivating layer which could protect copper surface [24,25]. Temperature is also relevant in the corrosion process by affecting the rate of corrosion because at higher temperatures copper corrosion increases but also scale stability and formation kinetics; the orientation of pipes also might affect the mixing process of water into the pipe affecting the rate of corrosion [23,26].

Maximum concentration of released copper is typically found after 7–12 h of stagnation [27] due to oxygen consumption. Similarly, the study of aged pipes flushed under different flow conditions has shown that the release of copper might be dependent on flow hydrodynamics; indeed, at the initial stages of water flushing through pipes, an increase in the copper concentration has been observed, evidencing that flow hydrodynamics might enhance an additional loss of material from the pipe surface [8,11]. Experimental results suggest that during these events, part of the increase in copper concentration is due to the presence of nanoparticles in the bulk water, probably induced by shear stress under unsteady conditions [8], but not necessarily in the form of the so-called erosion–corrosion, which is due to high velocities under fully turbulent flows, and is increased by irregular surfaces and exposure of the clean pipe material which were not observed in this study.

The quantification of the link between flow hydrodynamics and copper release in domestic pipes is difficult owing to the transient nature of the initial stages of flushing events which imply a transition from laminar to turbulent conditions. An indirect technique used to measure the flow influence on copper release to the bulk water includes measuring outlet concentrations from pipe experiments and flow volume (V) [8,11]. Moreover, it can be assumed that outlet copper concentrations can be correlated with global flow parameters such as mean shear stress (τ) or mean flow velocity (u). Mean shear stress is usually computed from the cross-section averaged flow velocity or by pressure head differences along the pipe (ΔP) [16,28,29].

In the present work, we report new experimental results on copper release from aged pipes under different flow conditions by taking care in controlling hydrodynamic, physical, biological and chemical characteristics of the flowing water. Our aim is to quantify the rate of copper release from the pipe surface to the bulk flow and to link it to hydrodynamic variables. For that purpose, we performed flushing experiments on copper pipes measuring the total mass of dissolved copper transported by the bulk flow out of a control volume, and several hydrodynamic parameters, such as flow discharge and pressure fluctuations within both edges of the pipe system. The relation between flow hydrodynamics and copper release is analyzed combining experimental observations and an integral form of a copper mass balance equation resulting in an advection–diffusion transport model.

2. Materials and methods

2.1. Experimental set-up

Experiments were conducted using copper pipes (1.95 cm internal diameter, type L, manufactured by MADECO-Chile under ASTM-B88). New pipes were pretreated, using a cleaning protocol previously reported by our group [30]. The protocol consists in a 3 step cleaning: first, 2 min with 0.1 M NaOH, followed by tap water (3 times), and finally washed with MilliQ water (3 times) to remove particles and corrosion by-products eventually formed due to atmospheric corrosion during the storing time.

The experimental set-up used for the flushing experiments consists in a tank with constant water head of 42 cm connected to a piping system formed by PVC and copper pipes (Fig. 1C). The copper pipe tested was located at the end of the system and consisted in a 1.0 m length pipe with an internal diameter of 0.0195 m providing a total volume capacity of 0.3 L (V_o). The total length of the experimental set-up was 3.2 m. At the end of the piping system, the water was collected for copper analysis. The piping system had three ball valves for flow control and was equipped with a continuous electromagnetic flow discharge measuring device and two high response pressure sensors (TP1, TP2, Fig. 1B) at each end of the copper pipe to be described later.

Before reaching the testing section, we ensured that the flow velocity profile was fully developed. For a laminar boundary layer, the required development length can be estimated from $L_e = 0.03R_eD$ where D is the pipe diameter [22]; for turbulent flows the following relationship is commonly used $L_e = 4,4R_e^{1/6}D$ [31]. Using, $D = 0.0195$ m, $\mathrm{Re} \approx 2000$ and 10,000 respectively, we obtained $L_e = 1.17$ m (laminar), and $L_e = 0.39$ m (turbulent). Hence the minimum length of the pipe must be 117 cm at 20 °C. To ensure full development under steady conditions, an additional pipe length was considered to reach 150 cm.

2.2. Flow-stagnation experiments

To examine the effect of different pipe aging times and flow conditions, 8 experiments were conducted (Table 1). For tests #1 to #6, and prior to the stagnation-flushing experiments, an aging process was conducted during 6 weeks. For aging, synthetic water was prepared with MilliQ water and $NaHCO_3$ (99.7% chemical grade, Merck KGaA, Germany) to adjust $[HCO_3^-]$ to 14 mM and pH = 8.5. The objective to have a water with a high inorganic carbon concentration was to induce the formation of a copper carbonate hydroxide film [8]. For the experiments, pipes were prepared, and kept in horizontal position connected to a peristaltic pump (Cole-Parmer Model No. 7553-75), and connected to a tank (Fig. 1A). The water inside the pipe was replaced every 48 h because oxygen is consumed in nearly 50 h [32]. The operating flow discharge for water replacement was set to 0.36 L/min in order to avoid hydrodynamic effects on the pipe surface (high shear stresses and turbulence). Reynolds number computed for this operation was Re = 432.