

# Evaluation of the effect of dosage, pH and contact time on high-dose phosphate inhibition for copper corrosion control using response surface methodology (RSM)

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## Abstract

The response surface methodology (RSM), particularly Box-Behnken design model, was used in this study to obtain the optimum operating conditions for reduction of copper corrosion by-product release using high-dose polyphosphate inhibition. Furthermore, the RSM was also applied to study the main and interactive effects of the parameters investigated. Both analysis of variance (ANOVA) and coefficient of determination ( $R^2$ ) showed that the RSM approach was appropriate for the optimization of high-dose polyphosphate inhibition. The main effects of polyphosphate dosage and pH were found significant in reducing copper release using high-dose polyphosphate whereas the effect of contact time was less significant. In terms of interactions between the effects, the relation between polyphosphate dosage and the pH was the most significant. The optimal polyphosphate dosage, pH and contact time were found to be 22–28 mg/L of polyphosphate as P, 9–10, and 36 h, respectively. The highest total copper release reduction was estimated as 95.86% under the optimum condition. Surface analysis using both SEM–EDX and XRD discovered that cupric phosphate was present on the copper surface treated with high-dose polyphosphate inhibition. This implied that cupric phosphate could be responsible for the reduction of copper release by forming a protective layer.

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## 1. Introduction

Copper leaching from water distribution systems has been a major worldwide concern over the past decades, mainly attributable to more stringent regulatory standards for potable water and rising consumer expectations. Excessive inhalation of copper can cause nausea, cramping, vomiting in the gastrointestinal tract, and cirrhosis of the liver [1]. Public concern on copper corrosion has further risen recently due to the occurrences of “blue water” or copper corrosion by-product release (hereinafter referred as copper release). Blue water is considered as a type of localized corrosion and related to the release of high concentrations

of copper precipitates under conditions expected to induce passivity and adherent corrosion products on the copper surface [2]. Though no definitive mechanisms have yet explained precisely how blue water can occur in practice, a number of suspected factors have been identified: (1) temperature [3,4]; (2) chlorine [4–7]; (3) natural organic matter [3,4,8,9]; (4) microbe [4,10,11]; (5) pH [5,12]; (6) stagnation [3]; (7) anions [13–15]; (8) cations [5,13]; and (9) alkalinity [5]. In the United States, USEPA has specified an “action limit” of 1.3 mg/L copper while WHO has established that copper in drinking water shall be less than 2 mg/L to avoid any health risks associated with the long-term consumption of high concentrations of copper.

Among the most commonly reported copper corrosion control strategies are dosing of corrosion inhibitors (particularly phosphate inhibition), pH/alkalinity adjustment,

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calcium adjustment, and chlorination [3,5,16,17]. Phosphate inhibition appears to be the most frequently adopted approach due to its benefit in preventing calcium scale buildup and the promise of providing a low-cost approach to meeting multiple water quality objectives [18]. Polyphosphate and orthophosphate are the commonly used phosphate inhibitors [19] by drinking water utilities to reduce copper release from distribution system materials and resolve discolored water complaints. According to Edwards et al. [20], about 67% of US water utilities uses polyphosphate or a blend of polyphosphate and orthophosphate as corrosion inhibitors, while the remaining 33% doses with orthophosphate. Phosphate inhibitors can basically reduce corrosion by producing a passivated film on the pipe wall, and thereby inhibiting the electrochemical processes that lead to corrosion.

Inhibitor performance is extremely dependent upon dosage and pH, and frequently pH must be adjusted with inhibitor addition [21]. On the other hand, contact time may also play an important role in inhibition. In practice, phosphate inhibitor doses usually ranged from about 0.07–1 mg/L as P [19]. A phosphate dosage of about 1 mg/L as P was effective for forming the protective film, according to Sheiham and Jackson [22]. Orthophosphate was reported to be more effective than polyphosphate when this low dosage was used in copper corrosion control [20,22,23]. However, the results from polyphosphate use at this dosage could be unpredictable, e.g., it either slightly decreased or increased corrosion, according to Cantor et al. [24]. Conversely, there was a contradictory study revealing that polyphosphate inhibition achieved a greater decrease in copper corrosion than orthophosphate inhibition at low dosage [25].

Polyphosphate has also been found to have a tendency to revert to orthophosphate [20,26,27]. This reversion could reduce the ability of polyphosphate to sequester metals but increase its ability to minimize metal leaching from the pipe wall [28]. According to some studies [26,28,29], various factors such as pH, time and temperature could affect the reversion of polyphosphate to orthophosphate. Furthermore, the role of polyphosphate in corrosion control seemed to be more complex than a simple reversion from one species to another [24].

The statistical response surface methodology (RSM) has been widely used in many engineering fields such as materials engineering, food engineering, chemical engineering, bioprocess engineering, and pharmaceutical engineering to evaluate the effects of individual factors, their interactive effects, and to optimize the engineering process. This RSM approach could potentially be applied in phosphate inhibition for copper corrosion control because it could result in process optimization, reduce process variability, provide closer confirmation of the output response to target requirements, and reduce process duration and overall cost.

While much research has been devoted to the vague low-dose polyphosphate in copper corrosion control [20,24,30],

comparatively little information is available on the effect of high-dose polyphosphate in copper by-product release reduction. On the other hand, though higher pH is claimed to be required for control of copper solubility [31], still very little is known about the effect of pH on polyphosphate performance in copper corrosion control, especially in the typical pH range of drinking water (i.e. pH 7–10). Furthermore, only few studies have been done on the effect of contact time on polyphosphate inhibition. Contact time is of importance in this study because a high-dose polyphosphate inhibition that is efficient in remedying copper corrosion on site would always be a better alternative in practice since it causes less interruption to water supply. To the best of our knowledge, the application of RSM has not been utilized in the optimization of phosphate inhibition for copper corrosion control.

The main objective of this study was to examine the main and combined effects of dosage, pH and contact time on high-dose polyphosphate inhibition in reducing copper release using a RSM approach. In addition, the RSM was also applied to identify the optimal conditions for the high-dose polyphosphate inhibition. The feasibility of using a higher dosage of polyphosphate was of interest in this study because it could reduce copper release from corroded copper pipes within a short period by forming the protective film. Microscopic examination on the surface morphology of the pipes after high-dose polyphosphate treatment was also discussed in this study.

## 2. Experimental

### 2.1. Base solution preparation

All the glasswares used in the tests were soaked overnight in 5% HNO<sub>3</sub> and rinsed with Milli-Q (18.2 MΩ cm resistivity) water. Plastic lab-wares were similarly rinsed several times with RO or DI water before use. Analytical reagent grade chemicals were used for preparation of all solutions throughout the study.

Tap water produced by a local water treatment plant was used as the base solution. The base water solution was examined for its physical and chemical characteristics. The color of water was measured using a Shimadzu UV-1201V Spectrophotometer while turbidity was determined using a HACH 2100N Turbidimeter. The pH of water was measured using a pH meter (Radiometer analytical pH meter model ION check 10) and dissolved oxygen measurement was performed using a YSI 5000 DO Meter. The conductivity of water was determined using a resistance/conductivity meter (HORIBA conductivity meter), the total organic carbon was measured using TOC Analyzer model Shimadzu TOC-Vcsh, and a TDS meter (model pIONner30) was used to measure the total dissolved solids. The alkalinity and acidity of water were determined using the standard titration method according to Standard Methods 2310 and 2320, respectively [32]. The hardness of water was determined using the calculation method

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