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Corrosion control and inhibition mechanism of sodium polyphosphate on highly eroded cement coating pipe surfaces

Xiaohui Bai[a,*] , Yang Wang[a], Ruilian Li[b], Jiao Chen[a], Qunli Rao[c], Shihu Shu[d]

[a]*State Key Laboratory of Microbial Metabolism, School of Life Sciences and Biotechnology, Shanghai Jiao Tong University, Shanghai 200240, China, Tel. +86-21-34203714, email: xhbai@sjtu.edu.cn (X. Bai), wangyang2016@sjtu.edu.cn (Y. Wang), Benita-chen@sjtu.edu.cn (J. Chen)*

[b]*College of Information and computer, Taiyuan University of Technology, Shanxi, 030024, China, email: ruilian_li@163.com*

[c]*Instrumental Analysis Center of Shanghai Jiao Tong University, Shanghai 200240, China, email: qlrao@sjtu.edu.cn*

[d]*State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology. Harbin, 150090, China, email: ssh314@126.com*

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ABSTRACT

Cement coating has been widely used in water distribution systems to protect metal pipes. However, little attention has been paid to the eroded cement coating surface and the iron release control. A complicated biochemical reaction exists between the bulking pipe water and the cement coating surface. While the treated water may meet the standards for drinking water quality, water chemical stability cannot always be guaranteed, resulting in the occurrence of discoloration and sand particles in tap water and complaints from customers about water quality. In this study, the effects of adding sodium hexametaphosphate on pipe water quality and corrosion inhibition from cement coating surfaces were investigated. It was shown that adding sodium hexametaphosphate not only provides an immediate remedy for the highly eroded cement coating surface and inhibits iron releasing, but it also improves water chemical stability and water quality by reducing hardness and total iron content in the bulking pipe water. A short-time inhibition treatment cannot provide residual protection for the eroded cement coating surface. Adding sodium hexametaphosphate does not promote bacteria regrowth in water distribution systems. Surface analysis using SEM, EDS and XRD showed that a protective layer developed on the cement coating surface and was composed of $CaFe_2P_2$, $Ca_2Fe(PO_4)_2(H_2O)_4$, $Ca(Fe_6(OH)_6(H_2O)_2(PO_4)_4)_2$ and $Fe_2P_2O_7{\cdot}2H_2O$ by removing Ca, P, and Fe from the bulk pipe water.

Keywords: Surface corrosion; Cement coatings; Corrosion inhibition; Sodium hexametaphosphate; Chemical stability

1. Introduction

Water suppliers should produce good quality and safe drinking water to their end consumers. However, iron leaching from water distribution systems has become a major worldwide concern during recent decades. Public concern regarding iron corrosion is mainly due to the occurrence of “red water,” or iron corrosion by-product release [1,2]. The corrosion of the pipe network not only deteriorates the water quality, which affects the water’s color, turbidity, hardness and accumulation of microbes but also leads to damage of the pipeline wall via thinning and even perforation [3–7].

Cement coating has been extensively used to protect metal pipes. According to a survey from AWWA, cement or asbestos cement occupies 17% of the 1.4 million kilometers of water mains in the United States [8]. Cement coating has also been used in most areas of China, especially for water mains in Shanghai, where 80% of the pipes

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*Corresponding author.

are coated with cement to prevent corrosion [9]. Although the cement coating is used to protect the pipeline, it can be eroded due to the chemical instability of the processed water, and consequently, metal pipelines can release iron and deteriorate drinking water quality. How to protect or remediate the corroded cement coat on the metal pipe surface without excavation is a big problem facing the waterworks.

Polyphosphate and orthophosphate are phosphate inhibitors commonly used by drinking water utility companies to reduce the release of copper from distribution system materials and resolve complaints of water discoloration [5,10–14]. Some researchers have found that sodium hexametaphosphate can inhibit the iron and steel corrosion via threshold treatment [15,16] and that phosphate inhibitors can inhibit copper corrosion [17,18]. Other studies have reported that phosphate inhibitors can be used to inhibit corrosion of metal pipes to prevent the occurrence of red water [19–22]. Many US waterworks have used phosphate inhibitors in water pipe networks to control metal pipeline corrosion [23]. However, to our knowledge, little attention has been given to the protective abilities of phosphate inhibitors in pipes coated with cement. The interface features between the drinking water and the cement coating are significantly different from those between the drinking water and the metal pipes.

Considering the prevalent use of cement coating pipes in water distribution systems, it is important to study the control strategies required to prevent the release of iron from the cement coating surface. In this study, two used sections of pipeline with and without cement coating were selected to investigate the effects of different dosages of sodium hexametaphosphate on the water quality and the development of protective layers on the corroded cement coated surface.

2. Materials and methods

2.1. Experimental materials

Two used pipe sections with and without a cement coating were cut from different areas of the water distribution system in Shanghai. The elemental composition of the ductile iron pipe is shown in Table 1. The elemental compositions of the new and used cement coatings in the pipe are shown in Table 2. The cement coating in the used pipe section became somewhat loose and yellow due to the corrosion by drinking water, as shown in Fig. 1, and the Ca content in the used cement coated pipe significantly decreased relative to that in the pipe coated with new cement, as shown in Table 2. The used pipe coated with cement was damaged by the corrosive drinking water, which resulted in the release of some iron into the cement coating. These two pipes were used in a real water supply network for approximately 15 years and were 232 mm in length and 95 mm in nominal diameter.

The water flowing into the pipes was tap water obtained from the laboratory. The water quality is shown in Table 3. Sodium hexametaphosphate was used as a corrosion inhibitor and was completely mixed with tap water before being pumped into the two pipes via two separate peristaltic pumps. The hydraulic retention time in the pipes was 10 min, which was controlled using glass rotor flow meters that represent a typical slow flowing status in a terminal water supply system. This experiment began in January 2013 and ended in December 2013. Water samples were taken once every 3 days for chemical analysis.

Fig. 1. Two pipe sections with cement coating (left) and without cement coating (right).

Table 1
Elemental composition of the ductile iron in the cement coated pipe

Element	Cr	Al	As	B	Ba	Co	Ca	Cd	Co
Percent (%)	0.1163	0.2090	0.0352	0.4159	0.0180	0.4581	0.5528	0.0070	0.0084
Element	Fe	K	Li	Mg	Mn	Pb	Na	Ni	P
Percent (%)	79.20	0.0340	0.0015	0.1233	0.6261	0.0047	0.0617	0.0067	0.0566
Element	S	Si	Sn	Sr	Ti	V	Zn	Cu	
Percent (%)	0.0983	0.2522	0.0396	0.0037	0.0751	0.0235	0.0227	0.0637	

Table 2
Elemental composition of the new and used cement coating on the metal pipe surface

Average percent(%)	C	O	Mg	Al	Si	S	Ca	K	Fe	Mn	P
New cement coating	9.39	52.16	0.74	2.14	8.52	0.88	22.75	0.57	2.53	0.31	0.01
Used cement coating	2.64	37.42	3.14	8.61	27.8	1.24	2.02	3.03	6.35	7.20	0.54

Table 3
Influent water quality for the pipe water

pH	Turbidity (NTU)	TDS (ppm)	Hardness (as $CaCO_3$, mg/L)	Total Fe (mg/L)	
7.11–7.23	0.24–0.3	431–448	146–149	0.06–0.08	
Total P(mg/L)	Alkalinity($CaCO_3$, mg/L)	Cl^-(mg/L)	SO_4^{2-}(mmol/L)	LSI	LR
0.01–0.04	64–69	3.16–3.18	0.89–1.17	–0.53 – –0.65	2.52–2.68

2.2. Methods

Chemical characteristics, including the pH, turbidity, TDS, alkalinity, chloride, sulfate, hardness, total phosphorus, total iron, Langelier Saturation Index (LSI), Larson Ratio (LR) and Heterotrophic Plate Count (HPC), in the influent and effluent water were determined using standard methods for the examination of water and wastewater (American Public Health Association, 1998). These two systems were run at 0 mg P/L, 1 mg P/L, 2 mg P/L, 3 mg P/L, 5 mg P/L and 10 mg P/L and lasted approximately one year.

The morphology of the cement coated surface was observed using Scanning Electron Microscopy (SEM) by employing a field emission scanning electron microscope (FE-SEM) produced by the Dutch FEI company (the model for Sirion 200), and the elemental composition of the protective layer was analyzed using quantitative energy dispersive spectroscopy (EDS) by employing a British Oxford's silicon drift detector spectrometer (model INCA X-Act). The resolutions of the FE-SEM were 1.5 nm at 15 kV, 2.0 nm at 10 kV and 3.0 nm at 5 kV. The resolution of the energy spectrum was greater than 129 eV at the Mn (K-alpha) line, and all elements ranging from B5 to U92 could be detected.

The main components of the protective layer at a dosage of 5 mg P/L were measured using a Bruker X-Ray Polycrystalline Diffractometer (model D8 ADVANCE). The voltage was 40 kV, and the current was 40 mA. The scanning speed was 4°/min, and the step size was 0.02°. XRD was used to explain the inhibition mechanism via analysis of the substances on the cement-coated surface [24,25].

3. Results and discussion

3.1. Effect of adding polyphosphate inhibitor on the water chemical stability

LSI is a water chemical stability index for assessing damage to the cement coat. As shown in Fig. 2, the LSI changed insignificantly between the influent and effluent water when no sodium hexametaphosphate was added to the pipe water. However, when the LSI was below 0 and the water was corrosive enough to corrode the cement, iron releasing occurred as shown in Fig. 6. When the sodium hexametaphosphate was added to the pipe water, the LSI in the effluent water increased, and the water was more chemically stable and prevented the corrosion of the cement coating. When the addition stopped, the LSI returned to the same level as that in the influent and the pipe water became corrosive again. The existence of a protective layer on the pipe surface can inhibit iron release for a few days, as shown in Fig. 6.

LR is a water chemical stability index used to assess metal damage. As shown in Fig. 3, LR exhibited the same trend as LSI, and these two indexes can be effectively used to evaluate the water chemical stability in a cement coating pipe system. When no sodium hexametaphosphate was added, the pipe water was chemically unstable and corrosive to the cement coating and the iron pipe and iron was released from the pipe surface, as shown in Fig. 6. When sodium hexametaphosphate was added to the pipe water, the LR decreased and the water became less corrosive. When the addition stopped, the LR increased to reach the level in the influent and became corrosive again. However, the formed protective layer delayed the iron release for a few days.

3.2. Effect of adding polyphosphate inhibitor on the pipe water quality

In Fig. 4, the hardness in the pipe water was significantly influenced by the addition of sodium hexametaphosphate. The differences in hardness between the influent and effluent water from these two pipes became higher, especially for a dosage greater than 2 mg P/L of sodium hexametaphosphate. The addition of sodium hexametaphosphate can attenuate hardness from the water by reacting with calcium ions. In all of the samples, the hardness in the effluent from the pipe with the cement coating was slightly higher than that without the cement coating, which suggests that the hardness of the water was slightly affected by the release of the cement coating on the pipe wall. When the addition of sodium hexametaphosphate stopped, the hardness in the effluent quickly returned to the original level in the influent.

As shown in Fig. 5, the total phosphorus concentration was very low in the tap water. When sodium hexametaphosphate was added to the pipe water, the total phosphorus in the water began to be consumed. Specifically, when the dosage of sodium hexametaphosphate was 5 mg P/L, the total phosphorus consumption immediately increased. However, when the addition of sodium hexametaphosphate stopped, the total phosphorus in the water immediately returned to the original influent levels. This suggests that some chemicals, such as calcium ions, may react with sodium hexametaphosphate in the water [26]. This was also supported by the loss of hardness in pipe water, as shown in Fig. 3.

As shown in Fig. 6, when no inhibitor was added to the pipe water, noticeable amounts of iron were released from the pipe wall, regardless of whether there was a cement coating on the pipe surface. This suggests that the tap water was chemically unstable and corrosive. When the inhibitor was added to the pipe water, the total iron began to decrease

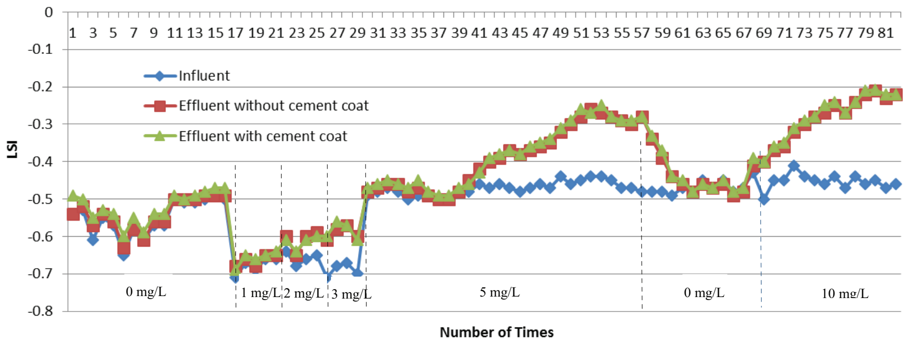

Fig. 2. LSI in drinking water after treatment with different dosages of sodium hexametaphosphate.

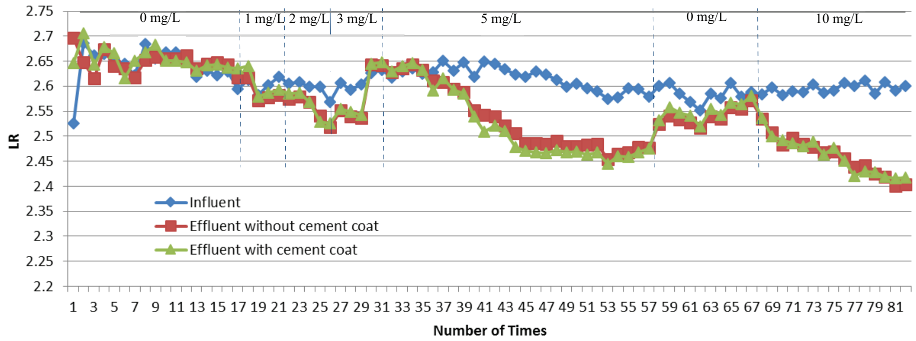

Fig. 3. LR in drinking water after treatment with different dosages of sodium hexametaphosphate.

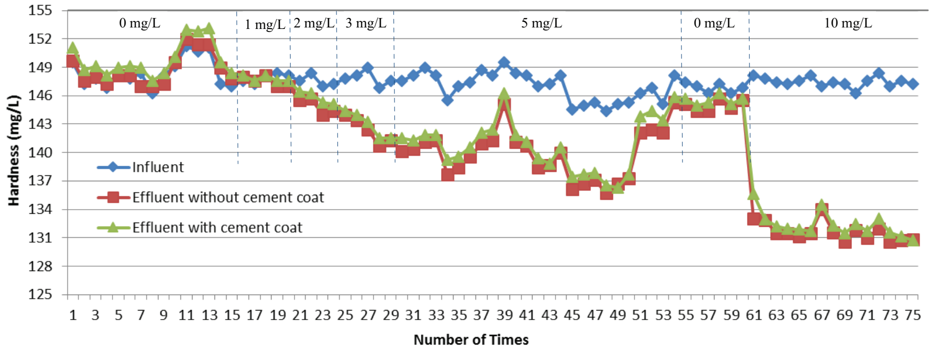

Fig. 4. Hardness in drinking water after treatment with different dosages of sodium hexametaphosphate.

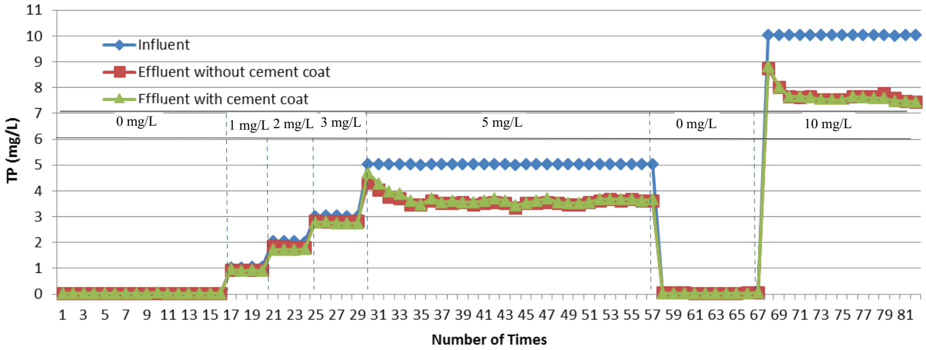

Fig. 5. TP in drinking water after treatment with different dosages of sodium hexametaphosphate.

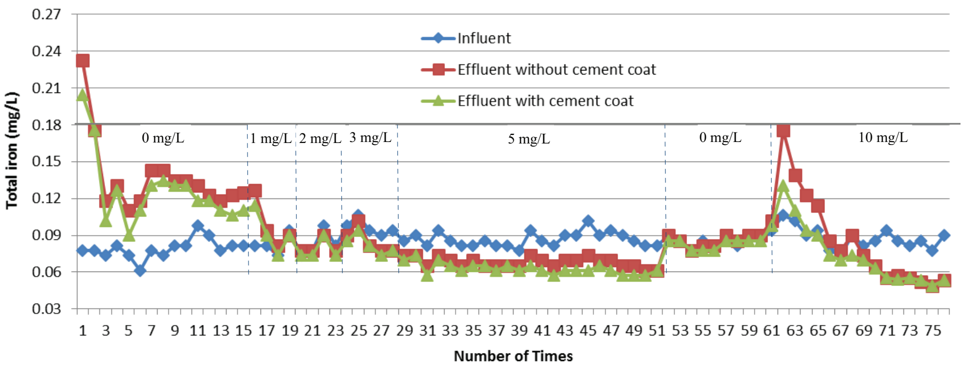

Fig. 6. Total iron in drinking water after treatment with different dosages of sodium hexametaphosphate.

immediately and was soon below the level in the influent, especially when the dosage was above 3 mg P/L. This verified that the release of iron from the pipe surface was inhibited and that some iron in the bulk water could react with sodium hexametaphosphate and was separated from the water. However, when the inhibitor addition stopped, the total iron in the effluent nearly returned to the same level as in the influent, and then the iron release was observed again. This observation potentially resulted from the protective layer that formed on the cement-coated surface, but the layer was not stable and compact enough to provide protection from corrosion over a long period of time. However, during our experimental period, the pipe without the cement coating released more iron into the bulk water than the pipe with the corroded cement coating. A new cement coating on the pipe surface provides protection from iron release, but the corroded cement coat can release iron from the metal surface by passing it through the cement coating.

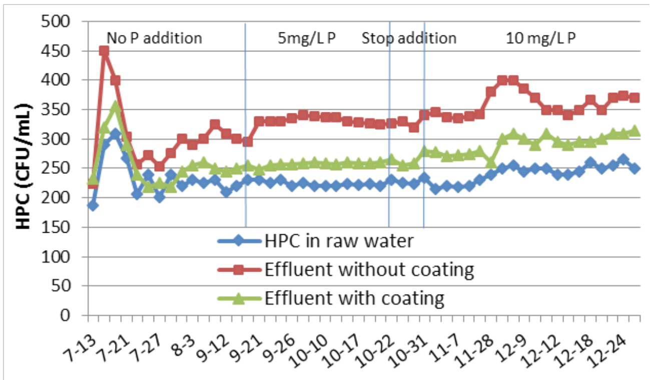

Fig. 7. HPC in drinking water after treatment with different dosages of sodium hexametaphosphate.

3.3. Effect of polyphosphate inhibitor on bacteria regrowth in pipe water

As a key nutrient component, phosphorous is an important impact factor for bacteria regrowth in water. However, little research has been done on the effects of adding sodium hexametaphosphate on bacteria regrowth or biological stability in water distribution systems. As Fig. 7 shows, when different dosages of sodium hexametaphosphate were added to the pipe water at different times, no significant increase in HPC was observed. This means that adding sodium hexametaphosphate can be used to inhibit the erosion of the cement coat and the release of iron from the pipe walls without affecting its influence on bacteria regrowth. However, HPC in the effluent without cement coating was higher than that in raw water and effluent with cement coating. That was perhaps because of the effects of biofilm attached to different types of inner surfaces of the metal pipes.

3.4. Formation and development of the protective layer on cement coating surfaces

The SEM micrograph (Fig. 8) showed that a protective layer developed on the cement coated surface when different dosages of sodium hexametaphosphate were added to the pipe water. As the dosage of sodium hexametaphosphate increased, the protective layer area on the cement coated surface increased, as shown in Fig. 8 A, B, C and D in which 0 mg P/L, 1 mg P/L, 3 mg P/L and 5 mg P/L sodium hexametaphosphate were added to the pipe water, respectively. These SEM micrographs showed that the surface morphology of the cement coating was coarse and uneven. A protective layer over the cement coated surfaces formed discontinuously. However, the passivated layer area enlarged and developed as the addition of sodium hexametaphosphate increased. The polyphosphate inhibitor may reduce corrosion by producing a passivated layer that covers the pores in the cement coated surface, thereby inhibiting the electro-chemical processes that lead to corrosion [27,28].

3.5. Composition of elements and compounds in the protective layer

The elemental composition of the protective layer that precipitated on the cement surface was studied using EDS. Table 4 shows the elemental mass percentages for the protective layer. The changes in elemental mass percentages matched their abundances in the bulk water. The mass percentage of P on the cement surface quickly increased at a sodium hexametaphosphate dosage of 5 mg P /L because more phosphate compounds precipitated (Fig. 3). The

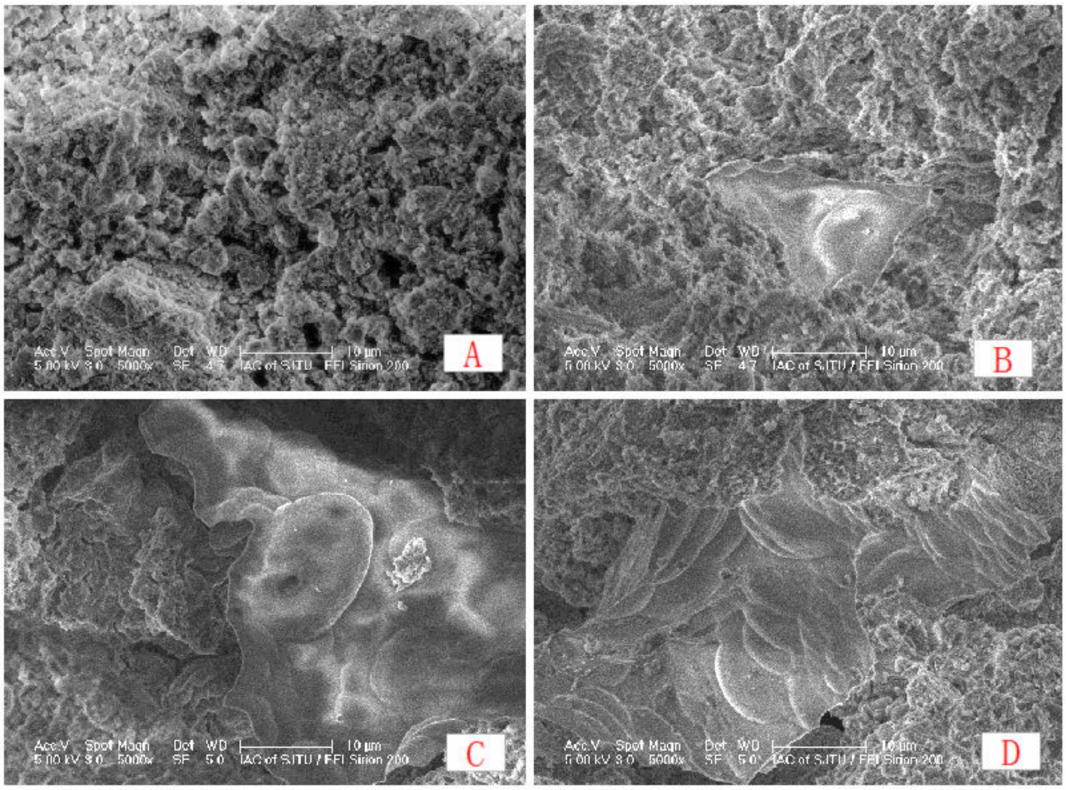

Fig. 8. SEM micrographs of the cement coated surface after treatment with different dosages of sodium hexametaphosphate.

Table 4
Elemental composition in the protective layer on the cement coated surface as a function of sodium hexametaphosphate dosage

Element (%)	Dosage of sodium hexametaphosphate			
	0 mg P/L	1 mg P/L	3 mg P/L	5 mg P/L
P	0.97	1.00	1.12	1.79
Ca	2.20	1.42	1.45	1.66
Si	13.75	11.33	11.91	12.13
Fe	5.25	2.86	3.60	3.67
Al	9.60	5.35	10.88	9.74
Mg	4.79	5.81	9.74	10.88
Others	63.44	72.23	61.30	59.95

higher initial mass percentage of Ca on the cement-coated surface was primarily due to the higher background Ca in the cement. When sodium hexametaphosphate was added to the pipe water, a protective layer formed and Ca in the bulk water combined and settled on the cement-coated surface. Additionally, the mass percentage of Ca in the inhibition layer began to increase (Fig. 2). The mass percent of iron exhibited the same trend as Ca. When no sodium hexametaphosphate was added to the pipe water, the iron in the metal pipe was released into the bulk water because of the corrosion of the metal pipe and damage to the cement coating by the chemically unstable pipe water, as shown in Table 3 and Figs. 5 and 6. When sodium hexametaphosphate was added to the pipe water, the cement-coated surface became covered by a discontinuous layer, which can inhibit the release of iron from the corroded cement coated surface into the pipe water.

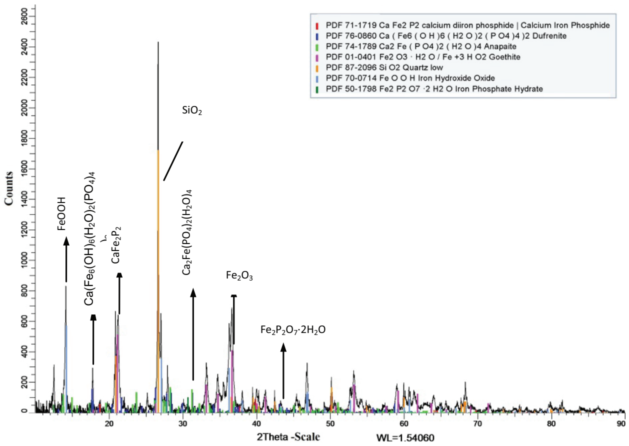

Fig. 9. XRD spectra of the protective layer on the cement coated surface.

The XRD analysis for the surface deposit composition at a sodium hexametaphosphate dosage of 5 mg P/L confirmed that $CaFe_2P_2$, $Ca_2Fe(PO_4)_2(H_2O)_4$, $Ca(Fe_6(OH)_6(H_2O)_2(PO_4)_4)_2$ and $Fe_2P_2O_7{\cdot}2H_2O$ are the main chemicals formed in the protective layer by the removal of Ca, P, and Fe from the bulk water, as shown in Fig. 9. The existence of these compounds implies that the protective mechanism of the passivated layer is quite complicated. Some corrosion products, such as FeOOH and $Fe_2O_3{\cdot}2H_2O$, were also found in the surface deposit due to the uncompleted development of a protective layer that left some areas of the surface uncovered.

4. Conclusions

Cement coatings cannot supply persistent protection against metal corrosion when eroded by corrosive treated water. The addition of sodium hexametaphosphate to the pipe water can provide an immediate remedy for highly eroded cement coating surfaces and can inhibit the release of iron. In addition, sodium hexametaphosphate can improve the chemical stability of the water, removing hardness and iron from pipe water. However, this inhibition treatment cannot provide residual protection when the inhibitor addition is stopped. Continuously dosing sodium hexametaphosphate above 3 mg P/L was required to maintain a low level of iron release at all times. Adding sodium hexametaphosphate does not greatly influence bacteria regrowth in water distribution systems. The surface analyses using SEM, EDS and XRD proved that a discontinuous protective layer developed on the cement coated surface after different dosages of sodium hexametaphosphate were added to the pipe water. This protective layer was primarily composed of $CaFe_2P_2$, $Ca_2Fe(PO_4)_2(H_2O)_4$, $Ca(Fe_6(OH)_6(H_2O)_2(PO_4)_4)_2$ and $Fe_2P_2O_7{\cdot}2H_2O$.

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