

Inhibitive effect of sodium molybdate on the corrosion behavior of galvanized steel in simulated concrete pore solution

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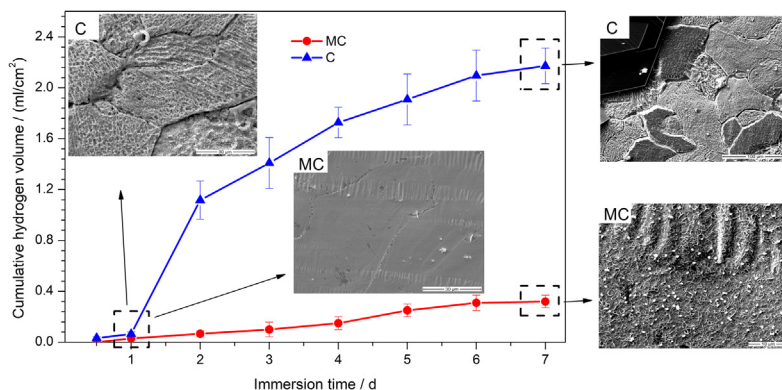
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HIGHLIGHTS

- The inhibitive effect of sodium molybdate on the galvanized coating was investigated.
- Rapid dissolution of the galvanized coating was suppressed.
- Hydrogen evolution of the galvanized coating was also suppressed.
- Mo-containing protective film formed at the corroding interface.
- The formation and properties of the corrosion products was also modified.

GRAPHICAL ABSTRACT



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ABSTRACT

To limit rapid dissolution of zinc and hydrogen evolution of zinc coating in fresh concrete environments at the early stage, the corrosion inhibition of sodium molybdate for galvanized steel in simulated concrete pore solutions with/without chloride ion addition was studied. The inhibitive effect of sodium molybdate was assessed by open circuit potential (OCP), potentiodynamic polarization test, polarization resistance measurement, scanning electron microscopy/energy dispersive spectrometry (SEM/EDS) and X-ray photoelectron spectroscopy (XPS) tests. The results showed that the formation of a Mo-containing film at the corroding interface occurred, which acted as a barrier effect to suppress the rapid dissolution and hydrogen evolution of the zinc layer in the high alkaline environments. Meanwhile, the presence of sodium molybdate also modified the formation and properties of the corrosion products formed on the sample surface.

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1. Introduction

Hot-dip galvanized steel is used as a preventive measure for corrosion control in reinforced concrete structures exposed to carbonation or mild contamination with chlorides [1–15]. The

corrosion behavior of galvanized steel has been widely studied in concrete and some different aqueous solutions as the concrete pores solution, such as $\text{Ca}(\text{OH})_2$ based solutions and alkaline solutions containing KOH and/or NaOH [3,9,13,15–21], these studies showed that the conventional hot-dip galvanized steel was corroded at an acceptable low rate under the lower alkaline environments ($\text{pH} < 13$) due to the formation of a compact passivation film on the surface of the zinc layer. As pH is above 13, the passivation layer is not formed since the size of calcium

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hydroxyzincate crystals are very big and do not cover all the surface of the substrate and the zinc corrodes until all zinc coating is consumed. Hot-dip galvanizing can significantly prolong the lifetime of the construction exposed to carbonation or mild contamination with chlorides. However, as concrete is an alkaline environment, the pH even increases to values above 13 in the first several hours of curing [22,23]. Under these conditions, as the zinc is thermodynamically unstable at this pH, immediately after galvanized steel has been embedded in fresh concrete environments, the zinc rapidly corrodes and hydrogen evolution occurs for a limited period (from several hours to a few days) until the passivating layers on the zinc surface are formed and the concrete hardens. It may result in a consumption of the zinc layer of a thickness around 10 μm at the initial corrosion stage [15,23]. And further data have proved that forming hydrogen irreversibly increased the porosity of cement on reinforcement/concrete interface which can significantly reduce bond strength [2]. These problems connected with the use of galvanized reinforcement in concrete have attracted attention recently [1,2].

Cr (VI) species were traditionally used as active inhibitors and conversion coatings in different coated systems. However, the search for alternatives started due to the health and environmental restrictions imposed on the use of Cr (VI). To avoid/limit corrosion of galvanized steel in the fresh concrete environment, some efforts have been proposed. For instance, galvanized steel pre-treated with conversion coatings or sol-gel coatings before it has been embedded in the simulated fresh concrete environment has been reported [24–30]. On the other hand, an application of non-toxic and environmental-friendly corrosion inhibitor for corrosion protection of galvanized/carbon steel in concrete environments has caused great interest from researchers [31–36]. Sodium molybdate (Na_2MoO_4), which is a commonly used corrosion inhibitor for corrosion protection of galvanized steel in the chloride-containing environment, has been applied as an inhibitor in concrete environments for corrosion protection of carbon steel [34,36].

The inhibitive effect of sodium molybdate for steel has been studied in concrete, which suggested that sodium molybdate is an effective inhibitor for steel in concrete environments. However, the study on the inhibitive effect of sodium molybdate on hydrogen evolution of the galvanized steel in the fresh concrete environments is not available, and the corrosion inhibition mechanism of sodium molybdate for the galvanized steel which corrodes rapidly in the fresh concrete environments in the early corrosion stage is not discussed. Consequently, our paper is aimed to investigate the inhibition effect of sodium molybdate on both rapid dissolution of zinc layer and hydrogen evolution in the fresh concrete environments in the early corrosion stage.

2. Experimental

2.1. Materials and exposure condition

Q235 cold rolled steel sheets (the composition of the steel is listed in Table 1) with a dimension of 40 mm \times 3 mm \times 0.8 mm were chosen as the substrate for galvanized coating. Before hot-dip galvanizing, each Q235 cold rolled steel sheet specimen was degreased, pickled, fluxed and dried as described previously [37], then the specimens were hot-dip galvanized at 450 $^\circ\text{C}$ for 1 min then cooled by water. SEM morphology of the galvanized coating is shown in Fig. 1. In Fig. 1, the grain boundaries and wrinkles on

the coating surface can be evidently observed. The specimens were mounted with epoxy resin leaving an exposed area of 2 cm^2 . Before the exposed test, the samples were degreased with ethanol in ultrasonic, cleaned carefully with distilled water and dried.

Saturated $\text{Ca}(\text{OH})_2$ solution with 0.2 M KOH (pH, 13.2 ± 0.1) was used to simulate fresh concrete pore (SCP) solution [3,25]. To keep a relative constant pH value of the SCP solutions during the test, a layer of calcium hydroxide was designed in the bottom of the plastic beaker, the pH value of the SCP solutions was about 13.2 ± 0.1 during the test. Meanwhile, the fresh SCP solution was carefully added in the plastic beaker to maintain the exposed solution with constant volume (500 ml). In order to simulate a marine exposure, Cl^- was added as NaCl at concentrations of 0.25 M (brackish water) [38]. The composition and pH of the exposed solution are listed in Table 2. The exposed experiment was carried out at temperature 20 ± 2 $^\circ\text{C}$. The samples immersed in the solutions in Table 2 were labeled as Ref., M, C, and MC respectively. Triple samples were repeated for the samples Ref., C, M and MC respectively.

2.2. Characterization methods

Electrochemical tests were performed using a CHI660B potentiostat, a three-electrode cell was used with the Hg/HgO (1 M NaOH) electrode as the reference electrode, a platinum sheet (8 cm^2 in size) as the counter electrode and the specimens (exposed area: 2 cm^2) as the working electrode. The open circuit potential (OCP) was registered as a function of exposed time, at regular intervals, up to 7 days of exposure. For the linear polarization resistance measurements, the applied potential was varied ± 10 mV from the OCP of the samples at a 0.1 mV/s scan rate. Potentiodynamic polarization curves (obtained about 5 mins of immersion after the potential established) were measured in the SCP solutions with a scan rate 0.1 mV/s.

A schematic section showing the experimental arrangement for hydrogen collection with the specimen as used as shown in Fig. 2 [39]. Specimens were mounted with epoxy resin during an exposed experiment during which the evolved hydrogen was collected using the funnel and the burette above the specimen. The burette which was initially full of the SCP solution was displaced by the evolved hydrogen. Cumulative hydrogen was carefully collected in the burette, no leakage of gas through funnel, burette and beaker occurred during the test.

Scanning electron microscopy (SEM), using a Quanta 200 microscope was used to characterize the corrosion products formed on the sample surface. X-ray photoelectron spectroscopy, XPS (Thermo Scientific K-Alpha) was used for coating chemistry analysis with THETA Probe, pass energy: 50 eV, analysis region: 400 \times 400 μm . X-ray diffraction (XRD) was performed for the corrosion products, using a Cu $K\alpha$ radiation, with a counting time of 0.1 s/step, and angular resolution of 0.02 $^\circ$ over the angular range 10–80 $^\circ$ (2θ).

3. Results and discussion

3.1. Characterization of the corrosion products

In Fig. 3, it illustrates the SEM morphology of the sample surface obtained after one day of exposure in the SCP solutions. In Fig. 3a and b, it can be observed that the coating was totally dissolved with grooves/pits on the surface of the samples Ref. and C,

Table 1
Chemical composition of Q235 cold rolled steel sheets.

Elements	C	Si	Mn	P	Al	Cr	Ni	S	Fe
wt%	0.038	0.03	0.21	0.012	0.01	0.01	0.01	0.01	Balance

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