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Influence of Vitamin B3 Intercalated Layered Double Hydroxides (VB3-LDHs) on the Corrosion of Steel Bars in Cement Mortar

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Vitamin B3 intercalated layered double hydroxides (VB3-LDHs) were added to cement mortar, and thermogravimetric analysis (TGA) and mercury intrusion porosimetry (MIP) were used to characterize the effect of VB3-LDHs on the properties of cement-based materials. Electrochemistry methods were applied to study the corrosion protection performance of VB3-LDHs on steel bars in cement mortar at different ages. The surface analysis of the steel bars was carried out using XPS. The results show that 3% VB3-LDHs have a good anti-corrosion effect, and the corrosion protection performance is substantially better than that of pure VB3. On the one hand, this is due to the dual effect of the slow release of VB3-LDHs and the adsorption of chloride ions. On the other hand, VB3-LDHs can effectively improve the compactness performance of cement mortar. The anti-corrosion effect is not ideal at a content of 6%, which may be caused by the poor dispersion of VB3-LDHs at a high content. XPS characterization indicated that VB3 can form an organic film on the surface of steel bars.

Keywords: Vitamin B3, layered double hydroxides, reinforced concrete, corrosion protection

1. INTRODUCTION

The corrosion problem caused by chloride ion attack is an important factor limiting the application of reinforced concrete [1-6]. Common corrosion protection methods include coatings [7], cathodic protection [8,9], and corrosion inhibitors [10,11]. Corrosion inhibitors have received extensive attention due to their economy and ease of construction [12,13,14]. Corrosion inhibitors are generally include inorganic and organic inhibitors. One of the main representatives of inorganic substances is

nitrite inhibitors. This type of corrosion inhibitor has excellent performance but high toxicity and has been banned in many countries [15,16]. Organic corrosion inhibitors usually include alcohol amine organics and plant extracts, etc. This type of corrosion inhibitor has less pollution, but the long-term corrosion resistance is not good [10,13,14].

Layered double hydroxides (LDHs), also known as hydrotalcite, has an outstanding anion exchange effect and has been widely used in catalysis, energy storage, drug delivery, water treatment, etc [17-20]. In recent years, LDHs have also received attention in the field of corrosion protection due to their excellent chloride ion adsorption capacity [21,22]. Researchers usually use the ion exchange performance of LDHs to prepare corrosion inhibitor intercalated LDHs. Corrosion inhibitor intercalated LDHs can slowly release the inhibitor while effectively adsorbing chloride ions in the environment. This dual effect can effectively reduce the amount of added corrosion inhibitor and reduce the chloride ion concentration in the environment [21,22].

In the research field of reinforced concrete corrosion protection, corrosion inhibitors inserted into LDHs are usually inorganic nitrite inhibitors. However, such corrosion inhibitors will seriously pollute the environment, and the application value is not high [23-25]. At present, there are almost no reports about green environmental protection corrosion inhibitor intercalated LDHs in the field of reinforced concrete anti-corrosion. Vitamin B3 is a green organic matter, and it has been verified that it has certain anti-corrosion properties for steel in a concrete environment [26]. In this paper, we studied the anti-corrosion effect of VB3-LDHs prepared by the roasting restoration method on steel reinforcement in cement mortar. VB3-LDHs were added to the mortar in different dosages, and its corrosion protection performance was determined through open circuit potential (OCP) and electrochemical impedance spectroscopy (EIS) tests. The influence of VB3-LDHs on cement hydration and microstructure development was analyzed by TG-DTG and MIP. The film formation mechanism of VB3 on the surface of steel bars was analyzed by XPS.

2. EXPERIMENT

2.1 Materials

Mg-Al-CO₃(Mg₄Al₂(OH)₁₂CO₃.4H₂O) was purchased from Sigma-Aldrich. Vitamin B3 was purchased from Shanghai Chemical Reagent Company. The cement was PO42.5 cement provided by Anhui Conch Cement Company. The water used in the experiment was commercial deionized water. The experimental sand was Xiamen ISO standard sand with a particle size of approximately 1.5-2 mm. Steel bars were purchased from Shandong Xinsheng Company, with a diameter of 10 mm and a length of 80 mm. The type of steel bars was Q235, which consists of C 0.17%, Si 0.30%, Mn 0.61%, P 0.05%, S 0.045% and Fe. The mortar mold was an ordinary PVC pipe with an outer diameter of 40 mm and a length of 120 mm.

2.2 Preparation of VB3-LDHs

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VB3-LDHs were synthesized by the calcination-restoration method, and the specific steps were as follows: CO₃-LDH_s was roasted in a muff furnace for 3h at 450°C to obtain calcined LDHs (CLDHs). CLDHs were added to the mixed solution of VB3 and NaOH according to a solid-liquid ratio of 1g/250 ml, and the concentration of VB3 and NaOH was 0.25mol/L. The mixture was stirred for 15 h at 50°C and a pH of 11 and then filtered and dried to obtain VB3-LDHs. N₂ is passed for 20 minutes before stirring.

2.3 Preparation of mortar sample

Pretreatment of the steel bars: The steel bars were ultrasonically cleaned in absolute ethanol for 10 minutes, rinsed with deionized water, and then quickly dried with a hair dryer. The whole process was repeated twice. The top of the steel bar was welded with the copper wire, and the two ends of the steel bar were sealed with a heat shrink tube. Then seal the junction between the heat shrinkable tube and the steel bar with epoxy resin (the quality of the curing agent diethylene triamine was 8 wt.% of the epoxy resin), and cured for 48 hours. The bars were prepared with 400-2000 mesh sandpaper, and stored in a desiccator.

Preparation and curing of the mortar sample: the mass ratio of the water and cement is 0.55. The mass ratio of cement to sand is 0.4. The contents of VB3-LDHs are 1%, 3% and 6% of the cement mass, and the pure VB3 content is calculated according to the equivalent amount of VB3 in 3% VB3-LDHs. To simulate a severe chloride ion erosion environment, 6% NaCl was added to the cement mortar. After preparation, the mortar sample was placed into a curing box at 20°C and 97% humidity for 24 hours, and then the mold was removed. After removing the mold, curing was continued under this condition for 28 days.

2.4. Electrochemical measurements

The electrochemical measurement adopted the traditional three-electrode system, the saturated calomel electrode was used as the reference electrode, the counter electrode was a platinum electrode, and the working electricity was caused by the steel bars in cement mortar (the electrochemical testing device is shown in Figure 1). Each mortar block was immersed in a separate bucket to prevent mutual interference. The immersion solution was deionized water, and the immersion time was 60 days. The electrochemical workstation used was a Chenhua CHI660e and the EIS parameters were fitted by ZsimpWin software. OCP and EIS tests were performed at specific immersion times. When the value of OCP was stable and changed to less than 2 mV within 5 min, an EIS test was conducted. The interference amplitude of the EIS test is 5mV, and the frequency was 100 kHz to 10 MHz. The temperature of the test environment was $25^{\circ}C\pm1^{\circ}C$.



Figure 1. The Electrochemical testing device.

2.5. Characterization

The XPS instrument was a Thermo Scientific K-Alpha + X-ray photoelectron spectrometer system. The TGA was carried out by a NETZSCH STA449F3 synchronous thermal analyzer in a nitrogen atmosphere of 30-790°C, and the heating rate was 20°C min⁻¹. The instrument used in the MIP tests is the American MacPritice AutoPore Iv 9510.

3. RESULTS AND DISCUSSION

3.1. Thermogravimetric analysis (TGA)

Figure 2 shows the TGA curve of the cement paste cured for 3 d. According to the following formulas (1)-(4) [27,28], the CH content of the blank sample, and sample with 1%, 3% and 6% VB3-LDHs can be calculated as 16.3%, 16.7 %, 17.4%, and 16.2%, respectively.

CH_1 mass percentage= mass loss of $CH \times 4.11$	(1)
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 $CaCO_3$ mass percentage= mass loss of $CaCO_3 \times 2.27$ (2)

 CH_2 mass percentage= $CaCO_3$ mass percentage×0.74 (3)

 CH_T mass percentage= CH_1 mass percentage+ CH_2 mass percentage (4)

where CH_1 , CH_2 , and CH_T represent the mass proportion of direct $CH(Ca (OH)_2)$, the mass proportion of CH converted into CaCO₃, and the total mass of the CH proportion, respectively. The ratio of the molecular weight between CH and H₂O is 4.11, the ratio of the molecular weight between CaCO₃ and CO₂ is 2.27, and the ratio of the molecular weight between CH and CaCO₃ is 0.74.

The cement hydration product containing 3% VB3-LDHs has the highest CH content. According to the relevant literature [27,28], the content of CH in hydration products reflects the hydration degree of cement. The higher the content of CH in the hydration products, the more sufficient the hydration degree. This shows that VB3-LDHs not only have good compatibility with the cement matrix, but also promote the hydration of cement.



Figure 2. TGA curve of cement paste with and without VB3-LDHs: (a) TG curve and (b) DTG curve *3.2. MIP*

Figure 3 shows the MIP data of cement paste with different contents of VB3-LDHs after curing for 3 d. Figure 3a shows that the total pore volume decreases after adding VB3-LDHs. The porosities of the blank sample and samples with 1%, 3%, and 6% are 42.8%, 27.4%, 24.9%, and 40.1%, respectively. The porosity after adding 3% VB3-LDHs is the lowest, followed by 1%. However, there is no obvious effect at the 6% content, which may be caused by the poor dispersion of VB3-LDHs in the cement paste due to the high content. This is similar to the trend of nano silica mixed into mortar [29]. Figure 3b shows that VB3-LDHs exhibit no significant change in the most probable pores of the cement paste after curing for 3 days, and have no significant effect on the pore size distribution.



Figure 3. MIP curve of cement pastes with and without VB3-LDHs: (a) cumulative pore volume and (b) pore size distribution

3.3. Electrochemical analysis

3.3.1. Open circuit potential (OCP)

The OCP value is considered to roughly reflect the corrosion tendency of metals [30]. Figure 4 shows that the OCP of the sample containing 3% VB3-LDHs is the most positive, and there is no obvious

negative shift during the corrosion age of 60 d, indicating that the VB3-LDHs at this dosage have the best anti-corrosion effect.



Figure 4. Open circuit potential of steel bars in cement mortars after immersion for 60 days

The anti-corrosion effect of pure VB3 is obviously weaker than that of VB3-LDHs, and OCP has a negative shift tendency from 28 d. The anti-corrosion performance of VB3-LDHs is substantially better than that of pure VB3. The anti-corrosion effect of 1% VB3-LDHs is also ideal, and the OCP starts to negatively shift substantially after 45 days. The anti-corrosion performance of 6% VB3-LDHs is the worst, even inferior to the blank sample. This may be due to the poor dispersion caused by the excessively high VB3-LDHs content, which causes the mortar itself to be insufficiently uniform and compact [31,32].

3.3.2. Electrochemical impedance spectroscopy (EIS)

Figure 5 shows that in the first 28 days, the capacitive reactance arc of the 3% VB3-LDHs is the largest, and the corresponding phase angle and modulus are also the largest. The capacitive reactance arc diameter of 1% VB3-LDHs is close to 3%. The 6% content of VB3-LDHs had no obvious corrosion inhibition effect. This may be due to the poor dispersion of the high content of VB3-LDHs in the matrix, which in turn makes the matrix nonuniform and dense. The capacitive reactance arc, phase angle and modulus of pure VB3 are significantly less than that of 1% and 3% of VB3-LDHs, which shows that the anti-corrosion effect of VB3-LDHs is significantly better than that of VB3. This is due to the slow-release effect of LDHs on corrosion inhibitors and the ability to adsorb chloride ions, which is consistent with the rule of LDHs loaded inorganic inhibitors and MOF loaded organic inhibitors [21,33]. On the other hand, VB3-LDHs can improve the density of mortar [34].

It can be seen from the EIS spectra of 60 d corrosion (Figure 6), 1% and 3% contents of VB3-LDHs still have a large capacitive arc, and the radius of the 3% capacitive arc is significantly larger than the 1% contents. However, in the first 28 d, this advantage is not significant, which may be due to the low consumption of corrosion inhibitor in the early stage. However, with prolonged with the prolongation of corrosion time, corrosion inhibitors are gradually consumed, which makes the advantage of high dosages more obvious [35]. Compared with the age of the first 28 d, the arc diameter, phase angle and modulus of pure VB3 and 6% VB3-LDHs were significantly reduced, indicating that the corrosion protection ability declined seriously under the long-term corrosion environment.



Figure 5. Nyquist plots and Bode plots of the steel bars in the cement mortars after immersion for 28 days



Figure 6. The Nyquist plot and Bode plot of the steel bars in cement mortars after immersion for 60 days

The fitting circuit is shown in Figure 7. The physical meaning of each component is as follows: R_s represents the resistance of the solution; R_{ct} is the charge transfer resistance, and C_{dl} is the electric double layer capacitance. R_cC_c is caused by the cement mortar cover layer; $R_{ct}C_{dl}$ is caused by the electrochemical reaction on the surface of the steel bar [36,37].



Figure 7. Fitting circuit used to fit EIS data

As seen from Table 1, the R_p value of pure VB3 is higher than that of the blank sample, indicating that it has a certain corrosion inhibition effect on steel bars [38,39]. However, the R_p values of the 1% and 3% VB3-LDHs were 47.9% and 47.3% higher than that of pure VB3 at the age of 28 d. At the age of 60 days, the R_p value of the 3% VB3-LDHs was the largest, which was 76.7%, 87.5% and 26.7% higher than that of the blank, pure VB3 and 1% VB3-LDHs, respectively. This shows that VB3-LDHs have a good long-term anti-corrosion effect at a 3% content. VB3-LDHs can provide a long-term anti-corrosion effect. First, LDHs has good ion adsorption, which can effectively adsorb chloride ions while releasing corrosion inhibitors [21]. In addition, VB3-LDHS can effectively improve the structural compactness of cement-based materials [34]. However, the anti-corrosion effect of 6% VB3-LDHs is poor. This may be due to poor dispersion when the VB3-LDHs content is high, which in turn causes the cement mortar structure to be insufficiently uniform and compact.

	Time	$R_s/\Omega \ cm^2$	C _c /10 ⁻⁵	R_c/Ω	C _{dl} /10 ⁻⁵	$R_{ct}/k\Omega$	$R_p/k\Omega$
			μF cm ⁻²	cm ²	μF cm ⁻²	cm^2	cm^2
Control	3d	185.7	0.001292	455.9	0.001187	7573	8214.6
	7d	205	0.0009164	1403	0.003907	6001	7609
	14d	205.1	0.0009499	1328	0.003852	4433	5966.1

Table 1. Fitting data for EIS

7916.8	6223	0.0008839	1483	0.003515	210.8	28d	
6076.7	5563	0.001024	243.9	2.51E-6	296.8	60d	
12020 4	11520	0.001287	205 5	0.001/3/	212.0	24	20/ VP2
12029.4	11320	0.001267	293.5	0.001434	215.9	50 74	3% V D3
12445.5	12020	0.001165	194.3	0.001203	231	/0	
121/1.8	11/30	0.0006102	168.7	0.001138	273.1	14d	
11368.3	11000	8.555E-6	137.2	0.001011	231.1	28d	
3270.1	2314	6.618E-6	581.9	0.005821	374.2	60d	
18841.2	18500	0.001121	171.5	0.001491	169.7	3d	1%
18338.4	17950	0.001547	168.6	0.001561	219.8	7d	VB3-
21992.6	21560	0.001304	166.8	0.001419	265.8	14d	LDHs
21814.6	21500	0.001155	95.51	4.117E-6	219.1	28d	
19138.5	18190	2.853E-6	614.9	0.001181	333.6	60d	
27035 5	26710	0.001539	155.8	0.001117	1697	3d	3%
22352.8	21960	0.001425	165.6	0.001057	227.2	7d	VB3-
23123.2	22700	0.001029	156.6	0.001175	266.6	14d	LDHs
21564.09	21280	2.836E-6	81.69	0.0009263	202.4	28d	
26102.6	25150	2.768E-6	620.8	0.001023	331.8	60d	
11331.2	11010	0.00152	195 5	0 00144	125.7	3d	6%
8200 7	7811	0.00132	196.8	0.001778	192.9	50 7d	VB3-
7482 4	7070	0.003037	179.6	0.001778	232.8	14d	LDHs
46167	4293	0.0017	134 5	3 979E-6	189.2	28d	
2856.2	1913	1.464E-6	552.8	0.007004	390.4	60d	
210 191 270 223 231 215 261 113 82 74 46 28	26710 21960 22700 21280 25150 11010 7811 7070 4293 1913	0.001133 2.853E-6 0.001539 0.001425 0.001029 2.836E-6 2.768E-6 0.00152 0.003037 0.0014 0.00322 1.464E-6	 95.31 614.9 155.8 165.6 156.6 81.69 620.8 195.5 196.8 179.6 134.5 552.8 	4.117E-0 0.001181 0.00117 0.001057 0.001057 0.0009263 0.001023 0.001023 0.00144 0.001778 0.003312 3.979E-6 0.007004	219.1 333.6 169.7 227.2 266.6 202.4 331.8 125.7 192.9 232.8 189.2 390.4	200 60d 3d 7d 14d 28d 60d 3d 7d 14d 28d 60d	3% VB3- LDHs 6% VB3- LDHs

3.3 XPS

Figure 8a shows the wide-scan XPS spectrum, and the main elements are Fe, O, and C. The content of Fe is 3.9%, the content of O is 36.92%, and the content of C is 55.15%. In order to get a more clear conclusion, it is necessary to divide the peaks of the XPS narrow spectrum and analyze different chemical bonds. Figures 8b, c, and d show the narrow-scan spectra of Fe 2p, O 1s, and C 1s, respectively. The peaks of Fe2p are located at 710.9, 709.2, and 706.6 eV, which are Fe₂O₃, FeO and Fe respectively [40,41]. Through the area ratio of each peak, the proportion of Fe₂O₃ can be calculated as 52.7% and the proportion of FeO is 23.7% respectively, which demonstrates the formation of a passive film on the surface of the steel bar. Figure 8(c) shows that O1s can be divided into two peaks: 531.4 eV corresponds to FeO/Fe₂O₃ [42]. The content of C-O is 74.9%, and the content of FeO/Fe₂O₃ is 25.1%, which further illustrates the formation of passivation films and organic film layers. The C1s spectrum of Figure 8(d) can be divided into three peaks: 288.3 eV and 285.4 eV correspond to C-O, 284.5 eV correspond to C-C/C-H [43], and the proportions of C-O and C-C/C-H are 18.8% and 81.2%, respectively, which indicates that the VB3 organic film is formed on the surface of the steel.



Figure 8. XPS spectra of steel bars in cement mortar containing 3% VB3-LDHs (a) Broad spectrum, (b) Fe 2p, (c) O 1s, and (d) C 1s

4. CONCLUSIONS

Through a series of characterization test methods, the improvement effect of VB3-LDHs on the corrosion protection of reinforcement in mortar and the hydration and structural characteristics of cement were studied, and the following conclusions were obtained:

1. TGA data show that the appropriate amount of VB3-LDHs has a certain promotion effect on the hydration of cement-based materials. The MIP results showed that: 1% and 3% VB3-LDHs can effectively reduce the porosity of cement-based materials. However, the content of 6% has little effect on the porosity, which may be due to the poor dispersion of VB3-LDHs at a high content.

2. The electrochemical results showed that VB3-LDHs with contents of 1% and 3% had significant anti-corrosion effects on the steel bars in the mortar in the first 28 days, and the Rp increased by 47.9% and 47.3% respectively compared with pure VB3. After 60 days of corrosion, the content of 3% is still above 25000, which is much higher than the other dosages of VB3-LDHS and pure VB3, indicating that 3% VB3-LDHs has the best long-term corrosion resistance. XPS analysis shows that VB3 released by VB-LDHs can form organic films on the surface of steel bars.

3. VB3-LDHs improve the anti-corrosion effect of VB3 mainly in terms of two aspects. One is that LDHs have good ion adsorption, which can effectively adsorb chloride ions while releasing corrosion inhibitors. In addition, VB3-LDHs can effectively improve the structural compactness of cement-based materials.

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