Supplementary Information

Passivation Performance and Mechanism of a Novel Self-Healing Composite Passivator on Pyrite

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Text S1 Electrochemical measurements methods

A three-electrode system was used in the electrochemical measurements: a passivated pyrite electrode, a Pt foil sheet electrode (10 mm × 10 mm), and a saturated calomel electrode (SCE) with a Lugin capillary were used as the working electrode, counter electrode and reference electrode, respectively. A 0.2 M Na_2SO_4 solution with pH 2.0 was served as the electrolyte. The electrochemical measurements were performed on a CHI650 electrochemical workshop. To make sure the electrochemical system was stable, an open circuit potential (OCP) test was performed before every electrochemical experiment. ZSimpwin software was used to match the acquired EIS data, which were carried out at OCPs in the frequency scope from 100 kHz to 0.01 Hz with a signal amplitude of 0.005 V. The Tafel tests were performed in the OCP ± 0.2 V/SCE range at a scan rate of 1 mV/s.

Text S2 Characterization

A Tecnai G2 F20 field emission transmission electron microscope (TEM) was used to examine the microstructures of the pyrite specimens. The morphological characteristics of the pyrite surface were analyzed using a scanning electron microscope (SEM, TESCAN MIRA4). The static contact angle tests were carried out using a static contact angle meter (DSA30, Germany). A Revetest scratch tester (CSM-MCT, Switzerland) was utilized to conduct the scratch tests. A Fourier transform infrared (FTIR) spectrometer (Thermo Fisher Model Nicolet iS5) was used to identify the differences in chemical structure between the passivated and raw pyrite. X-ray photoelectron spectroscopy (XPS) was carried out using a Thermo Scientific ESCALAB 250Xi spectrometer with Al Kα radiation. The C1s peak with a binding energy of 284.80 eV was utilized to calibrate the acquired data.

Text S3 8-HQ release experiment

The release characteristics of HH@PE-2, HH@PE-4, and HH@PE-6 in leach solutions at pH 1, 3, 5 and 6.5 are illustrated in Fig. S1, respectively. The results demonstrate that all the three different HH@PE samples can achieve pH response release of 8-HQ. However, as can be seen from Fig. S1a, when only two polyelectrolyte layers are wrapped, the release concentration of 8-HQ is still high even at pH=6.5, reaching 4.61 mg/L at 3 h. Whereas when six polyelectrolyte layers were wrapped (as seen in Fig. S1c), the release concentration of 8-HQ is very low under the pH=1, which is only 7.55 mg/L at 3 h. This suggests that too few polyelectrolyte layers are detrimental to the loading of 8-HQ under neutral environments, while too many polyelectrolyte layers are detrimental to the release of 8-HQ under acidic environments. For HH@PE-4 (Fig. S1b), it can be seen that the greatest release concentration of 8-HQ reaches 11.88 mg/L in the solution with pH=1 after 3h, while the corresponding data decrease sequentially to 9.67 mg/L, 4.60 mg/L and 3.00 mg/L in the solution with pH=3, 5 and 6.5, respectively. Consequently, it can be found that the HH@PE-4 can realize the best pH responsive release property of 8-HQ. Therefore, HH@PE-4 was selected as the nanofiller in the preparation of the composite passivator.

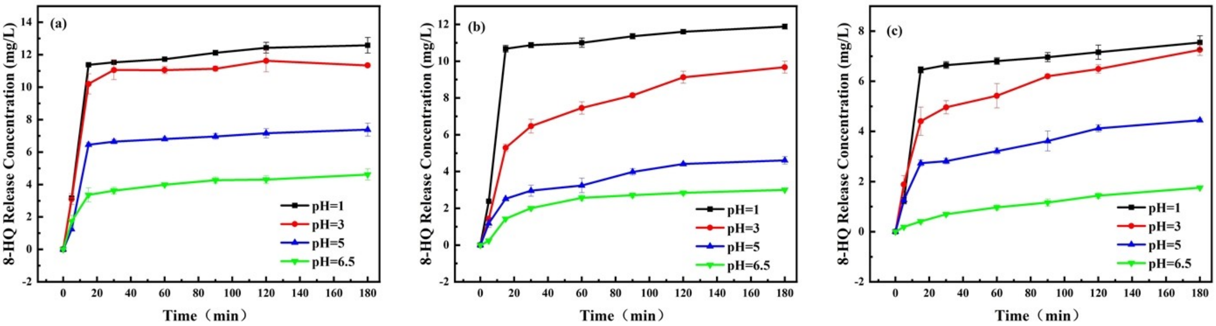

Fig. S1. Release concentration of 8-HQ in different pH solutions with (a) 2 polyelectrolyte layers, (b) 4 polyelectrolyte layers and (c) 6 polyelectrolyte layers.

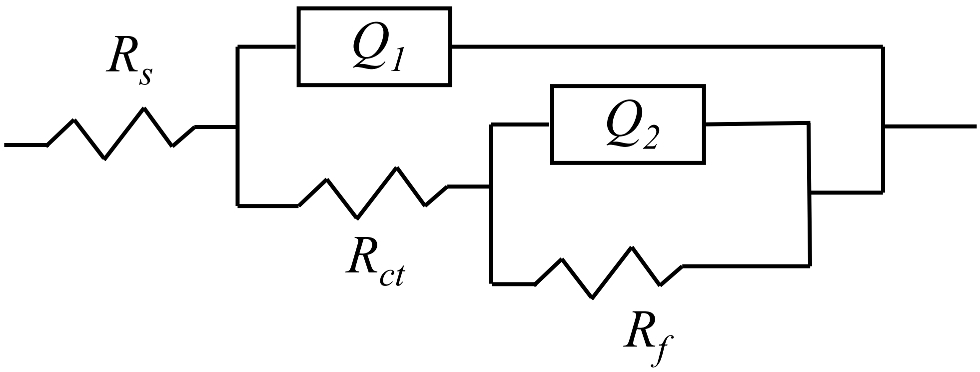

Fig. S2. Equivalent electrical circuit model for fitting the EIS data obtained from different pyrite electrodes.

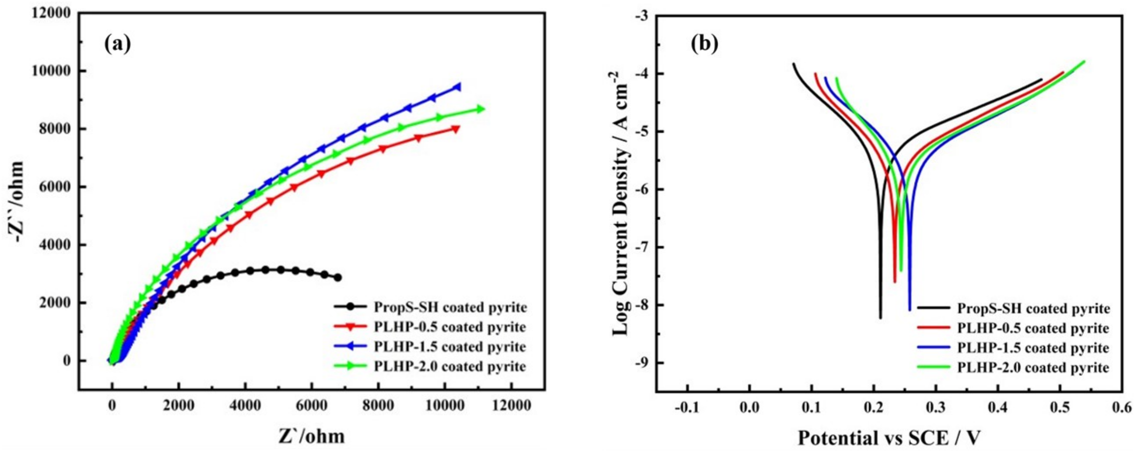

Fig. S3. (a) Nyquist plots of EIS data obtained from PropS-SH, PLHP-0.5, PLHP-1.0, PLHP-1.5 coated pyrite electrodes in 0.2 M Na_2SO_4 solution with pH 2.0; (b) Tafel polarization curves of PropS-SH, PLHP-0.5, PLHP-1.0, PLHP-1.5 coated pyrite electrodes in 0.2 M Na_2SO_4 solution with pH 2.0.

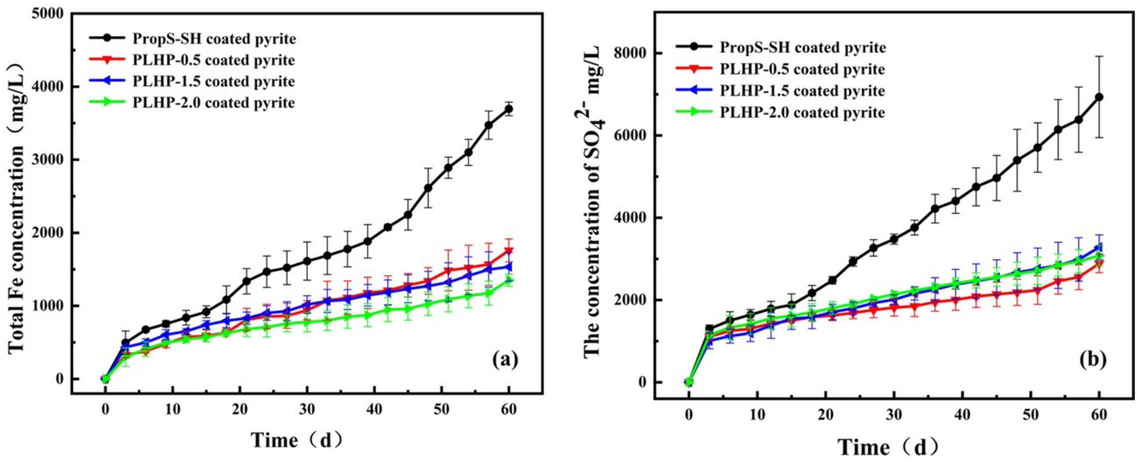

Fig. S4. Concentrations of (a) Total Fe and (b) SO_4^{2-} released as a function of time for PropS-SH, PLHP-0.5, PLHP-1.0, PLHP-1.5 coated pyrite samples in the chemical leaching solutions.

Table S1. Results of scratch adhesion tests for different coatings

Sample	**Critical load(N)**
	Lc
Raw pyrite	**1.98**
PL coated pyrite	**3.71**
PLHP-1.0 coated pyrite	**6.68**