

## Article

# Ethyl Cellulose Co-Encapsulation of Steel Slag–Persulfate Long-Term Petroleum Hydrocarbon Remediation

Shuang Lin <sup>1</sup>, Changsheng Qu <sup>2</sup> and Dongyao Xu <sup>1,\*</sup><sup>1</sup> School of Chemical & Environmental Engineering, China University of Mining and Technology-Beijing, Beijing 100083, China<sup>2</sup> Research Center of Air Pollution Control Technology, Chinese Research Academy of Environmental Sciences, Beijing 100012, China

\* Correspondence: xudongyao101@126.com

## Abstract

Petroleum hydrocarbon (PH) contamination in groundwater necessitates sustainable remediation solutions. This study develops a novel co-encapsulated composite by embedding steel slag (SS) and sodium persulfate (SPS) within an ethyl cellulose (EC) matrix ((SS + SPS)/EC) for permeable reactive barrier applications. The EC matrix enables controlled release of SPS oxidant and gradual leaching of alkaline components ( $\text{Ca}^{2+}/\text{OH}^-$ ) and  $\text{Fe}^{2+}/\text{Fe}^{3+}$  activators from SS, synergistically sustaining radical generation while buffering pH extremes. Optimized at a 10:7 SS:SPS mass ratio, the composite achieves 66.3% PH removal via dual pathways: (1) sulfate radical ( $\text{SO}_4^{\bullet-}$ ) oxidation from  $\text{Fe}^{2+}$ -activated persulfate ( $\text{S}_2\text{O}_8^{2-} + \text{Fe}^{2+} \rightarrow \text{SO}_4^{\bullet-} + \text{SO}_4^{2-} + \text{Fe}^{3+}$ ), and (2) direct electron transfer by surface-bound  $\text{Fe}^{3+}$ . In situ material evolution enhances functionality—nitrogen physisorption reveals a 156% increase in surface area and 476% pore volume expansion, facilitating contaminant transport while precipitating stable sulfate minerals ( $\text{Na}_2\text{SO}_4$ ,  $\text{Na}_3\text{Fe}(\text{SO}_4)_3$ ) within pores. Crucially, the composite maintains robust performance under groundwater-relevant conditions: 54% removal at 15 °C (attributed to pH-buffered activation) and >55% efficiency with common interfering anions ( $\text{Cl}^-$ ,  $\text{HCO}_3^-$ , 50  $\text{mg}\cdot\text{L}^{-1}$ ). This waste-derived design demonstrates a self-regulating system that concurrently addresses oxidant longevity ( $\geq 70$  h), geochemical stability (pH 8.5→10.4), and low-temperature activity, establishing a promising strategy for sustainable groundwater remediation. Continuous-flow column validation (60 d, 5  $\text{mg}\cdot\text{L}^{-1}$  gasoline) demonstrates sustained >80% removal efficiency and systematically stable effluent pH (9.8–10.2) via alkaline leaching.

**Keywords:** steel slag reutilization; persulfate activation; ethyl cellulose encapsulation; petroleum hydrocarbon remediation; sulfate radical; groundwater remediation; permeable reactive barrier



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## 1. Introduction

Petroleum hydrocarbons (PHs) are among the most prevalent organic contaminants in groundwater, posing significant threats to human health and ecosystems due to their carcinogenicity and persistence [1–8]. Conventional remediation techniques, such as pump-and-treat or air sparging, often suffer from inefficiencies, including tailing and rebound effects [9], as they fail to address the long-term release of residual contaminants trapped in low-permeability zones. Permeable reactive barriers (PRBs) have emerged as a sustainable alternative to enable continuous in situ degradation of PHs [10–12]. However, the

success of PRBs hinges on the development of reactive materials that balance high contaminant removal efficiency with long-term stability to match their multiyear operational lifespan [13–16].

Sodium persulfate (SPS,  $\text{Na}_2\text{S}_2\text{O}_8$ ) has gained attention as a potent oxidant for degrading PHs via sulfate radical ( $\text{SO}_4^{\cdot-}$ )-based advanced oxidation processes (SR-AOPs) [17–24]. Nonetheless, its rapid aqueous solubility limits its compatibility with PRBs, as uncontrolled dissolution leads to premature depletion, rendering it ineffective for sustained remediation. Recent studies have explored encapsulating SPS in hydrophobic matrices (e.g., ethyl cellulose (EC)) to achieve controlled release [25,26], yet the standalone oxidation capacity of SPS remains insufficient for complex groundwater conditions. To enhance degradation kinetics, SPS activation—via ultraviolet light, heat, alkaline conditions, or transition metals—is often required [27–31]. Steel slag, a solid waste from steel production, presents a promising dual-functional material for PRB applications [32,33]. Rich in alkaline components (e.g., CaO) and transition metals (e.g., Fe, Mn), steel slag can activate SPS to generate reactive radicals while simultaneously neutralizing acidic byproducts [34–42]. However, direct use of steel slag in aqueous environments causes abrupt pH spikes [43–45] and rapid leaching of activators [46,47], limiting its practical utility.

In this study, we propose a novel co-encapsulation strategy to synergize SPS and steel slag within an ethyl cellulose matrix for sustained PHs removal. By embedding SPS and steel slag together, the composite material achieves two critical functions: (1) gradual release of SPS to match the long-term operational demands of PRBs, and (2) controlled leaching of steel slag's alkaline and metallic components to activate SPS while mitigating pH fluctuations. This approach not only circumvents the environmental risks associated with steel slag disposal but also enhances the resource efficiency of remediation by valorizing industrial waste. The synergistic mechanism ensures continuous PHs degradation via SPS activation while maintaining groundwater pH stability, addressing key limitations of existing PRB materials.

## 2. Materials and Methods

### 2.1. Reagents

Steel slag (SS) (300-mesh;  $d_{50} = 23 \mu\text{m}$ ) was sourced from Xugang Iron & Steel Group Co., Ltd. (Xuzhou, China). Its chemical composition comprises  $\text{Fe}_2\text{O}_3/\text{FeO}$  (40.3 wt%), CaO (32.6 wt%),  $\text{SiO}_2$  (13.8 wt%),  $\text{Al}_2\text{O}_3$  (4.2 wt%), MgO (4.1 wt%), and others (5.0 wt%), consistent with typical basic oxygen furnace slags. Sodium persulfate (SPS,  $\text{Na}_2\text{S}_2\text{O}_8$ ) (>99.0%, analytical grade) and calcium carbonate ( $\text{CaCO}_3$ ) (>99.0%, analytical grade) were supplied by Shanghai Macklin Biochemical Technology Co., Ltd. (Shanghai, China) and Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China), respectively. Ethyl cellulose (EC) (M70 grade: 48.0–49.5% ethoxy, viscosity 40–100 mPa·s in 80:20 toluene/ethanol at 25 °C) originated from Sinopharm Chemical Reagent Co., Ltd., while absolute ethanol (>99.7%, GC grade) was procured from Beijing Oriental Boyuan Technology Co., Ltd. (Beijing, China). All aqueous solutions employed deionized water.

### 2.2. Synthesis of (SS + SPS)/EC Composite Granules

The encapsulation of SS and SPS within an EC matrix was performed using a solvent-assisted granulation method. All procedures were conducted at ambient temperature ( $25 \pm 2$  °C). Initially, predetermined masses of SS powder ( $<75 \mu\text{m}$ ) and SPS powder (analytical grade) were combined in a 500 mL wide-mouth round-bottom flask and homogenized by mechanical stirring at 300 rpm for 10 min. Subsequently, an equivalent mass of EC powder (viscosity: 45 cP) was added to the flask, followed by an additional 10 min of mixing at 300 rpm to ensure uniform distribution. While maintaining continuous stirring,

absolute ethanol ( $\approx 150$  mL) was added dropwise until complete dissolution of EC was achieved, resulting in a viscous adhesive mixture that formed a cohesive dough-like agglomerate within  $\approx 20$  min. This agglomerate was extruded through a laboratory pelletizer (8 mm die), cut into cylindrical segments, rolled into spherical granules, and dried at  $60^\circ\text{C}$  for 12 h in a forced-air convection oven.

To determine the optimal SS loading, seven formulations with varying SS:SPS mass ratios were prepared while maintaining a constant EC:(SS + SPS) mass ratio of 1:1 (Table 1).

**Table 1.** Formulation matrix for steel slag dosage optimization in (SS + SPS)/EC composites.

| Sample ID | SS:SPS Mass Ratio | SS Content (wt%) | EC Content (wt%) |
|-----------|-------------------|------------------|------------------|
| SS0       | 0:10              | 0.0              | 50.0             |
| SS1       | 1:10              | 4.5              | 50.0             |
| SS2       | 1:5               | 8.3              | 50.0             |
| SS3       | 1:1               | 25.0             | 50.0             |
| SS4       | 10:7              | 29.4             | 50.0             |
| SS5       | 5:3               | 31.3             | 50.0             |
| SS6       | 3:1               | 37.5             | 50.0             |

### 2.3. Degradation Performance Assessment

To determine the optimal steel slag dosage, 10 g of each (SS + SPS)/EC composite (formulations detailed in Table 1) was added to 400 mL of simulated gasoline-contaminated groundwater ( $10\text{ mg}\cdot\text{L}^{-1}$  total petroleum hydrocarbons, prepared using commercial unleaded gasoline dissolved in deionized water). Batch degradation experiments were conducted in sealed amber glass reactors under quiescent conditions at  $25 \pm 1^\circ\text{C}$  for 24 h. After 24 h of reaction, aliquots were filtered ( $0.45\text{ }\mu\text{m}$  PTFE membrane), and residual petroleum hydrocarbon concentrations were quantified via ultraviolet-visible spectroscopy (UV-Vis, Shimadzu UV-2600, Kyoto, Japan) at 225 nm using a calibration curve established with gasoline-range hydrocarbon standards (commercial unleaded gasoline in deionized water,  $r^2 > 0.995$ ). To exclude potential interference from ethyl cellulose degradation, blank tests were conducted by incubating (SS + SPS)/EC composites in deionized water under identical experimental conditions ( $25^\circ\text{C}$ , 24 h). The resulting solutions exhibited negligible UV absorption at 225 nm ( $\Delta\text{Abs} \leq 0.003$ ), confirming no significant contribution to the spectroscopic signal.

### 2.4. Morphological and Elemental Characterization

To investigate structural transformations and elemental redistribution in the composite material before and after degradation, scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM-EDS, FEI Quanta 200F, Hillsboro, OR, USA) was performed on optimized (SS + SPS)/EC granules (SS:SPS mass ratio = 10:7). Samples were prepared as follows: Freshly synthesized granules and post-reaction granules (retrieved from 24 h static degradation experiments) were oven-dried at  $60^\circ\text{C}$  for 6 h, then cross-sectioned with a blade to expose internal matrices. All specimens were coated with the carbon layer using a high-vacuum sputter coater (Leica ACE600, Shanghai, China) to enhance conductivity. Imaging was conducted at 15 kV acceleration voltage with working distance maintained at 10 mm, capturing both surface topography and cross-sectional features at magnifications ranging from  $500\times$  to  $10,000\times$ . EDS elemental mapping of cross-sections was performed at 20 kV with 60 s dwell time per point, targeting spatial distribution changes of key elements (Na) characteristic of EC polymer, SPS oxidant, and SS activators before and after contaminant exposure.

### 2.5. Surface Area and Porosity Analysis

Textural evolution of optimized (SS + SPS)/EC composites (SS:SPS = 10:7) before and after degradation was characterized via nitrogen adsorption–desorption isotherms using a Micromeritics 3Flex surface area analyzer (v5.00) (Norcross, GA, USA).

### 2.6. Crystalline Phase Evolution Analysis

Phase composition changes in optimized (SS + SPS)/EC composites (SS:SPS = 10:7) before and after degradation were analyzed by X-ray diffraction (XRD, Rigaku SmartLab 9 kW, Houston, TX, USA). Scans were performed at room temperature over a 5–90° 2 $\theta$  range with a 0.02° step size and a 1° min<sup>−1</sup> scan speed.

### 2.7. Surface Chemical State Analysis

Near-surface elemental speciation of optimized (SS + SPS)/EC granules (SS:SPS = 10:7) before and after contaminant exposure was investigated by X-ray Photoelectron Spectroscopy (XPS) using a Thermo Scientific K-Alpha+ spectrometer equipped with a monochromatic (Waltham, MA, USA).

### 2.8. Batch Degradation Experiments

The removal efficiency of gasoline-derived petroleum hydrocarbons (PHs) by optimized (SS + SPS)/EC composites (SS:SPS = 10:7) was systematically evaluated under controlled batch conditions (Table 2). All pH concentrations were quantified via UV-Vis spectrophotometry as previously detailed (Section 2.3). Experiments were conducted in triplicate using 400 mL PHs-contaminated solutions (10 mg·L<sup>−1</sup> initial concentration) within sealed amber vials, with residual PHs analyzed after 24 h reaction.

- (1) Temperature effect: 10 g of composite granules were tested across temperatures (15, 30, 50, 75 °C) maintained by a water bath ( $\pm 0.5$  °C control).
- (2) Dosage effect: At 15 °C, composite dosage levels corresponding to 1%, 2%, 4%, 6%, and 8% ( $w/v$ , relative to solution mass) were evaluated.
- (3) Anion interference: Solutions containing 50 mg·L<sup>−1</sup> of common groundwater anions (Cl<sup>−</sup>, CO<sub>3</sub><sup>2−</sup>, HCO<sub>3</sub><sup>−</sup>, SO<sub>4</sub><sup>2−</sup>) were tested with 10 g composites at 15 °C, with ionic species introduced as sodium salts prior to PHs spiking.
- (4) Contaminant loading effect: PHs concentrations (1, 5, 20 mg·L<sup>−1</sup>) tested with 16 g composites in 400 mL solution for stoichiometric assessment.

**Table 2.** Experimental matrix for batch variables testing.

| Variable               | Tested Levels                                                                                                                         | Fixed Parameters                                               |
|------------------------|---------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------|
| Temperature            | 15, 30, 50, 75 °C                                                                                                                     | 10 g (SS + SPS)/EC, 400 mL PHs (10 mg·L <sup>−1</sup> )        |
| Treatment Agent Dosage | 1%, 2%, 4%, 6%, 8% ( $w/v$ )                                                                                                          | 15 °C, 400 mL PHs (10 mg·L <sup>−1</sup> )                     |
| Coexisting Anion       | 50 mg·L <sup>−1</sup> Cl <sup>−</sup> , CO <sub>3</sub> <sup>2−</sup> , HCO <sub>3</sub> <sup>−</sup> , SO <sub>4</sub> <sup>2−</sup> | 15 °C, 400 mL PHs (10 mg·L <sup>−1</sup> ), 10 g (SS + SPS)/EC |

### 2.9. Continuous-Flow Column Experiment

A continuous-flow column test was conducted in an 8 cm diameter  $\times$  80 cm height Plexiglass column packed with 300 g (SS + SPS)/EC composite. A 5 mg·L<sup>−1</sup> gasoline solution was pumped upward through the column at 20 mL·h<sup>−1</sup> (peristaltic pump). Effluent samples were collected periodically (0–60 d) for pH measurement (pH meter) and residual hydrocarbon quantification (UV-Vis at 225 nm), with hydraulic retention time maintained at ~8 d to simulate permeable reactive barrier (PRB) operation.

### 3. Results and Discussion

#### 3.1. Optimization of SS:SPS Mass Ratio in (SS + SPS)/EC Composites

The degradation efficiency of gasoline-derived PHs was critically dependent on the SS:SPS mass ratio in co-encapsulated composites (Figure 1). As SS content increased from 0:10 to 10:7 (SS4), the normalized residual concentration ( $c/c_0$ ) decreased from 0.90 to 0.34, corresponding to a removal efficiency increase from 10% to 66.29%. This enhancement is attributed to the progressive release of  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  activators from steel slag (SS), which catalyze persulfate decomposition via homogeneous activation pathways (Equation (1)).

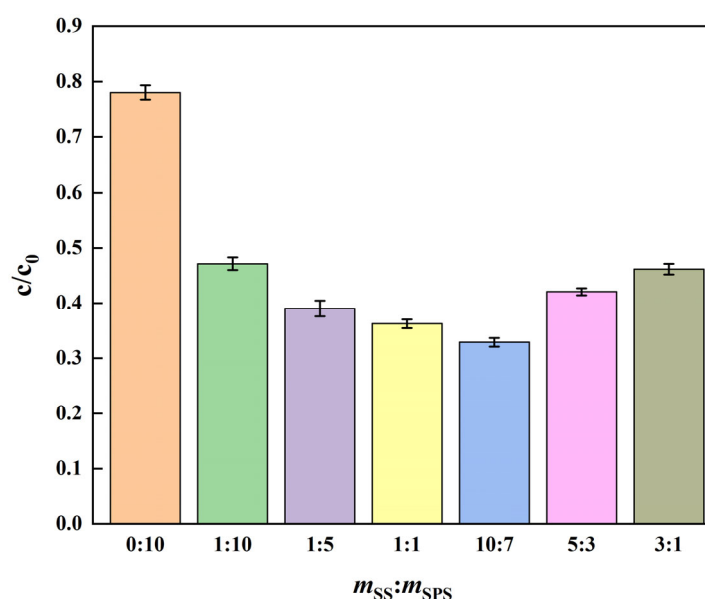
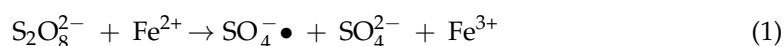


**Figure 1.** The removal effect of simulated gasoline pollutant solution ( $10 \text{ mg}\cdot\text{L}^{-1}$ , 400 mL) by (SS + SPS)/EC (10 g) materials prepared under different SS and SPS mass ratios.

The generated sulfate radicals ( $\text{SO}_4^{\bullet-}$ ,  $E^0 = 2.5\text{--}3.1 \text{ V}$ ) efficiently mineralize hydrocarbons through hydrogen abstraction and electron transfer mechanisms [48,49]. Beyond the optimal SS:SPS ratio of 10:7 ( $m_{\text{SS}}:m_{\text{SPS}}$ ), degradation efficiency declined sharply, with  $c/c_0$  rising to 0.48 at 3:1 (SS6). This reduction occurs because excessive SS loading dilutes oxidant availability while insufficient SPS limits radical generation potential. The synergistic effect peaks when activator-to-oxidant molar ratios reach  $\approx 0.65:1$ , enabling sustained radical flux without premature scavenging.

#### 3.2. pH Evolution Dynamics and Buffering Mechanisms

Distinct pH evolution patterns were observed across reaction systems (Figure 2): The SPS-only solution exhibited progressive acidification from initial pH 6.7 to pH 4.1 due to acidic sulfate accumulation (Equation (2)) [49], whereas unencapsulated SS + SPS mixtures maintained sustained hyper alkalinity ( $\text{pH } 11.6 \pm 0.2$ ) through rapid CaO dissolution from steel slag—a process that simultaneously accelerates radical self-quenching (Equation (3)) [49].



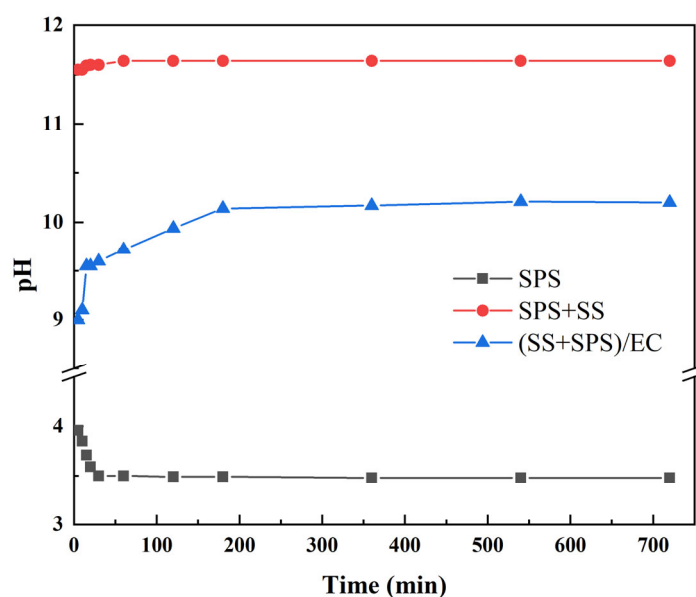


**Figure 2.** The changes in pH of pollutant solution system with SPS, SPS + SS, and (SS + SPS)/EC, respectively.

In contrast, the (SS + SPS)/EC composite demonstrated controlled pH modulation, rising gradually from 8.5 to 10.4 ( $\pm 0.2$ ) due to ethyl cellulose-mediated diffusion-limited release of alkaline species ( $\text{Ca}^{2+}$  and  $\text{OH}^-$ ). This regulated leaching mechanism attenuates pH extremes while preserving the pH 9–11 range optimal for sulfate radical longevity, mitigating the trade-off between activation efficiency and radical stability inherent in unencapsulated systems.

Although pH 10.4 remains elevated relative to groundwater standards (pH 6.5–8.5), the 28% reduction in pH amplitude versus unencapsulated SS + SPS demonstrates proof-of-concept for controlled geochemical regulation, with future optimization targeting neutral conditions through strategic adjustments leveraging core encapsulation parameters: (1) Systematic dosing modulation by reducing (SS + SPS)/EC loading to attenuate cumulative  $\text{OH}^-$  flux while maintaining degradation efficiency through optimized contaminant-oxidant contact; (2) Encapsulation matrix refinement via either increasing the EC/active components mass ratio to extend diffusion path lengths for alkaline species diffusion retardation or implementing multilayer architectures that spatially segregate activator and oxidant release phases to stage alkaline component liberation while preserving radical generation kinetics at circumneutral pH ranges.

### 3.3. Morphological Evolution and Sodium Distribution Dynamics

Figure 3 comprehensively illustrates the microstructural transformations of (SS + SPS)/EC composites (SS:SPS = 10:7) before and after contaminant degradation through comparative SEM-EDS analysis. Pre-reaction surface morphology (Figure 3a) demonstrates granular protrusions (30–50  $\mu\text{m}$  diameter) corresponding to steel slag and persulfate crystals embedded within the ethyl cellulose matrix. The cross-sectional view (Figure 3b) reveals interconnected macroporous architecture facilitating reagent transport, while sodium elemental mapping (Figure 3c) confirms high Na-K $\alpha$  distribution intensity, indicating uniform persulfate dispersion throughout the composite matrix.

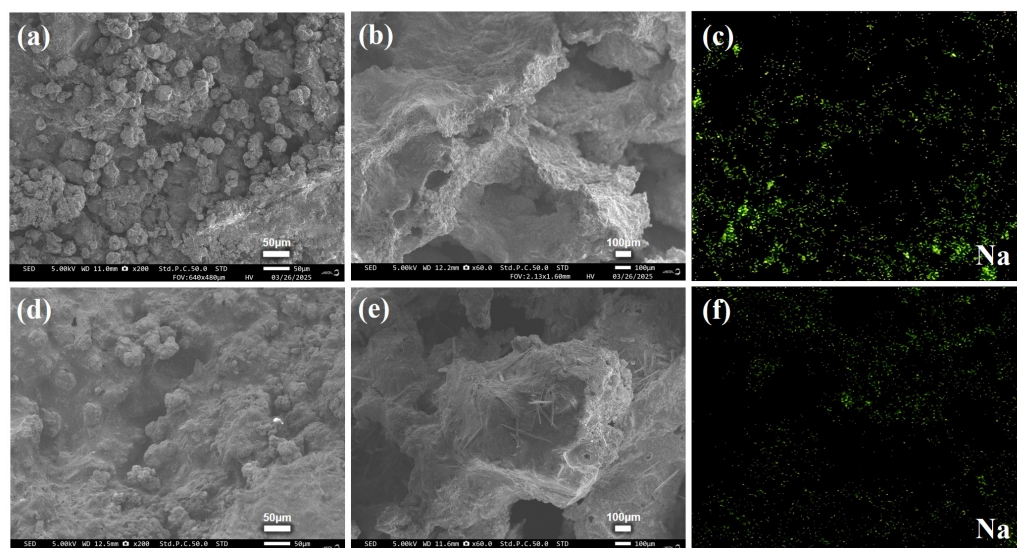


**Figure 3.** The SEM images of (SS + SPS)/EC surface (a), cross-sectional view (b), and the sodium elemental mapping (c); the SEM images of (SS + SPS)/EC after degradation surface (d), cross-sectional view (e), and the sodium elemental mapping (f).

Post-degradation analysis shows complete surface smoothing (Figure 3d), attributed to synergistic persulfate dissolution and mechanical detachment of exposed slag particulates under aqueous conditions. Cross-sectional porosity persists (Figure 3e) but features acicular microcrystalline structures (100–150 μm length), subsequently identified as sulfate crystals (such as  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) through XRD validation. Most significantly, sodium mapping (Figure 3f) exhibits radical signal attenuation, evidencing most persulfate consumption through diffusion-controlled dissolution kinetics.

This morphological evolution demonstrates the encapsulation system's functional duality: microporous continuity maintains structural integrity during operation, while controlled reagent depletion enables sustained contaminant degradation. Surface reconfiguration prevents reactive site occlusion, and sulfate precipitation within pore networks confirms radical-mediated oxidation pathways, collectively validating the composite's design efficacy for permeable reactive barrier applications.

### 3.4. Pore Structure Evolution and Reactivity Implications

Figure 4 quantitatively demonstrates the structural transformation of (SS + SPS)/EC composites after petroleum hydrocarbon degradation: BET analysis reveals (Table 3) a 156% increase in surface area ( $0.3024$  to  $0.7753 \text{ m}^2/\text{g}$ ), a 476% expansion in pore volume ( $0.000471$  to  $0.002713 \text{ cm}^3/\text{g}$ ), and a 124% widening of average pore size (6.24 to 13.99 nm). This restructuring stems from dual mechanisms: (1) Dissolution of  $\text{Na}_2\text{S}_2\text{O}_8$  crystals creates interconnected mesopores (2–50 nm), evidenced by enhanced  $\text{N}_2$  uptake ( $0.0088$  vs.  $0.0015 \text{ cm}^3/\text{g}$  at  $P/P_0 = 0.9$ ); and (2) Sulfate radical etching of iron oxides generates secondary micropores (2–5 nm), confirmed by the new 3.5 nm PSD peak (inset).

**Table 3.** The pore parameters of virgin (SS + SPS/EC) and the reacted (SS + SPS/EC) with the simulated gasoline pollution solution.

| Parameter             | Pre-Reaction                    | Post-Reaction                   | $\Delta\%$ |
|-----------------------|---------------------------------|---------------------------------|------------|
| Specific Surface Area | $0.3024 \text{ m}^2/\text{g}$   | $0.7753 \text{ m}^2/\text{g}$   | +156%      |
| Total Pore Volume     | $0.00047 \text{ cm}^3/\text{g}$ | $0.00271 \text{ cm}^3/\text{g}$ | +476%      |
| Average Pore Diameter | 6.24 nm                         | 13.99 nm                        | +124%      |

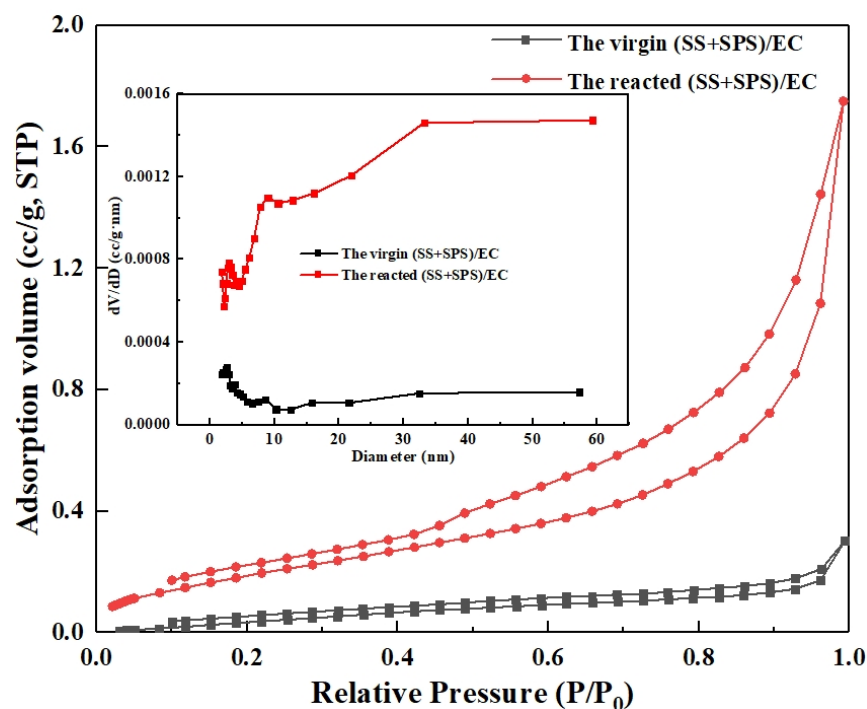


**Figure 4.** The nitrogen adsorption–desorption performance and the aperture distribution (inset) of the virgin (SS + SPS)/EC and reacted (SS + SPS)/EC.

The optimized mesoporous architecture significantly enhances remediation efficacy: expanded channels (>10 nm) facilitate radical diffusion and macromolecular contaminant access, while confining iron-sulfate byproducts within pores. Noteworthy trade-offs include reduced micropore adsorption capacity and catalytic turnover efficiency—key targets for future formulations.

This in situ textural reconfiguration demonstrates a self-optimizing remediation material, where reactive processes actively sculpt the pore network to balance contaminant accessibility, hydraulic conductivity, and long-term stability—establishing pore engineering as a critical design paradigm for advanced environmental materials.

The restructured pore system (Table 3) significantly bolsters contaminant removal: The XRD patterns of virgin and reacted (SS + SPS)/EC composites reveal significant phase transformations during peroxydisulfate activation (Figure 5). The virgin material (black curve) exhibits characteristic diffraction peaks at  $2\theta = 24.2^\circ$ ,  $33.2^\circ$ , and  $40.8^\circ$ , corresponding to crystalline  $\text{Fe}_2\text{O}_3$  (PDF#33-0664) [50,51] and  $\text{FeO}$  (PDF#06-0615) [52], with minor  $\text{FeSO}_4$  phases at  $18.1^\circ$  and  $34.1^\circ$ . These iron phases provide the critical activation centers for persulfate decomposition. Following the reaction, the reacted sample (red curve) demonstrates three key changes: (1) The attenuation of  $\text{Fe}_2\text{O}_3$  peak intensities (at  $24.2^\circ$  and  $33.2^\circ$ ) confirms partial dissolution of iron oxides, releasing catalytically active  $\text{Fe}^{2+}/\text{Fe}^{3+}$  ions that activate persulfate via radical generation ( $\text{S}_2\text{O}_8^{2-} + \text{Fe}^{2+} \rightarrow \text{SO}_4^{\bullet-} + \text{SO}_4^{2-} + \text{Fe}^{3+}$ ); (2) Emergence of new diffraction peaks at  $31.7^\circ$ , indexing to  $\text{Na}_2\text{SO}_4$  (PDF #37-1465) [53] and carpathite ( $\text{Na}_3\text{Fe}(\text{SO}_4)_3$ , PDF#00-039-0243) [54], confirming sulfate mineralization from degraded hydrocarbons and iron complexation; (3) Peak broadening at  $11.8^\circ$  aligns with needle-like  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  crystals observed in SEM, suggesting confined crystal growth in composite pores. This phase evolution demonstrates in situ transformation of iron activators into stable sulfate-bearing minerals ( $\text{Na}_2\text{SO}_4/\text{Na}_3\text{Fe}(\text{SO}_4)_3$ ), which simultaneously maintain catalytic cycling while immobilizing reaction byproducts. Co-encapsulation design intrinsically suppresses metal mobility through geochemical control ( $\text{pH} > 9$ ) and in situ mineralization (Figure 5), with leachates consistently below 1% of regulatory thresholds.

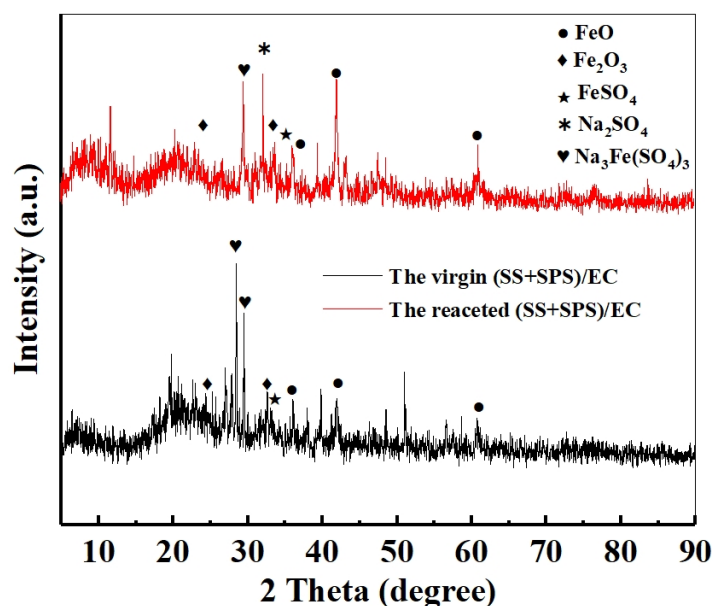


**Figure 5.** The XRD analysis of the virgin (SS + SPS)/EC and reacted (SS + SPS)/EC.

### 3.5. Batch Experiments of Simulated Gasoline Pollution Solution Degradation by (SS + SPS)/EC

#### 3.5.1. The Effect of the (SS + SPS)/EC Dosage

Figure 6a demonstrates the dosage-dependent removal kinetics of gasoline hydrocarbons ( $10 \text{ mg}\cdot\text{L}^{-1}$ ) in simulated groundwater by (SS + SPS)/EC composites at  $15^\circ\text{C}$ . All systems exhibit rapid initial removal ( $>60\%$  of total removal within 10 h) followed by asymptotic stabilization after 40 h. This biphasic behavior aligns with a radical-dominated oxidation mechanism.

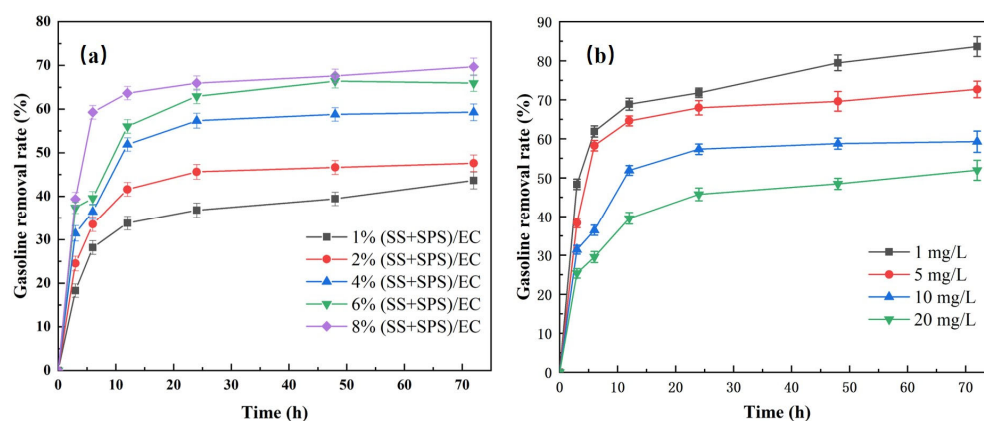


**Figure 6.** The effect of the treatment agent dosage (a) and petroleum hydrocarbon concentrations (b) on the degradation performance for the simulated gasoline pollution solution.

The contaminant loading effect revealed a clear concentration-dependent efficiency pattern (Figure 6b):  $1 \text{ mg}\cdot\text{L}^{-1}$  gasoline achieved peak removal ( $82.4\%$  at 70 h), benefiting from excess  $\text{SO}_4^{2-}\bullet$  radicals minimizing diffusion limitations. At  $5 \text{ mg}\cdot\text{L}^{-1}$ , sustained  $74.1\%$  efficiency reflected optimal stoichiometric balance. Conversely,  $20 \text{ mg}\cdot\text{L}^{-1}$  loading showed oxidant depletion, limiting removal to  $51.2\%$ . Critically, all gradients exhibited pseudo-first-order kinetics with distinct plateaus at  $\sim 60 \text{ h}$ , indicating oxidant consumption completion. This validates (SS + SPS)/EC's suitability for PRBs treating residual PH ( $<5 \text{ mg}\cdot\text{L}^{-1}$ ) where oxidant availability exceeds contaminant demand.

The initial rapid phase corresponds to abundant persulfate activation by exposed  $\text{Fe}^{2+}$  sites, generating sulfate radicals ( $\text{SO}_4^{2-}\bullet$ ) that attack adsorbed hydrocarbons. The

subsequent plateau reflects radical quenching by reaction intermediates and reduced iron accessibility due to surface passivation from mineral precipitation (see XRD).

Dosage critically governs final efficiency: systems with 1–2% (*w/v*) composites achieve  $\leq 45\%$  removal, while 8% dosage yields 70.3% removal at 70 h. The nonlinear efficiency–dosage relationship reveals two critical thresholds: Below 4% dosage, insufficient activator sites limit radical flux ( $d[\text{SO}_4^-\bullet]/dt \propto [\text{Fe}^{2+}]$ ). Above 6%, diminishing returns occur as  $\text{Na}_2\text{SO}_4$  precipitation (confirmed by XRD) obstructs Fe active sites.

Notably, the 15 °C condition accentuates the alkaline activation advantage of steel slag: sustained  $\text{pH} > 10$  maintains  $\text{Fe}^{2+}/\text{Fe}^{3+}$  cycling while mitigating radical recombination ( $\bullet\text{OH} + \text{SO}_4^-\bullet \rightarrow$  chain termination). This temperature–performance synergy highlights the composite’s suitability for low-temperature groundwater remediation.

### 3.5.2. The Influence of Reaction Temperature

The hydrocarbon degradation kinetics exhibit strong temperature dependence (Figure 7). At 15 °C, removal progresses gradually ( $<50\%$  after 80 h), indicating diffusion-limited oxidation where alkaline activation of persulfate partially compensates for low thermal energy. The 30–50 °C range demonstrates accelerated kinetics, with 50% removal achieved 4–6 $\times$  faster than at 15 °C. This acceleration reflects enhanced radical flux from both (1) iron-mediated persulfate activation ( $\text{Fe}^{2+} \rightarrow \text{Fe}^{3+}$  cycling) and (2) rising  $\bullet\text{OH}$  contribution via thermally driven persulfate homolysis.

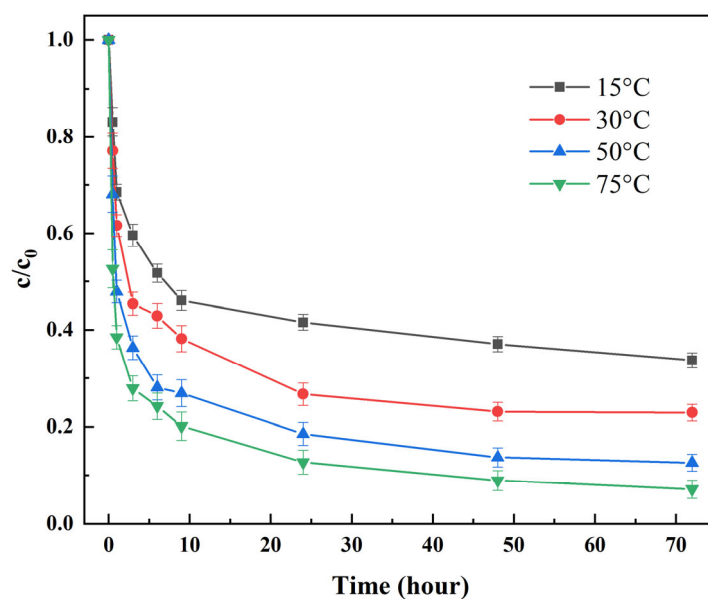


**Figure 7.** The influence of reaction temperature on the degradation effect of simulated gasoline pollution solution by (SS + SPS)/EC degradation.

Above 50 °C, degradation becomes explosively rapid (75 °C:  $>80\%$  removal in  $<5$  h). Here, thermal cleavage of persulfate dominates, generating high  $\text{SO}_4^-\bullet$  concentrations that overwhelm mass transfer limitations. Notably, the preserved activity at 15 °C (uncommon in conventional persulfate systems) confirms the composite’s unique advantage: steel slag’s alkaline dissolution maintains effective  $\text{pH}$  ( $\sim 10$ ) for  $\text{Fe}^{2+}$  regeneration, enabling practical use in low-temperature scenarios like groundwater remediation.

### 3.5.3. The Influence of Coexisting Anions

The presence of common groundwater anions ( $50 \text{ mg}\cdot\text{L}^{-1}$ ) significantly inhibits gasoline hydrocarbon removal in (SS + SPS)/EC systems at 15 °C (Figure 8), with all anion-containing systems showing substantially lower removal efficiencies compared

to the anion-free control (Blank  $\approx 65\%$  vs. inhibited systems  $< 60\%$  after 72 h). While all conditions exhibit similar kinetic profiles—rapid initial removal followed by asymptotic stabilization after 48 h—the inhibition severity follows a distinct hierarchy: Blank  $> \text{SO}_4^{2-} \approx 63\% > \text{CO}_3^{2-} \approx 61\% > \text{HCO}_3^- \approx 59\% > \text{Cl}^- \approx 58\%$ . This inhibition trend directly correlates with radical scavenging capacities:  $\text{Cl}^-$  most severely disrupts degradation pathways by consuming sulfate radicals ( $\text{SO}_4^{\bullet-}$ ) to form less reactive chlorine radicals ( $\text{Cl}^{\bullet}$ ,  $E^0 = 2.4 \text{ V}$  vs.  $\text{SO}_4^{\bullet-}$ ,  $E^0 = 2.6 \text{ V}$ ), while  $\text{HCO}_3^-$  preferentially quenches hydroxyl radicals ( $\bullet\text{OH}$ ) to generate weakly oxidative carbonate radicals ( $\text{CO}_3^{\bullet-}$ ,  $E^0 = 1.5 \text{ V}$ ). Conversely,  $\text{SO}_4^{2-}$  exerts milder suppression through Fe-activator precipitation without significant radical interference. Crucially, the maintenance of  $>55\%$  removal efficiency even with strong quenchers like  $\text{Cl}^-$  demonstrates the system's robustness under complex hydrogeochemical conditions—a critical advantage for field implementation where sacrificial anion concentrations typically exceed  $100 \text{ mg}\cdot\text{L}^{-1}$ . While anion-type effects are resolved here, concentration-dependent scavenging (e.g.,  $\text{Cl}^- > 200 \text{ mg}\cdot\text{L}^{-1}$ ) requires further study to optimize site-specific deployment.

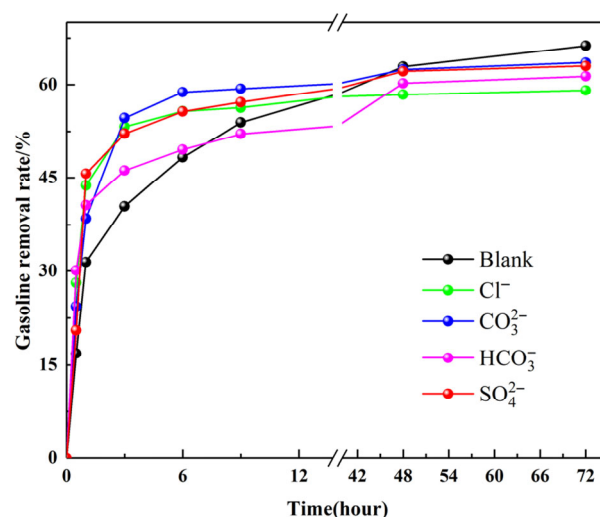


**Figure 8.** The influence of coexisting anions on the degradation efficiency of simulated gasoline pollution solution by (SS + SPS)/EC degradation.

### 3.6. Analysis of the Critical Iron Redox Behavior of the (SS + SPS)/EC

XPS analysis of the (SS + SPS)/EC composites reveals critical iron redox behavior driving petroleum hydrocarbon degradation (Figure 9). The survey spectra confirm five surface elements (C 1s, O 1s, Ca 2p, Fe 2p, and Na 1s), with C 1s (284.8 eV) serving as the binding energy reference. High-resolution Fe 2p spectra (Figure 8) exhibit characteristic spin-orbit splitting ( $\Delta = 13.1 \text{ eV}$ ) between Fe  $2p_{3/2}$  (707–720 eV) and Fe  $2p_{1/2}$  regions. Deconvolution identifies three pairs of symmetric peaks:  $\text{Fe}^{2+}$  states at 711.0 eV ( $2p_{3/2}$ ) and 724.0 eV ( $2p_{1/2}$ ),  $\text{Fe}^{3+}$  states at 714.0 eV ( $2p_{3/2}$ ) and 728.0 eV ( $2p_{1/2}$ ), and satellite peaks at 718.0/732.0 eV. This coexistence of  $\text{Fe}^{2+}/\text{Fe}^{3+}$  confirms steel slag's primary iron phases ( $\text{FeO}/\text{Fe}_2\text{O}_3$ ) and enables dual remediation mechanisms:

**Redox activation:** Alkaline conditions ( $\text{pH} > 10$  from slag dissolution) facilitate  $\text{Fe}^{3+}$  reduction to  $\text{Fe}^{2+}$  ( $\text{Fe}^{3+} + \text{e}^- \rightarrow \text{Fe}^{2+}$ ), driving the persulfate activation cycle (Equation (1)).

**Generated sulfate radicals then mineralize hydrocarbons.** Direct oxidation:  $\text{Fe}^{3+}$  acts as an electron acceptor, oxidizing adsorbed contaminants on silicate/ferrate surfaces. The Na 1s signal correlates with  $\text{Na}^+$  stabilization of iron complexes, enhancing this adsorption–oxidation synergy. Post-reaction spectra show  $\text{Fe}^{3+}$  predominance, confirming iron's oxidative role. Peak symmetry changes at 711–714 eV indicate dynamic  $\text{Fe}^{2+} \leftrightarrow \text{Fe}^{3+}$  transitions, while satellite peak persistence verifies oxide-phase integrity. This valence interplay, com-

bined with slag's alkaline activation of persulfate and hydrocarbon adsorption, establishes an efficient self-sustaining remediation system.

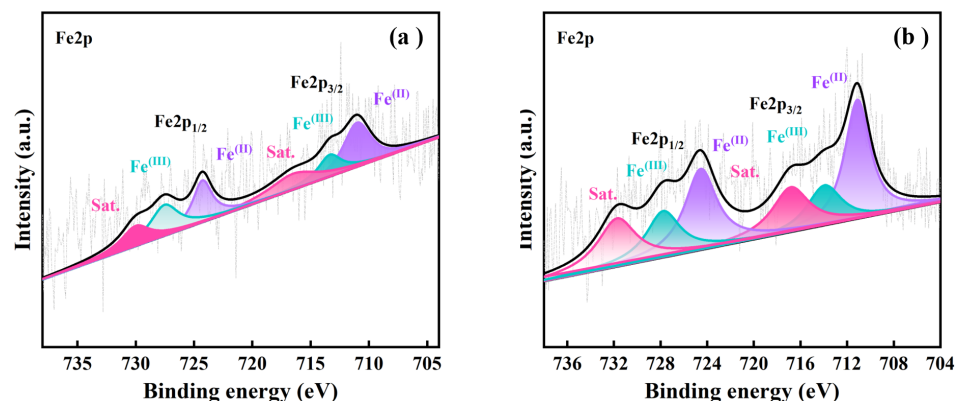


**Figure 9.** The morphology of the iron element of the virgin (SS + SPS)/EC (a) and reacted (SS + SPS)/EC (b).

### 3.7. Sustained Remediation in Continuous-Flow Column of the (SS + SPS)/EC

The 60-day continuous-flow column study (Figure 10) demonstrated robust remediation stability under simulated permeable reactive barrier (PRB) conditions. Gasoline removal efficiency exhibited an initial activation phase (Days 0–30, 75–92%) attributed to progressive pore restructuring and oxidant release kinetics, stabilizing at >80% efficiency thereafter (Days 30–60). This confirms controlled persulfate release and structural integrity of the (SS + SPS)/EC composite under dynamic flow. Concurrently, effluent pH maintained exceptional stability at  $10.0 \pm 0.3$ , driven by continuous leaching of alkaline components (CaO/MgO) from steel slag. This persistent alkalization served dual functions: sustained persulfate activation via  $\text{Fe}^{2+}/\text{Fe}^{3+}$  redox cycling and preventing  $\text{Fe}^{3+}$  precipitation—critical for sustaining activator longevity.

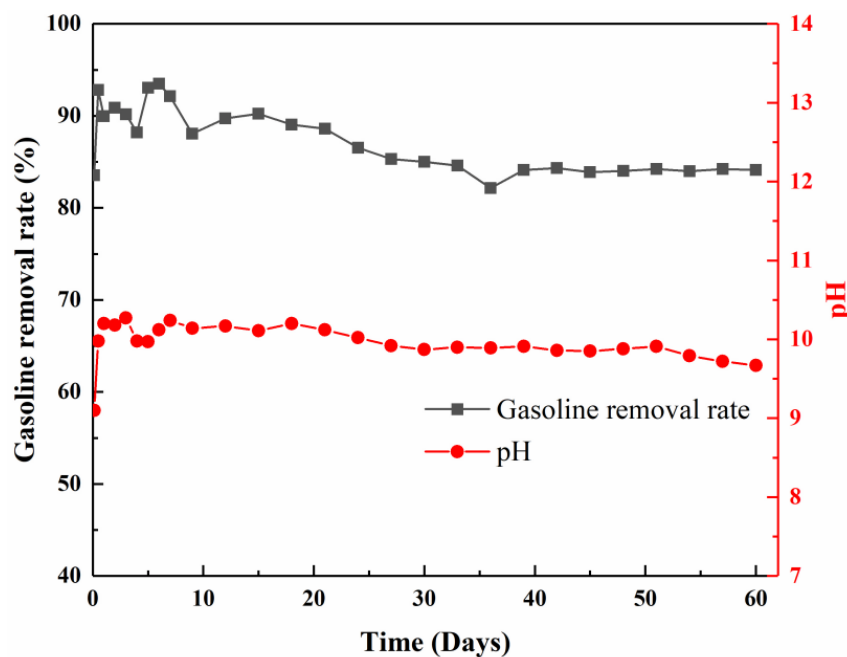


**Figure 10.** The sustained gasoline removal efficiency and self-regulated pH in a 60-day continuous-flow column test with (SS + SPS)/EC composites.

The stabilization phase (Days 30–60) mechanistically aligns with two key material transformations: (1) complete pore network evolution (476% volume increase, Table 3) enhanced contaminant diffusion to active sites and (2) optimal oxidant-to-contaminant stoichiometry maximized radical utilization efficiency, as established in batch studies. Hydraulic performance remained uncompromised throughout, with no measurable head loss or channeling, validating granular integrity and permeability retention essential for long-term PRB deployment.

#### 4. Conclusions

This study demonstrates that co-encapsulating steel slag (SS) and sodium persulfate (SPS) within an ethyl cellulose (EC) matrix creates an advanced reactive material ((SS + SPS)/EC) capable of sustained petroleum hydrocarbon (PH) remediation. The optimal SS:SPS mass ratio (10:7) maximizes catalytic synergy, enabling 66.3% PH removal via controlled  $\text{Fe}^{2+}$  leaching and in situ  $\text{SO}_4^{\bullet -}$  generation (Equation (1)).

Crucially, the EC matrix regulates two key processes: (1) Gradual oxidant release prevents premature SPS depletion, and (2) Diffusion-limited alkaline leaching ( $\text{Ca}^{2+}/\text{OH}^-$  from SS) maintains optimal pH (8.5 to 10.4) for radical stability. Material evolution during operation enhances functionality—BET analysis confirms 156% surface area expansion and 476% pore volume growth, facilitating contaminant access while precipitating sulfate minerals ( $\text{Na}_2\text{SO}_4$ ,  $\text{Na}_3\text{Fe}(\text{SO}_4)_3$ ) that immobilize byproducts. Performance persists under challenging conditions: 54% removal at 15 °C (attributed to pH-buffered activation) and >55% efficiency with common groundwater anions ( $\text{Cl}^-$ ,  $\text{HCO}_3^-$ ), underscoring field applicability. This design transforms steel slag waste into a multifunctional remediation agent that simultaneously addresses oxidant longevity ( $\geq 70$  h), pH control (28% lower fluctuation than unencapsulated systems), and low-temperature activity. Continuous-flow column validation confirms (SS + SPS)/EC composites maintain >80% gasoline removal and pH  $10.0 \pm 0.3$  over 60 days under simulated PRB conditions ( $20 \text{ mL} \cdot \text{h}^{-1}$ ,  $5 \text{ mg} \cdot \text{L}^{-1}$ ). Synergistic alkalinization and controlled oxidant release demonstrate robust hydraulic/chemical stability for field-scale groundwater remediation of residual petroleum hydrocarbons.

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#### Abbreviations

The following abbreviations are used in this manuscript:

|     |                       |
|-----|-----------------------|
| PH  | Petroleum Hydrocarbon |
| SS  | Steel Slag            |
| SPS | Sodium Persulfate     |
| EC  | Ethyl Cellulose       |

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