

Sulphate-Induced Corrosion of Mild Steel Rebar in Alkaline Media: Long-Term Kinetics, pH-Dependent Morphology, and Non-Passivating Behaviour

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Abstract

Sulphate-induced corrosion of mild steel reinforcement in alkaline media remains poorly characterised relative to chloride attack, despite the prevalence of sulphate-contaminated groundwater and industrial effluents in contact with reinforced concrete. This study investigated the long-term (six-month) gravimetric corrosion behaviour of commercial-grade mild steel (3 mm flat bar, 0.07% C) immersed in sodium sulphate solutions at three concentrations (200, 1000 and 2000 mg/L), combined with chloride additions (0, 50 and 500 mg/L), at pH 9 and pH 12, representative of carbonated and fresh concrete pore solutions, respectively. The objectives were to quantify sulphate- and chloride-driven corrosion kinetics, characterise the pH-dependent corrosion morphology, and determine whether the resulting corrosion products are passivating. Mass loss was measured at weekly and monthly intervals over six months, and corrosion morphology was examined macroscopically and by stereomicroscopy. Mass loss increased linearly with time at both pH values ($R^2 = 0.96\text{--}0.99$), confirming non-passivating corrosion kinetics, with a maximum mass loss of approximately 287 mg recorded after six months. Corrosion was consistently more severe at pH 12 than at pH 9 for equivalent solute concentrations. A pH-dependent shift in morphology was observed: uniform etch corrosion occurred at pH 9, whereas at pH 12, pronounced pitting and tuberculation were observed. Increasing sulphate concentration increased corrosion rates at both pH values, and a chloride addition of 500 mg/L caused a disproportionately sharp increase compared with lower additions. These findings indicate that sulphate attack in highly alkaline conditions can be more damaging to mild steel than chloride-induced corrosion, with direct implications for the durability design of reinforced concrete in sulphate-bearing environments. The work also supports the UN Sustainable Development Goals 3, 6, 9, 11, 12, and 13 by informing the development of more resilient, resource-efficient infrastructure.

Keywords

Sulphate Corrosion; Alkaline Media; Gravimetric Corrosion Testing; Corrosion Kinetics; Carbon Steel, Concrete pore solution, Tuberculation

1. Introduction

Metallic corrosion constitutes one of the most economically significant classes of material degradation encountered in civil, industrial, and infrastructure engineering. A landmark survey of the South African economy conducted between 1979 and 1983 estimated the annual cost of metallic corrosion in the mining, power generation, shipping, and

transportation sectors at R1 250 million, exceeding 4% of Gross National Product at the time (Slabbert, 1986). In the United States, metallic corrosion was estimated to cost approximately USD 300 billion annually at 1995 prices, equivalent to approximately 4.9% of GNP (Roberge, 2000). These figures underscore the critical importance of understanding, predicting, and mitigating corrosion in engineering systems.

Reinforced concrete is the world's most widely produced composite construction material, with annual production volumes exceeding that of steel by a factor of ten in tonnage (Roberge, 2000). While concrete exhibits high compressive strength, its tensile capacity is inherently limited, and the primary structural function of embedded mild steel reinforcement (rebar) is to augment tensile strength and improve the fracture toughness of the composite system (Hanson, 1995). The vast majority of reinforcing steel in use globally consists of unprotected carbon steel — mild steel — favoured for its broad availability, weldability, and low unit cost. Carbon steel reinforcement in good-quality, uncarbonated concrete is protected from corrosion by a thin, adherent passive film that forms under the highly alkaline conditions of fresh concrete pore solution (pH 12.6–13.8).

Sulphate and chloride ions are ubiquitous contaminants in natural soils, groundwater, marine environments, and industrial waste streams (Morcillo et al., 1997). Both anion species are capable of compromising the integrity of the passive layer on embedded steel; however, the overwhelming majority of rebar corrosion research has focused on chloride-induced depassivation. Sulphate attack on mild steel in concrete — particularly its long-term kinetics, quantitative mass loss behaviour, and morphological character — remains comparatively ill-defined in the peer-reviewed literature. This knowledge gap has practical consequences: current durability standards for reinforced concrete structures in aggressive ground conditions typically specify chloride threshold concentrations but afford less systematic attention to sulphate threshold values for reinforcement corrosion initiation.

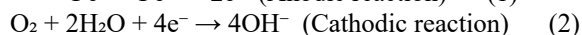
Potgieter et al. (1993) demonstrated that sulphate ions contribute to pitting corrosion of steel under specific electrochemical conditions. Vasquez et al. (1984) showed that alkaline solutions containing sulphur-bearing anions produce pitting of mild steel at potentials more positive than the active-passive transition of iron in basic media. Mursalo et al. (1988) demonstrated that the corrosivity of chloride towards mild steel increases with sulphate addition, while Cheng et al. (1990) concluded that sulphate ions can produce greater corrosion rate increases than chloride ions at equivalent concentrations in simulated concrete pore solutions. However, systematic, long-term gravimetric data for mild steel rebar across a well-defined matrix of sulphate and chloride concentrations at two environmentally representative pH values — pH 12 (fresh concrete) and pH 9 (carbonated concrete) — remain largely absent from the literature.

The present investigation directly addresses this gap. The specific objectives of this study were: (i) to determine the extent, rate, and kinetics of corrosion of mild steel in alkaline media attributable to sulphate ions both independently and in combination with increasing chloride concentrations; (ii) to characterise the type and mechanism of corrosion attack as a function of pH, using quantitative gravimetric and qualitative morphological data collected over six months; and (iii) to establish whether corrosion products formed under these conditions are passivating or non-passivating in nature, and to interpret the results within the context of concrete service-life durability. The hypothesis tested is that sulphate ions cause significant, progressive, and non-self-limiting corrosion of mild steel in alkaline media consistent with concrete pore solution conditions, and that the severity, rate, and morphological character of this attack are governed by pH.

2. Literature Review

2.1 Electrochemical Mechanism of Mild Steel Corrosion in Alkaline Media

Metallic corrosion is broadly defined as the deterioration of a material surface arising from chemical or electrochemical reaction with its environment (Roberge, 2000). In the wet corrosion process governing rebar embedded in concrete or concrete-representative solutions, the essential prerequisites are water, oxygen, and an ionic electrolyte. The anodic oxidation of iron and the cathodic reduction of dissolved oxygen proceed according to the following fundamental half-reactions:



The ferrous ions generated at the anode react with hydroxide ions to form ferrous hydroxide ($\text{Fe}(\text{OH})_2$), which is subsequently oxidised to a range of iron oxides and oxyhydroxides, including magnetite (Fe_3O_4), haematite ($\alpha\text{-Fe}_2\text{O}_3$), goethite ($\alpha\text{-FeOOH}$), lepidocrocite ($\gamma\text{-FeOOH}$), and green rusts (Genim et al., 1986; Refait et al., 2006). Under conditions of depleted dissolved oxygen — such as in sealed test systems after extended immersion — the Kuch mechanism (Kuch,

1988) may sustain corrosion by the reduction of previously deposited ferric oxyhydroxide scale acting as an electron acceptor: $\text{Fe} + 2\text{FeOOH} + 2\text{H}^+ \rightarrow 3\text{Fe}^{2+} + 4\text{OH}^-$. This autocatalytic mechanism enables continued iron dissolution even after ambient dissolved oxygen is substantially consumed.

The Pourbaix (E-pH) diagram for the Fe-H₂O system at 25°C (Genim et al., 1986) predicts that, at pH 9 and pH 12, the thermodynamically stable corrosion products include goethite, magnetite, and green rusts, depending on the prevailing electrochemical potential. At pH values exceeding 12, a passive ferric oxide film is thermodynamically favoured; however, Huet et al. (2005) demonstrated that iron (III) oxide phases forming on mild steel in concrete pore solutions are reactive, porous, and poorly crystallised solids that do not function as a true passivating barrier — in sharp contrast to the dense, stable passive film characteristic of austenitic stainless steels.

2.2 Role of Sulphate Ions in the Corrosion of Mild Steel

The role of sulphate ions in the corrosion of mild steel in alkaline environments has received considerably less systematic attention than chloride-induced attack. Vasquez et al. (1984) conducted potentiostatic and potentiodynamic studies on mild steel in alkaline solutions (pH ~11.3) containing sulphur-bearing sodium salts and observed macroscopic pitting consistent with a competitive surface reaction mechanism involving passive film formation and nucleation of a ferrous sulphide salt layer at higher anodic potentials.

Mursalo et al. (1988) established a critical distinction between mild steel and stainless steel in synthetic mine waters containing combined sulphates and chlorides: while sulphate ions exert a beneficial passivating effect on stainless steel, mild steel is adversely affected by both anion species simultaneously, undergoing general corrosion without passivation. Furthermore, the corrosivity of chloride towards mild steel was found to increase with sulphate addition, demonstrating a synergistic interaction with broad engineering significance.

Cheng et al. (1990) studied sulphate attack using simulated concrete pore solutions and concluded that sulphate ions modify the protective iron oxide film to a less protective sulphate film, and that sulphate ions may produce greater corrosion rate increases than chloride ions at normal ambient temperatures. Dehwah et al. (2002) showed that the rate of chloride-induced corrosion in Portland cement concrete specimens increased by factors of 1.1 to 2.4 in the presence of sulphate, attributable to decreased electrical resistivity of the concrete matrix from the combined ionic presence. Similarly, Mullick (2000) demonstrated that the electrical resistivity of concrete is lower in the presence of combined NaCl and Na₂SO₄ than with either salt in isolation.

Moayed and Newman (2006) investigated sulphate ion effects on the critical pitting temperature of high-alloy austenitic stainless steel 904L and identified five concurrent effects of sulphate in chloride solutions: slight inhibition of passive film breakdown; facilitation of stable pit growth at reduced temperatures; passivation of stable pits; increased bulk solution conductivity; and alteration of pit chemistry. Critically, the authors noted that, for mild steel, the absence of a protective passive film fundamentally alters the corrosion response, rendering these stainless-steel-specific findings inapplicable without significant mechanistic qualification.

Hartt et al. (2004) confirmed in comparative testing that corrosion rates of mild steel reinforcement increased progressively with time in chloride media, while stainless steel corrosion rates stabilised, confirming passive film stability on alloyed steels but not on carbon steel. Huet et al. (2005) further verified through electrochemical and XRD analysis that iron oxide products on mild steel in alkaline concrete conditions are porous, non-passivating, and allow sustained corrosion action.

2.3 Tuberculation and Localised Corrosion in Alkaline Media

Obrecht and Pourbaix (1967) distinguished between uniform corrosion products (generalised scales) and localised corrosion products (tubercles): dome-shaped mounds of corrosion product depositing over and capping discrete zones of intense subsurface metal loss. Tubercle formation is promoted by elevated concentrations of sulphate, chloride, and other aggressive anions, as well as by high bicarbonate alkalinity in soft waters. The mineral phases associated with tubercle formation include goethite, lepidocrocite, magnetite, siderite, ferrous hydroxide, ferric hydroxide, and various green rust phases (Sarin et al., 2001; Benjamin et al., 1996).

Fontana (1987) described the growth mechanism of corrosion tubes at pit boundaries: ferrous hydroxide forms at the interface between an active pit and the adjacent passive surface through the interaction of OH⁻ from the cathodic reaction

and soluble corrosion products migrating outward from the pit; this precipitate is further oxidised to $\text{Fe}(\text{OH})_3$, Fe_3O_4 , Fe_2O_3 , and higher oxyhydroxides, and the product rim grows outward in the characteristic form of a tube or convoluted appendage. The autocatalytic character of pitting sustains pit stability: the concentrated, oxygen-depleted solution within the pit is essential to its propagation, and pits preferentially develop in the direction of gravitational drainage. The build-up of corrosion scale on mild steel is explicitly non-protective because the phases produced are poorly crystallised, compositionally heterogeneous, and highly friable (Machiels and Munson, 2003).

Singh (2006) attributed the increase in corrosion rates at elevated sulphate concentrations to two concurrent effects: increased solution ionic conductivity and the formation of basic ferric sulphate, which drives intense localised dissolution. Baylis (1953) further noted the magnetic character of magnetite and proposed that magnetic attraction may cause precipitated ferric scale to accumulate on the steel surface, influencing the spatial distribution of subsequent tubercle growth. The Kuch (1988) mechanism — whereby ferric scale acts as a terminal electron acceptor under oxygen-limited conditions — provides an additional electrochemical pathway by which severe corrosion may persist indefinitely in sealed or near-stagnant systems.

2.4 Research Gaps Addressed by This Study

A synthesis of the literature reviewed above reveals the following critical research gaps: (i) the overwhelming majority of rebar corrosion studies are focused on chloride-induced depassivation, with comparatively few addressing sulphate as a primary corrodent; (ii) systematic, long-term gravimetric data for mild steel rebar in sulphate-dominated alkaline media spanning six months of continuous immersion are largely absent; (iii) the pH-dependent transition between uniform and localised corrosion morphologies for mild steel in sulphate-bearing media has not been rigorously characterised across the pH range relevant to concrete carbonation; and (iv) the non-passivating nature of corrosion product layers on mild steel in these conditions has not been confirmed quantitatively over extended test durations at two pH values representative of the concrete lifecycle.

The present study was designed specifically to address all four of these gaps through a six-month immersion programme spanning nine sulphate/chloride concentration combinations at pH 9 and pH 12, with triplicate determinations at six time intervals, yielding a comprehensive gravimetric and morphological dataset.

3. Experimental Methodology

3.1 Test Material and Chemical Composition

Commercial-grade mild steel flat bar of 3 mm nominal thickness was selected as the test material. The bar was low-carbon, hot-rolled steel representative of uncoated concrete reinforcement (rebar) material in widespread engineering use. The chemical composition as specified by the material supplier is presented in Table 1. Carbon content was independently confirmed by Leco combustion analysis at the Council for Geoscience (Pretoria), yielding a mean carbon content of 0.07% for the 3 mm flat bar, which is characteristic of structural mild steel.

Table 1. Chemical composition of 3 mm mild steel flat bar: supplier specification and Leco verification

Element	Fe	C	Si	P	Mn
% (mol/mol)	98.52	0.15	0.30	0.04	1.00
<i>Carbon content confirmed by Leco analysis (Council for Geoscience): 0.07% C for 3 mm flat bar.</i>					

3.2 Sample Preparation

Coupons were cut from the flat bar in nominal dimensions of 20 mm × 30 mm × 3 mm using a Labcon Struers precision steel cutter. A 2 mm diameter suspension hole was drilled into the upper edge of each coupon for attachment during immersion. Surface preparation followed ASTM G1 guidance for mass loss coupon testing: pickling and sandblasting were avoided due to potential chemical residue contamination and surface damage. Surfaces were progressively ground using silicon carbide emery paper (320 through 1200 grit, water-lubricated), rinsed sequentially in distilled water and analytical-grade ethanol, air-dried, and weighed to the nearest 0.01 g using a calibrated analytical balance. External dimensions were measured using a calibrated Vernier calliper to calculate the exposed geometric surface area for each coupon. Following preparation, coupons were stored in a desiccator over silica gel until commencement of immersion testing.

3.3 Test Solution Preparation

Test solutions were prepared by dissolving chemically pure sodium sulphate (Na_2SO_4) and sodium chloride (NaCl) in double-distilled deionised water to the target concentrations specified in Table 2. Solution pH was adjusted to either 9 or 12 by controlled addition of sodium hydroxide (NaOH) and verified with a calibrated combination electrode pH meter (standardised using NIST-traceable buffers at pH 7.0 and 10.0). Each solution was homogenised using a magnetic stirrer for 20 minutes prior to use. pH 9 is representative of the pore solution in fully carbonated concrete, while pH 12 approximates the pore solution of freshly cast, fully hydrated Portland cement concrete.

Table 2. Test solution matrix: sulphate and chloride concentrations, molar ratios, and test pH values

No.	Composition	$[\text{SO}_4^{2-}]$ (mg/L)	$[\text{Cl}^-]$ (mg/L)	$\text{SO}_4^{2-} : \text{Cl}^-$ Ratio	pH	Duration
1	SO_4^{2-} only	200	0	1 : 0	9 & 12	6 months
2	SO_4^{2-} only	1000	0	1 : 0	9 & 12	6 months
3	SO_4^{2-} only	2000	0	1 : 0	9 & 12	6 months
4	$\text{SO}_4^{2-} + \text{Cl}^-$	200	50	4 : 1	9 & 12	6 months
5	$\text{SO}_4^{2-} + \text{Cl}^-$	200	500	0.4 : 1	9 & 12	6 months
6	$\text{SO}_4^{2-} + \text{Cl}^-$	1000	50	20 : 1	9 & 12	6 months
7	$\text{SO}_4^{2-} + \text{Cl}^-$	1000	500	2 : 1	9 & 12	6 months
8	$\text{SO}_4^{2-} + \text{Cl}^-$	2000	50	40 : 1	9 & 12	6 months
9	$\text{SO}_4^{2-} + \text{Cl}^-$	2000	500	4 : 1	9 & 12	6 months

3.4 Immersion Test Procedure

Each prepared coupon was suspended by nylon thread through its drilled hole and immersed in 2 L of the appropriate test solution contained in a borosilicate glass beaker. Beakers were sealed with fitted Perspex lids and wrapped with cling film to minimise evaporative loss and limit oxygen ingress to the volume of air trapped at the time of sealing. Solutions remained stagnant and without external aeration throughout the test period — conditions representative of pore water in concrete. Ambient temperature was maintained at $25 \pm 3^\circ\text{C}$ (room temperature, with seasonal variation recorded). Triplicate coupons were exposed per test condition and per time interval; all reported results represent the arithmetic mean of triplicate determinations.

Samples were retrieved at the following programmed intervals: 1 week, 2 weeks, 1 month, 2 months, 3 months, and 6 months — yielding a total of 324 individual coupon measurements across all conditions. Upon retrieval, loosely adherent corrosion products were removed by gentle mechanical cleaning with a nylon brush, followed by rinsing in distilled water and ethanol, and thorough air-drying prior to re-weighing. The mass loss was calculated as the difference between the initial and post-exposure mass. Macroscopic digital photography and stereomicroscopy (Nikon SMZ 800; Olympus C-400 digital camera; magnification range $1\times$ to $6.3\times$) were performed on selected representative coupons at each time interval to characterise and document corrosion morphology.

3.5 Corrosion Rate Calculation

Corrosion rates were calculated from the measured mass loss values using the NACE Standard formula (NACE, 1984):

$$\text{Corrosion Rate (mpy)} = 534 \times W / (D \times A \times T) \quad (3)$$

where W is the measured mass loss (mg), D is the density of mild steel (7.82 g/cm^3), A is the exposed geometric surface area of the coupon (cm^2), and T is the total immersion time (hours). Corrosion rates expressed in mils per year (mpy) were converted to millimetres per year (mm/year) by multiplying by the factor 0.0254. All reported corrosion rate values represent the mean of triplicate determinations. Coefficient of determination (R^2) values from linear regression of mass loss versus time were computed for all conditions to quantify the linearity of the kinetic response.

4. Results and Discussion

4.1 Mass Loss Kinetics at pH 12

Table 3 presents the average mass loss of mild steel coupons as a function of immersion time for all nine test solutions at pH 12 over the full six-month duration. In all cases, the relationship between cumulative mass loss and test duration is linear, with R^2 values ranging from 0.96 to 0.99 across all conditions. This consistent straight-line behaviour is of fundamental mechanistic significance: it unequivocally demonstrates that corrosion products formed on the mild steel surface under these highly alkaline conditions are non-passivating in character, and that the corrosion process proceeded without deceleration or passivation throughout the entire duration of the experiment.

The highest cumulative mass loss values after six months were recorded for the 2000 + 50 mg/L and 2000 + 500 mg/L solutions, reaching approximately 280 mg and 277 mg, respectively. At the lowest base sulphate concentration of 200 mg/L, cumulative mass loss after six months was approximately 150 mg in the sulphate-only solution, increasing to approximately 253 mg when 500 mg/L chloride was added — an increase of approximately 69%.

A consistent and reproducible response pattern was observed across all three base sulphate concentrations: the addition of 50 mg/L chloride produced only a modest increase in mass loss relative to sulphate-only solutions, whereas the addition of 500 mg/L chloride produced a markedly disproportionate acceleration. This confirms that a ten-fold increase in chloride concentration from 50 to 500 mg/L exerts a substantially greater influence on corrosion rate than a commensurate proportional increase in sulphate concentration, indicative of a non-linear synergistic interaction between the two anion species at the corroding steel surface (Table 3 summarises the key six-month results).

Table 3. Summary of cumulative mass loss and estimated corrosion rate rankings at the six-month endpoint for all test conditions.

Test Condition	6-month Mass Loss pH 9 (mg)	6-month Mass Loss pH 12 (mg)	CR pH 9 (mm/year)	CR pH 12 (mm/year)	R^2
200 mg/L SO_4^{2-} only	~207	~150	Low	Low	≥ 0.97
1000 mg/L SO_4^{2-} only	~185	~210	Moderate	Moderate	≥ 0.97
2000 mg/L SO_4^{2-} only	~195	~240	Moderate	High	≥ 0.97
200 + 50 mg/L Cl^-	~185	~175	Low	Low	≥ 0.97
200 + 500 mg/L Cl^-	~210	~253	Moderate	High	≥ 0.96
1000 + 50 mg/L Cl^-	~190	~220	Moderate	Moderate-High	≥ 0.97
1000 + 500 mg/L Cl^-	~220	~260	Moderate-High	High	≥ 0.97
2000 + 50 mg/L Cl^-	~235	~280	High	Very High	≥ 0.97
2000 + 500 mg/L Cl^-	~247	~277	High	Very High	≥ 0.96

4.2 Mass Loss Kinetics at pH 9

The aggregate average mass loss at pH 9 for all nine test solutions over six months. Linear mass loss versus time relationships were observed at pH 9 for all concentrations, with R^2 values between 0.97 and 0.99, confirming non-passivating kinetics at the lower pH value as well. The maximum mass loss recorded after six months at pH 9 was approximately 247 mg, achieved in the 2000 + 500 mg/L solution — slightly lower than the maximum recorded at pH 12 for comparable concentrations.

A notable departure from the pH 12 behaviour was observed at pH 9: the addition of increasing chloride concentrations to sulphate solutions did not consistently produce greater mass loss at pH 9. In certain test conditions, mass loss values

exhibited a marginal reduction upon chloride addition relative to sulphate-only solutions at the same sulphate concentration. This contrasts markedly with the consistent positive chloride-sulphate synergism observed at pH 12, suggesting that the mechanism by which chloride amplifies sulphate-driven corrosion is contingent on the level of alkalinity and the specific character of the corrosion products formed at each pH.

4.3 Comparative Mass Loss: pH 9 versus pH 12

A direct comparison of mass loss at pH 9 and pH 12 for each of the nine test concentrations reveals that, for sulphate-only solutions at 1000 and 2000 mg/L, and for the majority of sulphate-chloride combinations, cumulative mass loss values after six months are substantially higher at pH 12. The sole notable exception was the 200 mg/L sulphate-only solution, where pH 9 mass loss (approximately 207 mg) marginally exceeded the pH 12 value (approximately 150 mg), consistent with the observation of Cohen and Mather (1991) that changes in sulphate concentration can alter the operative mechanism of attack.

The preponderance of greater mass loss at pH 12 is interpreted as follows. While higher alkalinity is conventionally expected to stabilise passive films on metallic surfaces, the absence of a true protective passive film on mild steel — confirmed independently by Huet et al. (2005) and Hartt et al. (2004) — means that the primary determinant of corrosion rate becomes the ionic conductivity of the electrolyte. At pH 12, equivalent solute concentrations yield higher ionic conductivity than at pH 9, and this increased solution conductivity directly accelerates the corrosion rate in the absence of passivation (Morcillo et al., 1997; Singh, 2006; Mullick, 2000). The linear mass loss relationships at both pH values confirm that mild steel undergoes progressive, non-self-limiting corrosion under all conditions examined.

4.4 Corrosion Rate Analysis

Corrosion rates were greatest at the six-month measurement endpoint across all conditions, consistent with the non-passivating linear kinetics established by the mass loss data: since mass loss is linear with time, the corrosion rate computed over the full six-month exposure represents the most meaningful steady-state value. At all three base sulphate concentrations, corrosion rates increase monotonically with sulphate concentration in the absence of chloride, more markedly so at pH 12 than at pH 9. Corrosion rates at pH 12 after six months substantially exceeded those at pH 9 for all high-sulphate conditions and for combined sulphate-chloride solutions. The pie chart distributions of corrosion rates at the six-month endpoint confirm that pH 12 conditions consistently produce higher corrosion rates across all concentration combinations, and that all pH 9 rates at six months are statistically comparable, confirming uniform attack at a steady, moderately elevated rate.

4.5 pH-Dependent Corrosion Morphology

4.5.1 Morphology at pH 9

At pH 9, a uniform, orange-coloured powdery corrosion product was observed blanketing the entire surface of all coupons throughout the full six-month immersion period, across all nine test concentrations. The product was loosely adherent, readily dislodged by minimal mechanical disturbance, and non-cohesive. After product removal, coupon surfaces appeared uniformly stained and finely roughened, but no discrete pits, localised zones of preferential attack, or tubercle formations were detectable by visual inspection or stereomicroscopy. Stereomicroscope examination confirmed the classification of this morphology as generalised uniform etch corrosion — a progressive, diffuse surface dissolution without preferential localised attack.

This uniform corrosion mode is fully consistent with the general corrosion behaviour of mild steel in alkaline media described by Mursalo et al. (1988), and indicates that under carbonated concrete conditions (pH 9), sulphate attack proceeds as a spatially homogeneous dissolution of the steel surface. The loosely adherent, easily removed corrosion products confirm the absence of any passivating layer. From a structural engineering perspective, uniform corrosion allows for deterministic service-life prediction: the rate of cross-sectional area loss can, in principle, be integrated over time to model load-bearing capacity reduction.

4.5.2 Morphology at pH 12

At pH 12, a qualitatively and mechanistically distinct and markedly more severe form of attack was observed across all nine test concentrations. A crusted, structured, voluminous corrosion product was deposited on coupon surfaces, consistent with tubercle and scaling corrosion as described by Fontana (1987), Sarin et al. (2001), and Obrecht and Pourbaix (1967). Different discrete areas on each coupon exhibited individualised attack zones, with tubercles forming at

each localised reaction centre. Several tubercles presented convoluted, finger-like corrosion product growths extending from the primary deposit, consistent with the corrosion tube growth mechanism described by Fontana (1987).

After mechanical removal of corrosion products from pH 12 samples, coupon surfaces exhibited not sharp, narrow deep pits but rather a characteristic pattern of layered material removal: successive surface strata removed from the metal substrate over progressive immersion intervals, resulting in measurable net mass loss. Stereomicroscope images confirmed the development of shallow, broad-based pitting on samples exposed for the full six-month duration, with severity increasing with immersion time and sulphate concentration. The scale formed at pH 12 was highly friable and detached readily from the metal surface upon minimal mechanical perturbation — clear macroscopic evidence that, despite its volumetric prominence, the accumulated corrosion product offers no protective function. This finding is wholly consistent with the results of Hartt et al. (2004), Dakhil et al. (1984), and Huet et al. (2005), all of whom reported that iron oxide products on mild steel in alkaline media do not constitute a passivating layer.

The voluminous, poorly adherent corrosion products observed at pH 12, by creating a concentration cell between the cathodic exterior and the anodic interior of each tubercle, may in fact accelerate further attack through the Kuch mechanism. The accumulation of ferric oxyhydroxide products enables oxygen-independent corrosion continuation even in the sealed, oxygen-limited test environment.

4.6 Non-Passivating Linear Kinetics: Mechanistic Interpretation

The linear mass loss versus time relationships universally observed at both pH 9 and pH 12 carry mechanistic significance of first importance. Linear kinetics are definitionally inconsistent with the development of any protective passive layer on the steel surface. This is in stark contrast to the behaviour of stainless steels, for which stable passive films produce decreasing or asymptotically stable corrosion rates over time (Hartt et al., 2004). For mild steel, the iron (III) oxide corrosion products that accumulate on the surface are characterised by poor crystallinity, high porosity, and weak adhesion — properties that prevent them from functioning as effective solid-state diffusion barriers against electrolyte access to the underlying metal.

Two complementary mechanisms account for the persistence of non-passivating linear kinetics throughout the six-month test. First, corrosion products are insufficiently dense and adherent to create an effective barrier layer: reactants (water, dissolved sulphate, residual oxygen) continue to access the bare metal surface beneath the friable crust. Second, the Kuch (1988) mechanism sustains corrosion by enabling ferric oxyhydroxide scale to function as a terminal electron acceptor under oxygen-depleted conditions, permitting continued iron oxidation even after dissolved oxygen in the sealed test vessels is substantially consumed. The implication is unambiguous: corrosion of mild steel rebar in sulphate-bearing alkaline media will not self-limit. Once initiated, it will persist at a steady rate unless the rebar is physically removed from the corrosive environment or effective protective measures are applied.

4.7 The pH Effect: Fresh versus Carbonated Concrete Conditions

The comparison of results at pH 9 and pH 12 reveals that the two pH regimes differ not only in the absolute magnitude of corrosion but in the fundamental character and mechanism of attack. At pH 9, representative of fully carbonated concrete, corrosion is spatially uniform and progresses at a moderate, steady rate. At pH 12, representative of freshly cast hydrated concrete, corrosion rates are generally elevated and the attack is strongly localised — more concentrated and, critically, more structurally damaging per unit of mass loss.

These findings carry important infrastructure lifecycle implications. In reinforced concrete, the pH of the pore solution is initially very high (pH 12.6–13.8) and decreases progressively as atmospheric CO₂ diffuses through the cover concrete and reacts with calcium hydroxide, eventually reaching pH 8–9 in fully carbonated zones. The present results demonstrate that the period of highest structural risk from sulphate-induced corrosion of the reinforcement corresponds to the early service life of the structure, when alkalinity is highest and localised pitting and tuberculation are most active. This localised attack may initiate significant structural damage before any surface indication of deterioration becomes visible, making early inspection and condition monitoring critical for structures in sulphate-bearing environments.

The greater corrosion severity at pH 12 appears paradoxical in the context of the conventional expectation that higher alkalinity should stabilise passive films. The explanation lies in the confirmed failure of iron oxides to form a truly protective layer on mild steel, which removes the alkalinity-passivation benefit entirely. In the absence of passivation, the

higher ionic conductivity of the pH 12 solutions relative to pH 9 at equivalent solute concentrations directly drives higher electrochemical corrosion rates (Morcillo et al., 1997; Mullick, 2000; Singh, 2006).

4.8 Synergistic Effect of Chloride on Sulphate-Induced Corrosion

The data demonstrate a threshold, non-linear response to chloride addition. At both pH values, the addition of 50 mg/L chloride to sulphate solutions produced only marginal increases in mass loss relative to sulphate-only solutions. The addition of 500 mg/L chloride — a ten-fold increase — produced a markedly disproportionate acceleration. This threshold behaviour strongly suggests that chloride ions begin to exert substantial synergistic effects on sulphate-induced corrosion only after reaching a critical concentration, below which their incremental influence on the dominant sulphate-driven dissolution is minor.

At pH 12, the mass loss values for the 200 + 500 mg/L combined solution (approximately 253 mg after six months) greatly exceeded those for 200 mg/L sulphate alone (approximately 150 mg) — a relative increase of approximately 69%. The corresponding differential at pH 9 was substantially smaller, confirming that the synergistic aggravation of sulphate corrosion by chloride is most pronounced under conditions of high alkalinity, where the attack mechanism is already localised and the corrosion cell geometry is most favourable to autocatalytic pit propagation. These results are broadly consistent with the findings of Dehwah et al. (2002) and Mullick (2000), and extend their applicability specifically to the context of mild steel rebar in sulphate-dominated solutions.

A critical practical implication follows: sulphate and chloride concentrations should not be evaluated independently in the context of rebar corrosion risk assessment. The combined presence of both ionic species at ratios and concentrations intersecting the non-linear synergism threshold identified here represents a substantially greater threat to mild steel reinforcement durability than either ion in isolation would predict.

4.9 Structural Implications of pH-Dependent Corrosion Morphology

The pH-dependent transition from uniform corrosion at pH 9 to localised pitting and tuberculation at pH 12 carries direct and significant structural engineering implications. Uniform corrosion reduces the gross cross-sectional area of the rebar at a predictable, spatially distributed rate, enabling analytical service-life modelling of load-bearing capacity degradation. Localised pitting and tuberculation, by contrast, are inherently less spatially predictable, are capable of propagating rapidly in depth at isolated points of attack, and generate voluminous corrosion products that exert expansive outward forces on the surrounding concrete matrix. The convoluted, finger-like tubercle formations documented in this study at pH 12 are wholly consistent with descriptions in the literature of corrosion-product-driven concrete spalling and cover cracking (Montemor et al., 2003).

The formation of these expansive products initiates a coupled degradation cycle: electrochemical dissolution of the reinforcement is accompanied by physical disruption of the concrete cover. Once the cover cracks and spalls, the effective diffusion pathway for aggressive ionic species is dramatically shortened, accelerating the rate of sulphate and chloride ingress and establishing a positive feedback cycle of accelerating deterioration. The sulphate-induced pitting and tuberculation documented at pH 12 in this study is therefore capable of initiating this destructive cycle at a comparatively early stage in the structural life — before any pH reduction attributable to carbonation provides a degree of morphological mitigation.

4.10 Comparison with Prior Literature and Study Limitations

The conclusion that sulphate attack at high pH is more aggressive than chloride-induced corrosion in isolation is directly supported by Cheng et al. (1990), who concluded that sulphate ions produce greater corrosion rate increases than chloride ions at normal temperatures. The linear, non-passivating kinetics observed here are consistent with the progressive corrosion rate increases reported by Hartt et al. (2004) for mild steel rebar in chloride media, and extend this mechanistic characterisation to sulphate-dominated and combined sulphate-chloride alkaline media for the first time over a six-month duration.

The universal absence of passivation under all test conditions — including sulphate-only solutions and at both pH values — is consistent with the thermodynamic predictions of the Pourbaix diagram for the Fe-H₂O system in the pH 9 to pH 12 range (Genim et al., 1986), which does not predict the formation of a stable passivating phase for mild steel under these conditions. It is further consistent with the conclusions of Huet et al. (2005), whose XRD and electrochemical analyses identified a thin, poorly adherent iron (III) oxide layer on mild steel in alkaline pore solutions that does not prevent

continued corrosion. The present six-month gravimetric programme provides direct quantitative macroscopic confirmation of these microscale observations at an environmentally representative scale.

Several limitations of the present study should be acknowledged. Test solutions employed sodium sulphate exclusively as the sulphate source; other cations associated with sulphate in natural groundwater and soils (calcium, magnesium, potassium) may produce different corrosion products and altered kinetics (Samthanam et al., 2001). Furthermore, as noted by Metha (1975) and Brown (1981), continuous laboratory immersion may not perfectly replicate field conditions, where sulphate concentration can diminish over time through consumption reactions. These boundary conditions suggest that the absolute corrosion rates reported here may differ quantitatively from those in actual cementitious environments; however, they are appropriate for the comparative mechanistic characterisation and pH-dependence analysis that constitute the primary objectives of the study.

5. Conclusions

This study has systematically characterised the long-term corrosion kinetics and morphological behaviour of mild steel reinforcement in alkaline sulphate and sulphate-chloride solutions at pH 9 and pH 12, representative of carbonated and freshly hydrated concrete pore environments, respectively. The following principal conclusions are drawn:

- Corrosion kinetics are linear and non-passivating at both pH 9 and pH 12 across all nine sulphate and chloride concentration combinations tested over six months ($R^2 = 0.96\text{--}0.99$). The absence of any passivating plateau in the mass loss data confirms that the corrosion products formed on mild steel under all conditions investigated are porous, weakly adherent, and non-protective. Once initiated, sulphate-driven corrosion of mild steel will continue unabated unless the rebar is removed from the corrosive environment or an effective barrier is applied.
- Mass loss and corrosion rates at pH 12 are generally greater than at pH 9 for equivalent solute compositions, particularly at higher sulphate concentrations. This demonstrates that sulphate attack on mild steel reinforcement is more aggressive under fully alkaline conditions representative of freshly hydrated concrete than under lower pH conditions representative of carbonated concrete.
- A critical pH-dependent transition in corrosion morphology was identified: pH 9 produced diffuse uniform etch corrosion across all concentrations and durations, while pH 12 consistently produced severe localised pitting, tuberculation, and convoluted scale formation. The localised attack at pH 12, with its associated expansive product formation, is substantially more structurally damaging than the uniform corrosion at pH 9 and is capable of initiating concrete cover cracking and spalling.
- Higher sulphate concentrations consistently increased corrosion rates and cumulative mass loss at both pH values. A non-linear synergistic relationship between sulphate and chloride was identified: the addition of 50 mg/L chloride produced modest corrosion acceleration, whereas 500 mg/L chloride (a ten-fold higher concentration) produced a dramatically disproportionate increase, particularly at pH 12. Sulphate and chloride concentrations therefore cannot be assessed independently in reinforced concrete durability evaluations.
- Sulphate ions, at concentrations as low as 200 mg/L at pH 12, cause significant and progressive corrosion of mild steel reinforcement. Under fully alkaline conditions (pH 12), sulphate attack is more aggressive and structurally dangerous than chloride-induced corrosion at equivalent concentrations. This finding directly challenges the current tendency in reinforced concrete durability practice to focus monitoring and threshold compliance predominantly on chloride concentrations while systematically underestimating the corrosion risk posed by sulphate-bearing environments.

6. Alignment with United Nations Sustainable Development Goals

wledge that underpins The research presented in this study carries implications that extend substantially beyond the discipline of corrosion science. The findings directly support and advance multiple United Nations Sustainable Development Goals (UN SDGs), adopted under the 2030 Agenda for Sustainable Development. The SDG framework recognises that durable, safe, and resource-efficient infrastructure is a prerequisite for equitable human development, poverty reduction, and environmental protection. The present investigation, by characterising the mechanisms and severity of sulphate-induced corrosion in reinforced concrete — the world's most widely used structural material — contributes fundamental knowledge to the engineering of more durable, safer, and more resource-efficient built environments at a global scale. The specific SDG alignments are articulated below.

SDG 3: Good Health and Well-Being

The structural failure of reinforced concrete buildings, bridges, hospitals, schools, and water infrastructure attributable to rebar corrosion represents a direct and demonstrable public safety hazard. Sudden or progressive structural collapse

caused by undetected rebar deterioration poses a lethal risk to occupants and surrounding communities. SDG 3 commits signatory nations to ensuring healthy lives and promoting well-being for all ages. This research directly advances SDG 3 by providing the scientific basis for identifying and mitigating a corrosion mechanism — sulphate-induced attack — that has been systematically underestimated relative to chloride-induced attack in current engineering practice. By demonstrating that sulphate-bearing environments cause non-self-limiting, progressive structural degradation beginning at concentrations as low as 200 mg/L at the alkalinity of fresh concrete, this study enables engineers, regulators, and asset owners to make more accurate safety assessments of structures in sulphate-rich ground conditions, thereby directly reducing risk to human life. The finding that localised pitting and tuberculation at pH 12 can cause significant subsurface structural damage before any external sign is visible is particularly significant from a public safety perspective, as it underscores the inadequacy of visual inspection alone as a health-and-safety monitoring tool.

SDG 6: Clean Water and Sanitation

Sulphate contamination of groundwater and industrial effluent is globally prevalent, particularly in regions underlain by sulphate-bearing geology, in proximity to mining operations, and in areas receiving agricultural runoff. Reinforced concrete is the primary structural material for water treatment plants, dams, reservoirs, sewage systems, pump stations, and water distribution infrastructure. The corrosion of mild steel rebar in sulphate-bearing environments is therefore a direct threat to the long-term integrity of water and sanitation infrastructure. SDG 6 calls for universal access to safe and affordable drinking water and sanitation services, and requires that these services be maintained reliably over time. This research directly informs the design, specification, and maintenance of reinforced concrete water infrastructure in sulphate-bearing environments by quantifying the kinetics and morphological severity of sulphate-driven corrosion. The six-month gravimetric dataset generated here, establishing non-passivating linear corrosion kinetics across a range of sulphate and chloride concentrations at two environmentally representative pH values, provides the empirical foundation for evidence-based durability specifications for concrete water infrastructure — directly enabling engineers to design structures capable of delivering sustained clean water and sanitation services over multi-decade service lives.

SDG 9: Industry, Innovation, and Infrastructure

SDG 9 explicitly calls for the development of quality, reliable, sustainable, and resilient infrastructure. Globally, reinforced concrete infrastructure represents an investment of many trillions of dollars; premature failure attributable to corrosion not only destroys this investment but imposes enormous additional costs through reactive maintenance, emergency repair, unplanned service disruption, and accelerated replacement. The present investigation is of profound significance to SDG 9 because it fills a critical and long-standing knowledge gap in the understanding of how sulphate ions — a prevalent environmental contaminant — degrade the structural integrity of reinforced concrete at concentrations representative of real-world exposure. The demonstration that sulphate attack is more severe and more structurally dangerous at pH 12 (the condition of freshly cast concrete, representing the greatest proportion of the structural service life before carbonation) than at pH 9 is a finding of direct engineering design significance. It implies that durability specifications based solely on chloride threshold values are insufficient to ensure infrastructure resilience in sulphate-rich environments, and that revised material specifications, concrete mix design, and cover concrete depth requirements are warranted. Furthermore, the non-linear synergism between sulphate and chloride identified in this study provides the empirical evidence base necessary for the development of combined ionic exposure limit values in engineering standards — an innovation directly advancing SDG 9's mandate for resilient, knowledge-driven infrastructure.

SDG 11: Sustainable Cities and Communities

SDG 11 seeks to make cities and human settlements inclusive, safe, resilient, and sustainable, with specific emphasis on inclusive and sustainable urbanisation and on protecting and safeguarding the world's cultural and natural heritage. Rapid urbanisation in the developing world — particularly across sub-Saharan Africa, South and Southeast Asia, and Latin America, where sulphate-bearing soils and groundwater are common — is producing unprecedented quantities of new reinforced concrete infrastructure at a time when understanding of sulphate-driven rebar corrosion is least mature. This research is of particular relevance to SDG 11 in the context of the Global South: many rapidly urbanising regions lack the dense network of long-term infrastructure monitoring data available in developed nations, making research-based durability design all the more critical. The present study, conducted at a South African university and directly addressing environmental conditions prevalent across the African continent and other developing regions, contributes foundational scientific knowledge that supports the design of durable, safe, and sustainable urban built environments in these high-growth contexts. Moreover, the identification of the positive feedback deterioration cycle — whereby sulphate-induced pitting and tuberculation cause concrete cover cracking, which in turn accelerates further ionic ingress — underscores the

importance of preventive durability design over reactive maintenance, a principle that is economically imperative in communities with constrained infrastructure maintenance budgets.

SDG 12: Responsible Consumption and Production

The production of steel and Portland cement — the two primary components of reinforced concrete — together account for approximately 15% of global anthropogenic CO₂ emissions and consume vast quantities of energy, water, and mineral resources. The premature failure of reinforced concrete structures due to corrosion dramatically amplifies the resource burden of the built environment: each tonne of corroded rebar that necessitates premature replacement represents not only the embodied carbon and energy of the original material, but also that of its replacement, together with the demolition and waste disposal burden of the failed structure. SDG 12 calls for the decoupling of economic growth from environmental degradation through sustainable patterns of production and consumption. The present research advances SDG 12 by establishing the scientific basis for accurately predicting and extending the service life of reinforced concrete structures in sulphate-bearing environments. Extended structural service life directly reduces the frequency of material replacement and the associated resource and emissions burden per unit of structural output. The findings challenge current over-reliance on chloride thresholds alone for durability specification, which may inadvertently lead to under-specified concrete in sulphate-rich environments and hence premature, environmentally wasteful structural failure. Correct durability specification, informed by the present findings, reduces unnecessary consumption of cement, steel, aggregates, water, and energy over the lifecycle of infrastructure assets.

SDG 13: Climate Action

Climate change is progressively altering the chemical composition of soils and groundwater, including through increased sulphate mobilisation from geological formations, greater frequency and intensity of acid mine drainage events, and changes in precipitation patterns that concentrate aggressive ions in near-surface environments. These trends are likely to increase the exposure frequency and severity of reinforced concrete infrastructure to sulphate-rich conditions globally. SDG 13 calls for urgent action to combat climate change and its impacts. The present research contributes to climate adaptation by providing the scientific knowledge base required to design and maintain reinforced concrete infrastructure that remains structurally sound under the more aggressive ionic exposure conditions projected under climate change scenarios. Furthermore, the finding that sulphate attack is most severe at the early-life high-alkalinity condition of concrete has implications for the deployment of supplementary cementitious materials (SCMs) such as fly ash, ground granulated blast-furnace slag (GGBS), and silica fume — materials that both reduce the carbon footprint of concrete and modify pore solution chemistry. Understanding the sulphate corrosion kinetics at pH 12 is essential for evaluating the protection afforded by such materials as a climate-aligned concrete technology strategy.

SDG 17: Partnerships for the Goals

SDG 17 recognises that the achievement of the 2030 Agenda requires multi-stakeholder partnerships, knowledge-sharing across national boundaries, and the strengthening of science-policy interfaces. This research, produced at a South African university through collaboration between industry-facing researchers and academic supervisors, represents precisely the kind of nationally rooted, globally relevant knowledge contribution that SDG 17 seeks to catalyse. The experimental data generated — a six-month gravimetric corrosion dataset with 324 individual measurements across 18 distinct test conditions — constitutes an open, replicable, and transferable scientific contribution that can directly inform revised corrosion standards, building codes, and infrastructure durability specifications across multiple jurisdictions. By closing a well-defined scientific knowledge gap, this study creates the conditions for productive collaboration between corrosion scientists, structural engineers, materials standardisation bodies, and infrastructure regulators globally, in service of the shared goal of durable and sustainable built infrastructure.

6.7 Overarching Significance of This Research

The significance of this research must be understood at three intersecting scales: scientific, engineering, and global development.

At the scientific scale, this study delivers the most comprehensive long-term gravimetric characterisation of sulphate-induced corrosion of mild steel rebar in alkaline media reported to date. The 324-measurement, six-month dataset spanning 18 conditions establishes, for the first time in a systematic manner, the linear non-passivating kinetic character of sulphate attack, the pH-dependent transition between uniform and localised corrosion morphologies, and the non-linear synergism between sulphate and chloride at two pH values representative of the concrete lifecycle. These findings resolve

longstanding ambiguities in the corrosion literature and extend the experimental evidence base for the Kuch mechanism and the Pourbaix diagram predictions to the specific context of sulphate-bearing concrete pore solution analogues.

At the engineering scale, the research reveals that current engineering practice in sulphate-rich environments is likely inadequate. The focus of durability standards, inspection protocols, and corrosion risk assessment frameworks on chloride thresholds — while neglecting sulphate thresholds for rebar corrosion — leaves a systematic blind spot in the structural integrity management of reinforced concrete infrastructure globally. This study provides the quantitative data necessary to close that blind spot: to define sulphate concentration thresholds for corrosion initiation, to characterise the kinetics for service-life prediction models, and to establish that combined sulphate-chloride exposure requires a fundamentally different risk assessment approach than single-ion evaluation. These are not marginal refinements — they represent a paradigm correction in reinforced concrete durability engineering.

At the global development scale, the alignment of this research with SDGs 3, 6, 9, 11, 12, 13, and 17 demonstrates that it is not merely a contribution to an academic discipline but a direct enabler of safer communities, cleaner water, more resilient cities, lower material consumption, and more effective climate adaptation. Reinforced concrete infrastructure is the physical skeleton of modern civilisation. Research that improves our ability to protect this material from corrosion — at lower cost, with greater accuracy, and over longer service lives — makes a measurable contribution to human well-being at a civilisational scale. This study, modest in its laboratory footprint but ambitious in its scientific rigour and scope, represents precisely the kind of fundamental research investment that the UN SDG framework recognises as essential to achieving the 2030 Agenda.

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CRedit Authorship Contribution Statement

Kasturie Premlalla: Conceptualisation, Data Curation, Formal Analysis, Investigation, Methodology, Validation, Visualisation, Writing – Original Draft. Douw Faurie: Writing – Review and Editing. Herman Potgieter: Supervision, Writing – Review and Editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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