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SIMULATION STUDIES ON CORROSION OF STONE COATED ROOFING SHEETS SOLD IN NIGERIA

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ABSTRACT. Acid rain condition were simulated on three selected stone-coated roofing sheets sold in Nigeria to assess the rate of metallic dissolution using (0.25, 0.50, 0.75, and 1.00 M) concentration of sulfuric acid, simulated acid rain (0.5 M of HCl, HNO_3 and H_2SO_4) for 30 days in a controlled environment. Six metals (Pb, Fe, Zn, Al, Si, and Cd) concentration were determined using same concentration of sulfuric acid and distilled water for three hours at elevated temperature of 55 ºC. The results showed that as concentration increases across the three samples, the corrosion rate of stone-coated roofing sheets increases. The analysis of variance (ANOVA) showed that the weight loss of the samples were significant by all input variables. The fourth order polynomial model conducted for weight loss and corrosion showed best fit with regression (R^2) which ranged from 0.95 and 0.99 across three samples except for 1.00 M of sulfuric acid at 0.8953, 0.8862, and 0.8933 for Sample 1, 2 and 3, respectively. Metal dissolution conducted for three hours showed that zinc had highest dissolution across different concentration, followed by iron respectively, aluminum dissolved at 0.75 M sulfuric acid; lead dissolved at 0.25 M, 1 M and distilled water while silicon and cadmium had relatively low dissolution accordingly. Acid rain has negative impact on corrosion stone-coated roofing sheets thus influencing its lifespan and durability.

KEY WORDS: Simulation, Acid rain, Metal dissolution, Corrosion, Stone-coated roof, Nigeria

INTRODUCTION

Since the 19th century, roofing materials is constantly been developed and manufactured to protect building from the impact of climatic elements, which are low cost and durable. Different type of roofing sheets such as corrugated iron, galvanized metals, ceramic tiles, polymer roofs, stone-coated roofs etc., have constantly been used in Nigeria to add beauty to buildings. Nevertheless, over time, roofing sheets degrades in quality compared to its original states due to corrosion. Corrosion is the gradual degradation (or denature) by chemical attack on an exposed surface, often a metal, to produces corrosion deposits [1]. Corrosion is an environmental phenomenon, which creates balance, but negatively has economic implication as it costs industries millions of dollars from coating, painting, galvanizing, or anodizing, etc. According to Awajiogak and Emmanuel [2], corrosion reduces the binding energy of metals thus; it is accelerated by acid rain most especially in oil producing environments. This acid rain is due to oxides of sulfur, nitrogen, carbon and phosphorus that are released into the atmosphere, thereafter combines with rain droplets producing acids that initiate corrosion [3, 4]

Industrialization has solved man's problem but create environmental factors from release of anthropogenic gases such as oxides of nitrogen, sulfates, carbon, etc. into the atmosphere which in-turn react with moisture forming reactive ions as wet deposition. Wet deposition of rainwater leads to corrosion of different metals roofing sheets respectively as carrion deposits referred to as rust and leaching [5]. The levels of metals leached by precipitation from roofing strongly depend on the composition of the roof, atmospheric pollution, temperature variation, age of

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roofing material, residence intensity, and aerial deposition of rainfall [6]. The reaction between roofing sheets and pollutants are very complex and depend on roofing sheet exposure, reactivity and the amount of moisture present. These lead to the deterioration of roofing materials, dissolution and partial oxidation of metals and bioaccumulation of toxic metals in biodiversity [7, 8].

Stone-coated roofing material are made from aluminium; zinc coated steel, acrylic resin protective coatings, natural/ceramic stone chips and acrylic over-glaze [9] as shown in Figure 1. Currently, stone-coated roofing material is currently gaining recognition in the building sector in Nigeria. The study aims to evaluate corrosion rate and metal dissolution from acid corrosion of stone-coated roofing sheet in Nigeria.

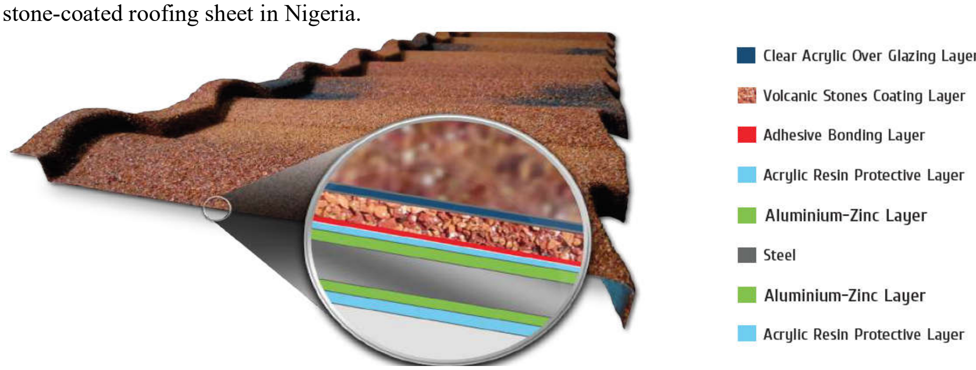


Figure 1. Stone coated sheet [10].

EXPERIMENTAL

Sample preparation

Three different brands of stone-coated roofing sheets were bought from Korban Technology at Aroma in Akwa South Local Government Area of Anambra State, Nigeria. They were assigned sample identity: Sample 1 (S1), Sample 2 (S2) and Sample 3 (S3). Each samples were cut into small sized sheets per brand having dimension of 4 cm × 4 cm and a hole of 2 mm diameter was drilled at the top center of each sheet, cleaned with ethanol to remove dirt, oil and grease and prevent further reaction to sheets.

Reagent preparation

Concentrated sulfuric acid (H_2SO_4) was prepared into different concentration of 0.25, 0.5, 0.75 and 1.00M, simulated acid rain (0.5 M of HCl, HNO_3 and H_2SO_4)and distilled water as reference standard.

Analytical method

The cut sheets were weighed to obtain initial weight, tied with plastic band, attached to retort stand before immersing into 250 mL conical flask containing 100 mL of different concentrations of sulfuric acid and simulated acid rain (0.5 M of HCl, HNO_3 and H_2SO_4), respectively, in controlled environment. The weight loss was determined every three (3) days for 30 days.

Same concentration of sulfuric acid and distilled water was heated to 55 ºC, using water-bath and sustained for three hours to determine metal deposit concentration. After the boiling process, the metal sheets were obtained from the conical flask, washed with running distilled water and bristle brush, air-dried with blow drier and reweighed as final weight. 5 g of solutions from corrosion process was weighed into a digestion flask and 20 mL of acetic acid mixture (650 mL concentrated nitric acid; 80 mL perchloric acid; 20 mL concentrated sulfuric acid) was added. The flask was heated until a clear digest was obtained. The digest were diluted with distilled water to the 50 mL beaker and 5 mL of concentrated nitric acid was added and made up to 100 mL in volumetric flask before transferring into a pre-treated bottles. Six metals (Pb, Fe, Zn, Al, Si, and Cd) were quantified using Agilent atomic absorption spectrophotometer. These were conducted in triplicates for precision measurement.

Statistical method

Microsoft Excel Package 2019 for Windows was used to execute several statistical analysis using different preinstalled Add-ins XRealStats (Fourth order polynomial regression with one-way ANOVA). Graphical presentations were prepared with cluster column using Excel Charts. ANOVA assess significance of two groups. Polynomial regression utilized for non-linear graph relationship between dependent variable y and independent variable x modelled to nth degree. It gives ascertain high degree of accuracy compared to statistical analysis.

Polynomial regression analysis as applied by Oji and Okon [11] were used to model the weight loss and the rates of corrosion of the stone-coated roofing sheets.

Weight loss equation: $y = at^4 + bt^3 + ct^2 + dt + e$ (1)

where y is weight loss; a, b, c, d, e are constant and t is exposure time. The Corrosion rate was determined from equation 1 differentiated to obtain

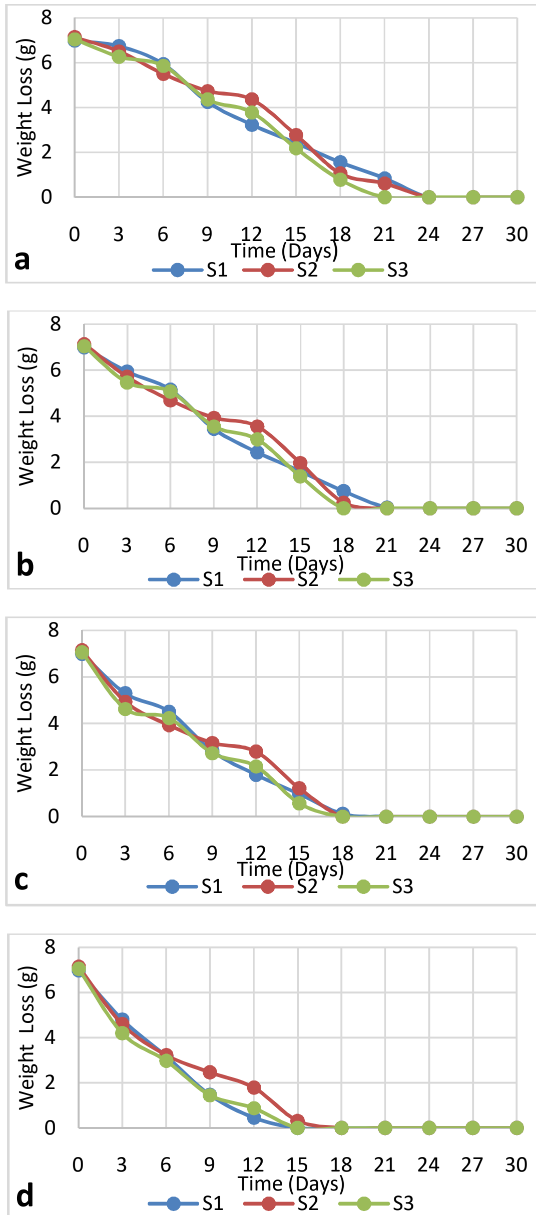
$$R = 4at^3 + 3bt^2 + 2ct + d \quad (2)$$

where R: corrosion rate; a, b, c, d are constant and t is exposure time.

RESULTS AND DISCUSSION

Weight loss from corrosion process

Figure 2a-e shows a line plot of weight loss (g) against time (days) across different concentration of sulfuric acid and simulated acid rain. A survey of each line plot 2a-d showed steady degradation, which is due to the fact that as sulfuric acid (H_2SO_4) concentration increases, the rate and impact of corrosion. A look at 2e for simulated acid rain (hydrochloric acid, nitric acid and sulfuric acid) showed that S1 dissolved on the 15th day while S2 and S3 melted on the 18th day. Sulfuric acid, nitric acid and hydrochloric acid is readily soluble in water as it dissociate into respective ions and free hydrogen, which initiate high reactivity rate with metallic compositions. Sulfur oxides (SO_x), nitrogen oxides (NO_x) and carbon oxides (CO_x) are precursor gases, which causes increase corrosion reaction on roofing sheet from anthropogenic releases [12]. Chlorides gets on stone-coated roofs from mining and seawater releases, which combines with dust particles, settle and form corrosive coating on roofing sheet degrading slowly in presence of rainwater [13]. As these combine together in aqueous medium, the pH decreases leading to creation of active site for hydrogen abstraction by metals ions, which binds readily with dissolved sufate ion or hydroxylion from distilled water used [14].


a
Weight Loss (g)
8
6
4
2
0
0 3 6 9 12 15 18 21 24 27 30
Time (Days)
S1 S2 S3
b
Weight Loss (g)
8
6
4
2
0
0 3 6 9 12 15 18 21 24 27 30
Time (Days)
S1 S2 S3
c
Weight Loss (g)
8
6
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0 3 6 9 12 15 18 21 24 27 30
Time (Days)
S1 S2 S3
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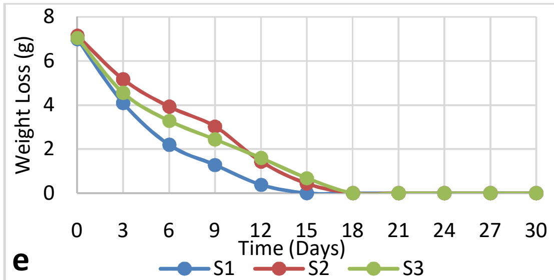


Figure 2. a–e: Line graph for 0.25, 0.50, 0.75 and 1.00 M H_2SO_4; and simulated acid rain (0.5 M HCl, HNO_3, H_2SO_4).

ANOVA and polynomial regression

Table 1 and 2 show ANOVA results and fourth order polynomial model for S1, S2 and S3. ANOVA study showed that various concentration of sulfuric acid and simulated acid rain assessed were significant at $p < 0.05$; regression showed that every concentration of sulfuric acid and simulated acid rain were between 0.95 and 0.99 except for 1 M H_2SO_4 were at 0.8953, 0.8862, and 0.8933 for S1, S2 and S3, respectively. The results were similar to corrosion studies by Oji and Okon [11], which exhibited increasing metal dissolution from corroding acid medium.

Table 1. Analysis of variance (ANOVA) across different concentration of sulphuric acid and simulated acid rain.

0.25 M H_2SO_4	R-square	SE	SS	MS	F-value	*p*-value	Sig.
Sample 1	0.9956	0.2226	55.7886	13.8852	245.9411	4.58E-06	Yes
Sample 2	0.9945	0.2831	57.9612	11.5281	143.8793	0.0001	Yes
Sample 3	0.9924	0.2832	58.36645	11.6091	144.7018	0.0001	Yes
0.50 M H_2SO_4							
Sample 1	0.9964	0.2024	45.1422	8.9957	219.5645	5.75E-05	Yes
Sample 2	0.9778	0.4531	46.2771	9.1729	88.8984	0.0003	Yes
Sample 3	0.9885	0.3208	44.9482	8.8978	77.5024	0.0005	Yes
0.75 M H_2SO_4							
Sample 1	0.9936	0.1614	36.4193	7.2612	256.3827	4.22E-05	Yes
Sample 2	0.9785	0.3794	33.4174	6.6031	65.7180	0.0006	Yes
Sample 3	0.9899	0.2518	31.3404	6.2054	79.2439	0.0004	Yes
1.0 M H_2SO_4							
Sample 1	0.8953	0.5515	25.6604	6.3848	1291.293	1.68E-06	Yes
Sample 2	0.8862	0.2656	25.58116	6.3071	67.8523	0.0006	Yes
Sample 3	0.8933	0.4638	20.3216	5.0468	160.9457	0.0001	Yes
Simulated acid rain							
Sample 1	0.9667	0.4214	26.6482	5.2654	65.5484	0.0006	Yes
Sample 2	0.9785	0.3026	39.7545	7.8957	114.3797	0.0002	Yes
Sample 3	0.9886	0.3531	54.8266	10.8732	94.3707	0.0003	Yes

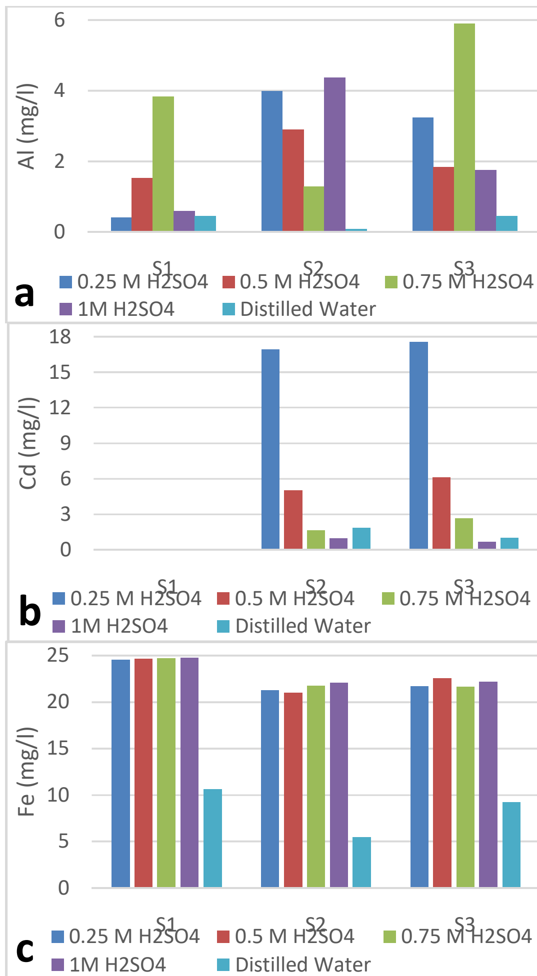
Where R: regression; SE: standard error; SS: sum of square, MS: mean square; Sig: significance.

Table 2. Modelling equations for weight loss and corrosion rate across different concentration of sulfuric acid and simulated acid rain.

0.25 M H_2S0_4	Weight loss equation	Corrosion rate equation
Sample 1	$y = 3.81E\text{-}06x^4 – 0.0003x^3 + 0.0095x^2 – 0.1269x + 0.3187$	$y = 2.77E\text{-}06t^3 + 6.04E\text{-}05t^2 + 0.0004t – 0.4087$
Sample 2	$y = –9.6E\text{-}06x^4 + 0.0008x^3 – 0.0232x^2 + 0.2896x – 1.7884$	$y = –6.1E\text{-}07t^3 + 0.0008t^2 – 0.0323t + 0.0511$
Sample 3	$y = –5.2E\text{-}06x^4 + 0.0004x^3 – 0.0092x^2 + 0.07852x – 0.4904$	$y = –4.6E\text{-}05t^3 + 0.00382t^2 -0.0958t + 0.5059$
0.50 M H_2S0_4		
Sample 1	$y = 8.55E\text{-}07x^4 – 9.8E\text{-}05x^3 + 0.0041x^2 – 0.0676x + 0.0421$	$y = 2.75E\text{-}05t^3 + 0.002t^2– 0.0391t – 0.1211$
Sample 2	$y = –1.2E\text{-}06x^4 + 0.0009 x^3 – 0.0261x^2 + 0.3171x – 1.89601$	$y = –3.0E\text{-}05t^3 + 0.0027t^2 – 0.0686t + 0.3082$
Sample 3	$y = –3.5E\text{-}06x^4 + 0.0002x^3 – 0.0034x^2 – 0.00437x – 0.01143$	$y = –7.2E\text{-}05t^3 + 0.0053t^2 – 0.1202t + 0.6504$
0.75 M H_2S0_4		
Sample 1	$y = 1.92E\text{-}06x^4 – 0.0002x^3 + 0.008x^2 – 0.1242x + 0.3748$	$y = –4.8E\text{-}05t^3 + 0.0324t^2 – 0.0602t + 0.0091$
Sample 2	$y = –8.3E\text{-}06x^4 + 0.0006x^3 – 0.0177x^2 + 0.2097x – 1.3279$	$y = –3.8E\text{-}05t^3 + 0.0029t^3 – 0.0678t + 0.2578$
Sample 3	$y = 9.13E\text{-}07x^4 – 0.0002x^3 + 0.0072x^2 – 0.1359x + 0.6621$	$y = –7.2E\text{-}05t^3 + 0.005t^3 – 0.1054t + 0.4879$
1.00 M H_2S0_4		
Sample 1	$y = 4.77E\text{-}06x^4 – 0.0004x^3 + 0.01206x^2 – 0.1315x + 0.0111$	$y = –1.3E\text{-}05t^3 + 0.0002t^2 + 0.0281t – 0.9006$
Sample 2	$y = –3.4E\text{-}06x^4 + 0.0003x^3 – 0.0063x^2 + 0.0768x – 0.774$	$y = –3.4E\text{-}05t^3 + 0.0022t^2 – 0.0381t – 0.1172$
Sample 3	$y = 2.7E\text{-}06x^4 – 0.0002x^3 + 0.0077x^2 – 0.0901x – 0.0194$	$y = –2.1E\text{-}05t^3 + 0.001t^2 + 0.0004t – 0.5362$
Simulated acid rain		
Sample 1	$y = 1.11E\text{-}05x^4 – 0.001x^3 + 0.031x^2 – 0.42x + 1.9076$	$y = –5.9E\text{-}05t^3 + 0.0035t^2 – 0.0495t – 0.2099$
Sample 2	$y = 6.28E\text{-}06x^4 – 0.0006x^3 + 0.0223x^2 – 0.3437x + 1.7733$	$y = –1.0E\text{-}04t^3 + 0.0066t^2 – 0.1337t + 0.5735$
Sample 3	$y = –5.9E\text{-}06x^4 + 0.0004x^3 – 0.01x^2 + 0.09x – 0.611$	$y = –6.3E\text{-}05t^3+ 0.0047t^2 – 0.1088t + 0.5233$

Metal dissolution

Figure 3 shows the bar chart from metallic dissolution of stone-coated roofing sheets at different sulfuric acid concentration with distilled water as control at elevated temperature for 3 hours. Aluminum had slight increase at 0.75 M H_2SO_4 concentration for S1 and S2 with low dissolution potentials for S3. Cadmium had low dissolution at S1 and high dissolution for S2 and S3 at 0.25 M H_2SO_4. Iron had highly soluble in across different H_2SO_4 concentration respectively. Lead showed increased dissolution in 0.25 M H_2SO_4 for S1 only with sharp decrease and slight increase as concentration progresses across all samples. Silicon dissolved at a low rate indicating less reactivity across the different concentration for S1; S2 and S3 had high reactivity and dissolution across different concentration. Zinc dissolved highest across different H_2SO_4 concentration for S1 to S3.In aqueous medium of sulfate ion, the influence of hydrogen ion (pH), electron potential (Eh), and ionic strength affects the combining prospects for metallic ion at different concentration that may be either at equilibrium or subject to redox chemical transformation, respectively [15]. Metals can react with sulfates ion to form aqueous complexes, which may be precipitated out or undergo further complexing reactions at different oxidation states for the case of aluminum, cadmium, lead, iron, zinc, and silicon ions, respectively [16-18]. Looking at the metal dissolution value of distilled water all analyzed metals have potential to form hydroxyl ion at elevated temperature that indicate that these metals can undergo redox-driven cycles across aqueous medium by adsorption under oxidizing conditions with moderate reduction to oxy-hydroxides respectively [19].



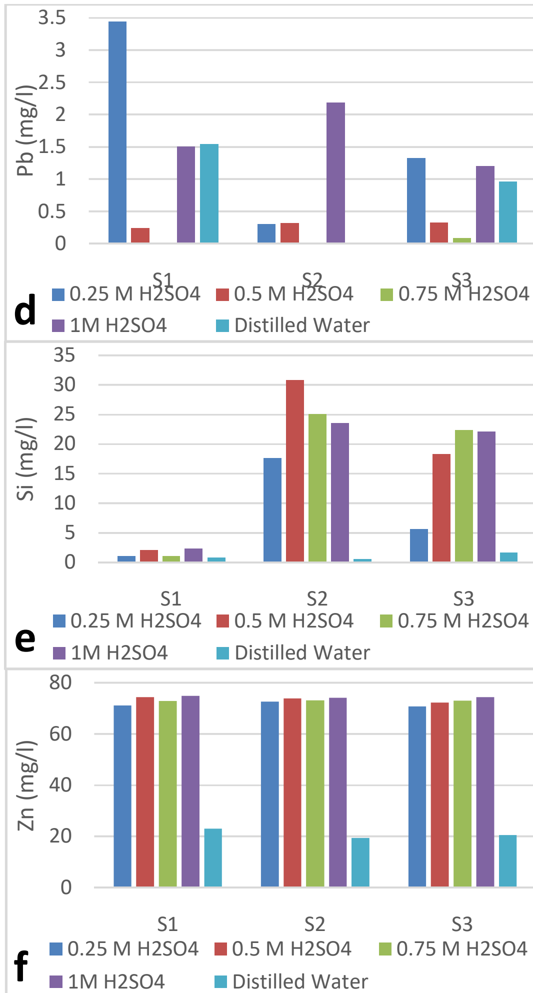


Figure 3. a–f: Bar chart indicating different metal dissolution.

CONCLUSION

From the simulation study, we assessed the impact of acid corrosion on three sampled stone-coated roofing sheets sold in Nigeria and discovered that the stone-coated roofing sheets had increased deposit with grey and brown coloration after several days as ANOVA showed $p < 0.05$ were significant. The extents of attack of the roofing sheet depended majorly on the concentration level of the pollutant. Fourth order polynomial regression conducted on weight loss and corrosion rate ranged from 0.8622 and 0.9956 across various concentration of sulfuric acid and simulated acid rain for a period of 30 days. Metal dissolution of stone-coated samples heated in aqueous medium of sulphuric acid and distilled water showed that elevated temperature impacts on corrosion potential from redox reaction on roofing sheet durability.

Therefore, manufacturing companies' needs to improve on the bonding processes to aid durability of stone-coated roofing sheet as Nigeria atmospheric condition is not favourable for the stone-coated roof material over long period leading to chemical leaching.

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