ABUAD Journal of Engineering Research and Development (AJERD)
ISSN (online): 2645-2685; ISSN (print): 2756-6811

Volume 8, Issue 2, 39-48

Modelling and Simulation of Spray Pyrolytic Graphene and Graphite Coatings on Polymeric Substrates for enhanced Oil and Gas Pipeline Repair Applications

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Received: 05/08/2025
Revised: 29/08/2025
Accepted: 29/09/2025
Available online: 03/10/2025

Abstract: *The oil and gas industry faces significant challenges in pipeline corrosion management, with high corrosion resistance and electrical conductivity for effective cathodic protection (CP). This study focuses on the mathematical modelling of electrical conductivity in nanoparticle-sized graphene, graphite, and graphene-graphite coated polyvinyl chloride (PVC) and glass reinforced polymer (GRP) substrates, prepared via low-temperature spray pyrolysis (50–60 °C). Single- and double-layer coatings with graphene-graphite blend ratios of 1:0.5 and 1:1 were applied on 50 × 10 × 2 mm substrates to obtain coating thickness of 0 µm, 56.1 µm, 77.2 µm, 80.6 µm, 80.8 µm, 92.6 µm, and 97.9 µm respectively for the PVC samples while the GRP samples have coating thickness of 0 µm, 110.3 µm, 114.0 µm, 109.0 µm, 115.7 µm, 117.3 µm, 124.7 µm. Electrical conductivity was measured using an LCR meter, and polynomial models were developed to correlate conductivity with coating thickness. Cathodic protection simulations assessed the performance of coated composites in a 15 km pipeline, highlighting the impact of non-conductive sections and the efficacy of bypass wire designs. Results showed that single-layer graphene-coated GRP achieved the highest conductivity (1.8×10^{-6} S/m), while double-layer hybrid coatings (1-0.5D) offered optimal durability. The mathematical models accurately predicted conductivity trends, with GRP exhibiting superior performance compared to PVC due to better graphene integration. CP modelling revealed that non-conductive GRP sections cause localized underprotection, mitigated by optimized bypass designs. These findings demonstrate that graphene-graphite-coated GRP composites, supported by predictive conductivity models and CP simulations, are promising alternatives to steel for pipeline repairs, enhancing corrosion control and longevity in oil and gas applications.*

Keywords: *Glass Reinforced Polymer, Polyvinyl Chloride, Graphene, Spray Pyrolysis, Simulation*

1. INTRODUCTION

The oil and gas industry relies heavily on carbon steel pipelines for crude oil conveyance due to their strength and electrical conductivity, which are essential for cathodic protection (CP) systems [1]. However, steel's susceptibility to corrosion necessitates frequent maintenance and repair, driving the need for lightweight, corrosion-resistant, and conductive alternatives [2]. Polymeric composites, such as polyvinyl chloride (PVC) and glass reinforced polymer (GRP), offer excellent corrosion resistance but lack the electrical conductivity required for CP and non-destructive testing [3]. Layer modification techniques, particularly spray pyrolysis of conductive nanomaterials such as graphene, have emerged as viable methods to enhance the electrical properties of composites [4, 5].

The study on spray-deposition of graphene/polymer thin coatings on polyimide sheets to reduce lunar dust adhesion reveals that In comparison to conductive and insulating surfaces, graphene-coated polyimide surfaces with charge dissipative resistivity (106 Ω/sq) showed the least amount of residual particle areal coverage [6]. GRP composites coated with graphene, offer a promising, multifunctional alternative to steel in pipeline repair, leak mitigation, and corrosion control applications in the oil and gas industry, according to a prior study on the adaptability of nanoparticle-sized graphene and graphite-coated PVC and GRP (Glass Reinforced Polymers) as substitutes for steel in corrosion control and pipeline rehabilitation {7}. Numerous research projects have been prompted by the need for conductive materials with adjustable mechanical properties in sectors like electronics, automobile, aircraft, and oil & gas. As a result, conductive composite materials have been created to solve the issues these sectors face [11-13]. There is however paucity of literatures with explicit modelling and simulation of the electrical conductivity of coated polymer composites for use in the oil and gas sector.

This study focuses on the mathematical modelling of electrical conductivity in graphene and graphite-coated PVC and GRP substrates, emphasizing their application in oil and gas pipeline repairs. Spray pyrolysis at low temperatures (50–60 °C) was used to deposit nanoparticle-sized graphene and graphite, addressing the challenge of integrating conductive phases into non-conductive polymer matrices [8]. Mathematical models were developed to predict electrical conductivity as a function of coating thickness, while CP simulations evaluated the performance of these composites in pipeline systems. The results provide insights into optimizing composite designs for corrosion control, particularly in retrofitting non-conductive sections of pipelines with conductive coatings to maintain CP efficacy [9,10].

2. METHODOLOGY

2.1 Sample Preparation

PVC and GRP samples (50 × 10 × 2 mm) were used as substrates. The surfaces were roughened with 80-grit sandpaper, cleaned with acetone for 10 minutes, sonicated in distilled water for 10 minutes, and dried at 50 °C for 30 minutes. Coating solutions were prepared by mixing nanoparticle graphene and graphite powders with a polyvinyl alcohol (PVA) binder at graphene-to-graphite ratios of 1:0.5 and 1:1, and then stirred at 80 °C for 4 hours. The compositions are listed in Table 1. A total of fourteen samples were used for each round of testing and this included the control PVC sample, single layer graphene coated PVC sample, double layer graphene coated PVC samples, single layer graphene and graphite coated PVC samples in the ratio 1:1, double layer graphene and graphite coated PVC samples in the ratio 1:1, single layer graphene and graphite coated PVC samples in the ratio 1:0.5, double layer graphene and graphite coated PVC samples in the ratio 1:0.5, control GRP sample, single layer graphene coated GRP sample, double layer graphene coated GRP samples, single layer graphene and graphite coated GRP samples in the ratio 1:1, double layer graphene and graphite coated GRP samples in the ratio 1:1, single layer graphene and graphite coated GRP samples in the ratio 1:0.5, double layer graphene and graphite coated GRP samples in the ratio 1:0.5

Table 1: Various compositions used

Control (PVC, GRP)	1 Layer deposit using graphene (G-S)	2 Layer deposition of graphene (G-D)	Deposition of one Layer graphene + local graphite 1:1 (1-1S)
Deposition of two-layer graphene + Local graphite 1:1 (1-1D)	Deposition of one Layer graphene + local graphite 1:0.5 (1-0.5S)	Deposition of two-layer graphene + Local graphite 1:0.5 (1-0.5D)	

A nebulizer spray pyrolysis technique (NSPT) was employed at 50–60 °C to deposit the coatings. The precursor solution was atomized into droplets, transported via compressed air (3 bars), and sprayed onto the heated substrate, followed by drying at room temperature.

2.2 Electrical Conductivity and Thickness

Electrical resistance was measured using an LCR meter (Figure 1), which has a frequency range of 100–100,000 Hz and ±0.05% accuracy. Conductivity was calculated from resistance values. The LCR (Inductance [L], Capacitance [C], and Resistance [R]) meter (BK 891) (Figure 1) used to evaluate the samples' electrical resistance. It measures dissipation factor, impedance, resistance, and inductance up to 9.999 GΩ, capacitance up to 9999 F, and a frequency range of 20 to 300 kHz. It runs on a fixed or variable frequency with a direct current (DC) voltage of 220 V. The equipment has an accuracy of around ±0.05%. The substance being tested is enclosed in a sample holder and is connected to the LCR meter via Kelvin clips. Keypads are used for inputting measurement variables, and the results, along with their visualization, are displayed on the LED screen. The characteristics derived from the LCR meter may be used to determine the energy storage density, electrical conductivity, dielectric constant, and dielectric loss of a tested material. The electrical conductivity of the developed coating was computed using Equation (1).

$$\sigma = \frac{1}{\rho} = \frac{d}{(R_p)A} \quad (1)$$

Where:
A = area
d = thickness
ρ = resistivity of electrical

The thickness of the coatings was measured by cutting each sample into two halves (perpendicular to its length) using a TechCut 4 low-speed saw from Allied High Tech. Productions Inc. The thickness of the coatings was measured using a digital thickness gauge, and the values were calculated by averaging the three thickness measurements on each sample.

2.3 Mathematical Modelling

Electrical conductivity (EC) as a function of layer thickness (t) was modelled using MATLAB (year). Polynomial equations were derived for both PVC and GRP composites based on experimental data. The models account for the non-linear relationship between coating thickness and conductivity, reflecting the influence of graphene and graphite dispersion on the conductivity.

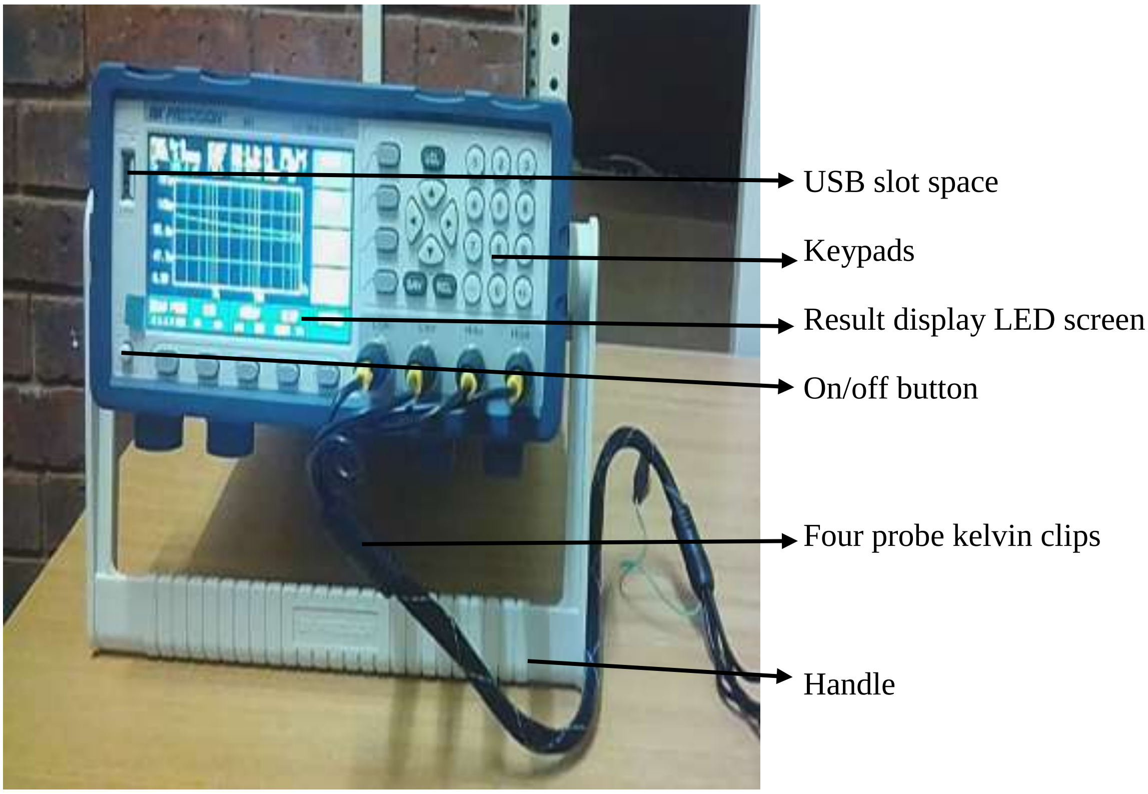

Figure 1: The LCR meter (model BK 891)

2.4 Cathodic Protection Modelling

COMSOL Multiphysics was used to evaluate the impact of sectional pipeline replacement using non-metallic composite materials on CP system performance. This Finite Element Modelling (FEM) approach simulates the electrochemical behavior of a buried steel pipeline before and after the replacement of a section of the line. A 3D model of the pipeline and surrounding soil environment was constructed, and pipeline properties included such as electrical conductivity, coating resistance, soil resistivity and operating potential ranges for effective CP. The pipeline was discretised into multiple segments, with one or more segments defined as non-metallic composites (electrically insulating) to represent the replaced sections. Boundary conditions were applied to simulate galvanic coupling and potential distribution across the pipeline. The simulation analysed the shift in CP potential distribution along the pipeline length, potential shielding or isolation effect introduced by the composite section. By comparing baseline (fully metallic pipeline) and modified scenarios, the model highlights areas where CP current no longer flows effectively past the non-conductive section, potentially exposing downstream steel segments to under protection or corrosion risk. This study seeks to introduce a conductive composite section for future pipeline repairs by surface modification with graphene. To simulate a conductive composite in COMSOL, an electrical bypass wire over the composite section is used.

A 15 km pipeline was simulated with a GRP section at the 6 km mark. The model evaluated electrolyte potential, pipe-to-soil potential, and current density distribution under CP. A bypass conductor was incorporated to mitigate conductivity disruptions caused by the uncoated GRP section. COMSOL was used to model the ICCP behaviour in a pipeline incorporating a GRP segment. The key modelling assumptions and system parameters are as follows:

- Pipeline Length: 15 km
- ICCP Anode Location: Installed at 0 km
- GRP Section Location: 6.0 km to 7.3 km
- Electrical Bypass: An insulated copper wire used to bridge the GRP section
- Soil Conditions: Assumed homogeneous resistivity

Analysis Parameters:

1. Electrolyte potential distribution
2. Pipe-to-soil potential shift
3. Current density before and after GRP integration

The simulation analysed both baseline and modified scenarios to determine the influence of the non-conductive section on CP system performance

Geometry Setup: A 15 km pipeline was modelled as a one-dimensional line, with a GRP section inserted at the 6 km mark. The GRP section was assigned a length of 1.3 km, reflecting a typical repair patch size. A bypass conductor was modelled as a parallel conductive path with a specified resistance of 0.01 Ω/m.

Material Properties Assignment: Steel sections were assigned an electrical conductivity of 4.03×10^6 S/m, while the uncoated GRP section was assigned 10^{-12} S/m. Coated GRP sections were assigned conductivities based on experimental measurements (e.g., 1.8×10^{-6} S/m for G-S). The surrounding electrolyte (soil) was modelled with a resistivity of 100 Ω·m.

Physics and Mesh Configuration: The Electric Currents module in COMSOL was used to simulate current flow and potential distribution. A fine mesh (element size of 0.1 m) was applied at transition zones (steel-to-GRP interfaces) to capture potential gradients accurately, while a coarser mesh (1 m) was used elsewhere to optimize computational efficiency.

Boundary Conditions Application: The anode was modelled as a point source at the pipeline's origin, supplying a constant current of 1 A. The pipe-to-soil potential was set to maintain a protective range of -0.85 V to -1.2 V vs $Cu/CuSO_4$. Grounded conditions were applied at the pipeline's end (Equation 4).

Simulation Execution and Post-Processing: The stationary solver was used to compute steady-state potential and current distributions. Results were extracted along the pipeline length to analyse electrolyte potential, pipe-to-soil potential, and current density, particularly at the GRP section transitions.

This procedure ensured accurate modelling of CP performance, highlighting the impact of uncoated GRP sections and the effectiveness of the bypass conductor design.

The limitations of this model is that rigorous modelling of intricate reactions like calcareous deposit formation in seawater or stray current effect in ICCP is often too complex, so simplified and approximate models are used. Another limitation is that thin electrolyte films in atmospheric corrosion introduce meshing difficulties and increase memory requirements often requiring assumptions like uniform current density to reduce computational load

2.4.1 Governing equations

The cathodic protection simulation was governed by electrochemical principles, specifically Laplace's equation for the electric potential in the electrolyte and Ohm's law for current flow in the pipeline and bypass conductor. The primary governing Equations are:

Laplace's Equation for Electrolyte Potential: $\nabla^2 \phi = 0$ (2)

Where (ϕ) is the electric potential in the electrolyte (soil). This equation assumes a homogeneous, isotropic medium with no charge sources in the electrolyte.

Ohm's Law for Current Density: $\mathbf{J} = -\sigma \nabla \phi$ (3)

Where ($\mathbf{J}$) is the current density (A/m²) and (σ) is the conductivity of the material (S/m). For the steel and coated GRP sections, (σ) was derived from experimental measurements, while the bypass conductor was assigned a high conductivity (10^8 S/m).

Boundary Condition at the Anode: $\mathbf{J} \cdot \mathbf{n} = I_0$ (4)

Where (I_0) is the constant current input (1 A) and ($\mathbf{n}$) is the normal vector to the anode surface.

Pipe-to-Soil Potential: $\phi_{\text{pipe}} - \phi_{\text{soil}} = E_{\text{protection}}$ (5)

Where ($E_{\text{protection}}$) is the target protection potential (-0.85 V to -1.2 V vs $Cu/CuSO_4$).

These equations were solved numerically using COMSOL Multiphysics to determine the potential and current distributions along the pipeline, particularly at the GRP section interfaces.

2.4.2 Assumptions and boundary conditions

The following assumptions were made during the CP modelling:

Note: The assumptions were in line with the COMSOl model industry standard for CP Modelling.

i. **Uniform Electrolyte Properties:** The soil was assumed to have a constant resistivity of 100 Ω·m, neglecting variations due to moisture or composition.
ii. **Steady-State Conditions:** The simulation assumed steady-state current and potential distributions, ignoring transient effects.
iii. **Ideal Bypass Conductor:** The bypass conductor was modelled with a constant resistance of 0.01 Ω/m, assuming negligible degradation over time.
iv. **Homogeneous Coating:** The graphene and graphite coatings were assumed to have uniform thickness and conductivity across the GRP section.
v. **Constant Anode Current:** A constant current of 1 A was supplied by the anode, simulating an impressed current cathodic protection (ICCP) system.
vi. **Anode Boundary:** A fixed current input of 1 A was applied at the pipeline's origin (x = 0 km).
vii. **Pipeline End:** A grounded condition (($\phi = 0$)) was applied at x = 15 km to simulate the return path to the CP system.
viii. **Pipe-to-Soil Interface:** The potential difference was set to maintain a protective range of -0.85 V to -1.2 V vs $Cu/CuSO_4$, based on standard CP criteria.

ix. **GRP Section Interfaces**: Continuity of current and potential was enforced at steel-to-GRP transitions, with the bypass conductor modelled as a parallel path to restore conductivity.

These assumptions and boundary conditions simplified the simulation while maintaining relevance to real-world pipeline CP systems.

3. RESULTS AND FINDINGS

3.1 Mathematical Model of Electrical Conductivity versus Layer Thickness

The plots and equations are obtained by modelling the experimental values of the layer thickness and the corresponding electrical conductivity using MATLAB. Equation 6 and 7 was a result of a mathematical model of the electrical conductivity in relation to the layer thickness of PVC and GRP samples. The plot in Figure 2 (for PVC sample) and Figure 3 (for GRP sample) shows that with an increase in layer thickness of the coating, there is a reduction in the electrical conductivity.

$$IE_{-PVC} = 5.52 * 10^{-7} - 1.12v * 10^{-6} * t + 6.45 * 10^{-7} * t^2 \quad (6)$$

$$IE_{-GRP} = 2.09 * 10^{-6} - 1.94 * 10^{-6} * t + 5.37 * 10^{-7} * t^2 \quad (7)$$

Where; IE: Electrical conductivity (S/m), and t: Layer thickness (μm)

Equation 1 shows a that there is direct proportionality between electrical conductivity and thickness. The models shows that an increase in the layer thickness will also lead to increase in the electrical conductivity when a point is reached. For the PVC sample, the rule of direct proportionality between electrical conductivity and layer thickness starts when the thickness is more than 0.8 μm but for the GRP sample there is a steep fall in the electrical conductivity with increasing thickness, although further increase in thickness will invariably lead to the electrical conductivity to be increased as is the case with the PVC samples

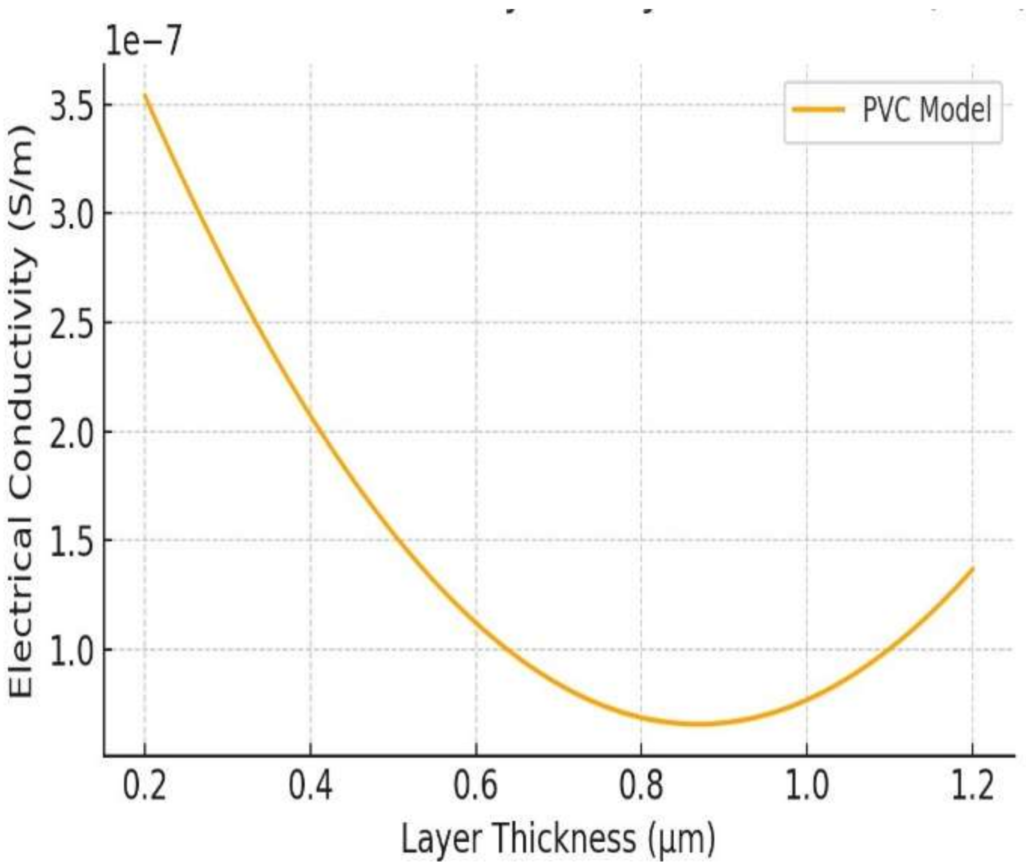

Figure 2: A plot of electrical conductivity as a function of layer thickness for PVC samples

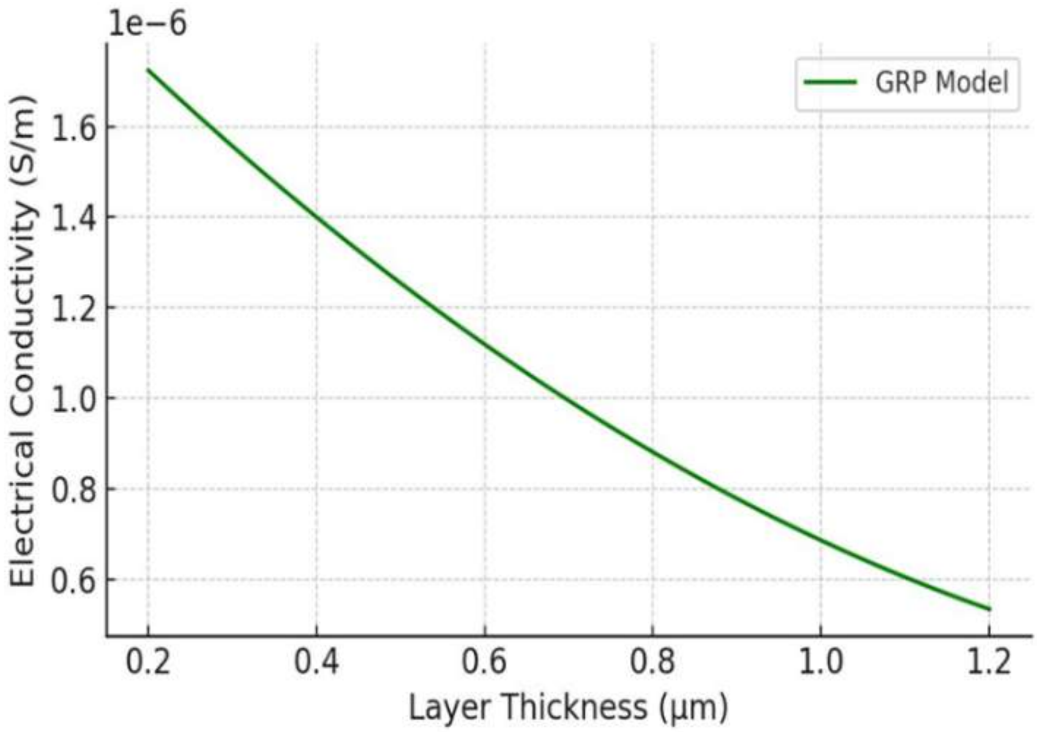

Figure 3: A Plot of electrical conductivity as a function of layer thickness for GRP samples

3.2 Modelling of Cathodic Protection (CP)

The simulation analyzed both baseline and modified scenarios to determine the influence of the non-conductive section on CP system performance. The protected 15 km pipeline was modeled as shown in Figure 4, and the composite section begins at the 6 km point. A conductor cable / bypass was modelled to represent the surface-modified composite.

3.2.1 Electrolyte potential distribution

The electrolyte potential remained relatively stable along the steel portion of the pipeline, up to the 6 km mark, as shown in Figure 7. However, a significant deviation in the potential gradient was observed at the GRP segment, specifically at the 6 km mark. This shift is attributed to the absence of current conduction through the GRP pipe body.

However, the electrical bypass wire (or surface modified composite) successfully restored current flow, but localized disturbances at transition points (entry/exit of the GRP section) suggest areas of possible under protection.

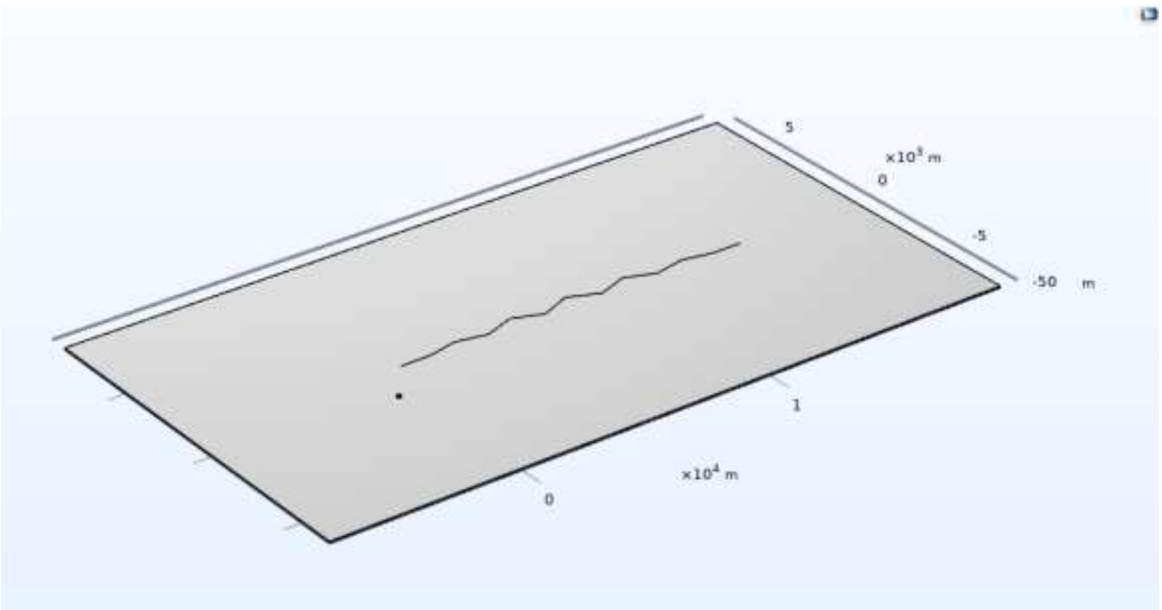

Figure 4: Simulation model of the pipeline

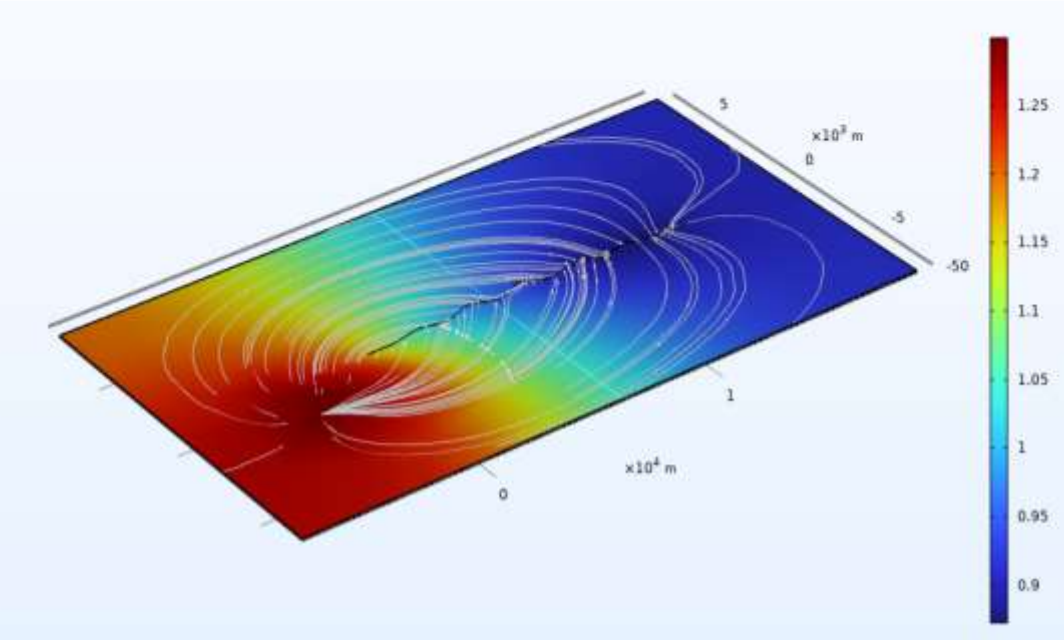

Figure 5: Electrolyte potential around the pipeline under cathodic protection

3.2.2 Pipe-to-soil potential shift

The simulation results showed that the pipe-to-soil potential remained within the protection range (-0.85 V to -1.2 V vs. $Cu/CuSO_4$) in most sections (Figure 5). However, from Figure 7, potential dips were observed at 6 km and 7.3 km, aligning with the start and end of the GRP section. The protection potential in steel decays uniformly the farther away we move from the CP groundbed. The steel pipeline serves as the current return path from the soil, through the pipeline and back to the CP rectifier. However, at the pipeline midpoint, a section has been replaced with composite. The current return path has been interrupted. This isolator (increased resistance) stops the flow of current and hence a dip in the protection potential is observed around the composite section. These dips may fall below the protective thresholds, increasing the likelihood of localized corrosion near GRP transitions if the bypass does not effectively compensate for current loss. The bypass wire is a #14 AWG copper conductor, introduced to allow some current movement between the two isolated sections in the Modelling environment. It is impossible to model a surface modified composite section in COMSOL so the wire bypass is the closest option available in COMSOL to simulate a conductive composite.

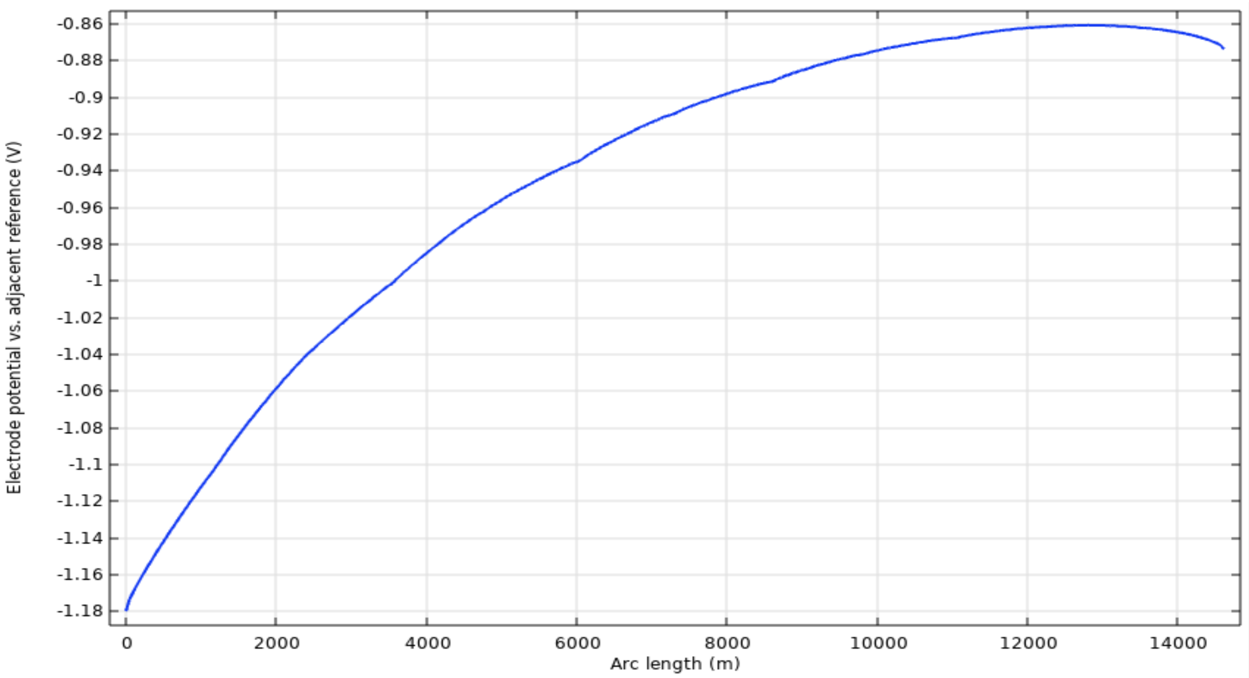

Figure 6: Protection potential chart showing uniform decay

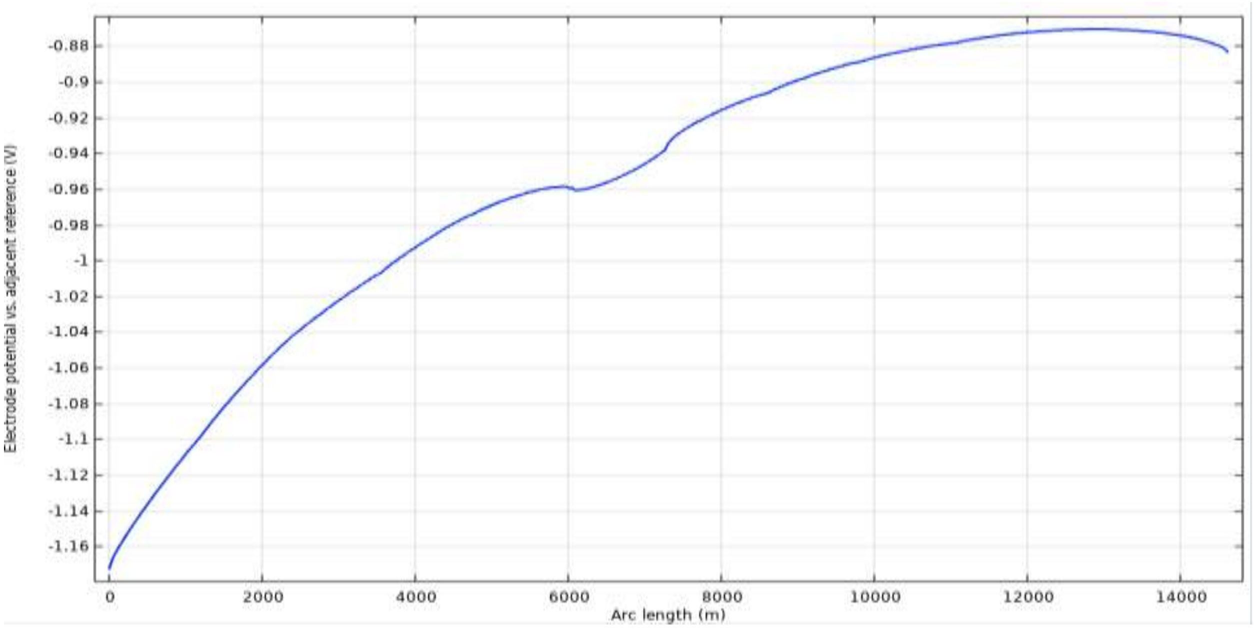

Figure 7: Protection potential chart showing a dip at composite section

3.2.3 Current density distribution

As seen in Figure 8, current density was uniformly distributed before the GRP segment. However, significant fluctuations occurred at the transition zones, indicating non-uniform current redistribution due to the presence of a non-conductive barrier, as shown in Figure 9. The transition zone is the zone between the coated region and the GRP sample at the core. The downstream steel section resumed receiving current after the bypass, but its effectiveness depended on the bypass wire resistance and soil properties. Observation: The current density reduction at transition points may require design improvements to maintain protective levels throughout. The design improvement is the aim achieved by this research by making the composites conductive enough for CP. It should be noted that polymer composites have mechanical properties that are a little less than steel but they are usually suitable for oil and gas operations. Polymer composites can hold the high pressures of typical fluid flows in oil and gas pipelines. So as a replacement candidate for steel, polymer composites are generally busied in the oil and gas industry if it can withstand the pressures required to maintain safe flow of oil or water in the pipes.

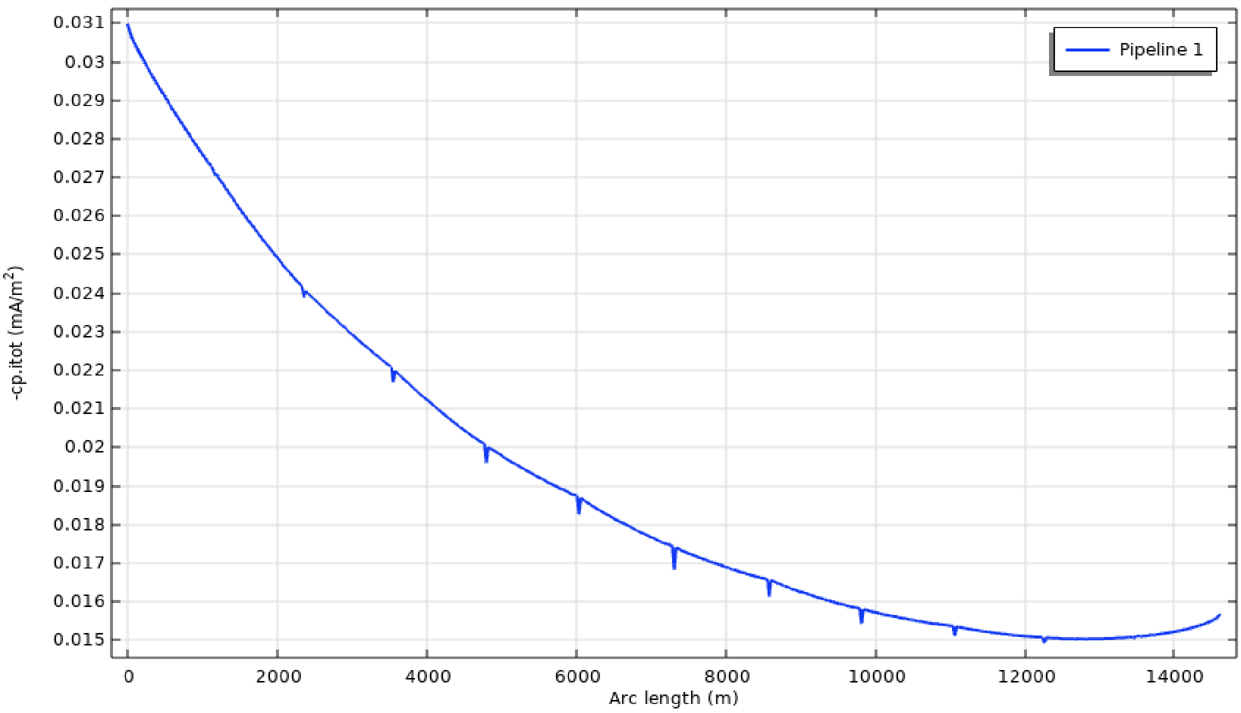

Figure 8: Protection current showing uniform decay

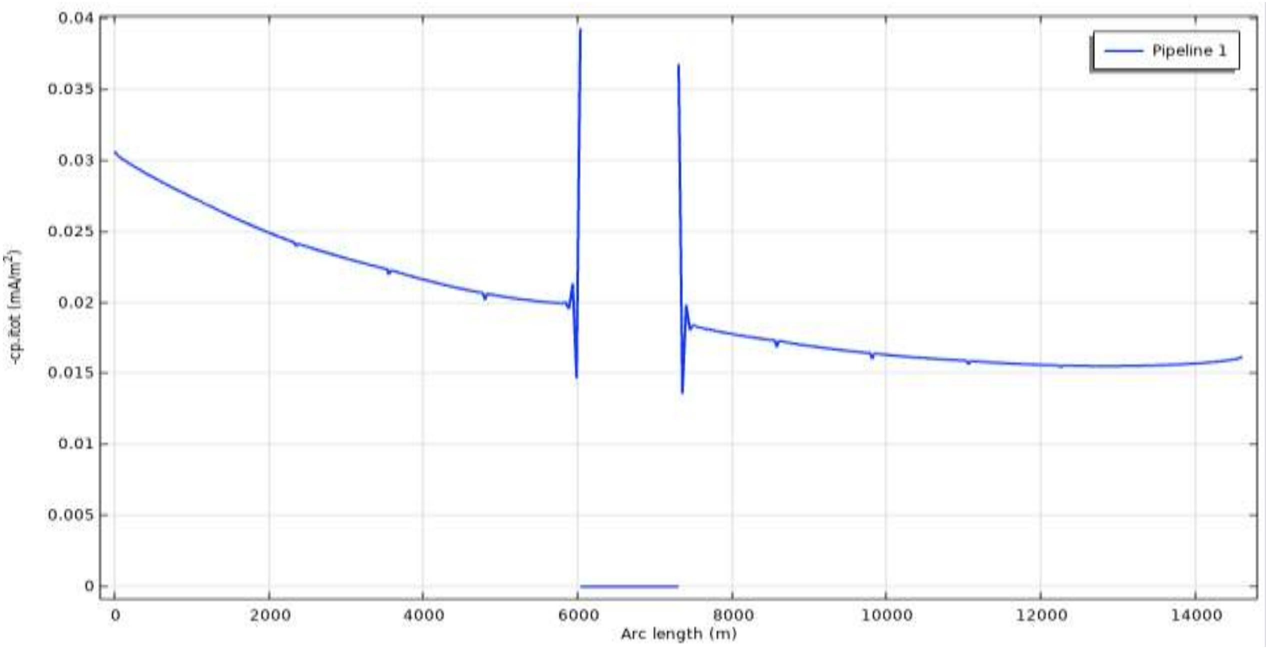

Figure 9: Protection current showing discontinuity /spike at composite section

The charts for the continuous carbon steel pipeline and the pipeline with a conductive composite section are the same. There is continuous current flow and potential distribution on the structure, with no corrosion risks. The corrosion threats introduced by the non-conductive composite section, as indicated by the current density spike and lowered protection potential, are eliminated with the conductive composite solution.

3.3 Application in Pipeline Repairs

The developed conductivity models and CP simulations have significant implications for the repair of oil and gas pipelines. Non-conductive GRP sections, when retrofitted into steel pipelines, disrupt CP systems, as evidenced by potential dips and current density fluctuations. The high conductivity of single-layer graphene-coated GRP (1.8×10^{-6} S/m) enables its use as a conductive patch for pipeline repairs, maintaining CP continuity. The double-layer 1-0.5D coating, with enhanced durability (57.32 HLD hardness), is ideal for high-pressure applications where mechanical stability is critical. The mathematical models provide a predictive tool for optimizing coating thickness to achieve desired conductivity levels, reducing trial-and-error in material design. CP simulations highlight the importance of bypass wire optimization in eliminating under protection risks, thereby ensuring long-term corrosion control in hybrid steel-composite pipelines.

4. CONCLUSION

This study successfully developed mathematical models for predicting electrical conductivity in graphene and graphite-coated PVC and GRP composites, with GRP demonstrating superior performance (1.8×10^{-6} S/m for G-S). CP modelling revealed that non-conductive GRP sections cause localized under protection, which can be mitigated through optimized bypass wire designs. The conductivity models and CP simulations provide a robust framework for designing conductive composite patches for pipeline repairs, offering a lightweight, corrosion-resistant alternative to steel. These findings enhance the feasibility of using GRP composites in oil and gas pipeline rehabilitation, addressing corrosion challenges while maintaining CP efficacy.

ACKNOWLEDGMENT

The authors recognise the support of Afolabi Mathew and Dr. A. Adeyinka-Aderanti in the successful completion of this research paper.

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