

# SELF-HEALING EPOXY/PDMS/GRAPHENE OXIDE NANOCOMPOSITES FOR ANTI-CORROSIVE APPLICATIONS

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This study discusses the synthesis, characterization and development of self-healing nanocomposite of amino-terminated PDMS (Polydimethylsiloxane), Epoxy (EPON828+ Diethylenetriamine (DETA)), and Graphene Oxide (GO). GO was prepared using a modified Hummer's method and was incorporated into the PDMS-Epoxy composite in various ratios (0.1 wt.%, 0.3 wt.%, and 0.5 wt.%) using toluene as the dispersing medium. Fourier Transform Infrared Spectroscopy was used for confirming the presence of the designed/prepared structures, and thermo-mechanical analysis was performed to test the change in glass transition temperature and initiation temperature of self-healing process. The composite resins were coated on mild steel substrates by curing freshly prepared resins over the substrates at elevated temperatures. The corrosion behavior of mild steel in 3.5 wt.% NaCl solution before and after the coatings was studied using Tafel Electrochemical Polarization test. The self-healing properties of the materials were also studied by applying cuts on the material and letting them heal under elevated temperatures, and the results showed that the prepared coating demonstrated an effective corrosion resistance for mild steel for various marine applications.

**Keywords:** corrosion, self-healing materials, PDMS, graphene oxide, coating

## 1. INTRODUCTION

Corrosion is one of the major challenges currently facing the world, with losses of trillions of dollars happening every year across the world (Bowman et al., 2016). One of the most common ways to inhibit corrosion is by coating the material with corrosion inhibiting materials such as paints, other protective

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metals etc. (Zarras and Stenger-Smith, 2014) However, most of the metals used as sacrificial coating have a limited life span because they themselves undergo corrosion over a period of time (Punith Kumar and Srivastava, 2013). Thus, protective coatings which are least susceptible to being corroded themselves and least likely to lead to pit formation are required to give proper corrosion protection to various structures.

Self-healing materials are synthetic materials that can repair a level of damage occurring to them without requiring substantial human intervention. While self-healing materials can be from all classes such as metals, ceramics etc. most self-healing materials are polymer-based materials based on dienes, amines, siloxanes etc. Self-healing materials can work by many mechanisms, such as by releasing healing agents from an internal capsule, reversible cross-linking of polymers, shape memory effect, etc. (Song et al., 2019). Self-healing materials can be used in many applications such as in paints, in coatings, in aerospace equipment etc. (Kirkland et al., 2012). The choice of which type of self-healing material and system to use depends on the requirement of various properties such as mechanical properties like impact resistance, glass transition temperature of the healing agent and the matrix, corrosion protection, electrical properties like conductivity etc.

Graphene and graphene oxide (GO) are being touted as promising materials for inhibiting corrosion of iron-based structures and for increasing their life span considerably (Bekas et al., 2016). Various composites of GO with epoxy and other additives are currently being tested all around the world for their corrosion resistant properties. For example, Madhusudhana et al. (2020) worked on corrosion resistance of GO coated on mild steel with the help of epoxy and determined that GO coating imparted excellent corrosion resistance in the mild steel specimen and that the epoxy covering added some impermeability to gases which added to its corrosion resistance (Madhusudhana et al., 2020). Similarly, Aneja et al. (2017) observed that graphene provided barrier-like properties and protected substrates from wet corrosion by increasing the activation energy required for water to diffuse in (Aneja et al., 2017). However, Graphene oxide may sometimes have minute pores or voids within its structure due to uneven oxidation during its synthesis, and this may result in incomplete corrosion protection (Srinivas et al. 2011). Thus, for complete and feasible corrosion protection, a composite of GO and another protective material can be used (Raghupathy et al., 2017). One of the best methods to achieve this would be to create a composite of GO with a self-healing material. This can provide better corrosion protection to the mild steel substrate from various corrosive media due to the presence of protective layers such as GO and epoxy, and it can also protect it from mechanical scratches and wear due to the self-healing material.

Composites of Self-Healing materials and graphene or graphene oxides have been synthesized and tested for various uses. For example, Rana et al have used capsule based click triggered self-healing material consisting of graphene oxide to combat fatigue in materials (Kargarfard et al., 2018). Similarly, Guadagno et al studied self-healing nanocomposites of epoxy with carbon nanotubes (CNTs) and graphene for aeronautical applications (Guadagno et al., 2018). However, there have not been many studies for the use of these materials for anti-corrosive applications, especially for marine environments.

In this paper, we work on testing the corrosion resistance of epoxy-PDMS (polydimethylsiloxane) self-healing composite coating with and without GO in it when coated on a mild steel substrate. Mild steel was chosen because it is one of the most common alloys used to construct various structures and machineries (de la Fuente et al., 2011). We worked on producing the composites using different ratios of PDMS and GO to test the optimum concentration of PDMS and GO required to provide maximum self-healing and corrosion resistance to the substrate. We also worked on understanding the effect of composition on the temperature of the composite at which self-healing is initiated, so as to understand the suitability of composites of a particular composition in various environments.

## 2. METHODOLOGY

### 2.1. Materials

Potassium permanganate, graphite powder, sulphuric acid (98 wt.%), sodium nitrate, toluene, and hydrogen peroxide (30 wt.%) were all supplied by SRL Pvt. Ltd. Chemicals. DETA (113 g/mol), EPON828 (855.1 g/mol), and PDMS (450–550 g/mol) were supplied by Sigma-Aldrich Chemicals. Water used for all reactions was de-ionized, ultra-pure water produced by double-distillation

### 2.2. Preparation of GO–Epoxy–PDMS composites

Graphene Oxide (GO) was prepared using a modified Hummer's method, as described by (Alam *et al.*, 2017). The self-healing mixtures were formed by mixing epoxy (EPON828 resin with DETA as hardener) with PDMS at different concentrations. The combined weight of DETA+PDMS was equivalent to 12.5 parts per 100 parts of epoxy resin (Ammar *et al.*, 2016). Two sets of self-healing mixtures were created with one set having DETA:PDMS ratio of 80:20 by weight and the other mixture having DETA:PDMS ratio of 60:40 by weight. 1 set each of these self-healing mixtures without any addition of GO was coated onto 1 cm × 1 cm mild steel substrates, by pouring the mixture onto the metal surface and curing it at 150 °C for 5 hours.

Both the self-healing composite mixtures were re-synthesized with different amounts of GO (0.1 wt.%, 0.3 wt.%, and 0.5 wt.%) added. For this, GO was first dispersed in toluene at a ratio of 30 mg/ml by sonicating it for 1 hour at room temperature. 1 g of epoxy resin and 250 mg PDMS (20% of DETA-PDMS mixture) were weighed. To this, the required amount of GO dispersed in toluene was added. Then, 100 mg DETA (80% of DETA-PDMS mixture) was weighed and added to this mixture while stirring continuously. This was stirred until it started becoming viscous, and then it was transferred into a silicone mould. A polished 1 cm × 1 cm mild steel substrate was inserted into the mould to deposit some of the mixture onto it and the liquid mixture was cured at 150 °C for 5 hours. This was repeated for the other ratio with 75 mg DETA and 50 mg PDMS (60:40 DETA-PDMS).

### 2.3. Characterization

The prepared GO sample was characterized using FTIR Spectroscopy (Frontier FT-IR/FIR, Perkin Elmer) to confirm the presence of the desired product. Thermo-mechanical analysis (TA Q-400em) was performed to understand the change in dimension with respect to increase in temperature, helpful to obtain the glass transition temperature. For testing self-healing, a small cut was created in the material using a blade and the material was kept for healing at successively higher temperatures to determine the temperature at which self-healing is initiated.

### 2.4. Corrosion analysis

The prepared samples coated on mild steel substrates were tested by the Tafel electrochemical polarization method using a Potentiostat to obtain their corrosion rate and tendency to corrode in 3.5 wt.% NaCl solution. The sample to be tested with 0.5 cm<sup>2</sup> exposed area was fixed in as the working electrode, with a platinum electrode acting as the counter electrode and AgCl electrode working as the reference electrode.

The Open Circuit Potential (OCP) of the samples was determined by dipping the substrate in the electrolyte for 1 minute without any applied potential and the polarization test was conducted by applying voltage in

a range from  $-9$  V to  $+2$  V at  $25$  mV/s scan rate to obtain the Tafel plots of the samples. Electrochemical polarization test of bare mild steel sample was also conducted in addition to that of the various composite coatings to obtain a reference value of corrosion.

The values of corrosion current density and corrosion potential,  $j_{\text{corr}}$  and  $E_{\text{corr}}$  were obtained from these plots. These values for the coatings were compared to that of the bare mild steel to obtain the corrosion resistance displayed by the various coatings. The corrosion inhibition efficiency (IE) of the composites coated on mild steel was obtained using the following equation.

$$IE = \left( \frac{j_{\text{corr,ms}} - j_{\text{corr,coating}}}{j_{\text{corr,ms}}} \right) \cdot 100 \quad (1)$$

where  $j_{\text{corr,ms}}$  and  $j_{\text{corr,coating}}$  stand for the corrosion current density of the bare mild steel sample and the coating to be studied, respectively.

### 3. RESULTS AND DISCUSSION

#### 3.1. Characterization of prepared materials

The GO sample prepared using modified Hummer's method was characterized with Infrared spectroscopy to determine the presence of GO. Figure 1 shows the FTIR analysis of prepared GO, (Johra and Jung, 2015) spectroscopy depicts peaks at  $3391$   $\text{cm}^{-1}$ ,  $1631$   $\text{cm}^{-1}$ , and  $1384$   $\text{cm}^{-1}$  denoting the presence of O–H bond, C=C bond, and C–O bond, respectively, which strongly indicates the presence of GO.

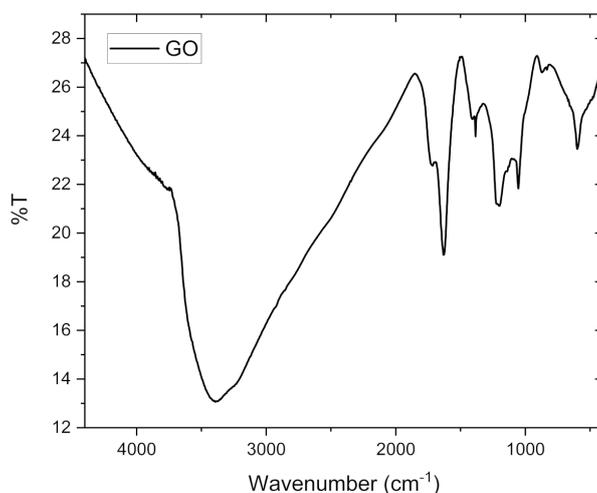


Fig. 1. FTIR Spectroscopy analysis of GO sample

The composites of Epoxy–PDMS and GO–Epoxy–PDMS at various ratios of PDMS and GO were synthesized using the method described in the methodology. The samples were characterized using FTIR spectroscopy to confirm the presence of GO and siloxane groups that provide the self-healing property to the composite. The FTIR spectroscopy images of 80:20 DETA:PDMS ratio epoxy composites with 0.5% and without GO are given in Figure 2(a) and 2(b) respectively. Both the samples with Epoxy–PDMS composite samples and the GO–Epoxy–PDMS composite samples display IR peaks at  $2055$   $\text{cm}^{-1}$  and  $1885$   $\text{cm}^{-1}$  which show the presences of  $\text{CH}_3$  symmetric bending as part of Si– $\text{CH}_3$  bond and Si–O–Si bonds respectively, thus confirms the presence of siloxane groups as shown in Figure 2. However, the samples of GO–Epoxy–PDMS composites display a few extra peaks at  $3598$   $\text{cm}^{-1}$  and  $1652$   $\text{cm}^{-1}$ , indicating the presence of GO (Groza and Surmeian, 2015). The transmittance is comparatively much lower for these samples due to their low transparency.

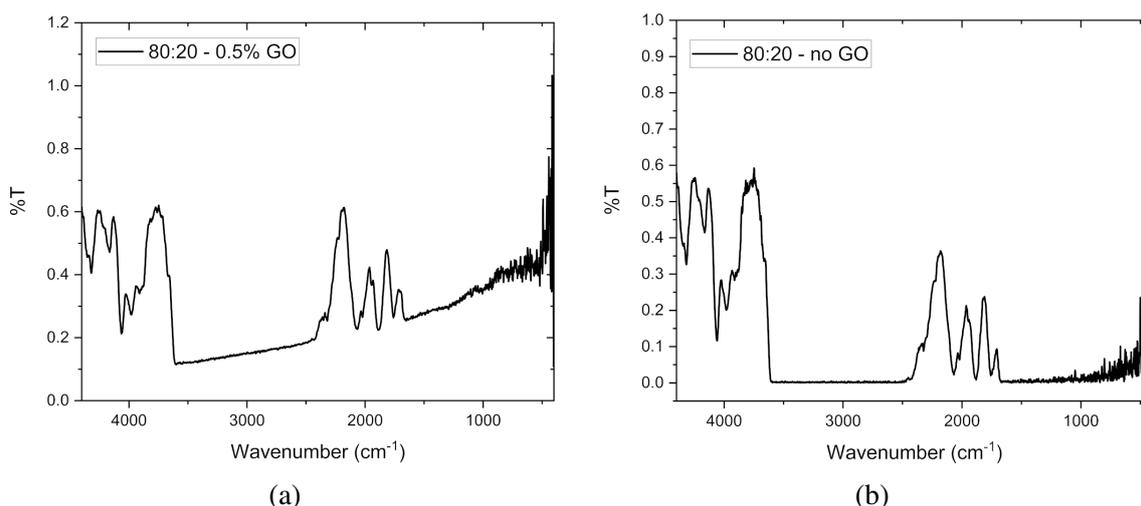


Fig. 2. FTIR spectroscopy images of (a) epoxy composite with 80:20 DETA: PDMS ratio and 0.5% GO and (b) epoxy composite with 80:20 DETA:PDMS ratio and no GO

The glass transition temperatures ( $T_g$ ) of the composites at various temperatures were obtained through a thermo-mechanical analyser, marked by a sharp drop in dimension change at that temperature during thermo-mechanical testing (Keddie et al., 1994). The graphs of deformation change vs. temperature, which depict the  $T_g$  value, for epoxy composites with 80:20 DETA:PDMS ratio and 60:40 DETA:PDMS ratio with various amounts of GO, are given in Figure 3(a) and 3(b) respectively. The  $T_g$  values observed from these are given in Table 1.

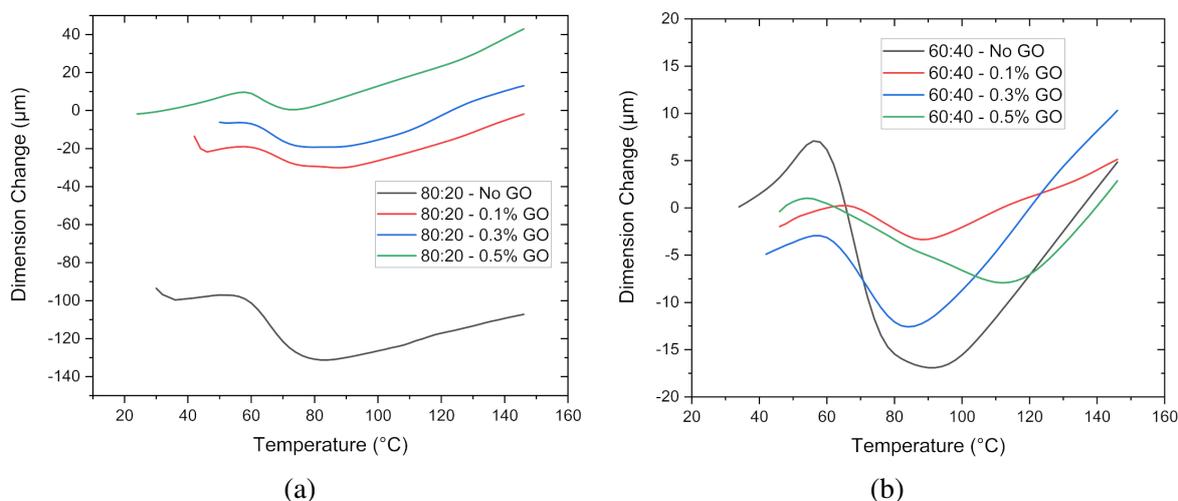


Fig. 3. Thermo-mechanical analysis graphs of (a) epoxy composite with 80:20 DETA:PDMS ratio and (b) epoxy composite with 60:40 DETA:PDMS ratio

Table 1.  $T_g$  values of various composite samples

	0% GO	0.1% GO	0.3% GO	0.5% GO
80:20 ratio of DETA:PDMS	58.23 °C	59.56 °C	60.6 °C	59.19 °C
60:40 ratio of DETA:PDMS	58.74 °C	64.26 °C	64.29 °C	57.59 °C

As observed, the  $T_g$  value first tends to increase with GO content, due to interfacial interactions between GO and the polymer. However, at higher contents of GO,  $T_g$  value decreases drastically, which indicates the occurrence of agglomeration as this prevents interfacial interactions from occurring (Liao et al. 2014).

The  $T_g$  values of samples with 60:40 ratio of DETA:PDMS is slightly higher with minimal amounts of GO, which indicates better interfacial interactions at higher concentration of PDMS.

### 3.2. Self-healing tests

The composites at various ratios of PDMS and GO were made to undergo self-healing test by making a small cut and allowed to heal it at different elevated temperatures. A slight self-healing was observed at lower temperatures, while a complete healing of the cut only took place at much higher temperatures comparatively. The temperature at which complete self-healing of the sample took place for each composite ratio is given in Table 2. Similar to the  $T_g$  values, there is not much difference observed in the self-healing temperature of the samples of various compositions, as mentioned earlier that the changes in composition are too small to have a considerable effect on the self-healing property. However, small changes are observed, with the temperature required for self-healing reducing slightly upon increasing the PDMS ratio and with slight increase of GO content. The temperature required for healing reduced for 0.5% GO for both ratios of PDMS, which could be due to agglomeration of the samples (Krishnakumar et al. 2020). The images of the composite with 60:40 ratio of DETA:PDMS with 0.1 wt.% GO content before and after self-healing are given in Figure. 4(a) and 4(b) respectively.

Table 2. Temperatures at which various composite samples underwent full self-healing

	0% GO	0.1% GO	0.3% GO	0.5% GO
80:20 ratio of DETA:PDMS	110 °C	110 °C	110 °C	100 °C
60:40 ratio of DETA:PDMS	100 °C	110 °C	110 °C	100 °C

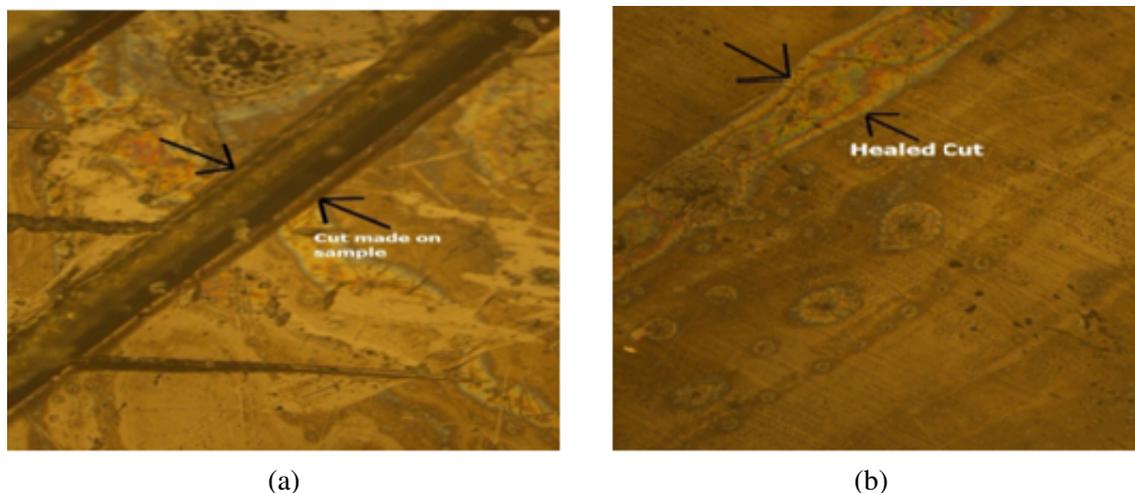


Fig. 4. Epoxy composite with 60:40 DETA:PDMS ratio and 0.1% GO: (a) before self-healing and (b) after self-healing (at 110 °C)

### 3.3. Corrosion analysis

The Tafel Polarization curves obtained for composite coatings with various amounts of GO using the Potentiostat are given in Figure 5. The curve for 60:40 ratio of DETA:PDMS is given in Figure 5(a) and that for 80:20 ratio of DETA:PDMS is given in Figure 5(b). The  $E_{\text{corr}}$  and  $j_{\text{corr}}$  were obtained from these plots by Tafel extrapolation through the use of Tafel Extrapolation extension to Origin by Originlab Technical Support. The values of  $E_{\text{corr}}$ ,  $j_{\text{corr}}$ , and corrosion inhibition efficiency of the various coatings as well as uncoated mild steel sample, obtained using this software, are given in Table 3.

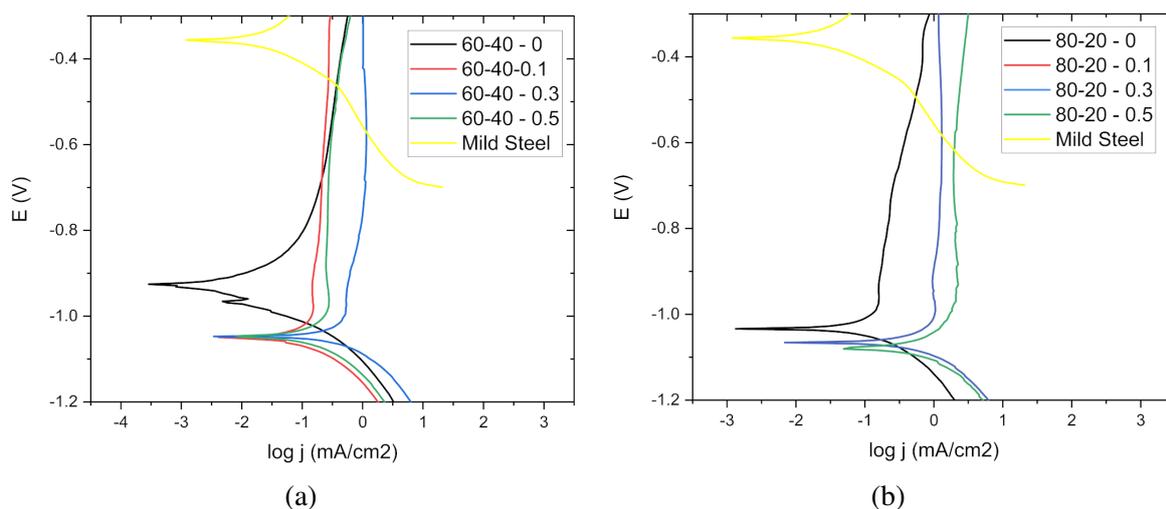


Fig. 5. Tafel Polarization Curves of working electrode potential vs current density depicting (a) self-healing composites with DETA:PDMS ratio of 60:40 with varying amounts of GO coated on mild steel and (b) self-healing composites with DETA:PDMS ratio of 80:20 with varying amounts of GO coated on mild steel. The polarization curve of uncoated mild steel sample is given with both for reference

Table 3.  $E_{corr}$ ,  $j_{corr}$ , and corrosion inhibition efficiencies of various composites prepared

Sample	$E_{corr}$ (V)	$j_{corr}$ (mA/cm <sup>2</sup> )	Inhibition efficiency
60:40 ratio of DETA:PDMS – 0 wt.% GO	-0.925	3.80281	59.08054
60:40 ratio of DETA:PDMS – 0.1 wt.% GO	-1.049	2.55313	72.52747
60:40 ratio of DETA:PDMS – 0.3 wt.% GO	-1.047	2.6066	71.9521
60:40 ratio of DETA:PDMS – 0.5 wt.% GO	-1.046	2.67964	71.16616
80:20 ratio of DETA:PDMS – 0 wt.% GO	-1.033	3.32732	64.197
80:20 ratio of DETA:PDMS – 0.1 wt.% GO	-1.075	2.55902	72.46412
80:20 ratio of DETA:PDMS – 0.3 wt.% GO	-1.082	2.46077	73.52135
80:20 ratio of DETA:PDMS – 0.5 wt.% GO	-1.08	2.40474	74.12419
Uncoated mild steel sample	-0.367	9.2934	0

The  $E_{corr}$  and  $j_{corr}$  values help determine corrosion resistance, as the lower the values, the lower the tendency of the material to corrode (Madhusudhana et al., 2020). Here, it is observed that all of the samples have considerably higher corrosion resistance as compared to uncoated mild steel samples, due to their excellent suitability to be used as corrosion resistant coatings in marine environments and other similar halide environments. It can also be observed that the corrosion inhibition increases on addition of GO, but the amount of GO added has a very negligible effect in the inhibition efficiency of the coating. This shows that the GO acted as a layer between the Epoxy–PDMS composites, thus decreasing the exposed area of mild steel and increasing the corrosion resistance. However, the further increase in GO was too small to have a considerable effect on corrosion resistance, as the corrosion inhibition efficiencies are very similar for all the coatings with GO present in them regardless of the composition. It is also observed from these values that the 80:20 ratio of DETA:PDMS composites have a slightly higher corrosion inhibition efficiency than 60:40 ratio of DETA:PDMS composites, which suggests the possibility of agglomeration at higher PDMS contents.

#### 4. CONCLUSION

This study is focused on the synthesis, development and characterization of self-healing nanocomposites (PDMS, Epoxy, and GO) and the corrosion resistance of mild steel substrates when coated with them. The sample with 60:40 of Epoxy–PDMS with 0.5 wt% of GO showed a better Tg compared to 80:20 with 0.5 wt%, due to the higher amount of GO and PDMS present in the sample. Whereas in the case of self-healing the presence of higher amount of GO (0.5 wt%) in both of the samples (80:20 and 60:40) displayed decrease in the initiation temperature required for self-healing of the composite. Interestingly, the sample with 0.5 wt% of GO in 80:20 showed the highest inhibition efficiency compared with 60:40 composite sample of all the ratios of GO, which makes it a suitable addition for coating on mild steel substrates to provide corrosion resistance. It can also be concluded that while the addition of GO has a positive impact on the corrosion resistance of the composite, further addition of GO provides negligible impact on this, thus making only a slight addition of GO necessary for obtaining good corrosion protection. GO–Epoxy–PDMS composite can be used as an effective corrosion resistant coating for mild steel for various marine applications.

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