



# Corn Oil-Derived Poly (Urethane-Glutaric-Esteramide)/Fumed Silica Nanocomposite Coatings for Anticorrosive Applications

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## Abstract

Poly (urethane-glutaric-esteramide) (PUGEA) was synthesized using corn oil and glutaric acid and further treated with toluene-2, 4-diisocyanate. Fumed silica with 1, 2, and 3 weight percentage was included in the synthesized PUGEA resin to obtain PUGEA-1, PUGEA-2 and PUGEA-3 nanocomposite resins, respectively. The chemical structure of glutaric poly-esteramide and PUGEA was characterized by Fourier-transform infrared spectroscopy and nuclear magnetic resonance (<sup>1</sup>H and <sup>13</sup>C NMR). The morphology, hydrophobicity and thermal stability of the PUGEA nanocomposites were investigated by using scanning electron microscopy and energy-dispersive X-ray spectroscopy, by measuring contact angle and by thermogravimetric analysis (TGA). Afterward, the PUGEA/fumed silica nanocomposite coatings were applied on mild steel and dried at room temperature, and they were physico-mechanically characterized by measuring their scratch and pencil hardness, bending, impact resistance and cross-hatch adhesion. The anti-corrosion effectiveness of PUGEA-2 was investigated by electrochemical impedance spectroscopy immersing it for 42 days in a 3.5 wt% NaCl aqueous solution. Finally, TGA reveals that the PUGEA-3 nanocomposite coating can be safely used up to 275 °C.

**Keywords** Corn oil · Fumed silica · Corrosion · Nanocomposite

## Introduction

Paints and coatings are commonly used to protect both the exterior and interior surface of mild steel. However, in the paint and coating industry, resins made from petroleum-based chemicals cause serious environmental harm and contribute to global warming [1–5]. Sustainable resources are best suited to reduce the environmental damage. Among sustainable resources, vegetable oil is the most important material to develop polymeric resins for paint and coating

[6–11]. In this context, different types of vegetable oils, such as castor, linseed, soybean, jatropha, pongamia, coconut, palm, and corn oil, are used to develop different types of resin, such as polyesters, epoxies, polyesteramide, polyetheramide, and polyurethane, among others [12–17].

Several aliphatic/aromatic acids used for the preparation of polyester, polyesteramide, and polyamide are applied commercially. Glutaric (pentanedioic) acid is a di-functional carboxylic acid used in several applications, such as fiber formation, phase change materials, photonic devices, superconductors, semiconductors, and in biodegradable, thermally stable, or corrosion-resistant materials [18, 19]. Polyurethanes are important materials for applications in coating, fibers and elastomers because of their excellent abrasion resistance, flexibility, mechanical properties and corrosion resistance [5].

The main disadvantage of polyesteramide films is that they provide a track for corrosive ions to reach the mild steel surface after immersion. The permeability of polyesteramide films is the main reason for substrate corrosion. Thus, polyesteramide resins are modified by urethane and by incorporating nanoparticles to decrease the film permeability to corrosive ions [20, 21]. The size and orientation of

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the nanoparticles affect the effectiveness of the barrier of the coated films. Different nano- and microparticles are used to improve the anticorrosive, physico-mechanical and antibacterial properties, such as silver (Ag) [22], iron oxide ( $\text{Fe}_2\text{O}_3$ ) [23], titanium oxide ( $\text{TiO}_2$ ) [24], zinc oxide ( $\text{ZnO}$ ) [15], silicon oxide ( $\text{SiO}_2$ ) [25–28], and carbon filler [29], among others [30]. Fumed silica has a superhydrophobic nature, high flexibility, and uniform dispersion; because of these, it can improve the mechanical, hydrophobic, and thermal properties of polymeric coatings for metallic substrates, and increase their impermeability to corrosive ions.

In this study, poly (urethane-esteramide) is prepared using corn oil-based diol by amidation of corn oil, esterification with glutaric acid and urethanation with toluene-2, 4-diisocyanate (TDI) [17]. During the synthesis of polyurethane, fumed silica is added to develop polyurethane nanocomposites. The prepared glutaric polyesteramide (GPEA) and poly (urethane-glutaric-esteramide) (PUGEA) are characterized by Fourier-transform infrared spectroscopy (FTIR), nuclear magnetic resonance ( $^1\text{H}$  and  $^{13}\text{C}$  NMR) and physicochemical analysis. The developed PUGEA/fumed silica nanocomposites are applied on mild steel panels and their coating properties are evaluated by standard methods. Their hydrophobicity is measured from their contact angle and their thermal behavior is characterized by thermogravimetric analysis (TGA). Corrosion studies of the PUGEA-2 nanocomposite are carried out by electrochemical impedance spectroscopy (EIS) in a 2.5 wt% NaCl solution at selected time intervals for up to 42 days at room temperature.

## Materials

Corn oil (Afia International Company, Jeddah, Saudi Arabia), glutaric acid (Sigma Aldrich, USA), toluene-2, 4-diisocyanate (Acros Organic, USA), diethanolamine (Riedel-deHaen, Germany), methanol, toluene, sodium (BDH Chemicals, Ltd. Poole, England), and fumed silica (surface area:  $395 \pm 25 \text{ m}^2/\text{g}$ , size:  $0.007 \mu\text{m}$ , refractive index: 1.46) (Sigma Aldrich, USA) were used to synthesize PUGEA.

### Synthesis of *N, N*-bis (2-hydroxy ethyl) Corn Oil Fatty Amide

*N, N*-bis (2-hydroxy ethyl) corn oil fatty amide (HECA) was synthesized according to our previous work [17].

### Synthesis of Glutaric Polyesteramide

2.0 mol of HECA and 2.0 mol of glutaric acid were dissolved in xylene and mixed with a magnetic stirrer in a four-neck round-bottomed flask placed on a hot plate. The flask was fitted with a Dean-Stark trap, thermometer, and

nitrogen inlet tube. The reactants were heated to  $160 \text{ }^\circ\text{C}$  and stirred until the removed water was fully collected in the Dean-Stark trap. The acid value was measured at regular time intervals to monitor the evolution of the reaction. After the required acid value was obtained, the solvent was evaporated in a rotary vacuum evaporator under reduced pressure to obtain GPEA.

### Synthesis of Poly (Urethane-Glutaric-Esteramide)

For the preparation of PUGEA, 10 g of GPEA dissolved in 20 mL of xylene were kept in a four-neck round flask, adding 12 wt%, 14 wt%, or 16 wt% TDI and mixing with a magnetic stirrer on a hot plate. The flask was fitted with a cold water condenser, thermometer and nitrogen inlet tube. The reactants were heated to a temperature of  $125 \text{ }^\circ\text{C}$ . The progress of the reaction was monitored by measuring the hydroxyl value and by thin-layer chromatography (TLC). The solvent was evaporated by a vacuum evaporator to obtain PUGEA.

### Synthesis of Fumed Silica-Modified Poly (Urethane-Glutaric-Esteramide)

PUGEA prepared with 14 wt% TDI was loaded with fumed silica before the urethane reaction. 15 g of GPEA were liquefied in 20 mL of xylene, and fumed silica was added at room temperature in different percentages (1 wt%, 2 wt%, and 3 wt%) to obtain PUGEA-1, PUGEA-2, and PUGEA-3, respectively. The compound was stirred for 30 min and then TDI (14 wt%) was added drop-wise. The flask was fitted with a cold water condenser, Dean-Stark trap, thermometer, and nitrogen inlet tube. The compound was heated to a temperature of  $125 \text{ }^\circ\text{C}$ , and the progress of the reaction was monitored by measuring the hydroxyl value and by TLC at regular time intervals. The solvent was evaporated by a vacuum evaporator; by measuring the drying time, it was found that the GPEA nanocomposite with 3 wt% fumed silica showed the best coating properties. The PUGEA-3 resin was dissolved in a 40% solution of xylene and brushed on mild steel panels. The coated panels were dried at room temperature for 15 days to complete the curing reaction. At this stage, dry to hard further subjected to physico-mechanical and corrosion tests were carried out on the coatings.

## Characterization

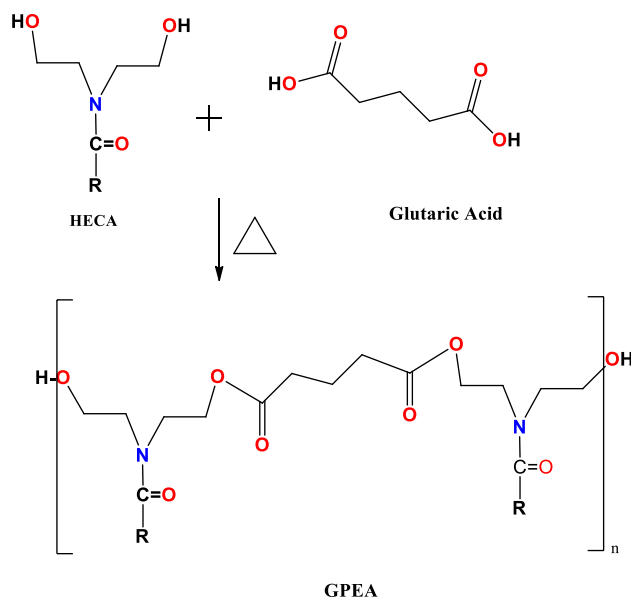
GPEA and PUGEA resins were analyzed with an FTIR spectrophotometer (Spectrum 100, Perkin Elmer Cetus Instrument, Norwalk, CT, USA), a  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR spectrometer (Jeol DPX400MHz, Japan) using deuterated chloroform ( $\text{CDCl}_3$ ) as solvent and tetramethylsilane (TMS) as internal standard. Thermal analysis of the PUGEA-1, PUGEA-2 and PUGEA-3 nanocomposites was carried out

by TGA (Mettler Toledo AG, Analytical CH-8603, Schwerzenbach, Switzerland) in a nitrogen atmosphere and a heating rate of 10 °C/min.

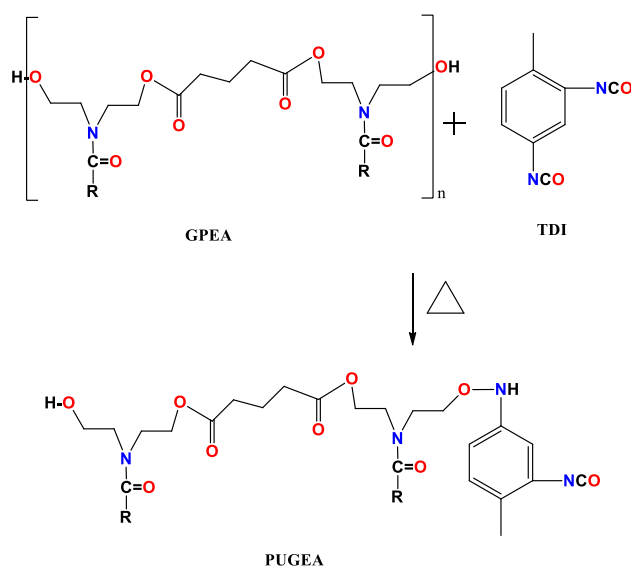
The morphology of the PUGEA-2 coating was studied by scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDX) (JEOL, JED-2200 Series, Japan). Physico-chemical characterization of the GPEA, PUGEA, and PUGEA/fumed silica nanocomposites was carried out by measuring the acid (ASTM D555-61) and hydroxyl (ASTM D1957-86) value. The coating properties of the PUGEA nanocomposites were assessed by measuring their refractive index (ASTM D1218), scratch hardness (BS 3900), adhesion crosshatch (ASTM D3359-02), pencil hardness (ASTM D3363-05), gloss (KSJ MG6-F1, KSJ Photoelectrical Instruments Co., Ltd. Quanzhou, China), and thickness (ASTM D 1186-B), and by carrying out impact (IS 101 part 5 s – 1,1988) and flexibility/bending (ASTM D3281-84) tests. The hydrophobicity was measured from the contact angle by a CAM 200 Attention goniometer. EIS studies on the corrosion of the PUGEA-2 nanocomposites were performed in a three-electrode system at room temperature in a corrosive medium (3.5 wt% NaCl) with a PUGEA-2-coated mild steel sample as the working electrode. The area of the exposed surface was fixed to 1.0 cm<sup>2</sup> using a PortHoles electrochemical sample mask. We used a platinum counter electrode, a KCl-filled silver reference electrode, and an Autolab potentiostat/galvanostat (PGSTAT204-FRA32) with the NOVA 2.0 software (Metrohm Autolab B.V. Kanaalweg 29-G, 3526 KM, Utrecht, the Netherlands). EIS measurements were carried out in the 10–10<sup>5</sup> Hz frequency range, with an amplitude of the sinusoidal voltage of 10 mV. The data of the impedance were analyzed and fitted with the NOVA 2.0 software. Each corrosion test was repeated three times to ensure reproducibility.

## Results and Discussion

Schemes 1 and 2 display the synthesis of GPEA and PUGEA by chemical reaction of HECA and glutaric acid. TDI was further added to form PUGEA; then, 1 wt%, 2 wt%, or 3 wt% fumed silica was added to the PUGEA. The coating properties were found to be deteriorated for ratios of the fumed silica above 2 wt%. The acid and hydroxyl values decreased when going from GPEA to PUGEA-3, being the former 25, 18, 14, 13, and 11, and the latter 250, 57, 55, 54, and 51 for GPEA, PUGEA, PUGEA-1, PUGEA-2, and PUGEA-3, respectively. The respective refractive index values were 1.5077, 1.5176, 1.5206, 1.5214, and 1.5216, showing a sensible increase with an increasing amount of fumed silica. The successful preparation of GPEA and PUGEA was confirmed by FTIR, <sup>1</sup>H NMR and <sup>13</sup>C NMR spectral analysis. We found that PUGEA resins are soluble in different organic solvents, such as toluene,



Scheme 1 Synthesis of GPEA



Scheme 2 Synthesis of PUGEA

xylene, dimethyl sulfoxide (DMSO), dimethylformamide (DMF), tetrahydrofuran (THF), and chloroform; sparingly soluble in methanol and ethanol; and insoluble in water. The PUGEA organic coating was cured by autoxidation [17].

## Spectral Analysis

### GPEA, FTIR (cm<sup>-1</sup>)

3404.39 (OH); 3008.39 (–CH=CH–); 2927.27 (CH<sub>2</sub>, asymmetrical); 2955.06 (CH<sub>2</sub>, asymmetrical); 1738.68 (C=O,

ester); 1628.53 (C=O, amide); 1465.52 (C–N, stretching); 1171.09, 1056.59 (–C(CO)O–C–, stretching); 773.15 (CH, bending) (Fig. 1).

#### PUGEA, FTIR (cm<sup>-1</sup>)

3296.04 (N–H, stretching); 3008.38 (–CH=CH–); 2925.96 (CH<sub>2</sub>, asymmetrical); 2855.02 (CH<sub>2</sub>, symmetrical); 2273.88 (free NCO); 1733.33 (C=O, ester); 1636.90 (CO, amide); 1464.78 (C–N, stretching); 1226.41, 1068.34 (–C(CO)O–C–, stretching); 1537.86, 880.52, 768.34 (aromatic); 729.15 (NH, bending) (Fig. 1). PUGEA-2 showed additional bands at 1057.30 and 1067.19 (O–Si–) [14, 31].

#### GPEA, <sup>1</sup>H NMR (CDCl<sub>3</sub>,δ)

0.756–0.770 (–CH<sub>3</sub>), 1.148–1.192 (–CH<sub>2</sub>–), 1.491 (–CO–CH<sub>2</sub>–CH<sub>2</sub>–), 1.905–1.943 (–CH<sub>2</sub>–CH=CH–), 2.177–2.434 (–CO–CH<sub>2</sub>, chain–, –CO–CH<sub>2</sub>–CH<sub>2</sub>–CH<sub>2</sub>–COO–), 2.643–2.655 (–CH=CH–CH<sub>2</sub>–CH=CH–), 3.384–3.620 (–N(–CH<sub>2</sub>–CH<sub>2</sub>–OH, –CH<sub>2</sub>–CH<sub>2</sub>–O–CO), 4.093–4.926 (OH), 5.224–5.252 (–CH=CH–) (Fig. 2).

#### PUGEA, <sup>1</sup>H NMR (CDCl<sub>3</sub>,δ)

7.967 (NH, urethane), 4.245–4.301 (–CH<sub>2</sub>–CONH), 3.635–3.678 (>N–CH<sub>2</sub>–CH<sub>2</sub>–CONH), 7.211–7.389 (aromatic, H–) (Fig. 3).

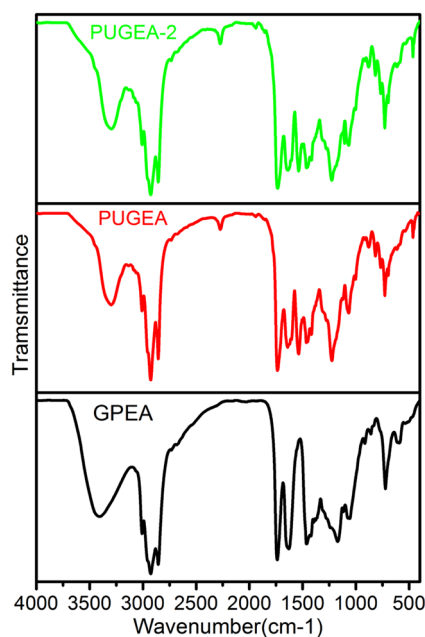


Fig. 1 FTIR spectra of GPEA, PUGEA, and PUGEA-2

#### GPEA, <sup>13</sup>C NMR (CDCl<sub>3</sub>,δ)

13.780–13.818 (–CH<sub>3</sub>), 19.859 (CO–CH<sub>2</sub>–CH<sub>2</sub>CH<sub>2</sub>CO–), 22.384 (–CH<sub>2</sub>–CH<sub>3</sub>), 24.711–25.035 (>NCO–CH<sub>2</sub>–), 25.331–26.895 (–CH<sub>2</sub>–CH=CH–), 28.812–29.461 (–CH<sub>2</sub>–CH<sub>2</sub>–CH<sub>2</sub>–), 31.216–34.030 (CH<sub>2</sub>, OCO–CH<sub>2</sub>–CH<sub>2</sub>–, –CH<sub>2</sub>–CH<sub>3</sub>), 45.030–47.364 (>N–CH<sub>2</sub>–CH<sub>2</sub>–OH), 50.636 (>N–CH<sub>2</sub>–CH<sub>2</sub>–OH), 59.869–61.691 (>N–CH<sub>2</sub>–OH, >N–CH<sub>2</sub>–CH<sub>2</sub>–O–CO–), 127.591–129.833 (–CH=CH–), 172.561 (CO, ester), 176.297 (>CO, amide) (Fig. 4).

#### PUGEA, <sup>13</sup>C NMR (CDCl<sub>3</sub>,δ)

163.492 (CONH), 158.440 (free NCO), 58.645 (–CH<sub>2</sub>CONH), 45.863 (>N–CH<sub>2</sub>–CH<sub>2</sub>–CONH), 124.892–130.033 (aromatic, C) (Fig. 5).

#### SEM and EDX Measurements

Figure 6 displays a SEM–EDX micrograph of the surface of PUGEA-2 and PUGEA-3. As observed, all the fabricated PUGEA coatings have a uniform, homogeneous close-packed structure adhering to the mild steel panel, leading to good physico-mechanical, anticorrosive, hydrophobic and thermal properties after the incorporation of fumed silica. In EDX analysis, the weight percentage of fumed silica on the coating surface of PUGEA-2 and PUGEA-3 was measured 2.07 and 2.89, respectively, which are consistent with the amount of fumed silica added to the composites. The surface morphology analysis of the PUGEA-2 and PUGEA-3 coatings evidences that the silica nanoparticles are uniformly distributed over the whole surface.

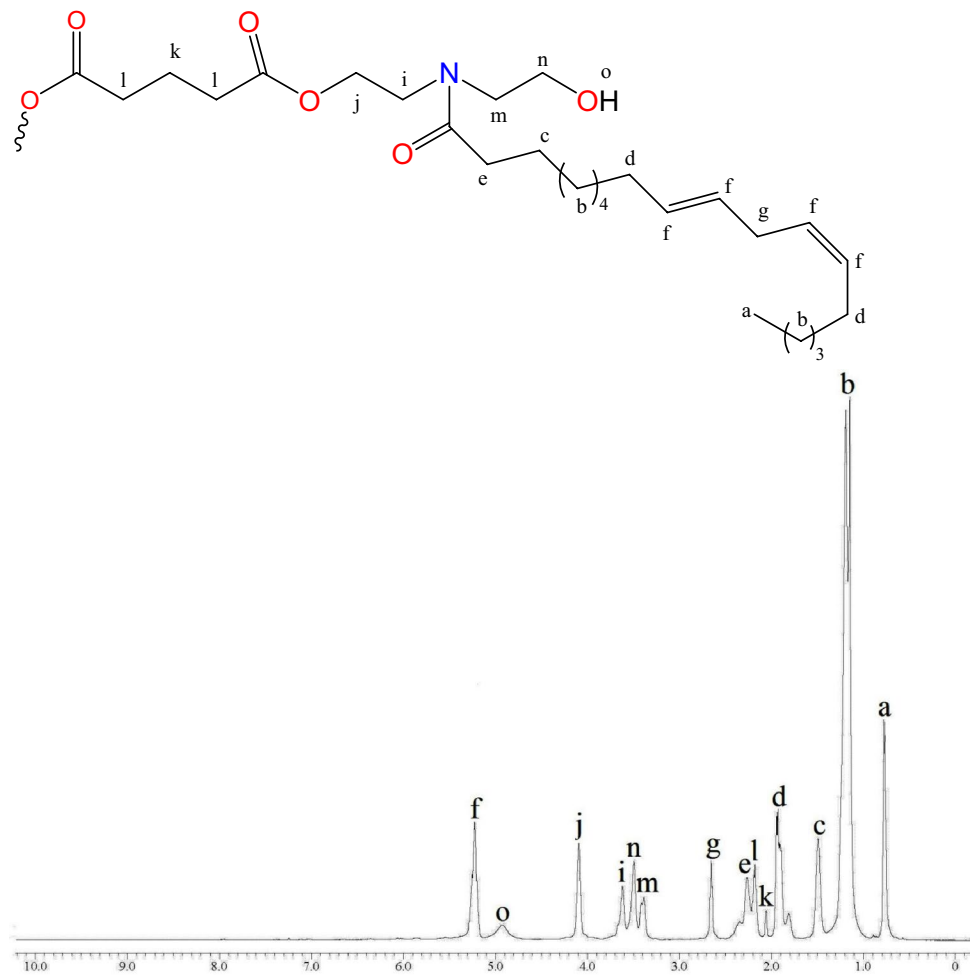
#### Contact Angle Measurements

As seen in Fig. 7, the contact angle of PUGEA was 86°. From the results, it is observed that the contact angle increases with an increase of the fumed silica. Amongst all the PUGEA nanocomposite coatings, PUGEA-3 has the highest contact angle, with a value of 108°. Thus, the improved hydrophobicity of the PUGEA coatings is due to the inclusion of fumed silica, which reduces their surface roughness. The high hydrophobicity of the PUGEA-3 coating makes it a good candidate to be used as an anti-corrosion coating material.

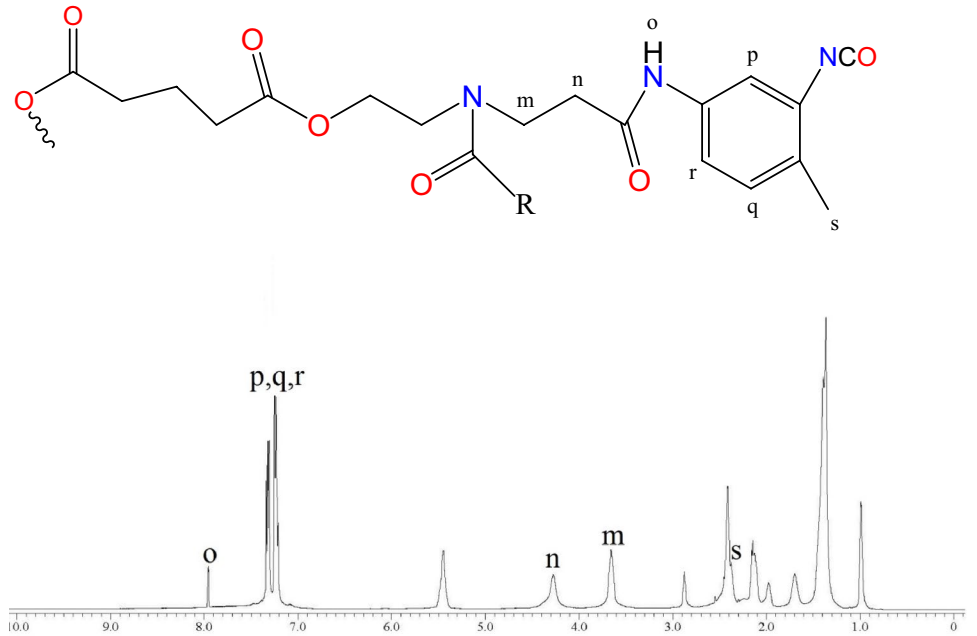
#### Physico-Mechanical Characterization

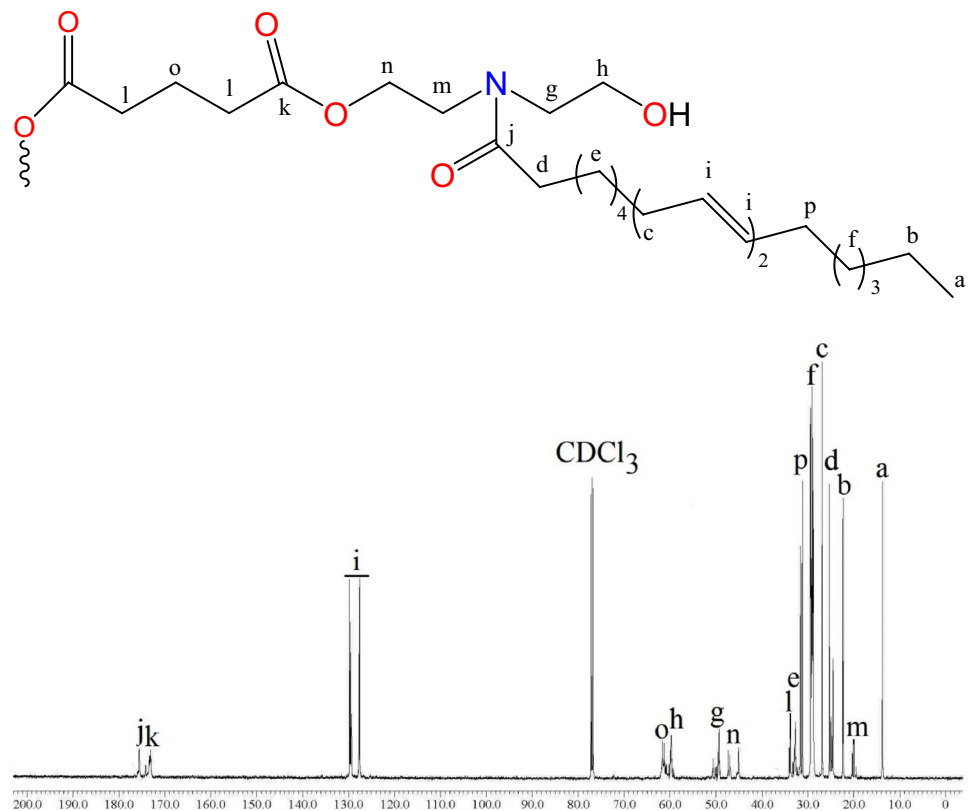
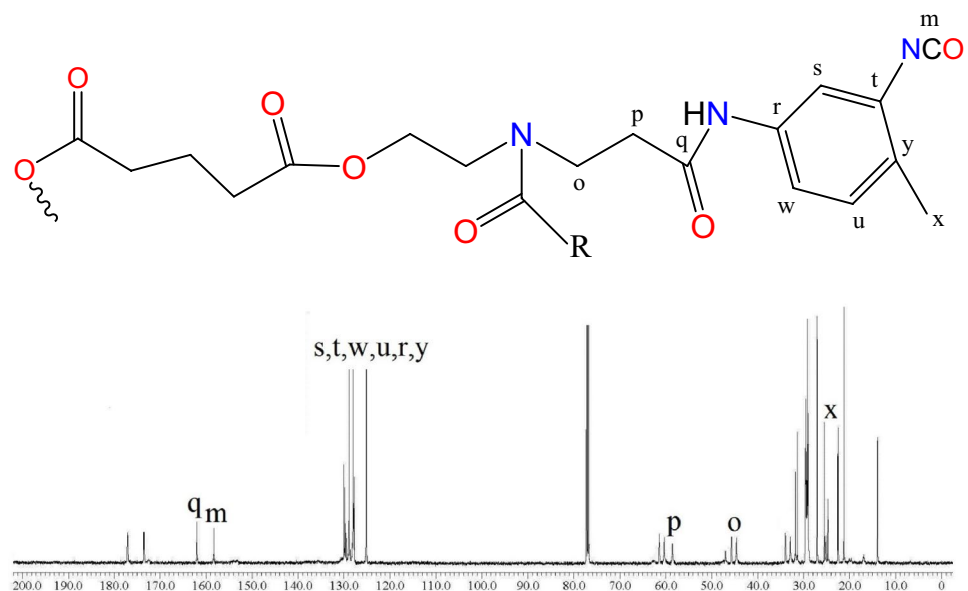
PUGEA coatings were applied on mild steel panels and left undisturbed to dry at room temperature. Curing of PUGEA occurs in three stages. The first stage is a physical process in which evaporation of solvent occurs. The second and third

**Fig. 2**  $^1\text{H}$  NMR spectrum of GPEA



**Fig. 3**  $^1\text{H}$  NMR spectrum of PUGEA



**Fig. 4**  $^{13}\text{C}$  NMR spectrum of GPEA**Fig. 5**  $^{13}\text{C}$  NMR spectrum of PUGEA

stages involve chemical processes wherein PUGEA chains undergo curing by atmospheric moisture at urethane groups and auto-oxidation at double bonds of fatty amide backbone, also leading to cross-linking of PUGEA chains. The coating thickness of PUGEA is  $175 \pm 5 \mu\text{m}$  regardless of the content of fused silica. The values of the scratch hardness are 2.0, 2.6, 2.9, and 2.8, and those of the pencil hardness

are 1H, 2H, 5H, and 4H for PUGEA, PUGEA-1, PUGEA-2, and PUGEA-3, respectively. The observed decrease in these values for PUGEA-3 means that the 2 wt% loading is the best. Gloss values at  $45^\circ$  of 70, 79, 87, and 90 were found for the four samples. All the coatings passed the impact resistance ( $150 \text{ lb inch}^{-1}$ ), bending (1/8 inch), and cross-hatch (100%) tests, with the PUGEA-2 coating showing

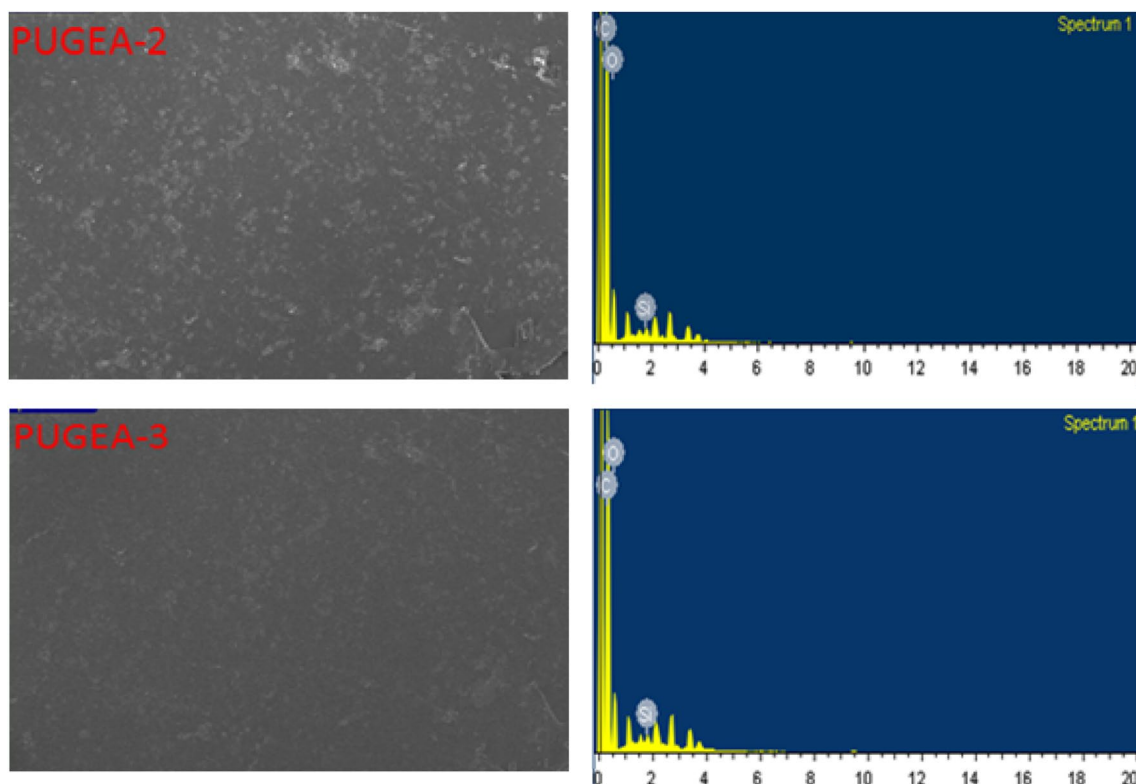
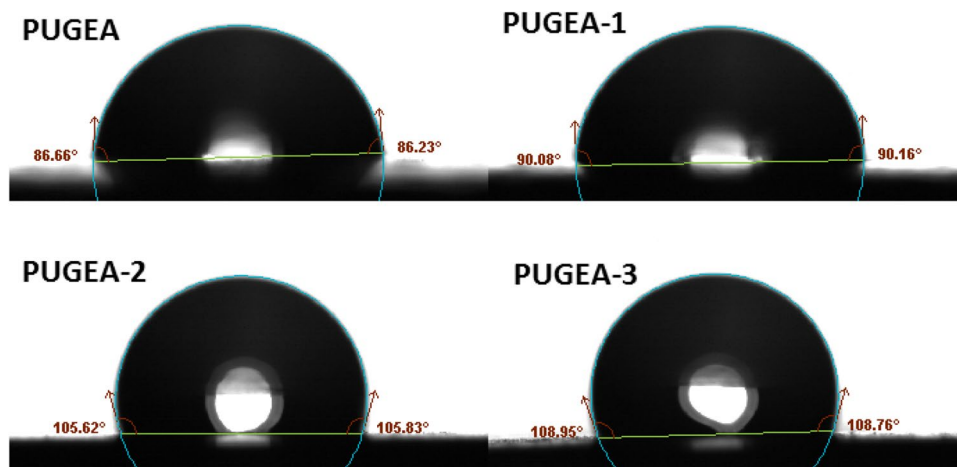


Fig. 6 SEM–EDX of PUGEA-2 and PUGEA-3

Fig. 7 Images of a water drop on the surface of GPEA, PUGEA-1, PUGEA-2, and PUGEA-3



the best physico-mechanical properties. The fumed silica nanoparticles increase the adhesion, impact resistance, and flexibility of the PUGEA coatings because of a good interaction between the polymeric chain and the inorganic fumed silica. Beyond a loading of 3 wt%, though, this interaction can deteriorate because of a silica overload. The silica nanoparticles appear as aggregates, promoting particle–particle interactions and improving compactness; by adding more silica, the increasing density of particles leads to stiff, rigid and brittle PUGEA coatings.

### EIS Measurements

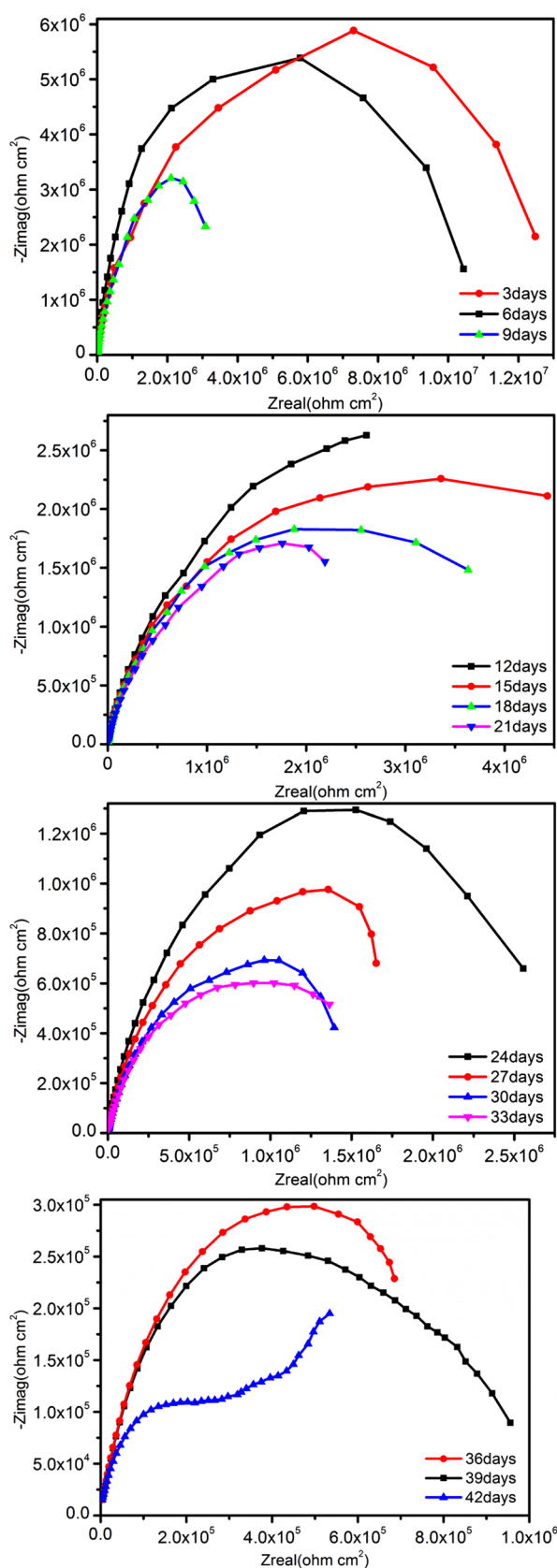
The corrosion performance of PUGEA-2-coated mild steel obtained following 42 days immersion in a 3.5% NaCl solution was analyzed by EIS on the base of Nyquist and Bode plots (Figs. 8 and 9) for selected immersion times. We fitted the impedance data by modeling an equivalent electrical circuit (Fig. 10) consisting of a parallel combination capacitance ( $C_c$ ), a charge transfer resistance ( $R_{ct}$ ), a solution resistance ( $R_s$ ) and a constant phase element (CPE)

instead of the standard Warburg element. The values of the simulated parameter are summarized in Table 1. The impedance of the CPE is obtained from the pre-exponential factor  $Y_0$  and the exponent  $n$ ; the  $\chi^2$  parameter is near to or less than 1, indicating a good fit. The PUGEA-2 sample shows a perfect coating up to 39 days of immersion, often displaying an excellent barrier performance resulting in a large impedance [32]. After 39 days the corrosive ions can reach the metal substrate; therefore, for increasing times, degradation of the PUGEA-coated metals results in a Warburg impedance instead of the CPE one. Blisters due to the electrolyte penetrating through the PUGEA-2 coating and creating a path to the underlying mild steel surface were observed after 42 days. At this stage, the coating degrades and permeable pores form, and the electrolyte can reach the mild steel substrate through them. The diagrams of Fig. 9 show a purely capacitive behavior, characteristic of a barrier film, up to 39 days of immersion, with the low-frequency limit of the impedance modulus and the phase angle being in the  $10^7$ – $10^5 \Omega \text{ cm}^2$  and  $90^\circ$ – $80^\circ$  ranges, respectively, for most of the measured frequencies. Only one time constant could be observed from inspection of the impedance spectra. The linear behavior observed in the low-frequency region indicates that diffusion determines the metal dissolution rate at the surface of mild steel. The results from Table 1 suggest that the solution resistance increases and the charge transfer resistance decreases while the immersion time increases up to 39 days. We observed that impedance decreases with increases immersion time [33–37].

The reason behind this behavior of the effectiveness of the PUGEA-2 coating on mild steel substrate lies in its structure. The adsorption of PUGEA-2 is influenced by the electronic structure of the polymeric chain and the electron density, and in turn, by the presence of functional groups such as urethane, ester, amide, alkyl chain and fumed silica. These contribute to inhibiting the corrosion because of their electron-donor nature, providing electrons to the metal surface. Because PUGEA coatings have an aromatic ring, they have a lone pair of valence electrons resulting from the nitrogen (urethane and amide), oxygen (ester) or silicon heteroatom. Thus, the good adhesion of the PUGEA-2 to the metal substrate depends on the nature of the functional groups and of the electron-donor heteroatom in the polymer matrix [38].

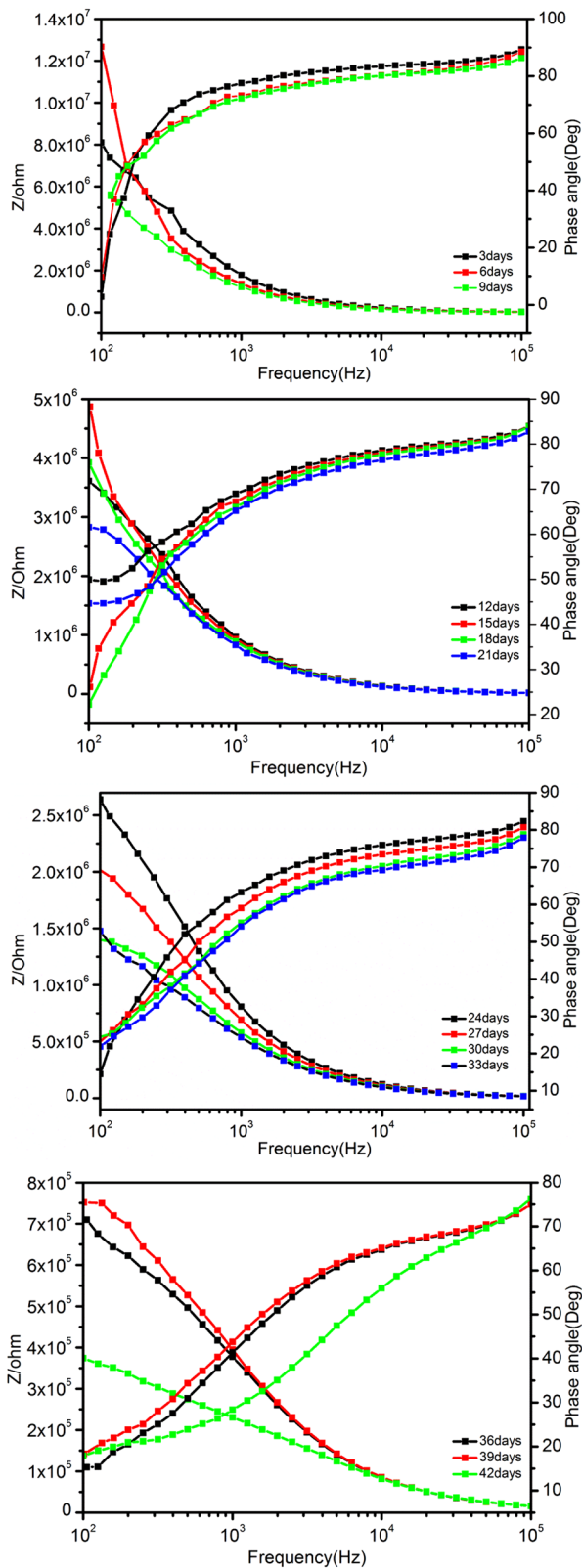
### Thermal Analysis

Figure 11 shows the thermal decomposition of PUGEA, PUGEA-2 and PUGEA3. Initial decomposition temperature, corresponding to 5% weight loss was used as a parameter for determining thermal stability. When the thermograms of PUGEA, PUGEA-2 and PUGEA-3 were compared, it was observed that 5% wt loss occurred at 247 °C, 260 °C and 350 °C, respectively, which was the first decomposition

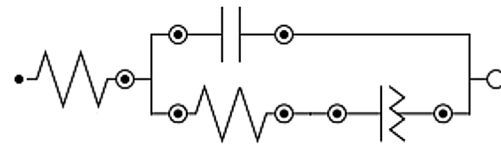


**Fig. 8** Nyquist plots of PUGEA-2 after immersion in the 3.5 wt% NaCl solution for 42 days





**Fig. 9** Bode and phase diagrams of PUGEA-2 after immersion in the 3.5 wt% NaCl solution for 42 days



**Fig. 10** Proposed equivalent circuit for fitting and simulation of the Nyquist plot

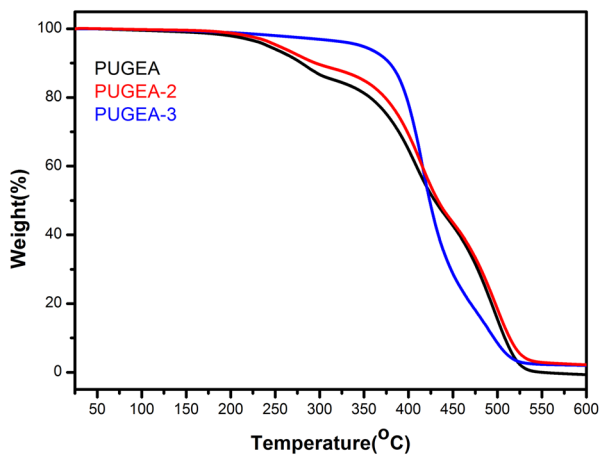
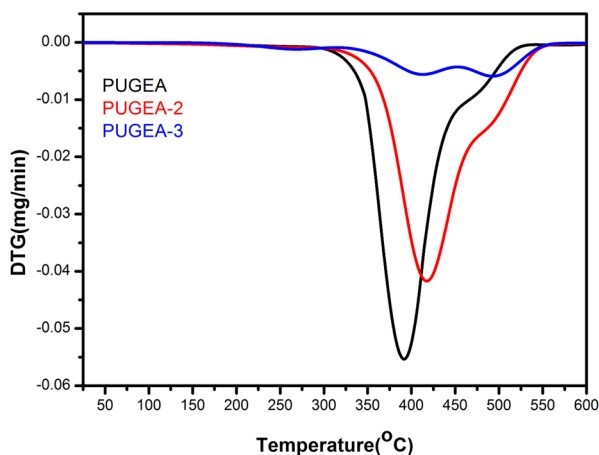
associated with weight loss due to trapped solvent. 50% weight loss of PUGEA, PUGEA-2 and PUGEA-3 at 430 °C, 426 °C and 423 °C occurred due to thermal degradation. DTG thermograms of PUGEA, PUGEA-2 and PUGEA-3 clearly showed decomposition in Fig. 12. One peak in PUGEA, PUGEA-2 and PUGEA-3 runs between 306 and 550 °C centered at 392 °C, 418 °C, and 414 °C, respectively. The peaks in this temperature range correspond to 50% weight loss in the TGA thermogram, due to urethane, ester, amide and hydrocarbon chain collectively. The increasing amount of silica in PUGEA resin increases the thermal stability that can be attributed to the homogeneous dispersion of fumed silica in the PUGEA matrix. Initially PUGEA-3 with higher silica content showed improved thermal stability over PUGEA-2, however beyond about 400 °C, PUGEA-2 shows better thermal stability relative to PUGEA-3. In general, the silica content forms a protective layer on polymer and plays significant role in enriching char formation, however, here, the increased silica loading did not bring about any significant increase in char yield formation as also found in previous results [39, 40]. Thus, the increase in fumed silica does not increase flame retardancy of PUGEA, however, good thermal stability is achieved upto 2wt% loading of fumed silica in PUGEA, beyond this loading the thermal stability is affected. Thus, the studies revealed that PUGEA-2 shows the best performance overall, to be employed as a corrosion protective coating materials.

## Conclusions

An air-dry nanocomposite was prepared from corn-oil based poly (urethane-esteramide) by incorporating fumed silica. The PUGEA/fumed silica nanocomposite shows improved hardness, gloss, adhesion, hydrophobicity, thermal stability and corrosion resistance respect to PUGEA. The PUGEA/fumed silica nanocomposite can serve as a coating to protect mild steel and is stable up to a temperature of 275 °C. Furthermore, it displayed an outstanding corrosion resistance in 3.5 wt% NaCl solution up to 39 days. From this study, it is concluded that the composite coating resulting from fumed silica and recycled corn oil is a cost-effective and environmentally friendly sustainable resource.

**Table 1** The electrochemical parameters obtained from the impedance of PUGEA-2 in the 3.5 wt% NaCl solution for 3–42 days at room temperature

Immersion time (days)	Solution resistance $R_s$ (k $\Omega$ )	Charge transfer resistance, $R_{ct}$ (k $\Omega$ )	Coating capacitance $C_c$ (pF)	CPE		$\chi^2$
				$Y_0$ (nmho*s <sup>n</sup> )	$n$	
3	3.32	-5.56	65.6	173.3	0.19	1.09
6	3.89	-1.03	73.8	8.98	0.37	1.00
9	4.25	-5.39	76.9	10.3	0.40	0.51
12	4.58	-331	85.6	12.7	0.42	0.14
15	4.62	-423	90.0	17.9	0.38	0.28
18	4.69	-711	95.8	33.7	0.30	0.30
21	4.96	-333	91.6	23.1	0.38	0.16
24	5.34	-1.15	103	70.0	0.20	0.41
27	5.99	-605	103	78.9	0.24	0.31
30	6.64	-458	104	109	0.24	0.31
33	6.95	-387	107	113	0.24	0.34
36	7.38	-307	121	153	0.02	1.03
39	7.27	-458	109	291	0.16	0.25
42	2.98	422	124	590	0.25	0.38

**Fig. 11** TGA thermogram of PUGEA, PUGEA-2, and PUGEA-3**Fig. 12** DTG thermogram of PUGEA, PUGEA-2, and PUGEA-3

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## References

- Sharmin E, Zafar F, Akram D, Alam M, Ahmad S (2015) *Ind Crops Prod* 76:215
- Shogren RL, Petrovic Z, Liu Z, Erhan SZ (2004) *J Polym Environ* 12:173
- Alam M, Akram D, Sharmin E, Zafar F, Ahmad S (2014) *Arab J Chem* 7:469
- Scala JL, Wool RP (2005) *Polymer* 46:61
- Ferrer MCC, Babb D, Ryan AJ (2008) *Polymer* 49:3279
- Maisonneuve L, Lebarbe T, Grau E, Cramail H (2013) *Polym Chem* 4:5472
- Dutta S, Karak N, Baruah S (2010) *J Appl Polym Sci* 115:843
- Koprululu A, Onen A, Serhatli E, Guner FS (2008) *Prog Org Coa* 63:365
- Lligadas G, Ronda JC, Galia M, Biermann U, Metzger JO (2006) *J Polym Sci Part A* 44:634
- Tsujimoto T, Uyama H, Kobayashi S (2003) *Macromol Rapid Commun* 24:711
- Behzadnasab M, Mirabedini SM, Esfandeh M, Farnood RR (2017) *Prog Org Coat* 105:212
- Chen S, Wang Q, Wang T, Pei X (2011) *Mater Deg* 32:803
- Siyabolu TO, Sasidhar K, Anjaneyula B, Kumar KP, Rao BVSK, Narayan R, Olaofe O, Akintayo ET, Raju KVS (2013) *J Mater Sci* 48:8215
- Alam M, Alandis NM, Ahmad N, Alam MA, Sharmin E (2019) *Open Chem* 17:206
- Alam M, Alandis NM, Ahmad N, Sharmin E, Zafar F, Alam MA (2016) *Korean J Chem Eng* 33:1736
- Alam M, Ashraf SM, Ray AR, Ahmad S (2010) *J Polym Environ* 18:208
- Alam M, Alandis NM (2014) *Ind Crops Prod* 57:17
- Huang Y, Lu J, Chen H, Du W, Wang X (2019) *J Cryst Growth* 507:1
- Dheep GR, Sreekumar A (2018) *Appl Therm Eng* 129:1189

20. Rozenberg BA, Tenne R (2008) *Prog Polym Sci* 33:40
21. Oprea S (2010) *Polym Bull* 65:753
22. Eksik O, Erciyes AT, Yagci Y (2008) *J Macromol Sci Part A* 45:698
23. Dhoke SK, Khanna AS (2009) *Corros Sci* 51:6
24. Mallakpour S, Dinari M (2012) *Prog Org Coat* 75:373
25. Yang J, Han CR, Duan JF, Xu F, Sun RC (2013) *Nanoscale* 5:10858
26. Sangermano MG, Malucelli G, Amerio E, Priola A, Billi E, Rizza G (2005) *Prog Org Coat* 54:134
27. Chiang HC, Hu HC, Tsiang RCC (2008) *J Polym Sci Part A* 46:8149
28. Zhou S, Wu L, Sun J, Shen W (2002) *Prog Org Coat* 45:33
29. Sangermano M, Periolatto M, Signore V, Spina PR (2017) *Prog Org Coat* 103:152
30. Madhup MK, Shah NK, Parekh NR (2017) *Prog Org Coat* 102:186
31. Dolatzadeh F, Moradian S, Jalili MM (2011) *Corros Sci* 53:4248
32. Garcia YG, Gonzalez S, Souto RM (2007) *Corros Sci* 49:3514
33. Zhang JT, Hu JM, Zhang JQ, Cao CN (2004) *Prog Org Coat* 51:145
34. Hosseini MG, Sabouri M, Shahrabi T (2007) *Prog Org Coat* 60:178
35. Grundmeier G, Schmidt W, Stratmann M (2000) *Electrochem Acta* 45:2515
36. Macedo MCSS, Mattos ICPM, Fragata FL, Jorcin JB, Pebere N, Mattos OR (2009) *Corros Sci* 51:1322
37. Zhu Y, Xiong J, Tang Y, Zuo Y (2010) *Prog Org Coat* 69:7
38. Srikanth AP, Nanjundan S, Rajendran N (2007) *Prog Org Coat* 60:320
39. Liu YL, Hsu CY, Wei WL, Jeng RJ (2003) *Polymer* 44:5159
40. Saavedra JT, Beceiro JL, Naya S, Artiaga R (2008) *Polym Degrad Stabil* 93:2133

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