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RESEARCH ARTICLE - CHEMISTRY

# Synthesis and Surface Activity of Nonionic Surfactants Derived from Gallic Acid

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**Abstract** A series of nonionic surfactants were synthesized from gallic acid and polyethylene glycols with different molecular weights. Chemical structures of the surfactants were confirmed using elemental analysis, infrared and nuclear magnetic resonance ($^1$H-NMR) spectroscopy. Surface activities of the surfactants in their solutions were determined by surface tension measurements. Increasing the polyethylene glycol chain length increased the surface activity of the surfactants in solutions. Measurements of interfacial tension between surfactants solutions and substrates with different polarity showed that the type of substrates plays functioning role on the interfacial properties. Thermodynamic properties of adsorption and micellization processes showed their tendency towards adsorption at solution interface and micellization in their solutions. Results showed reasonable surface activities compared to conventional nonionic surfactants.

**Keywords** Gallic acid · Nonionic surfactant · Interfacial activity · Adsorption · Micellization

## 1 Introduction

Recently, increasing attention to environmental issues has driven the industry towards environmentally friendly products from renewable sources. The use of synthetic, biodegradable and environmentally accepted surfactants from vegetable resources has increased over the past 25 years [1]. Gallic acid (trihydroxybenzoic acid, 3,4,5-trihydroxybenzoic acid) is found in gallnuts, sumac, witch hazel, tea leaves, oak bark and other plants [2]. Gallic acid has several applications in pharmaceutical industry and acts as efficient anti-fungal and anti-viral agents [3]. Gallic acid found in several plants including: oaks species (*Quercus alba*) and red oak (*Quercus robur*) [4], *Caesalpinia mimosoides*, stem bark of *Boswellia dalzielii* [5], Drosera, *Rhodiola rosea*, Triphala, *Toona sinensis*. The use of naturally occurring compounds including plant extracts, e.g., gallic acid, provides safe and environmentally friendly compounds, including biocides, corrosion inhibitors and surfactants [6]. The safety of these compounds is attributed to their ability towards biodegradation and low toxicity. These compounds can replace the ordinary compounds in several applications. Surfactants, including cationic, anionic and amphoteric, are widely used in the homecare and industrial applications. Their presence in the environment is a great problem due to the lack of biodegradation tendency. As a result, environmentally friendly surfactants have concerned significant interests for several researchers [6–9]. Nonionic surfactants are very important compounds which can be synthesized from natural resources and can be used in several applications and industries. This type of surfactants is important owns to its higher advantages over conventional surfactants in terms of biocompatibility and biodegradability in detergency and hygienic formulations [10–13]. The aim and importance of this study were to provide nonionic surfactants obtained from naturally occurring compound, gallic acid, which can be used in several applications including corrosion inhibitors and homecare formulations. The obtained surfactants are expected to be environmentally safe for the soil, water and human beings due to their naturally occur-

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ring origins. In this manner, a series of nonionic surfactant is prepared from gallic acid and characterized. The surface activity of the synthesized nonionic surfactants was measured in order to compare their properties by the conventional nonionic surfactants. The results will be used to determine the ability of these surfactants to be used as corrosion inhibitors and emulsifying agents.

## 2 Experimental

### 2.1 Chemicals

Gallic acid (99.9 %), *n*-hexane (99.8 %), 1-octanol (99.9 %) and polyethylene glycol (400, 600, 1,500 g/mole) (99.8 %) were obtained from Sigma-Aldrich, Germany. Xylene and paraffin oil were obtained from ADWIC chemicals company, Egypt. Paraffin oil is light paraffin oil (a mixture of hydrocarbons) with average carbon chains of 12 carbon atom.

### 2.2 Synthesis of Nonionic Surfactants

Gallic acid (0.2 mole, 34 g) and 0.11 mole of polyethylene glycol (400, 600 and 1,500 g/mole) were weighted accurately and esterified in two-necked flask equipped with mechanical stirrer and Dean-Stark connection in the presence of 150 mL of xylene solvent [14]. The reaction mixture heated at 134 °C for 6 h until the water of reaction (0.2 mole, 3.6 mL) was obtained [15]. After the reaction mixture is allowed to cool, solvent was evaporated under reduced pressure. The excess polyethylene glycol was removed by vacuum distillation to obtain three surfactants, which designated as: GA-400, GA-600 and GA-1500, Scheme 1.

### 2.3 Measurements

#### *2.3.1 Surface Tension Measurements*

The measurements of surface tension were taken using Krüss K6 tensiometer using platinum ring detachment method, Germany. Different aqueous solutions of the nonionic surfactants GA-400, GA-600 and GA-1500 in a concentration range of 10E-01–10E-05 M at 25 °C were used. The solutions were placed in a clean cup made from Teflon with a diameter of 28 mm, and the surface tension measurements started after 2 h to ensure solution interface stabilization and surfactants molecules adsorption at the water/air interface. The surface tension values were considered as an average of three replicates [16].

#### *2.3.2 Interfacial Tension Measurements*

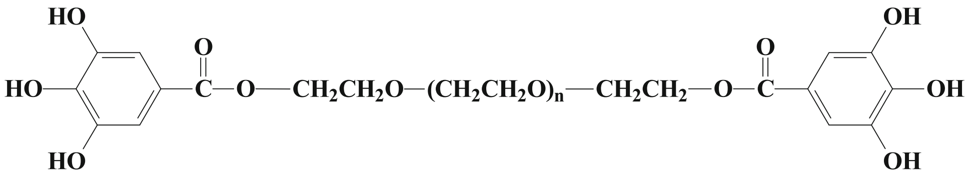
Interfacial tension measurements were taken using same procedures of surface tension measurements and performed between three liquids with different polarity: paraffin oil, *n*-hexane and 1-octanol and the aqueous solution of the surfactants (0.1 % by weight) at 25 °C [17,18].

#### *2.3.3 Emulsion Stability*

The stability of the emulsions was determined as follows: 10 mL (0.5 %) of aqueous solutions of the synthesized surfactant and 10 mL of paraffin oil mixed together in a closed graduated cylinder by vigorous stirring at 25 °C [19]. Then, the cylinder was allowed to stand for separation of the two layers of paraffin oil and aqueous surfactant solution. The steps were repeated three times, and the time of separating 9 mL of pure aqueous surfactant solution was counted. The emulsion stabilities of the surfactants were the average of the three readings.

## 3 Results and Discussion

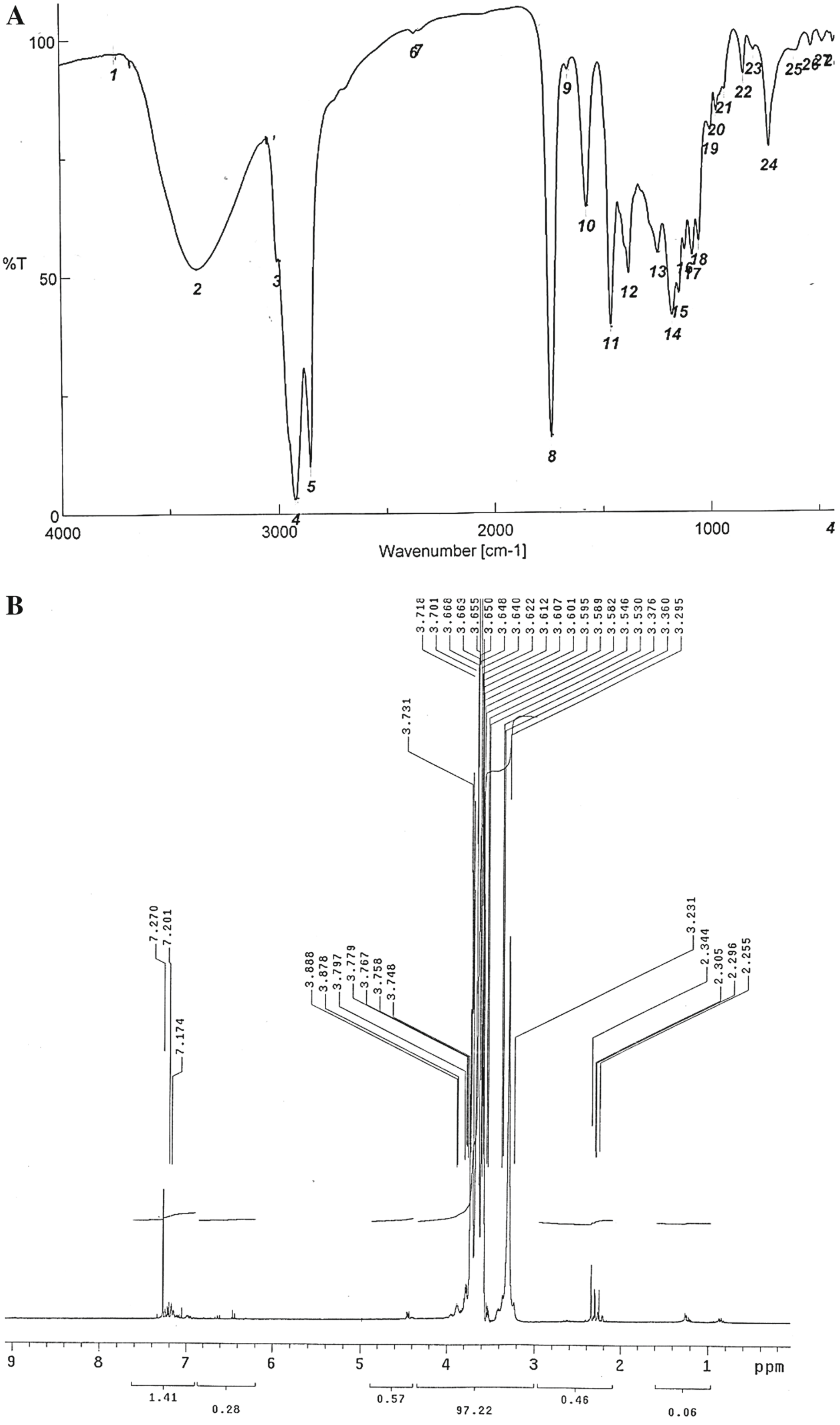
The structures of the synthesized surfactants were elucidated using IR spectra (Fig. 1a) which showed the following absorption bands: 3,590 and 3,477 $cm^{-1}$ were assigned to O–H group, 2,920 and 2,863 $cm^{-1}$ were ascribed to C–H of $CH_2$, 1735 $cm^{-1}$ assigned to C=O of ester group, 617 $cm^{-1}$ corresponded to C–O group, 1,453 $cm^{-1}$ assigned to C–H bond of $CH_2$ group, 883 and 822 $cm^{-1}$ bands ascribed to C–H bond of phenyl group, respectively, while $^1$H-NMR spectra of surfactants showed signals at (ppm): 7.17 ppm corresponding to H-O proton, 3.9–3.87 ppm assigned to phenyl H-C proton, 3.81 ppm ascribed to C–H proton of ester group ($COOCH_2$), 3.74 ppm assigned to C–H proton of $COOCH_2CH_2$, 3.71–3.29 ppm corresponding to the repeated methylene groups in polyethylene glycol chains, Fig. 1b.



**GA-400: n=6; GA-600; n=11; GA-1500; n=31.**

**Scheme 1** Structure of the nonionic surfactants: GA-400, GA-600 and GA-1500

**Fig. 1** **a** IR spectra of GA-400; **b** $^{1}$H-NMR spectra of GA-400 surfactant



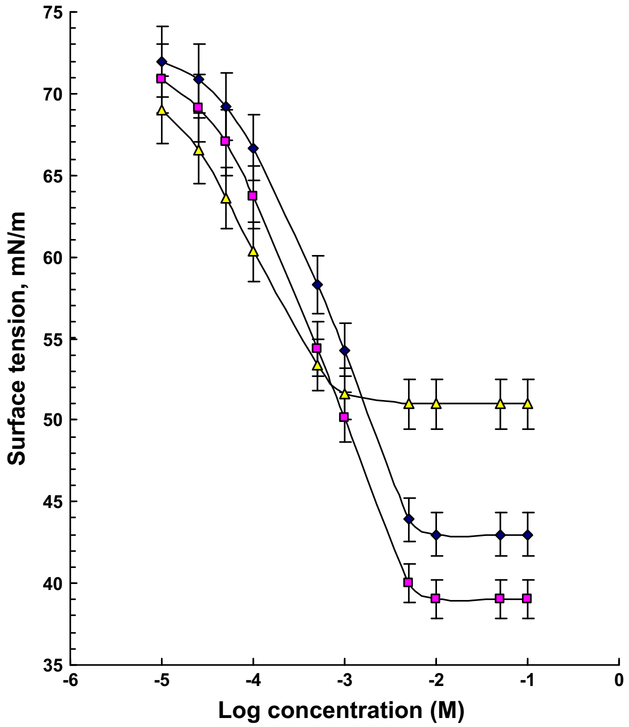


**Fig. 2** Surface tension versus concentration (−log C) for gallic acid-derived nonionic surfactants (*triangle* GA-400, *diamond* GA-600, *square* GA-1500) at 25 °C

### 3.1 Surface Activity

Surface activities of the synthesized surfactants were determined by measuring the surface tension of their aqueous solutions as a function of concentration, Fig. 2. It is clear from Fig. 2 that the values of surface tension are decreased gradually when the surfactant concentration increased (premicellar region). The variation in surface tension values is stopped at certain concentration which is called the critical micelle concentration (CMC).

After CMC, the surface tension values stay stable with no considerable change by increasing the concentration of the surfactants (post-region). The critical micelle concentration (CMC), as a characteristic property of aqueous surfactant solutions, is determined by extrapolation of pre-micellar region and post-micellar regions in surface tension vs. concentration profile. It is clear from surface tension profile that the increase in surfactant concentration depresses the surface tension of the solution. Depression in surface tension values of surfactant in their aqueous solutions is attributed to the increase in surfactant molecules adsorption at the solution interface. Adsorption of surfactant molecules at air–aqueous solution interface occurred due to the interaction between surfactant and water molecules. At constant concentration of the different surfactants, it is clear that the gradual decrease in the polyethylene glycol chain length decreases the surface tension values. That can be attributed to the increase in hydrophobicity by the decrease in PEG chains. That pumps the surfactant molecules to the interface and consequently decreases the surface tension. That can be observed from the surface tension values of surfactant molecules at the critical micelle concentration ($\gamma_{\mathrm{cmc}}$) (Table 1).

The effectiveness values ($\pi_{\mathrm{cmc}}$) are the depression in surface tension values of surfactant in their aqueous solutions at the critical micelle concentration compared to that of distilled water and can be calculated according to Eq. 1 as follows [18]:

$$\pi_{\mathrm{cmc}} = \gamma_{\mathrm{o}} - \gamma_{\mathrm{cmc}} \tag{1}$$

where $\gamma_{\mathrm{o}}$ is the surface tension of bidistilled water (71.8 mN/m) at 25 °C, $\gamma_{\mathrm{cmc}}$ is the surface tension of aqueous surfactant solution at critical micelle concentration.

Effectiveness is a useful surface active parameter and can be used to compare between the surface activities of two or more surfactants within the same series, i.e., surfactants have same function groups. Larger $\pi_{\mathrm{cmc}}$ value of aqueous surfactant solution indicates its higher surface activity than aqueous surfactant solution with smaller $\pi_{\mathrm{cmc}}$ value [19]. Increasing the polyethylene glycol chain length from 6 to 31 units is gradually decreases $\pi_{\mathrm{cmc}}$ values of the synthesized surfactants in their aqueous solutions.

The critical micelle concentration of surfactant is an important value that can be obtained from surface tension concentration profile of the surfactants in aqueous solutions. The critical micelle concentration values of the synthesized surfactants were extracted from Fig. 2 and recorded in Table 1. The obtained CMC values are in the range of 7.5E-04–5.62E-03 M, which are comparatively lower than the reported values of conventional nonionic surfactants [20].

It seems that the critical micelle concentration values are reduced by increasing the polyethylene glycol chain length. The micellization process is accompanied by saturation

**Table 1** Surface and thermodynamic parameters of gallic acid-derived nonionic surfactants (GA-400, GA-600, GA-1500) at 25 °C

| Compound | $\gamma_{\mathrm{cmc}}$ (mNm$^{-1}$) | $\pi_{\mathrm{cmc}}$ (mNm$^{-1}$) | Pc$_{20}$ (M × 10$^{-4}$) | CMC (M × 10$^{-4}$) | $\Gamma_{\max}$ (Mol cm$^{-2}$) | A$_{\min}$ (A$^2$) | ΔG$_{\mathrm{mic}}$ (kJ Mol) | ΔG$_{\mathrm{ads}}$ (kJ Mol) |
|---|---|---|---|---|---|---|---|---|
| GA400 | 39 | 33 | 10 | 56.2 | 2.68 | 61.8 | −12.84 | −14.29 |
| GA600 | 43 | 29 | 5.62 | 44.7 | 2.58 | 64.4 | −13.41 | −15.07 |
| GA1500 | 51 | 21 | 3.55 | 7.5 | 1.89 | 88.0 | −17.83 | −20.52 |

of surfactant solution interface with surfactant molecules. Adsorption of the synthesized surfactants at air/solution interface is occurred due to two forces. The first is the attraction force between surfactants and water molecules via hydrogen bonds which increased by increasing the ethylene glycol units, and the second is repulsion force between phenyl groups and water molecules. The first force increases the tendency of surfactant molecules to locate in the bulk of their solution, while the second increases their presence at the air/water interface [21–23]. The micellization of surfactants is the result of the two forces. The high tendency towards adsorption at interface causes saturation to the interface at low concentration, which consequently tends the molecules to form micelles at lower concentrations. It is clear that GA-1500 surfactant has higher tendency towards micellization, i.e., micelles formed at lower concentration, while the tendency towards micelle formation decreased by decreasing polyethylene glycol chain length (i.e., micelles formed at higher concentration), as represented in case of GA-400 and GA-600 surfactants, Table 1. Increasing the number of polyethylene glycol units (hydrophilic groups) in GA-1500 molecule increases their tendency towards the presence in the bulk of their aqueous solutions as a result of large ethylene glycol content. But, the increase in ethylene glycol units increases the hydrophobicity of the molecules due to the increase in methylene groups incorporated in the chains, which increases the adsorption of the molecules at air–aqueous interface to reach the saturation. The interface saturation forced the molecules to undergo into the bulk of the aqueous solution to from micelles [20,24].

The maximum surface excess ($\Gamma_{\max}$) is defined as the maximum concentration can be reached by surfactant at air–aqueous solution interface before the micellization occurred in the bulk.

The maximum surface excess values of the synthesized nonionic surfactants were calculated using the modified form of Gibb's-Duhem according to Eq. (2) as follows [25]:

$$\Gamma_{\max} = [\mathrm{d}\gamma/\mathrm{d}\log \mathrm{C}\,]/]\,2.303RT] \tag{2}$$

where $d\gamma/d\log$ C is the slope of the surface tension profile in the steeper region, $R$ is the universal gas constant and $T$ is the absolute temperature.

Increasing the surface concentration shows high tendency of surfactant molecules, and its high value is accompanied by high CMC values. Table 1 declares that GA-1500 derivative has the lowest maximum surface excess value. On contrarily, decreasing the hydrophilic chain, as represented in case of GA-400 and GA-600 derivatives, increases $\Gamma_{\max}$ to higher values, which is in agreement with the reported data [20,22].

The efficiency values ($\mathrm{Pc}_{20}$) are the logarithm values of surfactant concentration at which the surface tension of aqueous surfactant solutions reduced by20 $\mathrm{m\,Nm^{-1}}$. $\mathrm{Pc}_{20}$ value measures the efficiency of surfactant adsorption air–water interface. $\mathrm{Pc}_{20}$ value is useful in comparison between two or more surfactants of different types (i.e., anionic, cationic or nonionic). Larger $\mathrm{Pc}_{20}$ values indicate higher tendency of surfactant molecules to adsorb at air–water interface [23]. $\mathrm{Pc}_{20}$ values of GA-1500, GA-600 and GA-400 surfactants decreased by increase in polyethylene glycol chain length from 6 to 31. This result suggests that the longer the polyethylene glycol chains of the gallic acid-derived surfactants, the weaker the adsorption at the air–water interface. Comparing these results with our previous results of nonionic surfactants [24,26] indicates that gallic acid-derived surfactants are superior in reducing the surface tension due to their high adsorption tendency at air–water interface.

The area per surfactant molecule ($\mathrm{A_{min}}$) or the average area occupied by each surfactant molecule at the air–water interface is calculated according to Eq. (3) [27] as follows:

$$\mathrm{A_{min}} = 1/[N_\mathrm{A} \cdot \Gamma_{\max}] \tag{3}$$

where $N_\mathrm{A}$ is Avogadro's number and $\Gamma_{\max}$ is the maximum surface excess at constant temperature.

The average area per surfactant molecule ($\mathrm{A_{min}}$) at aqueous solution interface is particularly influenced by the hydrophilic group size. The hydrophilic groups of the synthesized surfactants are varied in size by changing the ethylene glycol units in each chain. The smallest area ($61.8\,\mathrm{A}^2$) at the air–water interface was obtained for GA-400 surfactant which had six repeated ethylene glycol units. Increasing the repeated ethylene glycol units from 9 units to 31 units increases the area occupied by surfactant molecules at their aqueous solution interface from 64.4 to $88.0\,\mathrm{A}^2$. Obviously, the influence of hydrophilic head group is marked in case of nonionic surfactants, while this difference is not pronounced in case of charged surfactants, especially in case of anionic surfactants [28].

The surface activity parameters including effectiveness, efficiency, maximum surface excess and minimum surface area of the synthesized surfactants determine their ability towards adsorption at the different interfaces. Higher effectiveness and efficiency and maximum surface excess values and lower minimum surface area indicate high tendency of surfactants molecules at the different interfaces (air–water, oil–water, solid–water interfaces). The high tendency of surfactants towards adsorption at the various interfaces increases their applicability in several applications and mainly as corrosion inhibitors and emulsifiers.

### 3.2 Interfacial Properties

The properties of surfactant molecules adsorbed in the interface between aqueous and nonpolar phases determine the emulsification tendency of these surfactants. This property can be measured using interfacial tension measurements.

**Table 2** Interfacial tension values of gallic acid-derived nonionic surfactants (GA-400, GA-600, GA-1500) against different nonpolar liquids

| Surfactant | Interfacial tension (mN/m) | | |
|---|---|---|---|
| | *n*-Hexane | Paraffin oil | 1-Octanol |
| GA400 | 29 | 22 | 10 |
| GA600 | 28 | 20 | 9 |
| GA1500 | 23 | 17 | 8 |

Interfacial tension values of GA-400, GA-600 and GA-1500 are arranged in Table 2. The nature of nonpolar phase has substantial consequence on the interfacial tension of *n*-hexane/water, paraffin oil/water and 1-octanol/water systems. The hydrophobicity of nonpolar medium increased in the following sequence: *n*-hexane > paraffin oil > 1-octanol. Interfacial tension values are increased in same trend. The values of interfacial tension of GA-400 are the highest among the three systems and ranged between 29, 22 and 10 mN/m for *n*-hexane, paraffin, oil and 1-octanol, respectively, while the lowest values were obtained in case of GA-1500 nonionic surfactants, which are 23, 20 and 8 mN/m for *n*-hexane, paraffin oil and 1-octanol. *N*-hexane has the maximum hydrophobic (nonpolar) character and has high repulsion by water phase. That is due to *n*-hexane has not any polar group; hence, its interaction with the adsorbed layer of the surfactant at the interface is very weak. That increases the interfacial tension of *n*-hexane/surfactant system considerably.

In case of paraffin oil, its chemical structure contains few function groups, e.g., double bonds and/or oxo-compounds. These groups interact by polyethylene glycol chains, which decrease the interfacial tension at water/paraffin oil interface. In case of 1-octanol, the presence of hydroxyl groups facilitates the development of hydrogen bonds between octanol and surfactants at interface. That decreases the interfacial tension of this system significantly [30]. Increasing the polyethylene glycol chain length from 9 to 31 units increases the interaction between adsorbed layer of surfactant molecules at the interface and the nonpolar phase. That was accompanied by decreasing interfacial tension values as shown in Table 2.

### 3.3 Micellization and Adsorption Thermodynamics

Thermodynamic functions describe the energetic parameters of the adsorption and micellization processes of surfactant molecules either at the interface or in the bulk of their solutions.

The free energies of micellization ($\Delta G_{\mathrm{mic}}$) and adsorption ($\Delta G_{\mathrm{ads}}$) were calculated using Eqs. (4–5) [31] as follows:

$$\Delta G_{\mathrm{mic}} = 2.303\mathrm{RT}\log(\mathrm{CMC}) \qquad (4)$$

$$\Delta G_{\mathrm{ads}} = \Delta G_{\mathrm{mic}} - (0.006\pi_{\mathrm{cmc}} \times \mathrm{A}_{\mathrm{min}}) \qquad (5)$$

Micellization and adsorption free energies are in negative values, which indicate their spontaneous occurrence in the solution, while large negative value indicates the majority of one process than the other. The standard free energies for adsorption and micellization of the synthesized surfactants are negative, which specifies spontaneous occurrence of both processes in the solution. The values of adsorption free energies are more negative than those of micellization process. That indicates the high tendency of the synthesized surfactants towards adsorption at the interface than the micellization. This result showed the applicability of these surfactants in several applications including: emulsification, detergency and corrosion inhibition application.

## 4 Conclusion

Results of this study revealed the following points:

1. Gallic acid-derived nonionic surfactants are simply prepared surfactants.
2. Surface activity measurements showed their high tendency towards surface tension reduction and low critical micelle concentration.
3. Surface and thermodynamic properties are dependent on the hydrophilic chain length.
4. Interfacial tension of the surfactants is strongly related to the type of the nonpolar medium and the polyethylene glycol chain length.
5. Surface activity parameters of the synthesized surfactants showed their high tendency towards adsorption at various interfaces. This qualifies these surfactants as efficient corrosion inhibitors and emulsifying agents.
6. The results will encourage several researches including the use of these surfactants as corrosion inhibitors to inhibit corrosion of carbon steel in acidic medium.

## References

1. Seddon, E.J.; Friend, C.L.; Roski, J.P.: Detergents and Dispersants chemistry and Technology of Lubricants, Springer 3rd Edn. pp. 213–236 (2010)
2. Reynolds, L.D.; Wilson, N.G.: Scribes and Scholars, 3rd Edn. pp. 193–194. Clarendon Press, Oxford (1991)
3. Fiuza, S.M.: Phenolic acid derivatives with potential anticancer properties-a structure–activity relationship study. Part 1: Methyl, propyl and octyl esters of caffeic and gallic acids. Bioorg. Med. Chem. **12**, 3581–3589 (2004)
4. Pirjo, M.; Heikki, S.; Lasse, L.; Juhani, K.; Terttu, V.: Analysis of oak tannins by liquid chromatography-electrospray ionisation mass spectrometry. J. Chromatogr. A **891**, 75–83 (2000)
5. Anchana, C.; Aphiwat, T.; Jeremy, D.K.; Nuansri, R.: Antimicrobial gallic acid from *Caesalpinia mimosoides* Lamk. Food Chem. **100**, 1044–1048 (2007)

6. Biswas, A.; Roy, A.: Surface-active characteristic of sodium anacardate isolated from cashew nut shell oil. Nature **182**, 1299–1300 (1958)
7. Solomon, K.R.; Thompson, D.G.: Ecological risk assessment for aquatic organisms from over-water uses of glyphosate. J Toxicol. Environ. Health B Crit. Rev. **6**, 289–324 (2003)
8. Infante, M.; Pinazo, A.; Seguer, J.: Non-conventional surfactants from amino acids and glycolipids: structure, preparation and properties. Colloid. Surf. A. Physicochem. Eng. Asp. **123**(124), 49–70 (1997)
9. Rondel, C.; Portet, B.; Alric, I.; Mouloungui, Z.; Blanco, J.F.; Silvestre, F.: Green production of anionic surfactant obtained from pea protein. J. Surfact. Deterg. **14**, 535–544 (2011)
10. Holmberg, K.: Novel Surfactants. Marcel Dekker, New York (1998)
11. Adewuyi, A.; Oderinde, R.A.; Rao, B.V.; Prasad, R.B.: Synthesis of alkanolamide: a nonionic surfactant from the oil of *Gliricidia sepium*. J. Surfact. Deterg. **15**, 89–96 (2012)
12. Leal, J.S.; Gonzalez, J.J.; Kaiser, K.L.; Palabrica, V.S.; Comelles, F.; Garcıa, M.T.: On the toxicity and biodegradation of cationic surfactants. Acta Hydrochim. Hydrobiol. **22**, 13–18 (1994)
13. Do, L.D.; Withayyapayanon, A.; Harwell, J.H.; Sabatini, D.A.: Environmentally friendly vegetable oil microemulsions using extended surfactants and linkers. J. Surfact. Deterg. **12**, 91—99 (2009)
14. Negm, N.A.; El Farargy, A.F.M.; Mohammed, D.E.; Mohamad, H.N.: Environmentally friendly nonionic surfactants derived from tannic acid: synthesis, characterization and surface activity. J. Surf. Deterg. **15**, 433–443 (2012)
15. Negm, N.A.; El-Tabl, A.S.; Aiad, I.A.; Zakareya, K.; Moustafa, A.H.: Synthesis, characterization, biodegradation and evaluation of the surface active properties of nonionic surfactants derived from Jatropha oil. J. Surfact. Deterg. **16**, 857–863 (2013)
16. Abdel-Salam, F.H.; El-Said, A.G.: Synthesis and surface active properties of Gemini cationic surfactants and interaction with anionic azo dye (AR52). J. Surfact. Deterg. **14**, 371–379 (2011)
17. Han, L.; Ye, Z.; Chen, H.; Luo, P.: The interfacial tension between cationic Gemini surfactant solution and crude oil. J. Surfact. Deterg. **12**, 185–190 (2009)
18. Negm, N.A.; Tawfik, S.M.: Isoxazolium cationic Schiff base surfactants: surface and antimicrobial properties. Chimica Oggi/Chem Today **30**, 5–9 (2012)
19. Negm, N.A.; Aiad, I.A.: Synthesis and characterization of multifunctional surfactants in oil-field protection applications. J. Surfact. Deterg. **10**, 87–92 (2007)
20. Negm, N.A.; Zaki, M.F.: Corrosion inhibition efficiency of nonionic Schiff base amphiphiles of p-aminobenzoic acid for aluminum in 4N HCl. Colloid Surface A Physicochem Eng Aspect **322**, 97–102 (2008)
21. Farıas, T.; de Menorval, L.C.; Zajac, J.; Rivera, A.: Benzalkonium chloride and sulfamethoxazole adsorption onto natural clinoptilolite: effect of time, ionic strength, pH and temperature. J. Colloid. Interface Sci. **363**, 465–475 (2011)
22. Farıas, T.; de Menorval, L.C.; Zajac, J.; Rivera, A.: Solubilization of drugs by cationic surfactants micelles: conductivity and $^1$H-NMR experiments. Colloids Surf. A **345**, 51–57 (2009)
23. Pei, L.; Cai, Z.; Song, Z.; Zhu, X.; Shang, S.: 3-Chloro-2-hydroxypropyl dimethyl dehydroabietyl ammonium chloride: synthesis, characterization, and physicochemical properties. J. Surfact. Deterg. (2013). doi:10.1007/s11743-013-1490-0.
24. Negm, N.A.; El-Tabl, A.S.; Aiad, I.A.; Zakareya, K.; Moustafa, A.H.: Synthesis, characterization, biodegradation and evaluation of the surface active properties of nonionic surfactants derived from Jatropha oil. J. Surfact. Deterg. **16**, 857–863 (2013)
25. Dianno, D.; Talmon, Y.; Zana, R.: Alkanediyl.-, Alph.-, Omega-bis(dimethyl ammonium bromide) surfactants (dimeric surfactants). 5. Aggregation and microstructure in aqueous solutions. Langmuir **11**, 1448–1453 (1995)
26. Negm, N.A.; El-Farargy, A.F.; Tawfik, S.M.; Abdelnour, A.M.; Hefni, H.H.; Khowdiary, M.M.: Synthesis, surface and thermodynamic properties of substituted polytriethanolamine nonionic surfactants. J. Surfact. Deterg. **16**, 333–342 (2013)
27. Negm, N.A.: Surface activities and electrical properties of long chain diquatemary bola-form amphiphiles. Egypt. J. Chem. **2**, 4–12 (2003)
28. Zhou, M.; Zhong, X.; Zhao, J.; Wang, X.: Synthesis and surface active properties of 1,1,1,1-tetra-(2-oxypropyl sulfonate-3-alkylether-propoxy)neopentanes. J. Surfact. Deterg. **16**, 285–290 (2013)
29. Atkin, R.; Craig, V.S.J.; Wanless, E.J.: Adsorption of 12-s-12 Gemini surfactants at the silica-aqueous solution interface. J. Phys. Chem. B. **107**, 2978–2985 (2003)
30. Singh, A.; Singh, V.K.; Quraishi, M.A.: Inhibition of mild steel corrosion in HCl solution using Pipali (*Piper longum*) fruit extract. Arab. J. Sci. Eng. **38**, 85–97 (2013)
31. Negm, N.A.; Mahmoud, S.A.: Synthesis, surface and thermodynamic properties of carboxymethyl (di-2-ethanol) ethyl alkanoate ammonium bromide amphoteric surfactants. Egypt. J. Petrol. **14**, 79–86 (2005)