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2-Hydroxyethyl substituted cationic surfactants with dodecyl hydrophobic chain: Properties and application

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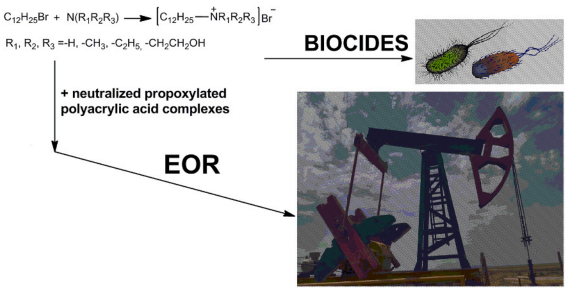
HIGHLIGHTS

- On the basis of $C_{12}H_{25}Br$ surfactants with different head groups were synthesized.
- Surface parameters of the synthesized cationic surfactants have been determined.
- The surfactants have good biological activity against the tested bacteria and fungi.
- Injection of 0.05 wt% complex solution offers 39–45% additional oil recoveries.

GRAPHICAL ABSTRACT



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ABSTRACT

Different head-group containing ionic-liquid cationic surfactants (ILCS) are synthesized by the reaction of 1-bromododecane and various alkylethanolamines ([mono(2-hydroxyethyl)amine, methyldiethanolamine, di(2-hydroxyethyl)amine, tris(2-hydroxyethyl)amine, 2-(methylamino)ethanol, N-2-(dimethylamino)ethanol, 2-(dimethylamino)ethanol]), which hold 2-hydroxyethyl group functionality. By tensiometric and conductometric measurements, surface parameters of the synthesized ILCSs are investigated. Variation of the colloid-chemical parameters with a change in the head group (e.g., replacing methyl-with ethyl- and 2-hydroxyethyl fragments) is recorded. The synthesized ILCSs antimicrobial properties are studied. Microbial assay analysis shows that C_{12}DEA is the most effective biocide relative to other ILCS. New polyelectrolyte-surfactant complexes are also synthesized via stepwise procedure: Acrylic acid and potassium acrylate are polymerized and obtained polymer is propoxylated, then exploiting di(2-hydroxyethyl)amine and tris(2-hydroxyethyl)amine, the synthesized propoxylated acrylic polymers are neutralized. Our studies reveal that the synthesized polyacrylate surfactant complexes are promising substances for oil recovery.

1. Introduction

In recent years quaternary ammonium-type cationic surfactants have been widely used as bactericides [1], inhibitors to suppress corrosion [2], floatation agents for minerals [3], fabric softeners [4], catalytic activating agents [5,6], emulsifying agents [7], oil recovery [8], and

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cosmetic additives [9]. Numerous studies were conducted to obtain the quaternary ammonium salt derivatives via tuning the tail and head groups. The results of the previous investigations show that, by making some changes in hydrophilic and hydrophobic groups of surfactants, it is possible to achieve an improvement in surfactivity, and biocidal properties [1,10–18].

D.Jordan et al. [19] studied the surfactants having a hydroxymethyl group possess the capability to form micelles at lower concentrations than other surfactants of a similar structure. Novel hydroxyl-group containing surfactants were obtained by replacing methyl groups (bonded to the nitrogen atom) in dodecyltrimethylammonium bromide with hydroxyethyl group. Zh. Zhang and coworkers [20] substituted methyl groups in dodecyltrimethylammonium bromide with one or two hydroxyethyl groups and identified that, in the formed surfactants, CMC and a degree of the counterion binding (β) decrease relative to the unsubstituted version. In the case of the quaternary ammonium surfactants, with similar head-group and alkyl chains (C_{14} and C_{16}), the same trend was recorded [21]. Liu et al. [22] synthesized ILCSs based on N-methyldiethanolamine and alkyl bromides. Antibacterial and cytotoxicity analysis of the synthesized ILCSs showed that the surfactant that contains dodecyl chain is highly effective compared to the other variations. A.R. Glennie synthesized (C_{12}-C_{16})-alkyl, dimethyl, and diethyl hydroxyethylammonium bromide-type surfactants and studied a series of their micellar properties [23]. Hydroxyethyl group was incorporated the surfactants polar head-group, and it was observed that the obtain modifications enhance the catalytic effectiveness of lipase via elevating the interfacial water concentration [24]. Since ionic liquid surfactants have better solubility in brine solution, they can be used for enhanced oil recovery (EOR) [25]. M. Nabipur et al. conducted a primary waterflood of tertiary oil recovery efficiency of various commercial surfactants and determined that 1-dodecyl-3-methylimidazolium chloride was the most suitable tertiary oil recovery surfactant, which improving oil recovery by 6.3% compared to other surfactants [26]. Imidazolium, pyrrolidinum, and pyridinium based ionic liquid surfactants were investigated extensively for the enhanced oil recovery application. The literature studies showed that only a few studies were available on the application of ammonium-based ionic liquid (mainly hydroxy-group containing) surfactants as EOR agents [27–29]. M. Ouyang et al. [30] determined that surfactants that retain hydroxyl groups exhibit more effective oil displacement capabilities.

The aforementioned studies show some properties of the ammonium-type ILCSs comprising dodecyl chain, methyl, ethyl, and 2-hydroxyethyl substituents (bonded to the nitrogen atom) in the head group were investigated. However, systematic investigations of surface and biocidal properties of the ternary and quaternary ammonium-type surfactants have not yet been explored. In parallel, the incorporation of some synthesized ILCSs with the acrylate oligomer derivatives to obtain polyelectrolyte complexes is of great interest to the petroleum industry since the complexes are effective EOR agents. The current study is devoted to scrutinize the colloidal-chemical and biocidal properties of secondary, ternary, and quaternary ammonium-type cationic surfactants comprising dodecyl chain as hydrophobic tail groups and different alkyl and 2-hydroxyethyl substituents containing head-groups. Additionally, polyelectrolyte complexes of those ILCSs were obtained and proposed as potential EOR agents.

2. Experimental section

2.1. Instruments and reagents

Bruker TOP SPIN spectrometer (300.12 MHz and 75.45 MHz) was used for ^{1}H NMR and ^{13}C NMR spectral analysis. Magnitudes of chemical shift (δ) in parts per million (ppm) were recorded downfield to tetramethylsilane. D_2O and DMSO were exploited in NMR experiments. Infrared-IR spectra were recorded (in KBr disks) using ALPHA FT-IR (Bruker) spectrometer (see Figs. 1S–12S in Supporting Information). The melting point of the obtained ILCSs was determined by Differential Scanning Calorimetry (DSC) Q20 V24.10 Build 122 was used. 1-bromododecane (97%), tris(2-hydroxyethyl)amine–TEA (98%), di(2-hydroxyethyl)amine–DEA (≥98%), mono(2-hydroxyethyl)amine–MEA (≥98%) were purchased from Sigma-Aldrich. 2-(methylamino)ethanol–MAE (99%), N-methyldiethanolamine–MDEA (≥98%), acrylic acid (90% stab.), propylene oxide (99%), 2-(dimethylamino)ethanol–DMAE (≥99.0%) and 2-(diethylamino)ethanol–DEAE (99%) were purchased from Alfa Aesar. The composition of the used Caspian sea water is as follows (ppm): $Na^+ + K^+$: 3240, Ca^{2+}: 350, Mg^{2+}: 740, Cl^-: 5440, SO_4^{2-}: 3010, HCO_3^-: 220, and total dissolved solids (TDS): 10100. The oil sample used for the study was taken from the Balakhani oil field (near Caspian Sea) and has the following characteristics: gravity of 27.4° API, density of 0.8878 g/cc, and kinematic viscosity 17.8 mm^2/s at 40 °C.

2.2. The synthesis procedure of ILCSs

ILCSs were synthesized according to the procedure described before [31]. Samples of (0.1 mol) 12.46 g of 1-bromododecane and (0.1 mol) 3.05 g of mono(2-hydroxyethyl)amine (or related amount of di(2-hydroxyethyl)amine, tris(2-hydroxyethyl)amine, N-methylethanolamine, N-methyldiethanolamine, 2-(dimethylamino)ethanol, and 2-(diethylamino)ethanol) were mixed under continuous stirring and refluxed with 40 mL of dry 2-propanol at 85 °C for 10 h. Then the reaction was stopped, the mixture was cooled down to −24 °C, and appeared white solid product was recrystallized in acetone several times; the product was then dried in a desiccator for 18 h. The obtained products yield were recorded in a range of 89–95%. The reaction between 1-bromododecane and hydroxyl-group comprising amines undergoes according to the following scheme:

$$C_{12}H_{25}Br + N(R_1R_2R_3) \rightarrow \left[C_{12}H_{25} - \overset{+}{N}(R_1R_2R_3)\right]Br^-$$

where R_1, R_2, R_3 = $–H$, $–CH_3$, $–C_2H_5$ and $–CH_2CH_2OH$

$$R_1 = R_2 = H, R_3 = -CH_2CH_2OH. \ (C_{12}MEA).$$

$$R_1 = H, R_2 = R_3 = -CH_2CH_2OH. \ (C_{12}DEA).$$

$$R_1 = R_2 = R_3 = -CH_2CH_2OH. \ (C_{12}TEA).$$

$$R_1 = H, R_2 = CH_3, R_3 = -CH_2CH_2OH. \ (C_{12}MAE).$$

$$R_1 = R_2 = CH_3, R_3 = -CH_2CH_2OH. \ (C_{12}DMAE).$$

$$R_1 = CH_3, R_2 = R_3 = -CH_2CH_2OH. \ (C_{12}MDEA).$$

$$R_1 = R_2 = CH_2CH_3, R_3 = -CH_2CH_2OH. \ (C_{12}DEAE).$$

C_{12}MEA is a jelly (highly viscous) substance turning to liquid state at 30-50 °C, C_{12}DEA, C_{12}TEA and C_{12}MAE are jelly substances also transitioning to liquid state at 48.3 °C, 41.5 °C, respectively. C_{12}DMAE, C_{12}MDAE, and C_{12}DEAE are at the solid form, melting correspondingly at 74.8 °C, 63.5 °C, and 63.7 °C temperatures.

2.3. Synthesis procedure of propylene oxide modified polyelectrolyte

To synthesize polyacrylic acid, acrylic acid and its potassium salt was polymerized by the radical polymerization method in the presence of $K_2S_2O_8$ initiator [32]. The average molecular weight of the obtained polyacrylic acid was determined by the viscosimetric method (M = 65, 000). 50% of the carboxyl groups in polyacrylic acid were available as potassium salt. In the second stage, 50% of the free carboxyl groups in the obtained polymer was propoxylated. The obtained polyelectrolyte was washed, solvent and unreacted starting materials were evaporated under vacuum. The final multifunctional polyacrylic polymer contains free carboxyl groups, potassium carboxylate, and hydroxy isopropyl groups ($-CH_2-CH(OH)-CH_3$). The scheme showing the synthetic protocol of polyacrylic acid is given below.

2.4. Determination of electroconductivity and surface activity the obtained ILCSs

The surface tension of the obtained ILCSs was determined by Du Nouy ring method (Sigma 702 tensiometer, Finland). 0.0001–0.35 % wt. water solutions of the ILCSs were prepared, and kept for one day, then their surface tension was measured at 298 K [33]. For these measurements, solutions were prepared using distilled water. The value of the surface tension at the air-water boundary of distilled water was 72.2 mN/m at 298 K. An error of these tensiometric measurements was less than ±0.15 mN/m.

Specific electrical conductivity-κ of the ILCSs water solutions were identified using “Anion-4120” conductometer. The 0.001–1% wt. solutions were prepared using water as solvent and after 48 h their κ values were determined [33]. The conductivity values of the used deionized water were in the interval of 1.5–1.8 μS/cm with ±0.25 μS/cm deviation.

2.5. Sand pack flooding experiment

A sand pack flooding experiment was conducted to determine the EOR capability of the synthesized polyelectrolyte complexes. Quartz sand was placed in a glass tube with a length of 64 cm and an inner radius of 0.8 cm, and pre-dried at 120–130°C. The exploited sand has a grain size in a range of 0.1–0.31 mm. The 41.5% fraction of the grain sizes is 0.1–0.16, 52.1% is 0.16–0.2 mm, and the remaining 6.4% is 0.2–0.31 mm. The sand in the tube was saturated with oil via a dropping funnel located at a height of about 1.4–1.5 m. After the saturation stage, the oil layer in the tube was characterized. It is necessary to emphasize that the air permeability coefficient of the assembled rock model in the conducted experiments has values in the range of 0.1–1.0 Darcy [34]. Next, the adsorbed oil by the sand was displaced with an aqueous solution of the studied polyelectrolyte complexes. Through the certain time range, the solution passed through the tube that contain send with adsorbed oil. Based on these studies, the dependence of the oil displacement coefficient on the pore volume of the solution is determined.

2.6. Determination of the size of the aggregates by the dynamic light scattering (DLS) measurement

Micellar aggregates size measurements were performed on a Particle Size Analyzer (Japan, HORIBA LB-550) with a 650−nm laser diode having a power of 5 mW served as a light source, at 25 °C temperature. Each solution was evaluated four times, then the automatic correlation function was analyzed by CONTIN to obtain the hydrodynamic diameter (D_h) [17]. The size measurement range was from 1 nm to 6000 nm.

2.7. Tests on the biocidal capability of the obtained ILCSs

The serial broth macro-dilution method was used with serial dilution for the determination of the minimum inhibitory concentration (MIC) [14,33]. Investigations were performed in test tubes comprising an ILCS at various concentrations and physiological saline. As a test crop, two Gram−positive bacteria (*Bacillus anthracoides, Staphylococcus aureus*), three Gram−negative (*Pseudomonas aeuroginosa, Klebsiella pneumoniae, Escherichia coli*) bacteria and fungus (*Candida ablicans*) laboratory strains were used. Suspensions were prepared, which containing 1 ml of the physiological solution 500 million microbial cells in each microbe daily culture. One drop of this suspension was included in each trial tube. After 10.0, 20.0, 40.0, and 60.0 min, samples were taken from each trial tube with a bacteriological loop were sown on the appropriate nutrient media (for the bacteria, meaty-peptone agar medium and, for *Candida ablicans*, Sabouraud agar medium was created). The sown specimens were incubated at 37 °C for 24 h.

3. Results and discussion

3.1. Colloidal-chemical characteristics of the obtained ILCSs and their polyelectrolyte complexes in aqueous solution

The values of surface tension (γ) at the border of the air and aqueous ILCS solutions were measured by tensiometric method. The graph that shows surface tension (γ) versus the surfactants concentration was

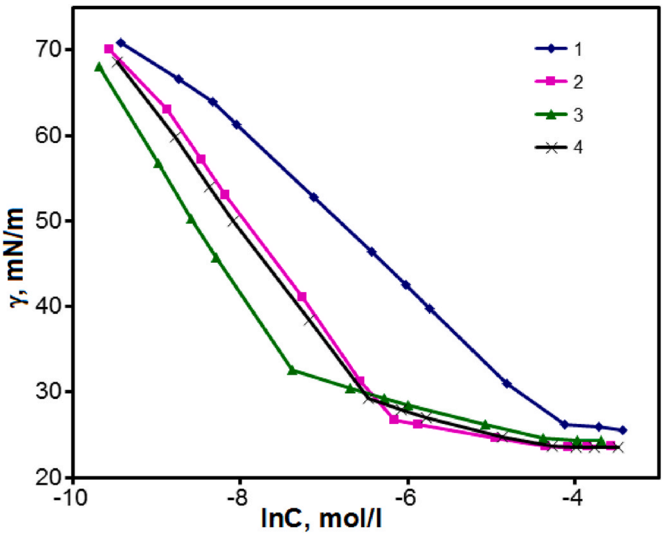
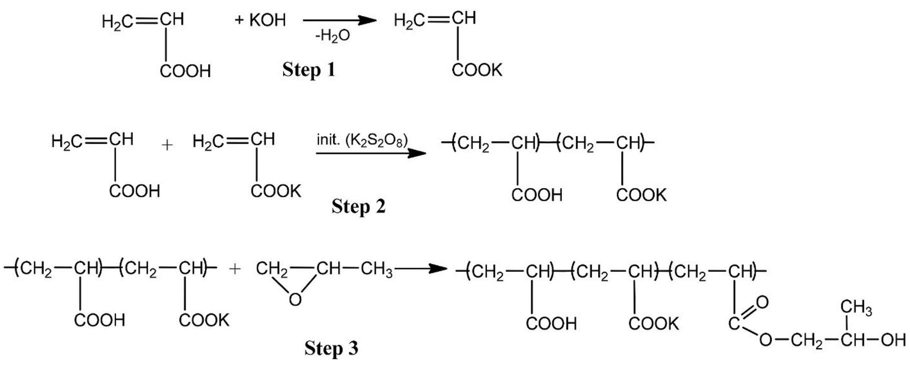


Fig. 1. Surface tension plots of C_{12}MEA (1), C_{12}DEA (2), C_{12}TEA (3), and C_{12}MAE (4) in water solution at 298 K versus ln(C) of ILCSs.



designed based on the obtained values (Figs. 1 and 2). As seen from the graphs, the concentration of those ILCSs in the solution rises, when γ decreases. Stabilization is observed after a certain value of concentration. The starting point of the stabilization part is taken as critical micellization concentration (CMC). But, in the case of the ILCSs obtained based on 2-(methylamino)ethanol, di(2-hydroxyethyl)amine, tris(2-hydroxyethyl)amine, after a certain concentration, γ values decrease slowly and, then, stabilization is observed. After the slow concentration decline, stabilization is observed. The starting point of concentration stabilization was accepted as CMC. Such a form of surface tension isotherm indicates observation of premicellar aggregates. The CMC values for the obtained ILCSs were described in Table 1. As seen from this table, the CMC values for the ILCSs based on mono(2-hydroxyethyl) amine, di(2-hydroxyethyl)amine, and tris(2-hydroxyethyl)amine are 16.6, 12.8, and 12.5 mM, respectively. As is seen from the obtained values, increasing the number of $–CH_2CH_2OH$ groups bonded to the nitrogen atom in these cationic surfactants the CMC values decline. A similar pattern is observed in the case of the cationic ammonium-type surfactants with the same hydrophilic fragments obtained based on 1-bromotetradecane [33], 1-chlorododecane [3], 1-bromononane [31], and 1-iodononane [31]. In the case of the quaternary ammonium-type ILCSs bearing $–CH_3$, $–CH_2CH_3$, and $–CH_2CH_2OH$ groups at the N atom, increasing the number of CH_2CH_2OH groups bonded to the N atom in the ILCSs head group, rises CMC value. When the $–CH_3$ group is replaced by the $–CH_2CH_3$ group, the situation is the same, i.e., the values of CMC were elevated. As shown in Table 1, the CMC value for the surfactants containing trimethylammonium group is smaller than that of the monoethanolammonium group-containing surfactants, but higher than those of the other surfactants.

As is seen from Fig. 1, in the case of the ILCSs synthesized based on DEA, TEA, and MAE, the concentration elevation after the CMC, results in decline in γ values. Similar results were observed in some gemini cationic surfactant having a long spacer chain (s ≥ 8). X. Huang et al. [39] confirmed by different physico-chemical methods that in gemini cationic surfactants with long spacers, at low concentrations, due to intermolecular H bonding between hydroxy-groups present in a hydrophilic fragment, dimers are formed and transformed into aggregates. The authors mention that premicellar aggregates formed in such surfactant types. Here is sharp decrease of γ at low concentrations is followed by the slow γ decline with an increase concentration. It was evident that, after the CMC concentration, stabilization occurs. Some scientists mark the formation of micelles at two different concentrations (CMC_1 and CMC_2) when plotting graphs [40,41].

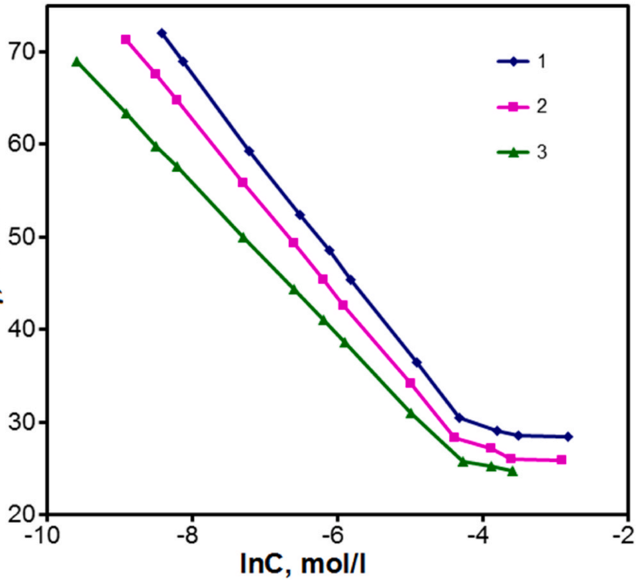


Fig. 2. Surface tension plots of C_{12}DMEA (1), C_{12}MDEA (2), and C_{12}DEAE (3) in water solution at 298 K versus ln(C) of ILCSs.

The concentration, where a sharp decline of surface tension is recorded (Fig. 1), which referred to critical aggregation concentration (CAC). As is seen from Fig. 1, the CAC values for the surfactants having diethanolammonium, methylethanolammonium, and triethanolammonium head-groups are 2.11 mM, head- −1.54 mM and, −0.63 mM, respectively. The formation of CAC in surfactants is the presence of NH^+ and OH groups, which are capable to generate intermolecular hydrogen bonding. Diethanolammonium and methylethanolammonium head groups containing surfactants possess both NH^+ and OH groups whereas, in the triethanolammonium head group, there are three OH groups. These groups are capable to form multiple hydrogen bonding. In the similar surfactants type, the formation of N···H-O, N-H···O, and O-H···O hydrogen bonds are confirmed by the X-ray structural analysis method [42]. X. Huang [39], A.B.Mirgorodskaya [5] and other scientists [43] also mention the formation of hydrogen bonding between head groups.

The nature of a hydrophilic fragment of ILCS impacts the value of maximum adsorption at the water-air interface -Γ_{max} as well as the minimal surface area occupied by a polar head group of one cationic surfactant molecule, A_{min}.

The Γ_{max} value for ILCSs is calculated using surface tension isotherm and Gibbs equation [44]:

$$\Gamma_{max} = -\frac{1}{nRT} \lim_{C \to C_{CMC}} \frac{d}{dlnC} \tag{1}$$

where R – is universal gas constant, (dγ/dlnC) is surface activity at absolute temperature (T), n indicates the number of ions in the ILCS molecule. In the obtained ILCSs, positively charged N^+-ammonium ion and negatively charged bromide ion-Br^- are available. So, n = 2.

According to Γ_{max} value, using the formula given below, A_{min} (in nm^2) can be calculated [44]:

$$A_{min} = 10^{16} / N_A \Gamma_{max} \tag{2}$$

In Table 1, the calculated values of Γ_{max} and A_{min} are given. As seen from the table, increasing number of $–CH_2CH_2OH$ fragments in the polar part of the surfactant molecule (according to the series: monoethanolammonium < diethanolammonium < triethanolammonium) increases Γ_{max} value while A_{min} value decreases. In the case of the ILCSs head group, which contain CH_3 and $–CH_2CH_2OH$ groups increasing $–CH_2CH_2OH$ fragments, i.e., dimethylethanolammonium is replaced with methyldiethanolammonium head-group, Γ_{max} decreases but A_{min} rises. The same pattern is observed when dimethylethanolammonium is substituted by diethylethanolammonium head group. For trimethylammonium head-group containing surfactant (namely in dodecyl trimethyl ammonium bromide - C_{12}TMA), Γ_{max} is much higher than the other tested surfactants.

The formed surface pressure-π at the air and the surfactants aqueous solution border is calculated by the following equation [44]:

$$\pi_{CMC} = \gamma_0 - \gamma_{CMC} \tag{3}$$

where γ_0 – surface tension at the water–air border, γ_{CMC} – surface tension at CMC of the ILCS at the border of air and the surfactants aqueous solution surface. For the obtained ILCSs, surface pressure varies in the range of 41.6–48.3 mN/m. The highest surface tensions data were obtained DEA and MAE based ILCSs. If $–CH_3$ substituents in trimethylammonium head-group are replaced by $–CH_2CH_2OH$ groups, the π_{CMC} value rises.

Adsorption efficiency at the water-air boundary (pC_{20}) is one of the essential parameters for comparing the adsorptivity of surfactants at the interface. The big adsorption efficiency value indicates that, the lower ILCS concentration, good adsorptivity of the surfactant at the interface and the surface tension value is going to decrease. pC_{20} can be calculated according the following equation [44]:

Table 1

Surface properties of the obtained ionic-liquid cationic surfactants in aqueous solution at 298 K[a].

Surfactants	β	CMC[b,c] × 10^3, mol·dm^{-3}		Γ_{max} × 10^{10}, mol·cm^{-2}	A_{min} × 10^2, nm^2	π_{CMC}, mN·m^{-1}	γ_{CMC}, mN·m^{-1}	pC_{20}	ΔG_{mic}, kJ·mol^{-1}	ΔG_{ad}, kJ·mol^{-1}
$C_{12}TMA$	0.78[j] 0.79[k]	15.4[j] 14.6[k]	14.7[d] 15.0[e] 14.5[k]	3.77[j] 2.85[k]	79[d] 44[j] 58.15[k]	36.4[j] 34.43[k]	36.4[j]	5.25[j]	−40.73[e]	48.63[k]
$C_{12}MEA$	0.45	16.1	16.6	1.90	87.5	45.8	26.2	3.06	−28.15	−30.56
$C_{12}DEA$	0.47	12.7	12.8	2.70	61.4	48.3	23.7	3.52	−30.50	−32.29
$C_{12}TEA$	0.56	12.0[i] 12.0	13.0[i] 12.5	3.12	53.4	47.4	24.6	3.78	−32.46	−33.99
$C_{12}MAE$	0.40	12.5	12.7	2.65	62.6	48.3	23.7	3.49	−28.74	−30.56
$C_{12}DMAE$	0.764[d] 0.66[f] 0.703[g] 0.732[h] 0.77	13.07[g] 13.3	13.4[d] 13.2[e] 14.5[f] 13.87[g] 14.3[h] 13.3	2.04	82[d] 81.3	41.6	30.4	2.95	−36.42[d] −41.37[e] −35.21[g] −35.5[h] −36.56	−38.59
$C_{12}MDEA$	0.758[d] 0.701[g] 0.71	12.89[g] 12.5	12.3[d] 12.46[e] 12.92[g] 12.3	1.93	86[d] 86.1	43.7	28.3	2.99	−36.65[d] −41.65[e] −35.26[g] −35.65	−37.91
$C_{12}DEAE$	0.683[h] 0.70	13.9	14.9[h] 14.0	1.65	100.6	46.3	25.7	3.27	−34.3[h] −34.90	−37.70

[a] The standard uncertainties-u are u(T) = 0.1 K and u(p) = 50 kPa. The combined expanded uncertainties U_c are $U_c(\beta) = 0.01$, $U_c(CMC) = 10^{-4}$ mol/dm^3, $U_c(\gamma) = 0.1$ mN/m, $U_c(A_{min}) = 0.5$ Å^2, $U_c(\Gamma_{max}) = 0.02$ mol/cm^2, $U_c(\pi) = 0.1$ mN/m, and $U_c(pC_{20}) = 0.002$ (0.68 level of confidence).

[b] CMC - surface tension method, in the column on the left.

[c] CMC - electroconductivity method, in the column on the right.

[d] Ref. [20].

[e] Ref. [35].

[f] Ref. [19].

[g] Ref. [21].

[h] Ref. [23].

[i] Ref. [36].

[j] Ref. [38].

[k] Ref. [37].

$$pC_{20} \cong -\log_{10} C_{(-\Delta\gamma=20)} \tag{4}$$

The pC_{20} values of the synthesized ILCSs are shown in Table 1. As can be seen from the table, these values vary in the range of 2.95–3.78. Increasing the –CH_2CH_2OH fragments in the ILCSs head group, increases the value of pC_{20}. Among the obtained ILCSs, the largest adsorption efficiency is displayed by the one having triethanolammonium head group.

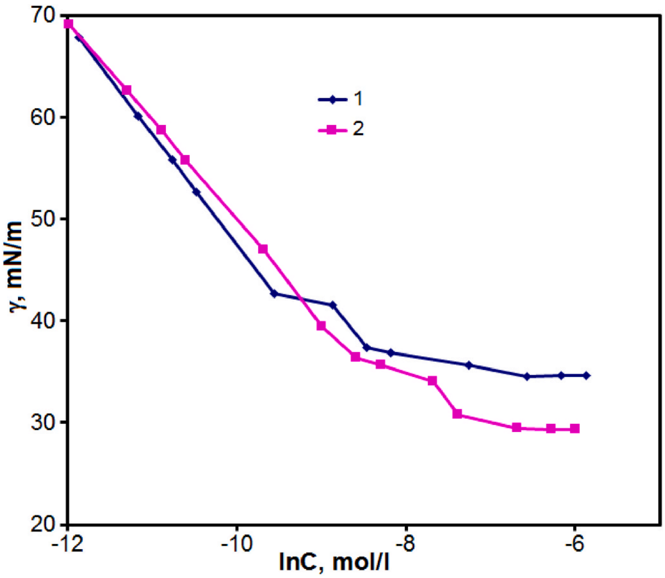


Fig. 3. Surface tension plots of ILCSs (1-$C_{12}DEA$, 2-$C_{12}TEA$) in the presence of 0.05% added polymers.

The surface tension values at the air-water boundary of the synthesized complexes based on $C_{12}DEA$ and $C_{12}TEA$ were determined (Fig. 3). The analysis of the obtained surface tension isotherms shows that there are three important points in the isotherm: T_1, T_2, and T_3. T_1 indicate the initiation of interaction between the surfactant with polyelectrolyte and is called critical aggregation concentration, T_2 is the saturation concentration of the polymer chain with surfactant molecules, and T_3 is called the CMC of the polymer-surfactant complexes. As can be seen from Table 2, the value of T_1 is smaller in the complex obtained based on $C_{12}DEA$. Since $C_{12}DEA$ is a ternary ammonium salt, the COO^- groups can bind to the substance easily. Bigger T_1-T_2 difference results in the wider interaction interval. In the complexes obtained based on $C_{12}TEA$, the interaction occurs in a small concentration range, the T_3 is smaller than for the complex obtained based on $C_{12}DEA$. This is due to three –CH_2CH_2OH groups in the head group in the case $C_{12}TEA$. If we compare the values of Γ_{max} and A_{min} given in Tables 1 and 2, we can see that when surfactant forms a complex with polymer, the value of Γ_{max} decreases, while the value of A_{min} increases.

3.2. *Specific electroconductivity of the obtained ILCSs*

The ILCSs aqueous solutions were prepared via dilution to the different concentrations and the specific electroconductivity values were determined by the conductometric method. Respective isotherms were designed based on the obtaining κ values (Figs. 4 and 5). It can be seen from these figures that, at low concentrations, κ values increase slowly. The related crossing point of the κ vs C variables is taken as the CMC value. According to the ratio of the slopes after (S_2) and before (S_1) CMC, a dissociation degree (α) of surfactants can be calculated:

$$\alpha = S_2/S_1 \tag{5}$$

A degree of binding (β) of the surfactant counter-ion is determined as

Table 2
Surface properties of the ILCSs/polyelectrolyte complexes in aqueous solution at 25 °C.

Complexes	$T_1 \times 10^4$, mol·dm^{-3}	$T_2 \times 10^4$, mol·dm^{-3}	T_1-$T_2 \times 10^4$ mol dm^{-3}	$T_3 \times 10^3$, mol·dm^{-3}	$\Gamma_{max} \times 10^{10}$, mol·cm^{-2}	$A_{min} \times 10^2$, nm^2	γ_{CMC}, mN·m^{-1}
C_{12}DEA-poly. com.	0.71	1.4	0.69	1.41	2.20	75.6	34.6
C_{12}TEA-poly. com.	1.9	4.6	2.7	1.26	1.96	84.6	29.3

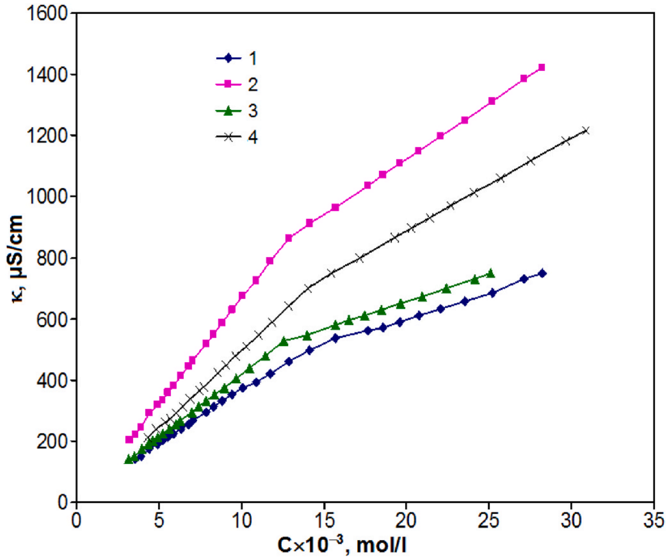


Fig. 4. Graphics of specific electroconductivity dependence ILCS concentration for water solutions of C_{12}MEA (1), C_{12}DEA (2), C_{12}TEA (3), and C_{12}MAE (4) at 298 K.

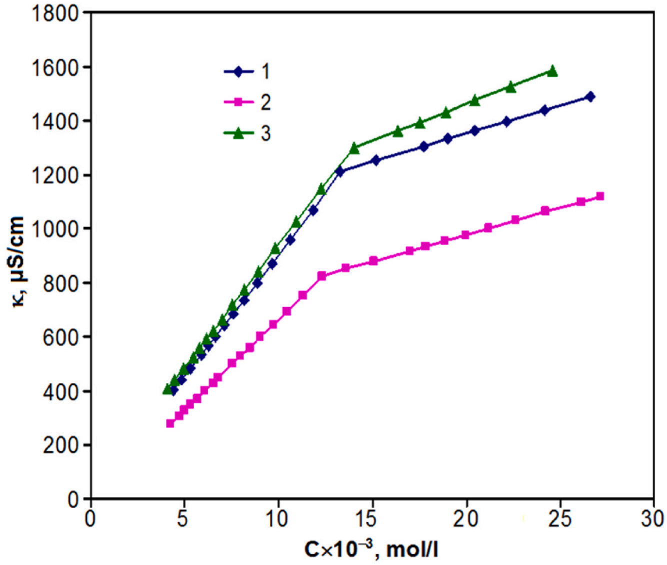


Fig. 5. Graphics of specific electroconductivity dependence ILCS concentration for water solutions of C_{12}DMEA (1), C_{12}MDEA (2), and C_{12}DEAE (3) at 298 K.

follows:

$$\beta = 1 - \alpha \tag{6}$$

In Table 1, the binding degree of the obtained ILCSs was shown. This table shows that the nature of the head-group in these ILCSs impacts the value of their dissociation degree. β rises with increasing the $-CH_2CH_2OH$ fragments in the head group. Likewise, in the case of the surfactants containing both $-CH_3$ and $-CH_2CH_2OH$ groups in the hydrophilic part, elevation of β is observed when the remaining methyl groups displaced with $-CH_2CH_2OH$ groups. A similar trend is observed among surfactants with nonylmonoethanol-, nonyldiethanol-, nonyltriethanolammonium bromide (or iodide) [31], tetradecylmonoethanol-, tetradecyldiethanol-, and tetradecyltriethanolammonium bromide [33]. In dimethylethanolammonium head group, the substitution of $-CH_3$ groups with $-CH_2CH_3$ leads to decrease in β value.

3.3. Size dimensions of aggregates of the obtained ILCSs

Figs. 6 and 7 show the dimensions of aggregates formed at CMC and 3 × CMC in the ILCSs aqueous solution derived from MEA, DEA, and TEA. As shown in Fig. 6, the mean diameter of the aggregates formed at CMC of C_{12}MEA, C_{12}DEA, and C_{12}TEA is 100 nm, 50 nm, and 70 nm, respectively. The three-fold increase in concentration led to a slight downshift in the size of the aggregates of C_{12}DEA and a slight increase in the case of C_{12}TEA. For C_{12}MEA, however, changing solution concentration from CMC to 3 × CMC resulted in a 1.5-fold increase in the mean size and formation of the small number of larger aggregates. Figs. 8 and 9 show the dimensions of aggregates formed at CMC and 3 × CMC in the aqueous solution of ILCSs obtained from MAE, MDEA, DMAE, and DEAE. As it can be seen from the figures, the mean size of the aggregates formed in C_{12}DMAE in aqueous solution has a very small value (4–5 nm), the 3-fold increase in concentration has almost no effect on the mean size of the aggregates. While the size of the aggregates in the CMC solution of C_{12}MAE is 75 nm, in 3 × CMC the diameter decreases to 60 nm. For C_{12}MDEA, a bimodal peak is observed at CMC. A three-fold increase in concentration reduces the mean size of the aggregates to 60 nm, and the bimodal peak becomes a monomodal peak. In C_{12}DEAE solutions, the mean size of the aggregates decreased from 40 nm to 9 nm as a result of a three-fold increase in concentration.

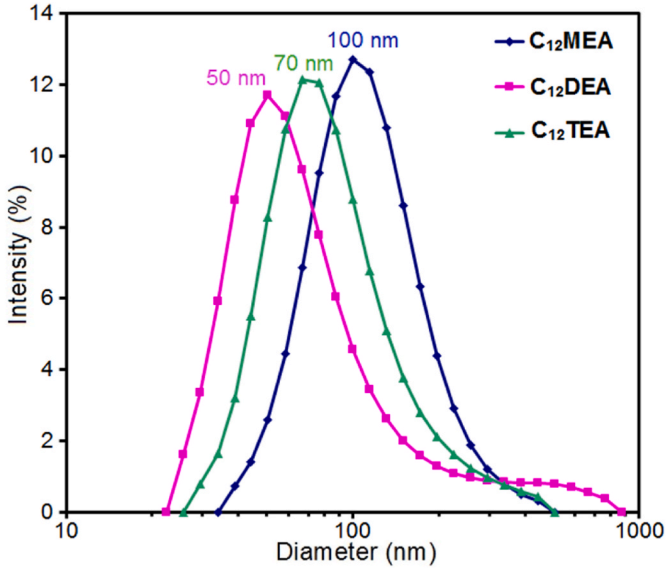


Fig. 6. The particle size distribution range of the ILCSs at CMC (25 °C).

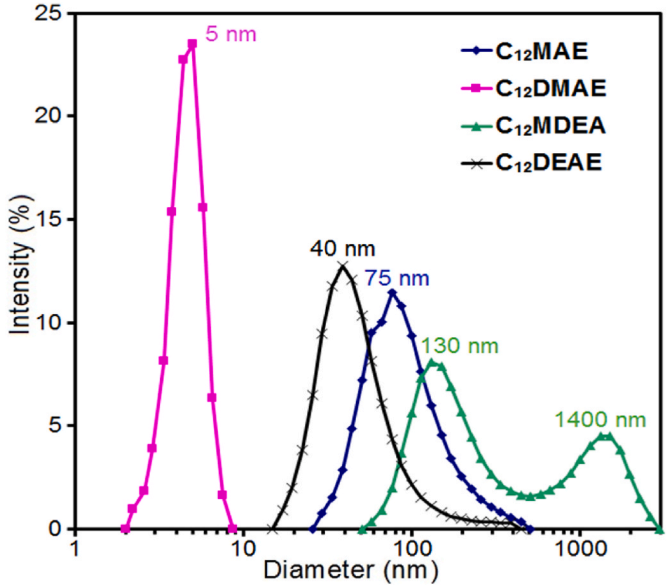


Fig. 7. The particle size distribution range of the ILCSs at CMC (25 °C).

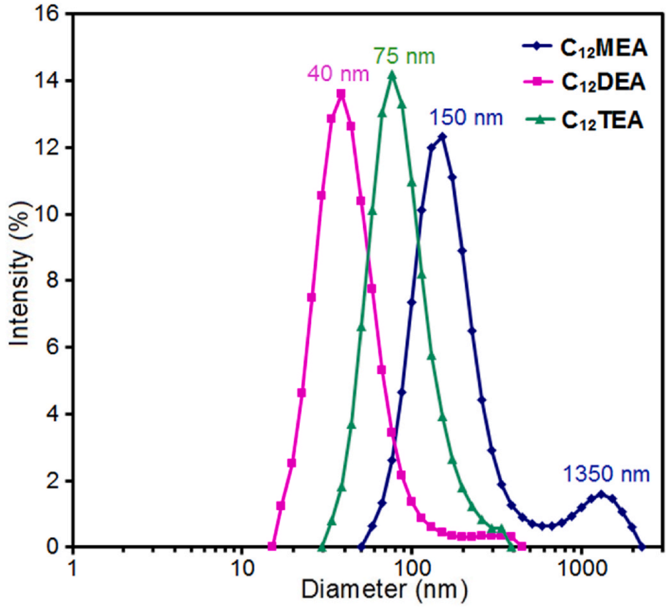


Fig. 8. The particle size distribution range of the ILCSs at 3 × CMC (25 °C).

3.4. Thermodynamic properties of the obtained ILCSs

Standard Gibbs energy values for the micelle formation process were computed by the following equation using CMC and the degree of binding values of the ILCSs [44]:

$$\Delta G^{\circ}_{mic} = (1+\beta)\, RT\ln X_{CMC} \tag{7}$$

where X_{CMC} is the mol fraction of critical micellization concentration, T is standard absolute temperature (298 K), and R is universal gas constant. X_{CMC} = CMC/55.4, where critical micellization concentration is expressed in mol/L, 55.4 takes origin from 1 L of distilled water which matches 55.4 mol of water at 25 °C.

Standard Gibbs energy values for the adsorption process of the synthesized ILCSs at the water–air interface were calculated using the following equation [44]:

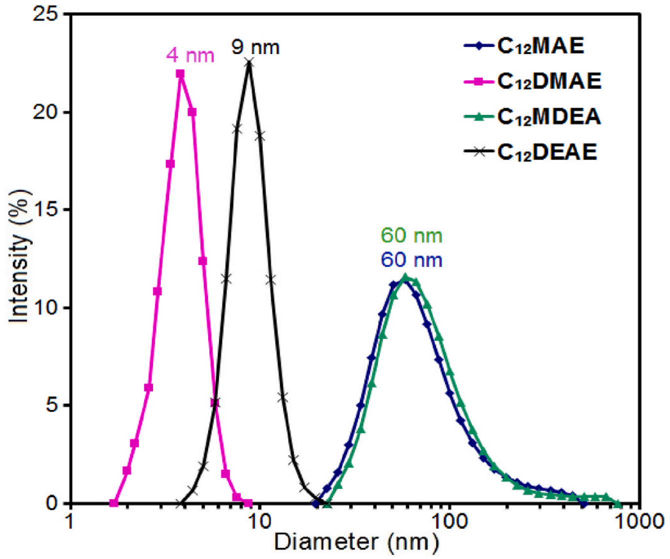


Fig. 9. The particle size distribution range of the ILCSs at 3 × CMC (25 °C).

$$\Delta G^{\circ}_{ad} = (1+\beta)RT\ln X_{CMC} - 0.6023\pi_{CMC}A_{CMC} \tag{8}$$

where A_{CMC} is A_{min} in Å^2 unit, π_{CMC} is the surface pressure (in mN/m unit) at CMC, and the border of the air and ionic surfactant aqueous solution.

The calculated values of ΔG°_{mic} and ΔG°_{ad} are included in Table 1. As shown in the table, both values are negative. It means that the adsorption and micellization processes of the synthesized ILCSs occur spontaneously. Meanwhile, ΔG°_{ad} values are more negative. Those particular points related to the fact that adsorption processes are more spontaneous than micellization processes.

As seen from Table 1 ΔG°_{mic} and ΔG°_{ad} values decline as the number of $–CH_2CH_2OH$ fragments increases in the surfactants head group containing mono-, di- and triethanolammonium cations. Among the surfactants having both methyl and $–CH_2CH_2OH$ fragments in the head group, complete substitution of $–CH_3$ group with $–CH_2CH_2OH$ or ethyl group, the values of ΔG°_{mic} and ΔG°_{ad} rise. Therefore, the surfactants with such head groups, ($-CH_2CH_2OH$ and CH_2CH_3 groups) adversely impact adsorption and micelle formation processes. In the synthesized ILCSs, the replacement of the $–CH_3$ with the $–CH_2CH_2OH$ group improves water-solubility of the surface-active substance which results in the weakening of spontaneity of adsorption and micellization processes.

3.5. Biocidal properties of the synthesized ILCSs

As seen in Table 3, among the synthesized ILCSs, $C_{12}DEA$ has the strongest antibacterial activity. A solution of this salt with a concentration range of 0.63–5.0 mg/ml completely stops the growth of all *Staphylococcus aureus, Bacillus anthracoides, Pseudomonas aeuroginosa, Escherichia coli, Klebsiella pneumoniae* bacteria, and even *C. albicans* fungus after 10 min period. Against *Bacillus anthracoides* bacterium, at concentrations in the range 1.25–5.0 mg/ml, this surfactant fully suppresses the activity of the mentioned bacterium starting from 10 min, whereas at the concentration of 0.63 mg/ml, after 20 min the growth of the indicated bacterium becomes sharply weakened.

$C_{12}MAE$ also exhibits a high antimicrobial capability. Thus, this surfactant prevents the development of *C. albicans* fungus and *Staphylococcus aureus*, *Bacillus anthracoides* bacteria at concentrations of 0.63–5.0 mg/ml. At a concentration of 0.63 mg/ml, $C_{12}MAE$ fully stops the growth of *Klebsiella pneumonia* after 20 min, completely inhibiting the activity of *Pseudomonas aeuroginasa* and *Echerichia coli* bacteria after 40 min.

Table 3
Biocidal properties of the obtained ILCSs by the method of consequent dilution (a = 5.0, b = 2.5, c = 1.25, d = 0.63 mg/ml).

Microorganisms	τ, min	$C_{12}MEA$				$C_{12}DEA$				$C_{12}TEA$				$C_{12}MAE$				$C_{12}DMEA$				$C_{12}MDEA$				$C_{12}DEAE$			
		a	b	c	d	a	b	c	d	a	b	c	d	a	b	c	d	a	b	c	d	a	b	c	d	a	b	c	d
Escerichia coli	10	-	±	±	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	-	-	+	+	-	-	-	+
	20	-	±	±	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	-	-	-	+	-	-	-	-
	40	-	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	-	-	-	-
	60	-	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	-	-	-	-
Pseudomonas aeruginosa	10	-	-	+	+	-	-	-	-	-	-	±	±	-	-	±	+	-	-	+	+	-	-	-	±	-	-	-	+
	20	-	-	+	+	-	-	-	-	-	-	-	±	-	-	-	+	-	-	-	+	-	-	-	-	-	-	-	+
	40	-	-	+	+	-	-	-	-	-	-	-	±	-	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-
	60	-	-	+	+	-	-	-	-	-	-	-	±	-	-	-	±	-	-	-	-	-	-	-	-	-	-	-	-
Klebsiella pneumoniae	10	-	-	+	+	-	-	-	-	-	-	-	-	-	-	-	+	-	-	-	+	-	-	-	±	-	-	-	+
	20	-	-	+	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	±	-	-	-	-
	40	-	-	+	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	60	-	-	+	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Staphylococcus aureus	10	-	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	20	-	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	40	-	-	-	±	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	60	-	-	-	±	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Candida albicans	10	-	-	-	±	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	20	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	40	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	60	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Bacillus anthracoides	10	+	+	+	+	-	-	-	+	-	-	±	±	-	-	-	-	-	-	-	-	+	+	+	+	-	+	+	+
	20	+	+	+	+	-	-	-	±	-	-	±	±	-	-	-	-	-	-	-	-	+	+	+	+	-	+	+	+
	40	+	+	+	+	-	-	-	±	-	-	-	±	-	-	-	-	-	-	-	-	+	+	+	+	-	+	+	+
	60	+	+	+	+	-	-	-	±	-	-	-	±	-	-	-	-	-	-	-	-	+	+	+	+	-	±	+	+

"+" – growth "±" – very weak growth "-" – no growth.

$C_{12}MDEA$ and $C_{12}DEAE$ demonstrate no antibacterial properties against *Bacillus anthracoides* at concentrations of 0.63–2.5 mg/ml. But they are effective toward the other bacteria and *Candida albicans* fungus at concentrations of 1.25–5.0 mg/ml.

$C_{12}MEA$ prevents the growth of only *Candida albicans* fungus. Complete inhibition is observed at all applied concentrations. Against bacteria (except *Bacillus anthracoides*), it is effective only at high concentrations (5.0 and 2.5 mg/ml). Regarding *Bacillus anthracoides*, $C_{12}MEA$ is ineffective at all chosen concentrations. So, $C_{12}MEA$ demonstrates only an antifungal effect.

In the case of the ILCSs containing both –CH_3 and –CH_2CH_2OH groups, the substitution of the –CH_3 with –CH_2CH_2OH and –CH_2CH_3 groups weakens antibacterial properties against *Bacillus anthracoides*.

Minimum inhibitory concentration was determined for ILCSs with high antimicrobial properties ($C_{12}MAE$, $C_{12}DEA$ and $C_{12}TEA$) (Table 4). It can be seen from the table that $C_{12}MAE$ and $C_{12}DEA$ can completely stop the growth of *Escerichia coli* and *Staphylococcus aureus* bacteria at lower concentrations. These substances are more effective than $C_{12}TMA$ (except *Pseudomonas aeruginosa*). The antimicrobial effects of ILCSs were widely studied [47]. Cationic surfactants are initially absorbed on the outer bacteria cell walls via electrostatic interactions, which occur between cationic quaternary ammonium groups and anionic bacterial strain groups. It follows interaction of hydrophobic groups in the ILCSs hydrocarbon chains with related hydrophobic part of bacteria cell wall. The interactions and potential osmotic deterioration of the cell resulted in outflow of the intracellular components and eventually, destroy inner bacteria membrane. As a result, cell death is inevitable because of adverse impact of such interactions, which then follows dehydration of bacteria membranes.

3.6. Oil recovery of the ILCSs neutralized with propoxylated polyacrylic acid complexes

One of the main properties of surfactants and polyelectrolytes for application in the EOR industry is because of their good solubility in brine. Although many surfactants and polyelectrolytes have high oil displacing abilities, their application is limited because they form precipitation in the brine/oil wells. The complexes obtained based on the neutralization of $C_{12}DEA$ and $C_{12}TEA$ with propoxylated polyacrylic acid do not precipitate in brine. Therefore, the oil displacement capabilities of those complexes were studied. 0.05 wt% solution of polyelectrolyte-surfactants complexes was prepared using sea water. 10 and 25% of the COO^- groups containing polyelectrolyte were mixed with surfactants mentioned above. The generated polyelectrolyte-surfactants complex was applied instead of brine in the secondary flooding. It was determined that when brine is used in secondary flooding, the oil recovery is 41%, but when the complex based on $C_{12}DEA$ (25% added due to free COO^- groups) is applied, the oil recovery increases up to 80%. When the solution of the complex obtained based on $C_{12}TEA$ (10% added due to free COO^- groups) is applied in secondary flooding, the oil recovery reaches 82%, and when the amount of $C_{12}TEA$ is taken up to 25% of the free COO^- groups, the oil recovery reaches 86%. Studies show that recovery increases by 41–45% when

Table 4
Minimum inhibitory concentration (MIC) of the cationic surfactants against microorganisms (in μg/ml).

Microorganisms	$C_{12}DEA$	$C_{12}TEA$	$C_{12}MAE$	$C_{12}TMA$[a]
Escerichia coli	5	1250	5	40
Pseudomonas aeruginosa	1250	1250	2500	346
Klebsiella pneumoniae	78	78	78	NT[b]
Staphylococcus aureus	5	10	2	8
Candida albicans	78	5	78	NT
Bacillus anthracoides	2500	1250	10	NT

[a] $C_{12}TMA$-dodecyl trimethyl ammonium bromide Ref. [45,46].
[b] NT - Not tested.

polyelectrolyte-surfactant complexes were applied instead of brine in secondary flooding. Increasing the number of –CH_2CH_2OH groups in the head fragment of the surfactant increases the oil displacement capabilities.

To the best of our knowledge, there is no well-known oil recovery process with ionic liquids [28]. In the recommended method, the interaction mechanism can be explained with the addition of ILCS to oil phase weakening the attraction at the interface of crude oil and water or crude oil rock. As a result of this interference by ILCSs in the interfaces, reduces interfacial tensions, and eventually helps the crude oil is easily mobilize through pores.

4. Conclusion

ILCSs were synthesized based on 1-bromododecane and amines containing –CH_3, –CH_2CH_3, and –CH_2CH_2OH fragments. Depending on the structure of the ILCSs head group, the changes in the colloidal chemical parameters was determined. Namely, increasing the number of –CH_2CH_2OH fragments in the head group, decreases the values of CMC, ΔG°_{ad}, ΔG°_{mic}, and A_{min}, and increases Γ_{max} and β values. In the case of the ILCSs containing both –CH_3 and –CH_2CH_2OH groups, replacing the remaining methyl with –CH_2CH_2OH groups resulted in reducing the CMC and β values and elevation of ΔG°_{mic} and ΔG°_{ad} values.

Studies of the antibacterial and antifungal properties of the obtained ILCSs revealed that C_{12}DEA, and C_{12}MAE exhibited higher effectiveness. The MIC value of these ILCSs was smaller than that of dodecyl trimethyl ammonium bromide.

Experiments of flooding showed that the oil recovery increased two-fold when brine solution was replaced with C_{12}DEA and C_{12}TEA containing propoxylated polyacrylic complexes.

CRediT authorship contribution statement

Shafiga M. Nasibova: Investigation, Resources, Writing – original draft. **Ravan A. Rahimov:** Writing – review & editing, Supervision, Methodology. **Sevda A. Muradova:** Investigation, Data curation. **Yusif Abdullayev:** Writing – original draft, Validation, Formal analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors are unable or have chosen not to specify which data has been used.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.matchemphys.2022.127268.

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