

***Investigation on the interaction between sodium dodecylsulfate and cationic polymer by using conductometric, densimeter and zeta potential technique***

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**Abstract:**

The interaction of solution of poly (diallyldimethylammonium) chloride, PDADMAc and sodium dodecylsulfate, SDS has been studied using conductometric, densimeter and zeta potential methods. the critical micellization concentration (cmc) was determined with conductance results and increased with the addition of PDADMAc comparing with solutions containing pure SDS. Further the degree of dissociation,  $\alpha$  and thermodynamic parameters viz. free energy of micellization,  $\Delta G_m^0$  and entropy of micellization,  $\Delta S_m^0$  has been evaluated. these results demonstrated that surfactant/polymer complex is governed by both electrostatic and hydrophobic interactions. The apparent molar volume,  $V_\phi$  and adiabatic compressibility,  $K_{\phi;s}$  have been determined for the SDS solution from density and speed sound data. the negative charge of the SDS solutions with polymer shows that these complexes probably do not contain surfactant in the form of micelles.

**Keywords:** Ionic surfactant; polymer solution, critical micellization concentration, Zeta Potential

**1. Introduction**

the effect of additives such as salt or polyelectrolyte on the properties of aqueous surfactant solutions has been a subject of intense research due to their extensive commercial applications and researcher viewpoints [1-7]. The surfactant-polymer mixtures have potential for domestic, industrial and technological applications such as, foods, paints, drug delivery, coatings and laundry products [8, 9]. Hence, the interaction of oppositely charged polymers with ionic surfactant is more convoluted comparing with nonionic polymers- surfactants. It was usually observed that at certain surfactant concentrations such interaction leads to precipitation of a polymer- surfactant complex. This precipitation was characterized for various oppositely charged polyelectrolyte-surfactant systems [10-15]. Phase separation could be prevented by addition of more nonionic groups to the polymer or to the surfactant, as reported by Bronich et al. and Dubin et al. [16-18]. Polyelectrolyte-surfactant interactions are controlled by a

balance between hydrophobic and electrostatic interactions and are modulated by temperature and ionic strength. While interactions between non ionic polymers and ionic surfactants rely mainly on hydrophobicity, attractive electrostatic forces dominates in the case of polyelectrolyte and oppositely charged surfactants in the presence of an oppositely charged polyelectrolyte, surfactant starts to form aggregates with polyelectrolyte as the surfactant concentration exceeds a critical value referred as a critical concentration aggregation. In this process, the counter ion of polyelectrolyte is replaced by the surfactant ion and the surfactant form a neutral ion pair [19]. This surfactant binding mechanism related cooperative and non cooperative steps as well. This cooperative binding depends on a range of factors, such as the length of carbon chain of surfactant, the salt concentration and the polyion charge density [20].

Poly(diallyldimethylammonium chloride) (PDADMAC), an important commercial water-soluble cationic functional polymer, is useful in water treatment as a flocculant or coagulant for the removal of organic and mineral contaminants such as arsenic, etc.[21-22] It is used in the textile and paper industries for making antibacterial fiber and to improve the wet strength of papers[23]. In several of these applications, surfactant is added in order to provide emulsification capacity or to control air/water interfacial or because the interaction between PDADMAC and SDS is important in determining the properties of these systems. Kong et al have used conductometric and rheometric techniques to examine SDS/PDADMAC complex [24]. Maroi et al. have applied the potentiometric and fluorimetric methods to the SDS/PDADMAC mixed solutions to evaluate the critical aggregation concentration [25].

Physico chemical of surfactant/ polymer systems are very important for industrial processes. The aim of this paper is to report physico chemical properties (conductance, densities and speed sound and zeta potential) for the following SDS-PDADMAC mixed solutions at  $T=(298.15, 308.15, \text{ and } 318.15 \text{ K})$ .

## 2. Experimental Section

Poly(diallyldimethylammonium)chloride PDADMAC of average  $M_w$  100,000–200,000 (21.8% aqueous solution) purchased from Sigma-Aldrich Co, and sodium dodecylsulfate SDS (minimum 98.5% by GC) purchased from Sigma Co. were used without additional purification. All other reagents were of analytical grade. Table 1 summarizes all chemicals used in this work, along molecular weight, mass fraction purity. The mixed polymer/SDS have

been prepared by using an electronic analytical balance with a precision of  $\pm 0.0001$  g (Shimadzu, model AY220).

Conductometric measurements were carried out using a digital conductivity meter CM-183 microprocessor based EC-TDS analyser with ATC probe. Prior this measurement, the conductivity cell was calibrated with KCl solutions in the concentration range of 0.01–1.0 mol  $\text{kg}^{-1}$ . The cell constant was determined to be  $1.0021 \text{ cm}^{-1}$ . No less than five measurements were taken for each concentration and only the mean value was taken into consideration. The small conductance due to water was subtracted from the measured data. Uncertainty of the measurements was less than 4%. After ensuring thorough mixing and temperature equilibration of (298.15, 308.15 and 318.15 K), the specific conductance ( $\kappa$ ) was considered.

Densities and speeds of sound of pure SDS and their mixtures with PDADMAc were measured by using an Anton Paar DSA 5000 (oscillating U-tube density and speed of sound analyzer) instrument and the temperature was controlled to  $\pm 1 \times 10^{-2}$  K by a built-in solid-state thermostat. Before each series of measurements, the densimeter was calibrated with doubly distilled, degassed water and with dry air at atmospheric pressure. The maximal error in the measurements of density and speed of sound relative to water ( $997.050 \text{ kg m}^{-3}$  and  $1496.687 \text{ m s}^{-1}$ ) is expected to be less than  $5 \times 10^{-3} \text{ kg m}^{-3}$  and  $5 \times 10^{-2} \text{ m s}^{-1}$ .

The Zeta potentials and size of the polymer-surfactant aggregates in aqueous solutions were determined using a Nano Zs Zetasizer 3000 (Malvern Instruments). A He-Ne laser beam of wavelength 632.8 nm was used. Each run was duplicated to check the reproducibility.

### 3. Results and discussion

#### 3.1. Conductance measurements:

**Figures 1(a, b, c)** shows the variation of conductance with SDS concentrations at temperatures ranged from 298.15 to 318.15 K. From this figure, it can be observed an increase in electrical conductivity with SDS molalities and having a gradual decrease in slope value. According to the Onsager theory of electrolyte conductivity, two linear regimes were expected in conductivity graph, one corresponding to pre-micellar region while the other corresponding to the post-micellar region [26]. The changes in conductivity values for the pure IL fit into two straight lines of different slopes ( $R = 0.99$ ), and the point of intersection of the lines of the abrupt change in slope gives the value of the (cmc). Here the cmc of pure SDS can be obtained from the intersection of the tangent lines before and after break in

conductivity . The cmc value of pure SDS was found to be at 8.8 mM at 298.15 K , it is in good agreement with the literature value [27]. We also obtain cmc values of SDS at the range of concentration 0.5 -1.5 g/L of PDADMAc and it is somewhat higher than pure SDS solution. the increase in cmc values upon the addition of PDADMAc is attributed to the fact that there is an increase in the binding sites available for the SDS monomers or micelle like aggregate (not the true micelles) where they can bind with the polymer. therefore it is quite obvious that on increasing concentration of PDADMAc below than 2 g/L polymer concentration , more amount of the SDS is required for binding. after binding of all sites , free surfactant micelles form. The degree of counter ion dissociation ( $\alpha$ ) was taken as the ratio of slopes of the lines of post to pre-micellar region the higher value of ( $\alpha$ ) of the complex micelles indicates due to the interaction of SDS with polymer. the cmc value of SDS upon the addition of PDADMAc at 2 g/L decrease compared with the cmc value of pure SDS , suggesting that surfactant micelles may form part of the polymer -surfactant complex bound at the interface.

temperature alters the cmc value of surfactant because of two opposite processes. high temperature causes the disruption in water structure and results in the increase solubilisation of surfactant molecules , along with delaying of aggregation process. therefore, the degree of hydration of ionic surfactant head domain decreases, this leads to an increase in cmc.

The temperature dependence of conductivity was employed to elucidate the thermodynamics of micellization of SDS at (0.25, 0.5 and 1 g/L) of PDADMAc concentrations. According to mass action model the standard free energies of micellization,  $\Delta G_m^0$  of SDS in the solutions were obtained by using the equation Eq.1 [28]:

$$\Delta G_m^0 = (2 - \alpha)RT \ln X_{cmc}$$

Where R is gas constant, T is temperature and  $\alpha$  is degree of counterion ionization, calculated from the ratio of slopes of post-micellar to pre-micellar regions of the specific conductance versus concentration graphs. The  $\alpha$  decreases with increase in polymer concentration due to lesser dissociation of ions and therefore results in more head group repulsions which further delays the micellization process.

From the temperature dependence of cmc, the standard enthalpy of micellization process was obtained using the equation Eq.2 [28]:

$$\Delta H_m^0 = RT^2 \left( (2 - \alpha) \frac{d \ln X_{cmc}}{dT} + \ln X_{cmc} \frac{d(1 - \alpha)}{dT} \right)$$

Further, the standard entropy of micelle formation,  $\Delta S_m^0$  is obtained using Eq.3[29]:

$$\Delta S_m^0 = \frac{\Delta H_m^0 - \Delta G_m^0}{T}$$

the method values were obtained by application of equation 2 to the fitted values of  $\ln(X_{cmc})$  with temperature. however  $d(\ln X_{cmc})/dT$  was determined as the slope of the straight line obtained by plotting  $\ln(X_{cmc})$  curve against temperature and subjecting the data to least squares treatment [30]. we have obtained the micellization enthalpies values of SDS/PDADMAc mixed solutions at temperatures of 298.15, 308.15 and 318.15 K. a number of trends are seen in table 1 :we find an increase of the micellization enthalpies as a function of increasing PDADMAc concentration at all temperatures. the micellization enthalpy consists of SDS-SDS interactions , PDAMADAc -SDS interactions , these interactions may be broken down into a hydrophobic portion as well as electrostatic contribution due to the mixing of the surfactant and PDADMAc in the head group region of the micelle. there is also a contribution from the interaction of the hydrophobic surfactant and PDADMAc chains with water, which results in the formation of structured water in solution. when the surfactant and PDADMAc aggregate; interaction between the hydrophobic chains lead to decrease in the enthalpy of the mixed systems of SDS and PDADMAc at 2g/L concentration of PDADMAc . It can be seen from table 1 that the free energy of micellization  $\Delta G_m^0$  values decrease with increasing PDADMAc concentration at the studied temperatures. these results demonstrated the the addition of PDADMAc promote micelle formation. we also find that  $T\Delta S_m^0$  values decrease with increasing polymer concentration and temperature. this is due to interactions between the surfactant and PDADMAc alky chains. thus when the mixed micelles are formed in 2 g/L concentration of polymer,  $\Delta H_m^0$  and  $T\Delta S_m^0$  values increase, this suggests that both the polymer and surfactant are contributing to the hydrophobic effects.

### 3.2. Density and speed sound measurements:

Apparent molar volumes ( $V_\phi$ ) of SDS solutions were calculated over the entire range of PDADMAC concentrations and temperature using the following equation Eq 4:

$$V_\phi = \frac{M}{\rho} - \left( \frac{1000(\rho - \rho_0)}{m\rho\rho_0} \right)$$

where,  $\rho$  and  $\rho_0$  are the densities of the mixed solutions of SDS and reference solvent (water, PDADMAC) respectively and  $m$ ,  $M$ , denote the molality of the solution of SDS, and molecular weight, respectively.

apparent molar volume values of SDS solutions at different temperature and concentration of polymer are graphically represented in figure 2. from this figure, it can be seen that  $V_\phi$  values increase with the increase of the molality of the surfactant, also it increases with increase of the temperature. As stated by Gheorghe et al. (2016) and dubey et al. (2019), these values could suggest the presence of strong intermolecular forces between surfactant and water molecules [31-32].

apparent molar isentropic compressibility ( $\kappa_\phi$ ) is expressed by using equation Eq 5 :

$$\kappa_\phi = \left[ \frac{1000(\rho_0 \kappa_s - \kappa_{s0} \rho)}{m\rho\rho_0} \right] + \frac{M\kappa_s}{\rho}$$

where  $\kappa_s$  and  $\kappa_{s,0}$  are the isentropic compressibility of the mixture and reference solvent respectively.  $\kappa_\phi$  increases with an increase in PDADMAC concentration and temperature, which means that the mixture lose their rigidity with increasing SDS molality and temperature. For diluted solutions, a simple linear relationship can be applied to describe the variation of the apparent molar volume,

$$V_\phi = V_\phi^0 + S_v m$$

where  $V_{\phi}^0$  and  $K_{\phi}^0$  are the limiting values of apparent molar volume and apparent molar adiabatic compressibility, respectively, and often regarded equal to the infinite dilution partial molar volume and infinite dilution partial molar adiabatic compressibility.

The presence of strong solute–solvent interactions is also reinforced by the positive values of  $V_{\phi}^0$  and negative values of  $K_{\phi}^0$  for the surfactant in aqueous PDADMAC solutions at different temperatures. The variation of  $V_{\phi}^0$  has been shown graphically in Figure 4 in which values of  $V_{\phi}^0$  are plotted for surfactant in aqueous solution of PDADMAC at different temperatures. The predominance of solute–solvent interactions over solute–solute interactions is evident from the sign (positive). The values of  $K_{\phi}^0$  become less negative with increase in temperature (as shown in Fig.5). The negative values of  $K_{\phi}^0$  (loss of compressibility of medium) indicate that the water molecules surrounding SDS are less compressible. The more negative values of  $K_{\phi}^0$  for surfactant at low temperature indicate prominence of strong attractive interactions between water and neighbouring species in medium. With increase in temperature the  $K_{\phi}^0$  values become less negative which means decrease in electrostriction and release of some water molecules to bulk. The release of water molecule into bulk by the hydration of ions occurring due to dissociation of SDS micelles results in strong attractive interactions[33].

### 3.3. Zeta potential:

Zeta potential ( $\zeta$ ) is a measure of the potential existing at the surface of a particle. The zeta potential of PDADMAC-SDS complex as function of SDS concentration is shown in Fig.6. These results show that with an increase in SDS concentration, the zeta potential of samples undergoes a charge reversal, going from positive to negative values. The addition of polymer at concentration below 1.5 g/L, the surface charge becomes more negative by adsorbing more surfactant, these observations are consistent with the contention that electrostatics play an important role. The polymer configuration initially tended to become compact by the electrostatic interaction with  $DS^-$  at a number of its positive  $Me_2N^+$  centers. The sign of zeta potential changes at concentration (2g/L) of PDADMAC, with  $\zeta=0$ . The zeta potential increases up to 1.5 g/L of PDADMAC in solution where  $\zeta=0$  and then decreases as the concentration of surfactant increases further. The fact of the positive value of the zeta potential decreases in the molality range above 6 mM of SDS shows that the electrostatic interactions cannot be the only interaction responsible for the uptake of the surfactant by the

polymer. Hydrophobic attractions between the polymer and the hydrophobic surfactant might account for the interaction. Also in Fig.6, one can observe that the  $\zeta$  potential is low and negative at  $1\text{g.L}^{-1}$  of PDADMAc concentration. It can be suggested that the electrostatic interactions play a minor role in the adsorption process. At some concentration of the added SDS, with ( $\zeta=0$ ), the electrical forces of repulsion are lowered sufficiently that the forces of attraction dominate. Under these conditions, the particles may approach each other more closely and form loose aggregates termed floccules.

#### 4. Conclusion:

This paper has examined the effects of polymer concentration and temperature on the physico chemical properties of aqueous SDS solutions by conductance, Density, Speed of sound and Zeta Potential methods. The obtained (cmc) using conductivity method increase in the presence of the polyelectrolyte which confirms the complexation between PDADMAc and SDS monomers. Using the cmc and  $\alpha$  values obtained from conductance, and it is observed that  $\alpha$  values are negative for all the studied systems indicating the spontaneous. Large and positive values of  $T\Delta S_m^0$  indicates the crucial role of hydrophobic forces in micellization. densities and sound speed data were experimentally determined for SDS mixed solutions at temperatures from 298.15 to 318.15 K. from the experimental data for the density and sound speed, it was possible calculate the limiting values of apparent molar volume ( $V_\phi^0$ ) and apparent molar isentropic compressibility ( $\kappa_\phi^0$ ). Positive values for  $V_\phi^0$  and negative values for  $\kappa_\phi^0$  have been observed, which increase upon increasing the temperature. It depicts decrease in electrostriction with increase in temperature. Further, Zeta Potential method help in better understand of the interaction between SDS and PDADMAc.



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**Table1:** Specifications of chemicals

Chemical name	Provenance	Mass fraction purity (%)
Sodium dodecylsulphate (SDS)	Sigma Aldrich,USA	$\geq 98.5\%$
Polydiallyldimethylammonium chloride (PDADMAc)	Sidma Aldrich	21.8% in aqueous solution

**Table 2:** The values of critical micelle concentration (cmc), degree of dissociation ( $\alpha$ ), free energy of micellization ( $\Delta G_m^0$ ), enthalpy of micellization ( $\Delta H_m^0$ ) and entropy of micellization ( $\Delta S_m^0$ ) of aqueous SDS solutions in different concentration of PDADMAc at different temperatures.

PDADMAc (g.L <sup>-1</sup> )	Cmc (mol.Kg <sup>-1</sup> )	$\alpha$	$\Delta G_m^0$ (kJ.mol <sup>-1</sup> )	$\Delta H_m^0$ (kJ.mol <sup>-1</sup> )	$T\Delta S_m^0$ (kJ.mol <sup>-1</sup> )
<b>T=298.15K</b>					
0	0.0086541	0.536	-42.304	--7.188	35.116
0.5	0.009300	0.533	-42.129	-6.982	35.147
1	0.0094360	0.517	-42.515	-6.885	35.63
1.5	0.009488	0.509	-42.745	-5.832	36.913
2	0.008540	0.496	-43.510	1.110	-44.62
<b>T=308.15K</b>					
0	0.008950	0.689	-39.041	-5.329	33.712
0.5	0.00909	0.668	-39.409	-5.110	34.299
1	0.00914	0.620	-40.907	-4.795	36.112
1.5	0.00924	0.605	-41.309	-4.665	36.644
2	0.00856	0.583	-42.359	1.128	-41.231
<b>T=318.15K</b>					
0	0.009748	0.7701	-37.540	-4.350	33.19
0.5	0.01010	0.76	-37.728	-4.118	33.61
1	0.01020	0.745	-38.152	-3.991	34.161
1.5	0.01030	0.530	-44.650	-3.885	40.765
2	0.00952	0.473	-46.700	2.751	-49.451

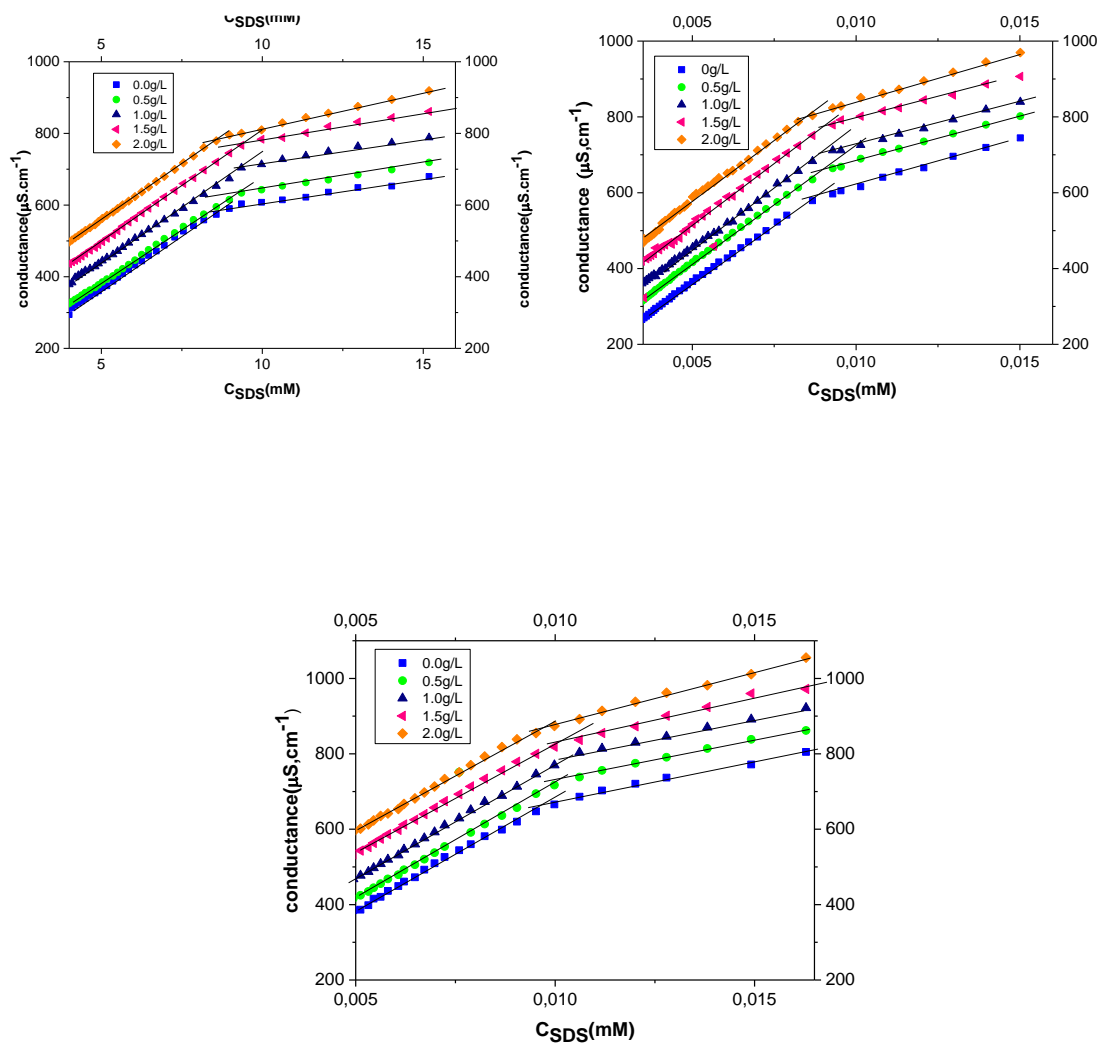


Fig. 1. Specific conductance ( $\kappa$ ) of aqueous SDS solutions in the presence of different concentrations of PDADMAc at temperatures (a) 298.15 K; (b) 308.15 K; (c) 318.15 K.

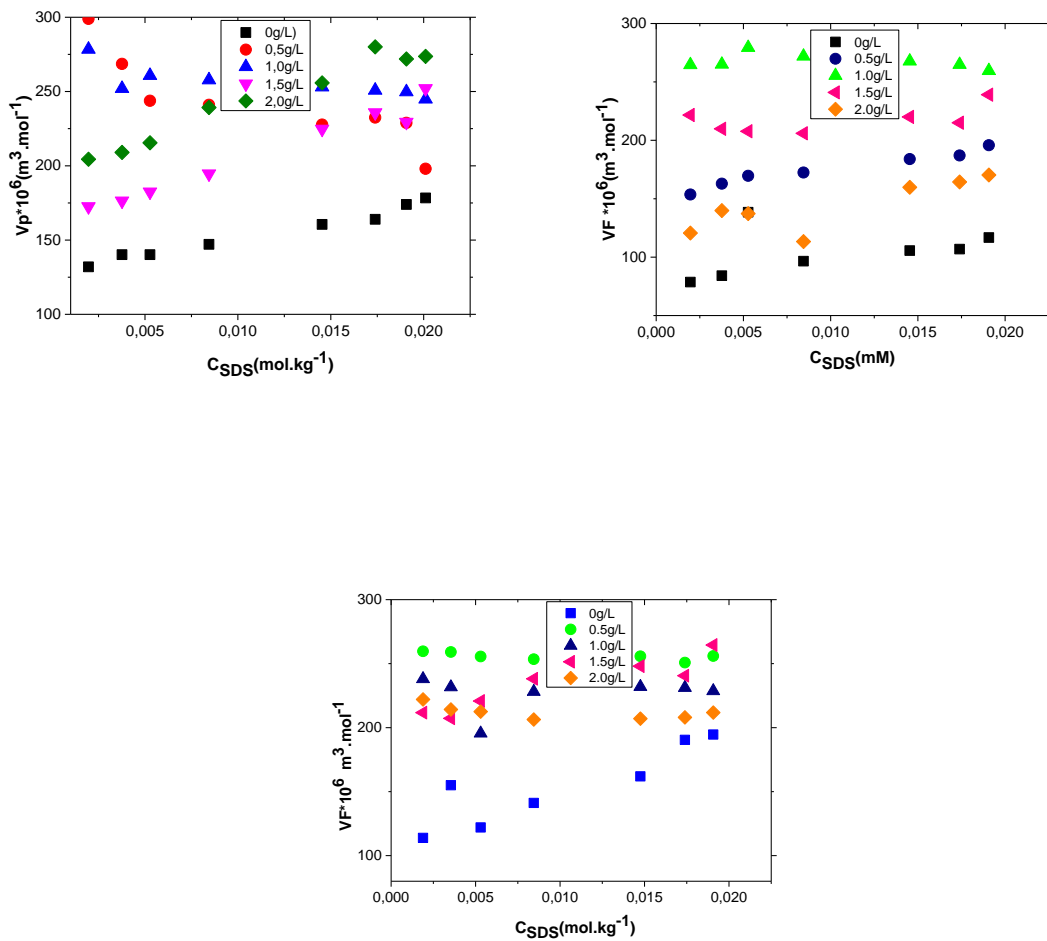


Fig.3. Plot of  $V\phi$  for surfactant in aqueous solutions of PDADMAc against molalities  $m$  of SDS at different temperatures; (a) 298.15, (b) 308.15 and (c) 318.15K



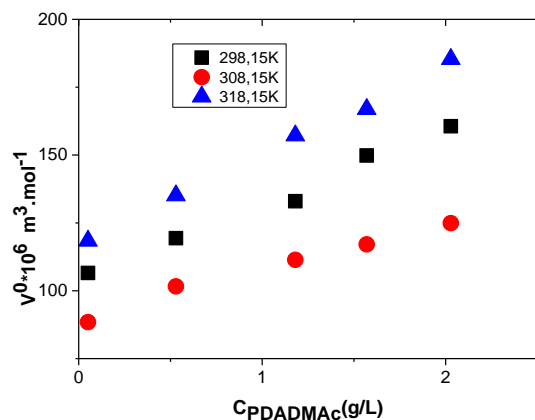


Fig. 4. Plot of  $V_{\phi}^0$  for surfactant SDS in aqueous solutions of PDADMAC at different temperatures.

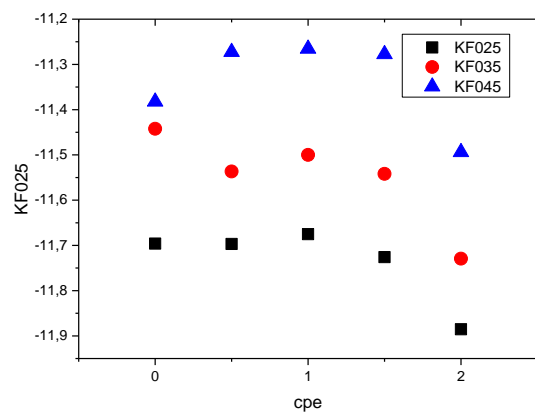


Fig. 5. The variation in limiting apparent adiabatic compressibility,  $K_{\phi}^0$  as a function of concentration of PDADMAC at different temperatures.

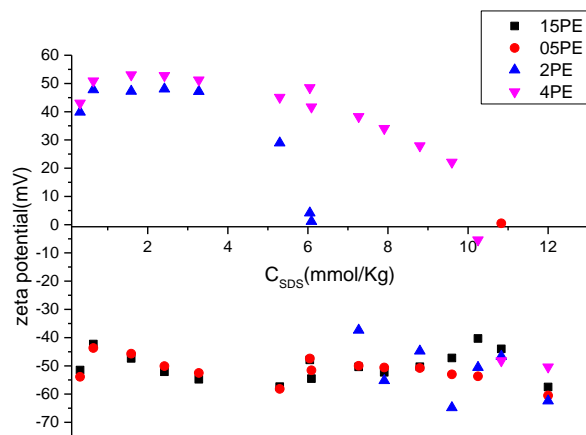


Fig.6. Zeta potential of SDS–PDADMAc samples as a function of the molality of surfactant.  $\Delta$   $1.5\text{g}\cdot\text{L}^{-1}$ ,  $\nabla$   $2\text{g}\cdot\text{L}^{-1}$ ,  $\circ$   $0.5\text{g}\cdot\text{L}^{-1}$  and  $\square$   $1\text{g}\cdot\text{L}^{-1}$  of PDADMAc