INTRODUCTION

Micelles are dynamic aggregates of amphiphilic or surfactant molecules possessing both hydrophilic and hydrophobic character. In such systems, both oil-soluble and water-soluble compounds can be dissolved due to the simultaneous presence of aqueous and non-aqueous domains. Since the reagents are localized in either aqueous or nonaqueous domain at the pseudophase, their effective local concentrations are increased. This can increase reaction rates. Electrochemical techniques have been applied as tool for the characterization of micelles and microemulsion systems.

In general voltammetry in microheterogeneous systems, such as micelles, microemulsions and macroemulsions is becoming increasingly popular as a general method for studying heterogeneous redox processes and for characterizing the discontinuous phase in such systems. These systems have important industrial and biomedical applications and warrant a thorough understanding of many subjects such as the related transport phenomena, structure of the pseudophase, electrode processes and many others. The micelles particle size is determined from the estimated diffusion coefficients using the Stokes-Einstein equation:

\[ D = \frac{KT}{6\pi\eta r_h} \]

where K is the Boltzmann constant, T is the absolute temperature, h is the viscosity coefficient and \( r_h \) is the hydrodynamic radius of the droplet. Generally, electrochemical (EC) techniques probed self-diffusion coefficients compared to...
quasi-elastic light scattering (QELS) technique and showed good agreement with those obtained from NMR because they possess very long diffusion time (\(\sim 1\) msec). Therefore, both EC and NMR techniques probe self-diffusion over a distance larger than the extension of any aggregate in solution. Whereas, in QELS measurements it showed collective data and the time scale is very short in the range of 0.1 to 10 msec.

This paper is devoted for the voltammetric detection of the onset of micellization and diffusion coefficient determinations of the synthesized, NaAAS surfactant using three different electrochemical techniques such as CV, RDV and CC. Reequilibrium kinetic considerations of ferrocene probe partitioning and solubilization studies are discussed. The protection of carbon steel L52 using NaAAS surfactant was measured by weight loss measurements.

**EXPERIMENTAL**

Material

Acrylonitrile, acrylic acid, oleic acid were obtained from Aldrich. Ferrocene, NaCl, HCl and NaOH were certified ACS reagents.

Acrylamidostearic acid (AAS) was prepared by reacting acrylonitrile and oleic acid according to the method of Greene\(^{21}\). The crude material was then dissolved in absolute ethanol and after filtering any insoluble residue, it is neutralized with sodium ethoxide. The addition of dry acetone to the neutralized ethanolic solution of acrylamidostearic acid, AAS, precipitates sodium acrylamidostearate (NaAAS). After repeatedly washing with dry acetone, it was then dried under vacuum at room temperature overnight. The recrystallized material was identified using FTIR, absorption bands appeared at 3080, 1620, 980 and 945 cm\(^{-1}\) corresponding to the terminal vinyl group (CH\(_2\) = CH\(-\)) while 3260, 1650 and 1560 cm\(^{-1}\) absorption bands were corresponded to the absorptions of amide group (-CONH\(-\)). Therefore the structural formula will be as shown below:

\[
\begin{align*}
\text{CH}_3\text{(CG}_2\text{)}_{8(7)}\text{-CH(CH}_2\text{)}_{8(7)}\text{-COONa} \\
\text{NHCOCH} = \text{CH}_2 \\
\text{(NaAAS)}
\end{align*}
\]

| Scan Rate mV/sec | \(i_{pc}\) \(\mu A\) | \(i_{pa}\) \(\mu A\) | \(E_{pc}\) \(\mu V\) | \(E_{pa}\) \(\mu V\) | \(i_{pa}/i_{pc}\) | \(\Delta E_p\) \(mV\) |
|-----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| 20              | 0.658          | 0.695          |                 |                 |                |                |
| 40              | 0.774          | 0.820          |                 |                 |                |                |
| 60              | 1.368          | 1.316          |                 |                 |                |                |
| 80              | 1.620          | 1.580          |                 |                 |                |                |
| 100             | 1.895          | 1.789          |                 |                 |                |                |
| 150             | 2.350          | 2.268          |                 |                 |                |                |
| 200             | 2.902          | 2.870          |                 |                 |                |                |

![Fig. 1: The plot of anodic peak potentials of ferrocene at different NaAAS concentrations](image)
Equipment

The electrochemical measurements such as cyclic voltammetry (CV), rotating disk voltammetry (RDV) and chronocoulometry (CC) were conducted using Electrochemical Analyser Model HQ-2030 and the rotating disk electrode from BAS. A three compartment cell consisted of glassy carbon as working electrode, platinum foil (1 cm²) as counter electrode and saturated calomel electrode (SCE) as the reference electrode used in electrochemical measurements.

The glassy carbon electrode was cleaned up by successive polishing with 0.3 and down to 0.05 mm sized alumina and thoroughly washed with distilled water, finally ultrasonicated in distilled water for 15 minutes to dislodge the retained aluminum oxide particles from the electrode surface. The area of the glassy carbon (GC) electrodes used for both cyclic voltammetry (CV) and rotating disk voltammetry (RDV) were estimated using cyclic voltammetry technique from 4 mM K₄[Fe(CN)₆] dissolved in 1 M KCl solution of known diffusion coefficient (D = 6.32x10⁻⁶ cm²/sec) 22. It was found to be 0.067 and 0.453 cm² for both techniques; respectively. In electrochemical measurements, the surfactant solutions were deaerated by bubbling a stream of nitrogen gas for at least 10 min using slow rate to avoid foaming.

Spectral measurements were conducted using PerkinElmer spectrophotometer Model Jasco V-500. All electrochemical measurements were recorded for the first scan and recorded at 25°C using HAAke water circulator bath.

Table 2: Electrochemical data obtained from rotating disk voltammetry around the critical micelle concentration of NaAAS surfactant containing 0.1 M NaCl at pH 10.0, using fixed 0.07 mM of ferrocene, 25°C

| [NaAAS] mM | Δi/ωΔω¹/₂ µA.sec | S/S₀ | Δd_a (cm²/sec) |
|------------|------------------|------|----------------|
| 0.00(*)    | 1.047            | 1.000| 6.70x10⁻⁶(*)   |
| 0.10       | 1.042            | 0.995| 4.00x10⁻⁶      |
| 0.20       | 1.010            | 0.965| 3.82x10⁻⁶      |
| 0.30       | 0.960            | 0.917| 3.54x10⁻⁶      |
| 0.40       | 0.748            | 0.715| 2.44x10⁻⁶      |
| 0.80       | 0.638            | 0.610| 1.92x10⁻⁶      |
| 1.00       | 0.618            | 0.590| 1.83x10⁻⁶      |
| 2.00       | 0.565            | 0.540| 1.60x10⁻⁶      |
| 4.00       | 0.502            | 0.480| 1.34x10⁻⁶      |
| 8.00       | 0.450            | 0.430| 1.14x10⁻⁶      |
| 10.00      | 0.441            | 0.422| 1.10x10⁻⁶      |

*Ref. 18.

Table 3: Apparent diffusion coefficients and its treatment according to slow- and fast-kinetic limits obtained for NaAAS surfactant using rotating disk voltammetry at 25°C

| [NaAAS] mM | [Fc] mM | D_a cm²/sec | D_a⁽ᵃ⁾ cm²/sec | D_a⁽ᵇ⁾ cm²/sec |
|------------|---------|-------------|----------------|----------------|
| 20         | 0.30    | 6.40x10⁻⁷  | 7.69x10⁻⁷      | -              |
| 40         | 0.55    | 6.30x10⁻⁷  | 3.06x10⁻⁷      | 2.30x10⁻⁸      |
| 60         | 0.81    | 6.21x10⁻⁷  | 3.99x10⁻⁷      | 2.20x10⁻⁷      |
| 80         | 0.98    | 6.10x10⁻⁷  | 5.68x10⁻⁷      | 2.83x10⁻⁷      |
| 100        | 1.25    | 5.90x10⁻⁷  | 4.47x10⁻⁷      | 3.35x10⁻⁷      |
| 150        | 1.50    | 5.90x10⁻⁷  | 4.71x10⁻⁷      | 3.79x10⁻⁷      |
| 200        | 2.00    | 5.80x10⁻⁷  | 4.91x10⁻⁷      | 4.20x10⁻⁷      |

a- slow-kinetic treatment
b- fast-kinetic treatment
Fig. 2: RDV voltammograms recorded at 1000 RPM for: A- $4 \times 10^{-4}$ M, B- $8 \times 10^{-4}$, C- $2 \times 10^{-4}$ M and D- $8 \times 10^{-3}$ M NaAAS
Table 4: The effect of anionic NaAAS concentrations on the weight loss measurements of carbon steel L52 corrosion in 1.0 M HCl

| [NaAAS] ppm | M       | Weight Loss (mg/d.cm²) | Inhibition efficiency (IE%) |
|-------------|---------|------------------------|----------------------------|
| 15.0        | 4.0x10⁻⁵ | 200.0                  | 64.28                      |
| 30.0        | 8.0x10⁻⁵ | 180.0                  | 67.86                      |
| 37.5        | 1.0x10⁻⁴ | 170.0                  | 69.64                      |
| 75.0        | 2.0x10⁻⁴ | 140.0                  | 75.00                      |
| 150.0       | 4.0x10⁻⁴ | 42.0                   | 92.50                      |
| 225.0       | 6.0x10⁻⁴ | 42.0                   | 92.50                      |
| 300.0       | 8.0x10⁻⁴ | 41.0                   | 92.68                      |
| 375.0       | 1.0x10⁻³ | 41.2                   | 92.64                      |

Fig. 3: The plots of limiting current of 7x10⁻⁵ M ferrocene in different [NaAAS] at different rotation speeds

Fig. 4: The plots of the slope ratio (S/So) at different surfactant concentrations

Fig. 5: The plots of the apparent diffusion coefficient and its corrections by slow and fast-methods at different [NaAAS]
Carbon steel sheets with dimensions of 1×2×0.2 cm were used in the weight loss measurements. A stock solution of 1×10^{-2} M NaAAS was prepared in absolute ethanol from Merk for the weight loss measurements. Each carbon steel sheet was immersed for six hours in 1M HCl solutions containing different NaAAS concentrations and recorded in quartet replicates.

RESULTS AND DISCUSSION

Determination of the onset of micellization

Cyclic voltammetry (CV)

The cyclic voltammograms of fixed ferrocene (Fc) concentration (7.0 × 10^{-5} M) solubilized in sodium acrylamidostearate (NaAAS) surfactant of different concentrations varied from 1.0×10^{-4} to 1.0×10^{-2} M were recorded in aqueous buffer solutions of pH 10.

The amount of ferrocene probe solubilized at the lower NaAAS concentration (1.0×10^{-4} M) containing 0.10 M NaCl as supporting electrolyte of pH 10 was measured from UV-Visible spectrophotometry. Standard solutions of ferrocene were prepared in ethanol-water mixtures and measured against water as blank. By appropriate dilutions it showed absorbance 0.30 at 250 nm wavelength and had a molar absorptivity e/10^4 M^{-1}.cm^{-1} which was found in a good agreement with that reported for CTAB. The amount of ferrocene was solubilized in 1.0×10^{-4} M NaAAS containing 0.10 M NaCl was performed by ultrasonication for at least seven hours. Solubilizations were performed three times and the amount of ferrocene dissolved in 1×10^{-4} M NaAAS was measured at 250 nm using the molar absorptivity given above from the standard graph and found to be 7.0×10^{-5} M. Therefore, the amount of ferrocene used in subsequent electrochemical measurements and CMC determinations was fixed at this concentration level to avoid the fluctuations in peak current and diffusion coefficient values as well as having a better estimate.

The voltammograms recorded at different scan rates from 20.500 mV/sec showed a single redox peak. Ferrocene is well behaved reversibly in these media at different surfactant concentrations. The peak potential separation ΔE_p (= E_{pa} - E_{pc}) is always kept in the range of 55.59 mV and the ratio of i_{pa}/i_{pc} did not exceed 1.10, Table (1). This behavior indicate the complete reversibility of ferrocene oxidations in these media with a net transfer of one electron forming ferrocenium ion (Fc⁺).

The peak current of reversible system is described by the Randles. Sevcik equation [24] for the forward step of the first cycle, plot

\[ i_p = 0.4463nFAC_0 (nF/RT)^{1/2}D_1^{1/4}v^{1/2} \]  ...(1)

The plots of \( i_{pa} \) versus \( v^{1/2} \) showed linear correlations passing through the origin at different NaAAS concentrations indicating the diffusion nature of the electrode process. The critical micelle concentration CMC is determined by observing the variation of anodic peak current for ferrocene oxidations against surfactant concentrations. The plots of \( i_{pa} \) obtained at 100 mV/sec versus [NaAAS] is linear and showed a break at 4.0×10^{-4} M indicating that physical and chemical changes taking place by the formation of micelles. Also, the plotting of \( E_{1/2} \) vs. [NaAAS] showed linear relationship consisting of two segments and the breaks occurs at 4.0×10^{-4} M which is further confirmed the obtained value of CMC, Fig. 1.

RDV experiments

The voltammograms of ferrocene solubilized in different concentrations of sodium acrylamidostearate (NaAAS) were recorded at different rotation speeds (ω) from 250 to 2000 RPM. The potential was scanned from -100 to 600 mV at scan rate 5 mV/sec. The rotating disk voltammograms were treated using the most practical form of completely treated rigorous hydrodynamic equation of Levich who first solved the mass transport equations of the rotating disk electrode. The limiting current of a reaction controlled only by mass transfer has given by the Levich equation:

\[ i_L = 0.62nFAD^{2/3}v^{1/6} \omega^{1/2}C_0 \]  ...(2)

where \( i_L \) is the limiting current (A), \( v \) is the kinematic viscosity (cm^2/sec) and \( \omega \) is the angular velocity (rad/sec). In this case ferrocene was used as the hydrophobic probe which has a solubility in aqueous solution of 5×10^{-5} M. Therefore, accurate
measurements fulfilled by making corrections for the contribution to the electrode surface by the amount of probe solubilized in aqueous continuous phase. The Levich equation may be rewritten in the pseudophase approximation as following:

\[ i_L = 0.62nFA\left(\frac{D_o^{2/3}(C_o - C_m) + D_m^{2/3}C_m}{\nu^{1/6}}\right)^{1/2} \]  

...(3)

where \(C_o\) and \(D_o\) are the total concentration of ferrocene and aqueous phase diffusion coefficient; respectively. \(C_m\) is the concentration of ferrocene solubilized micelles and \(D_m\) is the micelle diffusion coefficient. For very dilute aqueous solutions the slope was extracted from equation (2) as following:

\[ S_o = \frac{\partial i}{\partial \omega^{1/2}} \]  

...(4)

The analogous quantity derived from equation (3) divided by equation (4) it gives:

\[ S = \frac{C_o - C_m + D_m^{2/3}C_m}{C_o - D_m^{2/3}C_o} x \frac{\nu^{1/6}}{\nu_o^{1/6}} \]  

...(5)

The onset of micellization of NaAAS surfactant was detected from aqueous solutions of pH 10.0 containing 0.1 M NaCl at 25°C using RDV technique. The electroactive probe used was ferrocene and the surfactant concentration was varied from 1x10^{-4} to 0.01 M, Fig. 2. The results showed excellent linearity of \(i_L\) versus \(\omega^{1/2}\) passing through the origin indicating that ferrocene is well behaved and the electrode process is under mass transfer control within the surfactant concentration range, Fig. 3.

The plots of the slope ratio \((S/S_o)\) versus surfactant concentration, \(\log [Surf]\) at 0.07 mM concentration of ferrocene showed dissociation curve as represented in Fig. 4. The critical micelle concentration was determined from the intersect of the two portions and found to be at 4x10^{-4} M. The slope ratio \((S/S_o)\) was used for obtaining more accurate results avoiding fluctuation in electroactive probe concentration.

**Chronocoulometry (CC)**

An important aspect to be considered in electrochemical investigations in surfactant solutions is its adsorption on the electrode surface. Double potential step chronocoulometry (CC) is the most useful technique in determining the amount of adsorption of electroactive species\(^{28,29}\). In absence of product adsorption the difference of intercepts of \(Q\) versus \(t^{1/2}\) and \(Q\) versus \(\theta\) plots for both the forward and reverse reactions respectively, gives a direct measurement of the adsorbed species\(^{30}\). The amount of ferrocene probe adsorbed in NaAAS micelles was evaluated and found to be 1.0x10^{-15} M which considered a negligible amount as compared with the amount of probe being added (7.0x10^{-5} M). This is found to be in good agreement with the results obtained from both CV and RDV experiments.

**Solubilizations studies and diffusion coefficients using RDV**

**Diffusion coefficients**

From the electrochemical oxidation of ferrocene, the plots of \(i_L\) versus \(\omega^{1/2}\) at different NaAAS concentrations in the range from 20 to 200 mM) showed linear correlations passing through the origin. The concentration of ferrocene used at different surfactant concentrations were very close to the saturation concentration in each micellar solutions. Also, the amount of ferrocene solubilized in NaAAS micelles were determined spectrophotometrically as described above and listed in Table 3. The RDV voltammograms of saturated solution of ferrocene solubilized in different NaAAS concentrations were recorded at scan rate of 5 mV/sec and different rotation speeds. The plots of \(i_L\) versus \(\omega^{1/2}\) using were found to be linear and passing through the origin indicating that the electrode reaction taking place under mass transfer control, Fig. 3. The apparent diffusion coefficients \((D_a)\) were determined from the Levich eq. (2) using the slopes of the plots of \(i_L - \omega^{1/2}\) plots, Table 3.

**Reequilibrum and kinetic considerations from RDV**

The apparent diffusion coefficient \((D_a)\) values obtained according to the above equation is an average value, because the electroactive ferrocene has a solubility of 5x10^{-5} M in aqueous solution\(^5\). Therefore, the apparent diffusion
The apparent diffusion coefficient values \((D_a)\) obtained electrochemically from RDV and its corrected values \((D_d)\) according to the above equations (6) and (7) were plotted versus NaAAS concentrations, Fig. 5. Inspection of the diffusion coefficient values obtained by slow- and fast-kinetic corrections showed constancy of the diffusion coefficient values on increasing NaAAS concentrations at about \((4.69 \pm 0.4) \times 10^{-7} \text{ cm}^2/\text{sec}\). The micellar sizes were determined by substituting in Stockes-Einstein equation described above after correcting the viscosity coefficient showing constancy at about 58 Å diameter. This behavior indicates that globular self-assembly structure rather than rod-like takes place at higher NaAAS concentration. Since the size of NaAAS does not practically changes, the number of micelles increases yielding more repulsive interactions as reported for SDS and CTAB and others.

**NaAAS Surfactant as Corrosion Inhibitor**

**Due to the various industrial applications and economic importance of carbon steel, its protection against corrosion has attracted much attention.** The carbon steel (L-52) sheets used in this study had the following composition on the basis of weight percentages: 0.26 C, 1.35 Mn, 0.04 P, 0.05 S, 0.005 Nb, 0.02 V, 0.03 Ti and the remainder is iron. The use of inhibitors is one of the most practical methods for protection. Surfactant compounds were tested as corrosion inhibitors for carbon steel and found to have high inhibition efficiencies. Since some corrosion inhibitors used at lower concentrations may accelerate corrosion or causes emulsification at higher concentrations it is necessary to study the effect of surfactant concentration on the inhibition efficiency (IE) and its relation to micelles formations using weight loss measurements.

The loss in the weight of carbon steel sheets due to their immersion in 1 M HCl solutions containing different concentrations of anionic NaAAS surfactant was measured. It was found that the addition of NaAAS surfactant at any concentration level lowers the weight loss of carbon steel than its value in the corrosive medium. This result indicates that NaAAS acts as inhibitor for carbon steel in the presence of aggressive anions. The inhibitive effect could be attributed to the...
adsorption of surfactant molecules on carbon steel forming a barrier between the bar metal and the corrosive medium. The surface activity of NaAAS and its adsorption is facilitated by the presence of the carbonyl of the amide group in its structure as well as the unsaturation center. The adsorbed surfactant molecules onto metal surface is oriented via the hydrophilic amido group whereas the hydrocarbon domains is oriented toward the aqueous solution. Since the hydrocarbon chain is hydrophobic in its nature it repels the aqueous aggressive anions away from the metal surface and therefore inhibit the corrosion process. The inhibition efficiencies (IE%) of carbon steel strips immersed for 6 hours in 1M HCl solutions in absence and presence of different NaAAS concentrations was determined and given in Table (4). The inhibition efficiency (IE) is determined according to the following equation:

\[ \text{IE\%} = \left( \frac{W_f - W_i}{W_f} \right) \times 100 \]

where \( W_f \) and \( W_i \) are the weight loss of strips in the free and inhibited acidic solutions, respectively. Inspection of the values of weight loss measurements revealed that as the surfactant concentration increases as the weight loss decreased till reaching certain concentration at which the maximum efficiency was obtained. \( 4 \times 10^{-4} \) M (150 ppm) NaAAS displayed the maximum inhibition efficiency. This concentration is corresponding to the CMC of the anionic surfactant which is determined above electrochemically using both cyclic voltammetry and rotating disk voltammetry. At this concentration level the simple surfactant molecules changed to aggregates (spherical micelles) of particle size 58Å. Above the CMC the inhibition efficiency reached to almost constant value. This behavior is attributed to the constancy of micelles particle size where the morphological features of NaAAS micelles showed spherical drops with no conversion to rod like ones. Therefore, the inhibition efficiency increases via preferential adsorption of the highly negatively charged hydrophilic domains of the associated micelles via the amido-group while the hydrocarbon domains protrude brush-like into solution.

REFERENCES

1. Fendler J.H., Fendler E.J. “Catalysis in Micellar and Macromolecular Systems”, Academic Press, New York, (1975).
2. Pelizzetti, E., Pramauro E., J. Anal. Chem. Acta, 1: 169 (1985).
3. Burgess J., Colloids Surf., 48: 185 (1990).
4. Minero C., Pramauro E., Pelizzetti E., Colloids Surf., 35: 237(1989).
5. Georges J., Demetre S., Electrochimica Acta., 29: 521(1984); 31: 1519(1986).
6. Mahajan R.K., Kaur N. and Bakshi M.S., Colloids Surf., 225: 33 (2005).
7. Zana R. and Mackay R.A., Langmuir, 2: 109 (1986).
8. Shipovskov S., Ferapotova E., Ruzgas T. and Levashov A., Biochimica et Biophysica Acta, (2003).
9. Lu X., Zhang M., Kang J., Wang X., Zhuo L., and Liu H., J. Inorg. Biochem., (2004).
10. Matsubara T. and Texter J., J. Colloid Interface Sci. 112: 42 (1986).
11. Georges J. and Desmettre S., J. Dispersion Sci. Technol. 7: 21 (1986).
12. Texter J., Beverly T., Templar S. R. and Matsubara T., J. Colloid Interface Sci. 120: 389 (1987).
13. Morishita M., Shimizu Y., Kobayakawa K. and Sato Y., Electrochimica Acta, 53(22): 6651 (2008).
14. Shinoda K., Ed., Solvent Properties of Surfactant Solutions, Marcel Dekker, New York, Vol. 2 (1967).
15. Shah D. O., Ed., Macro and Microemulsions: Theory and Applications, Am. Chem. Soc. Symposium Ser., Washington D.C., 272 (1985).
16. Shah D. O. and Schecter R. S., Eds., *Improved Oil Recovery by Surfactant and Polymer Flooding*, Academic Press, New York (1977).

17. Spieser P. In *Reverse Micelles*, Luisi P. L., Straub B. E., Eds., Plenum, New York (1984).

18. Mackay R. A., *Adv. Colloid Interface Sci*. **15**, 131 (1981).

19. Lichtenberg D., Robson R. J. and Dennis E. A., *Biochim. Biophys. Acta* **737**, 285 (1983).

20. Einstein A., *Ann. Phys.*, **19**, 289 (1906).

21. Greene B.W., Sheetz D.P. and Filler T.D., *J. Colloid Interface Sci.*, **32**, 90 (1970).

22. Von Stackelberg M., Pilgram M. and Toome V., *Z. Electrochem.*, **57**, 342 (1953).

23. Ohsawa Y. and Aoyagui S., *J. Electroanal. Chem*., **136**, 353 (1982).

24. Adams A. J., “*Electrochemistry at Solid Electrodes*” Dekker (1969).

25. Bard A. J. & Faulkner L. R., “*Electrochemical Methods Fundamentals and Applications*”, John Wiley, New York, 218 (1980).

26. Mandal A. B., Nair B. U. and Ramaswamy D., *Langmuir* **4**, 736 (1988).

27. Levich V.G., *Acta Phyhsicochim. USSR* **17**: 157 (1942).

28. Anson F.C., *Anal. Chem.* **38**, 54 (1966).

29. Turner J.A., Turner B.A. and Darkinson B.A., *J. Electroanal. Chem.* **150**: 611 (1983).

30. Anson F.C. and Osteryoung R.A., *J. Chem. Educ.* **60**: 293 (1983).

31. Dayalan E., Qutubuddin S. and Texter J., *J. Colloid Interface Sci.* **143**: 423 (1991).

32. Texter J., *J. Electroanal. Chem.* **304**: 257 (1991).

33. Dayalan E., Qutubuddin S. and Texter J., *J. Colloid Interface Sci.* **158**: 249 (1993).

34. Corti M. and Degiorgio V., *J. Phys. Chem.* **85**, 711 (1981).

35. Dorshow R. B., Briggs J., Bunton C.A. and Nicoli D. F., *J. Phys. Chem.* **86**: 2388 (1982).

36. Mathur P.B. and Vasudevan T., *Corrosion*, **35**: 17 (1982).

37. Tedeschi R.J., *Corrosion*, **31**: 130 (1975).

38. Keera S.T., *Br. Corros. J.*, **36**(4): 26 (2001)

39. Osman M.M.: *Anti.Corros. Methods and Materials*, **45**(3): 176 (1998).

40. Al Sobagh A.M., Osman M.M., Omar A.A. and El.Gamal I.M.: *Anti.Corros. Methods and Materials*, **43**: 11 (1996).

41. Al Amrousiz F.A., Al Sobagh A.M. and Osman M.M.: *Fuel*, **75**: 1193 (1996).