

# Effect of Natural Products on the Corrosion of Titanium and Its Alloy in NaOH Solutions

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## Abstract

The corrosion behavior of pure titanium and Ti-6Al-4V alloy in different concentrations of NaOH was investigated by using different techniques. The effect of bovine serum albumin and egg albumin on corrosion behavior of pure titanium and Ti-6Al-4V alloy has been studied. It has been found that natural products such as (bovine serum albumin and egg albumin) inhibit the corrosion process of Ti-6Al-4V alloy more than pure titanium. The inhibition efficiency increases as the concentration of protein in the solution increases, as was confirmed by SEM.

**Keywords:** Egg albumin, Bovine albumin, NaOH

## 1. Introduction

Titanium and its alloys are attractive material due to their low density, high specific strength and high corrosion resistance. It exhibits exceptional resistance to a broad range of acids, alkalis, natural waters and industrial chemicals. This is due to the formation of a protective and self-adherent oxide film, amorphous, mainly TiO<sub>2</sub> (Young, 1961; Badawy, Felske, & Plieth, 1989; Kozlowski, Smyrl, Atanasoska, & Atanasoski, 1989; Schultze & Thietke, 1989). The morphological and chemical characterization of biomimetic bone like apatite formation on alkali treated (Ti-6Al-4V) titanium alloy. This discussion is an attempt to enhance the apatite – forming ability of titanium metal induced by the alkaline (NaOH) treatment (Faure, Balamurugan, Benhayoune, Torres, Balossier, & Ferreira, 2009). Sul (2009) studied the electrochemical oxide growth behaviour on titanium in acid and alkaline electrolytes. The influence of proteins on the fretting-corrosion behaviour of a Ti-6Al-4V alloy was studied by Hiramoto and Mischler (2006), those proteins were found to act as cathodic inhibitor by shifting the corrosion potential and the cathodic current towards more cathodic values. The adsorption of proteins on metal surfaces studied by ellipsometric and capacitance measurements, the effect of the nature of the metal on the amount and the configuration of adsorbed lysozyme and ovalbumin has been investigated (Bengt A. Ivarsson, Per-Olof Hegg, K. Ingemar Lundström, & Uif Jönsson, 1985). Titanium and titanium alloys are widely used as implant materials due to their excellent corrosion resistance and biocompatibility (Kovacs & Davidson, 1996; Rater, Johnston, & Lenk, 1987; Brems, 1988; Hunt & Stoichet, 2001; Williams, 1981; Fonseca & Barbosa, 2001; Semlitsch, Staub, & Weber, 1985; Healy & Ducheyne, 1992). The biocompatibility of metallic implant materials is closely related to their corrosion behavior. Corrosion and surface film dissolution are two mechanisms for introducing additional ions into the human body. It has been concluded that biocompatibility is determined, not only by corrosion products but, also, by the exchange currents and reaction products of different redox processes involving tissue compounds. It has been reported in some cases that, titanium implants (non-anodized), which have been inserted into the human body for a number of years, have shown high oxidation rates and relatively high amounts of titanium compounds have been found in tissue adjacent to the implant (Toshiaki Ohtsuka, Minoru Masuda, & Norio Sato, 1986). By anodizing titanium previous to its implantation in the body, the corrosion rate decreases and the rate of bone healing highly increases (Arsov, Kormann, & Plieth, 1991).

Titanium alloys in orthopaedic implants are susceptible to mechanical disruption of the passive film in a tribo-electrochemical cell on commercial pure titanium and Ti-6Al-4V alloy in inorganic buffer solutions in the pH range from 2.0 to 12.0 and calf bovine serum at pH 4.0 and 7.0 (Contu, Elsener, & Böhni, 2004).

The aim of the present work is to study the electrochemical behavior of pure titanium and Ti-6Al-4V alloys in NaOH and study the effect of proteins (bovine serum albumin and egg albumin) as corrosion inhibitor.

## 2. Experimental

Circular electrodes with working surface area of 1.14 and 1.54 cm<sup>2</sup> for pure titanium (99.999 %) and Ti-6Al-4V alloy respectively were used. The electrodes were fixed to glass tubing with araldite adhesive, the electrical contact was made through a thick copper wire soldered to the inner side of the electrodes. Prior to each experiment, the surface of the working electrode was performed by polishing with different grades of emery papers (800-1200), then diamond past (1 μm), washing by distilled water and ethanol and quickly inserted in the cell. The counter electrode was platinum sheet. The potential was recorded relative to saturated calomel electrode (SCE). Different concentrations of NaOH solutions (1 x 10<sup>-3</sup> - 6 M) and different concentrations of protein additives bovine serum albumin and egg albumin (0.25-1 %), were prepared. Potentiodynamic polarization studies were carried out in the potential range (-2000 to +1500 mV) at scanning rate 2 mV/S. This studies were performed on electronic potentiostat (PGZ 301 Dynamic EIS Voltammetry). Potentiodynamic cyclic polarization was measured also by using electronic potentiostat (PGZ 301 Dynamic EIS voltammetry), at scanning rate 10 mV/S. The cyclic polarization were swept from (-1200 to +1200 mV). The surface morphology of the electrodes before and after immersion in test solutions were observed by scanning Electron Microscopy (JEOL-JSM-5410, Japan).

## 3. Results and Discussion

### 3.1 Potentiodynamic polarization measurements

Typical potentiodynamic curves of pure titanium and Ti-6Al-4V alloy were obtained in different concentrations (1 x 10<sup>-3</sup> to 5 x 10<sup>-1</sup> M) of aerated NaOH solutions. In sodium hydroxide solution an active – passive transition is observed (Contu, Elsener, & Böhni, 2004). For both electrodes, they are representatively shown in Figures (1 and 2). As it is difficult to define a sufficient linear Tafel region in the anodic arm of the observed polarization plots, the value of the corrosion current density was obtained by using an interpolation method (Hefny, Dardir, & Abdel-Tawab, 2004). The electrochemical parameters obtained from the polarization curves in Figures (1 and 2) are given in Table 1. The results show that the corrosion potential shifts to more negative values by increasing concentrations of alkali. Also the corrosion current ( $I_{corr}$ ) and corrosion rate (C.R) increases markedly by increasing the alkali concentration. In order to increase its corrosion resistance and application, therefore, a thin layer of titanium oxide is produced on the surface by chemical and electrochemical methods, titanium has been anodized in 20 M of NaOH solution under potentiostatic conditions at constant voltage of 25 and 35 V (Afshar & Vaezi, 2003).

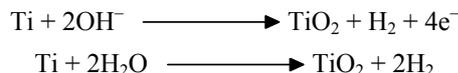
### 3.2 Potentiodynamic cyclic anodic polarization measurements

#### 3.2.1 Potentiodynamic cyclic anodic polarization measurements for pure Ti and Ti-6Al-4V alloy electrodes in NaOH solutions

The cyclic polarization curves of pure titanium and Ti-6Al-4V alloy in alkaline (NaOH) solution are shown in Figures (3 and 4). The concentration ranges from 4 to 6 M, using scan rate 10 mV/sec, in potential range from -1300 to +1300 mV (SCE) at room temperature. Values of associated electrochemical parameters, corrosion potential, protection potential, corrosion current, protection current, pitting current and corrosion rate are listed in Table 2. From the cyclic polarization curves Figures (3 and 4), the reverse scan came above the forward scan, indicating that the made of degradation with both pure titanium and Ti-6Al-4V alloy are due to uniform corrosion (Gurrappa & Venugopala Reddy, 2005). From Table 2, it is clear that, corrosion potential,  $E_{corr}$ , protection potential  $E_p$ , values decreased (shift) in the negative direction by increasing the concentration of NaOH. While the values of  $I_{corr}$ ,  $I_p$ ,  $I_{pit}$  and C.R., increases in both two electrodes except in alloy, the corrosion rate (C.R.) decrease by increasing the concentration of NaOH solution these results indicate that the Ti-6Al-4V alloy is resistant to uniform corrosion in alkali solutions than pure Ti and may form a passive layer on the surface of the electrode. At alkaline range, it was likely that oxygen reduction was the cathodic partial reaction, while the reaction leading to the formation of TiO was the anodic partial reaction (Contu et al., 2004).



The mechanism for the titanium oxide formation on Ti in NaOH solution is proposed (Pjescic, Mentus, Kommenic, & Blagojevic, 2000).



### 3.2.2 Effect of addition egg-albumin and bovine serum albumin on the potentiodynamic cyclic anodic polarization of pure Ti and Ti-6Al-4V alloy in 4.0 M NaOH solution

The anodic polarization curves of the two electrodes immersed in 4.0 M NaOH solution with and without different concentrations (0.25-1 %) of egg albumin and bovine serum albumin are reported in Figures (5-8). Sodium hydroxide with and without the additives of active-passive transition is observed for two test electrodes. It is clear from the curves that in pure Ti, the area of the loop is increased by increasing the concentration of egg albumin and bovine serum albumin, but in Ti-6Al-4V alloy, the area is almost the same. Comparison of the curves plotted for pure Ti and Ti-6Al-4V alloy in 4.0 M NaOH with and without different concentrations of some proteins additive show that egg albumin and bovine serum albumin shifted corrosion and protection potential to the more negative values in case of pure Ti and Ti-6Al-4V alloy by increasing the additives concentration. And the hysteresis loop of the reverse scan increase in case of pure Ti, but not affected in Ti-6Al-4V alloy. In contrast corrosion current, protection and pitting current decreases. It is clear from Tables (3, 4), inhibition efficiency (I.E %) increased with increasing of protein concentration. Maximum inhibition efficiency was obtained at concentration 1.0 % V/V) of egg albumin and bovine serum albumin. Ti-6Al-4V alloy was significantly more inhibited than pure Ti by the two additives, and egg albumin more efficient than bovine serum. This finding suggests that, egg albumin and bovine serum albumin behaves like as interface inhibitor that from adsorption layer interacting with the metal or alloy surface. Therefore, protein adsorbing onto the electrode surface reduce the effective surface area available for the cathodic reactions to occur (Contu et al., 2004).

### 3.3 Scanning electron microscope (SEM) for Ti-6Al-4V alloy

To confirm the degradation mechanism of Ti-6Al-4V alloy after being corroded in NaOH solution were observed under the scanning electron microscope (SEM). Figure (9, A-C) illustrate the morphology surface of Ti-6Al-4V alloy specimen after potentiodynamic cyclic anodic polarization treatment in 4.0 M NaOH in presence and absence of some proteins such as (egg albumin and bovine serum albumin). As can be seen from Figure 9 (A) in NaOH without addition of protein, the alloy was corroded uniformly and the mode of degradation is due to uniform corrosion, while Figure 9 (B) and (C) show the optical micrographs for the alloy surface in 4.0 M NaOH solution containing 1 % of both proteins. The effect of both proteins adsorbs onto the electrode surface. Therefore, protein molecules adsorbing onto the electrode surface reduce the effective surface area available for the cathodic reactions to occur. Egg albumin and bovine serum behaves like a corrosion inhibitor in sodium hydroxide. The inhibition efficiencies of egg albumin are higher than bovine serum. These results were supported by the reported data for the alloy in Table 4. SEM results are in good agreement with the cyclic polarization data. The structure and thickness of the anodic layer depend on the type of solution and the anodizing method (galvanostatic or potentiostatic). Thick anodic layers are produced in concentrated basic solutions (pH > 13) (Hugot-Le Goff, 1996).

## 4. Conclusion

- 1) The effect of NaOH solutions on the behaviour of two electrodes (pure titanium and Ti-6Al-4V alloy) were investigated by potentiodynamic polarization, corrosion potential shifts to more negative values by increasing its conc., and also  $I_{\text{corr}}$ . Increases markedly by increasing the alkali concentration.
- 2) The potentiodynamic cyclic anodic polarization technique were carried and for pure titanium and Ti-6Al-4V alloy in different conc. of NaOH solutions, the results indicated that the mode of degradation with both pure Ti and Ti-6Al-4V alloy are due to uniform corrosion.
- 3) The natural products, egg albumin and serum behaves similarly to an inhibitor. It is clear from the results of NaOH solutions containing different concs. from egg albumin and bovine serum that, Ti-6Al-4V alloy was significantly more inhibited than pure titanium. These results were confirmed by using scanning electron microscope (SEM).

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Table 1. The electrochemical parameters of pure titanium and (Ti-6Al-4V) alloy electrodes in different concentrations of NaOH solutions

| Electrodes        | Conc. M | $E_{corr.}$<br>mV | $I_{corr.}$<br>mA/cm <sup>2</sup> | Tafel slopes |           | Corrosion rate(C.R.) $\mu$ m/y |
|-------------------|---------|-------------------|-----------------------------------|--------------|-----------|--------------------------------|
|                   |         |                   |                                   | $\beta_a$    | $\beta_c$ |                                |
| Pure Titanium     | 0.5     | -958.5            | 3.5006                            | 395.90       | -490.00   | 40.94                          |
|                   | 0.1     | -957.3            | 0.0033                            | 301.50       | -277.10   | 38.61                          |
|                   | 0.01    | -929.9            | 0.0006                            | 431.70       | -195.50   | 7.098                          |
|                   | 0.001   | -735.4            | 0.0002                            | 327.10       | -171.00   | 2.112                          |
| (Ti-6Al-4V) Alloy | 0.5     | -858.0            | 52.71                             | 3001.6       | -2257.4   | 616.6                          |
|                   | 0.1     | -448.3            | 31.74                             | 4183.5       | -1480.5   | 371.2                          |
|                   | 0.01    | -396.7            | 25.96                             | 3472.2       | -2813.7   | 303.7                          |
|                   | 0.001   | -367.3            | 19.18                             | 5619.3       | -2745.3   | 224.3                          |

Table 2. The corrosion parameters of pure titanium and (Ti-6Al-4V) alloy electrodes in different concentration of NaOH solution

| Electrodes     | Conc. M | $E_{corr}$<br>mV | $E_{protect}$<br>mV | $I_{corr.}$<br>mA/cm <sup>2</sup> | $I_{protect}$<br>mA/cm <sup>2</sup> | $I_{pit}$<br>A/cm <sup>2</sup> | Corrosion rate<br>(C.R.) $\mu$ m/y |
|----------------|---------|------------------|---------------------|-----------------------------------|-------------------------------------|--------------------------------|------------------------------------|
| Pure Titanium  | 4.0     | -822.70          | -0.6169             | 102.7229                          | 3.691                               | 2.946                          | 120.1                              |
|                | 5.0     | -1000.7          | -0.9282             | 196.8602                          | 4.408                               | 3.569                          | 230.2                              |
|                | 6.0     | -1031.3          | -0.956              | 205.3385                          | 4.622                               | 3.700                          | 240.0                              |
| Titanium alloy | 4.0     | -574.0           | -0.2668             | 3.234100                          | 1.548                               | 2.372                          | 160.9                              |
|                | 5.0     | -714.70          | -0.5224             | 16.20310                          | 3.138                               | 3.870                          | 189.5                              |
|                | 6.0     | -974.9           | -0.8781             | 137.6454                          | 3.317                               | 4.040                          | 37.82                              |

Table 3. The corrosion parameters of pure titanium electrode in 4.0 M NaOH solution containing different concentrations of both egg albumin and serum bovine albumin

| Type of proteins     | Conc. % | $E_{corr}$<br>mV | $E_{protect}$<br>mV | $I_{corr.}$<br>mA/cm <sup>2</sup> | $I_{protect}$<br>mA/cm <sup>2</sup> | $I_{pit}$<br>mA/cm <sup>2</sup> | Corrosion rate (C.R.)<br>$\mu$ m/y | Inhibition<br>Efficiently I.E % |
|----------------------|---------|------------------|---------------------|-----------------------------------|-------------------------------------|---------------------------------|------------------------------------|---------------------------------|
| egg Albumin          | 0       | -822.70          | -0.6169             | 102.7229                          | 3.691                               | 2.946                           | 120.1                              | -                               |
|                      | 0.25    | -892.40          | -0.7892             | 37.71600                          | 3.922                               | 3.376                           | 112.7                              | 6.160                           |
|                      | 0.5     | -116.80          | -1.0390             | 24.35710                          | 4.030                               | 3.503                           | 99.80                              | 17.38                           |
|                      | 1       | -1159.8          | -1.1060             | 18.78710                          | 4.113                               | 3.779                           | 95.22                              | 20.71                           |
| Serum bovine Albumin | 0       | -822.70          | -0.6169             | 102.7229                          | 3.691                               | 2.946                           | 120.1                              | -                               |
|                      | 0.5     | -862.90          | -0.6990             | 37.28450                          | 3.850                               | 3.442                           | 110.8                              | 7.740                           |
|                      | 0.75    | -926.20          | -1.0080             | 35.14020                          | 3.900                               | 3.478                           | 98.70                              | 17.81                           |
|                      | 1       | -1183.0          | -1.01280            | 15.88870                          | 4.000                               | 3.550                           | 92.50                              | 22.98                           |

Table 4. The corrosion parameters of Ti alloy electrode in 4.0 M NaOH solution containing different concentrations of both egg albumin and serum bovine albumin

| Type of proteins     | Conc.% | $E_{corr}$ mV | $E_{protect}$ mV | $I_{corr.}$ mA/cm <sup>2</sup> | $I_{protect}$ mA/cm <sup>2</sup> | $I_{pit}$ mA/cm <sup>2</sup> | Corrosion rate (C.R.) $\mu$ m/y | Inhibition Efficiency (I.E.) % |
|----------------------|--------|---------------|------------------|--------------------------------|----------------------------------|------------------------------|---------------------------------|--------------------------------|
| egg Albumin          | 0      | -574.00       | -0.2668          | 3.2341                         | 1.548                            | 2.372                        | 160.9                           | -                              |
|                      | 0.25   | -861.00       | -0.669           | 3.1220                         | 1.445                            | 2.262                        | 104.0                           | 35.36                          |
|                      | 0.5    | -945.60       | -0.8059          | 2.9880                         | 1.350                            | 2.155                        | 97.67                           | 39.29                          |
|                      | 1      | 1023.9        | -0.8504          | 2.8700                         | 1.22                             | 2.090                        | 90.44                           | 43.79                          |
| Serum bovine Albumin | 0      | -574.00       | -2668            | 3.2341                         | 1.548                            | 2.372                        | 160.9                           | -                              |
|                      | 0.5    | -680.90       | -0.3057          | 0.6572                         | 1.300                            | 2.301                        | 134.9                           | 16.15                          |
|                      | 0.75   | -736.70       | -0.3446          | 0.1154                         | 1.197                            | 2.282                        | 120.8                           | 24.92                          |
|                      | 1      | -1183.0       | -0.3557          | 0.0260                         | 0.988                            | 2.282                        | 111.5                           | 30.70                          |

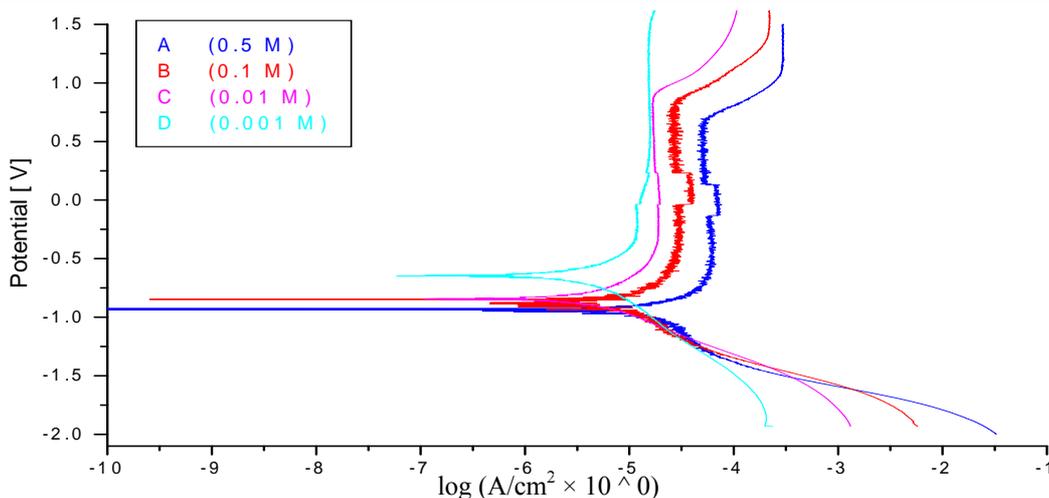


Figure 1. Potentiodynamic polarization curves of pure titanium electrode in different concentrations of NaOH solutions

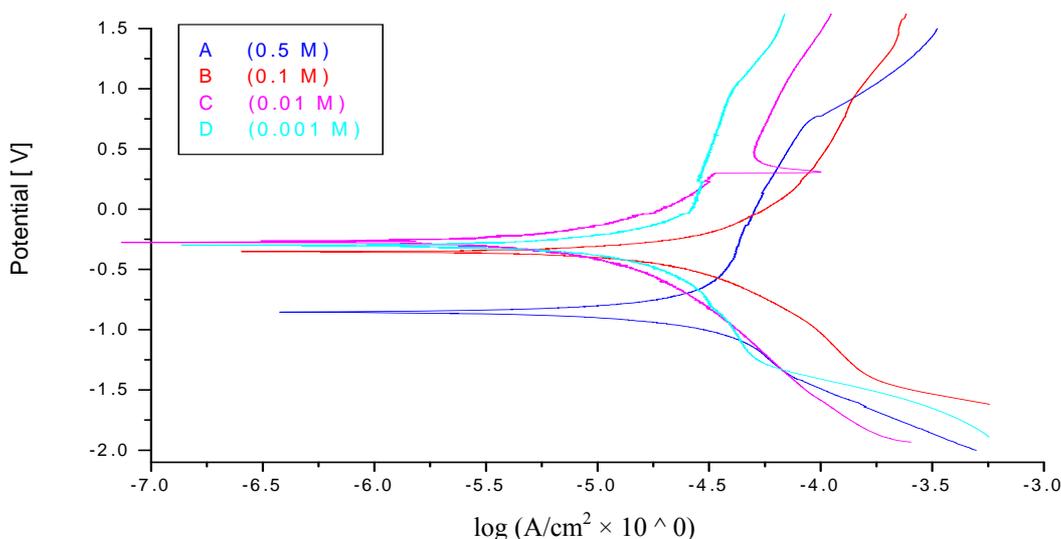


Figure 2. Potentiodynamic polarization curves of titanium alloy electrode in different concentrations of NaOH solutions

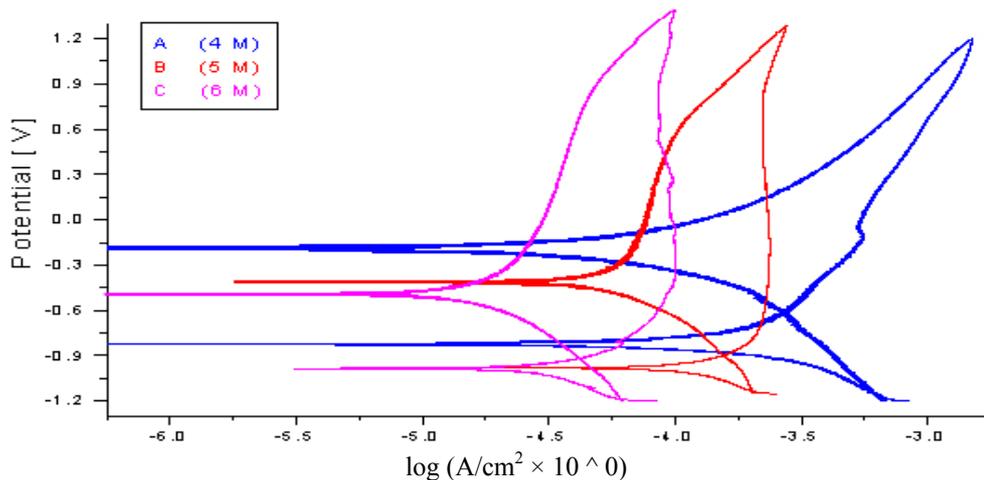


Figure 3. Cyclic anodic polarization curves of pure titanium electrode in different concentrations (4-6 M) of sodium hydroxide solutions

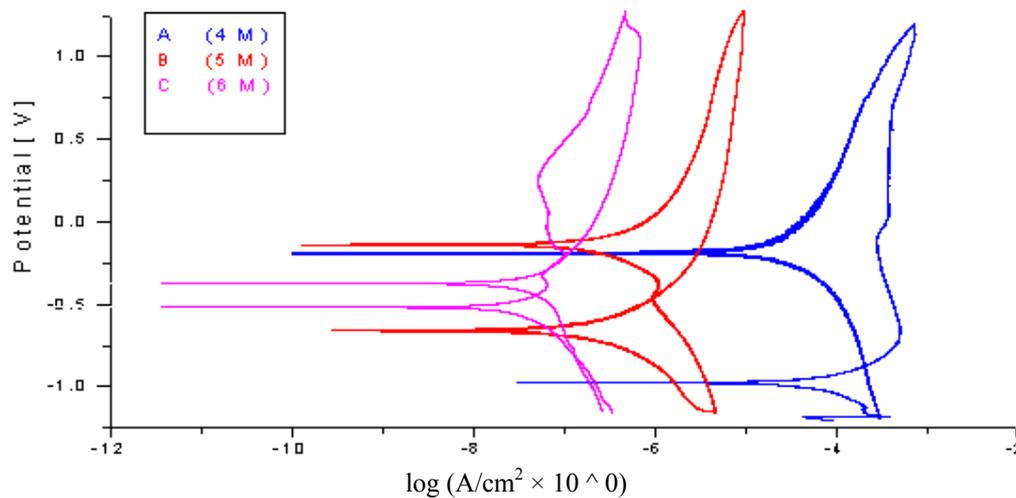


Figure 4. Cyclic anodic polarization curves of titanium alloy electrode in different concentrations (4 – 6 M) of sodium hydroxide solutions

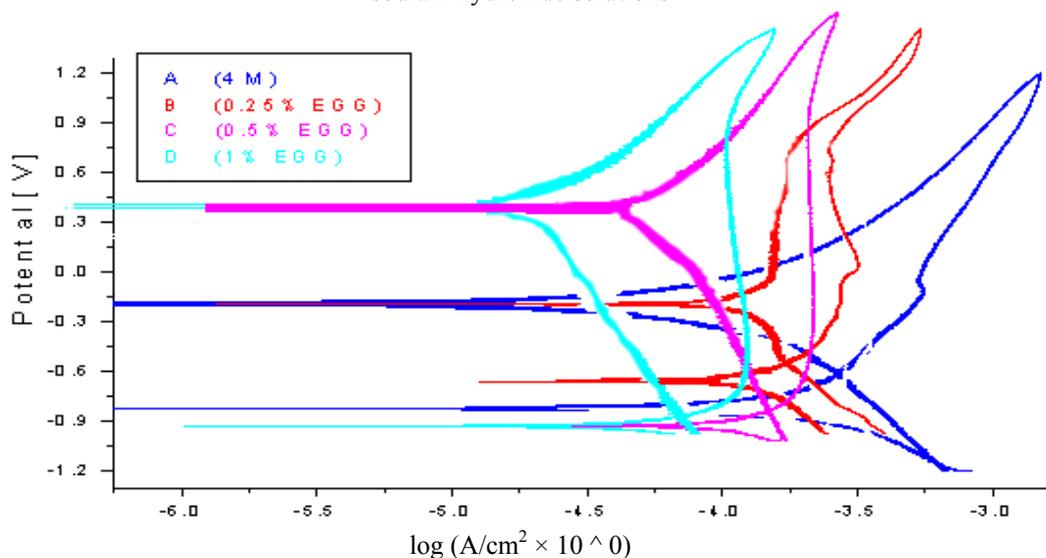


Figure 5. Cyclic anodic polarization curves of pure titanium electrode in (4 M) NaOH with and without different percentage (0.25-1 %) of egg albumin

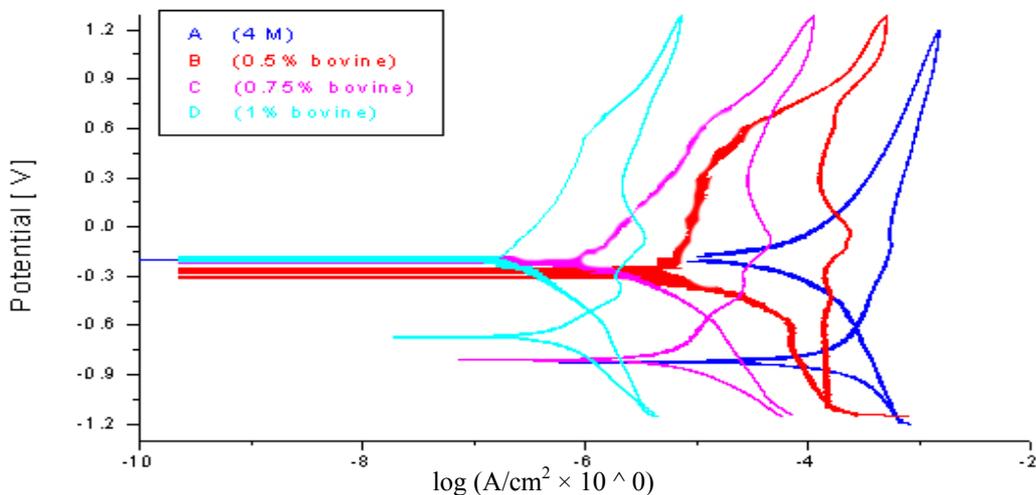


Figure 6. Cyclic anodic polarization curves of pure titanium electrode in (4 M) NaOH with and without different percentage (0.5-1 %) of bovine serum albumin

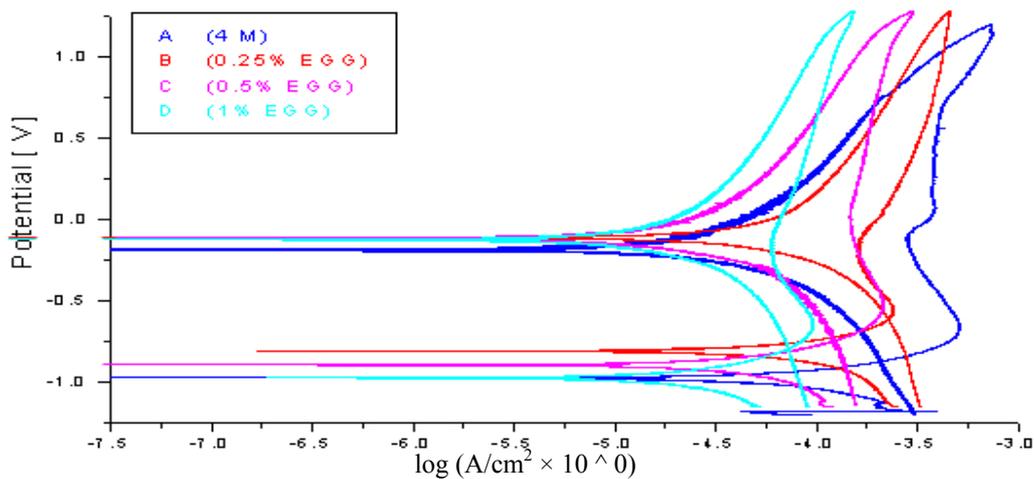


Figure 7. Cyclic anodic polarization curves of titanium alloy electrode in (4 M) NaOH with and without different percentage (0.25-1 %) of egg albumin

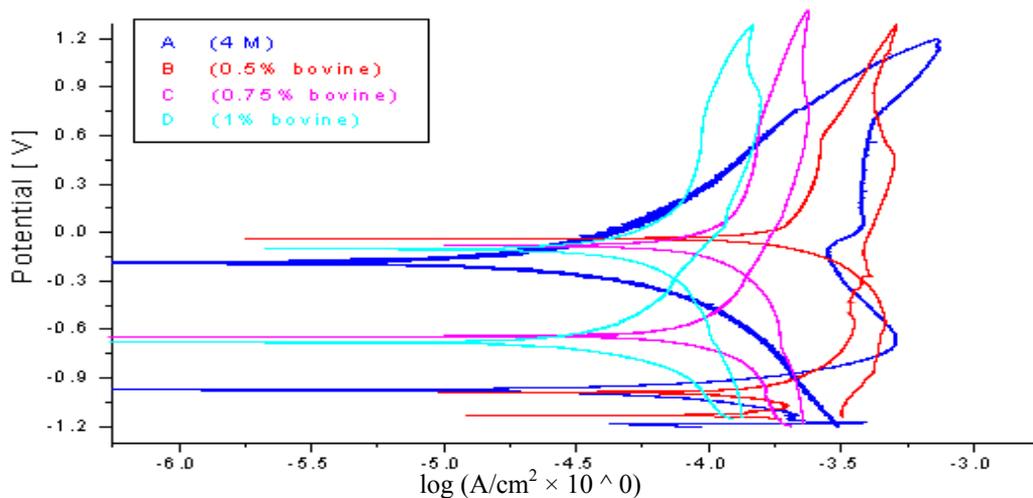
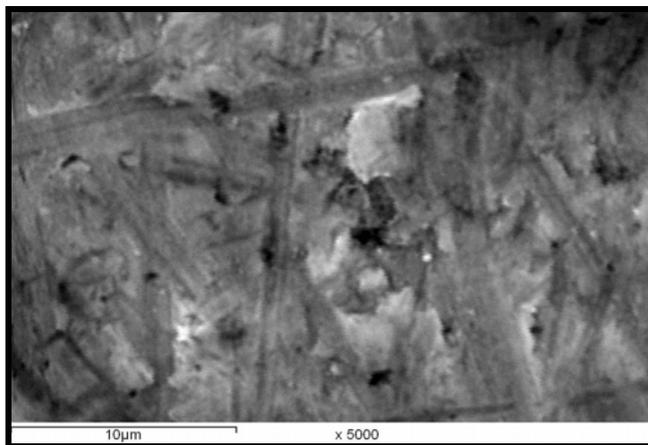
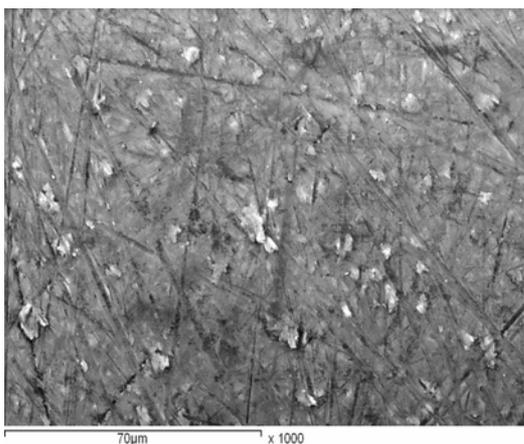


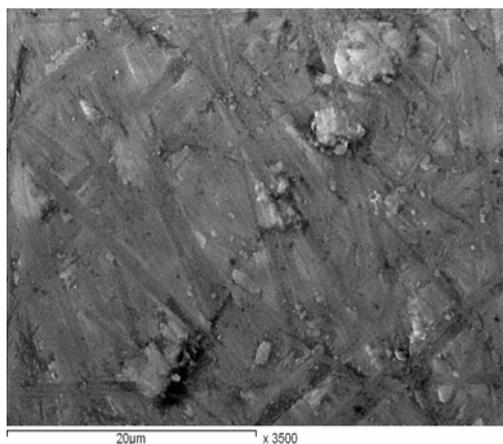
Figure 8. Cyclic anodic polarization curves of titanium alloy electrode in (4 M) NaOH with and without different percentage (0.5-1 %) of bovine serum albumin



(A)



(B)



(C)

Figure 9. Surface morphology of of titanium alloy (Ti-6Al-4V) electrode

(A) in 4.0 M NaOH solution (B) in 4.0 M NaOH solution +1 % egg albumin (C) in 4.0 M NaOH solution +1 % bovine albumin