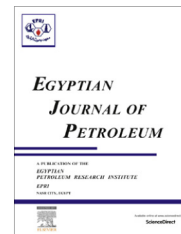


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FULL LENGTH ARTICLE

Corrosion behavior and electrochemical properties of carbon steel, commercial pure titanium, copper and copper–aluminum–nickel alloy in 3.5% sodium chloride containing sulfide ions

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Abstract In this research the electrochemical performance of Cu, Cu–10Al–10Ni, Cp-Ti and C-steel in 3.5% sodium chloride containing sulfide ions was investigated. Different electrochemical methods such as polarization technique and electrochemical impedance spectroscopy, EIS, were used. Surface examination and morphological studies were employed. The effect of immersion time of the different alloys was extensively studied by EIS. A comparison was made between the electrochemical stability of the different alloys in this media. Polarization measurements reveal that Cp-Ti and Cu–10Al–10Ni alloys possess lower corrosion rates than both Cu and carbon steel. EIS measurements, have shown that a more thicker and resistive passive film is formed on Cp-Ti alloy due to the formation of TiO₂ film of duplex nature. The incorporation of Ni in the Cu₂O barrier film leads to the stabilization of Cu–10Al–10Ni alloy and the stability is enhanced in the presence of Al. The thickness and resistance of the barrier layer formed on the alloy surfaces increase with the increasing the immersion time. The formation of such passive film on the surface of different alloys was discussed. An equivalent circuit model for the electrode/electrolyte interface under different conditions was proposed. The experimental impedance data were fitted to the theoretical data according to the proposed model.

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1. Introduction

Hydrogen sulfide, H₂S, corrosion has been a research topic for several decades [1]. Most studies on this subject were based on an engineering perspective because of fractures in metals caused by H₂S especially in the case of steel [2–4]. Also the electrochemical analysis technology has been widely used in

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recent decades to study the corrosion process and has been proven to be effective [5–10]. Copper and copper alloys belong to the group of alloys having sufficient resistance to corrosion in seawater. However, they corrode under specific conditions. Hydrogen sulfide is the most common pollutant which may cause an intensive corrosion of copper and copper alloys in seawater where the presence of sulfide ions lead to the modification of the oxide layer on copper and copper based alloys. Acceleration of corrosion is due to the enhancement of the cathodic process. One of the most important parameters playing a major part in the processes of corrosion of copper alloys in seawater polluted with sulfides is their concentration. Traverso et al. [11] have found that the most intensive corrosion is caused by sulfides at initial levels of 4 ppm. Gudas and Hack [12] stated that wrought 90/10 Cu–Ni is susceptible to accelerated attack in seawater containing only 0.01 ppm of sulfides. Mukhopadhyay, Baskaran [13] and Chauhan, Gadiyar [14] determined the chemical composition of corrosion products created on 70/30 Cu–Ni in sulfide polluted seawater. Instead of Cu_2O and green $\text{CuCl}_2 \cdot 3\text{Cu}(\text{OH})_2 \cdot 3\text{H}_2\text{O}$, a gray layer of $\text{Cu}_2\text{S}(\text{NiFe})_3\text{S}_4$ and Cu_2O was created. In our recent previous paper the stability of Cu–10Al–05Ni alloy in chloride solutions polluted by sulfide ions was studied [15]. Also the effect of Cl^- and H_2S on steel weld [16–19] was studied. The results demonstrated that H_2S could accelerate both the anodic iron dissolution and the cathodic hydrogen evolution in most cases, but some results showed that H_2S could also inhibit the corrosion of iron under certain special conditions [20,21]. Zimer et al. [22] recently studied the initial stage of pitting corrosion in AISI 1040 steel through temporal series micrographs coupled with an open-circuit potential and polarization curves in an alkaline sulfide solution. Due to a set of excellent properties, chemical and mechanical properties, Titanium and titanium based alloys are commonly employed in many functions such as chemical, marine, space and biomedical, dental and orthopedic applications [23–25]. The high corrosion resistance of titanium in aggressive environments is ensured by a compact and chemically stable oxide film that spontaneously covers the metal surface which is amorphous or poorly crystalline and mainly composed of titanium oxide, TiO_2 [26]. Though much effort has been put into separate individual studies for each alloy with the aggressive media, H_2S and/or NaCl, little research has been carried out on collection studies including different alloys in these media. In this work, some common alloys namely, Cu, Cu–10Al–10Ni, C-steel and commercially pure titanium, Cp-Ti, are investigated in both 3.5% NaCl and 3.5% NaCl polluted by sulfides. The mechanism of corrosion processes taking place at the electrode/solution interface will be discussed, and a categorization of the different investigated alloys according to their stability in this aggressive medium can be made.

2. Experimental details

2.1. Materials and sample preparation

The working electrodes were made from commercial grade Cu, Cu–10Al–10Ni, carbon steel and commercial pure Ti. The rods were prepared in metallurgical work shop. The prepared alloys are cast alloys without any special treatment. The metallic rods are mounted into glass tubes by two-component epoxy resin

leaving a surface area of 0.2 cm^2 to contact the solution. Chemical composition of Titanium in mass percent: 0.30 Fe, 0.25 O, 0.06 C, 0.03 N, 0.015 H and balance Ti. The chemical composition of carbon steel is: 0.34 C, 0.93 Mn, 0.26 Si, 0.0.2 S, 0.04 P, 0.01 Cu, 0.01 Cr, 0.02 Ni and balance Fe. The chemical composition of pure Cu and Cu–10Al–10Ni alloy electrodes is: Cu (99.928 Cu, 0.001 Al, 0.02 Ni, 0.007 Zn, 0.003 Mn, 0.033Sn, 0.002 Fe, 0.004 Si, 0.001 Mg and 0.001 Ti) and Cu–10Al–10Ni (76.00 Cu, 11.28 Al, 9.95 Ni, 0.10 Zn, 0.02 Mn, 0.14 Sn, 2.26 Fe, 0.24 Si, 0.01 Mg and 0.01Ti). The electrochemical cell is a three-electrode all-glass cell, with a platinum spiral counter electrode and saturated calomel, SCE, reference electrode. Before each experiment, the working electrode was polished using successive grades emery papers down to 2500 grit, washed thoroughly with bi-distilled water, then transferred quickly to the electrolytic cell. The electrochemical measurements were carried out in a stagnant, naturally aerated 3.5 (mass, m/volume , v)% NaCl containing 2 ppm S^{2-} solution. The source of sulfide ions in this research is Na_2S salt.

2.2. Electrochemical measurements

The corrosion behavior of the specimens under investigation were monitored using electrochemical impedance spectroscopy, EIS, and DC polarization techniques during immersion in 3.5% NaCl and 3.5% NaCl containing 2 ppm S^{2-} solution open to air and at room temperature. The polarization experiments and EIS, investigations were performed using a Voltalab PGZ 100 “All-in-one” Potentiostat/Galvanostat. The potentials were measured against and referred to the standard potential of the SCE (0.245 V vs. the standard hydrogen electrode, SHE). To achieve quasi-stationary condition, the polarization experiments were carried out using a scan rate of 5 mV s^{-1} . The total impedance, Z , and phase shift, θ , were measured in the frequency range from 10^5 to 0.1 Hz. The superimposed ac-signal amplitude was 10 mV peak to peak as assigned by measuring system. To achieve reproducibility, each experiment was carried out at least twice, where a range of 2–3 mV was considered to be reasonable for open-circuit potential measurements and 5 mV for the polarization experiments. The surface morphology of some electrodes was investigated by scanning electron microscopy, SEM (Model Quanta 250 FEG, FEI Company, Netherlands).

3. Results and discussion

3.1. Potentiodynamic polarization measurements

The electrochemical behavior of the different alloys was investigated by the polarization technique. Tafel extrapolation measurements were used. The shape of polarization curves can obtain information on the kinetics of the corrosion reactions. The polarization curves of the alloys were recorded after 240 min of electrode immersion in stagnant naturally aerated 3.5% NaCl containing 2 ppm S^{2-} ions at a scan rate of 10 mV/s and $25\text{ }^\circ\text{C}$. Polarization curves for Cu, Cu–10Al–10Ni, C-steel and Cp-Ti are presented in Fig. 1. The fitted values of corresponding kinetic parameters such as anodic and cathodic Tafel slope (β_a and β_c respectively), corrosion potentials, E_{corr} , and corrosion current densities, i_{corr} , are listed in Table 1. Polarization curve for Cu shows an obvious anodic

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