



# Correlative investigation of the corrosion susceptibility of C70600 and C26000 copper based alloys for application in seawater environment

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## ABSTRACT

Appropriate selection of Cu based alloys for petrochemical, energy generation, desalination and piping applications in marine operating conditions is of utmost importance. Correlative investigation of the corrosion susceptibility of two Cu based alloys (C70600 CuNi and C26000 CuZn) was done with potentiodynamic polarization technique and open circuit potential analysis in 0.05 M H<sub>2</sub>SO<sub>4</sub> solution at 0.5%, 1.5%, 2.5%, 3.5% and 4.5% NaCl concentration. The corrosion susceptibility of both alloys were generally similar at 0.5% (0.05 mm/y and 0.06 mm/y), and 1.5% (0.11 mm/y and 0.10 mm/y) Cl<sup>-</sup> ion concentration. Beyond 1.5% concentration C70600 proves to be substantially more corrosion resistant with optimal corrosion rate value of 0.74 mm/y compared to 2.68 mm/y for C26000 at 4.5% NaCl concentration due to variation in the constituent of the oxide layer precipitated on C70600 alloy which differs from C26000. C70600 exhibited dominant cathodic passivation and subsequent corrosion deterioration resulting in visible cathodic potential displacement. C26000, despite its relatively higher corrosion susceptibility underwent metastable pitting activity proportional to the Cl<sup>-</sup> ion concentration. Anodic passivation and selective dissolution of C26000 occurred following anodic polarization due to the presence of Zn in its outer matrix. Open circuit potential measurement shows Cl<sup>-</sup> ion concentration significantly influences the thermodynamic stability, passivation and corrosion resistance of the Cu alloys. However plot characteristics and potential readings showed C70600 to be more corrosion resistant and thermodynamically stable coupled with significantly lower tendency to corrode.

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## 1. Introduction

Corrosion is the degradation of the properties of metallic alloy components and parts due to electrochemical and chemical interactions with their operating environment. Corrosion results in component failure in mechanical systems resulting in progressive reduction of metal thickness, formation of pits and collapse of structures [1,2]. These are responsible for leakage of process fluids, industrial downtime, high maintenance cost and injury to personnel. A report by Electric Power Research Institute states the cost of corrosion to the electric power industry is about \$17 billion with 22% of the total cost being preventable [3]. The National Association of Corrosion Engineers states the total cost of corrosion to the United States economy is about 4% of its GNP [4]. The universal cost of corrosion worldwide in 2013 is about \$2.5 trillion constituting about 3–4% of the global GDP [5]. Cu based metallic alloys are extensively used for condenser tubing, desalination units, heat exchangers, hydraulic lines and piping material for electricity-

generating plants in marine operating conditions due to their excellent mechanical, electrochemical and physical properties such as machinability, corrosion and bio fouling resistance, thermal and electrical properties, versatility and relatively low cost [6–9]. The discharge of Cu anions and the structural configuration of oxide layer precipitated on the alloy surface impart bacteriostatic properties and resistance to micro-fouling and erosion corrosion. The alloys exhibit limited strength and thermal stability under extreme temperature conditions. The corrosion susceptibility of Cu based alloys occurs due to the precipitation of a passive, invisible and strongly adherent inner layer of Cu<sub>2</sub>O film, and a porous thick outer layer with respect to the environment in which they are exposed. Other factors such as Cl<sup>-</sup> anionic concentration, alloy composition, temperature and flow velocity etc. significantly influence the corrosion susceptibility of the alloy [10–14]. The most commonly applied copper based alloys are brass, bronze, aluminum bronze and cupronickels. These alloys are thermodynamically unstable and susceptible to corrosion in seawater [15]. Observation from

previous research shows that the  $\text{Cl}^-$  ion in seawater actively controls the corrosion reaction mechanisms of Cu resulting in the accelerated degradation of the alloy [16–19]. Localized corrosion deterioration on Cu alloy surfaces in seawater is insidious, undetectable, and selective wherever it occurs over microscopic areas [20]. Resistance of Cu to localized corrosion deterioration such as pitting corrosion, hydrogen embrittlement and fatigue corrosion is limited [21]. Failures of heat exchangers tube material for seawater application in refineries and desalination plants constructed from Cu alloys is due to pitting, erosion, and occasionally under deposit corrosion [22]. Deterioration and failure of Cu alloys in component systems such as brine heater, distillate collection systems and piping systems in desalination plants is also due to pitting corrosion [23–26]. Seawater is a major coolant in marine, petrochemical, mining desalination and offshore industry where Cu based alloys are extensively applied. C70600 (copper nickel alloys) and C260 (alpha brass) Cu alloys exhibits appreciable resistance to corrosion in seawater with respect to variation in composition of the electrolyte. C70600 are applied in environments significantly destructive to stainless steels especially seawater conditions. The presence of Ni in Cu enhances its strength, toughness, impingement resistance, durability and corrosion resistance. Ni improves the corrosion resistance of copper by its integration into continuous  $\text{Cu}_2\text{O}$  film. However, the resistance to corrosion is limited with respect to the concentration of  $\text{Cl}^-$  in seawater.  $\text{Cl}^-$  anion selectively corrodes Cu and Zn in C260 Cu alloy [27–29]. Previous research has focussed on the corrosion susceptibility of Cu based alloys in NaCl media with emphasis on the effect of  $\text{Cl}^-$  anions [30–34]. The corrosion susceptibility of the Cu based alloy in a 3.5 wt% NaCl solution was studied by Xia et al [35] who demonstrated that ultrasonic surface rolling process treatment of the alloy surface decreases the corrosion vulnerability of Cu-10Ni alloys. Zhu et al [36] examined the corrosion resistance of novel Cu alloy in artificial seawater. Observation shows the reaction products developed on the surface followed by the growth of dyoxide precipitates. Evaluation of the corrosion resistance of isomorphous Cu-Ni alloy in 3 wt% NaCl media by Huang et al. [37] showed increase in corrosion susceptibility of the alloy with increase in Ni concentration. Research of the effect of  $\text{SO}_4^{2-}$  anions on the corrosion resistance of C70600 and C260 is limited despite it being a major constituent of seawater. Consequent upon the above, this research investigates the corrosion susceptibility behaviour of C70600 and C260 in chloride-sulphate electrolyte.

## 2. Experimental methods

The nominal (wt. %) composition (Table 1) of cupronickel (C70600) and alpha brass (C26000) Cu alloy plates sourced from Vienna University of Technology, Vienna, Austria was confirmed with PhenomWorld scanning electron microscope at the Materials Characterization Laboratory, Department of Mechanical Engineering, Covenant University, Ota, Nigeria. C70600 and C26000 were cut into 5 samples with average surface area of  $0.25 \text{ cm}^2$  and  $0.20 \text{ cm}^2$ , joined to Cu wire with soldering lead and encased in vercoit acrylic mounts. The Cu alloy surface underwent metallo-

graphic preparation with SiC papers (80 to 1000 grits),  $6 \mu\text{m}$  diamond polishing fluid, and purified  $\text{H}_2\text{O}$  and  $\text{C}_3\text{H}_8\text{O}$ . 200 mL of 0.05 M  $\text{H}_2\text{SO}_4$  was prepared from analar grade reagent of  $\text{H}_2\text{SO}_4$  (98% purity). Standard grade NaCl was prepared in cubic concentrates of 0.5%, 1.5%, 2.5%, 3.5% and 4.5% per 200 mL of 0.05 M  $\text{H}_2\text{SO}_4$  solution. Potentiodynamic polarization test and open circuit potential analysis were carried out with a triple electrode configuration (vercoit mounted C70600 and C26000 electrodes, Ag/AgCl reference electrode and Pt rod counter electrode) stationed inside a glass container with 100 mL of the electrolyte and coupled to Digi-Ivy 2311 potentiostat (connected with a computer). Potentiodynamic polarization plots were obtained at sweep rate of  $0.0015 \text{ V/s}$  between from  $-1 \text{ V}$  and  $+1.25 \text{ V}$ . Corrosion current density  $C_D$  ( $\text{A/cm}^2$ ) and corrosion potential  $C_P$  (V) values were obtained from Tafel extrapolation of anodic-cathodic polarization plots. Corrosion rate  $C_R$  (mm/y) was computed from the mathematical equation [38];

$$C_R = 0.00327xC_DxE_Q/D \quad (1)$$

$E_Q$  represents equivalent weight (g) of the Cu alloys, 0.00327 is corrosion rate constant for dimension and time conversion in mm/y and  $D$  represents density (g). Open circuit potential analysis of C70600 and C26000 placed inside 100 mL of 0.05 M  $\text{H}_2\text{SO}_4$  electrolyte at 0%, 0.5% and 4.5% NaCl concentration was done at step potential of  $0.1 \text{ V/s}$  for 5400 s with 2 electrode configuration (Ag/AgCl reference electrode and vercoit mounted Cu alloy electrode) coupled to Digi-Ivy 2311 potentiostat.

## 3. Results and discussion

### 3.1. Potentiodynamic polarization studies

Potentiodynamic polarization plots of C70600 and C26000 Cu based alloy corrosion in 0.05 M  $\text{H}_2\text{SO}_4$  solution at 0%, 0.5%, 1.5%, 2.5%, 3.5% and 4.5%  $\text{Cl}^-$  ion concentration are shown in Fig. 1(a) and (b). The polarization data obtained is shown in Table 2. The corrosion rate data in Table 2 shows  $\text{Cl}^-$  ions (0.5% – 4.5% NaCl) significantly influence the corrosion resistance of C70600 and C26000 alloys when correlated to the values at 0%  $\text{Cl}^-$  ion concentration. At 0%  $\text{Cl}^-$  ion concentration, corrosion rate of C70600 and C26000 are comparable with values of 0.05 mm/y and 0.06 mm/y which correlates to corrosion current density of  $4.08 \times 10^6 \text{ A/cm}^2$  and  $5.09 \times 10^6 \text{ A/cm}^2$ .  $\text{Cl}^-$  ion concentration at 0.5% gave corrosion rate values of 0.11 mm/y and 0.10 mm/y for the Cu alloys. This observation shows the corrosion resistance of C70600 and C26000 alloy is generally similar for applications in environment with very low  $\text{Cl}^-$  ion concentration. Beyond 0.5%  $\text{Cl}^-$  ion concentration, C70600 alloy appears to be less vulnerable to corrosion. The corrosion rate values for C70600 varies from 0.17 mm/y at 1.5%  $\text{Cl}^-$  ion concentration to 0.74 mm/y at 4.5%  $\text{Cl}^-$  ion concentration while the corresponding corrosion rate values for C26000 varies from 0.80 mm/y to 2.68 mm/y. Increase in  $\text{Cl}^-$  ion concentration caused a proportionate increase in corrosion for both alloys. However, the rate of increase significantly varies due to variation in alloy composition. Surface deterioration occurs with  $\text{Cl}^-$  ions present, thus resulting

**Table 1**  
Constituents (wt. %) of tested Cu alloys.

<b>C70600</b>						
Element Symbol	Pb	Zn	Fe	Ni	Mn	Cu
% Composition	0.05	1	1.5	10	1	86.3
<b>C26000</b>						
Element Symbol	Pb	Zn	Fe	Ni	Mn	Cu
% Composition	0.07	28.38	0.05	–	–	71.5

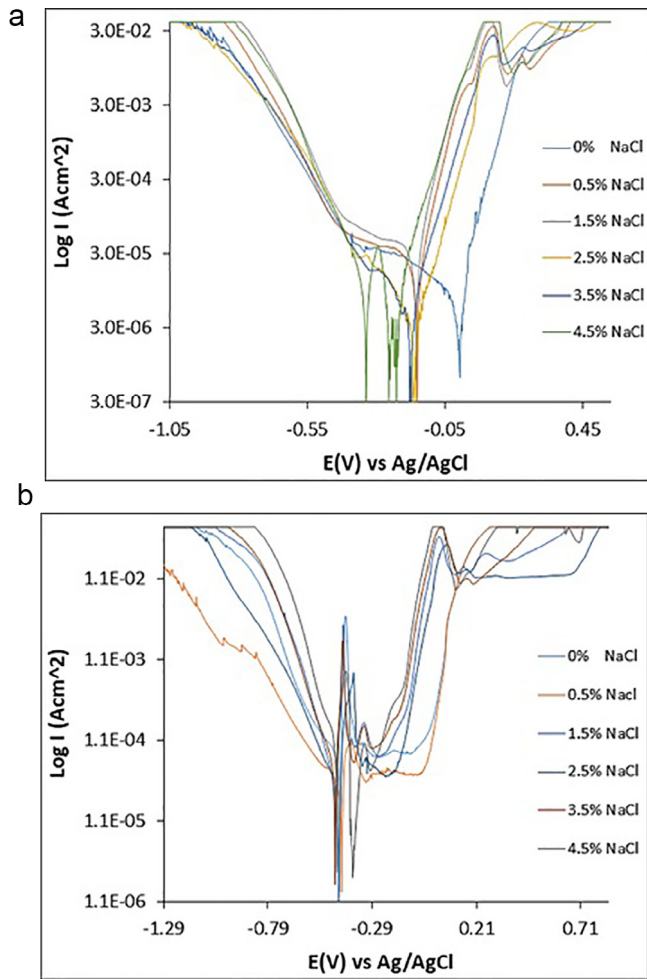


Fig. 1. Potentiodynamic polarization plots of (a) C70600 and (b) C26000 corrosion in 0.05 M H<sub>2</sub>SO<sub>4</sub> at 0% –4.5% NaCl concentrations.

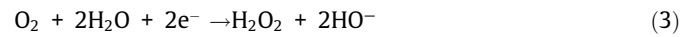
in the growth of an oxide film which hinders the transport of Cu cations [39]. The major product of Cu corrosion is CuCl according to the equation below.



CuCl reacts to produce Cu<sub>2</sub>O, which oxidizes to Cu(OH)<sub>2</sub>, Cu<sub>2</sub>(-OH)<sub>3</sub>Cl and CuCO<sub>3</sub>.Cu(OH)<sub>2</sub> depending on the Cl<sup>-</sup> ion concentration. The excess Cl<sup>-</sup> ions allows for its electrolytic transport from

the sulphate-chloride solution to the alloy surface resulting in significant redox reaction process. The lower corrosion rate values of C70600 alloy compared to the values obtained for C26000 shows the properties of the oxide membrane precipitated on C70600 alloy differs from C26000 due to variation in metallurgical structure and microstructural properties of the alloys. The presence of Ni ions in the lattice structure of Cu<sub>2</sub>O on C70600 decreases the ionic discharge of Cu ions into the electrolyte compared to Zn ions in C26000 [40–42]. Observation of the corrosion potential of C70600 alloy shows a progressive decrease in corrosion potential from 0.002 V at 0% Cl<sup>-</sup> ion concentration to -0.227 V at 4.5% Cl<sup>-</sup> ion concentration. This occurs due to the significant cathodic reactions mechanisms on C70600 surface during polarization. This is further confirmed from the plot in Fig. 1 where cathodic shift of the polarization plots occurred. The plots show significant cathodic passivation due to resistance to the rate of reduction reaction mechanisms whereby the mixed corrosion potential and corrosion current is lowered. The dominant cathodic mechanism is associated with O<sub>2</sub> reduction and limited H<sub>2</sub> evolution reactions on the alloy surface.

Significant cathodic shift occurred from 0% to 0.5% Cl<sup>-</sup> ion concentration and from 3.5% to 4.5% Cl<sup>-</sup> ion concentration. Both are associated with a threshold Cl<sup>-</sup> ion concentration for enhanced cathodic reaction mechanism. The reduction of O<sub>2</sub> results in the production of OH<sup>-</sup> ions according to the equations below;



The cathodic Tafel slope data indicates the cathodic reaction occurs under activation control mechanism at higher cathodic exchange current density. The corrosion potential of C26000 alloy varied with concentration of Cl<sup>-</sup> anions. However, the extent of variation is minimal. C26000 alloy exhibited anodic passivation behavior during potential scanning. The alloy exhibited metastable pitting activity following anodic polarization proportional with Cl<sup>-</sup> ion concentration. The metastable pitting behavior is due to formation of transient corrosion pits which collapses prior to passivation of the Cu alloy. Passivation is characterized by significantly lower anodic current before the alloy experience failure of the inert oxide at the transpassive region of the anodic polarization plot.

### 3.2. Open circuit potential measurement

The OCP plots for C70600 and C26000 Cu alloys in the chloride-sulphate electrolyte at 0%, 0.5% and 4.5% Cl<sup>-</sup> ion concentration is shown in Fig. 2(a) and (b). The plot configuration of C70600 and C26000 at 0% Cl<sup>-</sup> ion concentration shows they are the most electropositive relative to the plots at 0.5% and 4.5% Cl<sup>-</sup> ion concentra-

Table 2  
Potentiodynamic polarization data for C70600 and C26000 corrosion in 0.05 M H<sub>2</sub>SO<sub>4</sub> solution at 0% to 4.5% NaCl concentration (n = 1).

C70600								
Sample	NaCl Conc. (%)	C70600 C <sub>R</sub> (mm/y)	G <sub>i</sub> (A)	G <sub>j</sub> (A/cm <sup>2</sup> )	C <sub>P</sub> (V)	R <sub>p</sub> (Ω)	B <sub>c</sub> (V/dec)	B <sub>a</sub> (V/dec)
A	0	0.05	1.02E-06	4.08E-06	0.002	22590.0	-3.189	14.690
B	0.5	0.11	2.30E-06	9.22E-06	-0.152	19710.0	-6.678	11.750
C	1.5	0.17	3.77E-06	1.51E-05	-0.155	14530.0	-6.454	11.940
D	2.5	0.29	6.30E-06	2.52E-05	-0.140	4078.0	-2.343	15.290
E	3.5	0.47	1.03E-05	4.10E-05	-0.174	2504.0	-1.036	15.220
F	4.5	0.74	1.61E-05	6.42E-05	-0.227	1600.0	-1.960	13.130
C26000								
Sample	NaCl Conc. (%)	C26000 C <sub>R</sub> (mm/y)	G <sub>i</sub> (A)	G <sub>j</sub> (A/cm <sup>2</sup> )	C <sub>P</sub> (V)	R <sub>p</sub> (Ω)	B <sub>c</sub> (V/dec)	B <sub>a</sub> (V/dec)
A	0	0.06	1.02E-06	5.09E-06	-0.435	24113.0	-1.504	-8.533
B	0.5	0.10	1.59E-06	7.93E-06	-0.243	16210.0	-3.535	9.332
C	1.5	0.80	1.31E-05	6.55E-05	-0.451	1961.0	-5.712	-3.550
D	2.5	1.34	2.19E-05	1.09E-04	-0.468	1173.0	-9.410	-2.388
E	3.5	2.13	3.47E-05	1.74E-04	-0.467	740.4	-6.712	-12.450
F	4.5	2.68	4.36E-05	2.18E-04	-0.383	242.0	9.490	-12.900

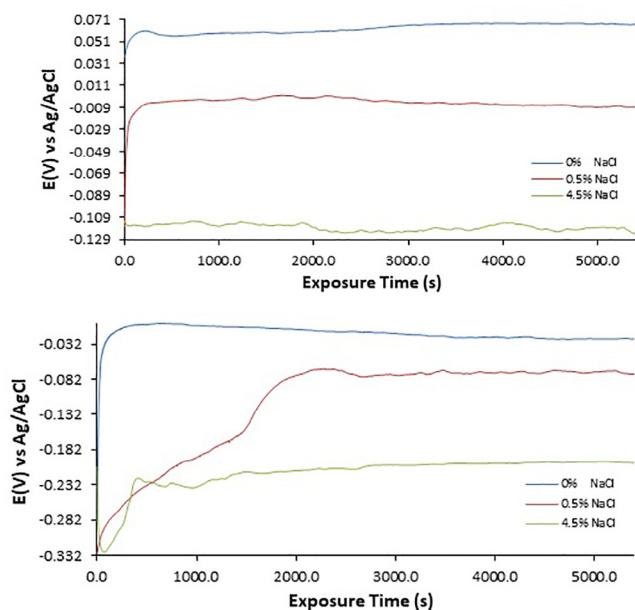


Fig. 2. OCP plots of (a) C70600 and (b) C26000 Cu alloy in 0.05 M H<sub>2</sub>SO<sub>4</sub> solution at 0%, 0.5% and 4.5% NaCl solution.

tion due to formation of Cu<sub>2</sub>O in the absence of chloride which interferes with the surface oxide formation. The absence of Cl<sup>-</sup> ions enhances the positive equilibrium potential of the alloys, inducing high thermodynamic stability. The plot in Fig. 2(a) started at 0.037 V (0 s) and increased at sharp incline to 0.059 V at 300 s due to spontaneous film initiation enhanced by the presence of Ni in C70600. Comparing the observation to the plot in Fig. (b), the plot initiated at relative electronegative potential of -0.284 V (0 s) and progressed for much longer period to -0.005 V at 400 s before attaining thermodynamic equilibrium. This observation shows the presence of Cu in Cu matrix enhances the corrosion resistance of Cu much quicker than the presence of Zn. The OCP plots for both alloys at 0% Cl<sup>-</sup> ion concentration culminated at 0.067 V and -0.024 V (5400 s) signifying higher resistance of C70600 to surface deterioration. Comparing the OCP plots at 0.5% Cl<sup>-</sup> ion concentration [Fig. 2(a) and (b)], its clearly visible that the OCP plot of C26000 attained thermodynamic equilibrium after prolonged instability compared to the OCP plot of C70600. C26000 plot initiated at cathodic potential of -0.329 V (0 s) and progressed to -0.068 V at 2200 s. The corresponding plot for C70600 initiated at -0.118 V (0 s) and attained thermodynamic stability at 200 s (-0.007 V). Comparing the time to attain thermodynamic equilibrium and significant corrosion resistance in the presence of chlorides, C70600 appears to be less susceptible to corrosion and thermodynamically balanced in the presence of low Cl<sup>-</sup> ion concentration. Cu<sub>2</sub>O formed more rapidly on the surface of the alloy matrix and reacts with Cl<sup>-</sup> ions which results in the formation of CuCl before conversion to Cu<sub>2</sub>(OH)<sub>3</sub>Cl. This is further confirmed from the final potential at -0.009 V and -0.075 V at 5400 s. However, the presence of chlorides at 0.5% concentration decreases the corrosion resistance of both Cu alloys compared to their performance at 0% Cl<sup>-</sup> ion concentration. At 4.5% Cl<sup>-</sup> ion concentration, the OCP plots in Fig. 2(a) and (b) is most electronegative relative to the plots at 0% and 0.5% Cl<sup>-</sup> ion concentration due to excess chlorides in the electrolyte. In Fig. 2(a), the OCP plot which initiated at -0.112 V was relatively stable with miniature active-passive transition behaviour before culminating at -0.123 V (5400 s). The corresponding OCP plot for C26000 in Fig. 2(b) was significantly more electronegative, initiating at -0.206 V and undergoing significant

corrosion -0.324 at 100 s before the gradual formation of Cu<sub>2</sub>O till -0.223 V at 400 s where thermodynamic stability was attained till 5400 s at -0.201 V. C70600 proves to be more resistant to the electrolytic transport of Cl<sup>-</sup> ion from the bulk solution to the alloy surface.

#### 4. Conclusion

C70600 copper alloy exhibited higher corrosion resistance than C26000 in sulphate-chloride solution above 1.5% Cl<sup>-</sup> ion concentration. At lower Cl<sup>-</sup> ion concentration their corrosion resistance was generally comparable. However, increase in Cl<sup>-</sup> ion concentration caused a progressive decrease in corrosion susceptibility of both alloys. The alloy content, metallurgical configuration and resulting oxide formation strongly influenced the performance of the alloys in the electrolyte. Cathodic reaction mechanism dominated the redox electrochemical process for C70600 corrosion due to resistance to O<sub>2</sub> reduction. C26000 underwent anodic passivation mechanism before corrosion failure. Open circuit potential study indicates C70600 exhibited significantly lower tendency to corrode, higher thermodynamic stability and more resistant oxide protection compared to C26000 signifying greater resistance to corrosion.

#### CRediT authorship contribution statement

**Roland Tolulope Loto:** Supervision, Conceptualization, Writing – original draft, Visualization, Investigation, Validation, Methodology, Data curation.

#### 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.

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