



Corrosion Behavior of Molybdenum Electrode in Comparison with Gold- Alloy for Dental Application in Some Food Stuff Solutions

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ABSTRACT

This research aimed to examine the opportunity of using Mo electrode in dental applications. Therefore, the behaviors of Mo electrode against Au- alloy in some food stuff solutions i.e. lemon, vinegar and tomato solutions were studied using open circuit potential and potentiodynamic polarization techniques at 30 and 50 °C. The obtained results revealed that under open circuit conditions, there was a tendency of Mo electrode for passivation in all tested solutions. It was in competition with gold electrode to reach the high steady state potential values. Under potentiodynamic polarization technique, the two electrodes showed typical Tafel behavior in all solutions. However, the corrosion rates were always higher for Mo electrode at the higher temperature in all solutions.

Keywords: Au, Mo, Food stuff solutions, Passivation, Corrosion.

INTRODUCTION

The slight malleability of gold alloys enables formation of teeth crowns have high surface quality that can couple safely with other teeth, which in numerous cases offer crowns better than porcelain crowns [1]. On the other hand, the high corrosion resistance, temperature resistance and wear resistance in addition to good thermal and electrical conductivity categorized molybdenum a widespread industrial metal [2]. The high corrosion resistance of Mo is attributed to the existence of skinny inert oxide layer on its surface [3, 4]. The steadiness of this oxide layer is governed by the composition and pH of the electrolyte which momentarily affect its potential. In acidic media, hydrogen diffuses to the molybdenum oxides which associated with high interface capacitance [5]. The behavior of Mo metal in mineral and organic acids created significant research attention [5,6].

It is worthy to mention that, each kilogram of persons mass contains about 0.07 mg of molybdenum. It arises in considerable amounts in the liver and kidneys and in minor amounts in the vertebrate. Molybdenum is also present within human tooth enamel and may help in preventing its decay [7,8].

The present paper studied the behavior of Mo in some food stuff solutions as lemon, vinegar and tomato at two temperatures, 30 and 50 °C and compared with that of Au- alloy in order to make a use of pure Mo in dental applications and to replace the high coast Au- alloy with lower coast Mo metal.

2. EXPERIMENTAL

2.1. Electrode preparation

The working electrodes were prepared from rods of massive cylindrical spectroscopically pure molybdenum supplied by Aldrich – Chemie, and gold alloy electrode of the following composition: 98.2% Au, 1.7% Ti and 0.1% Ir. The rods were fixed to appropriate wide glass tube using epoxy resin Araldite (Ciba– Geigy production, Switzerland) facing 0.196 cm² for Mo and 0.0184 cm² of gold alloy to contact the electrolyte. The surfaces of electrodes were mechanically polished via a standard procedure with emery papers of increasing grit: 320, 600, 800, 1000, 1500 and 2000. After polishing, the electrodes were thoroughly rinsed with bi-distilled water, degreased in ethanol for about 20 sec (to remove the greasy particles) and then again rinsed with bi-distilled water. The electrodes surfaces were finally wrapped against a soft cloth, washed with triply distilled water, and then immersed quickly in the test solutions.

2.2. Preparing the solutions:

Lemon, vinegar and tomato solutions were tested in this study. Extraction of lemon juice was done by cutting, squeezing and filtering its juice in an empty container until it reached the desired volume. The extract was used after dilution with water to be 50% (v/v). Vinegar was purchased from markets and was diluted with water to get a solution of 10% (v/v). To prepare a tomato juice, fresh tomatoes were procured and divided into fine parts in a mixer. The juice was collected into a clean bowl and used after dilution with tap water to 50% (v/v) solution. The pH of the solutions were measured using a

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sensitive pH meter which was calibrated at 25°C using certified commercial buffer at pH 4. The recorded pHs of the three solutions were 2.0, 2.2 and 4.5 for lemon, vinegar and tomato juices respectively.

2.3. Experimental techniques

2.3.1. Open circuit potential measurements

Open circuit potential (OCP) measurements were carried out in a double-walled glass cell filled with 25 ml of the test solution. The potentials vs. time were performed with digital multimeter (KEITHLEY, Model 175, USA) using saturated calomel electrode (SCE) as a reference electrode.

2.3.2. Potentiodynamic polarization measurements

Potentiodynamic polarization measurements were generated using an Electronic Potentiostat Wenking (Model POS 73). The measurements were performed with the aid of a glass cell (25 ml solution) containing three openings for electrodes. A saturated calomel electrode (SCE) provided with a Luggin capillary probe and a platinum sheet (4 cm²) were used as the reference and the auxiliary electrodes, respectively. The working electrodes were polarized by submission to a potentiodynamic scan from -5 to +5 V/SCE at scan rate of 3.33 mV/s and the corresponding currents were recorded. The potentiodynamic curves were recorded after the electrodes were immersed in the test solution for 90 min in order to reach a steady-state OCP value, followed by the polarization measurement. Values of corrosion current (I_{corr}) and corrosion potential (E_{corr}) were evaluated from intersection of the linear anodic and cathodic branches of Tafel plots. Each experiment was repeated at least three times at both 30 and 50 °C. The corrosion rate (C_R) in mpy, was calculated using Eq.1 [9]:

$$C_R = 0.13 \times I_{\text{corr}} \times e/d \quad \dots\dots\dots (1)$$

where 0.13 is the metric and time conversion factor, I_{corr} , is the corrosion current density in $\mu\text{A}/\text{cm}^2$, e and d are the equivalent weight and density of metal in geq/mol and g/cm^3 respectively.

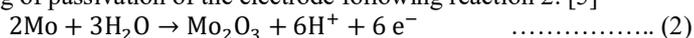
RESULTS AND DISCUSSION

Open circuit potential measurements

The variations of open circuit potential (OCP) of gold– alloy and molybdenum electrodes with time carried out in lemon, vinegar and tomato solutions are presented in Figures (1- 3). The electrode potentials were traced over a period of 90 minutes at two temperatures 30 and 50 °C. The potential at the moment of immersion (E_{imm}) and the steady state potential (E_{ss}) values were given in Table (1).

Figures (1 – 3) illustrate that in lemon, vinegar and tomato solutions, passivation of Mo occurred from the moment of electrode immersion. There was a positive shift in potential with time till steady state values. This indicated that the electrode surface became more passive after longer immersion time in the tested solutions. This behavior denoted that the initial surface films were flawed and undergoes repairing followed by thickening of the surface film until a steady state is attained [10]. In addition, the potential of molybdenum in tomato solution increased sharply and rapidly compared with its potential in lemon and vinegar solutions, indicating less stability in tomato solution.

The increase in potential of Mo electrode in the beginning of immersion in tested aqueous solutions, corresponded to the beginning of passivation of the electrode following reaction 2: [5]



Unexpectedly, the OCP curve of Au– alloy, in lemon solution, dropped sharply during the first 55 and 35 min at 30 and 50 °C, respectively. While in vinegar solution, the potential dropped at the first 3 min at both 30 and 50 °C. In tomato solution, the potential dropped at the first 10 and 5 min at 30 and 50 °C, respectively. Although decreasing of the potential of Au- alloy in the first minutes of immersion, the immersion potential (E_{imm}) of Au- alloy was always higher than that of molybdenum electrode indicating the more passivity of Au- alloy. But dropping of the alloy potential to the negative direction denotes the destruction of pre- immersion oxide film formed on the surface of the Au- alloy.

In lemon solutions (Figure 1), the potential time curves of the two electrodes crossed after 40 and 25 minutes at 30 and 50 °C, respectively. While in vinegar solutions (Figure 2), the curves crossed after 30 and 20 minutes, respectively. Interestingly, after the point of intersection, the molybdenum had more positive potential values than Au- alloy. In other words, the steady state potentials (E_{ss}) of Mo electrode were always higher than those of Au- alloy in lemon and vinegar solutions, indicating its more passivity. This might be due to that Mo_2O_3 was formed in the early stages of passivation and was located close to the metal/film interface [5] which meant that after a certain time, Mo could compete Au- alloy in its order of passivity in such solutions. Moreover, it was also observed that, increasing the temperature causes a decreasing of the electrode potential, indicating a decreasing of the passivity (Figures 1-3).

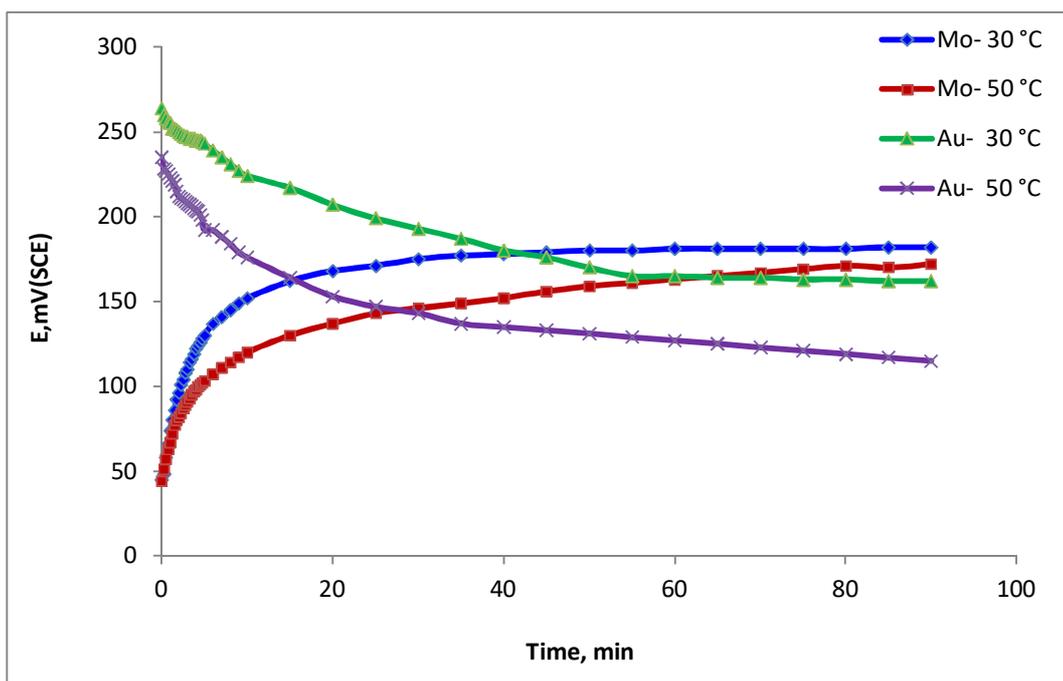


Figure 1: Potential – time curves of Mo and Au- alloy in lemon solution at 30 and 50 °C.

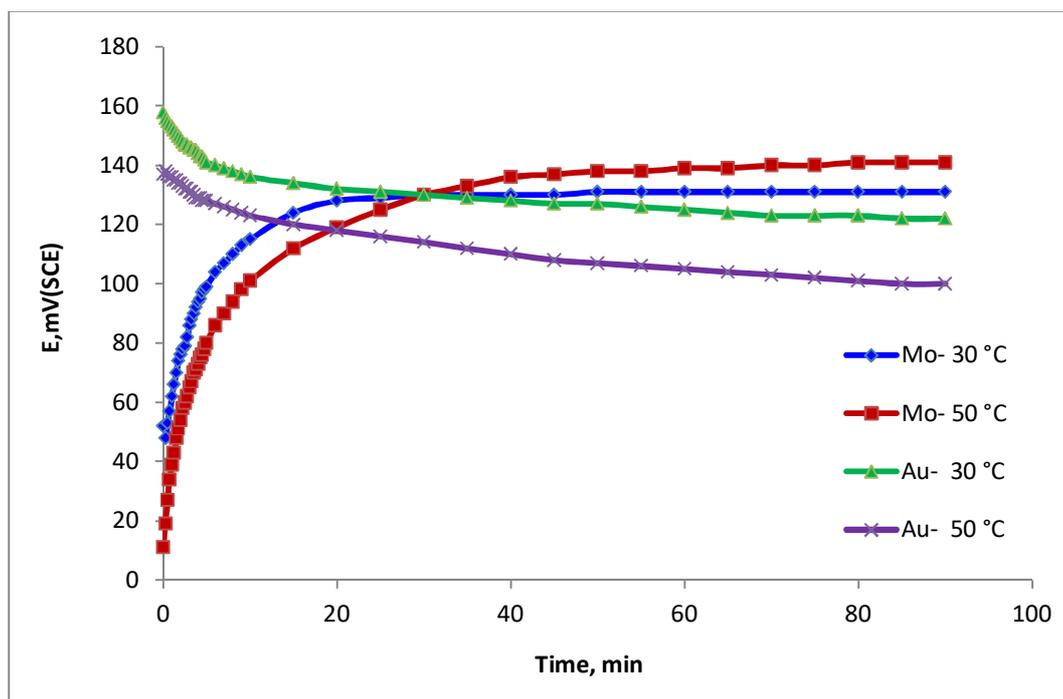


Figure 2: Potential – time curves of Mo and Au- alloy in vinegar solution at 30 and 50 °C.

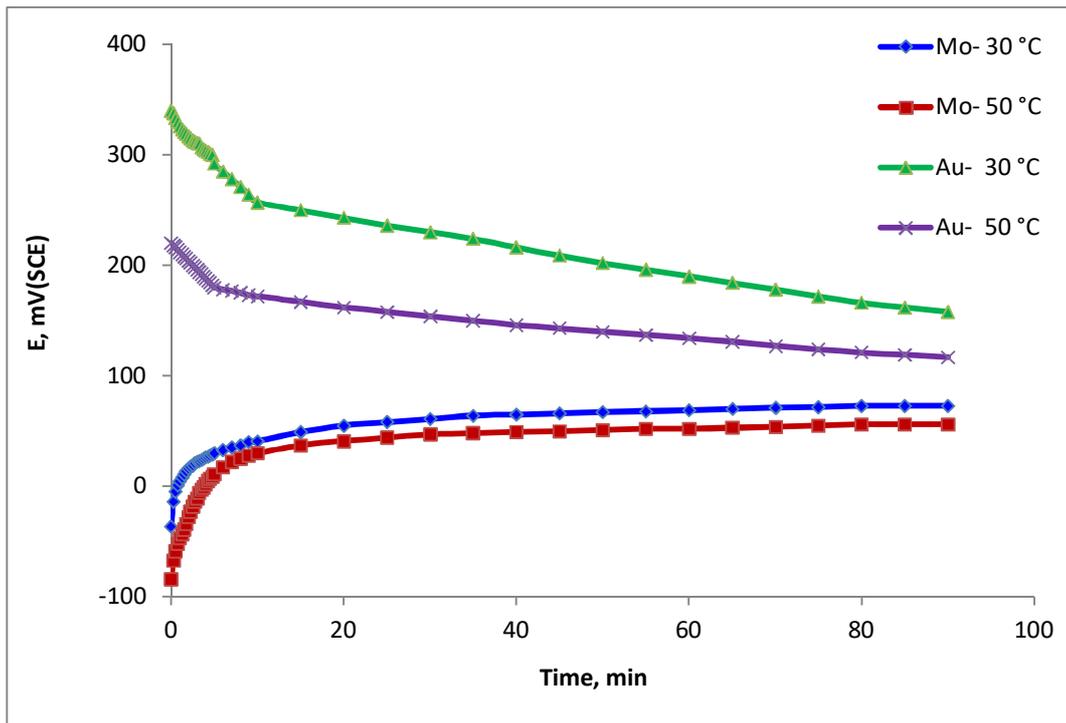


Figure 3: Potential – time curves of Mo and Au- alloy in tomato solution at 30 and 50 °C.

It is well-known that, gold metal is stable in air under normal conditions. It does not react with water or with aqueous acids or bases. The presence of alloying element (Ir and Ti) in small amount might be the reason behind the recorded reaction reactivity of the alloy illustrated by the first decrease in potential readings; as illustrated in Figures 1-3. However, the surface distribution of these elements presented on the Au- alloy substrate was not uniform and hence, some surface defects which stimulated corrosion of Au- alloy upon immersion in the test solutions were observed. Moreover, the potential differences among the alloying elements (act as cathodes) inside the Au matrix (acts as anode) resulted in building up micro-electrochemical cells and enhancing galvanic corrosion of the alloy [11].

Table 1: The immersion potential (E_{imm}) and the steady state potential (E_{ss}) for Au- alloy and Mo electrodes in the tested solutions at 30 and 50 °C

Temp.	Solution	Au- alloy		Mo	
		E_{imm}	E_{ss}	E_{imm}	E_{ss}
30 °C	Lemon	264	162	45	182
	Vinegar	158	122	11	141
	Tomato	340	158	-36	73
50 °C	Lemon	235	115	44	172
	Vinegar	137	100	52	131
	Tomato	220	117	-84	56

Molybdenum is a typical valve metal. Its anodic oxidation in aqueous solutions follows the familiar relationship [12]:

$$I_a = A \exp (BH) \dots\dots\dots(3)$$

Where, I_a is the imposed anodic current density, A and B are constants, and H is the effective field strength.

The quantity E/δ , may be considered instead of H, where E is the potential of the working electrode, and δ is the oxide film thickness. To keep the field strength (E/δ) constant, the positive increase in the electrode potential was parallel with increase in the oxide film thickness (Figures 1-3). The relation govern the electrode potential, |E|, and time for metals loaded with thin oxide films can be represented by:

$$|E| = cons. + 2.303 (\delta/\beta) \log t \dots\dots\dots(4)$$

Where δ represents the rate of oxide thickening per unit decade of time. The term β is identified as:

$$\beta = \frac{nF}{RT} \alpha \delta' \dots\dots\dots(5)$$

Where α is the charge transference coefficient encountered in normal electrochemical processes ($0 < \alpha < 1$) and δ' is the height of the energy barrier surmounted during charge transfer [14].

Concerning of the $E - \log t$ curves; Figures (4 – 6) showed that, the potential was varied linearly with the logarithm of time at the first 5 minutes of immersion. It indicated that the oxide film thickness on Mo electrode followed a direct logarithmic law in all tested solutions. The stable rate for increase in the electrode potential is consistent with the increase in the rate of oxide thickening. This step, which preceded the steady state potentials, symbolized the formation of the high corrosion resistance films of the metal. The value of α is taken as 0.5, while δ^0 as 1 nm and $n = 6$ for Mo electrode (according to reaction 2). Accordingly, β acquires the values of 114.9 and 107.8 for Mo electrode at 30 and 50 °C, respectively. The slopes of the E vs. $\log t$ plots for Mo and Au- alloy with their regression coefficients (R^2), and the computed values of film thickening (δ), for Mo electrode in the two studied temperatures are listed in Table (2).

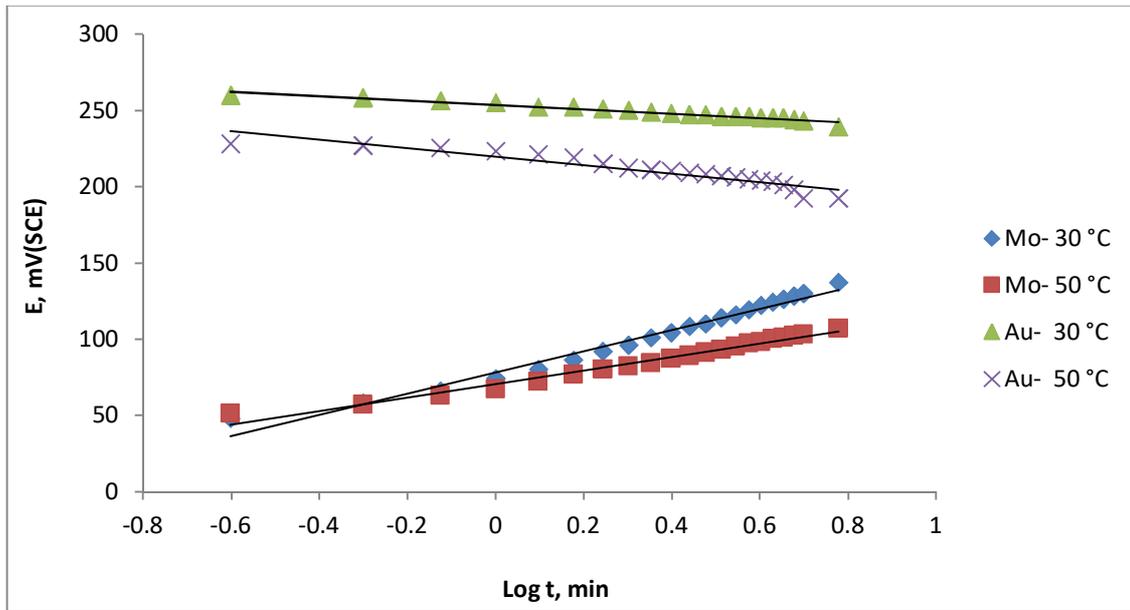


Figure 4: $E - \log t$ plot of Mo and Au- alloy in lemon solutions at 30 and 50 °C.

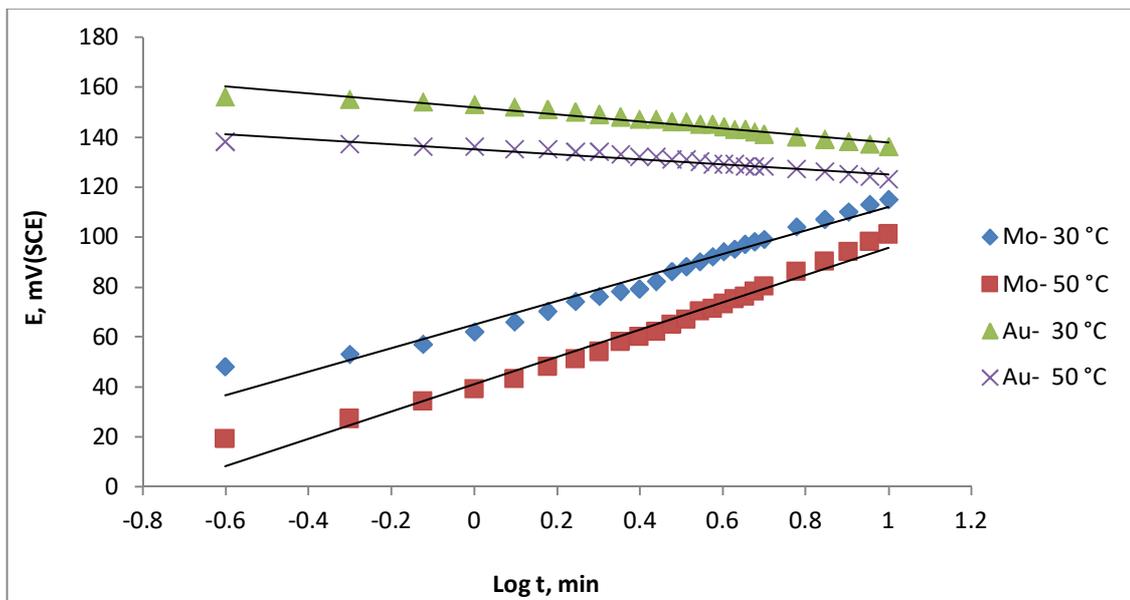


Figure 5: $E - \log t$ plot of Mo and Au- alloy in vinegar solutions at 30 and 50 °C.

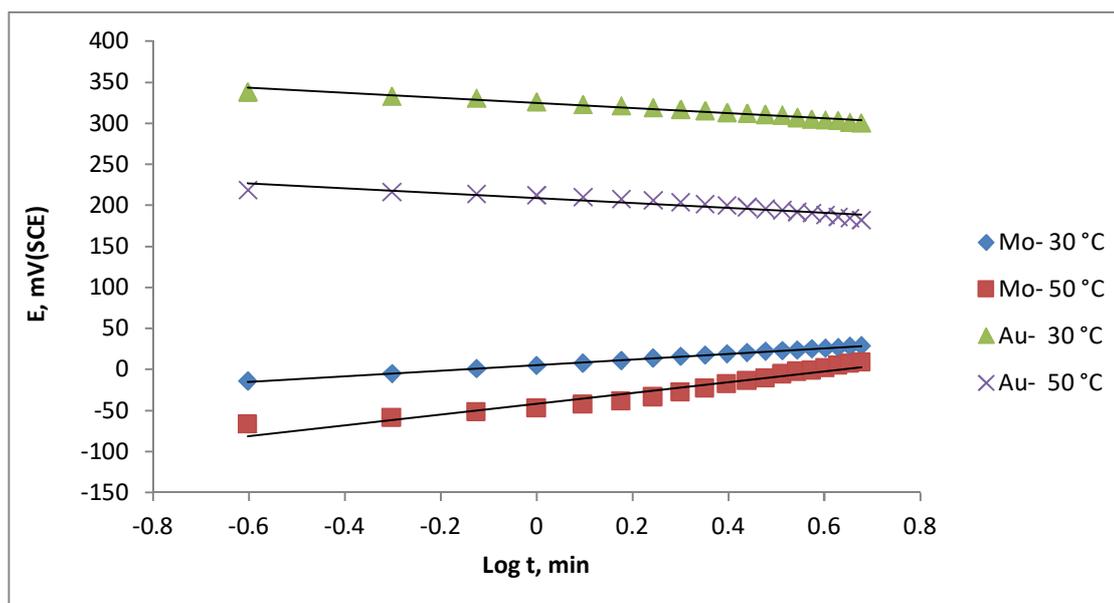


Figure 6: E – log t plot of Mo and Au- alloy in tomato solutions at 30 and 50 °C.

Table 2: Slopes of the straight lines, values of film thickening (δ) in nm/log t and regression coefficient R^2 for Au- alloy and Mo electrodes at 30 and 50 °C

Temp.	Solution	Au – alloy			Mo		
		slope	δ	R^2	slope	δ	R^2
30 °C	Lemon	-15.48	-	0.9180	68.54	3420	0.9764
	Vinegar	-13.31	-	0.9310	49.74	2482	0.9779
	Tomato	-30.40	-	0.9620	33.98	1696	0.9985
50 °C	Lemon	-25.06	-	0.9200	43.78	2049	0.9784
	Vinegar	-8.68	-	0.8930	43.53	2037	0.9784
	Tomato	-24.74	-	0.8820	66.76	3125	0.9383

Table 2 illustrated that the slopes of the linear relations, had positive values for Mo electrode, indicated the progress of oxide film thickening with time. However, negative slope values were observed for Au- alloy in all tested solutions at both 30 and 50 °C. These negative values confirmed the initial dissolution of metal- alloy surface with increasing immersion time. The maximum rate of oxide film thickness was observed for Mo electrode in lemon solution at 30 °C. The value was higher than those calculated for vinegar and tomato solutions at the same temperature by ~ 1.4 and 2 folds, respectively. Also, the value was higher than that calculated for the same solution at 50 °C with higher than 1.6 fold. On the other hand, the minimum rate of oxide film thickness was observed for Mo electrode in tomato solution at 30 °C with the best regression coefficient of linearity for all conditions. This means that the passivity of surface depends on nature of electrode, composition and temperature of solution.

In a nutshell, from the above results of open circuit potential technique, Mo electrode showed a comparable behavior with Au- alloy which might be an avenue to replace the expensive Au metal in dental applications.

Potentiodynamic polarization measurements

Potentiodynamic scanning (PDS) plots for the Au- alloy and Mo electrodes in the studied solutions are shown in Figures (7-9). The PDS profiles of each electrode were nearly similar in different solutions. The two electrodes showed typical Tafel behavior in all solutions. The respective potentiodynamic parameters including corrosion potential (E_{corr}), corrosion current density (I_{corr}), corrosion rate (C_R), anodic (β_a) and cathodic (β_c) Tafel slopes for Au- alloy and Mo electrodes are tabulated in Tables 3 and 4, respectively.

From Figure 7, in lemon solutions, it could be observed that the anodic and cathodic polarization curves of Mo electrode provided the higher current density values at 30 and 50 °C than those of Au- alloy electrode. Moreover, the cathodic branches of Mo electrode, at 30 and 50 °C, coincided together. On the other hand, the anodic and cathodic polarization curves of Au- alloy electrode at 50 °C are higher than its polarization curves at 30 °C. However, the anodic branches of the Au- alloy showed a higher deviation in the current density readings than in the cathodic ones. This confirmed the higher resistance of Au- alloy to corrosion under polarization conditions.

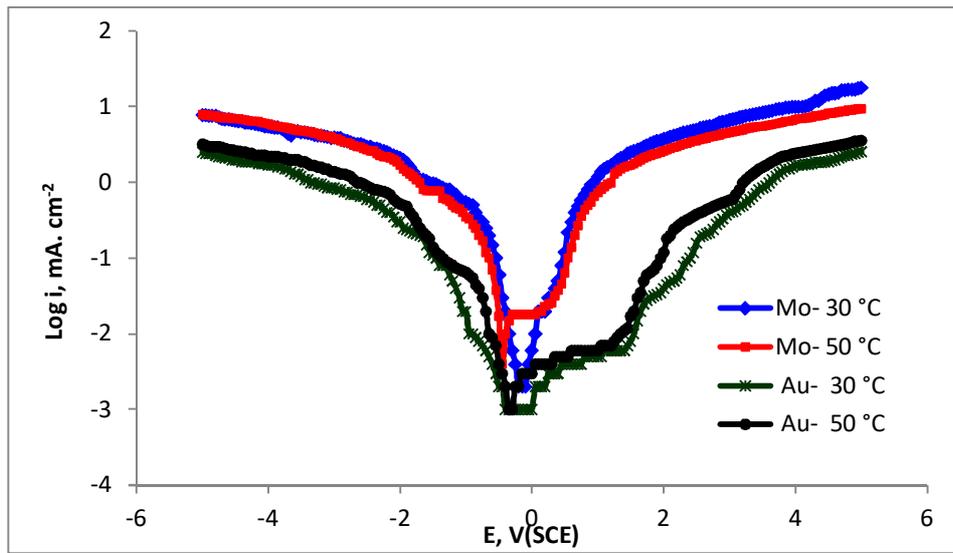


Figure 7: Potentiodynamic plot of Mo and Au- alloy electrodes in lemon solutions at 30 and 50 °C.

In vinegar solutions (Figure 8), the behavior of the electrodes was different. Each anodic and cathodic polarization branches had two parts, and the whole plot might appear as interior part (from -1.7 to +2.7 V/SCE) and outer part which separated in the anodic branch from 2.7 V/SCE to the end of the curves at 5 V/SCE and separated in the cathodic branch from -1.7 V/SCE to the end of the curves at -5 V/SCE. In the interior part, the high current density was observed for Mo at 50 °C then at 30 °C followed by Au- alloy at 50 then at 30 °C. The outer parts changed its order where Au- alloy had the highest current density at 50 °C followed by Au- alloy at 30 °C, then Mo at 50 °C. The lowest current density was recorded for Mo at 30 °C. This order was observed in both anodic and cathodic branches. These results revealed that Mo after certain polarization conditions was more passive than Au- alloy.

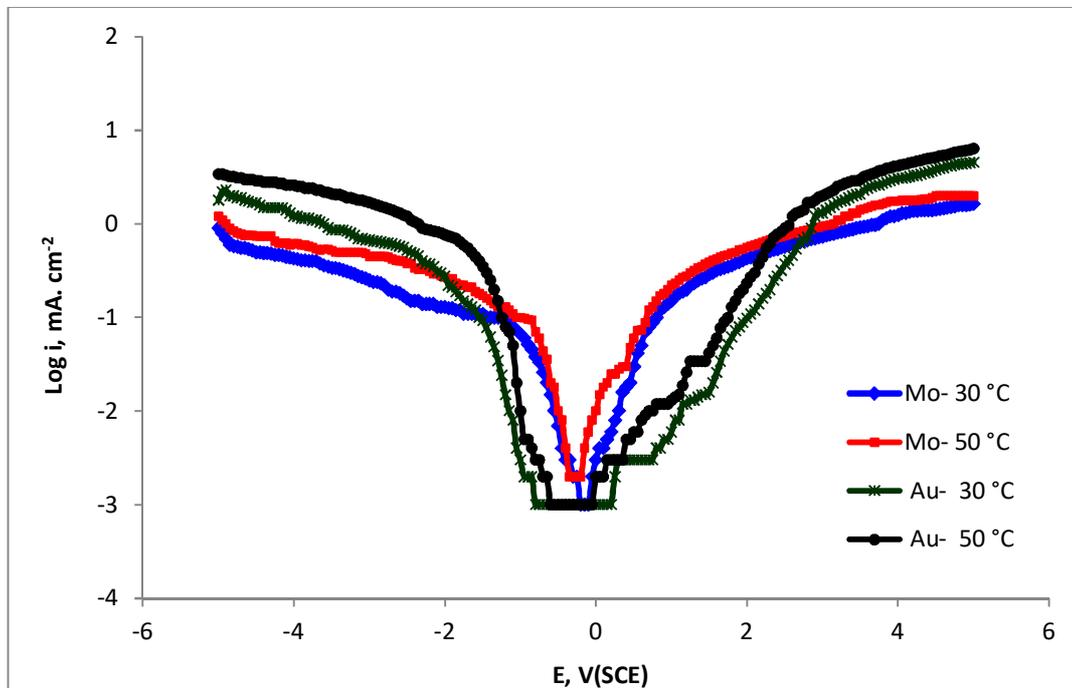


Figure 8: Potentiodynamic plot of Mo and Au- alloy electrodes in vinegar solutions at 30 and 50 °C.

In tomato solutions (Figure 9), Mo had the highest current density at 50 °C followed by at 30 °C in both anodic and cathodic branches. The behavior of Au- alloy was similar to that in lemon solutions.

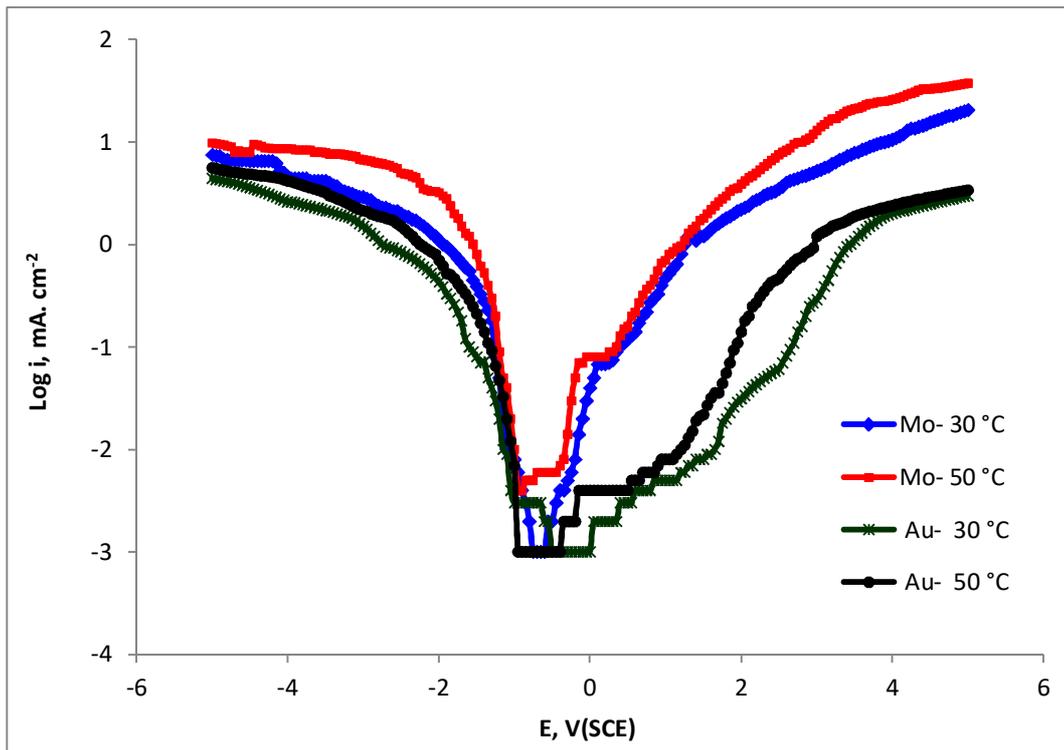


Figure 9: Potentiodynamic plot of Mo and Au-alloy electrodes in tomato solutions at 30 and 50 °C.

Table 3: Potentiodynamic parameters of Au-alloy electrode in lemon, vinegar and tomato solutions at 30 and 50 °C

Temp.	Solution	E_{corr} mV/SCE	I_{corr} mA/cm ²	β_c mV/dec	β_a mV/dec	C_R mpy
30 °C	Lemon	-100	0.0446	-2000	2500	4.57E-06
	vinegar	-150	0.0631	-2000	2500	6.47E-06
	tomato	-250	0.0700	-1600	1750	7.18E-06
50 °C	Lemon	-200	0.0790	-2000	3000	8.10E-06
	vinegar	-250	0.1000	-2000	2333	1.02E-05
	tomato	+150	0.0794	-1833	2250	8.15E-06

Table 4: Potentiodynamic parameters of Mo electrode in lemon, vinegar and tomato solutions at 30 and 50 °C

Temp.	Solution	E_{corr} mV/SCE	I_{corr} mA/cm ²	β_c mV/dec	β_a mV/dec	C_R mpy
30 °C	Lemon	-100	0.100	-1000	1000	2.02E-02
	vinegar	-200	0.016	-1250	1125	3.22E-03
	tomato	-600	0.063	-1100	1375	1.28E-02
50 °C	Lemon	-300	0.251	-2000	2000	5.08E-02
	vinegar	-300	0.050	-2200	2000	1.01E-02
	tomato	-700	0.126	-900	1500	2.55E-02

The corrosion potential (E_{corr}) of the Au-alloy electrode was constantly either equal or less negative than that of Mo electrode in all cases as seen from Figures (7-9) and Tables (3 & 4). For Mo electrode, all corrosion potentials in tested solutions at 30°C were less negative than those in its corresponding solutions at 50°C. This was also the case for Au-alloy, except in tomato solutions in which the behavior was reversed.

Table 3 illustrates that Au-alloy electrode had the highest corrosion rate in vinegar solution at 50 °C. The lowest corrosion rate was recorded for Au-alloy in lemon solution at 30 °C. Mo electrode has its highest corrosion rate in lemon solution at 50 °C (Table 4). The lowest corrosion rate for Mo electrode was recorded at 30 °C in vinegar solution. The corrosion rates were always higher for the two electrodes at the higher temperature in all solutions.

It could be concluded that the corrosion rates of Mo (in the two studied temperatures) decreased in the following order:

Lemon > tomato > vinegar

This may be due to the fact that lemon juice is more acidic (pH 2.0) than vinegar (pH 2.2) and tomato juice (pH 4.5). For Au-alloy, the corrosion rates decreased in the following order:

At 30 °C: Tomato > vinegar > lemon

At 50 °C: Vinegar > tomato > lemon

The observed differences in the behavior for the two electrodes could be attributed to different properties of the corrosion products as well as the pH- different effect. These observations illustrated that the anodic and cathodic current densities were influenced by the nature of solution and the type of electrode under certain temperature. In spite of these results which indicated that Mo had higher corrosion rates than those of Au- alloy in all conditions, these results were obtained under laboratory condition when Mo electrode was subjected to electric potential which would not face in dental applications.

The lower corrosion rates recorded in vinegar solutions than those in tomato solutions for Au- alloy at 30 °C and for Mo electrode at 30 and 50 °C could be owing to that vinegar is a solution consisting mainly of acetic acid (CH₃COOH) and water. Acetic acid abbreviated HAc is a short chain carboxylic acid, can give a significant contribution to the overall corrosion rate. The two main cathodic reactions in presence of vinegar solutions appeared to follow reactions 6 and 7 [15]:



Reactions 6 and 7 reflected overall reaction, and did not purport to indicate detailed mechanisms of the proton reduction. Moreover, for each electrode, it seemed established that HAc or Ac⁻ could inhibit the anodic partial reaction, particularly at lower temperature (30 °C). However, at the higher temperature (50 °C), HAc caused increased corrosion rates as illustrated in Tables 3 and 4.

On the other hand, a whole lemon contains about 3 grams of citric acid in addition to water, vitamins, minerals, and other sour flavoring molecules. However, malic and ascorbic acids are other contributors towards the chemical makeup of a lemon. Figure 10 illustrates the chemical structure of citric, malic and ascorbic acids. Malic acid presents in around 5% of the concentration of citric acid. Ascorbic acid or vitamin C levels in a lemon, at around 50 milligrams per 100 grams. The presence of the three different types of acids in the chemical structure of lemon might lead to the observed higher corrosion rates than in vinegar solution for Mo electrode at the two studied temperatures.

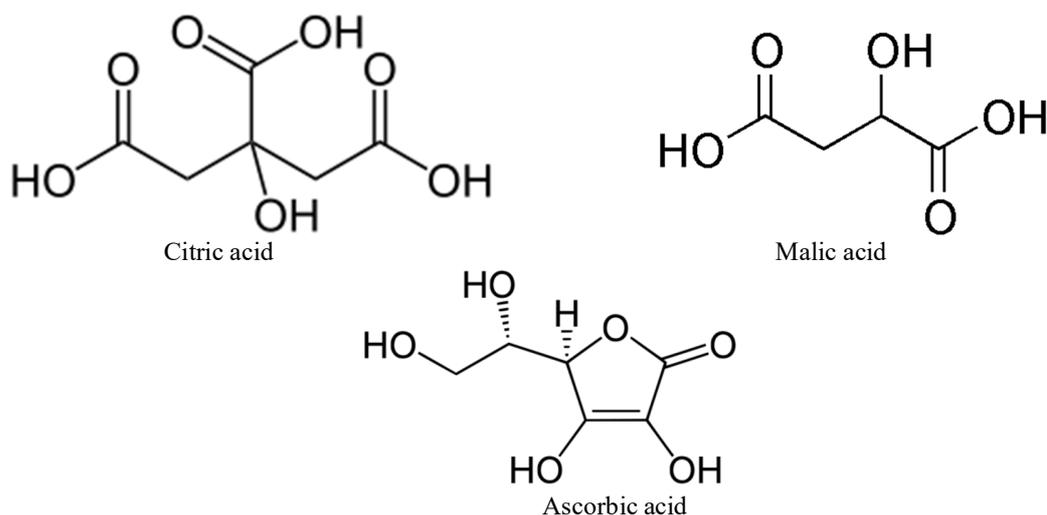


Figure 10: Chemical structure of citric, malic and ascorbic acid.

Tomato fruit is a rich source of ascorbic acid with an average of 25 mg/100 g [16]. Citric, malic, acetic, formic, trans-aconitic, lactic, fumaric, galacturonic, and α-oxo acids are also contributed to the structure of tomato fruit [17]. In 2010, Oluwole and Olawale [18] carried out an analysis for the tomato fluid and revealed the following composition: Ash (0.885%), Protein (1.91%), Lipid (0.77 %), Carbohydrate (4.58 %), Fibre (1.14 %) and Moisture (91.86%). Leaving the percentage of moisture aside, it could be observed that carbohydrate had the highest percentage of all other components.

As all the electrolytes used in this study are oxygen-containing solutions, the cathodic current recorded in the polarization branches can be attributed to the reduction of oxygen dissolved in the solution illustrated in equation 8 [19,20].



As the tomato solution has a great number of different types of compounds, they might work together to make tomato the first corrosive solution for Au- alloy at 30 °C and the second corrosive solution for both Mo (at 30 and 50 °C) and Au- alloy (at 50 °C).

However, at 50 °C, lemon and vinegar are the most corrosive solutions for Mo and Au- alloy respectively. The organic acids constituting lemon and vinegar may undergo higher dissociation at 50 °C, forming more aggressive ions that can cause higher corrosion rates than might otherwise be expected. These results indicated that the difference in temperature and the type of attack are the key parameters to understand the different observations.

CONCLUSION

The above mentioned results of open circuit potential and potentiodynamic polarization measurements illustrate that under certain condition Mo electrode might have protection properties better than Au- alloy. This might be an avenue for Mo electrode usage in dental applications rather than Au- alloy electrode which has a very high cost.

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