# Negative Effect of Laser Treatment with Nd:YAG on the Corrosion of Amalgam in Artificial Saliva

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## Abstract

This work involves study the corrosion behavior of amalgam in artificial saliva at pH=3 and  $\forall \forall \circ C$  after using Nd: YAG, where this laser used for healing the cancer. This study indicates that laser surface treatment affecting corrosion resistance of amalgam, and the results indicate that the laser treatment don't give the desired purpose because the nature of amalgam and the presence of mercury in this alloy which may be evaporate and it give toxicity for human and the energy of treatment must be low to avoid evaporation. In this work, laser treatment with two pulses under experimental conditions gave little improvement in corrosion resistance of amalgam compared with one and three pulses through the corrosion parameters of polarization behavior.

Key word: Amalgam, Laser treatment, Corrosion.

## Introduction

Hitherto, the possible danger from the internal source of corrosion products, dental amalgam fillings, has been mostly overlooked. Presently, this problem is devoted special attention [1]. Though a proof of amalgam biocompatibility has never been presented, amalgam is still by far the most extensively used material for dental restorations [1]. One of the possible reasons may be the very interdisciplinary character of the problem. For correct answer, specialist competence in the following fields is required: materials science, corrosion/electrochemistry, toxicology, medicine/diagnostics, physical biology, analytical chemistry. The common type of dental amalgam is an alloy containing typically in weight-%;  $\circ$  Hg,  $\uparrow \circ$  Ag,  $1 \cdot$  Sn, Cu, Zn.

Reported types of amalgam degradation are crevice corrosion,  $[, \xi, \circ]$  selective corrosion, [] galvanic corrosion in contact with dissimilar alloys  $[, \wedge]$  and mechanical wear []. Besides selective attack, stress corrosion has been proposed to be responsible for the marginal breakdown of amalgam restorations  $[, \cdot]$ . Further, cyclic loading, simulating chewing, strongly promotes corrosion of the amalgam surface [, 1, 1, 1].

There are many authors studied the behavior of dental alloys in artificial saliva to measure the resistance of these alloys toward many materials. Chang et al.  $[1^{\circ}]$  studied electrochemical behavior on microbiology related corrosion of

metallic dental materials in the presence of streptococcus mutants. While Latifa Kinani et al. studied effect of eugenol (occurs widely as a component of essential oils and is a major constituent of clove oil) on the titanium corrosion in artificial saliva enriched with eugenol at different concentration by utilizing electrochemical measurement and scanning electron microscopy [ $1\xi$ ].

Rajendran et al. were studied the corrosion behavior of mild steel (MS), zinc coated mild steel (MS-Zn) and stainless steel <sup>mill</sup>L (SS) in artificial saliva at pH=<sup>1</sup>,<sup>o</sup> and <sup>mill</sup>C in presence of D-Glucose by utilizing potentiodynamic polarization study and AC impedance spectra. The results of this study indicated that in the presence D-Glucose all three alloys showed better corrosion resistance than in the absence D-Glucose because in presence D-Glucose will form metal (Ni-Cr-Fe)-glucose complex on metal surface, it is compact, close packed and stable [<sup>1</sup>o]. In another study, Rajendran et al. studied corrosion behavior of same materials in the presence of spirulina powder (a tiny aquatic plant has been eaten by human since prehistoric times) using the same technique and they showed that the corrosion resistance was SS<sup>mill</sup>L > MS > MS-Zn [<sup>1</sup>].

The aim of this work is an attempting to improve the corrosion resistance of amalgam in artificial saliva at pH=3 and  $\gamma\gamma\circ C$  by leaser treatment.

#### **Materials and Procedure**

## Preparation of Samples and Electrolyte

The used alloy in this study was amalgam; (Ag  $\circ, \forall wt\%$ , Sn  $\uparrow, \forall wt\%$ , Cu  $\uparrow \xi, \forall wt\%$ ) which prepared by amalgamator. And then cold mounted using pyrax polymers to obtain only surface area.

The open side was polished mechanically to a mirror finish, rinsed in distilled water and stored in desiccators. The electrolyte reference used was modified Fusayama artificial saliva [ $\gamma\gamma$ ], which closely resembles natural saliva, with composition of ( $\cdot, \xi$  g/L KCl,  $\cdot, \xi$ g/L NaCl,  $\cdot, \gamma \cdot \gamma$  g/L CaCl<sub>x</sub>.  $\gamma$ H<sub>x</sub>O,  $\cdot, \gamma \cdot \gamma$  g/L NaH<sub>x</sub>PO $_{\xi}$ .  $\gamma$ H<sub>x</sub>O,  $\cdot, \gamma \cdot \gamma$  g/L Na<sub>x</sub>S.  $\gamma$ H<sub>x</sub>O and  $\gamma$ g/L urea).

## Corrosion Test:

Polarization experiments were performed in WINKING M Lab  $\forall \cdots$ Potentiostat/Galvanostat from Bank-Elektronik with electrochemical standard cell with provision for working electrode (amalgam), auxiliary electrode (Pt electrode), and a Luggin capillary for connection with saturated calomel electrode SCE reference electrode. Electrochemical measurements were performed with a potentiostat by SCI electrochemical software at a scan rate %mV.sec<sup>-1</sup>.

The main results obtained were expressed in terms of the corrosion potentials ( $E_{corr}$ ) and corrosion current density ( $i_{corr}$ ) in addition to measure the Tafel slops by Tafel extrapolation method.

## Laser Surface Modification (LSM)

The laser that used in this work is Nd:YAG laser with an output of second harmonic generation of 1.12 nm, laser energy of 1.1 mJ, 7.1 mJ, 7.1 mJ, and ... mm spot size. One advantages of laser surface modification LSM was the obtainment of smooth surface for treated alloy.

#### **Results and Discussion**

#### Surface Characterization

The microstructures of specimens of amalgam as shown in Fig. (1) for untreated and LSM treated specimens. They indicate laser spots in the specimens.

LSM treatment has effect on surface of specimens due to laser melting and rapid cooling, while the bulk of material (which known as saturated solid solution) remained unaffected.

#### **Open Circuit Potential Measurements (OCP)**

The electrical potential at the metal electrolyte interface is strongly dependent on the nature and the concentration of the electrolyte, pH and surface conditions. As a result, the electrochemical reactions at this interface vary with time.

The OCP-Time was measured with respect to SCE for  $\cdot$  minute with scan rate  $\cdot mV$  in aerated electrolyte of artificial saliva at pH=7 and  $\forall \forall \pm \uparrow \circ C$  for amalgam.

Fig.( $^{\gamma}$ ) shows OCP-time measurement for untreated and treated amalgam in artificial saliva. OCP-time measurement for untreated amalgam shows gradual rising in potential magnitudes for all duration test to reach - $^{q}$ . mV. The potential increase shows that the alloys become thermodynamically more stable with time. While the open circuit potential (E<sub>oc</sub>) for treated amalgam decreases due to gradually breakdown of passive layer and gradually decreased in potential magnitudes because the passive layer don't cover alloy surface completely, and the laser treatment affecting the phases in amalgam especially mercury. But generally the E<sub>oc</sub> were nobler than for untreated specimen.

#### Linear Polarization

This test is a simple method for evaluating the corrosion rate, sufficiently sensitive to measure even a low corrosion rate  $[\A]$ , the test was conducted at a scanning rate of  $\cdot$ ,  $\circ$ mA/sec. The current densities were calculated with reference to the samples geometrical area.

During anodic or cathodic polarization, or at open circuit, ion transfer reaction of metal ions and oxygen ions will take place. A clear process is the anodic corrosion of a passive metal in the steady state. In such a case, metal ions travel through the oxide film with a constant rate and are transferred in an ion transfer reaction at the interface oxide/electrolyte. The rate of that passive film

dissolution depends on the local potential drop at the interface, the pH and the activity of the metal ions at the oxide surface [19].

The polarizations test started at a cathodic potential in relation to the corrosion potential, therefore the surface passive film was at least partially removed due to the highly reducing initial potentials as shown in Fig. ( $^{\Upsilon}$ ) which illustrates the linear polarization for untreated three dental alloys in oral environments (artificial saliva) only.

After immersion in the electrolyte for  $\cdot \cdot$  min., a linear polarization test was carried out for the specimens in artificial saliva to determine the values of polarization data. The dissolution of amalgam take place in anodic region (upper section of curve).

While the lower section represents reduction reaction which includes evolution of hydrogen molecules because of the acidity of electrolyte (artificial saliva) as follows:

$$\mathcal{V}H^+ + \mathcal{V}e \rightarrow H_{\mathcal{V}}\dots\dots(\mathcal{V})$$

In addition to reduction of oxygen as follows [?:]:

 $O_{\mathsf{r}} + \mathfrak{E}_{\mathsf{e}} + \mathfrak{E}_{\mathsf{H}^+} \rightarrow \mathfrak{H}_{\mathsf{r}}O \dots (\mathfrak{r})$ 

The corrosion current densities  $(i_{corr})$  were obtained from the polarization curves by extrapolation of the cathodic branch of the curves to the corrosion potential  $E_{corr}$ . The average values of corrosion parameters are shown in Table(1).

These data indicate that laser treatment of the amalgam with one and three pulses shift  $E_{corr}$  toward noble direction, while laser treatment with two pulses shifts  $E_{corr}$  toward active direction. The corrosion current densities values ( $i_{corr}$ ) shift to higher values for treatment with one and three pulses, but the treatment with two pulses shift ( $i_{corr}$ ) to lower value. It is known that any factor that enhances the value of ( $i_{corr}$ ) results in an enhanced value of the corrosion rate on pure kinetic ground. The rate ( $C_R$  mpy) of corrosion in a given environment is directly proportional with its corrosion current density ( $i_{corr}$ ) in accordance with the relation [ $\gamma$ ]:

$$C_R\left(\frac{mm}{y}\right) = 3.27 \frac{e}{\rho} i_{corr.}$$
 .....( $\gamma$ )

where  $C_R(mpy)$ : corrosion rate in mil per year, *e*: equivalent weight of alloy (gm),  $\rho$ : density of alloy (gm/cm<sup>r</sup>) and *i*<sub>corr</sub>: corrosion current density (mA.cm<sup>-r</sup>).

The laser treatment leads to increment in the surface hardness which was very little with laser treatment in comparison with untreated specimen. This increment can be due to the fact that laser radiation has caused a smoother surface. Generally, change in surface hardness shows a microstructure modification in metallic bonds. The comparison of linear polarization curves indicate a few important points which are as follows: the corrosion rate for laser treated specimen reduced for treated amalgam only with two pulses. This implies that treated specimen release hydrogen easier and act as an electron donor to electrolyte [<sup>YY</sup>].

Increase of corrosion resistance probably means that most inclusions at the surface have been dissolved in the structure due to melting or alternatively, they are covered by molten material.

Treatment with one and three pulses shows no significant changes in its behavior. An interesting feature is the systematic shift of the pitting potential in the active region with a laser treatment despite the formation of a semipassive film at lower current densities, i.e. corrosion is observed. This indicates the formation of unstable passive film that is subtle to severe pitting with laser treatment. This is observed by NasserAl-Aqeeli in his work about the corrosion behavior of electrodeposited and laser irradiated Ni-Co nanostructured  $alloy[\Upsilon^{r}]$ .

## Cyclic polarization

Cyclic polarization data were recorded by potentiostat with electrochemistry software. The polarization scan was initiated after immersing the specimen for  $\cdot$  minutes and scanning the potential in the more noble direction at the scan rate of mV/sec. When  $-\text{V} \cdot \text{mV}$  were reached, the scanning direction was reversed.

Potentiodynamic measurements were carried out in order to determine the initiation and propagation of local corrosion, which is associated with the breakdown of passive protective film.

The breakdown potential  $(E_{br})$  is the one at which the anodic current increases considerably with applied potential. The potential, at which the hysteresis loop is completed upon reverse polarization scan, is known as the protective potential or repassivation potential.

Breakdown potential is a sign of local corrosion but the measure of pitting susceptibility is the difference between the breakdown potential and the repassivation one. The protection potential represents the potential at the intersection of hysteresis curve with passive domain. Below this potential the propagation of existing localized corrosion will not occur. If the difference between breakdown and the repassivation potential is increasing, the chance in the appearance of pitting is greater and its propagation in depth is more intense. In other words, the hysteresis loop increases as the susceptibility of material to corrosion increases.

From Figure ( $^{r}$ ), it can be seen that the forward scan is the same of reverse scan, this type of cyclic polarization curve is known to resist localized corrosion [ $^{7} \epsilon$ ]. It is also observed that the reverse scan curves meet the forward scan curve along the passive range. The potential for the reverse scan curves are

more positive than that for the forward scan in the laser treatment with two pulses.

### Conclusion

The laser treatment always used to improve the corrosion resistance, but in this work can be conclude that the laser treatment for amalgam don't give the desired purpose because the nature of amalgam and the presence of mercury in this alloy which may be evaporate and it give toxicity for human and the energy of treatment must be low to avoid evaporation. In this work, laser treatment with two pulses under experimental conditions gave little improvement in corrosion resistance of amalgam compared with one and three pulses through the corrosion parameters of polarization behavior.

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Treatment	-E <sub>oc</sub> (mV)	-E <sub>corr</sub> (mV)	i <sub>corr</sub> (A.cm <sup>-</sup> )	-b <sub>c</sub> (mV.dec <sup>-</sup> ')	$b_a (mV.dec^-')$	$C_R$ (mm/y)
Without	9	987,0	۷۹۷,۸۲ <sub>X</sub> ۱۰ <sup>-۹</sup>	۱۰٦,۰	00,0	۲,٦٨ $_X$ ۱۰ <sup>-۲</sup>
One pulse	91.	۹٠٦,٧	1.,02. X17	۸٦,٣٠	۳۷,۸	٣,0٤X1'
Two pulse	927	937,5	۲00,9٤ <sub>X</sub> ۱۰ <sup>-9</sup>	۱۰۲,۱	01,7	۲,0٤ <sub>X</sub> ۱۰ <sup>-۲</sup>
Three pulse	۸٥٣	140,7	۱٤,٠٥٠ <sub>X</sub> ۱۰ <sup>-٦</sup>	٦٨,٢٠	٥٤,٨	٤,٧٢ <sub>X</sub> ١٠-٢

**Table** (1): Corrosion parameters of untreated and treated amalgam with leaser in artificial saliva at pH=3 and temperature  $^{m_{VO}}C$ .



Treated with Two pulse



Treated with one pulse



Treated with Three pulseUntreated amalgm alloyFig. ( $^{1}$ ): Opticalmicrograph foruntreated and treated amalgam alloy at  $\xi \cdot \cdot X$ <br/>magnification.





-1150

0.00001

0.0001

0.001

0.01

0.1

Fig. (<sup>r</sup>): Linear polarization of untreated and treated amalgam in artificial saliva at

(c)



Fig. (<sup>£</sup>): Cyclic polarization of untreated and treated amalgam in artificial saliva at pH= $^{1}$  and  $^{\psi \circ \circ}C$ .

# التأثير السلبي للمعاملة الليزربة بليزر Nd:YAG على تأكل الحشوة الزئبقية في اللعاب الاصطناعي م.فاطمه عربص سلطان الجامعه التكنولوجيه- قسم العلوم التطبيقية- فرع علوم الليزر.

الخلاصة

يهدف هذا البحث الى دراسة سلوك التأكل للحشوة الزئبقية بعد المعاملة الليزرية بليزر Nd:YAG في اللعاب الصناعي عند اس هيدروجيني ٦ ودرجة حرارة ٣٧ درجة مئوبة حيث يستخدم هذا الليزر لمعالجة الاورام في اللثة. ان الدراسة الحالية اثبتت بان المعاملة الليزرية تؤثر على مقاومة التأكل بسبب طبيعة الحشوة الزئبقية الحاوية على الزئبق والذي قد يتبخر معطياً تأثيراً سمى وإن الطاقة الليزرية يجب ان تكون اقل ما يمكن. ان المعاملة الليزرية في هذا البحث بضربتين ليزريتين اعطت تحسناً قليلاً في مقاومة التأكل مقارنة مع المعاملة الليزرية نبضة وإحدة وثلاث نبضات كما تبينه متغيرات التأكل المقاسة من سلوك الاستقطاب.