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Influence of Khat Extract (*Catha edulis*) on Corrosion Resistance of Co-Cr Dental Alloys

Influência do extrato de Khat (Catha edulis) na resistência à corrosão de ligas dentais Co-Cr

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ABSTRACT

Objective: The aim of this experimental study was to assess the effect of khat extract with different concentrations on the corrosion resistance of cobalt-chromium (Co-Cr) dental alloys used for removable denture. **Material and Methods:** The corrosion resistance of three Co-Cr alloys (Neobond II®, Kera 501® and PD Casta H®) was evaluated in artificial saliva in presence of three different concentrations of khat extracts. Fusayama-Meyer artificial saliva was used as a reference solution. The corrosion properties of the alloys were analyzed using potentiodynamic polarization, electrochemical impedance spectroscopy and surface analysis. The data was presented in means, standard deviations, and related figures. Comparison between the different concentrations was done using 1-way ANOVA test. The surface analysis was performed using scanning electron microscopy (SEM). **Results:** the results showed that the corrosion resistance of the three alloys tested (Neobond II®, Kera 501® and PD Casta H®) decreased in artificial saliva containing khat extract compared with that of the reference solution. Additionally, the corrosion resistance of the three Co-Cr dental alloys decreased by increasing the concentration of khat extract. Furthermore, the results indicate that Neobond II® alloy showed the least corrosion resistance compared with that of Kera 501® and PD Casta H® alloys. **Conclusion:** Khat extract with different concentrations had negative impact on the corrosion resistance of Cr-Co alloys. More in-vivo studies are highly recommended to confirm the results of the present study.

KEYWORDS

Khat; Catha edulis; Partial denture; Corrosion resistance.

RESUMO

Objetivo: O objetivo deste estudo experimental foi avaliar o efeito do extrato de khat em diferentes concentrações na resistência à corrosão de ligas dentárias de cobalto-cromo (Co-Cr) utilizadas em próteses removíveis. **Material e Métodos:** A resistência à corrosão de três ligas de Co-Cr (Neobond II®, Kera 501® e PD Casta H®) foi avaliada em saliva artificial na presença de três concentrações diferentes de extratos khat. A saliva artificial de Fusayama-Meyer foi usada como solução de referência. As propriedades de corrosão das ligas foram analisadas usando polarização potenciodinâmica, espectroscopia de impedância eletroquímica e análise de superfície. Os dados foram apresentados em médias, desvios-padrão e figuras relacionadas. A comparação entre as diferentes concentrações foi feita usando o teste One-way ANOVA. A análise de superfície foi realizada em microscopia eletrônica de varredura (MEV). **Resultados:** os resultados mostraram que a resistência à corrosão das três ligas testadas (Neobond II®, Kera 501® e PD Casta H®) diminuiu na saliva artificial contendo extrato khat em comparação com a solução de referência. Além disso, a resistência à corrosão das três ligas dentais Co-Cr diminuiu com o aumento da concentração de extrato de khat. Além disso, os resultados indicam que a liga Neobond II® apresentou a menor resistência à corrosão em comparação com as ligas Kera 501® e PD Casta H®. **Conclusão:** O extrato de Khat com diferentes concentrações teve impacto negativo na resistência à corrosão de ligas de Cr-Co. Mais estudos in vivo são altamente recomendados para confirmar os resultados do presente estudo.

PALAVRAS-CHAVE

Khat; Catha edulis; Prótese parcial; Resistência à corrosão.

INTRODUCTION

Khat (*Catha edulis*) is an evergreen shrub grown in South Arabia (mainly Yemen and South Saudi Arabia) and some East African countries such as Ethiopia, Somalia, Djibouti and Kenya [1,2]. Millions of people in these countries as well as in Europe and North America chew khat on daily basis for its stimulating effects [1-3]. Khat chewing is practiced as a regular daily habit (particularly in Yemen and some East African countries) and forms a basis of social interaction [1,2,4]. The habit involves inserting khat leaves into the mouth, chewing them and keeping the bolus in the buccal fold for several hours [3]. The main chemical components of Khat leaves are alkaloids, mainly cathine and cathinone, amphetamine-like sympathomimetic substances [3]. Other chemical components include amino acides, tannins, fluoride, and traces of vitamins [4,5]. Long term khat chewing has been linked with several systemic and oral complications [3]. Reported oral and dental effects of khat chewing include, periodontal diseases, dental attrition, mucosal white lesions, xerostomia, temporomandibular joint disorders, among others [6-11]. In addition to its effects on oral soft and hard tissues, Khat chewing has been reported to have negative effects on dental restorations and prostheses [12-14]. A 2017 cross-sectional survey investigated the pattern of restorative failure among khat chewers and shammah users in Jazan, Saudi Arabia, and found significant association between khat chewing and various restorative failures in amalgam, composite, crowns and removable prostheses [14]. A more recent experimental study reported that khat extract was associated with the highest effects on the color stability of different types of composite Resins compared to other tested materials [15]. Additionally, one study investigating the influence of khat extract on the corrosion of Nickel cobalt alloys found that khat extract accelerates the corrosion of Nickel cobalt alloys [13].

Cobalt–chromium-based alloys (Co-Cr alloy) are widely used for the fabrication of removable partial dentures and metal ceramic prostheses [16]. Co-Cr alloy exhibits high strength, good cytocompatebility and are non-magnetic [17,18]. Additionally, Co-Cr alloys have also been found to be more resistant to corrosion than Ni-Cr alloys [16,18]. These properties make Co-Cr ideal alloys to be used in the oral cavity [16]. Given the nature of khat chewing, and the fact that khat leaves are kept in the oral mucobuccal fold for several hours, khat quid and juice usually comes in contact with dental prosthesis, leading to dissolution of these materials. The prolonged contact may have a negative effect on dental alloys including Co-Cr alloys, and as stated above, khat was found to induce corrosion of Ni-Cr alloy. Unfortunately, the literature is scarce regarding the potential effects of khat on dental materials, and so far no studies have investigated the influence of khat (*Catha edulis*) on corrosion resistance of Co-Cr dental alloys. Therefore, the present study aimed to assess the influence of methanolic extract of khat with different concentrations (0.5, 1, and 2g/L) on corrosion resistance of Co-Cr dental alloys used for removable partial dentures electrochemically and under the scanning electron microscope (SEM). It was hypothesized that Khat extract with different concentrations has no effect on Co-Cr dental alloys in terms of corrosion resistance.

MATERIAL AND METHODS

Co-Cr Dental Alloys

Three Co-Cr dental alloys used for removable partial dentures (Neobond II®, Kera 501® and PD Casta H® alloys) were used as test materials in this study. The chemical composition of these alloys is shown in table I. Form each type, two samples were fabricated. For electrochemical measurements, the working electrodes were cylindrically shaped (7 mm in diameter and 15 mm length) and covered with a polytetrafluoroethylene (PTFE) coating. The area exposed to the solution was 0.38 cm^2. The working electrodes were mechanically polished with abrasive paper of different grades (400-1000-1500), washed in distilled water and then dried with ethanol before corrosion test.

Table I - The chemical composition of materials

Material	Composition (wt %)
Neobond II®	Co: 52.2 %, Cr: 27.8 %, Ga: 2.5%, Fe: 0.5%, Si: 0.5%, C: 0.4%, Divers: 12.4%
Kera 501®	Co: 61 %, Cr: 30.25%, Mo: 5.5%, Mn: 0.25%, Al: < 0.4%, Si: 0.4%, Fe: 0.5%, Nb: 1%
PD Casta H®	Co: 60 %, Cr: 29 %, Mo: 6.2%, Ni: 2%, Fe: 2%, C, Si, Mn: < 1%

Khat Extracts

Fresh khat leaves were collected in summer, weighed, washed with distilled water three times and left to dry for three days in a clean dry room (20 ± 5 °C) protected from sunlight. After drying, the plant was weighed, packed in a closed foil packet and stored at 20 °C until use.

Khat was extracted from leaves as described by previous studies [19, 20]. Briefly, dried khat leaves (100 g) were chopped into small pieces, homogenized in 100 mL of 95% ethanol, centrifuged at 5000 rpm for 5 min and the supernatant then filtered with Whatman filter paper (no. 1). Ethanol (100 mL) was added to the remaining leaves and the procedure repeated. The ethanol khat extract was concentrated using a rotary evaporator (BUCHI 461 Water Bath, Switzerland) at 30 °C with a rotation speed of 70 rpm until 70% of the ethanol solvent had evaporated. The resulting viscous solution was diluted with 100 mL of distilled water and then stirred at 1000 rpm with for 1 h at ambient temperature. The filtrate was kept frozen at -70 °C for 24 h and then dried by lyophilization (CHRIST, Alpha, 2-4 LD Plus). Typically, 100 g of dried leaves yielded 7 g of khat extract powder.

Test solution

The reference electrolyte was Fusayama-Meyer artificial saliva [21, 22]. The composition of this solution, which closely resembles natural saliva is: KCl (0.4 g/l), NaCl (0.4 g/l), $CaCl_2.2H_2O$ (0.906 g/l), $NaH_2PO4.2H_2O$ (0.690 g/l), $Na_2S.9H_2O$ (0.005 g/l), Urea (1 g/l). The pH was measured with a glass electrode (pH meter HANNA Instrument, France). The pH of this reference solution measured was 5.3. The second medium used had the same contents as the reference solution but enriched with khat extract with a concentration 0.5 g/l. The pH measured was 4.6. The third medium was identical to the reference solution but enriched with khat extract with a concentration 1 g/l. The pH measured was 4.5. The last medium used had the same contents as the reference solution but enriched with khat extract with a concentration 2 g/l. The pH measured was 4.5.

Electrochemical measurements

Potentiodynamic measurements and electrochemical impedance spectroscopy (EIS) were performed using a potentiostat (Voltalab® PGZ 301 radiometer analytical, France). The analyses were carried out in a three-electrode cell, using a saturated calomel electrode as reference electrode, platinum as counter-electrode and Co-Cr samples was used as working electrode, controlled by corrosion analysis software model (Voltamaster 4). The measurements were performed after the establishment of a reasonable steady state condition, which was safely achieved after 30 min of immersion. The polarization curves were plotted in the potential range of -1000 mV/SCE to +1000 mV/SCE at scanning rate of 0.2 mV/s. The EIS measurements were carried out in the frequency range of 100 kHz to 0.01 Hz at the open circuit potential, after 30 min of immersion, by applying a superimposed potential variation with 10 mV amplitude. The corrosion parameters are: corrosion potential (E_{corr}), corrosion current density (I_{corr}), polarization resistance (R_p), charge transfer resistance (R_{ct}), resistance of solution (R_s) and double layer capacitance (C_{dl}) [23]. The values of the corrosion parameters are the mean of two experiments with a maximum error of 10-15%.

Surface analysis

Two specimens of each material were observed using scanning electron microscopy (SEM) (Philips, Quanta 200 X. TM-© Fei Company). One specimen of each material was subjected to immersion for 15 day in artificial saliva containing khat extract (*Catha edulis*) and compared to analysis of initial state materials.

Statistical analysis

The software program SPSS V25 for Windows was used for data analysis. The data was presented in means, standard deviations, and related figures. Comparison between the different concentrations was done using 1-way ANOVA test. A P-value < 0.05 was considered significant.

RESULTS

Polarization curves

The polarization curves of the three alloys (Neobond II®, Kera 501® and PD Casta H®) in artificial saliva in the absence and presence of different concentrations of khat extracts are shown in Figure 1 (A-c). It can be observed that the corrosion current densities of the three alloys increased by increasing concentrations of khat extract. At artificial saliva containing khat extract with a concentration 2 g/l, Kera 501® showed an active dissolution peak at – 0.1V (SCE) and a high passive current density. Conversely, the polarization resistance of the three alloys decreased by increasing concentrations of khat extract.

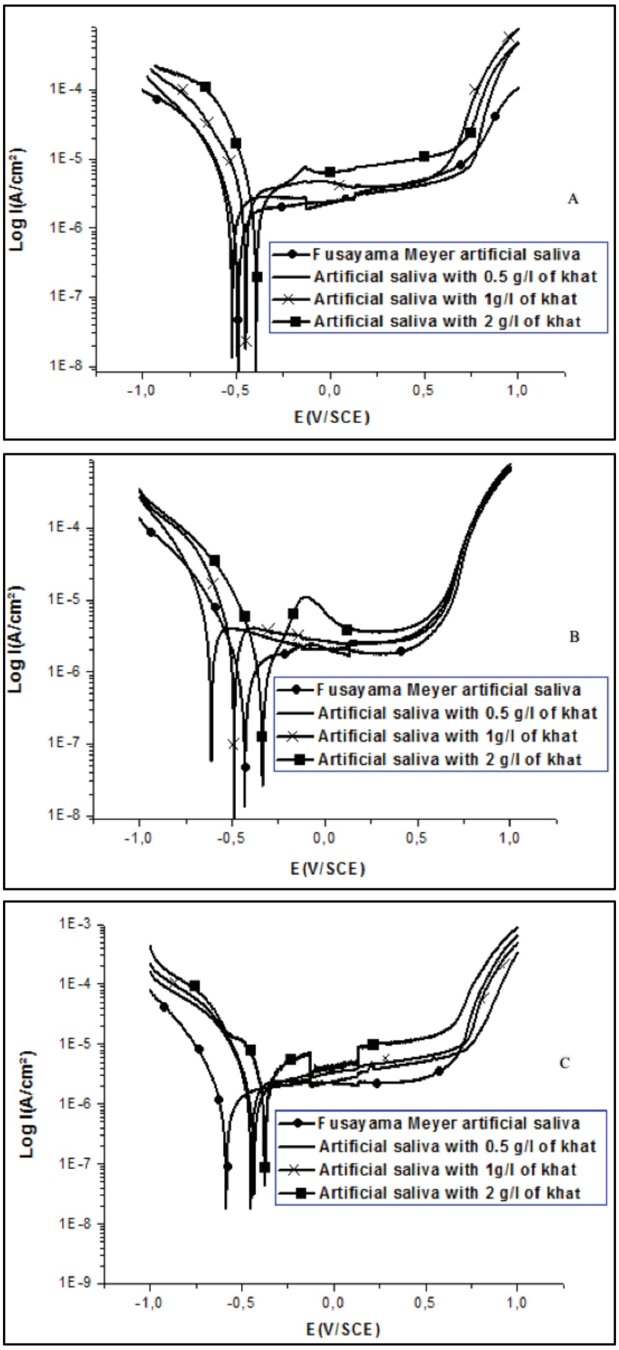

Figure 1 - The polarization curves of Neobond II® alloy (A), Kera 501® alloy (B), and PD Casta H® alloy (C) in artificial saliva in the absence and presence of different concentrations of khat extracts.

Similar polarization curves were obtained for both Kera 501® and PD Casta H® alloys in Fusayama-Meyer artificial saliva, and the polarization resistance values obtained for Kera 501® and PD Casta H® alloys were also of the same order. The corrosion current densities obtained for Neobond II® alloy in Fusayama-Meyer artificial saliva was high compared with that of Kera 501® and PD Casta H® alloys. More details are presented in Table II.

Table II - Corrosion parameters of the three dental alloys (Neobond II®, Kera 501® and PD Casta H®) obtained from potentiodynamic polarization in artificial saliva in the absence and presence of different concentrations of khat extract

	E_{corr} (mV/SCE)	I_{corr} (µA cm^{-2})	R_p (kΩ cm²)
Neobond II®			
Fusayama-Meyer artificial saliva	- 485.0±8.7	5.6±0.3	12.2±0.5
Saliva with 0.5 g/l of khat extract	- 521.0±6.5	10.5±0.4	7.6±0.4
Saliva with 1 g/l of khat extract	- 406.0±8.0	13.8±0.6	5.4±0.3
Saliva with 2 g/l of khat extract	- 446.5±8.3	16.8±0.5	3.6±0.4
ANOVA test	P= 0.001	P< 0.001	P< 0.001
Kera 501®			
Fusayama-Meyer artificial saliva	- 458.4±21.9	2.0±0.2	14.2±0.3
Saliva with 0.5 g/l of khat extract	- 593.7±23.5	9.9±0.3	9.1±0.3
Saliva with 1 g/l of khat extract	- 507.9±14.6	12.4±0.5	6.3±0.2
Saliva with 2 g/l of khat extract	- 354.8±13.8	15.8±0.4	3.9±0.2
ANOVA test	P= 0.001	P< 0.001	P< 0.001
PD Casta H®			
Fusayama-Meyer artificial saliva	- 541.6±18.7	1.8±0.2	14.8±0.6
Saliva with 0.5 g/l of khat extract	- 467.0±12.5	8.0±0.9	9.3±0.2
Saliva with 1 g/l of khat extract	- 447.4±9.8	11.4±0.3	7.1±0.2
Saliva with 2 g/l of khat extract	- 381.0±4.0	14.9±0.7	4.3±0.1
ANOVA test	P= 0.001	P< 0.001	P< 0.001

Electrochemical impedance spectroscopy

Figure 2 (A-C) shows the the Nyquist diagrams of Neobond II®, Kera 501® and PD Casta H® alloys in artificial saliva in the absence and presence of different concentrations of khat extracts. The plots of the three alloys are open semicircle and the diameter of the open semicircle decreases by increasing the concentration of khat extract. Diagrams can be represented by an equivalent circuit consisting of a double layer capacitance and charge transfer resistance in parallel (Figure 3). The values of charge transfer (R_{ct}), double layer capacitance (C_{dl}) and solution resistance (R_{sol}) of Neobond II®, Kera 501® and PD Casta H® alloys are reported in Table III. From the results of Nyquist diagrams, the Neobond II®, Kera 501® and PD Casta H® alloys showed low charge transfer resistance in artificial saliva containing different concentrations of khat extracts compared with that of reference solution. Additionally, the charge-transfer resistance (R_{ct}) decreases by increasing the concentration of khat extract. In Fusayama-Meyer artificial saliva, the Neobond II® alloy has low charge transfer resistance compared with that of Kera 501® and PD Casta H® alloys. The values of double layer capacitance (C_{dl}) of the Neobond II®, Kera 501® and PD Casta H® alloys increases by increasing concentration of khat extract compared with that of reference solution.

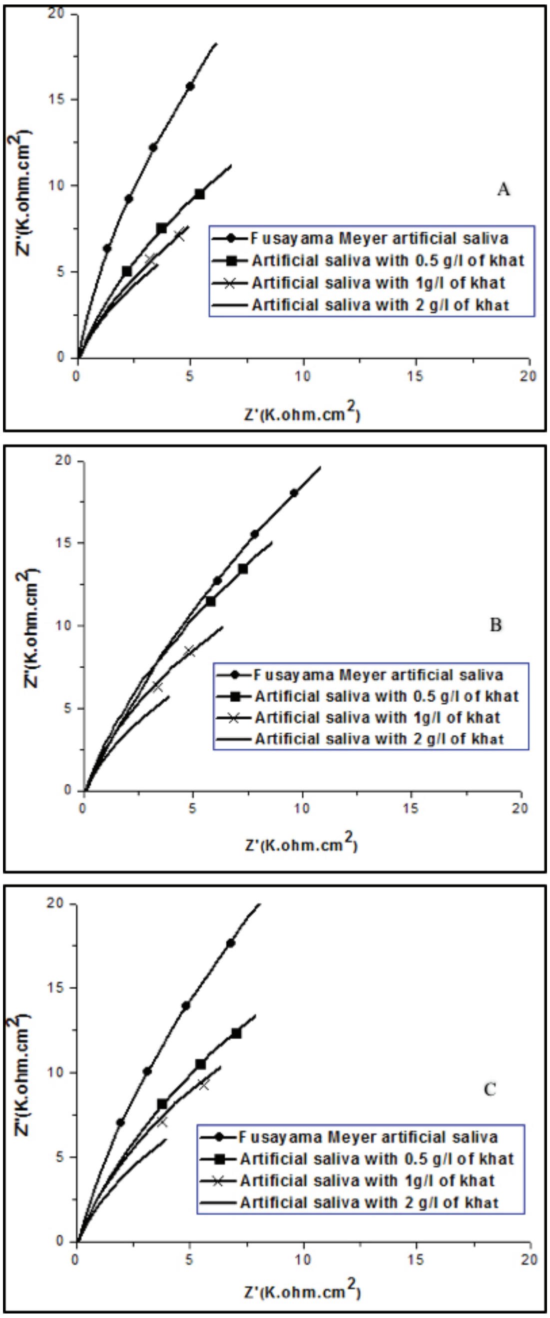

Figure 2 - Nyquist complex impedance plot for Neobond II® alloy (A), Kera 501 alloy (B), PD Casta H® alloy (C) in artificial saliva in the absence and presence of different concentrations of khat extracts.

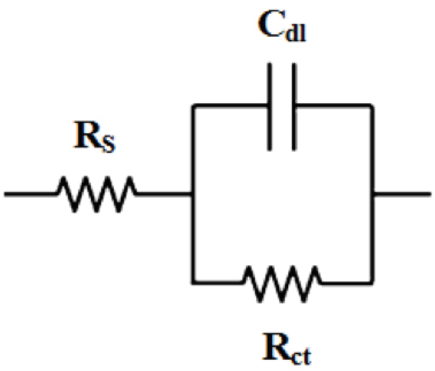

Figure 3 - Equivalent circuit, where R_s is the solution resistance, R_{ct} the charge transfer resistance and Cdl the double layer capacitance.

Table III - Corrosion parameters of the three alloys obtained from impedance measurements in artificial saliva in the absence and presence of different concentrations of khat extracts

	Rsol (Ω cm²)	Rct (kΩ cm²)	Cdl (μF cm⁻²)
Neobond II®			
Fusayama-Meyer artificial saliva	79.3±1.1	120.1±1.4	13.0±0.4
Saliva with 0.5 g/l of khat extract	75.4±1.3	48.7±1.5	22.8±0.5
Saliva with 1 g/l of khat extract	73.5±1.3	32.1±1.5	33.5±0.7
Saliva with 2 g/l of khat extract	75.3±1.0	23.5±1.0	47.8±0.5
ANOVA test	P= 0.033	P< 0.001	P< 0.001
Kera 501®			
Fusayama-Meyer artificial saliva	76.3±2.1	137.2±0.9	11.8±0.3
Saliva with 0.5 g/l of khat extract	72.4±1.3	68.1±1.1	22.1±0.8
Saliva with 1 g/l of khat extract	69.3±1.0	41.6±1.4	31.9±1.3
Saliva with 2 g/l of khat extract	67.3±0.9	25.0±1.8	44.5±1.1
ANOVA test	P= 0.011	P< 0.001	P< 0.001
PD Casta H®			
Fusayama-Meyer artificial saliva	140.8±1.4	156.9±1.4	9.9±0.8
Saliva with 0.5 g/l of khat extract	78.1±2.2	67.5±2.7	22.6±2.0
Saliva with 1 g/l of khat extract	69.6±0.4	44.5±0.6	34.5±1.7
Saliva with 2 g/l of khat extract	66.7±1.6	28.2±1.1	43.5±1.3
ANOVA test	P< 0.001	P< 0.001	P< 0.001

Surface analysis

Scanning electron microscopy (SEM) was used to analyze the surface of the Neobond II®, Kera 501® and PD Casta H® after immersion in artificial saliva containing khat extract for 15 days and compared to analysis of initial state materials. In the case Neobond II®, the comparison of SEM photomicrographs (Figure 4A and 4B) showed a uniform corrosion, which covered the surface of the alloy after immersion in artificial saliva containing khat extract. In the case Kera 501®, the comparison of SEM photomicrographs (Figure 5A and 5B) showed a pitting corrosion on the surface after immersion in artificial saliva containing khat extract. In the case PD Casta H®, the comparison of SEM photomicrographs (Figure 6A and 6B) showed slight localized corrosion after immersion in artificial saliva containing khat extract.

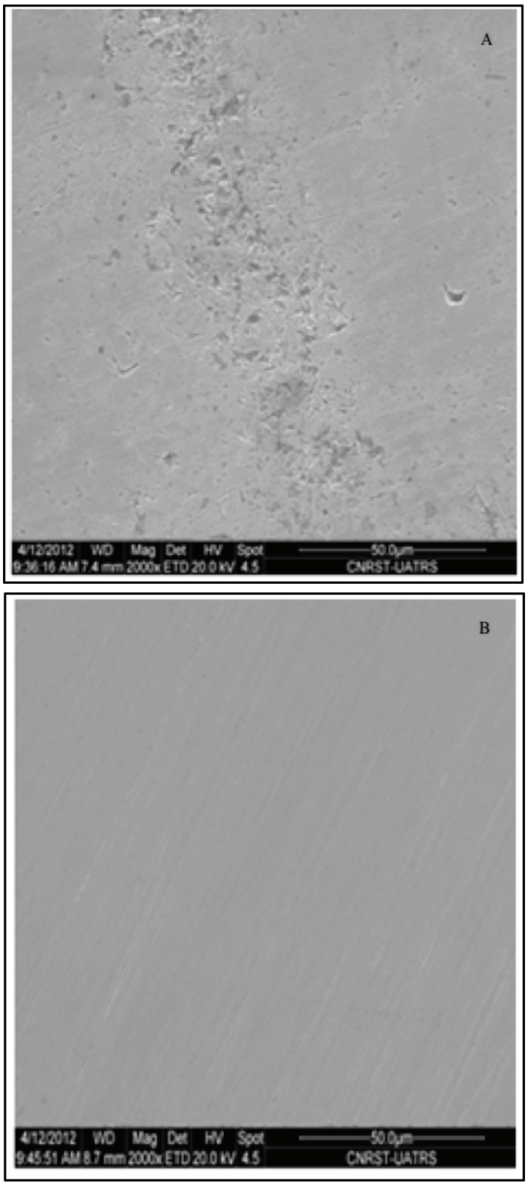

Figure 4 - The scanning electron microscopy of Neobond II® in artificial saliva containing khat extract (A) and in initial state (B).

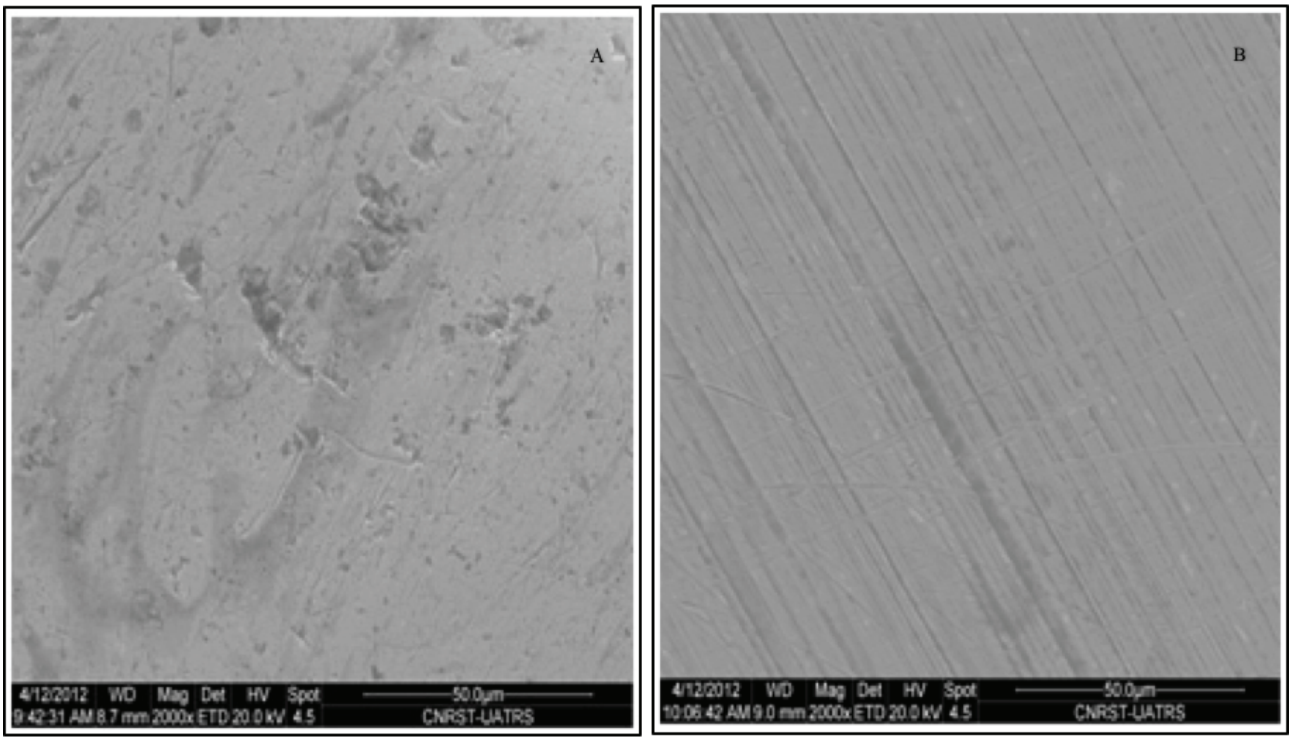

Figure 5 - The scanning electron microscopy of Kera 501® in artificial saliva containing khat extract (A) and in initial state (B).

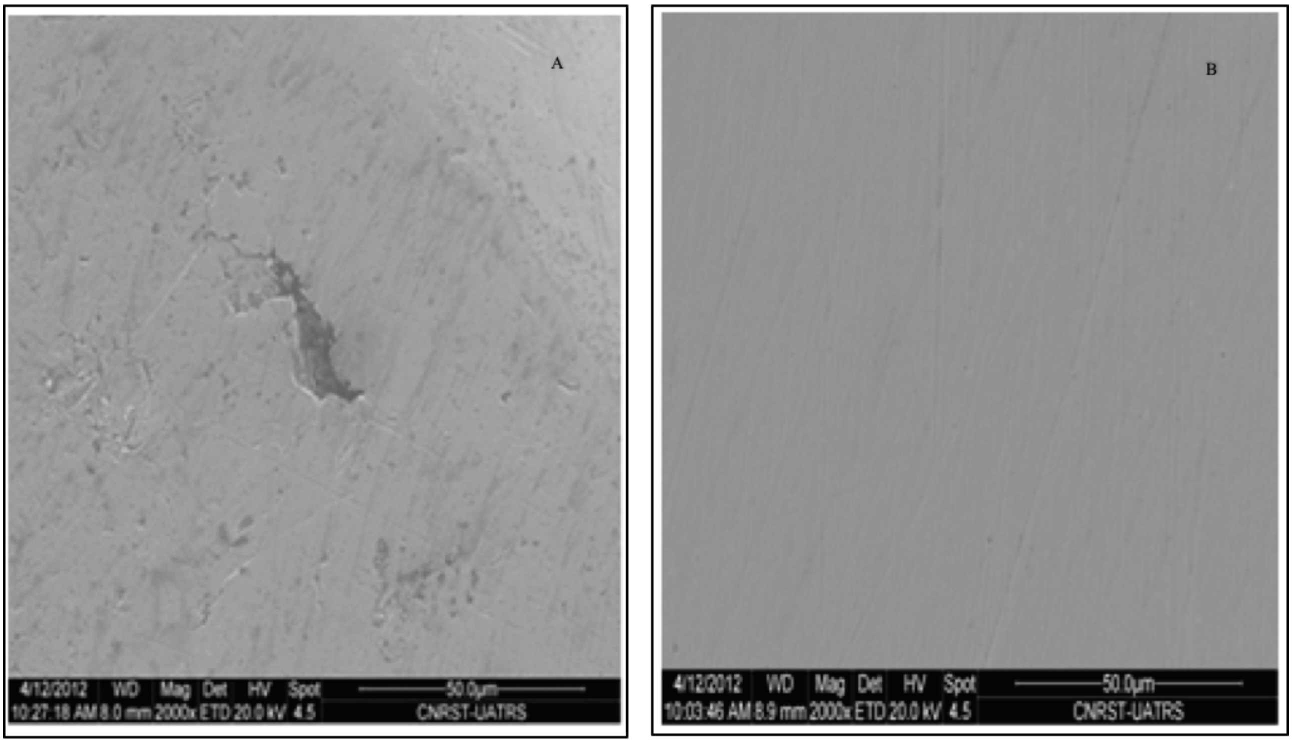

Figure 6 - The scanning electron microscopy of PD Casta H® in artificial saliva containing khat extract (A) and in initial state (B).

DISCUSSION

Khat chewing is a widespread social habit in some parts of the world. This habit has been reported to have several detrimental effects on oral and dental tissues [8,9,23,24]. There is an extensive literature on khat-related oral conditions [6-9,12,23,24]. However, the literature is scarce regarding the potential effects of khat on dental prosthesis and restorations. Hence, the present study investigated the effects of Khat extract on the corrosion behavior of Co-Cr dental alloy used for removable partial dentures. To the best of our knowledge, this is the first study that evaluated the effects of khat on the corrosion resistance of Co-Cr dental alloys. Overall, the results of the present study showed that the corrosion resistance of the three alloys tested (Neobond II®, Kera 501® and PD Casta H®) decreased in artificial saliva containing khat extract compared with that of reference solution. Additionally, the corrosion resistance of the three Co-Cr dental alloys decreased by increasing concentration of khat extract. Furthermore, the results indicate that Neobond II® alloy which contain no Mo show least corrosion resistance compared with that of Kera 501® and PD Casta H® alloys.

Khat leaves contain a complex group of alkaloids namely, phenylalkylamines, phenylpentylamines, and cathedulins [3]. Khat also contains amino acids, tannins, vitamins and minerals (Calcium, Manganese, Iron, Zinc and Fluoride) [3,5,25]. Chemicals components of khat, particularly fluoride may have a critical role in corrosion of metallic materials [5]. Khat chewing involves chewing fresh khat leaves and storing the bolus inside the mouth for several hours. As a result, the chemical substances of khat usually dissolve in the saliva and may affect the metallic materials and prosthesis [15]. Altabachew et al. [5] assessed the Fluoride content of Ethiopian khat (Catha edulis) chewing leaves; and found that the fluoride concentration was 12 $\mu g\ g^{-1}$ in matured leaves and 6.5 $\mu g\ g^{-1}$ in young leaves [5]. Interestingly, Ameer et al [26] showed that in artificial saliva containing different concentration of fluoride, the corrosion rate of Co-Cr and Ni-Cr dental alloys increased by increasing concentration of fluoride. In line with that, Prioteasa et al [27] showed that by adding of fluoride ions to Fusayama-Meyer artificial saliva, the corrosion resistance of Co-Cr and Ni-Cr dental alloys is diminished. The results of the latter two studies substantiate the theory that fluoride and other chemical components of khat may be responsible for the decrease in corrosion resistance of Co-Cr alloys seen in the present study. The prolonged close contact of khat with Co-Cr alloy in khat chewers may lead to accelerated corrosion of Co-Cr alloy, with subsequent release of corrosion products including metal ions. This is very important because corrosion products of dental alloys contain metal ions and may be the reason for allergic and some other diseases reported among khat chewers.

Unsurprisingly, Neobond II® alloy, which contain no Mo showed the least corrosion resistance compared with that of Kera 501® and PD Casta H® alloys. Geis-Gerstorfer et al [28] showed that alloys, which contain no Mo are known to be susceptible to pitting corrosion. On the other hand, Kera 501® and PD Casta H® alloys, which contain a high Co content (61 wt. %, 60 wt. % respectively), showed the best corrosion resistance compared with that of Neobond II® alloy which contain Co content (52.2 wt. %). Thus, in Fusayama-Meyer artificial saliva the corrosion resistance of the three Co-Cr dental alloys are in the following order: Kera 501® ≈ PD Casta H® > Neobond II®. The results of surface analysis by using the scanning electron microscopy showed corrosion on the surface of Co-Cr alloys after immersion in artificial saliva containing khat extract; such results substantiate the results obtained from potentiodynamic polarization curves and Nyquist impedance diagrams. On the basis of the results obtained, khat chewing should be avoided in patients treated with Co-Cr dental alloys. Further in-vitro and in-vivo studies are highly recommended.

CONCLUSION

The findings of the present study suggest that the Khat (Catha edulis) has a negative effect on the corrosion resistance of Co-Cr alloy. The corrosion resistance of the three Co-Cr alloys decreased in artificial saliva containing khat extract, and the degree of corrosion resistance was associated with the concentration of the khat extract.

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