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Evaluation the Effect of Diazonium Salt and Aluminum Oxide on the Shear Bond Strength of Heat Cure Acrylic Resin at Co/Cr and Titanium Alloys Interface

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Abstract

Bonding between metal components and denture base resin plays an important role in the success of removable partial denture. Removable partial denture prostheses often present a mechanical failure. The bonding between metal framework and acrylic resin is usually a mechanical interlock, since they do not chemical bond spontaneously. The aim of this study was to evaluate the effect of different surface treatments (diazonium salt and aluminum oxide) on the shear bond strength of PMMA at the Co/Cr and titanium alloys interface. Sixty-disc shape specimens were prepared and divided into 2 main groups according to the type of material, 30 specimens for Co/Cr alloy and 30 specimens for titanium alloy, then subdivided into 3 groups each one consist of 10 specimens, according to type of surface treatments. Each group had control group, aluminum oxide group (110 µm), and diazonium salt group. The specimens were thermocycled (3000 cycles) after applications of PMMA. Shear bond strength test conducted with crosshead speed 0.5mm/min. Data analyzed via One-way ANOVA and GHD test. Co/Cr alloy revealed that diazonium salt surface treatment had highest mean value of shear bond strength followed by aluminum oxide surface treatment at $p<0.01$. For titanium alloy, the results showed the diazonium salt surface treatment had highest mean value of shear bond strength followed by aluminum oxide surface treatment group at $p<0.01$. Aluminum oxide treatment slightly increased shear bond strength for both Co/Cr and titanium, while diazonium salt treatment statistically increased shear bond strength for Co/Cr and titanium alloy.

Introduction:

Removable partial denture is a combination of metal framework that fitted over the residual ridges and attached to natural teeth and acrylic denture base for teeth and gum replacement have been an inexpensive and predictable treatment option for the rehabilitation of partially edentulous patients [(1)]. Poly methyl methacrylate (PMMA) is extensively used indenture materials for removable partial dentures because of its biocompatibility, excellent esthetic and mechanical properties. Cobalt chromium alloy and titanium alloy are commonly used to fabricate the metal framework prosthesis for removable partial denture because for their biocompatibility and corrosion resistant as well as desirable mechanical properties [(2, 3)]. The bonding between the metal framework and the acrylic denture base resin plays an important role in successful of the removable partial denture prosthesis [(4, 5)]. Any defect or separation between the acrylic resin and the metal framework, especially at the finishing line, cracks or crazing may occur in the acrylic resin, leading to discoloration, deterioration of the acrylic resin, and the creation of a reservoir for oral debris and microorganisms. The lack of bond can directly affect the metal-resin interface [(2)]. Several methods have been tested to increase the bond strength between acrylic resin and alloys in dental prostheses such as electrolytic etching, sand blasting, silica coating, chemical bonding and metal primer [(6)]. Mechanical bonding is based on macromechanical or micromechanical retention. The macromechanical retention has several types like lattice, mesh, beads, nail heads, pins and loops that retain acrylic resin to the frame. Lattice design is particularly susceptible to permanent deformation, and the open lattice design provides optimum retention for acrylic resin. Mesh can be used in every particular clinical situation, beads or nail heads are the weakest types among acrylic retentive designs. Macromechanical retention was correlated with over contouring and space problems; the lack of chemical adhesion makes the interface subject to break over time [(5)]. The sandblasting technique or air abrasion is a mechanical method for cleaning and modifying the surface roughness and is commonly have been associated with enhanced bond strength, sandblasting made by spraying sand or micron size particles are hard and sharp with a moderately high speed in the surface layer [(7)]. Chemical bonding it is a surface chemistry modification, and a chemical-active substance, which is normally liquid and has bonding rapprochement to both surfaces, binds two dissimilar surfaces. Chemical bonding may be interfacial or adhesive in nature. The interfacial chemical bonding system has been developed as a substitution to etching processes. The successful dental adhesion process involves different factors which including contact angle, purity, or cleanliness of bonding surfaces [(8)]. Aryldiazonium salts are organic compounds have been used as a chemical bond to modify material surfaces for many applications [(3)]. The aryl diazonium salts can be induced to provide aryl layers that can "graft onto" many different interesting surfaces with electrochemical, ultrasonic, chemical, or photochemical techniques that can be covalently bound to a wide range of metallic surfaces [(9)].

Materials and Methods:

Co/Cr alloy and titanium alloy specimen`s preparation

A metal mold designed to reproduce wax pattern in disc shape with a dimension of 2mm thickness, and 10mm diameter for the 30 Co/Cr specimens and 30 titanium specimens. Co/Cr alloy (Adentatec, Germany) casted by using centrifugal casting technique, following the manufacturer’s instructions, titanium alloy (Baoji Jinsheng, China) casted by using vacuum-pressure machine, following the manufacturer’s instructions. All the surfaces of specimens were ground finished using silicon carbide paper (number p60) (Trojan, China) in the grinding machine (160E, Mapao, China) under running water on a 300 rpm for 10 seconds in order to provide uniform and flat surface [(10, 11)] and cleaned ultrasonically for 3 minutes with deionized water and dried with air [12].

Application of Heat Cured Acrylic Resin

After obtaining 30 disk shape specimens for each Co/Cr alloy and titanium alloy, a special metal split mold was used for all disk specimens to add modeling wax (Shanghai, China) in a specific area placed centrally with dimensions of 5mm diameter and 2mm thickness [6]. These specimens (Co/Cr alloy/titanium alloy disk modeling wax assemblies) were flasked in a standard flasking technique for acrylic dentures with dental stone (Easy Dental, Bulgaria) [13, 14], and then the specimens were dewaxed and cleaned using boiled water [2]. Before packing, the specimens of each Co/Cr and titanium divided into 3 groups according to the surface treatment that they received (n=10 for each group):

- Control groups: Without surface treatment.
- Aluminum oxide groups: Abraded with alumina oxide (Al_2O_3) 110 µm particle size (Renfert, Germany) with an airborne particle sandblasting machine (Rotex, Turkey) at 2 bar pressure for 15 seconds with 10mm of distance that was standardized by using a specially designed holder [15] and then ultrasonically cleaned (Quigg, Turkey) with deionized water for 10 minutes and dried by air [16, 17].
- Diazonium salt group: by using Aryldiazonium salts which is done in a two-stages protocol (priming and adhesive)[3]. The first stage (priming) was as follows: Diaminodiphenyl sulfone (0.248.3 g) and $NaNO_2$ (0.07 g) dissolved in a glass beaker containing 250 ml of 1 Molar HCl [18] the temperature of solution should be between (0-5) °C [19]. The solution mixed with a magnetic stirrer for 5 minutes to produce Aryldiazonium salt, the Co/Cr and titanium specimens immersed in solution by using (DC) power supply (JYD Inc, China) and two-electrode cell and left to react with a magnetic stirrer for 45 min under 0.42±0.05 Volt [20]. All the specimens were ultrasonication in distilled water and acetone for 5 min to remove any ungrafting matter, this first step leads to spontaneous grafting of apolyaminophenylene (PAP) layer on each Co/Cr and titanium specimens. These samples referred to as (metal–PAP). The second (adhesive) step conducted in order to optimize the adhesion of monomer (MMA) to (metal–PAP) samples as follows: $NaNO_2$ (0.034 g) dissolved in in a glass beaker containing 250 ml of 1 Molar HCl [3], sodium dodecyl sulfate (SDS) (0.026 g) and MMA were applied [21], and then Co/Cr and titanium specimens (metal–PAP specimens) immersed in solution by using (DC) power supply, and left to react with a magnetic stirrer for 45 min under 0.42±0.05 Volt [20]. All the specimens were ultrasonication in distilled water and acetone for 5 min to remove any ungrafting matter.

The heat cured acrylic resin (RODEX, Turkey) mixed with powder: liquid ratio of 3:1 and packed by placing the flask in a hydraulic press (Rotex, Turkey), with 5 MPa pressure was applied slowly [7]. All the specimens were heat processed according to the manufacturer's instructions [22, 23]. The 60 specimens of all groups for each Co/Cr alloy and titanium alloy were subjected to 3,000 thermocycles in artificial saliva between 5°C and 50°C with a dwell time of 1 minute using a thermocycling system (Cooler: Beko, Turkey; Herter: Windom, China; Custom made holder) [17, 24].

Preparation of specimens for Bond Strength Test

The specimens inserted in the center of autopolymerizing acrylic resin base (RODEX, Turkey) by using a special silicon mold. To obtain the bond strength the specimens mounted in the specimen holder of Instron universal testing machine and specimen's surface in parallel to the loading piston, and load with a crosshead speed of 0.5 mm/min, Fig.(1) [6]. The load at failure was recorded in (N) and dividing by the bonded surface area (mm^2) to obtain the bond strength in (MPa) [25]. After the test, type of failure defined as adhesive, cohesive, and mixed, Fig.(2). The specimens examined under a stereomicroscope with 20X magnification images of the fracture site to verify the type of failure [26].

Statistical Methods

The study data analyzed via One-way ANOVA and Games Howell Difference test used to determine the pair differences.

Results:

Shear bond strength test

The bond strength for all tested groups is graphically presented in Fig. (3); the results indicate that the mean bond was maximum for Co/Cr diazonium salt treatment group (9.786 ± 1.504) and minimum for titanium control group (1.679 ± 0.966). According to the results of one-way ANOVA tests as shown the difference was statistically highly significant among all groups at $p<0.01$, Table (1). On applying Games Howell test for multiple pair wise comparisons between tested groups in Table (2).
All control groups and aluminum oxide groups for each Co/Cr alloy and titanium alloy exhibit predominantly adhesive failure, however, a mixed failure were mostly observed in diazonium salt group of each Co/Cr alloy and titanium alloy.

Fourier Transforms Infrared Spectroscopy (FTIR)

All various functional groups present are studied through FTIR analysis, the characteristic bonds of benzene ring for diazonium that bind with Co/Cr alloy are peaks at 3127.01 cm^{-1}, 1597.73 cm^{-1}, 3283.21 cm^{-1}, 1407.78 cm^{-1}, and 1254.74 cm^{-1}. The functional groups of monomer that entanglement the diazonium are peaks at 1646.91 cm^{-1}, 1091.51 cm^{-1}, and 2923.59 cm^{-1}, Fig.(4). The functional groups of diazonium that characterize the chemical reactions with titanium alloy are peaks at 3118.33 cm^{-1}, 1518.67 cm^{-1}, 3238.86 cm^{-1}, 1407.78 cm^{-1}, and 1261.22 cm^{-1}. The functional groups of monomer that entanglement the diazonium are peaks at 1638.23 cm^{-1}, 1107.9 cm^{-1}, and 2927.41 cm^{-1}, Fig. (5).

Discussion:

The bonding between the metal framework and the acrylic denture base resin plays an important role in successful of the removable partial denture prosthesis; deficiencies in bonding at the metal-resin interface consider a significant problem that leads to failure of the prosthesis [17].

Aluminum oxide surface treatment

The surface treatment with aluminum oxide particle abrasion has been recommended methods to establish an acceptable surface condition to improve bond strength, eliminate the contaminated layer, debris and/or metal oxides and increase the surface area by creating micromechanical roughness [27, 28]. In addition, this treatment increases the surface wettability of the material [29].
In this study the aluminum oxide group of each Co/Cr alloy and titanium alloy with 110 µm Al_2O_3 particle size exhibit a significantly slightly higher bond strength than the specimens without surface treatment, this is due to the variation of surface morphology that advances a micromechanical interlocking sites as well as greater wettability, which allows the heat cured acrylic resin be mechanically joined and increase the mechanical bonding.

Diazonium salt surface treatment

Diazonium salts can graft easily by covalent bonding on the metal surface [30]. The diazonium chemistry used to bind PMMA with (Co/Cr alloy and titanium alloy) strongly. The chemical mechanism began in the first step when the amino one ends NH_2 (~ C_6H_4-NH_2) of diaminodiphenyl sulfone transformed into diazonium cation (~ C_6H_4-N_2^+) by adding $NaNO_2$, the electrochemical reduction will reduce the diazonium cation into benzene ring free radical (~ C_6H_4•) and grafted on the surface of each (Co/Cr alloy and titanium alloy) specimen and then forming (PAP) layer. The second step designed to change the second end of amino NH_2 of

diaminodiphenyl sulfone like in the first step into diazonium cation (~ C_6H_4-N_2^+) and then into (~ C_6H_4•). The free radical of carbon benzene ring (~ C_6H_4•) will react with double bond (C=C) of monomer, (SDS) disperse the monomer in the aqueous reaction solution by forming micelles around the monomer droplets, the polymerization of monomer leads to the formation of an adhesive layer [21, 31].
The non-significant difference that is shown between Co/Cr alloy and titanium alloy diazonium salt group could be explained due to diazonium salt grafted in the same manner on the different metal substrates by covalent bond that formed between diazonium layers Polyaminophenylene (PAP) and metal substrate, also diazonium salt is capable to increase the wettability of metal substrate [3].

Conclusion:

In present study, aluminum oxide surface treatment increased shear bond strength of heat cured acrylic resin to both Co/Cr alloy and titanium alloy. Diazonium salt surface treatments for Co/Cr alloy showed highest shear bond strength with heat cured the acrylic resin among all studied groups. Also, the shear bond strength have been improved with diazonium salt surface treatment for titanium alloy.

Fig. (1): Specimens mounted in the universal testing machine.

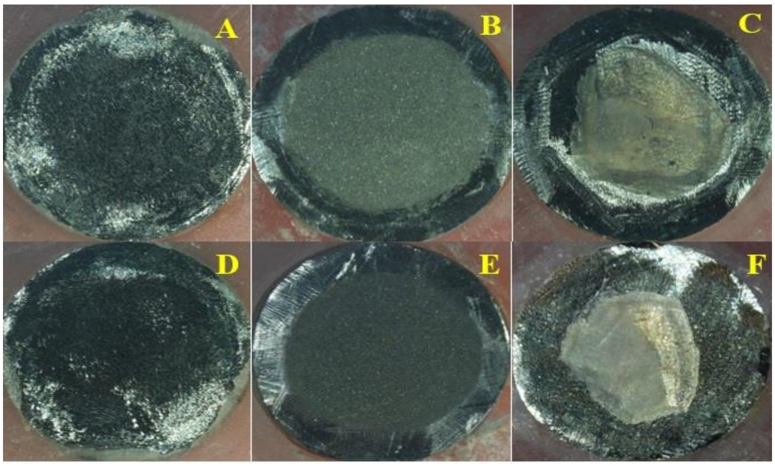

Fig. (2): The mode of failure image.
(A): Co/Cr control group. (B): Co/Cr aluminum oxide group. (C): Co/Cr diazonium group.
(D): titanium control group. (E): titanium aluminum oxide group. (F): titanium diazonium group.

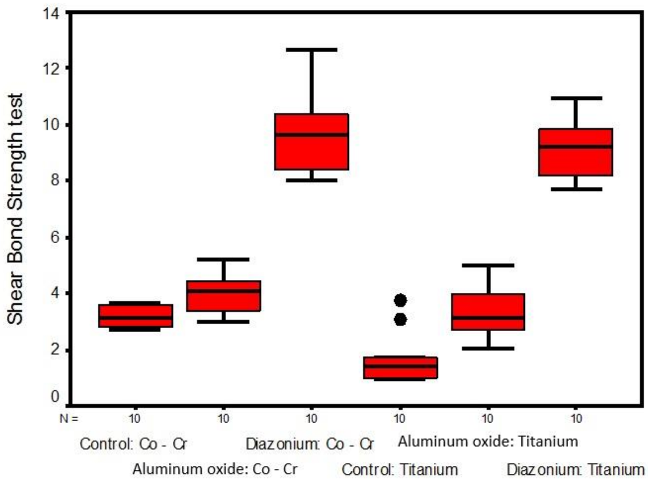

Fig. (1): Box plot for shear bond strength test.

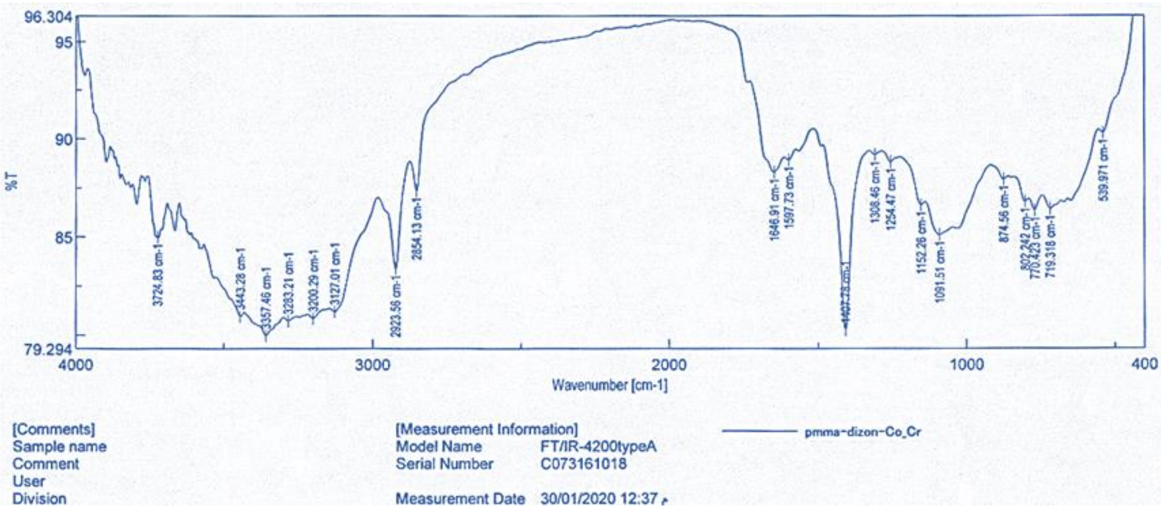

Fig. (2): FTIR spectrum of Co/Cr alloy.

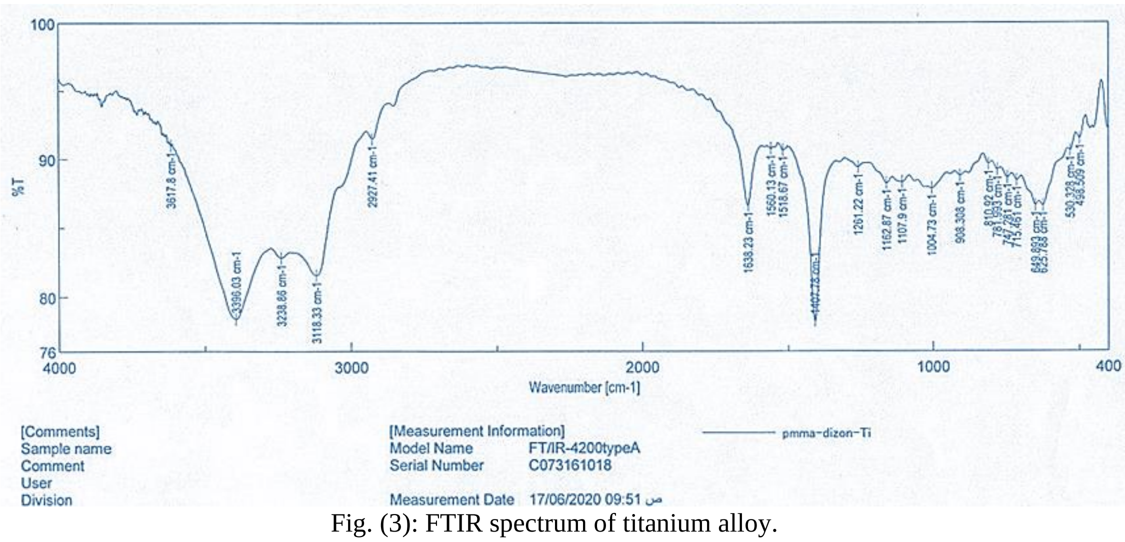

Fig. (3): FTIR spectrum of titanium alloy.

Table (1): Descriptive analysis statistics with Levene's and one-way ANOVA tests

Groups	No.	Materials		ANOVA Difference rest	
		Co/Cr alloy	Titanium alloy	Levene`s Test	F-test
		Mean ± SD	Mean ± SD		
Control	10	3.159 ± 0.366	1.679 ± 0.966	p=0.018 (S)	p=0.000 (HS)
Aluminum oxide	10	3.991 ± 0.717	3.358 ± 0.92		
Diazonium salt	10	9.786 ± 1.504	9.138 ± 1.13		
*** HS: Highly Sig. at p<0.01; S: Sig. at p<0.05**					

Table (1):- Games Howell Difference test for all studied groups.

Groups	Co/Cr control	Co/Cr aluminum oxide	Co/Cr diazonium salt	Titanium control	Titanium aluminum oxide	Titanium diazonium salt
Co/Cr control	-	NS	HS	HS	NS	HS
Co/Cr aluminum oxide	-	-	HS	HS	NS	HS
Co/Cr diazonium salt	-	-	-	HS	HS	NS
Titanium control	-	-	-	-	HS	HS
Titanium aluminum oxide	-	-	-	-	-	HS
Titanium diazonium salt	-	-	-	-	-	-
* HS: Highly Sig. at p<0.01; NS: Non Sig. at p>0.05						

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