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By Abdullah Jasim Mohammed

Original Article

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Abstract

Objectives. This study was aiming to evaluate the influence of Titanium dioxide nanoparticles addition on the bonding performance of platinum primer (G611) between maxillofacial silicone and acrylic substrate.

Materials and methods. 180 samples were divided on two tests (shear and peel bond strength) and they were divided into two main groups (cold cure, and 3D printed) acrylic resin each group were subdivided into 5 subgroups (N=9) according to the concentration of nano-filler addition (0%, 0.5%, 1%, 1.5% and 2%) by weight. The data obtained were analyzed using one-way ANOVA and Tukey's post hoc tests at P<0.05.

Results. Statistical analysis revealed (0.5% and 1%) concentrations had significantly the highest bond strength in both groups, whereas (2%) had the lowest bond strength in both groups.

Conclusion. Reinforcement of the platinum primer G611 with Titanium dioxide (TiO₂) nanoparticles (1%) concentration by weight resulted in a significant

improvement in the bonding performance of the platinum primer G611 larger concentrations (1.5%, and 2%) resulted in declining bonding strength.

Keywords: Silicone elastomer, maxillofacial prosthesis, Acrylic resin, Nanoparticles, 3-D printing

Introduction:

A maxillofacial appliance is a prosthetic that fills in the gaps left by congenital or acquired abnormalities brought on by trauma or surgical procedures (mainly because of neoplasms). In general, maxillofacial prostheses have been held in place by spectacles, anatomical undercuts, and medical-grade adhesives. But the development of osseo-integrated implants provided a better method for maintaining maxillofacial prostheses [1-6]. The housing is frequently required for an implant-retained facial prosthesis in order to keep attachments in the proper place. This housing is typically made of acrylic resin materials [5,7] Although acrylic resins and maxillofacial elastomers are all polymers, their chemical composition are different, for this reason a bonding agent is frequently required to enhance adhesion between the two materials [8]. These adhesives prepare the substrates by etching them or encouraging covalent and hydrogen bonds. They also increase the substrate's wettability by enabling the polymeric substances to infiltrate the resin surface.[9] Although silicone elastomers have improved greatly in terms of their physical and mechanical characteristics, rubber delamination from the retaining substrate is still a persistent issue [10]. Previous studies have been conducted to evaluate the best method for increasing this bond strength, using shear and peel forces tests in their evaluation. These tests are nearest to clinical circumstances to measure the bond strength between the acrylic substrate and the facial elastomers [9,11,12]. The lateral displacement of the prosthesis is believed to be caused by forces with a horizontal component, the peel strength test is thought to simulate these displacing force [13]. Many of these studies were focusing on the alteration of the topography of the substrate surface, either mechanically or by adding various types of primers that contain different kinds of

solvents [7,14–16]. However, little attention has been given for the alteration of the mechanical and physical properties of the liquid primers by adding nano-fillers to increase their bonding capacity. Therefore, the purpose of the current study was to evaluate the influence of adding Titanium dioxide nano-fillers to the bonding agent of the platinum primer used for bonding platinum cured facial elastomers to an acrylic substrate. Previous studies showed that the addition of filler particles to various adhesive systems could alter their chemical or rheological properties of these adhesive liquids [17,18]. Nanoparticles in the adhesive systems can raise the bond strength by different means. Where, fillers could work by blocking and exhausting the crack propagation at the interface of the adhesive and substrate. Consequently, could alter the energy level required for adhesive-substrate interface failure [19–21]. Titanium dioxide nanoparticles have excellent mechanical characteristics. The unique photoactive properties of (TiO₂) nanoparticles and their excellent physical and mechanical characteristics made them an ideal additive to improve the effectiveness of polymeric and adhesive materials [22,23].

Materials and methods:

The effect of adding titanium dioxide nanoparticles (Skyspring Nanomaterials, Inc., Houston, USA) to the G611 platinum primer (Principality Medical limited, Newport, UK) on the bonding performance of the primer was evaluated using shear and peel bond strength tests to measure the bond strength between both self-cure acrylic resin (Veracril, New stetic Co., Colombia), 3D printed denture base resin (Senertek, Senerlabs, Turkey) and a Cosmesil facial silicone (Cosmesil M511; Principality Medical limited, Newport, UK).

Titanium dioxide nanoparticles (0.0038g, 0.0076g, 0.0114g, and 0.0152g) using digital balance (with four digits precision; PG 503-S MonoBloc inside, Mettler Toledo Ltd, Switzerland) were added to (1ml) of the G611 platinum primer to form nano-filled liquid. The concentration percentages of the nanoparticles were (0.5%, 1%, 1.5%, and 2%) by mass respectively. To ensure optimal distribution of the

nanoparticles in the liquid primer. One hundred-eighty samples were fabricated, and they were divided to two tests (shear and peel bond tests). Ninety samples for each test were divided to cold cure resin and 3D printed resin groups which were subdivided to five subgroups (0% addition as control group), and four groups followed the concentration abovementioned (0.5%, 1%, 1.5%, and 2%). One hundred-eighty samples were fabricated, and they were divided to two tests (shear and peel bond tests). Ninety samples for each test were divided to cold cure resin and 3D printed resin groups which were subdivided to five subgroups (0% addition as control group), and four groups followed the abovementioned concentrations (0.5%, 1%, 1.5%, and 2%). The surface of the samples were cleaned with acetone liquid. The G611 platinum primer were added using a brush, the primer were left to dry for 30 minutes according the manufacturer's recommendations. The facial silicone M511 (part A) was mixed with part B catalyst (ratio of 10:1), and left for 24 hours at room temperature for curing.

The fabrication method of the samples used for shear bond test was described in previous studies as follows [5,24]. An auto-polymerizing acrylic resin were mixed and packed inside PVC tubes (12.5mm external diameter, 11mm internal diameter, and 20mm height). The 3D printed resin samples were fabricated using 3D printing machine (LD-002H, Creality Co., Shenzhen, China) a disk with 10mm diameter, and 3mm thickness were fabricated and fixed at the top of the PVC tubes with aid of cold cure resin. After complete polymerization, and printing the surface were finished and the primer filled with nanoparticles were applied on the substrate surface. A plastic rings were fixed on the top of each PVC tube with (10mm external diameter, 8mm internal diameter, and 3mm height), and packed with facial silicone material under 10Kg of weight. The samples were placed in the universal testing machine (GESTER International Co., Quanzhou, China). And the shear bond strength was measured with a cross head speed 10mm/min. The shear bond strength was calculated by dividing the failure load (Newton) by the attached surface area (mm²).

For evaluation of the peel bond strength of the nano-filled modified primer a rectangular auto-polymerizing acrylic using conventional flasking procedure, and 3D printed resin samples using DLP 3D-printing method, both with (75mm length, 10mm width, 3mm thickness) were fabricated according to ASTM D 903 – 98 [2,5]. A layer of modeling wax with the same dimensions was placed over the resin one to create a room for the silicone material. After setting of the stone the twin sample was removed, and the space created was used to pack the silicone material. the primer was added as a thin layer on the area of the attachment which represent the terminal 25mm (length) and 10mm (width) from the original samples' dimensions, a separating tape were attached to the rest of the acrylic samples and the M511 silicone material packed in the prepared flasks over the acrylic samples, pressed constantly for 24 hours at 3500psi. The peel strength of the samples were tested with the universal testing machine with crosshead speed set on 10mm/min. The acrylic part was grasped with the lower jaw of the machine and the free end of the silicone turned upward and grasped with upper jaw. The peel strength (PS) (Newton/mm) were estimated using the following equation [13]:

PS = F/W((1+L/2) +1)

Where F is the failure force (Newton), W is the width of the sample, and L is the ratio between the stretched and non-stretched length of the silicone part. The statistical analysis was accomplished using SPSS (17) (SPSS Inc., Chicago, IL, USA), ANOVA and Tukey's tests. The significance level was defined at P < 0.05.

Results:

Shear and peel bond strength were tested to assess the impact of titanium dioxide nanoparticle addition to the platinum primer G611. Tables (1) show the mean and standard deviation of the shear bond strength for the main groups of self-cured and 3D printed resin. One way ANOVA statistical test showed that there was a significant statistical difference among all concentrations added per weight (0% control, 0.5%, 1%, 1.5%, and 2%) (significance 0.000) at P< 0.05. Tukey's test for

group were conducted and showed that the shear bond strength for (1%) addition was the highest followed by (0.5%), (1.5%), (0% control), and (2%) respectively. The mean and standard deviation of the peel bond strength is presented in Table (2) for the two main groups. One way ANOVA and Tukey's multiple comparison tests revealed that there is a significant difference among all concentrations added (0.000) at P < 0.05, with one exception that the difference between (0%) control group and (1.5%) addition had insignificant difference. The highest mean of peel strength was occurred with (1%) addition followed by (0.5%), (0% control), (1.5%) and (2%) respectively.

Discussion:

The adherence of different types of materials can occur due to mechanical adhesion, micromechanical adhesion, and molecular bond. The bond created by the primers depends on the mechanical and physical strength of the primers mainly because of its molecular attraction, and the chemical affinity of such primers to the facial silicone and the acrylic substrate [26-28]. The bonding performance of these primers can be influenced by their ability to wet the substrate surface, so they would be capable to fill the surface micro-irregularities, thereby increasing the surface area of the attachment, in addition to the micromechanical engagement of these irregularities [29,30]. The addition of titanium dioxide nanoparticles improved the bonding ability of the platinum primer G611 employed in the investigation, as evidenced by the shear and peel bond strengths assessed in this study. For both experiments, the increased bonding strength was at its highest at (1%) by weight concentration. However, this effect was already noticeable at lower concentrations (0.5% and 1%), but as nanoparticle concentrations were increased continuously, the bond strength began to steadily decline starting at (1.5%) concentration, reaching its lowest shear and peel bond strength levels at (2%) concentration. The addition of nano-fillers to various adhesive agents can change their chemical, and physical properties, where, these nano-fillers can absorb some of the energy of the applied force that tend to separate the silicone from the acrylic substrate, or they could increase the bonding performance by bridging the initial crack propagation by their higher ductility, consequently higher energy level would be required to break the bond between the silicone and the acrylic resin [18-20]. Also the increased bonding ability of the adhesive agents by the addition of nano-fillers could be the result of decreased viscosity of the bonding agents as long as the incorporation of nano-fillers in small concentrations can increase flow properties of the liquids, consequently, this would increase the surface wettability by the bonding agent, which is an essential property for the adhesive systems that could improve their bonding performance [9,31,32]. Because of their small size particles, more surface area would be available, they can be added in small amounts to achieve significant improvement in the bonding performance. For each individual adhesive system there is an optimal amount of nanoparticles to achieve the best effect on the bonding strength [33,34]. Higher concentration of nanoparticles can result in adverse effect due to clustering of the nanoparticles before being dispersed in the liquid, the high surface energy nanoparticles make them more vulnerable to aggregate. This agglomeration increase greatly when nano-fillers are added in higher concentration, this may reduce the distance among nanoparticles making them more prone to attract to each other, resulting in weakness of the adhesive agents, and declining bonding strength of the adhesive systems [35–37]. The results of the current study are in agreement with the results of previous studies who found that the addition of nanoparticles to an adhesive liquid can promote the strength of the bonding agent and strengthens the interface between bonded materials, and can influence the degree of conversion of the adhesive agent and the efficiency of polymerization [18,19,33,38]. Also, the incorporation of nanoparticles to an adhesive system can decrease the viscosity of the liquid, and also can increase the bonding capacity of the bonding agent [29-32]. Using one type of primers and one type of the silicone materials, in addition to the use of one kind of acrylic resin were the important limitations in the current study, furthermore, the surface treatment of the acrylic substrate was neglected in this study, which can be suggested to be evaluated in future studies as a combined effect of nano-fillers

and surface treatment, which could be a promising line for achieving optimum bonding strength of the facial silicone to the acrylic substrate. Detachment of the silicone elastomer from their acrylic substrates can initialize critical clinical problem, the addition of 1% TiO₂ particles can increase the bonding performance of the platinum primer used to bond the maxillofacial elastomer to the acrylic substrate. subsequently, it will increase the service time of the maxillofacial prostheses.

Conclusions:

Reinforcement of the platinum primer G611 with Titanium dioxide (TiO₂) nanoparticles in (0.5%, and 1%) concentrations by weight resulted in a significant increase in the means of both shear and peel bond strength. Indicating that Titanium dioxide nanoparticles can be added in a small concentrations (1%) to achieve better bonding performance of the platinum primer G611. The addition of the (TiO₂) nano-fillers in larger amounts (1.5%, and 2%) concentrations can result in declining bonding strength. The addition of (TiO₂) nano-fillers in (1%) concentration had the highest means of shear and peel tests, whereas, (2%) concentration means of shear and peel strength indicates that the addition of (TiO₂) nano-fillers can significantly compromise the bonding performance of the platinum primer used in this study.

Conflict of interests: No conflict of interest.

Ethics approval: Approval allowed from Research and Ethics Committee, Dentistry College, Mosul University, IRAQ. (code No. UoM.Dent/H.DM.18/23).

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References:

1. Parel SM, Branemark PI, Tjellstrom A, Gion G. Osseointegration in

- maxillofacial prosthetics. Part II: Extraoral applications. J Prosthet Dent. 1986;55(5):600–6.
- Wolfaardt J, Gehl G, Farmand M, Wilkes G. Indications and methods of care for aspects of extraoral osseointegration. Int J Oral Maxillofac Surg. 2003;32(2):124–31.
- Schoen PJ, Raghoebar GM, van Oort RP, Reintsema H, van der Laan BFAM, Burlage FR, et al. Treatment outcome of bone-anchored craniofacial prostheses after tumor surgery. Cancer Interdiscip Int J Am Cancer Soc. 2001;92(12):3045–50.
- Watson RM, Coward TJ, Forman GH. Results of treatment of 20 patients with implant-retained auricular prostheses. Int J Oral Maxillofac Implants. 1995;10(4).
- Hatamleh MM, Watts DC. Mechanical properties and bonding of maxillofacial silicone elastomers. Dent Mater. 2010;26(2):185–91.
- Frangou MJ, Polyzois GL, Tarantili PA, Andreopoulos AG. Bonding of silicone extra-oral elastomers to acrylic resin: the effect of primer composition. Eur J Prosthodont Restor Dent. 2003;11(3):115–8.
- Polyzois GL, Frangou MJ. Bonding of silicone prosthetic elastomers to three different denture resins. Int J Prosthodont. 2002;15(6).
- 8. Qudah S, Harrison A, Huggett R. Soft Lining Materials in Prosthetic Dentistry; A Review. Int J Prosthodont. 1990;3(5).
- Mutluay MM, Ruyter IE. Evaluation of bond strength of soft relining materials to denture base polymers. Dent Mater. 2007;23(11):1373–81.
- McMordie R, King GE. Evaluation of primers used for bonding silicone to denture base material. J Prosthet Dent. 1989;61(5):636–9.

- Gibbs CH, Mahan PE, Lundeen HC, Brehnan K, Walsh EK, Holbrook WB.
 Occlusal forces during chewing and swallowing as measured by sound transmission. J Prosthet Dent. 1981;46(4):443–9.
- Minami H, Suzuki S, Ohashi H, Kurashige H, Tanaka T. Effect of surface treatment on the bonding of an autopolymerizing soft denture liner to a denture base resin. Int J Prosthodont. 2004;17(3).
- Al-Athel MS, Jagger RG. Effect of test method on the bond strength of a silicone resilient denture lining material. J Prosthet Dent. 1996;76(5):535–40.
- Hattori M, Sumita YI, Muthiah L, Iwasaki N, Takahashi H, Aimaijiang Y, et al. Effect of fabrication process on the bond strength between silicone elastomer and acrylic resin for maxillofacial prosthesis. Dent Mater J. 2014;33(1):16–20.
- Al-Shammari FAK. Effect of different primers on the peel bond strength between silicone elastomer and acrylic resins. Br J Med Med Res. 2015;8(12):1034–44.
- Polyzois GL, Frangou MJ, Andreopoulos AG. The Effect of Bonding Agents on the Bond Strengths of Facial Silicone Elastomers to a Visible Light-Adivated Resin. Int J Prosthodont. 1991;4(5).
- Reddy AK, Kambalyal PB, Patil SR, Vankhre M, Khan MYA, Kumar TR. Comparative evaluation and influence on shear bond strength of incorporating silver, zinc oxide, and titanium dioxide nanoparticles in orthodontic adhesive. J Orthod Sci. 2016;5(4):127.
- 18. Tankut N, Bardak T, Sozen E, Tankut AN. The effect of different nanoparticles and open time on bonding strength of poly (vinyl acetate) adhesive. Measurement. 2016;81:80–4.
- 19. Belli R, Kreppel S, Petschelt A, Hornberger H, Boccaccini AR, Lohbauer U.

- Strengthening of dental adhesives via particle reinforcement. J Mech Behav Biomed Mater. 2014;37:100–8.
- Dos Santos AF, de Lucena FS, Borges AFS, Lisboa-Filho PN, Furuse AY. Incorporation of TiO2 nanotubes in a polycrystalline zirconia: Synthesis of nanotubes, surface characterization, and bond strength. J Prosthet Dent. 2018;120(4):589–95.
- Ramos-Tonello CM, Lisboa-Filho PN, Arruda LB, Tokuhara CK, Oliveira RC, Furuse AY, et al. Titanium dioxide nanotubes addition to self-adhesive resin cement: Effect on physical and biological properties. Dent Mater. 2017;33(7):866–75.
- 22. Carp O, Huisman CL, Reller A. Photoinduced reactivity of titanium dioxide. Prog solid state Chem. 2004;32(1–2):33–177.
- Koziej D, Fischer F, Kranzlin N, Caseri WR, Niederberger M. Nonaqueous TiO2 nanoparticle synthesis: a versatile basis for the fabrication of selfsupporting, transparent, and UV-absorbing composite films. ACS Appl Mater Interfaces. 2009;1(5):1097–104.
- Hatamleh MM, Watts DC. Fibre reinforcement enhances bonding of soft lining to acrylic dental and maxillofacial prostheses. Eur J Prosthodont Restor Dent. 2008;16(3):116–21.
- Method ST. Standard Test Method for Impact Strength of Adhesive Bonds 1.
 Test. 2003;01(Reapproved):5–8.
- Hatamleh MM, Watts DC. Bonding of maxillofacial silicone elastomers to an acrylic substrate. Dent Mater. 2010;26(4):387–95.
- Shetty US, Guttal SS. Evaluation of bonding efficiency between facial silicone and acrylic resin using different bonding agents and surface alterations. J Adv Prosthodont. 2012;4(3):121–6.

- Chang PP, Hansen NA, Phoenix RD, Schneid TR. The effects of primers and surface bonding characteristics on the adhesion of polyurethane to two commonly used silicone elastomers. J Prosthodont Implant Esthet Reconstr Dent. 2009;18(1):23–31.
- AlHamdan EM, Al-Saleh S, AlRefeai MH, Farooq I, Abrar E, Vohra F, et al. Adhesive bond integrity of Y-TZP post with calcium fluoride infiltrated resin dentin adhesive: An SEM, EDX, FTIR and micro-Raman study. Surf Interface Anal. 2021;53(11):956–62.
- Alqarawi FK, Alkahtany MF, Almadi KH, Ben Gassem AA, Alshahrani FA, AlRefeai MH, et al. Influence of different conditioning treatments on the bond integrity of root dentin to rGO infiltrated dentin adhesive. SEM, EDX, FTIR and microRaman study. Polymers (Basel). 2021;13(10):1555.
- Al-Saleh S, Alateeq A, Alshaya AH, Al-Qahtani AS, Tulbah HI, Binhasan M, et al. Influence of TiO2 and ZrO2 nanoparticles on adhesive bond strength and viscosity of dentin polymer: A physical and chemical evaluation. Polymers (Basel). 2021;13(21):3794.
- Torres-Mendez F, Martinez-Castanon G-A, Torres-Gallegos I, Zavala-Alonso N-V, Patino-Marin N, Nino-Martinez N, et al. Effects of silver nanoparticles on the bonding of three adhesive systems to fluorotic enamel. Dent Mater J. 2017;2015–299.
- Bardak T, Tankut AN, Tankut N, Sozen E, Aydemir D. The effect of nano-TiO2 and SiO2 on bonding strength and structural properties of poly (vinyl acetate) composites. Measurement. 2016;93:80–5.
- Silvestre J, Silvestre N, De Brito J. Polymer nanocomposites for structural applications: Recent trends and new perspectives. Mech Adv Mater Struct. 2016;23(11):1263–77.

- 35. Kim J, Cho B, Lee I, Um C, Lim B, Oh M, et al. Effect of the hydrophilic nanofiller loading on the mechanical properties and the microtensile bond strength of an ethanol-based one-bottle dentin adhesive. J Biomed Mater Res Part B Appl Biomater An Off J Soc Biomater Japanese Soc Biomater Aust Soc Biomater Korean Soc Biomater. 2005;72(2):284–91.
- Sadat-Shojai M, Atai M, Nodehi A, Khanlar LN. Hydroxyapatite nanorods as novel fillers for improving the properties of dental adhesives: Synthesis and application. Dent Mater. 2010;26(5):471–82.
- Solhi L, Atai M, Nodehi A, Imani M, Ghaemi A, Khosravi K. Poly (acrylic acid) grafted montmorillonite as novel fillers for dental adhesives: synthesis, characterization and properties of the adhesive. Dent Mater. 2012;28(4):369–77.
- 38. Wagner A, Belli R, Stotzel C, Hilpert A, Muller FA, Lohbauer U. Biomimetically-and hydrothermally-grown HAp nanoparticles as reinforcing fillers for dental adhesives. J Adhes Dent. 2013;15(5):413–22.

Table (1) Means and standard deviation of the shear strength of the nano-filled modified primer G611 for the cold cure and 3D printed resin groups.

Bonding strengths (shear and peel) Mean (SD) **Titanium** Ν Shear bond strength Shear bond strength dioxide of the self-cure resin of the 3D printed resin Nanoparticles (%) (N/mm^2) (N/mm^2) Control (0%) 3.568 (0.244) a 2.785 (0.242) a 9 6.188 (0.223)b 5% 9 4.330 (0.409) b 8.371 (0.368) c 7.234 (0.332) c 10% 5.128 (0.273) d 15% 9 2.520 (0.489) a 20% 9 2.639 (0.419) e 1.857 (0.437) d

N: Number of samples, SD: Standard deviation. Means labeled with different letters (a-e) indicate a significant difference among the means within one column.

Table (2) Means and standard deviation of the peel strength of the nano-filled modified primer G611 for the cold cure and 3D printed resin groups.

		Mean (SD)	
		Peel bond strength	Peel bond strength
Titanium dioxide	N	of the self-cure resin	of the 3D printed resin
Nanoparticles		(N/mm²)	(N/mm^2)
(%)			
Control (0%)	9	5.201(0.161) a	4.874 (0.258) a
5%	9	8.183(0.319) b	8.252 (444) ^b
10%	9	9.440.(0.398) °	9.733 (0.344) °
15%	9	4.874(0.607) a	4.789 (0.418) ^a
20%	9	3.425(0.298) ^d	1.651 (0.336) ^d

N: Number of samples, SD: Standard deviation. Means labeled with different letters (a-d) indicate a significant difference among the means within one column.