Iranian Journal of Materials Science and Engineering, Vol. 20, Number 3, September 2023

RESEARCH PAPER

Effect of Incorporation of Amorphous Calcium Phosphate on Bonding Strength of a Dental Adhesive

Ali Hasanzade Salmasi[1], Mahban Zarei[2], Shadab Safarzadeh Khosroshahi[3,*], Soolmaz Heidari[4], Farhood Najafi[5], Mojtaba Ghomayshi[6], Katayoun Lesani[7]

* *shadab.kh@gmail.com*

[1] Postgraduate Student, Department of Endodontics, School of Dentistry, Shahid Sadoughi University of Medical Sciences, Yazd, Iran
[2] DDS Candidate, University of Pacific, Arthur Dugoni School of Dentistry, San Francisco, USA
[3] Department of Operative Dentistry, Dental Material Research Center, Islamic Azad University of Medical Science, Tehran, Iran
[4] Department of Operative Dentistry, Dental Caries Prevention Research Center, Qazvin University of Medical Sciences, Qazvin, Iran
[5] Department of Resin and Additives, Institute for Color Science and Technology, P.O. Box: 16765-564, Tehran, Iran
[6] Student Research Committee, Qazvin University of Medical Sciences, Qazvin, Iran
[7] Postgraduate Student, Department of Oral and Maxillofacial Radiology, School of Dentistry, Shahid Sadoughi University of Medical Sciences, Yazd, Iran

Received: March 2023 Revised: September 2023 Accepted: September 2023
DOI: 10.22068/ijmse.3203

Abstract: *Amorphous calcium phosphate (ACP) which is a transient phase in the natural bio-mineralization process has recently gained the spotlight. This study aimed to assess the effect of the incorporation of nano-ACP (NACP) in a dental adhesive with/without surface treatment with silane coupling agent on bond strength. NACP was synthesized by the wet chemical precipitation technique. To characterize the structure of NACP, X-ray diffraction, scanning electron microscopy, and energy-dispersive X-ray spectroscopy were used. Forty molars were randomized into 4 groups of 10. The teeth were restored with composite resin and the bonding agent (one of the four groups). Adper Single Bond 2 was used as the control group. In the 4 wt% NACP group, NACP fillers were added to the bonding agent. In 0.4 wt% and 4 wt% SNACP groups, silanized NACP fillers were added to the bonding agent. Finally, the mode of failure of specimens was determined. Data were analyzed by one-way ANOVA and Tukey's post-hoc tests. P< 0.05 was considered statistically significant. The addition of 4 wt% non-silanized NACP decreased the bond strength compared with the control group (P< 0.05). The bond strength of the groups with silanized fillers was not significantly different from that of the control group. The addition of silanized NACP to dental adhesive had no significant adverse effect on bond strength, which is a promising finding to pave the way for the synthesis of bonding agents containing bioactive fillers.*

Keywords: *Amorphous Calcium Phosphate, Silane, Bond Strength.*

1. INTRODUCTION

Bioactive materials are products that form a hydroxyapatite layer in contact with physiological body fluids [1]. Of bioactive materials, calcium phosphate compounds are of particular interest. Calcium phosphate ceramics have long been used in tissue engineering as bone graft materials due to their optimal osteoconductivity and excellent biocompatibility. Crystalline calcium phosphates are synthetic biomaterials with a structure and composition similar to the mineral phase of bone and teeth. Thus, they form optimal chemical bonds with the mineral tissue in the human body [2, 3]. On the other hand, some studies showed that amorphous calcium phosphate (ACP) has a higher remineralizing ability than other crystalline calcium phosphates such as hydroxyapatite and tricalcium phosphate [4, 5]. Perhaps one reason for this issue is the higher solubility of the amorphous calcium phosphate, which leads to the faster release of calcium and phosphate ions. The disordered and non-crystalline structure of ACP is responsible for more solubility and faster degradation. [6, 7]. ACP is not a stable component of tooth structure or bone. It is a transient phase in the process of natural biomineralization which is converted to hydroxyapatite [4, 8]. At present, it is commonly used for the enhancement of enamel remineralization. ACP is available in a stable form in combination with casein phosphopeptide (CPP-ACP) or as an unstable compound in toothpaste. Moreover, it is used as a

[DOI: 10.22068/ijmse.3203]

cariostatic, and remineralizing agent in polymer composites, bleaching agents, and mouthwashes [8]. The majority of restorative materials available in the market have inert fillers, which do not elicit any physiological response (such as hydroxyapatite formation) in the host. The release of calcium and phosphate ions from the fillers increases the durability of bonded restorations [9]. The released calcium and phosphate ions can deposit again obstruct the nano-sized voids and prevent nano-leakage through the interface. These ions are released in greater amounts at a pH of 4, which is an advantage for products containing calcium phosphate to prevent demineralization [10]. Evidence shows that the addition of ACP by up to 40% has no significant effect on immediate or delayed (after 28 days) bond strength in aqueous environment [10, 11]. Zhang et al. [10] indicated that resin-based materials containing 20-30% nano-ACP (NACP) enable considerable release of calcium and phosphate ions. They used 10% hydroxyethyl methacrylate to enhance the properties of the bonding agent containing ACP. The solubility and degradation rate of dental adhesives are related to some factors including the composition of the adhesive and curing process [12]. The low solubility of dental adhesives with a mainly hydrophobic resin structure is an advantage due to the prevention of microleakage in the future [13, 14]. On the other hand, when bioactive fillers are added to the basic composition of adhesives, they may not be released easily due to being trapped in the resin structure with low solubility [15]. In this study, amorphous calcium phosphate was used instead of other crystalline counterparts, with the assumption that its release may be higher due to higher solubility and perhaps higher remineralizing ability. Furthermore, silane coupling agent monomer (3-methacryloxypropyltrimethoxysilane) was used for the preparation of ACP fillers in this study [16]. It has been confirmed that silanization of fillers, especially SiO_2 fillers in the resin matrix increases the physical and mechanical properties of composite resins [17, 18]. Silane can couple the resin matrix with an inorganic component since it contains two or more functional groups [18]. Lung et al. [19] indicated that silanization of hydroxyapatite fillers incorporated into a composite resin improved the surface hardness and decreased the water sorption of the composite. Thus, this study aimed to assess the effect of the incorporation of silanized NACP fillers on the bond strength of a commercial dental adhesive.

2. EXPERIMENTAL PROCEDURES

2.1. NACP Synthesis

NACP was synthesized as explained by Karimi et al [20]. The required precursors were purchased from Merck kGaA (Darmstadt, Germany). A deep eutectic solvent was used for the synthesis of NACP. In brief, choline chloride (ChCl) and urea were mixed in a 1:2 molar ratio and heated at 100°C to obtain a homogenous liquid. Next, 0.67 g of $CaCl_2$ was dissolved in 50 cc of the solvent. Afterward, 0.81 M K_2HPO_4 aqueous solution was immediately added to the solution to acquire a gel-like suspension with a calcium/phosphorous molar ratio of 1.5. The obtained suspension was stirred for 20 min, filtered, and rinsed with distilled water containing 0.001% of v/v 25% ammonia solution several times. The obtained suspension was freeze-dried (Pishtaz Equipment Engineering, DKLD-150-CAG) for 65 h at -67°C and 0.5 Torr pressure, and kept in a freezer until the experiment. X-ray diffraction (XRD, Siemens D-500 diffractometer, Cu-Kα radiation, λ= 1.5406A°) was performed to confirm that the obtained material is amorphous.

2.2. Silanization of NACP

For this purpose, 3-(Methacryloyloxy) propyltrimethoxysilane (Silane A 174, Merck. KGaA, Darmstadt, Germany) with the following molecular structure was used to modify the surface of NACP particles (Fig. 1).

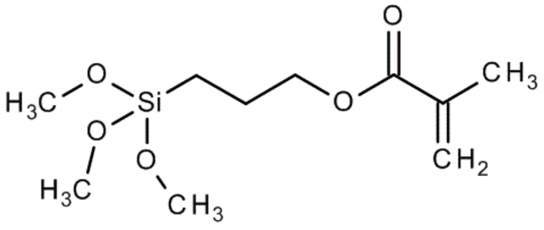

Fig. 1. Chemical structure of Silane A 174 [21]

In brief, 1 g of silane was dissolved in 10 cc of 70% ethanol. Next, 9 g of NACP was added to the solution, and it was ultrasonicated for 6 min (SonoPlus-UW 2200, Germany). Afterward, 0.1 cc of 25% ammonia solution was added and the sol-gel process was carried out in an alkaline

[DOI: 10.22068/ijmse.3203]

environment for binding of NACP to silane. This process can be explained as follows:

1- Sol: Hydrolysis of silane groups in an alkaline medium

2- Gel: condensation of hydroxyl groups in silane with each other and with hydroxyl groups of ACP along with liberation of water as by product (Fig. 2)

The modified deposits were filtered by a filter paper and rinsed with ethanol twice. The obtained substance was then dried at room temperature. All products were purchased from Merck (Merck. KGaA, Darmstadt, Germany).

Adper Single Bond 2 (3M ESPE; St Paul, MN, USA) bonding agent served as the control group in this study to evaluate the effect of incorporation of NACP and silanized NACP (SNACP) on bonding strength. The experimental groups are described in Table 1.

2.3. Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray (SEM/EDS) Analysis

SEM analysis (VEGA; TESCAN-XMU, Czech Republic) was performed to determine the morphology and size of particles; while, EDS (Mira 3-XMU, Czech Republic) was carried out for elemental analysis.

The accelerating voltage was 5 kV for SEM and 15 kV for EDS. The specimens were gold sputter-coated for electrical conduction and reduction of charge when observing the image of particles. Since phosphorous interferes with gold in the conduction of EDS, the gold coating was not performed for EDS. EDS analysis was performed at three random points of each specimen.

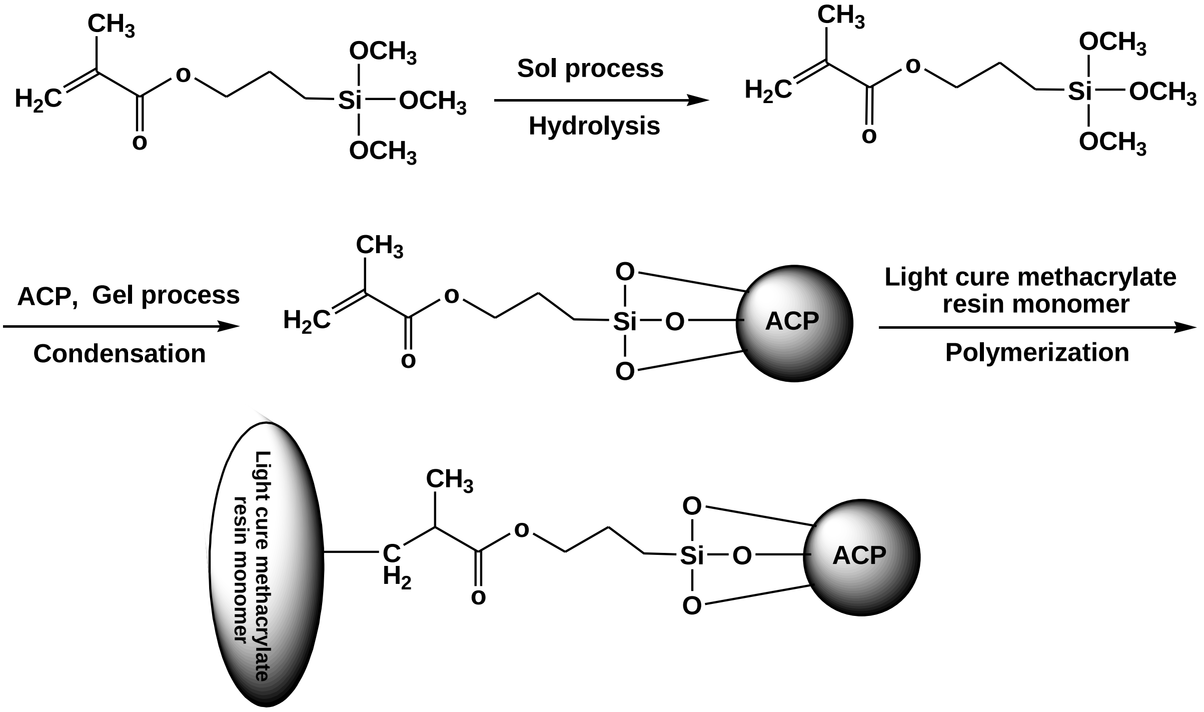

Fig. 2. The mechanism of silane function is schematically explained [22]

Table 1. Groups and their compositions

Groups	Composition
Adper Single Bond 2 (manufacture: 3M ESPE; St Paul, MN, USA)	BisGMA, HEMA, dimethacrylates, ethanol, water, photoinitiator system, methacrylate functional copolymer of polyacrylic and polyitaconic acids (according to the manufacturer's information)
4 wt% NACP	0.24 g of NACP was added to 6 g of Adper Single Bond 2 and ultrasonicated for 3 minutes.
4 wt% SNACP	0.24 g of silanized NACP was added to 6 g of Adper Single Bond 2 and ultrasonicated for 3 minutes.
0.4% SNACP	0.024 g of silanized NACP was added to 6 g of Adper Single Bond 2 and ultrasonicated for 3 minutes.

[DOI: 10.22068/ijmse.3203]

2.4. Microtensile Bond Strength Testing

Forty sound human molars were collected and visually inspected to ensure the absence of carious lesions, restorations, fractures, cracks, or anomalies. The teeth were immersed in chloramine T solution for one week and stored in distilled water at 4°C until the experiment. The occlusal enamel was then cut by a low-speed handpiece and a double-blade diamond disc (Fig. 3 A, B). The tooth surface was then polished with 600-grit silicon carbide abrasive paper. A matrix band was applied and dentin was etched with 37% phosphoric acid gel (Condac 37; FGM, Joinville, Brazil) for 15 s as recommended by the manufacturer. It was then rinsed under running water for 15 s. The teeth were then gently air-dried. The bonding agent was applied as recommended by the manufacturer in all groups. For this purpose, two coats of adhesive were applied on the dentin surface using a micro brush. The adhesive was gently thinned with air spray, and light-curing was performed for 20 s by a curing unit (Coltolux II, Coltene) with a light intensity of 600 mW/cm^2. Z250 composite resin (A2 shade; 3M ESPE, St. Paul, MN, USA) was then incrementally applied on the tooth surface. Five 1-mm-thick increments of the composite were applied and cured for 40 s as recommended by the manufacturer (Fig. 3 C, D). The matrix band was then removed and the teeth were incubated in distilled water (Memmert; Schwabach, Germany) at 37°C until the experiment.

The teeth were sectioned into microbars (8 x 1 x 1 mm^3) by a microtome with a 0.3-mm blade (Presi, France) (Fig. 3 E, F). The microbars underwent 5000 thermal cycles (TC-300, Vafaei Industrial, Iran) between 5-55±2°C with a dwell time of 20 s and a transfer time of 10 s [23]. The microtensile bond strength was measured by a microtensile tester (Bisco Inc., Schaumburg, IL, USA). The specimens were bonded to the jig with cyanoacrylate glue (Nano Bond 123, Turkey). Each specimen was adjusted in such a way that the interface area was placed exactly in the middle of the gap between the two jaws. The tensile load was applied at a crosshead speed of 0.5 mm/min until failure (Fig. 3 G, H). Data were separately recorded for each group, and the microtensile bond strength was calculated in megapascals (MPa) by dividing the load at fracture in Newtons by the cross-sectional area in square millimeters.

Finally, the specimens were inspected under a stereomicroscope (Omax 40X-2000X Microscope, China) to determine the mode of failure. The mode of failure was categorized as an adhesive (at the substrate-adhesive interface or within the adhesive layer), cohesive (within dentin or composite resin), or mixed (a combination of adhesive and cohesive failures) [25].

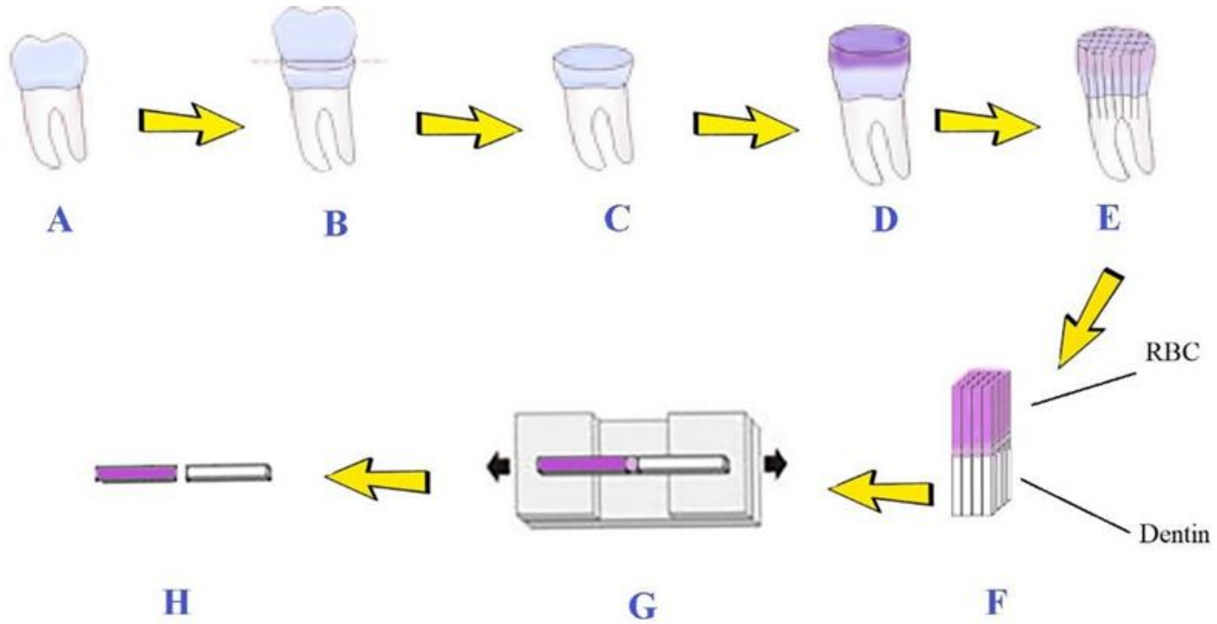

Fig. 3. The process of the microtensile bond strength test is shown schematically from A to H (RBC: resin-base composite) [24].

[DOI: 10.22068/ijmse.3203]

Statistical analyses:

Data were analyzed by SPSS version 21 (SPSS Inc., IL, USA) using one-way ANOVA and Tukey's post-hoc tests. The normality of data distribution was evaluated by the Kolmogorov-Smirnov test (P >0.05). P< 0.05 was considered statistically significant.

3. RESULTS AND DISCUSSION

This study sought to evaluate the changes in microtensile bond strength of Adper Single Bond 2 following the incorporation of NACP as filler. The XRD pattern of NACP revealed an amorphous pattern with a wide peak at 25-35° indicative of ACP [20] (Fig. 4).

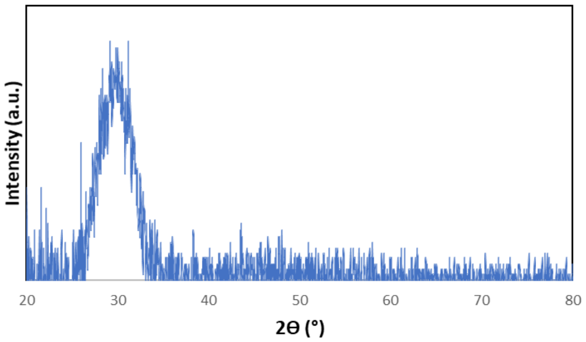

Fig. 4. XRD pattern of amorphous calcium phosphate

The incorporation of high amounts of fillers into dental adhesives increases their viscosity and prevents adequate penetration of resin into dental porosities. Thus, a limited amount (<10%) of fillers can be added to dental adhesives in order not to compromise the bond strength [26, 27]. Adper Single Bond 2 (3M ESPE), which is a 5th generation etch-and-rinse dentin bonding agent was used in this study, which contains 10 wt% spherical silica nanoparticles as filler measuring 5 nm in diameter [28]. The presence of optimal filler content in dental adhesives improves their bond strength and mechanical properties.

The intermediate adhesive layer along with the hybrid layer decreases the adverse effects of polymerization shrinkage and enables optimal distribution of stress following load application [27]. Controversy exists regarding the optimal amount of bioactive fillers such as hydroxyapatite for incorporation into commercial dental adhesives without compromising their bond strength. Leitune et al. [29] indicated that the addition of 2 wt% hydroxyapatite significantly increased the bond strength. However, Kavrik and Kucukyilmaz [26] reported that the addition of 7% hydroxyapatite to a bonding agent yielded the maximum bond strength. In the present study, 4 wt% NACP was the maximum amount of added filler. The microtensile test was performed for the measurement of bonding strength in this study, which has a lower rate of cohesive fracture than other tests, and enables the measurement of bonding strength in very small areas [30].

Table 2 presents the microtensile bonding strength of the four groups. The bonding strength of the control group was significantly higher than that of 4% NACP group (P< 0.05). Also, the bonding strength of both silanized groups was significantly higher than that of the 4% NACP group (P< 0.05). In the present study, the results indicated significantly lower bonding strength of the 4 wt% NACP group than the control group (pure bonding agent), which may be due to the addition of excessive amounts of filler. The cumulative effects of the two fillers present in the dental adhesive with 4 wt% NACP (10 wt% nano-silica fillers and 4 wt% NACP fillers) increase the viscosity of the bonding agent and results in the formation of filler clusters on the dentin surface, which prevents optimal penetration of resin into the inter-tubular spaces and dentinal tubules. However, the 4 wt% SNACP groups had no significant difference with the control group in bond strength. The 4 wt% SNACP group showed a significant difference from the 4 wt% NACP group in bond strength. The higher bond strength of silanized groups may be due to the properties of silane. Organosilanes, especially the functional types, have a unique chemical structure for surface modification of mineral substrates.

Table 2. Comparison of the microtensile bonding strength between four groups. The differences between 4 %wt NACP with other groups were significant p< 0.05).

Group	Mean (MPa)	Standard deviation
Adper Single Bond 2 (control group)	34.3	7.9
4 %wt NACP	23.8	7.3
0.4 %wt SNACP	34.4	9.4
4 %wt SNACP	32.6	10.9

[DOI: 10.22068/ijmse.3203]

Silane has a coupling action and can form a chemical bond between the organic and mineral phases, and improve the polymer properties as such [31]. Silane contains silanol groups, which react with mineral fillers. It also has organofunctional groups, which are co-polymerized with methacrylate monomers in the process of curing [32]. Silane increases the mechanical properties and hydrolytic stability [18]. Agglomeration of filler nanoparticles compromises the mechanical properties of dental adhesives. Silanization of filler particles decreases their surface energy and increases the surface wetting. It has been demonstrated that in case of not using silane, agglomeration can occur in the presence of 1 wt% filler while following the addition of silane, this rate reaches to over 5 wt% filler. In case of no silanization of fillers, a gap forms at the filler-resin matrix interface, which can cause bond failure in non-silanized samples. Nonetheless, the amount and size of filler particles can affect the surface/volume ratio of fillers and are important factors that play pivotal roles in the reinforcement of dental adhesives by the filler particles [33]. The higher the filler-resin matrix bond strength, the lower the water sorption, and the higher the hydrolytic stability would be [8, 34]. Studies on the quality of composite resins containing ACP fillers have indicated that in comparison with silanized glass fillers, ACP fillers with their hydrophilic and biodegradable properties can lead to water sorption in the long term and decrease the bonding durability. Also, the agglomeration of ACP particles and their poor adhesion to the resin matrix can compromise the mechanical properties of these composites and increase their water sorption at the interface [35]. Thus, surface treatment of ACP bioactive fillers should be considered for their incorporation in dental adhesives.

In the present study, the addition of silane resulted in a higher bond strength and indicated that if a higher percentage of fillers is to be added to the bonding agent, they can be silanized to ensure a higher bond strength.

Some studies have shown that up to 40 wt% NACP can be added to commercial bonding agents without compromising their bond strength. This controversy may be due to the use of this filler along with some other components such as a new monomer or the use of different types of bonding agents [11, 36-38]. However, some other studies discussed that non-silanized calcium phosphate fillers can only be added to bonding agents in small amounts in order not to compromise the bond strength [29, 39]. Although the addition of high amounts of mineral fillers may not compromise the bond strength, it may decrease the degree of conversion due to incompatibility of the refractive index of fillers and resin matrix, and affect the physical and mechanical properties of the bonding layer, irrespective of the use of silane for filler preparation [40, 41].

Figure 5 presents the SEM micrographs and the results of EDS analysis of NACP and SNACP. Agglomerated spherical calcium phosphate nanoparticles with a rough surface and porosities in some areas were seen more in the NACP group compared to the 4 wt% SNAP group. The size of NACP particles ranged from 20 to 50 nm. The EDS analysis revealed the presence of calcium (Ca), phosphorous (P), and oxygen (O) elements in NACP and 4 wt% SNACP. The Ca/P molar ratio was close to 1.5. The presence of Si and C in SNACP group is related to the silane treatment.

As shown by SEM assessment, the size of ACP filler particles was in the range of nano-scale. The use of nano-sized particles enhances the mechanical properties and can lead to greater release of calcium and phosphate ions due to increased surface area. On the other hand, nano-sized particles can better lodge into the porosities created by etching [26].

The results of fractographic analysis are reported in Table 3. Figure 6 shows adhesive and cohesive failure patterns. Evaluation of the mode of failure of specimens following the micro-tensile bond strength test indicated that

Table 3. Modes of failure in different groups (%).

Group	Adhesive	Cohesive	Mixed
Adper Single Bond 2	75	15	10
4% NACP	74	12	14
0.4%wt SNACP	53	42	5
4%wt SNACP	75	8	17

[DOI: 10.22068/ijmse.3203]

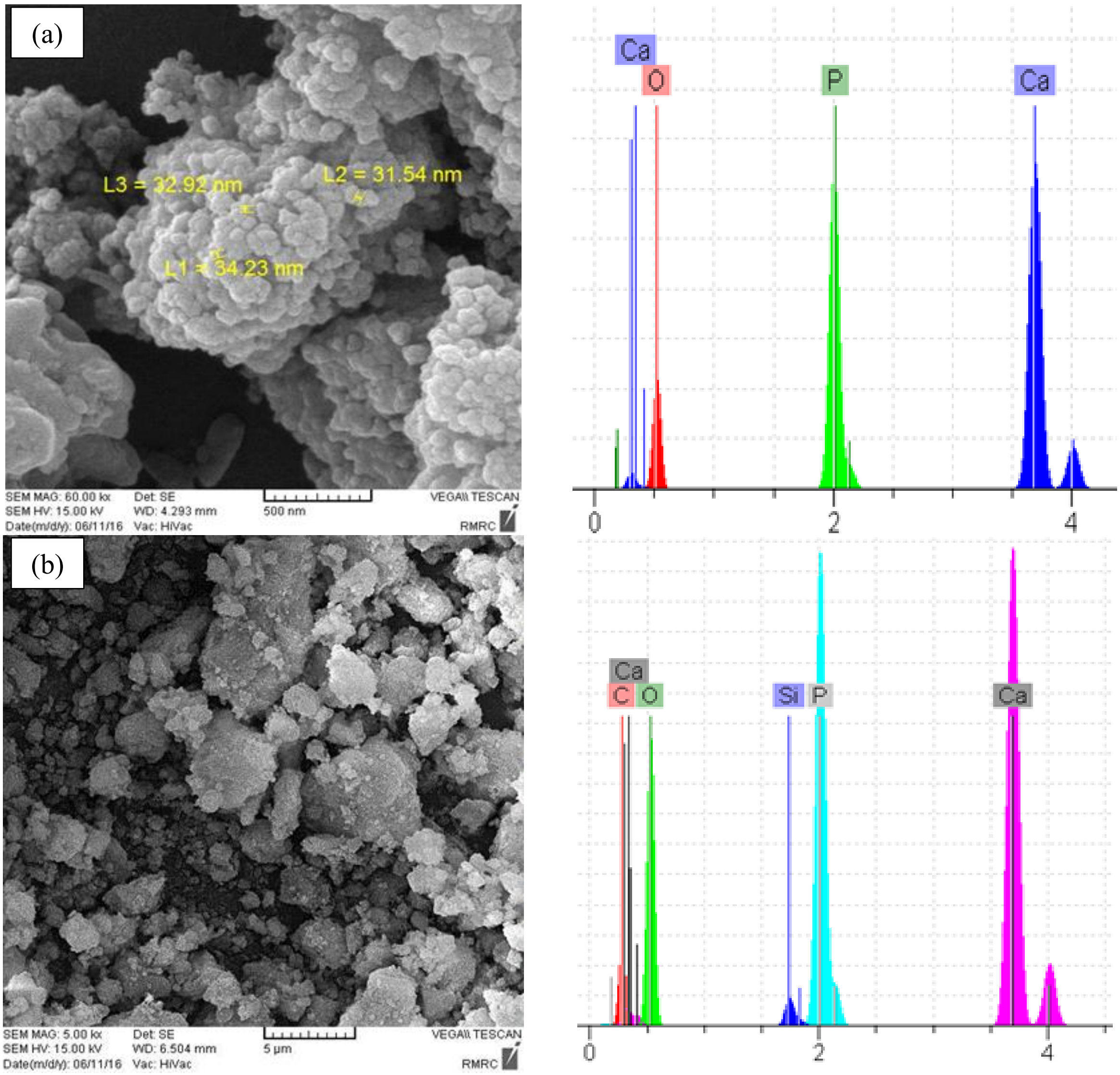

Fig. 5. SEM/EDS analyses of (a) NACP and (b) 4 wt% SNACP

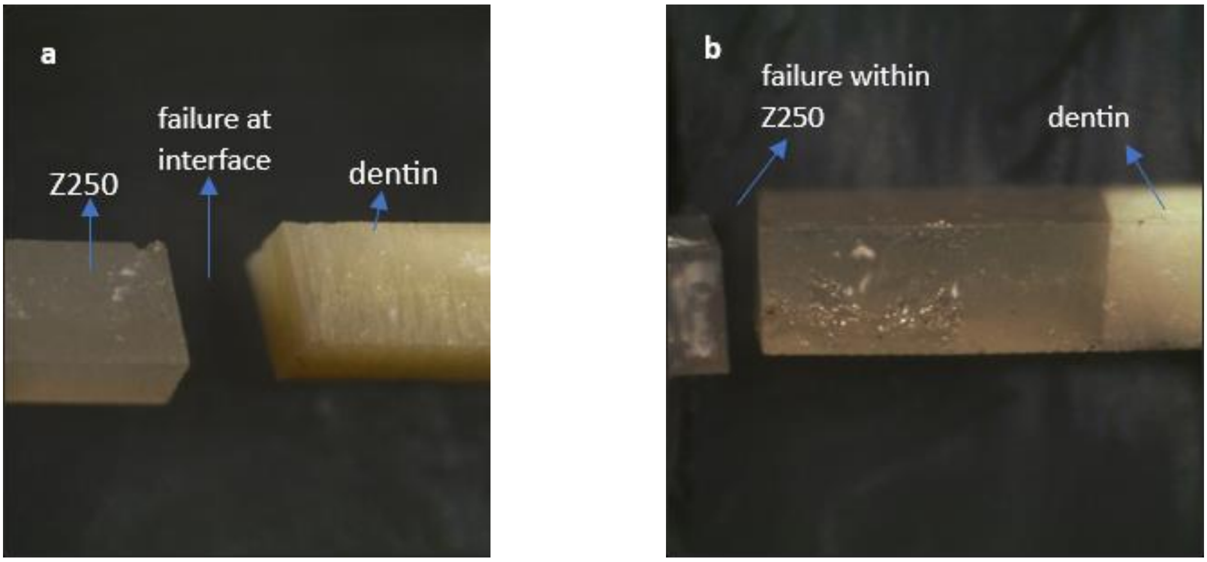

Fig. 6. The modes of failure a (adhesive) and b (cohesive)

[DOI: 10.22068/ijmse.3203]

The adhesive failure was the most frequent mode of failure. However, cohesive failure had a higher frequency in 0.4% SNACP group, compared with other groups. Cohesive failure is probably due to the excessively high bond strength. Strong bond to dentin results in higher frequency of failures within the weaker substrate [42] as noted in 0.4% SNACP group, which had the maximum bond strength.

4. CONCLUSIONS

Adding calcium phosphate fillers without silanization significantly reduced the bond strength. The addition of 4 wt% silanized NACP to Adper Single Bond 2 did not compromise the bond strength. It yielded a bond strength comparable to that of the control group. Calcium phosphate fillers may have the ability to release calcium and phosphate ions in an aqueous environment. These ions can prevent tooth demineralization and help the remineralization process to occur. However, other analyses are needed to evaluate the release of these ions and their efficacy in the remineralization process. Also, microleakage and bonding durability should be further investigated in future studies. This is a promising finding to pave the way for the synthesis of bonding agents containing bioactive fillers.

REFERENCES

[1]. Almulhim KS, Syed MR, Alqahtani N, Alamoudi M, Khan M, Ahmed SZ, Khan AS. Bioactive Inorganic Materials for Dental Applications: A Narrative Review. Materials. 2022; 15(19):6864.

[2]. Tang Z, Li X, Tan Y, Fan H, Zhang X. The material and biological characteristics of osteoinductive calcium phosphate ceramics. Regen Biomater. 2018 Feb; 5(1):43-59.

[3]. Al-Sanabani JS, Madfa AA, Al-Sanabani FA. Application of calcium phosphate materials in dentistry. Int J Biomater. 2013; 2013:876132.

[4]. Zhao J, Liu Y, Sun WB, Zhang H. Amorphous calcium phosphate and its application in dentistry. Chem Cent J. 2011 Jul 8; 5:40.

[5]. Zhao J, Liu Y, Sun W-B, Yang X. First detection, characterization, and application of amorphous calcium phosphate in dentistry. J Dent Sci. 2012; 7(04):316–323.

[6]. Vecstaudza J, Gasik M, Locs J. Amorphous calcium phosphate materials: Formation, structure and thermal behaviour. J. Eur. Ceram. Soc. 2019 Apr; 39:1642– 1649

[7]. Mok ZH, Mylonas P, Austin R, Proctor G, Pitts N, Thanou M. Calcium phosphate nanoparticles for potential application as enamel remineralising agent tested on hydroxyapatite discs. Nanoscale. 2021 Dec 13; 13(47):20002-20012.

[8]. Diane R. Bienek, Anthony A. Giuseppetti and Drago Skrtic (July 2nd 2019). Amorphous Calcium Phosphate as Bioactive Filler in Polymeric Dental Composites, Contemporary Topics about Phosphorus in Biology and Materials, David G. Churchill, Maja Dutour Sikirić, Božana Čolović and Helga Füredi Milhofer, IntechOpen.

[9]. Braga RR. Calcium phosphates as ion-releasing fillers in restorative resin-based materials. Dent Mater. 2019 Jan; 35(1):3-14.

[10]. Zhang L, Weir MD, Hack G, Fouad AF, Xu HH. Rechargeable dental adhesive with calcium phosphate nanoparticles for long-term ion release. J Dent. 2015 Dec;43(12):1587-95

[11]. Chen C, Weir MD, Cheng L, Lin NJ, Lin-Gibson S, Chow LC, Zhou X, Xu HH. Antibacterial activity and ion release of bonding agent containing amorphous calcium phosphate nanoparticles. Dent Mater. 2014 Aug; 30(8):891-901.

[12]. Moreira Fdo C, Antoniosi Filho NR, Souza JB, Lopes LG. Sorption, solubility and residual monomers of a dental adhesive cured by different light-curing units. Braz Dent J. 2010; 21(5):432-8.

[13]. Sofan E, Sofan A, Palaia G, Tenore G, Romeo U, Migliau G. Classification review of dental adhesive systems: from the IV generation to the universal type. Ann Stomatol (Roma). 2017 Jul 3; 8(1):1-17.

[14]. Gupta A, Tavane P, Gupta PK, Tejolatha B, Lakhani AA, Tiwari R, Kashyap S, Garg G. Evaluation of Microleakage with Total Etch, Self Etch and Universal Adhesive Systems in Class V Restorations: An In vitro Study. J Clin Diagn Res. 2017 Apr;

11(4):ZC53-ZC56.
[15]. Kasraei S, Haghi S, Valizadeh S, Panahandeh N, Nejadkarimi S. Phosphate Ion Release and Alkalizing Potential of Three Bioactive Dental Materials in Comparison with Composite Resin. Int J Dent. 2021 May 7; 2021: 5572569.
[16]. Weir MD, Chow LC, Xu HH. Remineralization of demineralized enamel via calcium phosphate nanocomposite. J Dent Res. 2012 Oct; 91(10):979-84.
[17]. Yon MJY, Matinlinna JP, Tsoi JKH, Vallittu PK, Lassila LVJ. Effect of long-chain silane on mechanical properties of experimental resin composites. Research Square; 2023.
[18]. Matinlinna JP, Lung CYK, Tsoi JKH. Silane adhesion mechanism in dental applications and surface treatments: A review. Dent Mater. 2018 Jan; 34(1):13-28.
[19]. Lung CY, Sarfraz Z, Habib A, Khan AS, Matinlinna JP. Effect of silanization of hydroxyapatite fillers on physical and mechanical properties of a bis-GMA based resin composite. J Mech Behav Biomed Mater. 2016 Feb; 54:283-94
[20]. M. Karimi, S. Hesaraki, M. Alizadeh, A. Kazemzadeh, Synthesis of calcium phosphate nanoparticles in deep-eutectic choline chloride-urea medium: investigating the role of synthesis temperature on phase characteristics and physical properties, Ceram. Int. 42 (2016) 2780–2788.
[21]. Merck Millipore (Merck KGaA, Darmstadt, Germany) 2023, https://www.merckmillipore.com/DE/de/product/Silane-A-174,MDA_CHEM-107673?ReferrerURL=https%3A%2F%2Fwww.google.com%2F
[22]. Rawls HR, Whang K, Resin-Based Composites. In Shen Ch, Rawls HR, Esquivel-Upshaw JF. Philip's Science of Dental Materials. 13th ed. St. Louis, Missouri: Elsevier; 2022. P. 248
[23]. Morresi AL, D'Amario M, Capogreco M, Gatto R, Marzo G, D'Arcangelo C, Monaco A. Thermal cycling for restorative materials: does a standardized protocol exist in laboratory testing? A literature review. J Mech Behav Biomed Mater. 2014 Jan; 29:295-308.
[24]. Hass V, Folkuenig MS, Reis A, Loguercio AD. Influence of adhesive properties on resin-dentin bond strength of one-step self-etching adhesives. J Adhes Dent. 2011 Oct; 13(5):417-24.
[25]. Tichy A, Brabec M, Bradna P, Hosaka K, Tagami J. A competing risk model for bond strength data analysis. Dent Mater. 2020 Dec; 36(12):1508-1515.
[26]. Kavrik F, Kucukyilmaz E. The effect of different ratios of nano-sized hydroxyapatite fillers on the micro-tensile bond strength of an adhesive resin. Microsc Res Tech. 2019 May; 82(5):538-543.
[27]. Azad E, Atai M, Zandi M, Shokrollahi P, Solhi L. Structure-properties relationships in dental adhesives: Effect of initiator, matrix monomer structure, and nano-filler incorporation. Dent Mater. 2018 Sep; 34(9):1263-1270.
[28]. 3M ESPE Dental Products (2004), https://multimedia.3m.com/mws/media/276868O/adper-single-bond-2-technical-profile.pdf.
[29]. Leitune VC, Collares FM, Trommer RM, Andrioli DG, Bergmann CP, Samuel SM. The addition of nanostructured hydroxyapatite to an experimental adhesive resin. J Dent. 2013 Apr; 41(4):321-7.
[30]. Fernandes Jr VVB, Oliani MG, Nogueira Jr L, Silva JMF, Araújo RM (2016) Analysis and Comparison of Different Bond Strength Tests. JSM Dent 4(5): 1076.
[31]. Antonucci JM, Dickens SH, Fowler BO, Xu HH, McDonough WG. Chemistry of Silanes: Interfaces in Dental Polymers and Composites. J Res Natl Inst Stand Technol. 2005 Oct 1; 110(5):541-58.
[32]. Michelotti G, Niedzwiecki M, Bidjan D, Dieckmann P, Deari S, Attin T, Tauböck TT. Silane Effect of Universal Adhesive on the Composite-Composite Repair Bond Strength after Different Surface Pretreatments. Polymers (Basel). 2020 Apr 19; 12(4):950.
[33]. Wagner A, Belli R, Stötzel C, Hilpert A, Müller FA, Lohbauer U. Biomimetically- and hydrothermally-grown HAp nanoparticles as reinforcing fillers for dental adhesives. J Adhes Dent. 2013 Oct; 15(5):413-22.

[DOI: 10.22068/ijmse.3203]

[34]. Szczesio-Wlodarczyk A, Sokolowski J, Kleczewska J, Bociong K. Ageing of Dental Composites Based on Methacrylate Resins-A Critical Review of the Causes and Method of Assessment. Polymers (Basel). 2020 Apr 10; 12(4):882.

[35]. Zhao J, Liu Y, W-b Sun, Yang X (2012) First detection, characterization, and application of amorphous calcium phosphate in dentistry. J Dent Sci 7(4):316–323.

[36]. Melo MA, Cheng L, Zhang K, Weir MD, Rodrigues LK, Xu HH. Novel dental adhesives containing nanoparticles of silver and amorphous calcium phosphate. Dent Mater. 2013 Feb; 29(2):199-210. PMC3552134.

[37]. Liang K, Weir MD, Reynolds MA, Zhou X, Li J, Xu HHK. Poly (amido amine) and nano-calcium phosphate bonding agent to remineralize tooth dentin in cyclic artificial saliva/lactic acid. Mater Sci Eng C Mater Biol Appl. 2017 Mar 1; 72: 7-17.

[38]. Zhang K, Cheng L, Wu EJ, Weir MD, Bai Y, Xu HH. Effect of water-ageing on dentine bond strength and anti-biofilm activity of bonding agent containing new monomer dimethylaminododecyl methacrylate. J Dent. 2013 Jun; 41(6):504-13.

[39]. 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 May; 26(5):471-82.

[40]. Bendary IM, Garcia IM, Collares FM, Takimi A, Samuel SMW, Leitune VCB. Wollastonite as filler of an experimental dental adhesive. J Dent. 2020 Nov; 102:103472.

[41]. Halvorson RH, Erickson RL, Davidson CL. The effect of filler and silane content on conversion of resin-based composite. Dent Mater. 2003 Jun;19(4):327-33

[42]. Lohbauer U, Wagner A, Belli R, Stoetzel C, Hilpert A, Kurland H-D, et al. Zirconia nanoparticles prepared by laser vaporization as fillers for dental adhesives. Acta biomater. 2010 Dec; 6(12):4539-46.

[DOI: 10.22068/ijmse.3203]