

Microtensile Bond Strength of Nanofilled Resin Composite Restoration Bonded to Dentin Using Self-etch adhesive Containing Nanohydroxyapatite versus Conventional One after One Month (In-vitro study)

Ahmed Mohamed Osama¹, Mona Ismail Riad² and Maha Elbaz³

¹MSc Restorative and Esthetic Dentistry, Faculty of Dentistry, Cairo University

²Professor of Restorative and Esthetic Dentistry, Faculty of Dentistry, Cairo University.

³Lecturer of Restorative and Esthetic Dentistry, Faculty of Dentistry, Cairo University.

At87_1@yahoo.com

Abstract: Objectives: The aim of this study was to evaluate the effect of addition of hydroxyapatite nanoparticles to self etch adhesives on microtensile bond strength of resin composite to tooth structure after 24 hours and evaluate the effect of one month storage on the bond strength. **Methods:** A total of 80 non-carious human molar teeth were selected A flat dentin occlusal surface was prepared 0.5 mm below DEJ perpendicular to the longitudinal axis of each tooth using a low speed diamond saw under water coolant. The specimens was divided into two equal groups of 40 specimens each, according to type of self etch adhesive used, where (A1) represents self etch adhesive containing nano hydroxyapatite and (A2) represents self etch adhesive without nano hydroxyapatite. Each group will be subdivided into two subgroup according to storage period, where (T1) represent 24 hour storage time and (T2) represent one month storage time. A Teflon mold was used to build up resin composite 4mm in thickness; they were light cured according to manufacturer's instructions. Teeth were sectioned in a longitudinal and a horizontal direction to obtain 0.9±0.1 mm in thickness beams. All samples were tested by universal testing machine. A Two-way ANOVA, followed by Tukey's post-hoc test was used. **Results:** Group (A1) after 24 hours produced the highest statistical mean microtensile bond strength (28.21±7.52 MPa) where group (A2) after 1 month storage period showed the least statistical mean microtensile bond strength at. **Conclusion:** The addition of hydroxy apatite nanoparticles to self etch adhesive system didn't interfere with immediate bonding to dentin and the bond strength of the tested adhesive either containing or non containing hydroxy apatite nanoparticles deteriorates with aging.

[Ahmed Mohamed Osama, Mona Ismail Riad and Maha Elbaz. **Microtensile Bond Strength of Nanofilled Resin Composite Restoration Bonded to Dentin Using Self-etch adhesive Containing Nanohydroxyapatite versus Conventional One after One Month (In-vitro study)**. *N Y Sci J* 2018;11(7):29-35]. ISSN 1554-0200 (print); ISSN 2375-723X (online). <http://www.sciencepub.net/newyork>. 5. doi: [10.7537/marsnys110718.05](https://doi.org/10.7537/marsnys110718.05).

Keywords: Microtensile; Bond Strength; Nanofilled Resin; Composite; Restoration; Dentin; Self-etch adhesive Containing; Nanohydroxyapatite; In-vitro study

1. Introduction:

All-in-one self etch adhesive require solvents and hydrophilic monomers to produce a strong bond at the moist dentin surface. The increase in acidic monomer concentration in self etch adhesive systems has compromised the resin-dentin bond, since a semi-permeable hybridization is formed. Degradation of hybrid layer at dentin adhesive interface is a concern for long term success of restorative procedures. Achievement of a more hydrolytic stable hybrid layer and high bond strength to the tooth is a recurrent goal of adhesive systems development nowadays (Tjaderhane, 2015).

The improvement of the physical and mechanical properties of the adhesive material can be achieved by fortifying the adhesive with filler (Belli et al., 2014). The addition of fillers reduces the amount of organic matrix in the same volume of material thus reducing water sorption and solubility. Considering that fillers are less prone to degradation by hydrolysis, the materials with filler addition could present a

decreased long term degradation (Giannini et al., 2012).

Hydroxyapatite (HA) [Ca₁₀(PO₄)₆(OH)₂] has been widely used as remineralizing agent and considered one of the most biocompatible ceramics (Kantharia et al., 2014). Its presence in hybrid layer is desirable to reconstruct the (HA) depleted zone after acid etching (Provenzi et al., 2014). Hydroxyapatite nanoparticles can be used as a filler to increase the mechanical properties of resin adhesives, considering the bioactivity and bond of these adhesives to the dental structure.

Hydroxyapatite nanoparticles may be a promising option of material for the preparation of new dental adhesives with superior properties without interfering with its immediate bond strength (Pepla et al., 2014). Thus this study was conducted to evaluate the effect of addition of the nano hydroxyapatite to self etch adhesive agent on micro tensile bond strength to dentin.

2. Materials and Methods:

The materials and preparation used as well as their description, composition and manufacturers are listed in (Table 1).

Preparation of hydroxyapatite nanoparticles:

Hydroxyapatite nanoparticles was prepared by hydrothermal method as reported by (Wagner et al. 2013). 0.09 Mole diammonium hydrogen phosphate (NH₄)₂HPO₄ and 0.15 Mole calcium nitrate 4-hydrate Ca (NO₃)₂·4H₂O were prepared. The phosphate was added drop-wise into the calcium nitrate solution, resulting in the precipitation of Hydroxyapatite (HA). The precipitate was aged for 22 hours at room temperature. Next, the precipitate (HA) was centrifuged and then washed with de-ionized water. The resulting powder was dried in a freeze-drier system for 10 hours. Finally to obtain hydroxyapatite nano-particles (HA-NP) the dried powder was calcined in an electrical box furnace at 900 °C for 1 hour at the rate of 5 °C/min in air (Paz et al. 2012). The size of the resultant particles was characterized using Transmission Electron Microscope and Visible Near Infra-Red (Vis-NIR) Spectrophotometer was used to monitor the absorption rate of tested particles.

Incorporation of Nano hydroxyapatite into adhesive system:

The (HA-NP) filler were incorporated into solution of Self-etch adhesive in 5% (w/vol)

concentration (Wagner et al., 2013). Thus to obtain 5% (w/vol) concentration of (HA-NP) in 5 ml of the adhesive the weight of the (HA-NP) would be 25 gram. After weighting 0.25 gram of (HA-NP) with high accuracy balance, the (HA-NP) were added to the resin solution in a dark glass box to avoid light and mechanically stirred by motorized mixer for one hour. To further increase the dispersion of the fillers, the resin mixtures being ultrasonicated for 30 minutes.

Selection of Teeth:

A total of 80 non-carious human maxillary first premolar teeth with straight two root canals, fully developed apices and extracted for orthodontics reason were selected from patient at age 20-35 years. The teeth were free cracks, showed no apparent hypoplastic defect.

Preparation of the teeth:

Porximal surfaces were reduced to expose the dentino enamel junction, then a line was placed 0.5 mm below the dentino enamel junction around the circumference of the tooth. Occlusal surfaces of the teeth were grinded until the level of the line using diamond disk mounted on a laboratory low speed hand piece. The teeth were cut horizontally and flattened under copious water coolant. This procedure was done to obtain flat dentin surface 0.5 mm below the dentino enamel junction.

Table (1): Materials' Description, composition, manufacturers and lot number

| Material | Specifications | Composition | Manufacturers | Batch number |
|---|---|--|---------------------------|--------------|
| Single bond Universal | A single step visible light cured self-etch adhesive | (MDP) [*] , Dimethacrylate resins, (HEMA) ^{**} , Vitrebond Copolymer, filler Ethanol, Water, Initiators, Silane | 3M ESPE, St Paul, MN, USA | 589528 |
| Composite Filtek TM Z350 XT (shade A3) | A Visible light activated cured nanohybrid methacrylate based resin composite | (BIS-GMA) ^{***} , (BIS-EMA) ^{****} , silanated silica, zirconia and ceramic, diurethane dimethacrylate, polyethyleneglycol dimethacrylate, (TEGDMA) ^{*****} , (BHT) ^{*****} and pigments | | N652030 |

Bis-GMA: Bisphenol A diglycidylmethacrylate, TEGDMA: triethylene glycol dimethacrylate, Bis-EMA: Bisphenol A polyethylene glycol diether dimethacrylate, UDMA: Urethane dimethacrylate HEMA: 2 hydroxyethyl methacrylate.

Acrylic blocks preparation:

A specially fabricated cylindrical Teflon mold (15mm diameter and 40mm height) was used to fabricate acrylic resin blocks. The mold consists of two halves and surrounded by metal ring for easy removal of the resultant blocks. Every prepared tooth was impressed vertically in a centralized position to expose only 5mm of the tooth, during the dough setting stage of the acrylic resin mix, the surveyor was

used to adjust the vertical axis of the tooth to be parallel to the long axis of the mold.

Adhesive systems application:

After preparation of the tooth, conventional self-etch adhesive was applied to the half number of the teeth and the modified adhesive applied to the other half of the tooth. The bottle was shaken, and then two layers of adhesive were applied and rubbed on dentin surface for 20 seconds with a micro brush. Air

blow was applied for five seconds to evaporate solvent and finally light cured for 10 seconds according to manufacturer's instructions using light-emitting diode (LED) Curing Light with light intensity of 1200 mw/cm².

Resin Composite application:

A specially constructed two halves split Teflon round mold with a central square hole (5mm in diameter and 4mm in height) was fabricated and secured firmly on a specially constructed metal ring (15 mm diameter and 45 mm height). Nano filled visible light resin composite was built up into two increments each 2mm in thickness on dentin surface. Each increment was packed using teflon tipped instrument then light cured for 20 seconds.

Specimen Preparation:

The acrylic blocks were used to facilitate mounting the teeth in IsoMet cutting machine. Stabilization of the selected teeth were done using metal square with opposing screws that holds the acrylic cylinder tightly. The mounted tooth was placed vertically parallel to the sectioning direction, thus maintaining the perpendicular relation between the cutting disc and the occlusal wall. After mounting the block in the gripping attachment, the composite specimen was serially sectioned, using a 0.3-mm thick diamond coated disc, at 2050 rpm; 8.8 mm/min feeding rate under copious coolant, mounted in an automated diamond saw, which was used for all sectioning procedures in this study. Resultant beams were 0.9±0.1 mm in thickness. A digital caliber was used to check the thickness of all beams.

Grouping of specimen:

A total of 80 dentin specimens were used. The specimens were divided into two equal groups of 40 specimens each, according to type of self etch adhesive used (A), where (A₁) represents self etch adhesive containing nano hydroxyapatite and (A₂) represents self etch adhesive without nano hydroxyapatite. Each group were subdivided into two subgroups according to storage period (T), where (T₁)

represent 24 hours storage period and (T₂) represent one month storage period.

Storage of the dentin specimen:

The microtensile bond strength of resin composite to dentin was measured at two time intervals (24 hours and 1 month). The specimens which stored for one month were immersed in distilled water that was changed every 4 days and kept in incubator adjusted at 37°C to simulate oral environment.

Microtensile bond strength measurement:

Geraldeli's jig was used to mount beams onto the universal testing machine. Each beam was aligned in the central groove of the jig and glued in place by its ends using cyanoacrylate based glue. The jig was in turn mounted into the universal testing machine with a load cell of 500 N. Tensile load was applied, at a cross-head speed of 0.5 mm/min, until bonding failure of the specimen occurred. Microtensile bond strength values were calculated in (MPa).

Ultramorphological Examination:

Two represented beams of each group were selected for ultra-morphological examination. Environmental Scanning Electron Microscope (ESEM) was used for examination of tooth restoration interface with backscattered electron mode at ×2000 and ×4000 magnification.

Statistical analyses:

Student's t-test used to compare between mean of two related groups of numerical (parametric) data. ANOVA (analysis of variance) used to compare between more than two groups of numerical (parametric) data followed by post-hoc tukey. P value <0.05 was considered statistically significant (S).

3. Results:

Group (A₁) after 24 hours produced the highest statistical mean microtensile bond strength (**28.21±7.52 MPa**) where group (A₂) after 1 month storage period showed the least statistical mean microtensile bond strength at (**15.87±6.64 MPa**).

Table (2): Mean and standard deviation (SD) for fracture resistance (N) for interaction between variables in descending order.

| | | Tensile Stresses at Maximum Load (MPa) | | Rank | p-value |
|--------------|------|--|------|------|---------|
| | | Mean | SD | | |
| Interactions | A1T1 | 28.21 | 7.52 | a | ≤0.001* |
| | A2T1 | 24.50 | 8.58 | ab | |
| | A1T2 | 23.02 | 9.67 | b | |
| | A2t2 | 14.59 | 5.74 | c | |

*=significant, NS= Non-Significant

Means with different letters within each column indicating significant difference at p≥0.05.

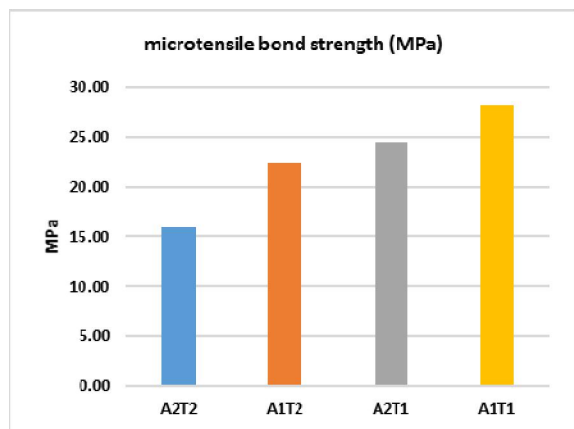


Figure (1): Histogram showing the microtensile bond strength (MPa) for interaction between variables.

4. Discussion:

Addition of fillers to adhesive resin could increase mechanical properties and decrease long term degradation of hybrid layer (Giannini et al., 2012).

Why hydroxyapatite?

Hydroxyapatite Nanoparticles is considered one of the most biocompatible and bioactive materials, and has gained wide acceptance in medicine and dentistry recently. Nano-sized particles have similarity to the apatite crystals of tooth enamel in morphology and crystal structure and have been studied as biomimetic materials for the reconstruction of tooth enamel suffering from mineral loss because of their unique potential for remineralization. It is considered as a good bone substitute, however (HA-NP) differs slightly from the biological apatite in terms of structure, composition, crystallinity, solubility and biological reactivity. Its presence in hybrid layer is desirable to reconstruct the hydroxyapatite depleted zone after acid etching that formed when dentin demineralization depth exceeds the dentin diffusion/impregnation depth, an area of hydroxyapatite-depleted collagen fibers is left exposed and inadequately infiltrated or hybridized. This zone of exposed collagen may be unstable and subjected to hydrolysis (Provenzi et al., 2014). Hydroxyapatite nanoparticles have been used to increase the mechanical properties of resin adhesives, considering the bioactivity and bond of these adhesives to the dental structure. A bioactive dental material would have several benefits including remineralization of adjacent tooth substance, potential closure of gaps between material and tooth and potentially better bond strength over time (less degradation of bond) (Pepla et al., 2014).

Preparation of hydroxyapatite

Hydroxyapatite nanoparticles was prepared by hydrothermal method as reported by Paz et al., 2011 and Wagner et al., 2013. There are other methods for (HA-NP) preparation a side from the hydrothermal method such as: dry methods, high-temperature processes, synthesis methods based on biogenic and combination procedures. The characteristics of a powder synthesized by a dry method are not strongly influenced by the processing parameters, hence most dry methods do not require precisely controlled conditions, making them suitable for mass production of powders. Hydroxyapatite powder generated from a typical dry method is usually large in size and irregular in shape. Therefore, hydrothermal method have conventionally been applied to the preparation of hydroxyapatite particles having a nanosized structure with a regular morphology. Hydrothermal method have advantages in their ability to control the morphology and the mean size of powder (Mehdi Sadat-Shojai, 2013). Based on these facts, hydrothermal method for nano-HAP preparation was adopted in this study.

Microtensile bond strength testing

Microtensile bond testing was preferred in the current study in order to overcome some of the inaccuracy of the conventional tensile and shear testing. As in the conventional tensile testing when the load is applied perpendicular to the bonded surface, non-uniform compressive stresses may be introduced along the bonded interface if alignment is not maintained between the adherend and the substrate during bonding or testing. While in the shear bond testing the load was applied parallel to the bonding surface, bending moments may develop if the force is distributed over relatively extended area of the adherend (Goracci et al., 2004).

The used beams were stick shaped with cross-sectional area of approximately 1 mm². It was found that the stress distribution improved as the thickness of the specimen decreased. (El Zohairy et al., 2004).

Microtensile bond strength results

Regarding microtensile bond strength results of resin composite to dentin where self etch adhesive containing 5 % (wt /vol) hydroxyapatite nano particles (HA-NP) and conventional self etch adhesive tested after 24 hours aging period showed a higher values with non-significant difference when compared to each other, while it showed a high significant difference when compared to that after one month aging period. This might be attributed to filler particles provide obstacles at the crack pathway, consuming energy during crack extension, as failure may originate from flaws in the adhesive resin, with the crack propagating in it (Belli et al., 2014). Also it

increased the surface energy of the dentin improving the resin infiltration inside and around collagen fibers.

These finding was in agreement with **Dumont et al. 2013**, who attributed improvements in the mechanical properties of the polymers to the insertion of inorganic particles. As the volumetric fraction of the inorganic matrix increases, the link of bond force is strengthened, because the diminishment of the organic portion minimize the polymerization shrinkage and increases the modulus of elasticity of the adhesive layer.

Akhavan et al. 2013, also attributed the adhesion improvement at the interface between the restorative material and the tooth structure to the nano filler through increasing mechanical strength of the adhesive layer and providing structural reinforcement. In fact, the nanofillers are stress absorbing and have the role of an elastic layer between dental composite and enamel. However, the amount of nanofiller and distribution of particles are the critical parameters which should be optimized in experiments.

However, the finding was in disagreement with **Elkassas et al, 2014**, who attributed the diminished micromechanical and chemical bonding efficacy to the dentin substrate to the addition of hydroxyapatite nano particles, as a reaction might have occurred between the acidic functional monomer 10 MDP with the added (HA-NP). The neutralization reaction might have increased the adhesive pH value due to the buffering effect induced by hydroxyapatite, diminishing its ultra-mild etching capacity and minimizing its micromechanical interlocking to the dentin substrate (**Zhang et al, 2012**). Furthermore, the reaction between the 10 MDP monomer with added Hap nanoparticles might have decreased the relative concentration of the functional monomers available for the subsequent interaction with the dentin substrate.

Also, these finding was also in disagreement with **M. Sadat-Shojai et al 2010**, as the microtensile bond strength increased significantly to the maximum value for specimens containing 0.2 wt% HA nanorods and then steeply decreased with increasing filler content. They attributed the reduction in microshear bond strength with increasing the filler content, due to incomplete curing of the adhesive. Moreover, at higher filler contents nanofillers tend to agglomerate.

Regarding Microtensile bond strength results of resin composite to dentin where conventional self etch adhesive showed a non-significant difference when compared to self etch adhesive containing 5 % (wt /vol) hydroxyapatite nano particles after 24 hours aging period, while it showed a high significant difference compared to that tested after one month aging period. This might be attributed to the fact that self-etch adhesives demineralized dentin only

partially, leaving hydroxyapatite partially attached to collagen. Since hydroxyapatite remains available for chemical interaction between residual hydroxyapatite and the functional monomers in self-etch adhesives. The functional monomers in self-etch adhesives can chemically interact with hydroxyapatite within a clinically manageable time, and this chemical interaction has been hypothesized with better resistance towards degradation by prevention of micro- and nanoleakage. (**Tekce et al., 2015 and Yoshida et al., 2004**).

Proenca et al. 2007, attributed the superior performance of the self etch adhesive system to the synergy of an unsaturated methacrylate phosphate ester (10- MDP) as the acidic monomer in combination with HEMA, which is believed to improve the wetting of the tooth surface and chelate to calcium ions of dentin and a higher exposure of the peritubular dentin, which is highly mineralized.

The microtensile bond strength results of both tested group after one month aging period showed the lowest values with significant difference when compared to each other, and showed a significant reduction in microtensile bond strength when compared to that tested after 24 hours aging period. This results might be attributed to the adhesive was HEMA based which is hydrophilic helping water sorption that initially caused a softening of the polymer resin component by swelling the network and reducing the frictional forces between the polymer chains. Water sorption may eventually cause irreversible damage to the material by formation of microcracks through repeated sorption desorption cycles. This is followed by hydrolytic degradation of the polymer with scission of the ester linkages and gradual deterioration of the infrastructure of the polymer over time (**Fukuoka A., 2011**).

These finding was in agreement with (**Yiu et al., 2004**) as the reduction in tensile strength of the hydrophilic resins that occurred mainly during the first month of water storage might be referred to the phase separation of the hydrophilic (HEMA) and hydrophobic domains in the resin matrices that further open up the polymer network, leading to more micro void formation with increased uptake of 'free' water.

Regarding the microtensile bond strength results of resin composite to dentin where self etch adhesive containing 5 % (wt /vol) (HANP) showed a higher significant difference when compared to that of conventional self etch adhesive (A2) after one month aging period. This result might be attributed to the addition of fillers to the adhesive resin that decreases the relative amount of polymeric matrix, decreasing the stress of contraction and hydrolytic degradation (**Provenzi et al., 2014**).

Leitune et al, 2013 also found that the addition of the filler results in reduction of the hydrophilicity of the adhesive resin leads to increase in restoration durability and longevity.

This result was in disagreement with **Elkassas et al, 2014**, as the nanofillers were incorporated into the adhesive without any surface pretreatment or silanization causes weak adhesion between the nanoparticles and matrix that might led to an easy plucking-out of filler particles in the wet environment. Although this action might have biological merit, it allows additional vacancies, which promoted a solvent effect, maximizing the plasticizing and hydrolytic effect of water, which was reflected in a significant reduction in the flexural strength values. Water storage was also associated with a significant reduction in μ -SBS as the hydrolytic degradation of the modified adhesives further negated the quality at the resin-dentin interface.

Conclusion:

Under the limitation of the current study the following conclusions could be derived:

1. The addition of hydroxy apatite nanoparticles to self etch adhesive system didn't interfere with immediate bonding to dentin.
2. Bond strength of the tested adhesive either containing or non containing hydroxy apatite nanoparticles deteriorates with aging.

References:

1. Akhavan A., Sodagar A., Mojtahedzadeh F., Sodagar K. (2013). Investigating the effect of incorporating nanosilver/ nanohydroxyapatite particles on the shear bond strength of orthodontic adhesives. *Acta Odontol Scand*, 71(5):1038-1042.
2. Belli R., Kreppel S., Petschelt A., Hornberger H., Boccaccini A. R., Lohbauer U. (2014). Strengthening of dental adhesives via particle reinforcement. *J Mech Behav Biomed Mater*, 37:100-108.
3. Dumont V. C. s., Silva R. M., Almeida-Jnior L. E., Roa J. P. B., Botelho A. M., Santos M. H. (2013). Characterization and evaluation of bond strength of dental polymer systems modified with hydroxyapatite nanoparticles. *Journal of Materials Science and Chemical Engineering*, 1(7):11-18.
4. El Zohairy A. A., de Gee A. J., de Jager N., van Ruijven L. J., Feilzer, A. J. (2004). The influence of specimen attachment and dimension on microtensile strength. *J Dent Res*, 83(5):420-424.
5. Elkassas D. W., Haridy M. H. (2014). Degree of conversion, flexure strenght, and bond strenght durability of a contemporary universal dentin adhesive fortified with silver and hhydroxyapatite nanoparticles. *Egyptian. Dental. Journal*, 61(3):110-121.
6. Fukuoka A., Koshiro K., Inoue S., Yoshida Y., Tanaka T., Ikeda T., Meerbeek B. V. (2011). Hydrolytic stability of one-step self-etching adhesives bonded to dentin. *J Adhes Dent*, 13(3):243-248.
7. Giannini M., Liberti M. S., Arrais C. A., Reis A. F., Mettenburg D., Rueggeberg F. A. (2012). Influence of filler addition, storage medium and evaluation time on biaxial flexure strength and modulus of adhesive systems. *Acta Odontol Scand*, 70(6):478-484.
8. Goracci, C., Sadek, F. T., Monticelli, F., Cardoso, P. E., & Ferrari, M. (2004). Influence of substrate, shape, and thickness on microtensile specimens' structural integrity and their measured bond strengths. *Dent Mater*, 20(7):643-654.
9. Kantharia N., Naik S., Apte S., Kheur M., Kheur S., Kale, B. (2014). Nano-hydroxyapatite and its contemporary applications. *Journal of Dental Research and Scientific Development* 1(1):1-15.
10. Leitune V. C., Collares F. M., Trommer R. M., Andrioli D. G., Bergmann C. P., Samuel S. M. (2013). The addition of nanostructured hydroxyapatite to an experimental adhesive resin. *Journal of Dentistry*, 41(12):115-124.
11. Paz A., Guadarrama Bello D., López M., E. Gonzalez, J., Brizuela N., Aragón Fernández J. (2011). A comparative study of hydroxyapatite nanoparticles synthesized by different routes. *Quim Nova J*, 35(9):1724-1727.
12. Pepla E., Besharat L. K., Palaia G., Tenore G., & Migliau G. (2014). Nano-hydroxyapatite and its applications in preventive, restorative and regenerative dentistry: a review of literature. *Annali di Stomatologia*, 5(3):108-114.
13. Proenca J. P., Polido M., Osorio E., Erhardt M. C., Aguilera F. S., Garcia-Godoy F., Toledano, M. (2007). Dentin regional bond strength of self-etch and total-etch adhesive systems. *Dent Mater*, 23(12):1542-1548.
14. Provenzi C., Leitune V. C., Collares F. M., Trommer R., Bergmann C. P., Samuel, S. M. (2014). Interface evaluation of experimental dental adhesives with nanostructured hydroxyapatite incorporation. *Applied Adhesion Science*, 2(1):2-7.
15. Sadat-Shojai M., Atai M., Nodehi A., Khanlar L. N. (2010). Hydroxyapatite nanorods as novel fillers for improving the properties of dental adhesives: Synthesis and application. *Dent Mater*, 26(5): 471-482.

16. Sadat-Shojai M., Khorasani M.-T., Dinpanah-Khoshdargi E., Jamshidi A. (2013). Synthesis methods for nanosized hydroxyapatite with diverse structures. *Acta Biomaterialia*, 9(8):7591-7621.
17. Tekce N., Demirci M., Tuncer S., Uysal Ö. (2015). Microtensile bond strength and sealing efficiency of all-in-one self-etching adhesives. *Biotechnology & Biotechnological Equipment*, 29(3): 570-578.
18. Tjaderhane L. (2015). Dentin bonding: can we make it last? *Oper Dent*, 40(1):4-18.
19. Wagner A., Belli R., Stotzel C., Hilpert A., Muller F. A., Lohbauer U. (2013). Biomimetically- and hydrothermally-grown HAp nanoparticles as reinforcing fillers for dental adhesives. *J Adhes Dent*, 15(5):413-422.
20. Yiu C. K., King N. M., Pashley D. H., Suh B. I., Carvalho R. M., Carrilho M. R., Tay, F. R. (2004). Effect of resin hydrophilicity and water storage on resin strength. *Biomaterials*, 25(26): 5789-5796.
21. Yoshida Y., Nagakane K., Fukuda R., Nakayama Y., Okazaki M., Shintani H., Van Meerbeek B. (2004). Comparative study on adhesive performance of functional monomers. *J Dent Res*, 83(6): 454-458.
22. Zhang Y., Wang Y. (2012). Hydroxyapatite effect on photopolymerization of self-etching adhesives with different aggressiveness. *J Dent*, 40(12): 1289-1299.

7/16/2018