THE EFFECT OF CRYSTAL GROWTH ON SHEAR BOND STRENGTH OF METALLIC BRACKETS AND ATOMIC FORCE MICROSCOPIC TOPOGRAPHIC EVALUATION OF TREATED ENAMEL

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ABSTRACT :

Objective: *To compare; the shear bond strength of metallic brackets, the residual adhesive after debonding using two concentrations of sulfated polyacrylic acid and the conventional acid etchant, and the topography of treated enamel using Atomic Force Microscopic examination (AFM).*

Methodology: *A total of 75 premolars were divided into three groups; 20 teeth each for shear bond strength test, and 5 teeth each for AFM. Three solutions were used; Solution I, 37% phosphoric acid, and two concentrations of sulphated polyacrylic acid (Solutions II, and III). Brackets were bonded, and the shear bond strength was measured using a universal testing instrument at a crosshead speed of 1mm/min. The residual adhesive was explored using a stereomicroscope at 20 times magnification, and quantified using the ARI. The topographic pattern of treated enamel was evaluated, using AFM, and an untreated enamel as a negative control.*

Results: *Solutions II and III, demonstrated lower shear bond strength (10.6MPa, 6.1MPa), compared with that of acid etchant (17.5 MPa). A significant difference in the distribution of ARI scores was found between group I and II, III ($P \leq 0.001$), with*

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more cement remaining on the teeth of group I. The AFM images showed a noticeable difference in the surface topography, with crystal deposits on the enamel surfaces treated with solution II.

Conclusions: *The concentration of 30% sulfated polyacrylic acid can be considered as an alternative to phosphoric acid. The shear bond strength value obtained was above the minimum of the clinically accepted for orthodontic use.*

Keywords: *Crystal growth; shear bond strength; residual adhesive; orthodontic brackets*

INTRODUCTION

Adhesives for orthodontic brackets, such as composite resins, have been considered as one of the most significant developments in clinical orthodontics. Nowadays, the use of acid-etching systems when bonding brackets to the enamel surface with orthodontic adhesives has been widely accepted by most orthodontists as a routine technique.[1,2] This bonding system provides a strong adhesion of the brackets to the tooth surface, hence preventing bracket failures. However, acid etching causes the dissolution of the interprismatic material in the enamel, producing an irregular enamel surface and it has been reported to cause up to 55 μm of enamel loss.[3]

The permanent loss of enamel calcium during the acid etching procedure may render the enamel surface more susceptible to demineralization during and after the orthodontic treatment.[4] This may also explain the iatrogenic effects on the enamel surface, especially during bracket removal. Reports of enamel fractures and cracks when debonding raised questions about the safety of using this strong adhesive technique.[1,5] There is an evident need to develop a mechanical or chemical retention system that would not or minimally alter the outer enamel surface. Building up a crystal layer for mechanical retention instead of creating a rough surface by means of strong etching has been studied.[6] The crystal bonding technique involved the application of polyacrylic acid containing residual sulfate ions which reacted with the enamel surface to produce a deposit of crystalline calcium sulfate.[7]

This method was studied as an alternative to the conventional phosphoric acid etch technique creating a micromechanical retentive surface through the formation of a crystalline interface.[8] It has been claimed that different ion solutions containing sulfate induce crystal growth and might be a better alternative to conventional acid etching for enamel pretreatment. Crystal formation depends mainly upon sulfate ion concentration and is independent on the molecular weight and concentration of the polyacrylic acid solution.[9]

This investigation was designed to evaluate the relevance of crystal growth on the enamel surface, and our hypothesis was that crystal growth can be produced by two different concentrations of sulfated polyacrylic acid with different pH, and can be effective alteration to conventional phosphoric acid etching. So, this study was conducted to compare the shear bond strength of the metallic brackets bonded to enamel using two concentrations of sulphated polyacrylic acid experimental solutions (in which the ion solution used was lithium sulfate), with that bonded using the conventional acid etch technique. Also, to determine the mode of bond failure, and, to verify the topographic difference of the enamel surfaces treated by these solutions and 37% phosphoric acid etching using AFM.

MATERIALS AND METHODS

The protocol of the study was approved by the Local Ethical Committee of Mansoura University. The used solutions were; 37% phosphoric acid (solution I), 30% sulfated polyacrylic acid, with 0.5M sulfate ions (solution II, pH= 0.24), and 20% sulphated polyacrylic acid, with 0.1M sulfate ions (solution III, pH= 1.35). The experimental solutions II and III were prepared at the Department of Pharmaco-chemistry, Faculty of Pharmacy, Mansoura University-Egypt.

A total of 75 freshly extracted first maxillary premolars teeth were collected from orthodontic clinics. The criteria for tooth selection included intact buccal enamel with no pretreatment with chemical agents, no cracks, no hypoplasia and no caries. These teeth were washed with water and stored in a solution of 0.1% thymol at 4C° until their use. The used brackets were metal mesh base premolar edgewise with an 0.018- inch slot (Victory series, 3M Unitek, Monrovia, Calif). The average bracket base area was 9.94 mm^2 according to the manufacture's specification.

For shear bond strength test, the teeth were divided into 3 groups; 20 teeth each according to the used solution. Each solution; I, II and III was applied on the tooth surface for 15 seconds, washed for 15 seconds, and thoroughly dried. For group I, the 37% phosphoric acid (3MUnitek) was used, and successful frosted appearance was required. For all groups, a thin uniform layer of Transbond XT primer (3M Unitek) was applied to the etched enamel surface, and Transbond XT adhesive (3M Unitek) was applied to the bracket base. The bracket was placed on the buccal surface of the tooth and pressed firmly to express any adhesive from the margins of the bracket base, and removed with an explorer before curing. Then, the composite was light-cured with an Ortholux LED curing unit (3M Unitek) for 10 seconds (5 seconds mesially, 5 seconds distally). All teeth were stored in distilled water at 37C° for one week. Each tooth was then embedded in a specimen holder ring with a self-curing acrylic resin. The buccal enamel surface was parallel to and projected above the rim of the cylindrical specimen holder ring.

Assessment of shear bond strength

A universal testing machine (Lloyd Instruments; West Fareham, UK) was used to determine shear bond strengths. A load was applied to the occlusal brackets' wings with a force in the occluso-gingival direction parallel to the buccal enamel surface. The force required to shear off the bracket was recorded in Newton (N) at a crosshead speed of 1.0 mm per minute. The shear bond strength in Megapascal (MPa), was then calculated by dividing the shear force by the bracket base area (9.94 mm^2).

Assessment of bracket bond failure

After bracket debonding, the bracket bases and the enamel surfaces were examined with a stereomicroscope at 20 times magnification to evaluate the mode of bond failure. The adhesive remnant index (ARI) was used to assess the quantity of adhesive remaining on the enamel surface.[10]

Enamel surface topographic examination

The remaining 15 premolars, 5 teeth each, were used for AFM examination to assess the topographic pattern of the enamel surfaces treated with the three different solutions. Plastic rings with standard 2cm height were prepared and filled with mixed self cure acrylic resin. The teeth were mounted in the acryl with their buccal surfaces facing up, and evenly flushed with the acrylic resin surface. The enamel surfaces were also mechanically polished under water cooling. Each solution was applied on the tooth surface for 15 seconds, washed for 15 seconds, and thoroughly dried. Then, the enamel surfaces were observed with the AFM (Thermomicroscpe Auto-rob cp; Santa Barbra, CA, USA), to evaluate their topography and roughness, and images were captured for an area of 25.0μm^2. As a negative control; untreated premolar enamel surface was subjected to AFM examination and an image was captured for further comparisons.

STATISTICAL ANALYSIS

Mean shear bond strengths, and roughness values (Ra) of the three groups were compared by one-way analysis of variance (ANOVA). Chi-square test was used for comparison of ARI scores. Tukey's post-hoc test was done for inter-groups comparisons. Statistical tests were conducted by using SPSS (SPSS Inc, Chicago, Ill). All tests were made at the significance level of $P \leq 0.05$.

RESULTS

Shear bond strength

Bonded brackets on enamel surfaces treated with the experimental solutions, II and III demonstrated inferior shear bond strength (10.6, 6.1MPa), compared with those bonded using 37% phosphoric acid (17.5 MPa). Their shear bond strengths were 61 and 35% of those treated with solution I. A high significant difference was noted among the shear bond strength of the three groups, $P \leq 0.001$ (Table 1).

Bracket bond failure

The recorded categories of bond failure were adhesive, cohesive, and mixed. The results revealed that 40% of brackets bonded to teeth etched with solution I, tended to fail by a mixed mode of failure, while only 10%, and 20% of those bonded on enamel treated with the experimental solutions failed similarly. Also, adhesive bond failure at bracket/ composite interface was considerably recorded in group I brackets (25%). Thus, most of the composite adhesive remained on the enamel surfaces (45%, 30%) etched with phosphoric acid scored 3, and 2 according to ARI. A significant difference in the distribution of ARI scores was found between group I and II, III ($P \leq .001$). Brackets bonded on solutions II and III treated enamel recorded the highest ratio of adhesive bond failure (90%, 70%) at enamel/ composite interface leaving the tooth surface clean (score 0, ARI). On the other hand, few brackets in group I showed similar failure mode (20%), leaving enamel surface with minimal adhesive remnants (score 1, ARI). Despite, there was no cohesive failure records within the enamel surface treated with both experimental solutions, two teeth (10%) were recorded in the solution I group (Table 2). No significant differences in ARI scores were found between group II and III ($P \geq 0.05$).

Enamel surface topographic examination

Compared with the untreated enamel surface (Figures 1, 2 A), the AFM images of the enamel etched with solution I (Figures. 1, 2 B), and that treated with the experimental solutions II and III (Figures. 1, 2 C, D), showed a noticeable topographic difference. The etched enamel surface showed marked deep irregularities, with a typical-honey comb appearance, and preferential dissolution of enamel prism cores and peripheries compared with marked crystalline overgrowth in group II. No similar topography was revealed by the image of the enamel treated using solution III, but only numerous and shallow pores were shown. The qualitative results of the crystalline structure produced by solution II, were interlacing crystalline rods, while the quantitative data were 7 to 9 micrometers (length) and 1.68 nano-micrometers to 2.68 micrometers (width).

Roughness of the enamel surfaces (as measured by AFM), produced by the three solutions was significantly different ($p \leq 0.001$). The high roughness value was recorded for solution II (347.8 nm), followed by solution I (245.2 nm), and the least value recorded was for solution III (174.8 nm), as shown in table 4. All roughness values were higher than that of the untreated normal enamel (146.8 nm).

Table 1: Shear bond strength (MPa) of brackets bonded after treatment of enamel surface with the three different solutions.

Solution N= 20	Mean ±SD	Tukey's test (5%)	95% Confidence interval for mean
(a) Solution (I)	17.50*** ±0.40	bc	17.32-17.70
(b) Solution (II)	10.60***±34	ac	10.42-10.73
(c) Solution (III)	6.1***±46	ab	5.93- 6.36
P	≤ 0.001		

*** A significant difference at ($P \leq 0.001$). SD: standard deviation.

Table 2. Mode of bracket bond failure presentation in the three different groups.

Mode of failure	Solutions (I)	Solution (II)	Solution (III)
Cohesive failure (composite)	1 (5%)	0 (0%)	0 (0%)
Cohesive failure (enamel)	2 (10%)	0 (0%)	0 (0%)
Adhesive failure (composite/ enamel)	4 (20%)	18 (90%)	14 (70%)
Adhesive failure (composite/ bracket)	5 (25%)	0 (0%)	2 (10%)
Mixed mode of failure	8 (40%)	2 (10%)	4 (20%)
Total no.	20	20	20

Table 3: Adhesive Remnant Index (ARI) scores.

ARI score	Solution (I)	Solution (II)	Solution (III)
0	0 (0.0%)	15 (75%)	12 (60%)
1	5 (25.0%)	3 (15%)	5 (25%)
2	6 (30%)	2 (10%)	2 (10%)
3	9 (45%)	0 (0%)	1 (5%)
Total no.	20	20	20

- Score 0; no adhesive remaining on the tooth;
- Score 1; less than half of the adhesive remaining on the tooth;
- Score 2; more than half of the adhesive remaining on the tooth; and
- Score 3; all adhesive remaining on the tooth with a distinct impression of the bracket base.

Table 4. Enamel surface roughness mean values: Ra in (nm)

Solution N= 20	Mean ±SD	Tukey's test (5%)	95% Confidence interval for mean
(a) Solution (I)	245.2^{***}±5.02	bc	238.96 – 251.4
(b) Solution (II)	382.4^{***}±4.9	ac	396.34- 388.45
(c) Solution (III)	174.8^{***}±3.9	ab	169.88- 179.71
P		≤ 0.001	

*** A significant difference at (p≤ 0.001). Untreated normal enamel surface: Ra = 146.8 nm

Ra: The arithmetic mean roughness of surface.

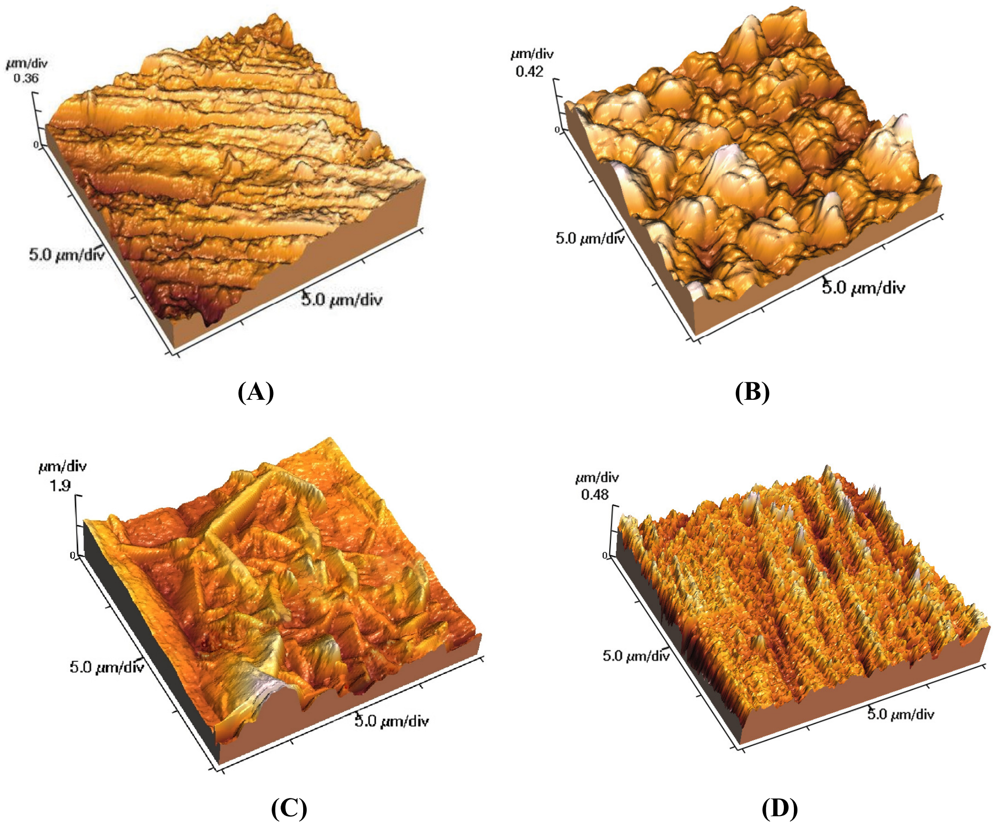

Figure 1. 3-D AFM images of untreated enamel (A), etched enamel with 37% Phosphoric acid etching (B), enamel treated with 30% sulfated polyacrylic acid (C) enamel treated with 20% Sulfated polyacrylic acid (D).

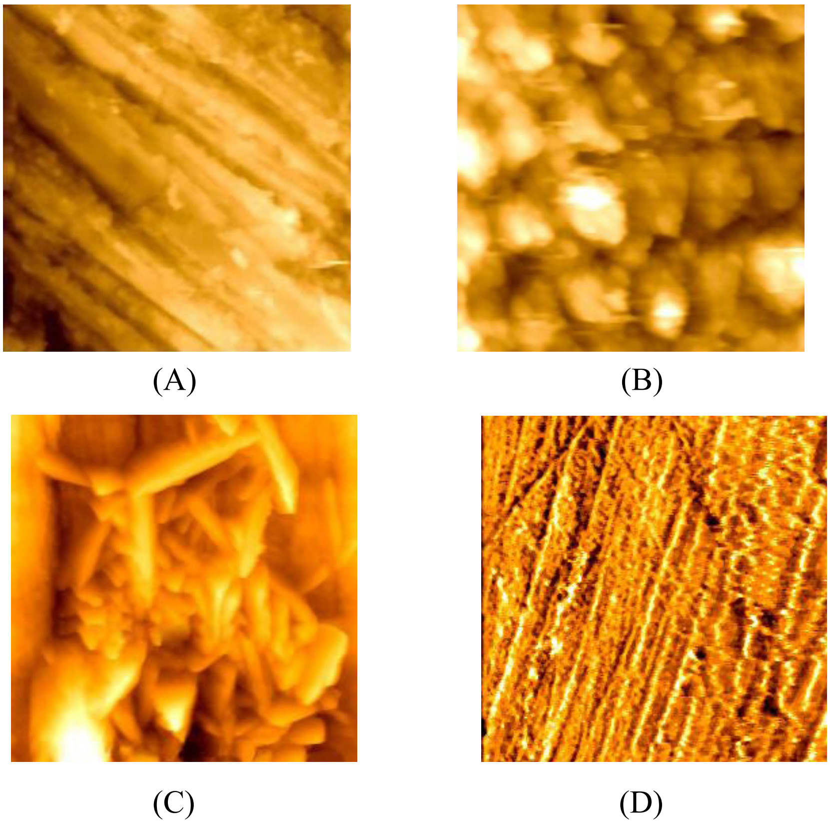

Figure 2. 2-D AFM images of untreated enamel (A), etched enamel with 37% Phosphoric acid etching (B), enamel treated with 30% sulfated polyacrylic acid (C) enamel treated with 20% Sulfated polyacrylic acid (D).

DISCUSSION

As a possible alternative to the conventional acid etching, crystal bonding technique with the application of polyacrylic acid containing residual sulfate ions react with the enamel surface to produce ionized carboxyl groups.[10]Strong ionic bonding between calcium ions at the enamel surface and the carboxyl groups provides crystal enucleation sites for the gypsum crystals. This in turn provides micromechanical retention for the bonding resin.[11,12] Selection of the experimental solutions in this study was based on the conclusions drawn from different studies.[3,9,12]

So, it was our aim to study the possibility of using two concentrations of sulfated polyacrylic acid, with different pH, to evaluate shear bond strength of bonded brackets and to verify crystal formation, if any, on the enamel surface using the Atomic Force Microscope.

The mean shear debonding force following 15 seconds etching using solution I, was significantly greater than that of 15 seconds application of solutions II and III. Several factors may account for this difference, the morphology of the treated enamel being the most likely. Etching with 37% phosphoric acid showed a typical honey comb appearance with a preferential dissolution of enamel prism cores and peripheries, similar to that obtained by previous studies.[3,13] On the other hand, using 30% sulphated polyacrylic acid, with 0.5 sulfate ion produced crystal growth, with slight etching effect in agreement with different studies.[3,13-16] However, application of 20% sulfated polyacrylic acid, with (0.1) sulfate ion produced no crystal growth, but slight etching effect, in agreement with previous study [9] and contrary to the result of others, who reported evidence of crystal growth.[3] The causes of no crystal formation may be related to the lower concentration of the sulfate ion,[9] or the alkalinity of the prepared solution (pH=1.35), as it was reported that the mild acidic solutions were preferred to release calcium ions to form a salt with the crystal-forming ionic solution.[14]

The achieved shear bond strength of the experimental solution II was 61% of that of the conventional acid etching solution. This agrees with earlier studies,[11,12,15,16] and disagrees with others,[17,18] who showed significantly lower shear bond strength. However, the higher bond strength associated with the conventional acid etching may be higher than clinically necessary. Thus, it could further increase the risk of enamel damage, or could leave maximal remnants at removal of the adhesive. Although the crystal growth technique produced lower bond strength, this may seem advantageous for maximal clearance of an adhesive and still works within the clinically accepted bond strength (6-10 MPa).[19, 20]

Roughness of enamel induced by solution II was significantly greater than that of phosphoric acid. This may be related to the combined effect of both etching and crystal deposits produced in solution II treated enamel. In spite of that, shear bond strength was in a reverse pattern, which may be related to the difference in enamel roughness morphology. Conventional etching using solution I revealed deep pits for resin penetration into the many pores in the etched surface, producing strong bond of resin, while solution II produced crystal growth structure with interlacing long crystalline rods forming retentive mesh for micromechanical retention. A weak chemical bond between the enamel and the deposited crystals or the properties of the resultant growth crystals may play a role in the weaker bond strength of the experimental solution II.[13]

On the other hand, the significant difference found between bond strength of brackets on enamel treated with solution II, and III may be related to the deposited crystal structure. Although the concentration of polyacrylic acid was higher, the crystal deposits might play the major role in enhancing the bond strength of solution II compared to solution III brackets.

The weaker bond strength and the unique roughness pattern on enamel treated by solution II, compared with the high bond strength and the deep pores resulted from conventional acid etching, may play a role on the increased rate of adhesive bond failure at the enamel/ composite interface.[10,18] This mode of failure could help in leaving the debonded enamel surface clean, more than other types of bond failure.

As revealed by ARI scores in this study, enamel fracture occurred only when the brackets debonded from enamel treated by solution I,[21] indicating that the increase in enamel fracture might be related to an excessive force. It was previously reported that enamel failure occurred when the bond strength exceeded 13.5 MPa,[22] which could explain the results of this study.

Despite the significantly lower shear bond strength of the experimental solution II, the findings of this study strengthened the value of the sulfated polyacrylic acid in bracket bonding with relatively clinically acceptable shear bond strength. Further studies using polyacrylic acid solutions with different concentrations, contact times, and altered nature of sulfate ion, to induce better crystal growth properties for trials to achieve a more reliable and acceptable bond strength are highly recommended.

CONCLUSIONS

- Crystal growth appeared only with 30% sulfated polyacrylic acid (pH, 0.24) as confirmed with AFM.
- Sulfated polyacrylic acid solutions provided significantly lower shear bond strength than that produced by 37% phosphoric acid. 30% sulfated polyacrylic acid produced significantly higher bond strength compared to the 20% sulfated polyacrylic acid.
- Increased rate of adhesive bond failure at the enamel/ composite interface occurred with 30% sulfated polyacrylic acid, leaving the enamel surface clean.
- 30% sulfated polyacrylic acid can be considered as an alternative to phosphoric acid etching.

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