Evaluation of the Adhesive Strength in Dentin after Irradiation with Ti:Sapphire Ultrashort Laser Pulses

Tarciso Penha Junior
Universidade Paulista - UNIP
São Paulo, SP, Brazil
tarciso.penha@uol.com.br

Mônica de Abreu Pessoa Rodrigues
Universidade Paulista - UNIP
São Paulo, SP, Brazil
monicaapr95@gmail.com

Denise Maria Zezell
IPEN-CNEN/SP
São Paulo, SP, Brazil
zezell@usp.br

Nilson Dias Vieira Jr.
IPEN-CNEN/SP
São Paulo, SP, Brazil
nilsondiasvieirajr@gmail.com

Ricardo Elgul Samad
IPEN-CNEN/SP
São Paulo, SP, Brazil
resamad@gmail.com

Maristela Dutra-Correa
Universidade Paulista - UNIP
São Paulo, SP, Brazil
maristeladutracorrea@gmail.com

***Abstract*—This study was done to evaluate whether the irradiation of dentine with ultrashort laser pulses prior to adhesive procedures contributes to increase the adhesive resistance to microtraction essays. Twenty-four human teeth (third molars) were used, divided into 4 groups (n=6): standard adhesive procedures with etch-rinse adhesive were used in the control group, and in the experimental groups, the dentin was irradiated by 25 fs pulses prior to the adhesive procedures, with varying fluences. One tooth from each group was used to evaluate its surface roughness. After 24h, the teeth were sectioned with perpendicular cuts, producing toothpicks that were submitted to the microtraction test. The results showed that the groups irradiated with fluences under 4 J/cm² presented similar results among themselves, comparable to the control group, while the group irradiated with 8 J/cm² showed lower adhesive strength. We propose that the adhesive strength and surface roughness reduction resulting from the fluence increase is probably related to the shielding arising from an electron plasma formation during the ablation, decreasing the material removal efficiency. Nevertheless, the lower energy densities did not affect the adhesiveness, maintaining values similar to the control.**

Keywords—adhesion, dentin, femtoseconds laser pulses, Ti:Sapphire laser irradiation, microtensile bond strength

I. INTRODUCTION

A dental restorative procedure involves the replacement of dental tissue by resinous materials, established by the formation of a hybrid layer and chemical interactions [1, 2], which may influence the quality and durability of the restoration [3, 4]. Research with new restorative materials, application techniques and different adhesive strategies for surface treatment have presented satisfactory results [5]. However, regardless of the adhesive strategy adopted, the interface is exposed to several factors that may favor its early degradation, such as mechanical fatigue, which may result in the formation of gaps and tensions resulting from contraction of post-gel polymerization, causing restoration failure [6].

The association of laser irradiation of the dental surface with adhesive techniques showed an increase in the adhesion resistance that can be measured by microtraction techniques, but the generation of heat can promote degradation of the collagen fibers, like the one originated from the interaction with Nd:YAG nanosecond pulses [7, 8]. On the other hand, Ti:Sapphire laser ultrashort pulses irradiation presents efficient ablation with low heat generation [9-11], due to the highly nonlinear interaction mechanism that excites electrons and promotes ablation before the electronic relaxation transmits significative amount of heat to the material [12, 13]. Femtosecond (fs) ultrashort laser pulse have been used in dentistry and medicine for surface treatment, promoting adhesion in ceramic brackets [14], removal of debris in necrotic tissues and changes in the healing phase [15]. However, treatment of dental surface (dentin) with ultrashort laser pulse has not been investigated.

The objective of this study was to evaluate whether the irradiation of dentine with ultrashort pulses from a Ti:Sapphire lasers before the adhesive procedures, contributes to the increase of the adhesive resistance to microtraction. This evaluation was done submitting the samples to Microtensile Bond Strength measurements.

II. MATERIALS AND METHODS

This research was approved by the Research Ethics Committee of the Universidade Paulista - UNIP (CEP: 1,785,244/2016). Twenty-four human teeth were used, randomly divided into 4 groups (n = 6) that received different surface treatments: C-Control: acid conditioning + primer + adhesive + restoration with composite resin; in the other groups the dentin was irradiated with ultrashort laser pulses with different fluences prior to the adhesive/restorative procedures, being: L2J-Laser (2 J/cm^2); L4J-Laser (4 J/cm^2) and L8J-Laser (8 J/cm^2). Initially, the dentin was exposed and the smear layer was standardized [16], followed by laser irradiation. For this purpose, they were placed in a computer-controlled x-y-z translation stage (3 UTS100CC translators, Newport, USA) for the irradiation with fs pulses generated by a Ti:Sapphire amplified laser system (Femtopower Compact Pro CE-Phase HP / HR, Femtolasers, Vienna, Austria). This system generates 25 fs (FWHM) pulses, centered at 785 nm with 40 nm of bandwidth, in a pulse train with maximum repetition rate of 4 kHz and 800 µJ maximum pulse energy, in a Gaussian beam with $M^2<2$. The beam was focused by a 75 mm focal length achromatic doublet, to a 20 µm beamwaist, positioned on the surface of the samples with normal incidence. No cooling system was used in the targets. The specimens were irradiated with pulse energies of 25 µJ, 50 µJ and 100 µJ, corresponding to fluences (energy densities) of 2.0 J/cm^2, 4.0 J/cm^2 and 8.0 J/cm^2, respectively. For each irradiation, the pulses energy was measured with an energy meter (sensor J-25MT-10KHZ with display LabMax TOP, Coherent, USA). The irradiation was performed by scanning the entire dentin surface with a constant velocity of 8 mm/s, in parallel lines with a displacement of 40 µm between the lines, and a repetition rate of 4 kHz. After irradiation, one sample from each group had its surface roughness determined from

978-1-7281-3092-7/19/$31.00 ©2019 IEEE

measurements performed with a 3D optical profilometer (ZeGage, Zygo Corporation, USA), and the other 5 samples were used in the microtraction tests.

After irradiation the restorative procedures were performed with etch-rinse adhesive (SBMP-3M ESPE, USA) and restored with resin (Z350 XT - 3M ESPE, USA) and light-cured (Optilight Max, Gnatus Equipamentos Médico-Odontológicos Ltda, Brazil). The materials were used according to the manufacturers' guidelines. After the photopolymerization, the teeth were stored in distilled water at 37ºC for 24h [17]. To make samples for the microtraction test, perpendicular cuts to the interface were then made, producing sticks with $0.7 \times 0.7\ mm^2$ that were individually tested until fracture using standard procedures [18, 19]. The type of the failure was analyzed in a stereomicroscope. The data were treated statistically to verify the normality and homoscedasticity, and Analysis of Variance (ANOVA one-way) was applied, followed by the Tukey test, for comparison between the groups.

III. RESULTS

Fig 1. shows, for each group, a topographic map and a scanning electron microscope micrography of a sample. In both the topographic maps and the micrographies the tracks etched by scanning the lasers pulses can be observed.

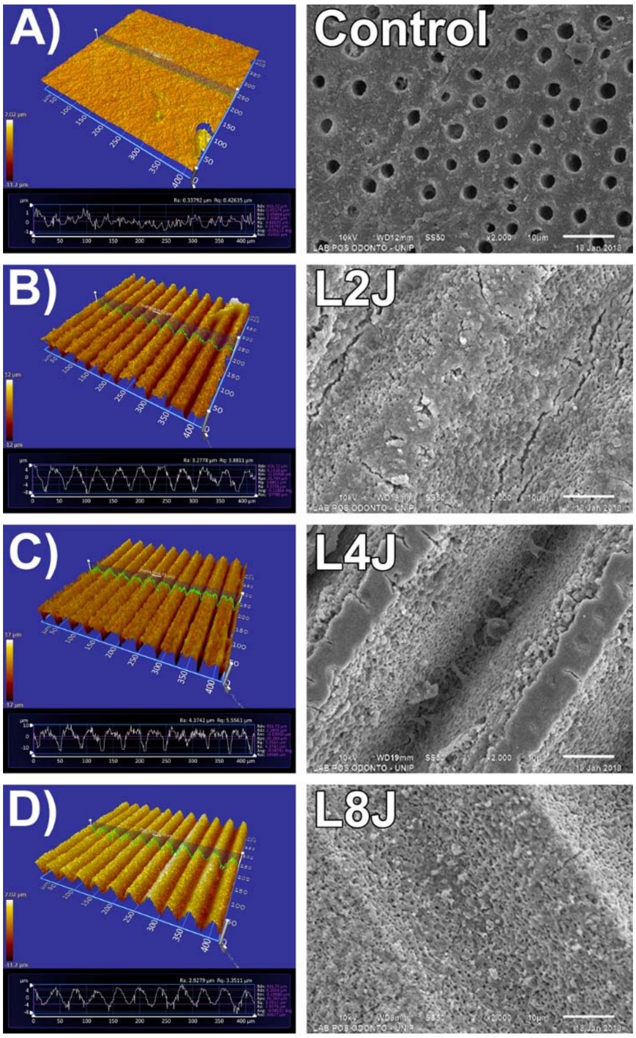

Fig. 1. micrographies of the 4 groups. In the topographics maps the laser scan lines can be clearly seen, and the profile show in green is reproduced below each image; in the micrographies the scale bar has 10 µm of length. A) Control Group. B) L2J group. C) L4J group. D) L8J group.

Table I summarizes the results obtained for the 4 groups. The first and second columns presents the roughness (Ra) and surface roughness (Sa) obtained from a sample from each group, and in the last column the Microtensile Bond Strenght (µTBS) values averaged over all samples of each group are shown. The µTBS values, which are also presented in Fig. 2, are similar between the Control and L2J and L4J groups; the L8J group presented a lower value of adhesive strength relative to the Control and L2J groups ($p < 0.05$) and also relative to the L4J group ($p < 0.01$).

TABLE I. SAMPLES RESULTS

	Ra (µm)	*Sa (µm)*	*Microtensile fracture Pressure (MPa)*
Control	0.34	0.44	35.0±5.1
L2J	3.28	3.40	34.9±3.7
L4J	4.37	4.40	37.5±5.8
L8J	2.93	2.96	28.1±4.1

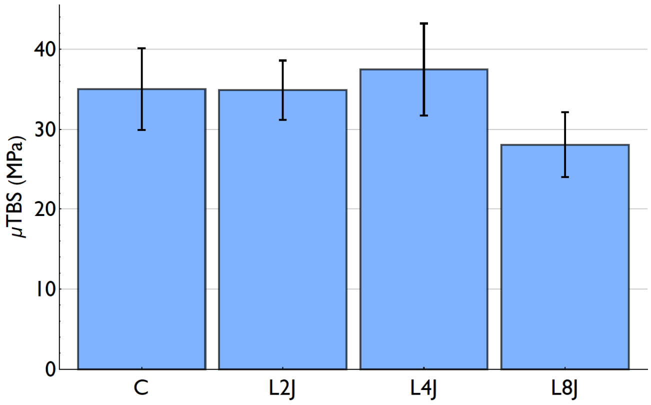

Fig. 2. Microtensile Bond Strenght (µTBS) mean values for each group.

Fig. 3 shows the percentage of fault types in the tooth-restoration interface. The faults were classified according to the following criteria: Adhesive: faults occur in the restorative-tooth interface; Cohesive: occur in the same substrate (resin or dentin), and Mixed: presence of fragments of both substrates. As expected, adhesive failure was predominant for all groups, since the interface is the weakest component of the system and the force needed to break it determines the quality of the restorations.

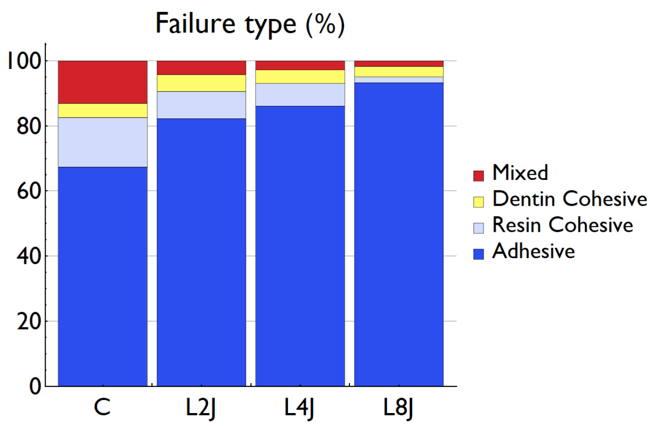

Fig. 3. Percentage of the failures in each group, showing the predominance of the Adhesive type.

IV. Discussion and Conclusions

To hypothesize why the highest fluence resulted in a decrease of the Microtensile Bond Strength, it is necessary to understand the physical mechanisms of ultrashort laser pulses ablation. When an ultrashort laser pulse (in the tens of femtoseconds time scale) hits the surface of a solid, it directly ionizes and excites the material electrons, creating a hot plasma of free electrons [20] inside and just outside this surface [21]. After the pulse has finished, the electrons relax their energy to the material molecules, breaking the atomic bonds, ejecting material away from the surface and generating an ionized plasma [11, 22-24], which dissipates rapidly, transferring little or no heat to the surrounding tissues. Although the plasma reaches a very high temperature, it carries a small amount of energy, and the cumulative thermal effect and the heat diffusion into the tissue is usually negligible, preserving its properties [22]. The small thermal effects can also be understood as a consequence of the very short pulse duration ($\sim 10^{-14}$ s) relative to the conduction of heat within materials, which usually occurs in the millisecond (10^{-3} s) time scale.

The interaction of Ti:Sapphire femtosecond pulses with hard dental tissues can alter their physical, chemical and structural characteristics. The ablation progression follows the geometry of the Gaussian beam, forming a conical cavity [9, 24], and the formation of ablation debris increases with the pulses focused energy density on the tissue surface [25]. Accordingly, in the present study, greater amounts of ablation debris were present on the irradiated surfaces with the higher energy density (8 J/cm²).

During the interaction of a pulse with the hard tissue (or any solid target), the density of the electrons plasma generated in the material grows, and it can influence the progress of the ablation; this density increase may approach the laser wavelength plasma frequency and reflect parts of the pulse, decreasing the ablation efficiency [11], in what is known as shielding effect [26]. In this study, the increase in pulse energy from 25 µJ to 50 µJ (2 and 4 J/cm² fluences, respectively) did not affect the microtensile adhesive strength, since the results were similar to each other and to the Control. However, when the pulse energy increased to 100 µJ (8 J/cm² fluence), the adhesive resistance was reduced with statistical difference relative to the other groups. We propose that the intense reflection originated in the electron's plasma prevented a significant fraction of the pulses energy to be transferred to the surface of the material, decreasing the ablation efficiency (material removal) and the surface modification. This effect is confirmed by the surface roughness values: the laser irradiation dramatically increases the surface roughness, and rising the pulse energy from 25 µJ to 50 µJ resulted in an approximately 35% roughness increase; when the pulse energy reached 100 µJ, the roughness produced was lower than the one obtained with 25 µJ, indicating that the surface relief was less modified at this higher energy. This could be a consequence of the plasma shielding or thermal effects that cause some melting and decreasing of the roughness. This melting is evidenced in Figs. 1b-d, in which the dentinal tubules cannot be seen at the samples' surfaces, in opposition to the control group (Fig. 1a) in which they are evident. More studies are necessary to prove this hypothesis.

Although the surface roughness has an influence on the adhesivity of the restoration to the tooth, it is clearly not the only mechanism to contribute this force, otherwise the non-irradiated tooth would present the lowest adhesion. The interaction between the irradiated dental structure and the restorative material has not yet been exhaustively explored. The present study showed that the irradiation with ultrashort laser pulses with different energy densities, was not enough to improve (increase) the adhesion between the irradiated dental surface and the restorative material. Therefore, future research should address this theme to find how this interaction occurs in scales smaller than the ones presented here, and if there is any modification in the chemical composition of the dental structure that could affect the adhesion.

In conclusion, irradiation of the dentin surface with low fluences did not affect the adhesiveness of the restoration, maintaining values similar to the control. However, the increase of the laser pulses energy resulted in a reduction of the adhesive strength and created a surface roughness that did not increase monotonically with the fluence, probably a consequence of the shielding effect.

References

[1] B. Van Meerbeek, J. De Munck, Y. Yoshida, S. Inoue, M. Vargas, P. Vijay*, et al.*, "Buonocore Memorial Lecture - Adhesion to enamel and dentin: Current status and future challenges," *Oper. Dent.,* vol. 28, pp. 215-235, May-Jun 2003.

[2] D. H. Pashley, F. R. Tay, L. Breschi, L. Tjaderhane, R. M. Carvalho, M. Carrilho*, et al.*, "State of the art etch-and-rinse adhesives," *Dent. Mater.,* vol. 27, pp. 1-16, Jan 2011.

[3] M. Sozzi, C. Fornaini, A. Cucinotta, E. Merigo, P. Vescovi, and S. Selleri, "Dental ablation with 1064 nm, 500 ps, Diode pumped solid state laser: A preliminary study," *Las. Ther.,* vol. 22, pp. 195-9, 2013.

[4] J. Liu, H. Chen, W. Ge, Y. Wang, Y. Sun, Y. Wang*, et al.*, "A Roughness Study of Ytterbium-Doped Potassium Yttrium Tungstate (YB: KYW) Thin-Disk Femtosecond Ablated Dentin," *J. Las. Med. Sci.,* vol. 5, pp. 32-38, 2014 2014.

[5] J. Perdigao, A. Sezinando, M. A. Munoz, I. V. Luque-Martinez, and A. D. Loguercio, "Prefabricated veneers - bond strengths and ultramorphological analyses," *J. Adhes. Dent.,* vol. 16, pp. 137-46, Apr 2014.

[6] A. O. Carvalho, M. T. d. Oliveira, T. Nikaido, J. Tagami, and M. Giannini, "Effect of adhesive system and application strategy on reduction of dentin permeability," *Braz. Oral Res.,* vol. 26, pp. 397-403, 2012.

[7] A. K. Marimoto, L. A. Cunha, K. C. Yui, M. F. Huhtala, D. C. Barcellos, A. Prakki*, et al.*, "Influence of Nd:YAG laser on the bond strength of self-etching and conventional adhesive systems to dental hard tissues," *Oper. Dent.,* vol. 38, pp. 447-55, Jul-Aug 2013.

[8] J. Gan, S. Liu, L. Zhou, Y. Wang, J. Guo, and C. Huang, "Effect of Nd:YAG Laser Irradiation Pretreatment on the Long-Term Bond Strength of Etch-and-Rinse Adhesive to Dentin," *Oper. Dent.,* vol. 42, pp. 62-72, Jan/Feb 2017.

[9] M. Dutra-Correa, G. Nicolodelli, J. R. Rodrigues, C. Kurachi, and V. S. Bagnato, "Femtosecond laser ablation on dental hard tissues—Analysis of ablated profile near an interface using local effective intensity," *Las. Phys.,* vol. 21, pp. 965-971, 2011.

[10] F. d. A. M. G. Rego Filho, M. Dutra-Correa, G. Nicolodelli, V. S. Bagnato, and M. T. de Araujo, "Influence of the hydration state on the ultrashort laser ablation of dental hard tissues," *Lasers. Med. Sci.,* vol. 28, pp. 215-22, Jan 2013.

[11] E. G. Gamaly, A. V. Rode, B. Luther-Davies, and V. T. Tikhonchuk, "Ablation of solids by femtosecond lasers: Ablation mechanism and ablation thresholds for metals and dielectrics," *Phys. Plasmas,* vol. 9, pp. 949-957, Mar 2002.

[12] M. Lenzner, J. Kruger, S. Sartania, Z. Cheng, C. Spielmann, G. Mourou*, et al.*, "Femtosecond optical breakdown in dielectrics," *Phys. Rev. Lett.,* vol. 80, pp. 4076-4079, May 1998.

[13] H. Chen, H. Li, Y. Sun, Y. Wang, and P. Lu, "Femtosecond laser for cavity preparation in enamel and dentin: ablation efficiency related factors," *Sci. Rep.,* vol. 6, p. 20950, Feb 11 2016.

[14] E. A. Erdur and F. A. Basciftci, "Effect of Ti:sapphire laser on shear bond strength of orthodontic brackets to ceramic surfaces," *Lasers Surg. Med.,* vol. 47, pp. 512-9, Aug 2015.

[15] M. O. D. Santos, A. Latrive, P. A. A. De Castro, W. De Rossi, T. M. T. Zorn, R. E. Samad*, et al.*, "Multimodal evaluation of ultra-short laser pulses treatment for skin burn injuries," *Biomed. Opt. Express.,* vol. 8, pp. 1575-1588, Mar 1 2017.

[16] D. H. Pashley, L. Tao, L. Boyd, G. E. King, and J. A. Horner, "Scanning electron microscopy of the substructure of smear layers in human dentine," *Arch Oral Biol,* vol. 33, pp. 265-70, 1988.

[17] B. R. Kim, M. H. Oh, and D. H. Shin, "Effect of cavity disinfectants on antibacterial activity and microtensile bond strength in class I cavity," *Dent Mater J,* vol. 36, pp. 368-373, May 31 2017.

[18] D. H. Pashley, H. Sano, B. Ciucchi, M. Yoshiyama, and R. M. Carvalho, "Adhesion testing of dentin bonding agents - A review," *Dent. Mater.,* vol. 11, pp. 117-125, Mar 1995.

[19] International Organization for Standardization, "Testing of adhesion to tooth structure (ISO Standard No. 11405:2015)", 2015. Retrieved fom https://www.iso.org/standard/62898.html

[20] S. Nolte, C. Momma, H. Jacobs, A. Tunnermann, B. N. Chichkov, B. Wellegehausen*, et al.*, "Ablation of metals by ultrashort laser pulses," *J. Opt. Soc. Am. B,* vol. 14, pp. 2716-2722, Oct 1997.

[21] M. P. Seah and W. A. Dench, "Quantitative electron spectroscopy of surfaces: A standard data base for electron inelastic mean free paths in solids," *Surf. Interface Anal.,* vol. 1, pp. 2-11, 1979.

[22] A. V. Rode, E. G. Gamaly, B. Luther-Davies, B. T. Taylor, J. Dawes, A. Chan*, et al.*, "Subpicosecond laser ablation of dental enamel," *J. Appl. Phys.,* vol. 92, pp. 2153-2158, Aug 2002.

[23] G. Dumitru, V. Romano, H. P. Weber, M. Sentis, and W. Marine, "Femtosecond ablation of ultrahard materials," *Appl. Phys. A-Mat. Sci. Proc.,* vol. 74, pp. 729-739, 2002.

[24] J. Neev, L. B. Da Silva, M. D. Feit, M. D. Perry, A. M. Rubenchik, and B. C. Stuart, "Ultrashort pulse lasers for hard tissue ablation," *IEEE J. Sel. Top. Quantum Elec.,* vol. 2, pp. 790-800, 1996.

[25] Q. T. Le, C. Bertrand, and R. Vilar, "Femtosecond laser ablation of enamel," *J Biomed Opt,* vol. 21, p. 65005, Jun 1 2016.

[26] A. Bogaerts, Z. Chen, R. Gijbels, and A. Vertes, "Laser ablation for analytical sampling: what can we learn from modeling?," *Spectrochim. Acta B,* vol. 58, pp. 1867-1893, 2003.