New Trends in Dental Adhesion—A Systematic Review

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Abstract: Restorative dentistry is based on a continuous development of materials that make the best possible connection with dental tissues. Thus, dental adhesives have been researched with increasing interest. A successful adhesive system depends very much on its chemistry, as well as on the clinical procedures that must be appropriate for that type of dental adhesive. This review presents a retrospective of dental adhesives focusing on research into new types of adhesive systems that involves the use of nanoparticles. Dental adhesive systems are used in most clinical procedures related to restorative dentistry and can be classified into “etch-and-rinse” adhesives and “self-etch” adhesives. Recently, both types of adhesive systems have been modified, being loaded with different types of nanoparticles to try to improve them in terms of the thickness of the adhesive layer, but also to induce other special qualities. Therefore, in order to avoid compromising the restorative procedures by losing the restorations or by affecting the dental pulp due to cytotoxicity, several factors must be considered in choosing the adhesive system.

Keywords: dental adhesive; nanoparticles; novel adhesives

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Introduction

Today, dental adhesives have a wide range of uses. Direct composite restorations in the frontal or posterior areas require the use of dental adhesives. These adhesives are also needed during the cementation of ceramic or composite prosthetic restorations made in the dental laboratory such as inlays, onlays, crowns or veneers [1].

The use of composite materials for the restoration of dental cavities resulting from the removal of decayed tissues is increasing, due to the links between the aesthetic and mechanical properties of polymers reinforced with different loads [2].

The main purpose of adhesive systems is the adhesion of composite materials to structures. In order to achieve the closest possible connection, the surface of the composite materials and the dental surfaces must be as close
as possible. In addition to this goal, adhesive systems must have characteristics such as low viscosity, substrate umectability and fluidity [3].

One of the biggest causes of failure of composite resin-based restorations is micro infiltration. Micro-infiltrations can be defined as the passage of fluids, bacteria, molecules or ions between the restoration material and the walls of the cavity [4].

The most used adhesive systems are of the “etch and rinse” and “self-etch” type [5]. Recent studies have shown similar behaviors at the tooth–restoration material interface, between the etch and rinse adhesive system and the self-etch adhesive system, from the perspective of the presence of micro infiltrations [6].

Dental adhesives must make a connection between the tooth and the restorative material that does not allow the appearance of micro-infiltrations. Different techniques are used to analyze the occurrence of microinfiltrations, among which the most accessible is that of penetration with different dyes [7].

The method of penetration with different dyes (such as silver nitrate) consists of immersing the sample in a dye for a specific period of time, following the sectioning of the sample and its optical analysis to confirm the presence or absence of infiltration areas. These tests are semi-qualitative or qualitative and make the studies limited due to the evaluation of the sample, this being analyzed only in the plane where it was sectioned, the dye infiltration not being uniform, the degree of microinfiltration being difficult to measure subjectively. Therefore, other techniques are needed for the analysis of microinfiltrations [8–10].

In addition to the method of impregnation with different dyes, there is testing for marginal integrity using penetration procedures with radioactive isotopes, bacteria or pressure from an air source [11].

In addition to the analysis techniques by the impregnation method, electronic detection methods by electrochemical studies and electronic monitoring of microinfiltrations can also be used. As microscopic evaluations we can use electron scanning microscopy, replication and scanning electron microscopy, fluorescent microscopy, confocal microscopy and optical coherence tomography [12].

Studies performed using stereomicroscopy have demonstrated the presence of microinfiltration areas, mainly in composite restorations performed by direct techniques, compared to prosthetic restorations performed in the dental laboratory [13–15]. In the case of class II cavities, studies performed using scanning electron microscopy have shown more microinfiltrations in the cervical areas compared to the occlusal areas [16]. The analysis of the interface surface between the restoration materials and the dental structures performed with the help of optical coherence tomography showed that there are areas where the interface is optimal, but there are also areas that showed microinfiltrations. Almost all the samples that were analyzed presented such areas, the results being validated with the help of micro-computer tomography investigations [17].

In order to have the best results in the investigation of different adhesives, it is necessary to increase the reflectance. The use of a normal adhesive can influence future results due to the inaccuracy of differentiating the adhesive layer from air inclusions [18].

**Innovative Dental Adhesives**

In recent years, research on adhesive systems has focused on their structural modification. Thus, different research groups have modified the content of different types of adhesive systems by adding to their content particles or nanoparticles that wanted to show changes in different directions including antibacterial effects,
regeneration effects, increased adhesion effects and also visible effects in reducing the thickness of the adhesive film applied on different dental surfaces.

Thus, the functionalization of carboxylic acid with titanium dioxide produces high levels of reactive oxygen species when irradiated with visible light in the field of 400–800 nm. Under these irradiation conditions, reactive oxygen species failed to generate oxidatively induced DNA for cell-free systems at TiO$_2$ concentrations of 0.5 mg/L. On the other hand, the incorporation of TiO$_2$ nanoparticles that have been functionalized with acrylic acid has allowed the use of reactive oxygen species to improve the photopolymerization process of adhesives. Dental adhesives that were loaded with TiO$_2$ nanoparticles, and that functionalized with acrylic acid under the action of visible light, showed an increase in shear strength by about 29% [19].

TiO$_2$ has proven to be the most widely used photocatalyst in dentistry due to its biocompatibility, strong oxidizing effect, as well as low cost and long-term photostability. In addition, it is known to be effective against Gram-negative and Gram-positive bacteria in UV irradiation conditions, the irradiation energy required for proper sterilization being dangerous for the human body [20–23].

By doping the TiO$_2$ nanoparticles with nitrogen using a solvothermal method, a photocatalyst that has higher levels of visible light absorption was produced. This was generated by the presence of nitrogen in the crystal lattice of titanium. The bacterial efficacy of TiO$_2$ nanoparticles was analyzed by embedding them in a dental adhesive. Specimens that were doped with 50, 67 or 80% TiO$_2$ nanoparticles showed strong antibacterial behavior compared to control samples, both in dark irradiation and light irradiation conditions. This conclusion denotes that TiO$_2$ nanoparticles can be used against cariogenic biofilms [24, 25].

Once the acid biofilm has generated dental caries, their treatment involves removing carious lesions and replacing them with a material that restores the defect formed [26,27]. The use of nanotechnology will help inhibit the appearance of cavities by improving remineralization and controlling acid biofilm. To combat bacterial biofilm, silver nanoparticles were integrated into dental adhesives along with quaternary ammonium methacrylate. The dental lesions were remineralized and the acids were neutralized by calcium/phosphate ions released by amorphous calcium phosphate nanoparticles. The combination of amorphous calcium phosphate nanoparticles, quaternary ammonium methacrylate and silver could develop a new class of dental adhesives with dual, remineralizing and antibacterial benefits. Following the same protocol, other nanoparticles such as ZnO, TiO$_2$ and polyethyleneimine were tested, the results being promising for their incorporation into various dental materials such as adhesives and composites [28].

HEMA (2-hydroxyethyl methacrylate) is widely used in dental adhesive systems, especially for binding to dentin. It behaves as an element of compatibility between hydrophilic and hydrophobic components, stabilizing the adhesive system. Its use entails several disadvantages, including water retention in the adhesive layer which can lead to the failure of adhesive restorations. Thus, finding a replacement for HEMA is necessary, with subsequent studies showing that Janus nanoparticles are better than HEMA in stabilizing dentinal adhesives and reducing phase separation. Janus reactive amphiphilic nanoparticles are synthesized by selective etching at the interfaces of a Pickering emulsion. Janus reactive nanoparticles improve the dentin binding interface without cytotoxic effects and can be promising materials to replace HEMA in the content of adhesive systems [29].

The connection between dental adhesives and restorative materials may fail due to imperfections present at the interfaces [30]. The use of nanoparticles by incorporating them in the content of an adhesive system can lead to a change in its mechanical and physical properties. An adhesive system was doped with 5, 10,
15 or 25% by weight glass filler, analyzing the measurements on water solubility and absorption, as well as the dynamic viscosity during the polymerization compared to the undoped system. Tensile strength and fracture energy measurements were used to calculate the tensile strength of the adhesive. The results showed an increased efficiency in increasing the healing degree, as well as accelerating the polymerization reaction without compromising the viscosity of the adhesive film for samples loaded with 5–10% nanoparticles. Nanoparticles increased tensile strength and breaking strength, even though nanoparticles tended to clump together. The strength of the bond between the nanoparticle-loaded adhesive and the restorative composite also increased when loaded with up to 10% by weight loaded adhesives were used, improving the mechanical properties [31].

Adhesive systems are in a continuous race for improvement, to increase their retention to hard dental structures. The development of adhesive dentistry has led to increased aesthetic requirements and an exponential increase in the number of adhesive restorations performed in dental offices [32].

The main reason for the failure of an adhesive restoration is the appearance of microcracks in the adhesive layer and their colonization with bacteria, following the appearance of secondary caries [33].

The thickness of the adhesive layer is the basis of the viability of the adhesive restorations, an increased thickness can lead to the failure of the therapeutic work.

The thickness of the adhesive layer, according to previous studies can be between 0.02 and 0.4 mm, the increased thicknesses of the adhesive layer can generate microcracks [34–36].

In order to achieve a uniform thickness of the adhesive layer, the adhesive systems were loaded with ferric nanoparticles, being applied on the dental surfaces by conventional techniques [37]. Dental adhesives were also loaded with glass microparticles and microspheres, to formulate antibacterial dental adhesives [38]. The presence of nanoparticles in the adhesive system was confirmed by analyzing the teeth using EDX (Energy Dispersive X-ray analysis). After the polymerization of the adhesive, the teeth were analyzed with the help of the latest generation imaging systems, resulting in the measurements of the adhesive surfaces between the two components. The results of the measurements were within the limits generated in the literature, with small decreases in the thickness on certain surfaces. To improve the results, a magnetic field of constant intensity was applied on the dental surfaces, varying only the contact time of the instrument on the dental surfaces. After applying the magnetic field, the thickness of the adhesive layer decreased by 30% for application times of 2 min and by 86.5% for the teeth on which the magnetic field was applied for 5 min [39]. Thus, the inclusion in the therapeutic stages of the magnetic field at certain intensities can lead to the decrease of the thickness in the adhesive layer and to its uniformity in the surface [40].

The applicability of the magnetic field was also tested in the prophylactic procedures for sealing ditches, pits and cracks, generating very good results regarding the thickness of the adhesive layer, uniformizing the adhesive surface and having a much better control over the materials used [41].

Conclusions

Recent advances in dentistry have led to an increase in cosmetic restorative treatments. The development of adhesive materials will lead to the restoration of the natural aspects of the teeth, aiming mainly at the most aesthetic aspect of the teeth [42].
Matching the color with that of natural teeth, but also maintaining the optical properties for longer, will be the major requirements of aesthetic adhesive materials.

Obtaining optical, morphological and biological results are the objectives of aesthetic restorations, imitating dentin and natural enamel [43].

The continuous evolution of these techniques and materials managed not to extract the teeth and to keep them on the arch, these improvements being evident in the adhesive dentistry [44].

The use of filler nanoparticles to strengthen dental adhesive systems can be a great advantage by improving mechanical properties, but also by utilizing new antibacterial and remineralizing properties.

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References
1. D’Arcangelo, C.; Vanini, L.; Casinelli, M.; Frascaria, M.; De Angelis, F.; Vadini, M.; D’Amario, M. Adhesive Cementation of Indirect Composite Inlays and Onlays: A Literature Review. Compend. Contin. Educ. Dent. 2015, 36, 570–577. [PubMed]
2. Ramakrishna, S.; Mayer, J.; Winterrmadel, E.; Leong, K.W. Biomedical applications of polymer-composite materials: A review. Compos. Sci. Technol. 2001, 61, 1189–1224. [CrossRef]
3. Pinzon, L.M.; Watanabe, L.G.; Reis, A.F.; Powers, J.M.; Marshall, S.J.; Marshall, G.W. Analysis of interfacial structure and bond strength of self-etch adhesives. Am. J. Dent. 2013, 26, 335–340. [PubMed]
4. Waldman, G.; Vaidyanathan, T.; Vaidyanathan, J. Microleakage and Resin-to-Dentin Interface Morphology of Pre-Etching versus Self-Etching Adhesive Systems. Open Dent. J. 2008, 2, 120–125. [CrossRef] [PubMed]
5. Sezinnado, A. Looking for the ideal adhesive—A review. Revista Portuguesa de Estomatologia, Medicina Dentária e Cirurgia Maxilofacial 2014, 55, 194–206. [CrossRef]
6. Borges, M.A.P.; Matos, I.C.; Kátia Dias, R.H.C. Influence of two self-etching primer systems on enamel adhesion. Braz. Dent. J. 2007, 18, 113–118. [CrossRef]
7. Hilton, T.J. Can modern restorative procedures and materials reliably seal cavities? In vitro investigations. Part 2. Am. J. Dent. 2002, 15, 279–289.
8. Carrera, C.A.; Lan, C.; Escobar-Sanabria, D.; Li, Y.; Rudney, J.; Aparicio, C.; Fok, A. The use of micro-CT with image segmentation to quantify leakage in dental restorations. Dent. Mater. 2015, 31, 382–390. [CrossRef]
9. Wu, W.; Cobb, E.; Derman, K.; Rupp, N.W. Detecting margin leakage of dental composite restorations. J. Biomed. Mater. Res. 1983, 17, 37–43. [CrossRef]
10. Deshpande, S.; Singh, A.; Mali, P. Microleakage of restorative materials: An in vitro study. J. Indian Soc. Pedod. Prev. Dent. 2006, 24, 15–18. [CrossRef]
11. Mortensen, D.W.; Boucher, N.E.; Ryge, G. A Method of Testing for Marginal Leaks of Dental Restorations with Bacteria. J. Dent. Res. 1965, 44, 58–63. [CrossRef]
12. Kumar, M.; Lakshminarayanan, L. Methods of detecting microleakage. J. Conserv. Dent. 2004, 7, 79–88.
13. Hasanreisoglu, U.; Sönmez, H.; Üçtaşli, S.; Wilson, H. Microleakage of direct and indirect inlay/onlay systems. J. Oral Rehabil. 1996, 23, 66–71. [CrossRef]
14. Milleldeig, P. Microleakage of indirect composite inlays: An in vitro comparison with the direct technique. Acta Odontol. Scand. 1992, 50, 295–301. [CrossRef]
15. Soares, C.J.; Celiberto, L.; Dechiichi, P.; Fonseca, R.B.; Martins, L.R.M. Marginal integrity and microleakage of direct and indirect composite inlays—SEM and stereomicroscopic evaluation. Braz. Oral Res. 2005, 19, 295–301. [CrossRef]
16. Ozel, E.; Tunar, E.B.; Firatli, E. The effects of cavity-filling techniques on microleakage in class II resin restorations prepared with Er:YAG laser and diamond bur: A scanning electron microscopy study. Scanning 2015, 38, 389–395. [CrossRef]
17. Sinescu, C.; Marsavina, L.; Negrutiu, M.L.; Rusu, L.C.; Ardelean, L.; Ionita, C.; Podoleanu, A.G.; Rominu, M.; Topala, F.I. Confocal Microscopy Combined with Time Domain Optical Coherence Tomography and Micro Computer Tomography in Interface Evaluation of Class II Direct Composite Restoration. Rev. Clin. 2011, 62, 1039–1043.
18. Rominu, M.; Sinescu, C.; Negrutiu, M.; Pop, D.M.; Bradu, A.; Petrescu, E.; Rominu, R.O.; Haiduc, C.; Dobre, G.; Podoleanu, A. Class V Cavities Diagnostic by En-Face Optical Coherence Tomography. The Necessity of Increasing the Scattering for Adhesive Layer Media. Timisoara Med. J. 2010, 60, 39–43.
19. Sun, J.; Petersen, E.J.; Watson, S.S.; Sims, C.M.; Kassman, A.; Fruhktbyen, S.; Skrtic, A.; Ok, M.T.; Jacobs, D.S.; Reipa, V.; et al. Biophysical characterization of functionalized titania nanoparticles and their application in dental adhesives. Acta Biomater. 2017, 53, 585–597. [CrossRef]
20. Cai, Y. Titanium Dioxide Photocatalysis in Biomaterials Applications. Doctoral Thesis, Department of Engineering Sciences, Nanotechnology and Functional Materials, Uppsala University, Uppsala, Sweden, 2013; p. 57.
21. Cai, Y.; Stromme, M.; Zhang, P.; Engqvist, H.; Welch, K. Photocatalysis induces bioactivity of an organic polymer based material. RSC Adv. 2014, 4, 57715–57723. [CrossRef]
22. Foster, H.A.; Dittra, I.B.; Varghese, S.; Steele, A. Photocatalytic disinfection using titanium dioxide: Spectrum and mechanism of antimicrobial activity. Appl. Microbiol. Biotechnol. 2011, 90, 1847–1868. [CrossRef] [PubMed]
23. Musk, P.; Campbell, R.; Staples, J.; Moss, D.J.; Parsons, P.G. Solar and UVC—Induced mutation in human cells and inhibition by deoxynucleosides. Mutat. Res. 1989, 227, 25–30. [CrossRef]
24. Florez, F.L.E.; Hers, R.D.; Larson, P.; Johnson, M.; O’Rear, E.; Rondinone, A.J.; Khajotia, S.S. Antibacterial dental adhesive resins containing nitrogen-doped titanium dioxide nanoparticles. Mater. Sci. Eng. C 2018, 93, 931–943. [CrossRef] [PubMed]
25. Dinh, C.-T.; Nguyen, T.-D.; Kleitz, F.; Do, T. Shape-Controlled Synthesis of Highly Crystalline Titania Nanocrystals. ACS Nano 2009, 3, 3737–3743. [CrossRef]
26. Watts, D.C.; Marouf, A.; Al-Hindi, A. Photo-polymerization shrinkage-stress kinetics in resin-composites: Methods development. Dent. Mater. 2003, 19, 1–11. [CrossRef]
27. Drummond, J.L. Degradation, Fatigue, and Failure of Resin Dental Composite Materials. J. Dent. Res. 2008, 87, 710–719. [CrossRef]
28. Cheng, L.; Zhang, K.; Weir, M.D.; Melo, M.A.; Zhou, X.; Xu, H.H. Nanotechnology strategies for antibacterial and remineralizing composites and adhesives to tackle dental caries. Nanomedicine 2015, 10, 627–641. [CrossRef]
29. Han, B.; Xia, W.; Liu, K.; Tian, F.; Chen, Y.; Wang, X.; Liang, F.; Yang, Z. Janus Nanoparticles for Improved Dentin Bonding. ACS Appl. Mater. Interfaces 2018, 10, 8519–8526. [CrossRef]
30. Bhandary, S.; Hegde, M.N. An evaluation and comparison of shear bond strength of composite resin to dentin, using newer dentin bonding agents. J. Conserv. Dent. 2008, 11, 71–75. [CrossRef]
31. Belli, R.; Kreppel, S.; Perschelt, A.; Hornberger, H.; Boccaccini, A.R.; Lohbauer, U. Strengthening of dental adhesives via particle reinforcement. J. Mech. Behav. Biomater. 2014, 37, 100–108. [CrossRef]
32. Caldas, I.P.; Alves, G.G.; Barbosa, I.B.; Selza, P.; De Noronha, F.; Selza, M.Z. In vitro cytotoxicity of dental adhesives: A systematic review. Dent. Mater. 2019, 35, 191–205. [CrossRef]
33. Yue, S.; Wu, J.; Zhang, Q.; Zhang, K.; Weir, M.D.; Imazato, S.; Bai, Y.; Xu, H. Novel dental adhesive resin with crack self-healing, antimicrobial and remineralization properties. J. Dent. 2018, 75, 48–57. [CrossRef]
34. Choi, K.; Condon, J.; Ferracane, J. The Effects of Adhesive Thickness on Polymerization Contraction Stress of Composite. J. Dent. Res. 2000, 79, 812–817. [CrossRef]
35. Van Meerbeek, B.; Van Landuyt, K.; De Munck, J.; Hashimoto, M.; Peumans, M.; Lambrechts, P.; Yoshida, Y.; Inoue, S.; Suzuki, K. Technique-Sensitivity of Contemporary Adhesives. Dent. Mater. J. 2005, 24, 1–13. [CrossRef]
36. Grossman, E.S.; Setzer, S. Bonding agents: Adhesive layer thickness and retention to cavity surfaces with time. SADFF 2001, 56, 266–272.
41. Oancea, R.; Zaharia, C.; Gabor, A.-G.; Sinescu, C.; Mioc, M.; Vaduva, D.B.; Simon, C.P.; Socoliu, V.; Rominu, M.; Negrutiu, M.-L. Imagistic Analysis of Dental Adhesives Loaded with Nanoparticles Used on Teeth Sealing of Pits and Fissures with Resin Based Materials. Mater. Plast. 2019, 56, 449–453. [CrossRef]

42. Migliu, G.; Besharat, L.K.; Sofan, A.A.A.; Sofan, E.A.A.; Romeo, U. Endo-restorative treatment of a severely discolored upper incisor: Resolution of the “aesthetic” problem through Componeer veneering System. Ann. Stomatol. 2016, 6, 113–118. [CrossRef] [PubMed]

43. Migliu, G.; Piccoli, L.; Besharat, L.K.; Romeo, U. Benchmarking matching color in composite restorations. Ann. Stomatol. 2016, 7, 29–37. [CrossRef] [PubMed]

44. Migliu, G. Evaluation of over-etching technique in the endodontically treated tooth restoration. Ann. Stomatol. 2015, 6, 10–14. [CrossRef]