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The influence of the inorganic fillers on the light-curing transmission through resin-matrix composites: A systemic review

Subtítulo

Daniela Minerva Evangelista Carpio

Dissertação conducente ao Grau de Mestre em Medicina Dentária (Ciclo Integrado)

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Trabalho realizado sob a Orientação de Professor Doutor Júlio Souza e Co-orientação da Dra. Rita Fidalgo Pereira

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AGRADECIMENTOS

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Resumo

Objetivo:

O objetivo deste estudo é efetuar uma revisão sistemática sobre o efeito da componente inorgânica na transmissão da luz na polimerização de resinas compostas.

Materiais e métodos:

Foi realizada uma revisão bibliográfica no PubMED utilizando os seguintes termos de pesquisa: “fillers” OR “particle” AND “light curing” OR “polymerization” AND “light transmission” OR “light absorption” OR “light intensity” OR “light attenuation” OR “light diffusion” AND “resin composite”.

Resultados:

As propriedades mecânicas das resinas compostas são afetadas pelo tamanho e morfologia das partículas inorgânicas. A resistência das resinas composta mostrou diferenças significativas na profundidade de polimerização. Os valores mais altos de resistência à flexão biaxial (154 MPa) foram registados para materiais com Bis-GMA, UDMA, TEGDMA enquanto que os valores mais baixos (77 MPa) foram registados para Bulk fill™. A utilização da lâmpada de halogéneo nos estudos promoveu um grau de conversão mais elevado do que os alcançados pela lâmpada LED. Compostos contendo fotoiniciadores inovadores, como o lucirin TPO™ (TPO), demonstraram um grau de conversão significativamente mais elevado.

Conclusões:

A quantidade de luz transmitida através da matriz de resina é influenciada pelo tamanho, conteúdo, microestrutura e forma das partículas inorgânicas. A diminuição do grau de conversão afeta negativamente as propriedades físicas e mecânicas das resinas compostas. Os monómeros residuais são progressivamente libertados para os tecidos orais circundantes e podem provocar uma resposta celular tóxica.

Abstract

Purpose:

The objective of this study was to perform a systematic review on the effect the inorganic fillers on the light curing transmission through the resin-matrix composites.

Method:

A bibliographic review was performed on PubMED using the following search terms: “fillers” OR “particle” AND “light curing” OR “polymerization” AND “light transmission” OR “light absorption” OR “light intensity” OR “light attenuation” OR “light diffusion” AND “resin composite”.

Results:

The mechanical proprieties of resin matrix composites are highly affected by the size and morphology of inorganic particles. The strength of the resin-matrix composites showed significant difference regarding the depth of polymerization. The highest values of biaxial flexural strength (154 MPa) were recorded for materials with Bis-GMA, UDMA, TEGDMA while the lowest values (77 MPa) were recorded for Bulk fill™. The use of the halogen lamp in the studies promoted higher degree of conversion than those achieved by LED lamp. Composites containing novel photoinitiators such as lucirin TPO™ (TPO), demonstrated significantly higher degree of conversion.

Conclusions:

The amount of light transmitted through the resin-matrix is influenced by the size, content, microstructure and shape of the inorganic filler particles. The decreasing of the degree of conversion affects negatively the physical and mechanical properties of the resin-matrix composites. Residual monomers are progressively released to the surrounding oral tissues and can cause a toxic celular response.

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List of abbreviations, acronyms, initials and acronyms

Bis-GMA = Bisphenol A-glycidyl dimethacrylate

TEGDMA = Triethylene glycol dimethacrylate

UDMA = Urethane dimethacrylate

Bis-EMA = ethoxylatedbisphenol A dimethacrylate

CQ = Camphorquinone

TPO = Lucerin TPO

LED = Light-Emitting Diodes

QTH = Quartz–Tungsten–Halogen

LCU = Light Curing Unit

SEM & FEGSEM = Scanning electron microscopy

FIB = Focused ion beam

AFM = Atomic force microscopy

DC = Degree of conversion

RDB = Residual double bonds

1. Introduction

Dental resin-matrix composites have become the most requested materials for direct and indirect restorations as a result of the highly development of their mechanical and optical properties (1–3) However, concerns have been reported regarding light transmission during polymerization. Light scattering caused by inorganic fillers is a phenomenon that can cause changes in light transmission through the resin-matrix composites during the light curing procedure. Therefore, the chemical composition, size, and morphological aspects of inorganic fillers should be clarified. As reported the origin of the mechanism of cytotoxicity is caused by the unbound free monomers released by resin-matrix composites during polymerization and long-term performance. The release of the monomers as a consequence of unproper polymerization can cause cytotoxicity of the surrounding tissues and an inflammation reaction over time (4–6). The release of monomers may cause disruption of pulp and gingival cells and are probably also involved in the allergic potential of the material (5,7).

The resin-matrix composites are composed of inorganic vitreous fillers dispersed in an organic matrix. The organic matrix is composed of dimethacrylate monomers such as bisphenol A-glycidyl dimethacrylate (Bis-GMA), triethylene glycol dimethacrylate (TEGDMA), urethane dimethacrylate (UDMA) and ethoxylatedbisphenol A dimethacrylate (Bis-EMA) (8–11). A photoinitiator system, such as, camphorquinone (CQ) or lucerin TPO is also added in the organic matrix to induce the polymerization under visible light irradiation in the range between 420-500nm (1,8) Nowadays, the most used light sources are Light-Emitting Diodes (LEDs) within an intensity of 400-1765 mW/cm2 for 20-40 s. The wavelength of commercially available light curing unit ranges from 0.4µm up to 0.8µm. The time of light exposure depends on the light curing intensity to reach the energy required for the polymerization of the resin-matrix composite (9,10). The degree of conversion is the percentage of double carbon bonds that react to become converted into single bonds to form a polymeric resin. Also, DC represents the proportion of polymerized monomers after light curing (12–14). The degree of

conversion has been used for complementary analyses of the physical properties of resin-matrix composites and correlation with clinical performance and biocompatibility (15).

The inorganic fillers content of resin matrix composites consists in silanized inorganic particles at different size and morphological aspects (i.e., spherical and irregular fillers). The most used inorganic fillers are composed of silica (16). Commercially available resin-matrix composites have a filler content ranging from 40 up to 90 wt% (17,18). A combination of different inorganic fillers (i.e., silica and glass ceramics) at different sizes can be found in the chemical composition of recent materials (1). For instance, spherical silica particles at 20-60 nm in combination with micro-scale glass-ceramics (i.e., zirconium or barium silicates at 1-2 µm) are common fillers added into the chemical composition of resin-matrix composites (19). Nano- and micro-scale particles are combined in the resin-matrix composites microstructure to provide a mechanical reinforcement under further mastication loading (20). Thus, a high content of nano- and micro-scale particles have a high surface provides a low organic matrix volume under polymerization (21). However, the effects of the fillers' morphological aspects such size and shape on the polymerization are not entirely clarified in literature. Also, the chemical composition, content, and microstructure of the fillers affect the light transmission that should be understood regarding the development of novel resin-matrix composites.

1.1. Purpose and hypothesis

The objective of this study was to perform an integrative review on the effect the inorganic fillers on the light curing transmission through the resin-matrix composites. It was hypothesized that size and chemical composition of inorganic fillers can affect light curing transmission through resin-matrix composites.

2. Method

A bibliographic review was performed on PubMED (via National Library of Medicine) considering such database includes the major particles in the field of dentistry and biomaterials. The present search of studies was carried out in accordance with previous integrative or systematic review articles (22–24). The following search terms were applied: “fillers” OR “particle” AND “light curing” OR “polymerization” AND “light transmission” OR “light absorption” OR “light intensity” OR “light attenuation” OR “light diffusion” AND “resin composite”. Also, a hand-search was performed on the reference lists of all primary sources and eligible studies of this systematic review for additional relevant publications. The inclusion criteria encompassed articles published in the English language from January 2011 up to August, 2021, focusing on the effects of the size and chemical composition of fillers on the light curing transmission through resin-matrix composite. The eligibility inclusion criteria used for article searches also involved: *in vitro* studies; meta-analyses; randomized controlled trials; animal assays; and prospective cohort studies. The exclusion criteria were the following: papers without abstract; case report with short follow-up period; pilot studies; studies on the effect of fillers through other composite materials applied in different biomedical or engineering fields. Studies based on publication date were not restricted during the search process.

2.1. Study selection and data collection process

The selection of studies was carried into three steps. At first, studies were scanned for relevance by title, and the abstracts of those that were not excluded at this stage were assessed. Three of the authors (JCMS, DC, RP) independently analyzed the titles and abstracts of the retrieved, potentially relevant articles meeting the inclusion criteria. A fourth author intervened in case of disagreements during the scanning of the articles. The total of articles was compiled for each combination of key terms and therefore the duplicates were removed using Mendeley citation manager (ed. Elsevier). The second step comprised the evaluation of the abstracts and non-excluded articles, according to the eligibility criteria on the abstract review. Selected articles were

individually read and analyzed concerning the purpose of this study. At last, the eligible articles received a study nomenclature label, combining first author names and year of publication. The following variables were collected for this review: authors' names, journal, publication year, aims, organic matrix type, fillers (size, chemical composition, types), light curing (methods and equipment), light transmission analyses, polymerization evaluation (degree of conversion), and related mechanical characterization. PICO question was adjusted to the issue where "P" was related to the materials and "I" referred to the methods of analyses while "C" was related to comparison of findings and "O" o the main outcomes. Data of the reports were harvested directly into a specific data-collection form to avoid multiple data recording regarding multiple reports within the same study (e.g., reports with different set-ups). This evaluation was individually carried out by two researchers, followed by a joint discussion to select the relevant studies.

3. Results

The initial search in the available database yielded a total of 136 articles of which 56 duplicate articles were eliminated. Of the remaining 80 articles, the titles and abstracts were read seeking concordance with the inclusion criteria of the present study and then 53 studies were discarded because they did not meet the inclusion criteria. The evaluation of titles and abstracts resulted in the selection of 27 potentially articles although nine articles were excluded because they did not provide comprehensive data. The results of the selection of articles are shown in Figure 1.

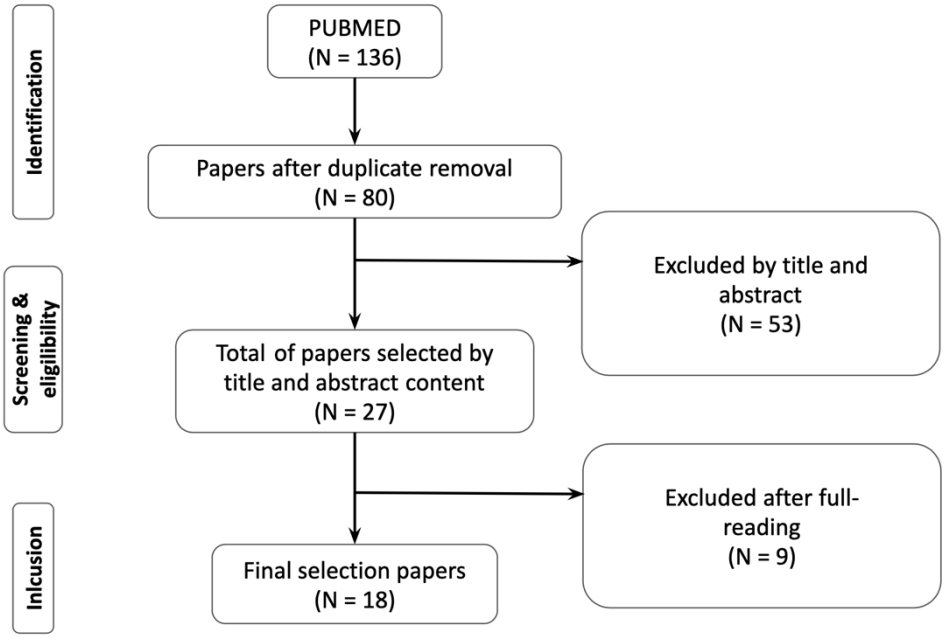

Figure 1. Flow diagram of the search strategy used in this study.

Of the 18 articles included in this review, five studies (27.78%) evaluated the chemical composition, properties, and light-curing transmission of bulk-fill resin-matrix composites (9,25,26). One study focused on the effect of filler amount on the effectiveness of light-transmission of resin-matrix composites (27). Two studies (11.11%) assessed different light-curing units considering the light-transmission through resin-matrix composites such as mercury arc lamp (28) and a diode-pumped solid state (DPSS) laser (29). Three studies (16.67%) assessed the effect of spherical glass fillers (SGMFs) to decreasing the shrinkage of resin-matrix composites in direct dental restorations (30,31) while one study also performed the evaluation of optical properties (32). Only two articles (11.11%) assessed zirconia as a filler for enhancement of the mechanical properties and the degree of conversion depending on the light transmission through the material (33,34). One *in vitro* study (35) evaluated the presence of two different photoinitiations: lucirin TPO^{TM} (TPO) or camphorquinone (CQ). Those studies analyzed the resin-matrix composite under different thermal specific conditions, by increasing and decreasing the temperature (36,37). Different amount and size of silica

fillers was carefully evaluated by one study (10,38) regarding light-transmission and polymerization of the organic matrix.

The major findings are shown in Table 1 and described as follow:

- The particle size and the fraction of the inorganic filler are directly related during the light transmission. The size and the morphology of the inorganic fillers highly affected filler loading which also can affect the mechanical properties of the resin-matrix composites (9,10,16,28,30);
- The strength of the resin-matrix composites showed significant difference regarding the depth of polymerization. The resin-matrix composites which contained Bis-GMA, UDMA, TEGDMA revealed the highest flexural modulus (5.11 GPa) while Bulk fill™ composites revealed the lowest flexural modulus (2.5 GPa). The highest values of biaxial flexural strength (154 MPa) were recorded for materials with Bis-GMA, UDMA, TEGDMA while the lowest values (77 MPa) were recorded for Bulk fill™. The Bulk fill™ composites did not reveal a significant difference among depth of polymerization (9,28,30,34,39);
- The translucency of the glass-ceramic spherical fillers promoted light diffusion within light-curing resin-matrix composites, mainly in critical situations such as in the case of deep proximal cavities (26,31);
- The polymerization efficiency of resin-matrix composites under the ramp-curing mode was higher when compared to the low-intensity curing mode. The use of the halogen lamp in the studies promoted a higher degree of conversion than those achieved by LED lamp (9,30,37);
- The light transmission decreased as the thickness of the composite (regular or Bulk fill™) increased. In addition, a progressive decrease in the degree of conversion of the composites occurred with increasing silica particle size. Composites containing novel photoinitiators such as lucirin TPO™ (TPO), showed significantly higher degree of conversion when compared to composites containing camphorquinone (CQ) (35,37).

4. Discussion

The present integrative review reported the major results of relevant previous studies taking into account the effect of the inorganic fillers of resin-matrix composites on the degree of conversion of monomers during light-curing transmission. The type, size, shape, and content of fillers do affect the degree of conversion and shrinkage of the organic matrix. Also, the light source, intensity level (light-curing mode), the photoinitiator efficacy, and the thickness do affect the mechanical properties of the resin-matrix composite. Therefore, the findings validate the hypothesis of this study. A detailed discussion of the main factors that affect the properties of the resin-matrix composites on light curing transmission is given as follow.

4.1. Resin-matrix composites

The traditional formulation of resin-matrix composites involves silanized inorganic filler particles embedded in an organic matrix (14,40) Organic matrix is often composed of dimethacrylate monomers bisphenol A-glycidyl dimethacrylate (Bis-GMA), triethylene glycol dimethacrylate (TEGDMA), urethane dimethacrylate (UDMA), and ethoxylatedbisphenol A dimethacrylate (Bis-EMA). The most common photoinitiator system consists in camphorquinone (CQ) associated with a tertiary amine. Recent photoinitiators such as lucirin TPO^{TM}, are also mentioned in reviewed studies, as shown in Table 1 and Figure 2 (1,8). Commercially available resin-matrix composites can have spherical or irregular filler inorganic particles (18), with average size between 40 nm up to 60 µm (8). A combination of micro- (1-10 µm) and nano-scale (40-60 nm) fillers is often found in the resin-matrix composites microstructure, (1,17). Commercially available resin-matrix composites have a filler content ranging from 40 up to 90wt%. Glass-ceramic fillers based on aluminum, barium silicate, borosilicate, colloidal silica, ytterbium fluoride, and zirconium silicate can be found in the resin-matrix composites inorganic content (1), as shown in Table 1. Recently, resin-matrix composites with bioactive glass fillers have been studied aiming to achieve optimal bioactivity without compromising other important properties, such as degree of conversion of the monomers. However, the effect of bioactive glass fillers on the degree of conversion and other mechanical properties needs to be further investigated (26,31).

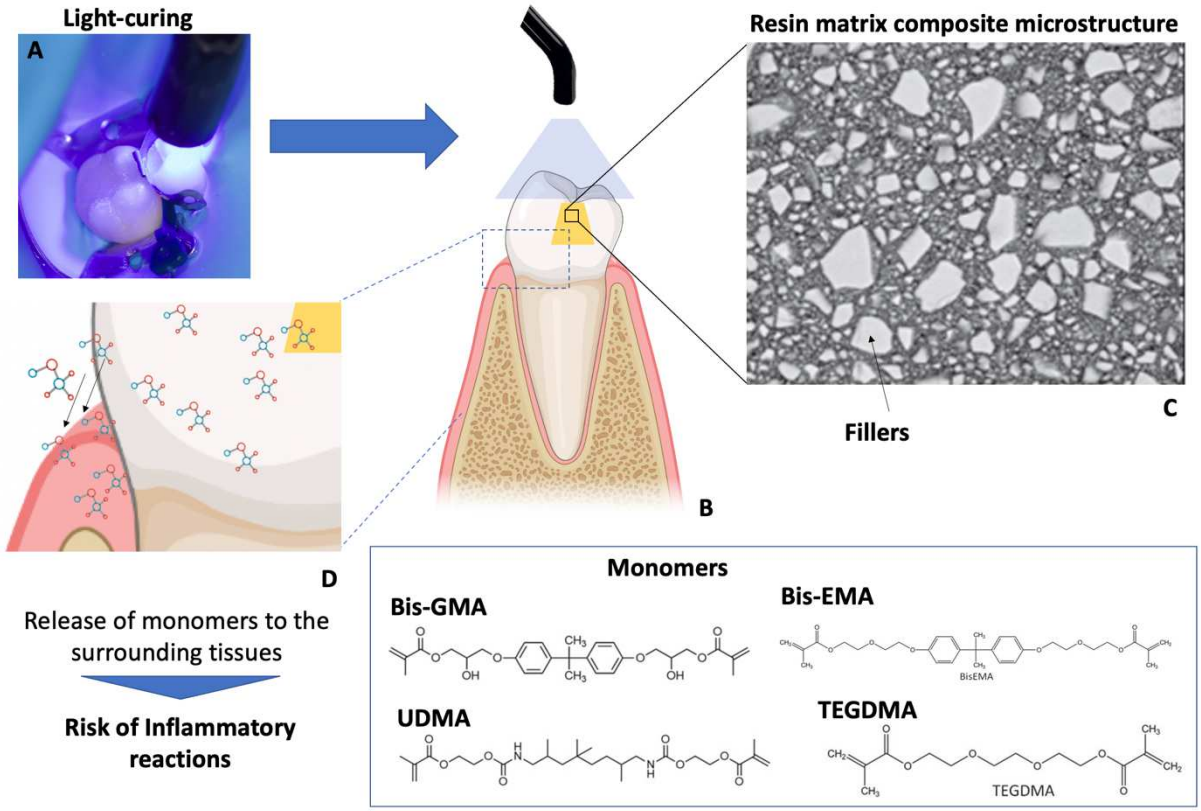

Figure 2. A) Light-curing. (B) Polymerization of resin-matrix composite and the inflammatory reaction. (C) Resin matrix composite microstructure. (D) Chemical composition of monomers (36).

Physicochemical properties of resin-matrix composites are dependent on the proportion and chemical composition of organic matrix and inorganic fillers (14,41). A continuous occlusal loading results in a progressive degradation and generation of micro-cracks, which causes failures of the resin-matrix composites (42). Previous studies have performed static and dynamic mechanical tests to estimate the polymerization status of the resin-matrix composites, as seen in Figure 3. Static mechanical assays involve compressive, flexural and tensile tests although flexural strength is quite assessed by biaxial, three-, or four-point bending strength tests (43). Flexural strength and elastic modulus increased with the amount of inorganic fraction (34,44). The fracture toughness has been also assessed to evaluate the resistance to the propagation of cracks through the resin-matrix composites (45). As expected, resin-matrix

composites with high content of fillers showed enhanced mechanical properties (45). Also, the volume of the organic matrix decreased that affects its shrinkage, light transmission, and degree of conversion on polymerization.

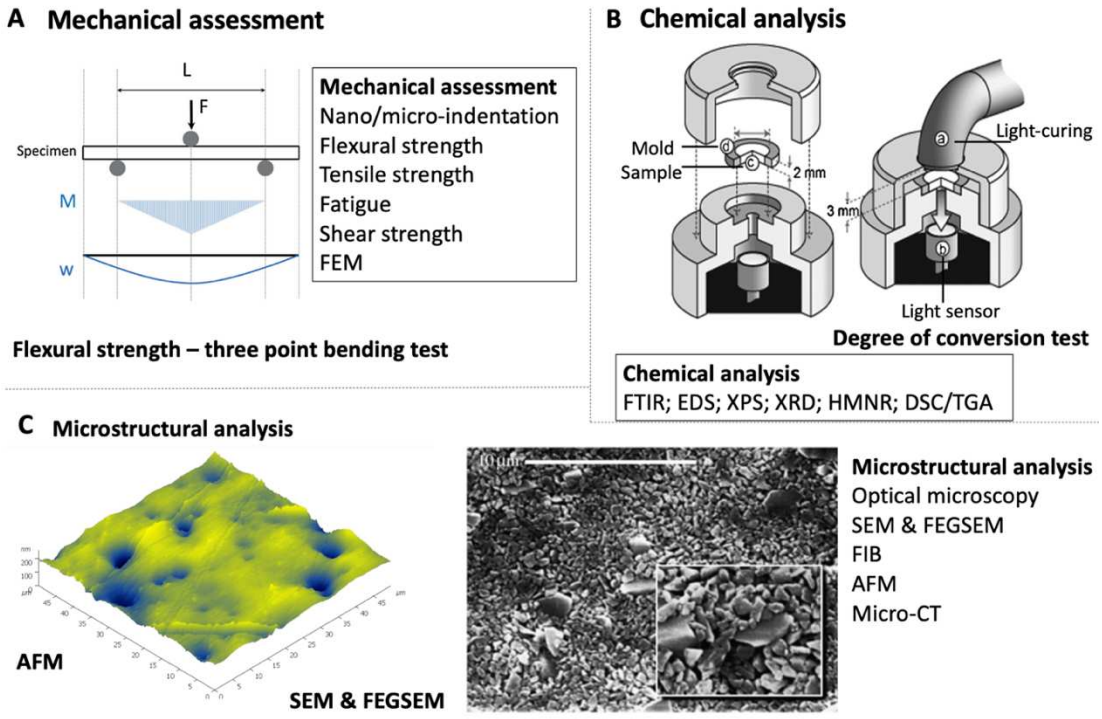

Figure 3. Set up testing for (A) Flexural strength by three-point bending test. (B) Specimen preparation (34). (C) Images acquired by atomic for microscopy (AFM) and scanning electron microscopy (SEM) (12,42)

4.2. Polymerization

Resin matrix composites polymerization is a complex process and it is dependent on intrinsic and extrinsic factors, such as: wavelength of emitted light, type of photoinitiator, bulb intensity, exposure time, distance, and type of the resin-matrix composite (12). In the majority of previous studies, resin-matrix composites were light-cured using Light-Emitting Diodes (LEDs) and Quartz–Tungsten–Halogen (QTH). Light-curing units' (LCU) intensity was reported at approximately 900-1765 mW/cm^2 for LED and 100-700 mW/cm^2 for QTH y (9,10). Light-curing irradiation time exposure was

performed for 20, 40, or 60 s between each resin-matrix composite increment. Distance between LCU and surface of resin-matrix composite was around 1-3 mm and the LCU angulation tip has been perpendicularly positioned to the restoration surface plane (12,18) Additionally, LCU requires a proper and precise calibration prior to any procedure to optimize the polymerization.

In general, the following polymerization modes were performed: high intensity, low intensity and pulse mode. However, that it depended on the brand. The dental pulp may be affected due to the high heating generated over a long period of exposure. For this reason, the use of low intensity is necessary to preserve pulp vitality and to avoid postoperative sensitivity (46,47). Inadequate operation of light-curing units can cause iatrogenesis and can minimize the long performance of the restoration (48). Even though recent resin-matrix composites show a minimum shrinkage level as a result of molecular cross-linking and densification (1,8,49,50), shrinkage stresses still take place during polymerization (1,50). Polymerization shrinkage occurs due to the cross-linking of monomers and then stresses are generated at the restorative interfaces (9). Other factors determine the magnitude of the shrinkage stresses such as pre-gel flowing, cavity design, molecular weight, and chemical composition (14,49) Differences in shrinkage stresses regarding filler particle size, content and shape were statistically significant. Lower shrinkage stresses were noticed in materials containing spherical fillers when compared to irregular filler particles (28,49). Several studies recommend incremental filling techniques to decrease the shrinkage stresses and to achieve an optimum degree of conversion of the organic matrix (9,50,51). The average maximum thickness of around 2 mm is recommended for each resin-matrix composite increment (9). On conventional resin-matrix composites, the light could not reach the depth of the restoration when the material thickness is over 4-5 mm and therefore the polymerization is not entirely accomplished (9,46). The light transmission reaches its maximum value of around 68 % immediately after the light curing irradiation. However, the inorganic particles can interfere in the light transmission depending on their translucency, size and microstructure. The visible light transmission gradually decreases as the material polymerization occurs (18).

The previous selected studies reported results on different physicochemical methods of characterization related to the influence of inorganic fillers on the material polymerization, as shown in Figure 3 (17,41). On mechanical assessment, flexural strength and microhardness values has been recorded to evaluate the degree of conversion (DC) of the monomers (17,41). On the morphological inspection of filler particles, the following tests were used: scanning electron microscopy (SEM & FEGSEM), optical microscopy, focused ion beam (FIB), atomic force microscopy (AFM), and Micro-CT (17). The degree of conversion (DC) represents the proportion of polymerized monomers after setting that is measured by subtracting the residual double bonds (RDB) value from 100 (15,26,52). Moreover, those parameters (DC and RDB) could be used to generate a prognostic on the behavior of dental restorations (9,52) The DC values of the resin-matrix composites range between 52 and 75% (26,36,52), although DC might reach a higher value for 24 h from the polymerization procedure (15,17). The DC magnitude is proportional to the resin-matrix composite polymerization shrinkage since a high amount of monomers are binding (26). However, DC is altered by the chemical composition of the resin-matrix composite and the light source. Additionally, the particle size of inorganic fillers can impact the light-curing transmission and evidently influence the degree of conversion (26). On the decrease of DC, a small crosslink density of the polymer take place decreasing their wear resistance and color stability that is responsible for failures and change of optical properties (26).

In fact, light transmission through a resin-matrix composites depends on light reflection, scattering and absorption, that vary accordingly to the chemical composition of the material. Increasing the size of silica particles reduces the extent of polymerization at the deeper region of the resin-matrix composites. Filler particles with diameter approaching half the wavelength of light transmittance tends to increase light scattering and transmittance. Another study revealed that the translucency and depth polymerization properties of resin-matrix composites were inversely affected by the particle size. Such findings were validated by the light scattering and reflection from the presence of smaller particles (50). On the other hand, a high filler content reduces light transmission due to the increase of light refraction at interfaces between the filler

particles and the resin matrix with different refractive index (9). A previous study reported that the inorganic filler and organic matrix must have similar refractive index match to achieve a high translucency in bulk fill composites. Bulk fill resin composites have reduced filler content aiming to achieve a high light transmission when compared to traditional resin-matrix composites (38).

The release of monomers from resin-matrix composites to the surrounding tissues can occur due to several factors, such as: insufficient polymerization, thermal oscillations, fatigue, wear, and corrosion (4,5,53). Further research is needed to precisely evaluate the degree of conversion of monomers and their chemical effects on the surrounding tissues (5,6,11). Monomers can be released to saliva and oral tissues, causing a risk of an inflammatory reactions, as illustrated in Figure 2. Cytotoxicity is related to initial short-term release of free monomers during monomer-polymer conversion, immediately after polymerization and over time (4,7). Studies have shown that most monomers react with the polymeric network during initial polymerization step. Thus, the percentage of free monomers is approximately 1.5-5% although such a low amount of monomer molecules could be enough to provide a cytotoxic effect (5,6). In vitro studies revealed alterations in gingival and pulp cells caused by free monomers (4–6).

5. Conclusion

Within the limitations of the in vitro selected studies, the following concluding remarks can be drawn as follow.

The amount of light transmitted through the resin-matrix is strongly influenced by the size, content, microstructure and shape of the inorganic filler particles. As a consequence, the light source could not be enough for a proper degree of conversion of monomers into a highly cross-linked polymer. Thus, the decrease in the degree of conversion negatively affects the physical properties the resin-matrix composite with a significant impact on their strength and optical properties. Residual monomers are

progressively released to the surrounding medium triggering a toxic cell response in the oral tissues. The studies highly recommend the control of the following light curing factors: light intensity, irradiation time, distance between material surface and light curing unit, and the compatibility between light wavelength and photoinitiator compounds. Also, the selection of the resin-matrix composite become a key role considering the effects of the size, shape, and chemical composition of the inorganic fillers. Further studies should carefully evaluate the correlation of the polymerization depth and the physical properties of recent resin-matrix composites taking into account the development of highly effective and less defective composites for dental applications.

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