Analysis of shrinkage stresses arising during polymerization of orthodontic adhesive systems

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Abstract: The aim of the study was to evaluate the shrinkage stress during cross-linking of composite orthodontic adhesive systems. The elastic-optical method was used for shrinkage stress analysis. Based on the obtained results, basic statistics were determined, including averages and standard deviations. For the comparative analysis of the mean values, the Tukey HSD test was used at the significance level $\alpha = 0.05$. The average value of shrinkage stress during cross-linking ranged from 7.2 to 11.5 MPa.

Keywords: orthodontics, adhesive systems, shrinkage stress, polymerization.

Analiza naprężeń skurczowych powstających podczas polimeryzacji ortodontycznych systemów adhezyjnych

Streszczenie: Celem pracy była ocena naprężeń skurczowych podczas sieciowania kompozytowych ortodontycznych systemów adhezyjnych. Do analizy naprężeń skurczowych zastosowano metodę elastooptyczną. Na podstawie uzyskanych wyników wyznaczono statystyki podstawowe, w tym średnie oraz odchylenia standardowe. Do analizy porównawczej wartości średnich użyto testu Tukeya HSD na poziomie istotności $\alpha = 0{,}05$. Średnia wartość naprężeń skurczowych podczas sieciowania ortodontycznych systemów adhezyjnych wynosiła od 7,2 do 11,5 MPa.

Słowa kluczowe: ortodoncja, systemy adhezyjne, naprężenia skurczowe, polimeryzacja.

There is no doubt that fixed orthodontic appliances are commonly used in treatment of malocclusion in both developmental and adult patients.

Although the first fixed orthodontic appliance was described by Luis Bourdet and then by Pierre Fauchard in the 18th century [1], it was introduced into use in mid-19th century in the United States. In Europe and the rest of the world, orthodontic appliances became widely used much later. One of the problems associated with using the first fixed orthodontic appliances was a lack of adequate adhesive resins and the need to attach ring-shaped abutments to all teeth. Such a situation had a negative impact not only on treatment mechanics. The bands on all teeth irritated periodontal tissues, favored accumulation of plaque, and after treatment was completed, it was necessary to close gaps between teeth.

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The introduction of composite resins to dentistry revolutionized treatment techniques in both conservative dentistry and orthodontics [2, 3].

Good mechanical properties, appropriate aesthetics of fillings, comfort and control of working time made it difficult to imagine a dental practice not using this type of materials. Composites were well received by dentists and have replaced amalgams as fillings of cavities in hard tissues of teeth.

Orthodontic adhesive systems used today are similar in their chemical structure to materials used as fillings in restorative dentistry. Most frequently they are materials whose polymerization is initiated by visible light – a curing unit, or bases on double polymerization systems [4, 5].

The introduction of adhesive techniques to orthodontics resulted in replacing bands used as elements of fixed appliances by brackets bonded to the labial surfaces (or, depending on the work technique, lingual surfaces) of premolars, canines, and incisors. In some clinical cases, bands used on molars may be replaced by tubes attached with light-cured resin. In the case of attaching abutments to impacted teeth [6], the use of adhesive techniques made it possible to replace previously applied traumatizing methods, such as drilling holes for ligature wire in tooth crowns or directly binding the cervix of tooth to which extrusion forces were applied.

Unfortunately, so far, no dental material has been produced that meets all the expectations, both in terms of clinical use and biological safety of use in patients.

Composite materials are not devoid of disadvantages, which include [7]: i) incomplete conversion of the polymer network during relatively long polymerization process, ii) chemical instability associated with incomplete cross-linking of the material, resulting in release of potentially harmful monomers into the body, iii) insufficient mechanical strength resulting in loss of mass of composite during its use in the oral cavity, iv) susceptibility to discoloration, v) sensitivity of the material and adhesive systems to application conditions, *e.g.*, moisture, vi) polymerization shrinkage responsible for marginal leakage between tooth tissues and the composite, and vii) development of internal forces/stresses in mass of the material responsible for its detachment from enamel or dentin during the cross-linking process.

As in the case of restorative materials based on composites, during the polymerization process internal stresses are generated in the mass of orthodontic adhesives, caused by a change in the spatial relations of their components.

Although the phenomenon of polymerization shrinkage of orthodontic adhesive systems does not seem to have such negative effects as in the case of composite materials for filling cavities, some authors point to its potential influence on development of micro-leakage between the bracket base and the enamel surface as well as development of demineralization white spot lesions [8–10].

Although forces generated during the polymerization process of orthodontic adhesive systems do not cause direct damage to the tooth enamel [10], the phenomenon of polymerization shrinkage of such resins is responsible for formation of stress not only in composite material itself, but also in enamel adjacent to the bracket bases [11].

Some authors also note that in the case of orthodontic adhesive systems, the C – factor (configuration factor expressing the ratio between bonded surface and free surface) is very high in the case of orthodontic adhesive systems [12], which may adversely affect the value of connection between bracket bases and the enamel surface of teeth during the polymerization process of the material. However, it should be remembered that an analysis of unfavorable phenomena arising during adhesive procedures in clinical conditions should not be based only on an assessment of the configuration factor. This approach does not consider properties of individual composites, their shape/geometry under the bracket bases, properties of the base and stress distribution in the enamel-resin-bracket system.

In the clinical setting, orthodontic adhesive systems based on composites are subjected to continuous masticatory forces and forces transmitted by arches when teeth change their position. Potential weakening of the enamel structure during adhesive procedures due to internal stresses of bonding materials may increase the risk of tooth tissue damage during brackets' debonding [11] and contribute to premature loss of brackets during orthodontic treatment.

The aim of the study was to assess the shrinkage stresses of four orthodontic adhesive systems based on composite materials, appearing after initiating the process of polymerization of materials with visible light.

T a b l e 1. Characteristic of orthodontic adhesive systems

Name	Composition
Resilience Light-Cure Bracket Adhesive	Bisphenol A Diglycidyl Methacrylate (bis-GMA) triethylene glycol dimethacrylate (TEGDMA) Camphorquinone
Ortho Connect	ethoxylated bisphenol-A dimethacrylate (Bis-EMA) 10–30 wt% urethane dimethacrylate (UDMA) 10–30 wt% phosphoric acid ester monomer** 1–5 wt% photoinitiator** ≤ 1 wt% stabilizer** ≤ 1 wt%
Grēngloo	Poly(oxy-1,2-ethanediyl), α, α'-[(1-methylethylidene)di-4,1-phenylene]bis [ω-[(2-methyl-1-oxo-2-propen-1-yl)oxy] 1–5 wt% Silica, amorphous, fumed, cryst. -free 1–5 wt% 2,3-epoxypropyl methacrylate Propylidynetrimethanol, ethoxylated, esters with acrylic acid 7,7,9(or 7,9,9)-trimethyl-4,13-dioxo-3,14-dioxa-5,12-diazahexadecane-1,16-diyl bismethacrylate phenyl bis(2,4,6-trimethylbenzoyl)-phosphine oxide bisphenol A
Transbond XT Light Cure	Bis-GMA 45–55 wt% TEGDMA 45–55 wt% 4-(dimethylamino)-benzeneethanol < 0.5 wt%

EXPERIMENTAL PART

Materials

The study assessed four orthodontic adhesives: Transbond XT Light Cure (3M Unitek, USA), Grēngloo (Ormco, USA), Resilience (Ortho Technology, USA) and Ortho Connect (GC, Japan). The characteristic of the orthodontic adhesive systems is presented in Table 1.

Methods

To evaluate contraction stress, transparent and photosensitive plates made of epoxy resin Epidian 5 (Organika-Sarzyna SA, Poland) with a thickness 4 mm were prepared. After that they had holes with diameter of 3 mm made in them. Then Prime & Bond ONE (Dentsply Sirona, USA) bonding system was applied on the orifices and cured with The Cure TC-01 (Spring Health Products, USA) lamp for 10 seconds. So prepared holes were filled with one layer of the tested material. Such holes were prepared for each material. Polymerization was performed for 40 seconds with The Cure TC-01. The light-curing unit had irradiance output of 1250 mW/cm^2, which was confirmed with the use of a radiometric system (Digital 200 light meter by Rolence Enterprice Inc., Taoyuan, Taiwan). The samples were then stored in the absence of light at 36.6°C for 24 hours. After that time, the generated fringes in the plates were visualized in circular transmission polariscope FL200 (Gunt, Hamburg, Germany). Photoelastic images in parallel and perpendicular orientation of filter polarization planes were recorded by a digital camera (Canon EOS 600D, Canon Inc., Japan). Next, dimension of interference fringes was determined using the Met-Ilo program.

Stress and deformation analysis was carried based on the theory of elasticity formulas in a two-dimensional state stresses and three-dimensional state deformations. Next, the elastooptic constant of the plate was determined experimentally (k_σ = 1.45 MPa). Reduced shrinkage stress of analyzed material was calculated based on fringe's dimension and elastic constant, using the modified Timoshenko equation:

$$\sigma_r = \frac{a^2 \cdot p_s}{b^2 - a^2} \cdot \left(\frac{b^2}{r^2} - 1 \right) \tag{1}$$

$$\sigma_\theta = \frac{a^2 \cdot p_s}{b^2 - a^2} \cdot \left(\frac{b^2}{r^2} + 1 \right) \tag{2}$$

where:

σ_r – is radial stress,
σ_θ – is circumferential stress,
p_s – is the shrinkage stress around composite filling,
a – is the radius of the internal orifices in the plate,
b – is the radius of the largest of isochromatic fringes,
r – is the radius contained in the region from a to b.

the reduced shrinkage stress subtract was:

$$\sigma_{int} = \sigma_r - \sigma_\theta \tag{3}$$

According to the hypothesis of maximum tangential stresses τ_{max}:

$$\sigma_2 - \sigma_1 = \tau_{max} \tag{4}$$

it was assumed that $\sigma_2 - \sigma_1 = \sigma_r - \sigma_\theta$

$$\sigma_{int} = \tau_{max} \tag{5}$$

The greatest stress acting on the joint of the materials was the reduced shrinkage stress σ_{int}.

Based on the analysis of data obtained for each of the samples, mean values and standard deviation were calculated. A comparison of the means was carried out based on one-way analysis of variance and the Tukey's HSD test at the significance level α = 0.05. Homogenous groups of the means were distinguished, and the groups were indicated using subsequent letters (the means marked with the same letter do not differ significantly between each other). The analyses were performed using Statistica 13 software (StatSoft, Poland).

RESULTS AND DISCUSSION

Exemplary images of interference fringes recorded 24 hours after initiating the polymerization process with light are presented in Figures 1–4.

The mean values of radial stressed ranged from 2.96 MPa in the case of the Transbond XT up to 4.97 MPa in the case of the Resilience. The mean values of radial stress and related results of statistical tests are presented in Table 2.

In the case of the analysis of circumferential stress values, similar relationships were noted. Internal forces generated by the Resilience material samples were on average -6.57 MPa, while by the Transbond XT adhesive system – 4.28 MPa.

The mean values of circumferential stress and related results of statistical tests are presented in Table 3.

The significantly highest mean value of main/reduced stresses was recorded at the level of 11.54 MPa in the case of Resilience material, and the lowest for Transbond XT resin at the level of 7.24 MPa. In the case of Grēngloo and Ortho Connect adhesives, the mean values were 9.94 and 9.60 MPa, respectively, and did not differ significantly from each other. The above values, together with the corresponding statistical test results, are presented in Table 4.

Polymerization shrinkage and consequent shrinkage stresses appearing at the composite-tooth tissue interface still constitute one of the unresolved clinical problems in the use of dental materials based on dimethacrylate matrix. Internal stress results from organic matrix mole-

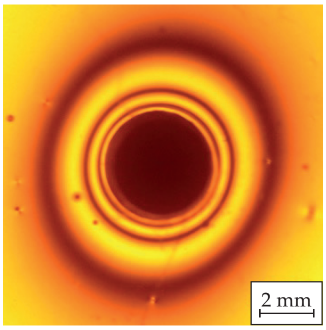

Fig. 1. Interference fringes for Ortho Connect material

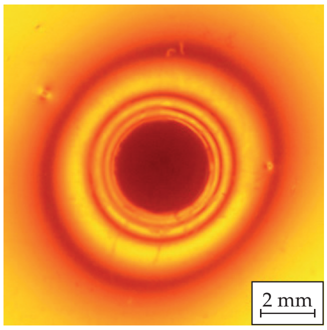

Fig. 2. Interference fringes for Grēngloo material

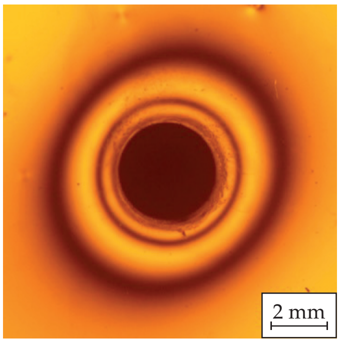

Fig. 3. Interference fringes for Transbond XT material

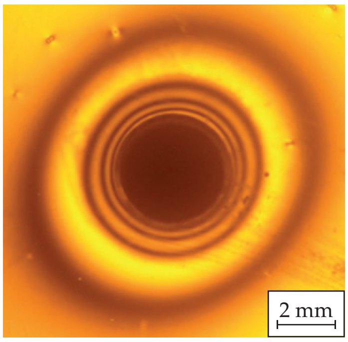

Fig. 4. Interference fringes for Resilience material

T a b l e 2. The results of statistical tests comparing the average values of radial stresses; $p = 0.05$

Material	Average σ_r MPa	Homogeneous group based on Tukey's test	Standard deviation	Ortho Connect	Grēngloo	Resilience	Transbond XT
Ortho Connect	4.01	b	0.54	–	0.785	<0.001	<0.001
Grēngloo	4.20	b	0.54	0.785	–	0.005	<0.001
Resilience	4.97	c	0.42	<0.001	0.005	–	<0.001
Transbond XT	2.96	a	0.36	<0.001	<0.001	<0.001	–

T a b l e 3. The results of statistical tests comparing the average values of circumferential stresses; $p = 0.05$

Material	Average σ_r MPa	Homogeneous group based on Tukey's test	Standard deviation	Ortho Connect	Grēngloo	Resilience	Transbond XT
Ortho Connect	-5.59	b	0.42	–	0.909	<0.001	<0.001
Grēngloo	-5.73	b	0.57	0.909	–	0.002	<0.001
Resilience	-6.57	a	0.41	<0.001	0.002	–	<0.001
Transbond XT	-4.28	c	0.48	<0.001	<0.001	<0.001	–

T a b l e 4. The results of statistical tests comparing the average values of main/reduced stresses; $p = 0.05$

Material	Average σ_{int} MPa	Homogeneous group based on Tukey's test	Standard deviation	Ortho Connect	Grēngloo	Resilience	Transbond XT
Ortho Connect	9.60	b	0.92	–	0.846	<0.001	<0.001
Grēngloo	9.94	b	1.11	0.846	–	0.002	<0.001
Resilience	11.54	c	0.82	<0.001	0.002	–	<0.001
Transbond XT	7.24	a	0.83	<0.001	<0.001	<0.001	–

cules coming closer to each other and from reduction of free spaces between them [13]. Weaker, primary van der Waals forces' interactions existing between monomers unbound with each other change into strong forces generated by C-C covalent bonds, formed during polymerization of the polymer network [14, 15].

In the case of restorative procedures, shrinkage stresses generated within the mass of the material during that time may endanger effective bonding of the material with the tooth if their levels exceed the values of opposing forces generated by dental adhesive systems. As mentioned above the configuration factor (C-factor) in the case of orthodontic brackets may be high, additionally affecting strength of the connection [15, 16]. For this reason, an analysis of generated stresses should also be considered as one of the factors in the context of using orthodontic adhesives based on methacrylate matrices.

In the authors' own study, the highest reduced stresses, which are equivalent to the stresses appearing on the orthodontic adhesive-enamel interface, are demonstrated by the Resilience Light-Cure Bracket Adhesive, the stresses of which amount to 11.5 MPa, and according to the research by Kuśmierczyk D. *et al*. [17] the degree of conversion of the said material is 68% 24 hours after curing. On the other hand, during the study, the lowest values of shrinkage stresses were recorded for the Transbond™ XT Light Cure Adhesive Kit material and they amounted to 7 MPa. The observed differences in shrinkage stresses of the above-mentioned materials can be related not only to their composition but also to the degree of conversion. A study by dos Santos R.L. *et al*. [18] showed that Transbond XT material was characterized by one of the lowest degrees of conversion (approx. 45%, regardless of the difference in curing time compared to *e.g*., Eagle Bond or Fill Mágic, for which DC% was respectively approx. 70% and 60%).

It is also known that there is a relationship between degree of conversion and polymerization shrinkage and shrinkage stress [20, 21].

In a study by Goncalves *et al*. [22], the authors confirmed the thesis that shrinkage stress of a composite material increases with an increase in material conversion and polymerization shrinkage. Confirmation of a low degree of conversion of the Transbond XT material can also be found in a study by Fernandes de Araujo *et al*. [23].

The above-mentioned materials have a matrix based on bis-GMA and TEGDMA, unfortunately there is no information on the weight ratio of these monomers and the degree of filling of the material. It is worth mentioning here that a change of the bis-GMA/TEGDMA ratio in a composite impact's polymerization shrinkage value and material viscosity [24]. In bis-GMA/TEGDMA mixtures, the conversion rate increases with increasing TEGDMA content.

Increased amount of TEGDMA in a mixture with Bowen monomer from 0 to 80 wt% causes an increase in the conversion rate from 53 to 80% [25].

A similar increase in degree of conversion (DC) from about 45% to 70% is observed after increasing the amount of TEGDMA from 0 to 40 wt% [26]. The positive effect of TEGDMA on the degree of conversion is related to the high concentration of terminal double bonds, low resin viscosity, as well as the size and mobility of the molecule [26]. It should also be noted that TEGDMA copolymer is characterized by over 2.5 times higher DC compared to bis-GMA [27].

It is also worth mentioning that the degree of conversion of double bonds (DC) of pure resins affects the mechanical and dynamic properties of cured material. There is a positive correlation between DC increase and hardness, compressive and bending strength, and bending modulus [28].

Similar observations apply to commercial composites for reconstruction of dentin core with different polymerization mechanisms [29]. In the above work it was found that with increased DC, microhardness, compressive and bending strength increases, and the amount of leached bis-GMA decreases. The degree of conversion also corresponds to cytotoxicity of orthodontic adhesives [18].

The other two orthodontic adhesives assessed in the study, Ortho Connect and Grēngloo™, have shrinkage stress of 9 MPa, statistically significantly different from the stresses observed for the two materials discussed earlier. Both adhesives contain in their matrix ethoxylated bisphenol A dimethacrylate (bis-EMA, molecular weight 540 g/mol) and urethane dimethacrylate (UDMA, molecular weight 470 g/mol). Compared to oligoethylene glycol dimethacrylates, e.g., TEGDMA, which, thanks to their favourable stereochemical structure, long chains, and flexibility, demonstrate a relatively high degree of conversion, UDMA and bis-EMA are characterized by a lower concentration of C = C double bonds, as well as a lower degree of conversion [30]. Therefore, in the case of the above-mentioned materials, reduced shrinkage stresses can be expected, which was confirmed by performed tests.

In a study by Choi A. *et al*. [31], the degree of conversion of Ortho Connect Flow was approximately 50%, which may also explain lower shrinkage stresses of this material observed in the present study, compared to internal forces confirmed for Resillence orthodontic adhesive system.

The results of our own study correspond to the "post-gel shrinkage" relationships described by Michael J. Rasmussen *et al*. [11] for Transbond XT Light Cure, Grēngloo™ and Ortho Connect 2-in-1 primer and adhesive. According to the cited authors, Transbond XT Light Cure (-0.38% vol.) demonstrated the lowest polymerization shrinkage (-0.38% vol.), followed by Grēngloo™ and Ortho Connect (-0.48% vol and -0.53% vol. respectively). The authors of the above-mentioned study associated significantly lower polymerization shrinkage of Transbond XT Light Cure with slower polymerization of the material and based the measurement method on a strain gauge and deformation curves of materials recorded during their polymerization.

The extent of shrinkage stress generated during polymerization of dental composites is also influenced by the amount of filler. The less filler, the higher the amount of organic phase, which is responsible for shrinkage and stresses. In available literature it is difficult to find data on filler content in the tested materials. We were able to find only a more precise composition of Transbon® XT Light-curing and Grēngloo™ adhesive. According to Vinagre A.R. *et al.* [32], Transbond® XT Light-curing contains 77 wt% in the form of Quartz, submicron silica. Similar information can be found in the publication of Berza *et al.* [33]. According to the quoted authors, Transbond XT Light Cure adhesive contains silane treated quartz and silane treated silica in an amount of 70–80 and < 2 wt%, respectively, while Grēngloo ™ adhesive contains aluminosilicate glass and silica glass (62–74 wt%, 0.5–74 wt%) 3% by weight. However, due to slight differences, it seems justified to omit these values in considerations on stresses recorded in the present study.

Although available literature contains a relatively large number of studies devoted to the issue of polymerization shrinkage of composite materials used for filling cavities, few studies are devoted to this matter in relation to orthodontic adhesives. The chemical structure and resulting physical properties of adhesive systems based on a polymer network are reflected in their functional properties determining clinical usefulness. Commonly used orthodontic adhesive systems, the polymerization of which is initiated by visible light, provide the dentist with control over positioning of brackets in the oral cavity conditions, and ensure sufficient bonding strength of brackets to the tooth surface, achieved immediately after completion of curing. Since resins are placed under bases of brackets, the influence of the oxygen inhibition phenomenon on cross-linking of monomers seems to be significantly reduced [34]. Unfortunately, we also know that the structure of these types of materials also determines their unfavourable properties. The phenomenon of polymerization shrinkage discussed in the article, together with accompanying stresses forming in the mass of materials, is only one of those properties. Another aspect that should be considered is incomplete cross-linking of the said materials, observed both immediately after initiating the polymerization process and sometime after its completion. This phenomenon makes composite materials used both for filling cavities and for fixing orthodontic brackets, susceptible to degradation [35, 36].

Orthodontic adhesive systems, which constitute a chemically heterogeneous group of materials, are used in the oral cavity environment, where they are exposed to variable temperature, pH, occlusal or orthodontic forces. Available literature includes studies devoted to the problem of potential biological impact on the human body of substances released in the process of degradation of composite materials [37–41]. Emission of bisphenol A [42] is particularly interesting - a monomer showing parahormonal activity, the potential harmful effect of which on organisms of mammals, especially in early stages of development, is confirmed in literature [43–45].

Although in clinical conditions internal stresses arising during polymerization of orthodontic adhesive systems do not seem to be as important as it is in the case of restorative materials, they should be considered when assessing performance of the mentioned orthodontic adhesives.

The limitations of the present study, resulting, among other things, from conducting it in laboratory conditions that do not take into account environmental conditions of the oral cavity and geometry of the tested materials different from that observed in clinical conditions, should be considered when interpreting its results, especially with regard to clinical application. There is no doubt, however, that a comparative analysis of internal stress of orthodontic adhesive systems may enhance the process of their assessment in the context of their performance properties. Although orthodontists do not have better adhesive systems than those based on light-cured composites, they should look critically at the materials used in everyday practice, considering not only their performance but also the safety aspect of their use.

CONCLUSIONS

Under the test conditions, the assessed orthodontic adhesive systems generated internal stresses during polymerization with visible light, which should be considered when assessing their clinical effectiveness.

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