# Chemical stability of orthodontic adhesives based on polymer network depending on external environment's temperature

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**Abstract**: In the present study the authors assessed chemical stability of four light-cured orthodontic adhesives: Contec LC, Transbond XT, Transbond Plus, Resilience, with respect to temperature of the external environment. Polymerized samples of orthodontic adhesives were treated with pH 7 phosphate-citrate buffer solutions based on HPLC-grade water at 20, 36 and 50 °C. After 1 hour, 24 hours and 7 days of sample incubation, the obtained eluates were analyzed using the high performance liquid chromatography method (HPLC) which confirmed the presence of triethylene glycol dimethacrylate (TEGDMA) monomer in solutions obtained after incubation of Contec LC, Resilience and Transbond XT samples. The presence of ethylene glycol dimethacrylate (EGDMA) monomer was also detected in eluates obtained from the Resilience adhesive. The eluates obtained after storage of Transbond Plus adhesive system were free of the sought substances. TEGDMA monomer concentrations were highest in the eluates obtained after 1 hour of incubation, the lowest after 7 days of storage of orthodontic adhesive samples, regardless of the temperature of the phosphate-citrate buffer. In addition, there were statistically significant differences in concentrations of monomers depending on the tested adhesive system. The rate of degradation of orthodontic adhesives based on a polymer network may also be adversely affected by an increase in ambient temperature.

**Keywords:** orthodontic adhesive systems, HPLC, chemical stability, monomers, temperature.

## Stabilność chemiczna klejów ortodontycznych opartych na sieci polimerowej w zależności od temperatury środowiska zewnętrznego

**Streszczenie:** Oceniano stabilność chemiczną czterech światłoutwardzalnych klejów ortodontycznych: Contec LC, Transbond XT, Transbond Plus oraz Resilience w warunkach zmiennych wartości temperatury środowiska zewnętrznego. Spolimeryzowane próbki klejów poddawano działaniu roztworów buforu fosforanowo-cytrynianowego na bazie wody o czystości HPLC o pH 7 i temperaturze 20, 36 i 50 °C. Po upływie 1 h, 24 h i 7 dni inkubacji próbek uzyskane eluaty analizowano metodą chromatografii cieczowej wysokociśnieniowej HPLC, która potwierdziła obecność monomeru dimetakrylanu glikolu trietylenowego (TEGDMA) w roztworach otrzymanych po inkubacji próbek materiałów Contec LC, Resilience i Transbond XT. W eluatach uzyskanych z kleju Resilience wykryto ponadto obecność monomeru dimetakrylanu glikolu etylenowego (EGDMA). Eluaty otrzymane po inkubacji systemu adhezyjnego Transbond Plus były wolne od poszukiwanych substancji. Największe stężenia monomeru TEGDMA były w eluatach uzyskanych po 1 h inkubacji, a najmniejsze po 7 dniach przechowywania próbek klejów ortodontycznych, niezależnie od temperatury buforu fosforanowo-cytrynianowego. Wykazano też istnienie istotnych statystycznie różnic stężeń oznaczonych monomerów w zależności od badanego systemu adhezyjnego. Zaobserwowano, że wzrost temperatury otoczenia może wywierać niekorzystny wpływ także na tempo degradacji klejów ortodontycznych opartych na matrycy polimerowej.

**Słowa kluczowe:** ortodontyczne systemy adhezyjne, HPLC, stabilność chemiczna, monomery, temperatura.

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The oral cavity, which constitutes the initial section of the digestive tract and the respiratory system, performs a number of important functions necessary for the proper functioning of the human body. Dental treatment, including orthodontic, provides, among other things, correct reconstruction of missing tissues of teeth and restoration of optimal occlusal conditions. For proper rehabilitation of the stomatognathic system, various materials are used that are permanently or temporarily introduced into the oral environment. They come into direct contact with tissues and are subject to the effects of masticatory forces, saliva, drinks, foods, or activity of microorganisms. Environmental conditions undoubtedly affect the dynamics and intensity of degradation of materials used in all fields of dentistry, including orthodontics [1–3], which may be associated with the risk of losing their physical properties that are important in the context of safety and efficiency of treatment [4]. Insufficient stability of dental materials' chemical structure and their susceptibility to degradation may contribute, apart from incomplete polymerization [5–7], to release of potentially harmful substances to the patient's body [8, 9]. Orthodontic adhesive systems, whose task is to fasten components of fixed appliances to tooth enamel, are based on composite materials. Monomers or oligomers, which are derivatives of methacrylic acid, form an organic matrix of orthodontic adhesives, and their composition is supplemented by inorganic fillers and a number of additional compounds with various functions, such as: polymerization initiators, catalysts, antioxidants, light stabilizers, plasticizers or dyes [8, 10–13].

Components of adhesives, products of their decomposition and manufacturing impurities of materials are not indifferent to living organisms, and their harmful actions are multi-faceted. Many studies confirm their cyto- and genotoxicity [2, 3, 14–16], negative impact on the reproductive system and fertility of animals [17], paraestrogenic action [18–20] and the ability to stimulate the growth of karyogenic bacteria [2]. Composite materials used in dentistry, including orthodontic adhesive systems, can irritate surrounding tissues and cause allergic reactions in treated patients [21].

One of physical variables that characterizes the oral environment and can affect the degree and rate of degradation of dental materials based on a polymer matrix is temperature. According to Volchansky *et al*. [22], temperature recorded in the oral cavity is not constant and varies depending on the site of measurement. In their study the authors used a digital thermometer and a thermocouple sensor, temperature on the surface of the mucosa was measured distally to the second molar and in the area of mandibular incisors on the labial side. Then the results were compared with values of temperature measured sublingually with closed and open mouth. It was confirmed that the temperature around anterior teeth of the mandible is statistically significantly lower than that measured in the retromolar and sublingual area. The temperature measured by Volchansky *et al*. [22] in the sublingual area with closed mouth equaled on average 36.3–36.9 °C.

Mean temperatures recorded by Choi *et al*. [23] on the palatal surface of superior incisors in 24-hour measurements equaled 33.99 °C. The study included 17 generally healthy volunteers, who had individual splints made containing a thermocouple that was worn by the subjects around the clock except during meals and baths.

Farella *et al*. [24] studied the oral cavity temperature of 11 healthy volunteers using wireless temperature sensors built into a vacuum-formed splint. The authors observed statistically significant differences between measurements obtained in the palatal area of upper incisors during daytime activity and during sleep. The probes were worn round-the-clock, except during meals requiring chewing and the time of hygienic procedures. Mean temperatures recorded during sleep were significantly higher than those recorded during the day.

In a study conducted by Barclay *et al*. [25], the authors used vacuum-formed splints for upper and lower dental arches, with built-in 28 thermistors in different parts of the arch, both on the vestibular and the palatal side. The adopted conditions of the experiment included drinking coffee at 77.5 °C and ice water at 1 °C. The authors observed that consumption of foods and beverages can be associated with occurrence of extreme temperatures in the range of 0–70 °C within anterior teeth.

Airoldi *et al*. [26] also assessed temperature changes in the oral cavity induced by consumption of hot and cold beverages. Six sensors for the lower arch and twelve for the upper arch were attached at various locations on Hawley retainer. Temperatures were recorded when drinking hot tea and cold water at 60 °C and 5 °C, respectively. Airoldi *et al*. observed temperature fluctuations within upper incisors in the range 7.1–57.4 °C.

Moore *et al*. [27], assessing daily temperature fluctuations in the oral cavity, observed that in the area of upper incisors the temperature is maintained at 33 to 37 °C for about 79 % of the measurement time, below 33 °C for 20 % of the time and above 37 °C for 1 % of the measurement duration.

Observations of the quoted authors indicate that temperature recorded in the oral cavity is maintained for the majority of time at a similar level, but it can periodically change in a relatively wide range.

The aim of the study was to assess chemical stability of four light-cured orthodontic adhesives with respect to temperature of the external environment.

## EXPERIMENTAL PART

### Materials

Four light-cured orthodontic adhesives: Contec LC (Dentaurum, Germany), Transbond XT (3M Unitek, USA), Transbond Plus (3M Unitek, USA), Resilience (Ortho Technology, USA) were tested.

**T a b l e 1. Composition of tested orthodontic systems declared by producers**

| Trade name | Basic ingredients | Filler content | Producer |
|---|---|---|---|
| Contec LC | 17–19 wt % of Bis-GMA<br>22–23 wt % of TEGDMA | Silicates | Dentaurum GmbH & Co. KG, Germany LOT: 90370 |
| Resilience – light-activated orthodontic adhesive system | Bis-GMA<br>TEGDMA<br>Camphorquinone | No data | Ortho Technology, Inc. Tampa, Florida USA LOT: H002658 |
| Transbond Plus – color change adhesive | 5–15 wt % of PEGDMA<br>5–15 wt % of 1,2,3-propanetricarboxylic acid 2-hydroxy-reaction products with 2-isocyanatoethyl methacrylate<br>2 wt % of Bis-GMA | 35–45 wt % of silane treated glass<br>35–45 wt % of silane treated quartz<br>< 2 wt % of silane treated silica | 3M Unitek Monrovia, Kalifornia USA LOT: N686102 |
| Transbond XT – light-cure adhesive paste | 10–20 wt % of Bis-GMA<br>5–10 wt % of bisphenol A bis(2-hydroxyethyl ether) dimethacrylate<br>< 0.2 wt % of diphenyliodonium hexafluorophosphate | 70–80 wt % of silane treated quartz<br>< 2 wt % of silane treated silica | 3M Unitek Monrovia, Kalifornia USA LOT: N619082 |

Orthodontic adhesive systems evaluated in the study and chemical composition declared by their producers are presented in Table 1.

## Sample preparation

The evaluated materials were placed in Teflon matrices with 5 mm diameter and 2 mm deep, and then polymerized for 20 seconds with LED 55 curing light (TPC Advanced Technology, USA) at 1200 mW/cm$^2$.

Adhesive resins, after removal from the matrices, were stored for 24 hours without light, and then placed in separate, aseptic tubes with a total volume of 15 cm$^3$. In order to avoid any influence of contamination with chemical compounds originating from the external environment, the tubes were rinsed three times with HPLC-grade water before use.

## Incubation of orthodontic adhesive systems in phosphate-citrate buffer solution

Samples of each of the assessed orthodontic adhesive systems were randomly divided into three groups of 5 samples each. The tubes in which the polymer network-based materials were placed were filled with 10 cm$^3$ of phosphate-citrate buffer solution based on HPLC-grade water (Sigma Aldrich, USA) at pH 7 and temperatures of 20, 36 and 50 °C, respectively, and then placed in an incubator shaker maintaining initial fluid temperatures.

After one-hour incubation of orthodontic adhesives in solutions, the obtained eluates were collected and the tubes with materials were filled again with 10 cm$^3$ of buffer solution with previously described parameters. The above procedure was repeated after 24 hours and 7 days of incubation. The control group in the study consisted of buffered solutions containing no samples of orthodontic adhesives. Eluates obtained in subsequent time intervals were frozen at -18 °C to minimize the probability of secondary polymerization reactions present in the solutions of chemical compounds.

## Methods of testing

### Chromatographic measurements

After the observation, the defrosted eluates were analyzed for the presence of camphorquinone (CQ), bisphenol A (BPA), triethylene glycol dimethacrylate (TEGDMA), urethane dimethacrylate (UDMA), 2,2-bis[4-(2-hydroxy-3-methacryloxypropoxy)phenylene]propane (Bis-GMA), ethylene glycol dimethacrylate (EGDMA) and 2,2-dimethoxy-2-phenylacetophenon (DMPA) using the ultra-high performance liquid chromatography method (UHPLC).

Chromatographic measurements were conducted with the use of NEXERA UHPLC system (Shimadzu Corporation, Japan) equipped with two LC-30AD pumps, SIL-30AC autosampler, SPD-M20A diode detector, CTO-20AC furnace and CBM-20A controller. During the analysis, Kinetex C18 columns and SecurityGuard ULTRA C18 2.1 mm ID precolumns (Phenomenex USA) were used. Phase A was HPLC-grade Chromasolv wa-

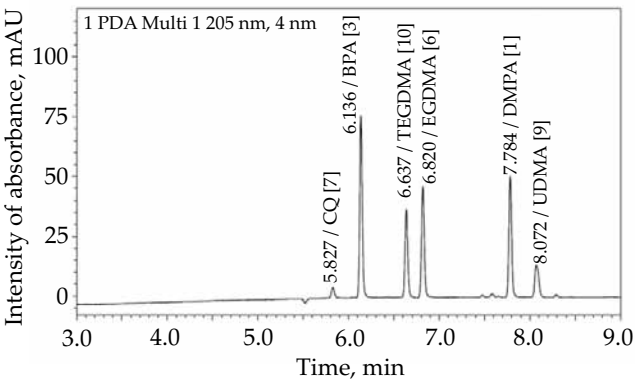


**Fig. 1. Retention times for sought substances**

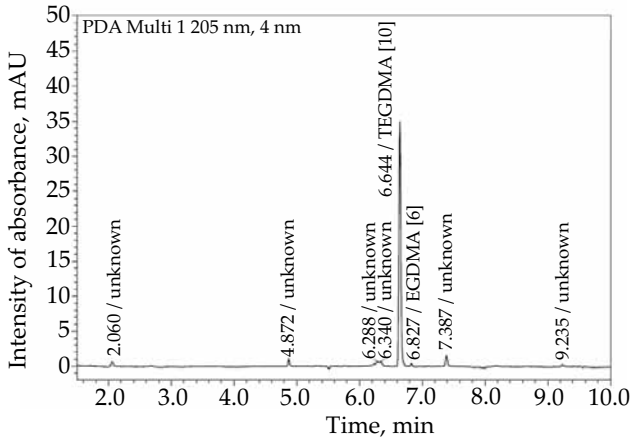


**Fig. 2. Exemplary HPLC chromatogram for Resilience adhesive (temp. 50 °C, pH 7, 1 h)**

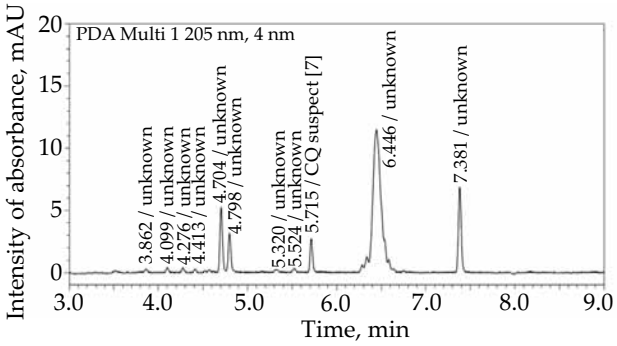


**Fig. 3. Exemplary HPLC chromatogram for Transbond Plus adhesive (temp. 50 °C, pH 7, 1 h)**

ter (Sigma-Aldrich, USA) and phase B was HPLC-grade Chromasolv acetonitrile (Sigma-Aldrich, USA). Analysis time of a single sample was 16 minutes and the phase flow rate was 0.3 $cm^3$/min. The quantitative analysis was made at the wavelength of 205 nm.

For calibration, CQ, BPA, TEGDMA, UDMA, Bis-GMA, EGDMA, DMPA reference standards from Sigma-Aldrich (USA) were used (Fig. 1).

#### Statistical analysis

Statistical analyzes were performed using Statistica 13 program (StatSoft, Poland). Comparisons of averages were conducted using the analysis of variance and multiple comparisons by the Fisher procedure (LSD). In order to determine the effect of temperature on substance concentrations, a simple linear regression analysis was performed and Pearson's correlation coefficients were calculated. In all analyzes, the significance level was assumed at $p = 0.05$.

## RESULTS AND DISCUSSION

### Results

TEGDMA presence was confirmed in solutions obtained after incubation of samples of Contec LC, Resilience and Transbond XT materials. EGDMA was detected in eluates from Resilience adhesive (Fig. 2).

The eluates obtained from Transbond Plus adhesive system were free of the sought substances. Some of the chromatographic analyzes performed for Transbond Plus had peaks similar to the CQ reference standard, but their position did not clearly confirm the compound's presence. In addition, the chromatograms obtained after an analysis of the eluates of all evaluated materials, demonstrated numerous peaks which did not correspond to the chemicals sought in the present study (Fig. 3).

TEGDMA concentrations for individual orthodontic adhesives were the highest in the eluates obtained after 1 hour of incubation, and the lowest after 7 days of sample storage, regardless of the temperature of the phosphate-citrate buffer. The highest concentrations of the monomer were identified in solutions obtained after incubation of Contec LC samples, and the lowest in solutions of Transbond XT. Also it should be noted that in the case of Transbond XT adhesive samples at 20 °C and after 7 days of material's incubation, regardless of the temperature of the environment, no TEGDMA monomer was found at the assumed detection level. Differences between mean TEGDMA concentrations determined in the eluates of the tested adhesive systems in particular temperature ranges and observation times are statistically significant ($p < 0.05$).

Table 2 compares mean concentrations of TEGDMA in solutions obtained from individual orthodontic adhesives in subsequent observation periods for each of the assumed temperature values.

In the case of Contec LC adhesive system, the highest concentrations of TEGMA were observed in eluates obtained after 1 hour of incubation. At 36 °C it averaged 8.578 μg/$cm^3$, at 50 °C it was 6.687 μg/$cm^3$, and at 20 °C the mean value was 4.551 μg/$cm^3$. Analysis of the correlation coefficient did not show that the effect of temperature on the increase in concentration of TEGDMA released from Contec LC material in the initial observation period was statistically significant. However, a significant relationship was confirmed between the temperature increase and the amount of TEGDMA released from Contec LC adhesive system in subsequent observation periods, *i.e.*, after 24 hours and 7 days of sample incubation.

In the case of Resilience adhesive, after 1 hour and 24 hours of sample incubation a significant positive correlation was observed between an increase in TEGDMA concentrations in solutions and an increase in their temperature. This dependence was not observed in the case of assays performed on eluates obtained after 7 days of material's incubation.

For Transbond XT adhesive system, the highest mean TEGDMA concentration of 0.049 μg/$cm^3$ was observed after 1 hour of material storage at 36 °C. At 50 °C after 1 hour and 24 hours of incubation, mean TEGDMA concentrations were recorded at 0.012 and 0.009 μg/$cm^3$, respectively. In the remaining temperature ranges, no

**T a b l e 2. Mean concentrations of TEGDMA detected in eluates of the tested orthodontic adhesives after 1 h, 24 h and 7 days of elution in a solvent at 20, 36 and 50 °C; pH = 7**

| Material | 1 h | | | 24 h | | | 7 days | | |
|---|---|---|---|---|---|---|---|---|---|
| | Mean concentration μg/cm³ | SD | Range μg/cm³ | Mean concentration μg/cm³ | SD | Range μg/cm³ | Mean concentration μg/cm³ | SD | Range μg/cm³ |
| Temperature 20 °C | | | | | | | | | |
| Contec LC | 4.551 c | 0.691 | 3.403–5.080 | 1.588 c | 0.342 | 1.250–2.157 | 1.346 c | 0.053 | 1.286–1.428 |
| Resilience | 2.337 b | 0.223 | 2.067–2.602 | 0.417 b | 0.082 | 0.354–0.554 | 0.299 b | 0.084 | 0.228–0.444 |
| Transbond XT | 0.000 a | 0.000 | | 0.000 a | 0.000 | | 0.000 a | 0.000 | |
| *p* (based on the analysis of variance) | < 0.001* | | | < 0.001* | | | < 0.001* | | |
| Temperature 36 °C | | | | | | | | | |
| Contec LC | 8.578 c | 1.761 | 6.42–10.61 | 2.233 c | 0.403 | 1.65–2.78 | 1.982 c | 0.324 | 1.65–2.38 |
| Resilience | 2.640 b | 0.377 | 2.23–3.11 | 0.513 b | 0.198 | 0.39–0.87 | 0.342 b | 0.145 | 0.25–0.60 |
| Transbond XT | 0.049 a | 0.017 | 0.02–0.07 | 0.000 a | 0.000 | | 0.000 a | 0.000 | |
| *p* (based on the analysis of variance) | < 0.001* | | | < 0.001* | | | < 0.001* | | |
| Temperature 50 °C | | | | | | | | | |
| Contec LC | 6.687 c | 0.941 | 5.100–7.433 | 3.806 c | 0.485 | 3.432–4.657 | 2.476 c | 0.210 | 2.221–2.785 |
| Resilience | 3.020 b | 0.226 | 2.803–3.379 | 0.590 b | 0.045 | 0.539–0.656 | 0.238 b | 0.006 | 0.230–0.244 |
| Transbond XT | 0.012 a | 0.001 | 0.011–0.014 | 0.009 a | 0.006 | 0.000–0.016 | 0.000 a | 0.000 | |
| *p* (based on the analysis of variance) | < 0.001* | | | < 0.001* | | | < 0.001* | | |

* – Statistically significant differences are present (as $p < 0.05$); SD – standard deviation; a–c – homogeneous groups.

TEGDMA was detected irrespective of the time the adhesive was stored in the solution.

Table 3 presents a comparison of mean TEGDMA concentrations in eluates of individual adhesives in subsequent time intervals depending on the value of ambient temperature.

Analysis of EGDMA concentrations determined after 1 hour and 7 days of incubation of Resilience samples in phosphate-citrate buffer showed a significant positive correlation between an increase in released monomer concentrations and an increase in external environment's temperature. In the case of Resilience solutions obtained after 24 hours of storage, this relationship was not statistically significant.

Mean EGDMA concentrations of monomer released from samples of Resilience orthodontic adhesive system in subsequent observation periods and temperature ranges are shown in Table 4.

## Discussion

The aim of the conducted study was to assess the influence of temperature on the chemical stability of four polymer-based orthodontic adhesive systems. In most publications regarding release of components from orthodontic adhesives, sample incubation is carried out in solutions at a constant temperature, typically around 37 °C [28–30].

**T a b l e 3. Distribution of mean TEGDMA concentrations (μg/cm³) in eluates obtained from Contec LC, Resilience and Transbond XT for three periods of observation depending on the changing temperatures of the solutions**

| Leaching time | Mean concentrations, μg/cm³ | | | Correlation coefficient (*r*) | Regression coefficient (*b*) | Probability value (*p*) |
|---|---|---|---|---|---|---|
| | 20 °C | 36 °C | 50 °C | | | |
| Contec LC | | | | | | |
| 1 h | 4.551 | 8.578 | 6.687 | 0.469 | 0.075 | 0.078 |
| 24 h | 1.588 | 2.233 | 3.806 | 0.894 | 0.073 | < 0.001* |
| 7 days | 1.346 | 1.982 | 2.476 | 0.917 | 0.038 | < 0.001* |
| Resilience | | | | | | |
| 1 h | 2.337 | 2.640 | 3.020 | 0.735 | 0.023 | 0.002* |
| 24 h | 0.417 | 0.513 | 0.590 | 0.530 | 0.006 | 0.042* |
| 7 days | 0.299 | 0.342 | 0.238 | -0.244 | -0.002 | 0.380 |
| Transbond XT | | | | | | |
| 1 h | 0.000 | 0.049 | 0.012 | 0.255 | 0.000 | 0.359 |
| 24 h | 0.000 | 0.000 | 0.009 | 0.705 | 0.000 | 0.003* |
| 7 days | 0.000 | 0.000 | 0.000 | – | – | – |

* – Statistically significant differences are present (as $p < 0.05$).

**T a b l e 4. Mean concentrations of EGDMA (μg/cm³) leached from Resilience adhesive in aqueous solutions at pH 7 and various temperatures values after 1 h, 24 h and 7 days of observation**

| Leaching time | Mean concentrations, μg/cm³ | | | Correlation coefficient (*r*) | Regression coefficient (*b*) | Probability value (*p*) |
|---|---|---|---|---|---|---|
| | 20 °C | 36 °C | 50 °C | | | |
| 1 h | 0.010 | 0.018 | 0.023 | 0.869 | 0.0004 | < 0.001* |
| 24 h | 0.004 | 0.000 | 0.007 | 0.340 | 0.0001 | 0.215 |
| 7 days | 0.005 | 0.005 | 0.007 | 0.676 | 0.0001 | 0.006* |

* – Statistically significant differences are present (as $p < 0.05$).

Kotyk *et al*. [31] investigating leaching of BPA from orthodontic materials, including Transbond XT, set incubation temperature at 37 °C. Before placing samples in solutions to perform assays, the authors subjected the tested materials to 10 shaking cycles at 60 °C and 4 °C, for 5 minutes each. The aim of this activity was to simulate mechanical and thermal conditions to which orthodontic appliances and adhesive systems are exposed in the oral cavity. Kotyk *et al*. analyzed the eluates obtained by gas chromatography and mass spectrometry (GC/MS). In the case of solutions obtained from incubation of Transbond XT, the authors obtained detectable amounts of BPA only after 3 days of observation at an average level of 2.75 μg/g. In that study, an assessment of possible influence of thermal aging of composite materials on the dynamics of BPA release is difficult, because the authors did not determine its concentrations when incubating samples not subjected to extreme temperatures. Direct comparison of BPA concentrations described by Kotyk *et al*. with results of other authors' studies is not possible due to differences in analytical methods used, preparation and selection of samples, type and volume of the leaching solutions, or in the way of result presentation.

Studies from the available literature where thermocycling was used as a method of aging composite materials focus primarily on the influence of the temperature variable on physical properties of composite adhesive systems.

Bishara *et al*. [32] subjected samples of two orthodontic adhesives to thermal cycles in the range of 2 ± 2 °C to 50 ± 2 °C with 3000, 6000, and 12 000 repetitions. The authors assumed them as equivalent to 15, 30 and 60 days of storage of materials in an environment of 100 % humidity and temperature of 37 °C, which would correspond to the conditions prevailing in the oral cavity. Bishara *et al*. confirmed weakening of resistance to shearing forces of both tested adhesives subjected to thermocycling.

Pereira *et al*. [33] assessed the size of microleakage for 2 composite filling materials in Class V cavities. They did not find a statistically significant effect of thermocycling (5000 cycles of 5 seconds at 5 °C and 55 °C) on the size of microleakage.

Tuncer *et al*. [34] subjected samples of Filtek Z250 composite material to coffee at 37 and 70 °C and to cola drink at 10 and 37 °C. The study by the quoted authors showed that beverages at higher temperatures caused a stronger color change, but did not significantly affect the hardness and roughness of the material.

Temperatures measured on the surface of the teeth, excluding the periods of food and drink consumption, show mean values which are lower than usually assumed as the oral cavity temperature, generally supposed to equal about 37 °C. This phenomenon is caused by such factors as:

– air flow during breathing and speech,

– ambient temperature,
– degree of lip closing,
– breathing track [23, 24, 27],
– and individual characteristics that affect body temperature, such as daily hormone fluctuations, health status, age, medications, *etc*.

Considering the above, in this study the adopted initial temperature value was 36 °C. The temperature range and time of oral exposure to extreme fluctuations in temperature values are individual for each patient. It largely depends on nutritional habits of individuals, their tolerance to the warmth of food and drinks, the method of food and fluids consumption: for example, the size of mouthfuls, time of keeping in the mouth, drinking from a cup or with a straw [25, 26], and it is difficult to reproduce in laboratory conditions. In the current experiment, additional temperatures of 20 and 50 °C were adopted, similarly to those suggested by Michailesco *et al*. [35] for thermocycling tests, in order to observe separately the influence of low and high temperatures associated with possible eating habits of patients.

The results of the presented study indicate that the adopted temperatures do not affect the type of substances released from the examined adhesive systems in the range of compounds sought in the experiment. As far as the seven sought compounds are concerned, only TEGDMA and EGDMA monomers were identified in the eluates. Also, it should be noted that during the analysis of chromatograms, numerous peaks indicated that other chemical compounds were also released into the external environment, not only those assumed as indicators. This observation indirectly confirms the chemical instability of orthodontic adhesive resins and suggests further research to identify components released from dental materials. Hope *et al*. [36] suggest the selection of mass spectrometry as the detection method that increases sensitivity and specificity of identification of eluted substances.

Comparison of mean TEGDMA concentrations observed in solutions obtained from incubation of individual adhesive systems confirms the thesis that their level depends on the type of adhesive system. Most probably this is due to the differences in composition and chemical structure of individual polymer-based orthodontic adhesives. Differences in the degree of conversion [37] of materials evaluated in the current study may also affect their durability and dynamics of components' release to the external environment. The adopted study method does not allow to determine explicitly whether and to what extent the elution of components from polymerized samples of orthodontic adhesives is caused by the presence of free monomers in the material or it results from subsequent degradation process of adhesive systems. It seems that both components can coexist together, and their mutual proportions may change with time. The high levels of mean concentrations of monomers released after one hour of observation are probably influenced by incomplete polymerization of the tested material. In subsequent observation periods researchers should pay more attention to the release of monomers from disintegrating polymer network. Release of TEGDMA monomer into solutions is confirmed by observations of other authors [28, 29, 38, 39]. Low molecular weight of the mentioned monomer makes its transfer into the external environment easier than in the case of other compounds of higher mass and more complex structure [8, 29]. Due to its widespread use in synthesis of dental materials based on a polymer network, it can be considered as a monomer that enables comparison of structural stability of various composites.

**Summary**

The analysis of the impact of environment temperature increase on chemical stability of the evaluated orthodontic adhesive systems, which was measured by concentrations of TEGDMA and EGDMA monomers in eluates, confirmed the existence of a significantly positive correlation between the above variables. The results of the observation allow to formulate the theory that patients preferring hot foods and beverages may be exposed to increased release of components from orthodontic adhesives into the oral environment, and to resulting consequences. Unfortunately, available literature does not offer any studies whose authors assessed the relationship described in the present study, hence it is impossible to directly refer the obtained results to other research.

## CONCLUSIONS

– Under the conditions of the study, orthodontic adhesive systems are not chemically stable.
– An increase in ambient temperature may have an adverse effect on chemical stability of orthodontic adhesives based on a polymer network.

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