

Doctoral (Ph.D.) Dissertation

**Material science and biocompatibility
examinations of pre-heated resin-based
composites**

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List of abbreviations

ACN = acetonitrile

AFM = addition-fragmentation monomer

ANOVA = analysis of variance

AUDMA = aromatic urethane dimethacrylate

BHT = butylated hydroxytoluene

BisEMA = ethoxylated bisphenol-A dimethacrylate

BisGMA = bisphenol A-diglycidil ether methacrylate

C = ceramic

CI = Confidence Interval

CQ = camphorquinone

D = dentin

DC = degree of conversion

DOC = depth of cure

DDMA = 1,12-dodecane dimethacrylate

DEJ = dentin-enamel junction

df = degree of freedom

DSC = Differential scanning calorimetry

LC = light-cured

LCU = light curing unit

LED = Light Emitting Diode

PPD = 1-phenyl-1,2-propanedione

PTFE = polytetrafluoroethylene

RBC = resin-based composite

RP-HPLC = Reversed-phase High-Performance Liquid Chromatography

S.D. = standard deviation

SPSS = Statistical Package for Social Science

TEGDMA = triethylene glycol dimethacrylate

UDMA = urethane dimethacrylate

UV = ultraviolet

VIS = visible

vol% = volumetric %

wt% = weight %

I. Introduction

Dental resin-based composites (RBC) were introduced in the early 60s. These materials are an aesthetic alternative to amalgam as their mechanical properties have become suitable for treating the posterior region as a result of numerous developments. Thanks to their outstanding optical properties, they can be an alternative to glass ceramics in the anterior region. Today, RBCs have become extremely widespread, and countless publications deal with their material science and biocompatibility. Searching the term *resin composite* on PubMed database, 40985 results can be found; searching only in the title of articles, we get 1569 results.

Although the RBC can be used as a long-term restoration, there are some undesirable characteristics despite the countless developments. The disadvantageous properties of the RBCs are closely related, among other things, to insufficient polymerization and the amount of unreacted monomers. While there have been concerns over the use of amalgam restorations with regard to mercury release and the amalgam waste management, there are also research results that prove the cytotoxic, genotoxic, mutagenic and estrogenic effects of the released monomers from RBC materials. In addition to the monomers, thermal effect during polymerization should also be taken into account as a potentially harmful *noxa*.

Numerous studies can be found in relation to traditional RBC materials, but the literature is incomplete regarding some properties of newer type of RBCs and application methods. Therefore, bulk-fills and pre-heating became the focus of my researches, providing the basis of this dissertation.

On one hand, the purpose was to examine the temperature changes during the application and polymerization, furthermore to detection the unreacted monomers from pre-heated bulk-fill RBCs, and to evaluate the effect of pre-heating on the degree of conversion (DC). On the other hand, the comparison of intrapulpal thermal changes resulting from cementation of ceramic inlay with different adhesive resin cements and pre-heated sculptable submicron RBCs was the focus of the interest. Assessing the influence of dentin and ceramic layer thicknesses on pulpal temperature rise, supplemented by a qualitative comparison of the thermal properties between dentin and ceramics was also a scope of the present study.

I.1. Resin-based dental composites

Dental resin-based composites are the most extensively employed restorative materials due to their manifold advantages, including excellent esthetics and mechanical properties (Figure 1).



Figure 1. *Upper left second premolar tooth was prepared to remove caries and restored anatomically and esthetically with resin-based composite
[photo made by Edina Lempel]*

I.1.1. The chemical composition of resin-based composites

RBC materials from a chemical point of view are consist of four major components: organic polymer matrix, inorganic filler particles, coupling agent, and the initiator-accelerator system [36].

The organic matrix is built from a system of mono-, di- or tri-functional monomers [73]. The most popular monomers have become dimethacrylates such as 2,2-bis[4-[2-hydroxy-3-(methacryloyloxy)propyl]phenyl] propane (Bis-GMA) or 1,6-bis[2-(methacryloyloxy)ethoxycarbonylamino]-2,4,4-trimethyl-hexane (UDMA) [85]. These monomers result in low methacrylate conversion, and their high viscosity limits the amount of fillers that can be added to the system. The diluent molecule most often applied to reduce viscosity is triethylene glycol dimethacrylate (TEGDMA), which can be used to increase the degree of conversion (DC) and the amount of fillers, but at the same time results in a higher shrinkage upon polymerization [37]. The disperse phase of RBCs consist of an inorganic filler material which determines the physical and mechanical

properties of the RBC. Incorporating as high proportion as possible of fillers is a fundamental goal to improve the physical properties of the organic matrix. In addition to the amount of fillers, their size and shape are also important determinants. Common fillers are quartz, silicon dioxide, boron silicates, lithium aluminium silicates. Barium, strontium, zinc, aluminium or zirconium are used as radio-opaque filling materials [73,137].

Bifunctional polar molecules, mainly in the form of organosilanes, are used to connect the organic matrix to the inorganic fillers. At one end of this bifunctional molecule, the vinyl group has methacrylate characteristics, and at the other end there is an alkoxy group that can interact and attach to glass or quartz surfaces. Thus, the resin is able to form a bond between the filler components with the help of the silane molecules in an RBC system [105,110].

The crosslinking reaction is usually based on a visible light initiated photopolymerization. For this purpose, the most often used system is composed of camphorquinone (CQ) and an amide. This system initiates the free-radical polymerization of the matrix monomers upon irradiation with light at a wavelength of 468 nm. An alternative to CQ is 1-phenyl-1,2-propanedione (PPD) which does not have a yellow coloring effect unlike the previous one, but is less effective in terms of initiating polymerization [37,85].

Inhibitors are also added to the RBC to prevent spontaneous or accidental monomer polymerization. These inhibitors react with free radicals faster than free radicals react with monomers, thus preventing chain propagation before the free radicals are able to initiate the polymerization. Butylated hydroxytoluene (BHT) is a commonly used inhibitor. The functions of these inhibitors are to increase the storage life of RBCs and to ensure sufficient manipulation time [14].

Figure 2 shows the structural formulas of the above-mentioned molecules (Figure 2).

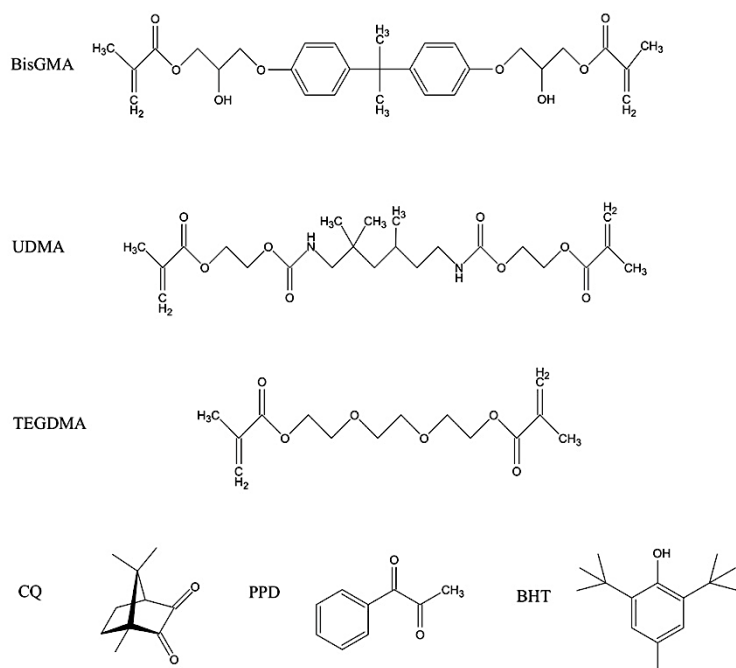


Figure 2. The structural formulas of bisphenol A-diglycidyl ether methacrylate (BisGMA), urethane dimethacrylate (UDMA), triethylene glycol dimethacrylate (TEGDMA), camphorquinone (CQ), 1-phenyl-1,2-propanedione (PPD), and butylated hydroxytoluene (BHT) molecules [own editing]

I.1.2. The polymerization of resin-based composites

The polymerization of methacrylate-based composites is a free-radical addition polymerization of the corresponding methacrylate monomers. During the clinical use of RBCs, visible-light-activated systems are used [14].

The polymerization reaction includes three steps: initiation, propagation, and termination. Free radicals, which are responsible for initiating the process, are generated during the initiation phase. These free radicals contain chemical groups with unpaired electrons, which are highly reactive, so can react with the carbon-carbon double bonds of the monomers. As the free radical concentration increases, the polymerization rate also increases. This phenomenon called as autoacceleration [36].

The activated monomers are triggering the chain-reaction by being added onto the double bonds of additional monomers. This second stage of polymerization is called as propagation, which resulting a growing polymer chain. The propagation reaction

becomes diffusion-controlled only if significantly higher conversion rate occurs. The process is limited, continues until the RBC becomes glassy. This process referred to as vitrification. As the polymer vitrifies, the propagation reaction slows down and the polymerization ceases, that is, autodeceleration occurs, results in unreacted methacrylates that remain in the RBC restoration.

Branching and cross-linking occur in the termination phase. This phase is diffusion-controlled and slowed by the network, even at low conversion. As the formation of polymer network progresses, the mobility continues to decrease, and the system becomes more viscous until the reaction stops. The process starts when the glass transition temperature reaches the polymerization temperature [14,36,37].

The polymerization of RBCs is an exothermic reaction. The released heat during the propagation increases the activity of the growing chains as well as the free radicals, results in further densification and vitrification of the structure [112].

Some disadvantage of RBC materials may compromise the longevity of RBC restorations such as polymerization stress, mismatch in thermal expansion, marginal leakage, toxicity, to mention just a few of them. To overcome these problems, attempts have been made to improve the mechanical properties, as well as to ameliorate the clinical procedure [138].

I.1.3. Pre-heating of resin-based composites

During restorative procedure it is favorable to use an easy-to-handle, non-technique sensitive, durable and esthetic restorative material with quick and efficient polymerization. In case of RBC restorations, expediting polymerization, increasing of maximum layer thickness and the degree of monomer conversion can be considered as the main objectives [61]. In order to achieve a durable, successful RBC restoration, the most important factors, among others, include good mechanical properties and handling characteristics, reduced polymerization stress, good marginal adaptation and high DC [102].

According to the literature data, there is a clear correlation between the DC and the physico-chemical characteristics of RBCs [67,94]. Meanwhile, DC is influenced by several factors, such as light exposure conditions, composition, shade, opacity and thickness of the RBC layer, pre-cure temperature also plays an important role in the polymerization process [6,40]. As the success of RBC restorations depends on their

polymerization and DC, the influence of temperature has become one of the central issues of several studies [27,160].

I.1.3.1. The polymerization of pre-heated resin-based composites

The polymerization of RBCs is an exothermic reaction and the released heat is mainly produced in the propagation phase [112]. The process of monomer conversion and the properties of the set polymer are influenced by the polymerization temperature [39]. An elevation in temperature promotes molecular mobility and increases collision frequency of reactive radicals, resulting in a higher conversion and delayed autodeceleration [40, 112]. Not only the exothermic reaction, but also the absorbed light energy contributes to the system temperature during the polymerization of light-cured RBCs [131]. While some authors attribute a greater significance to the heat emitted by the light curing unit in the temperature rise, others regard the heat generated in the exothermic reaction as more important [95,109].

I.1.3.2. Advantages and disadvantages of pre-heating

As reported by several investigations, pre-heating may have a beneficial impact on marginal adaptation, gap formation and microleakage by reducing the viscosity of RBCs [62,154,160]. Improved handling properties, such as flowability can facilitate the application of the filling material, consequently making the procedure less time-consuming. Reduced viscosity also improves marginal seal and surface microhardness, thereby contributing further to the overall clinical success [51,103,104]. There may be a lack of efficiency associated with the use of conventional heating devices as some authors have shown a rapid decrease in RBC temperature after removal from the device, as well as during dispensing and handling [42,102]. Moreover, during the cooling phase, the system bears a loss of energy, so vitrification takes place earlier and causes decreased DC [96].

I.1.3.3. Pre-heating methods

There are numerous methods for pre-heating dental RBCs mentioned by the literature. In addition to various devices specially developed for this purpose, several alternative methods are known, such as hand holding, hot water bath, the light of dental unit, use of microwave oven, hair dryer, wax warmer, and so on [16].

The most widely reported device to pre-heat dental RBCs is the Calset Composite Warmer (AdDent, Inc., Dandury, CT, USA) (Figure 3), which can warm up the material to 37 °C, 54 °C and 68 °C, and maintains a constant temperature. However, the efficacy of this device was reported to be lower than promised [16,75].



Figure 3. *Calset Composite Warmer (AdDent, Inc., Dandury, CT, USA) [78]*

Another similar device is Thermo-Flo™ Composite Warming Kit (Vista dental, USA) (Figure 4). The unit is designed to heat the RBC material to 68 °C [16,77].



Figure 4. *Therma-Flo™ Composite Warming Kit (Vista dental, USA) [76]*

Ena Heat Composite Heating Conditioner (Micerium, Avengo, Italy) (Figure 5) – used during our experiments – has two selectable temperatures: 39°C (T1 setting) and 55°C (T2 setting). The first temperature is recommended to heat RBC for build-up and to heat anesthetics and sodium hypochlorite; the second temperature setting is advised to heat RBCs for luting [74].



Figure 5. *Ena Heat Composite Heating Conditioner (Micerium, Avengo, Italy) [74]*

In case of these methods, however, the current temperature of the material is lower than the preset temperature. An additional disadvantage is the rapid temperature drop of the RBC after removal from the warming device and during the transfer to the prepared cavity [42].

I.1.3.4. New developments and findings in pre-heating of resin-based composites

Several types of RBC dental materials have been developed over the years, including bulk-fills, which can be placed in larger increments to reduce chairside time and technique sensitivity [31]. The primary advantage of bulk-fill RBCs over conventional ones is the increased depth of cure [5,99]. According to a literature review by Van Ende et al., the maximum layer thickness which still ensures adequate material characteristics as recommended by the manufacturers, is 4 mm or in some cases even 5 mm. Although most studies have confirmed the improved depth of cure for bulk-fill RBCs, some controversial data can still be found [151,93]. Besides the DC, polymerization shrinkage stress is another important issue addressing the clinical failures. Ausiello et al. demonstrated, that bulk filling, especially in deep cavities induces higher shrinkage stress along the cavity walls compared to a multilayer technique (i.e. no shrinking glass-ionomer basing and shrinking bulk-fill RBC cover) [18].

Recently, a thermoviscous bulk-fill RBC (VisCalor Bulk) and a new heating device (VisCalor Dispenser) was introduced with the aim of combining the advantages of bulk-filling and pre-heating (Figure 6).



Figure 6. *VisCalor Dispenser (VOCO, Cuxhaven, Germany) [80]*

This delivery system can warm up the filling material in seconds using near infrared technology and allows immediate application without removal of the capsule from the heating device thereby maintaining its increased temperature [79]. Although, VisCalor Bulk is a relatively new RBC, it has already been the subject of several investigations [34,50,108,158,159]. Yang et al. examined the effects of temperature on stickiness and

packability, and the effect of pre-heating time on pre-cure properties. The study found pre-heating to lead to a reduced extrusion force and increased flowability without premature polymerization, while stickiness and packability remained within a clinically acceptable range [158].

The effect of pre-heating and exposure duration on other properties of VisCalor Bulk have been investigated in another study undertaken by Yang et al. This article reported a longer exposure duration not to have an effect on the DC, maximum rate of polymerization and polymerization shrinkage, however it did lead to an increase top surface microhardness. The application of 3 min pre-heating and 20 s irradiation provided adequate hardness without unfavorable changes in polymerization shrinkage strain and polymerization kinetics [159]. The study of Marcondes et al. examined viscosity and thermal kinetics of pre-heated RBCs including VisCalor Bulk as well as the effect of ultrasound energy on film thickness. VisCalor Bulk showed the greatest extent of viscosity reduction at 69 °C, while film thickness could not be reduced below 50 µm without the use of ultrasound. This study also claimed that in order to take full advantage of the pre-heated RBCs, the ideal working time is merely 10-15 s [108]. Demirel et al. investigated the effect of different insertion techniques on internal void formation and found VisCalor Bulk to show the lowest void percentage with the utilization of the pre-heating technique [50]. Colombo et al. evaluated microhardness and depth of cure of four bulk-fill RBCs. According to the measured ratio of top to bottom hardness, all tested materials - including VisCalor Bulk - showed an adequate degree of polymerization. In addition, in case of VisCalor Bulk, acid storage led to one of the highest mean percentage loss in microhardness of the external side [34].

Besides the physico-mechanical properties, the chemical characteristics are also important determinants of the clinical performance and biocompatibility of an RBC [64]. Although, there is a strong inverse correlation between DC and monomer elution, the amount of released monomers may be influenced further by other factors such as the quality of the monomer system, filler type, content, porosity as well as employed solvent [93,144]. Elution from bulk-fill RBCs was found to be comparable to that of conventional materials despite their increased increment thickness, since the monomer release is more dependent on the hydrophobicity of the base monomers and the final network characteristics of the resin-matrix [8]. Detection of unreacted monomers from pre-heated RBCs is not a well investigated topic so far.

I.1.3.5. Further application of pre-heated resin-based composites and its considerations

Besides the advocacy of pre-heating in direct restorative procedures, pre-warmed RBCs are also introduced in cementation of indirect restorations. Posterior indirect partial restorations (inlay, onlay, overlay) are widely used in dental clinical practice to overcome issues resulting from the use of direct RBCs [47,89,114]. Adhesive cementation of ceramic inlays is recommended to improve the esthetic and mechanical properties of the restoration [2,13,107,122,157]. Resin-based adhesive cements can be classified as self-cured, dual-cured, or light-cured based on their polymerization mechanism [145]. An alternative innovation is the use of chairside pre-heated conventional restorative RBCs as a luting agent for indirect ceramic restorations. Owing to their reduced viscosity, low film thickness and good adaptation can be achieved [104]. In addition to color stability, favorable mechanical and physical properties are further benefits as a result of their high filler load [33,68,116]. An increased pre-polymerization temperature can improve the monomer-to-polymer conversion; however, this might be compromised during the luting procedure because of the rapid cooling of the luting RBC before it is light-cured [39,40, 84]. The durability of adhesively bonded restorations depends on the DC of the adhesive cement [59,120]. However, indirect ceramic restorations attenuate the light passing through them (Figure 7) [136]. The interposed material might impair the mechanical and esthetic properties of the luting agent caused by the reduced DC, thus compromising the durability of the indirect restoration [87,129]. To overcome or attempt to compensate for this drawback, the light intensity should be sufficiently high, or the exposure time should be as long as possible, considering light attenuation as a function of the restoration thickness [26,87].

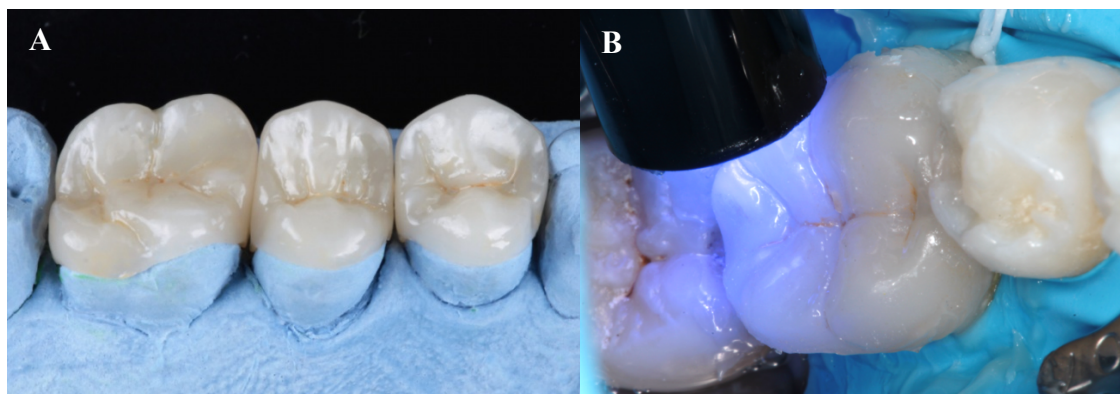


Figure 7. *Indirect ceramic restorations on the gypsum cast (A). Light curing of resin adhesive cement through the indirect ceramic restoration (B) [photo made by Edina Lempel]*

I.1.3.6. Biological considerations of pre-heating

An increase in irradiance delivered from the curing unit or upon extending the exposure time as a strategy to enhance the polymerization degree might cause an unfavorable temperature rise within the pulp chamber [162]. The temperature rise during polymerization is due to the absorption of energy by the irradiated objects together with the heating of the curing unit itself [23]. However, the polymerization of RBC materials is an exothermic reaction that leads to further heat generation [20,91]. Bouillaguet et al. [23] reported that by using infrared imaging, the highest temperature increases were recorded inside the RBC material and not outside the tooth during photocuring. The exothermic reaction is a material-, consistency-, and thickness-dependent phenomenon [91]. Although most of the heat generated during RBC polymerization is dissipated, the increase in pulpal temperature may exceed the putative pulpal damage threshold [134]. Several studies have consistently reported that the remaining dentin thickness is a critical factor in relation to the intrapulpal temperature increase due to the heat dissipating effect [70,162, 163]. According to the second law of thermodynamics, during light curing, heat flows from the external tooth surface or polymerizing material to the pulp chamber as the temperature difference is equalized by diffusion [32]. The thermal conductivity of human dentin was calculated at approximately 0.36–0.67 W/mK by de Magalhães et al. [48]. Although heat transfer or heat flux occurs at a lower rate in dentin, inducing a thermal insulating effect, the potential for pulpal damage is expected to be great in deep cavities, where the tubular surface area increases, and the light attenuation effect is weak [35,100,132]. Thus, clinically, it would be optimal for dentin preservation, or continuous high-energy output photo-curing should be avoided to protect pulp tissues from thermal injury [90]. Onisor et al. [121] conducted active cooling to reduce the heat during prolonged polymerization employed for luting indirect adhesive restorations with light-cured materials. Additionally, the shielding effect of dentin resulted in a lower pulpal temperature increase than that of the interposed ceramic restoration during light curing [90]. In addition to the interposed materials and the distance between the cavity floor and pulp, the effect of pulpal blood circulation, volume, and perfusion of the fluid in the dentinal tubules as well as the surrounding tissues play important roles in heat conduction and protection against the rise in pulpal temperature [86,133]. The putative pulpal damage threshold was based on the study conducted by Zach and Cohen. A temperature rise in pulpal tissues of 5.5 °C may lead to irreversible changes [106,164]. In support of the

above observation, a recent *in vivo* study showed that increased pulpal temperatures may induce inflammatory reactions, even if the temperature rise does not exceed the previously defined 5.5 °C threshold [69]. Although several investigations have been conducted on the effects of the light curing unit, RBC type, and remaining dentin thickness on the pulpal temperature rise, data are lacking in the dental literature regarding the effects of different dentin thicknesses on pulpal temperature change during cementation of different thicknesses of indirect ceramic restorations with adhesive resin cements or pre-heated restorative RBCs [20,66,83,91,100,123,139]. Furthermore, reliable, comparative data on the thermophysical properties of dentin and ceramic are essential to obtain precise calculations of the thermal changes in teeth and provide safer dental procedures, such as ceramic inlay cementation.

II. Objectives

II.1. Investigation of the effect of pre-heating on monomer elution and degree of conversion of contemporary and thermoviscous bulk-fill resin-based dental composites

Not only the physico-mechanical properties, but the chemical characteristics are also important determinants of the clinical performance and biocompatibility of an RBC. According to the literature data, there is a strong inverse correlation between DC and monomer elution, nevertheless both the amount of released monomers and the polymerization degree are influenced by several other factors such as the pre-cure temperature.

Detection of unreacted monomers from pre-heated bulk-fill RBCs is not a well-investigated topic so far, therefore, one of our purposes was to examine the temperature changes during the application and polymerization of a relatively new thermoviscous (VisCalor Bulk) and a high-viscosity full-body bulk-fill RBC (Filtek One Bulk Fill Restorative) in relation to different pre-cure temperatures. Further aims were to evaluate the effect of pre-heating on the DC using micro-Raman spectroscopy as well as determine the amount of released monomers using RP-HPLC.

Two null hypotheses were formulated: (1) Pre-heating had no effect on RBCs' post-cure DC%, and (2) pre-cure temperature did not affect the amount of released unreacted monomers.

II.2. Investigation of the effect of ceramic and dentin thicknesses and the type of resin-based luting agents on intrapulpal temperature changes during the luting of ceramic inlays

Besides the direct resin-based composite restorations, another common method for restoring the posterior region in the dental clinical practice is the use of indirect partial restorations (inlay, onlay, overlay) made of ceramic. The cementation of indirect ceramic restorations is performed with resin-based adhesive cements, in addition, alternative innovation is the use of pre-heated conventional restorative RBC as a luting agent. However, during the luting process, the energy emitted by the curing unit, the released

heat during the polymerization process and the pre-polymerization temperature of the pre-heated RBCs might cause an unfavorable temperature rise within the pulp chamber. Therefore, the other objective of our research project was to compare the intrapulpal thermal changes resulting from cementation of ceramic inlay with light- and dual-curing adhesive resin cements and pre-heated sculptable submicron restorative RBC. The aim was to assess the influence of simultaneously variable dentin and ceramic layer thicknesses on pulpal temperature rise, supplemented by a qualitative comparison of the thermal properties between dentin and ceramics.

The null hypotheses of the *ex vivo* study were threefold: (1) intrapulpal temperature change does not differ significantly using distinct luting agents during ceramic inlay cementation; (2) pulpal temperature rise is not influenced significantly by ceramic and dentin layer thicknesses; and (3) there is no significant difference between the thermal conductivity and heat capacity of dentin and ceramics.

III. Materials and methods

III.1. Investigation of the effect of pre-heating on monomer elution and degree of conversion of contemporary and thermoviscous bulk-fill resin-based dental composites

III.1.1. The investigated resin-based composites

Two brands of high-viscosity bulk-fill RBCs – the VisCalor Bulk and the Filtek One Bulk Fill Restorative – were investigated in this *in vitro* study (Table 1).

Table 1. The specifications of the investigated materials and their acronym codes

<i>Material (shade)</i>	<i>Manufacturer</i>	<i>Pre-Cure Temperature</i>	<i>Code</i>	<i>Resin System</i>	<i>Filler Type</i>	<i>Filler Loading</i>
VisCalor Bulk (A2)	Voco, Cuxhaven, Germany	25 °C	VCB_25	Bis-GMA, aliphatic dimethacrylates	Inorganic nanohybrid filler (not defined by the manufacturer)	83 wt%
		65 °C	VCB_65			
Filtek One Bulk Fill Restorative (A2)	3M ESPE, St. Paul, MN, USA	25 °C	FOB_25	AFM, UDMA, AUDMA, DDMA	20 nm silica, 4–11 nm zirconia, cluster Zr-silica, 0.1 µm ytterbium-trifluoride	58.5 vol%
		55 °C	FOB_55			76.5 wt%

III.1.2. Sample preparation

All of the samples were prepared by one operator in 4 mm layer thickness. According to the method of sample preparation there were two experimental groups for each of the two investigated materials. The pre-cure temperature of the RBC samples in the first group was 25 °C (room temperature) (FOB_25 and VCB_25), while RBCs in the second group

were pre-heated before the sample preparation. In case of VCB, pre-heating was performed by VisCalor Dispenser (VOCO, Cuxhaven, Germany) using T1 setting, which pre-warmed the device and RBC together to 65 °C in 30 s (VCB_65). Pre-warming of FOB was undertaken by Ena Heat Composite Heating Conditioner (Micerium, Avegno, Italy) using T2 setting, which pre-warmed the device and the RBC to 55 °C in 55 min and 15 min, respectively (FOB_55). Five specimens were prepared in each group, from each material for both the micro-Raman spectroscopy measurements as well as for the monomer elution measurements (n=40).

The samples were prepared in a cylindrical polytetrafluoroethylene (PTFE) mold with an internal diameter of 5 mm and a height of 4 mm, placed on a thermostatically controlled (30 ± 1 °C) glass slide to represent the isolated tooth. A polyester Mylar strip was positioned between the mold and the glass slide. A capsule dispenser gun was used to apply VCB_25, FOB_25 and FOB_55 materials into the mold. In the case of VCB_65 a VisCalor dispenser was used for both warming and application. The condensation of the RBCs was performed with room temperature hand instrument. Before irradiation, the RBC sample was covered with a Mylar strip to avoid the contact with oxygen. All specimens were irradiated with Light Emitting Diode (LED) curing unit (LED.D, Woodpecker, Guilin, China; average light output given by the manufacturer 850-1000 mW/cm²; $\lambda = 420\text{--}480$ nm; 8 mm exit diameter fiberglass light guide) in standard mode for 20 s, powered by a line cord at room temperature of 25 ± 1 °C, controlled by an air conditioner. The irradiance of the LED unit was monitored before and after polymerization with a radiometer (CheckMARC, Bluelight Analytics, Halifax, Canada). The tip of the fiberglass light guide was in direct contact, centrally positioned and parallel to the mold.

III.1.3. Temperature measurements

Temperature measurements during the application and the polymerization of RBCs were recorded with a registration device (El-EnviroPad-TC, Lascar Electronics Ltd., Salisbury, UK) attached to 0.5 mm diameter Cu/CuNi thermocouple probes (K-type, TC Direct, Budapest, Hungary) — positioned at the bottom of the temperature regulated mold — with a frequency of one measurement per second and resolution of 0.1 °C (Figure 8). The

quantity of heat emitted by the LED curing unit was also determined through the 4 mm empty mold.

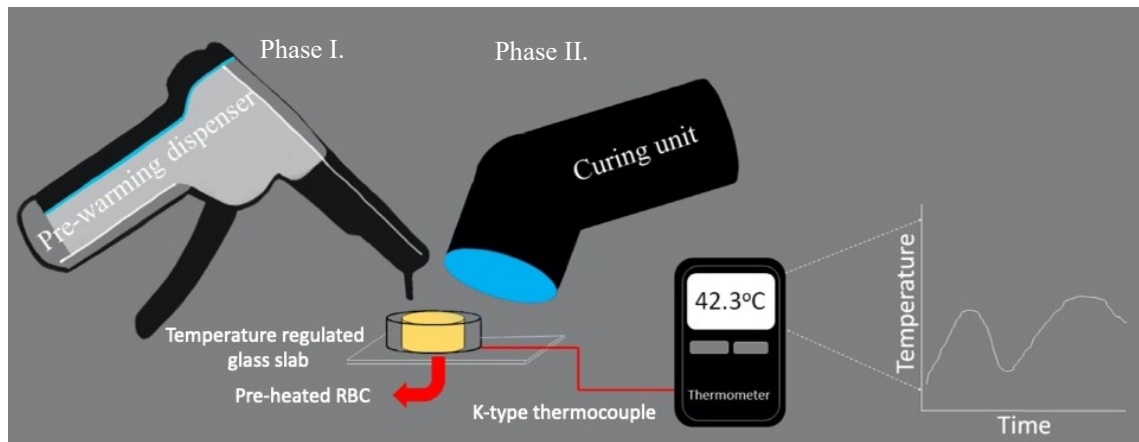


Figure 8. Schematic figure of the experimental set-up for sample preparation and temperature measurement [own editing]

III.1.4. Micro-Raman Spectroscopy Measurements

RBC samples made to estimate the temperature change of the RBC during sample preparation were then used to measure the DC. Confocal Raman spectrometer (Labram HR 800, HORIBA Jobin Yvon S.A.S., Longjumeau Cedex, France) was used to evaluate the 24 h post-cure DC values of the polymerized RBC samples. Setting parameters during the measurements were the following: 20 mW He-Ne laser with 632.817 nm wavelength, magnification x 100 (Olympus UK Ltd., London, UK), spatial resolution $\sim 15 \mu\text{m}$. Spectral resolution of $\sim 2.5 \text{ cm}^{-1}$ provided satisfactory results since the two peaks analyzed were $\sim 30 \text{ cm}^{-1}$ apart. Spectra were taken at three locations of the RBC samples (center, periphery and between these two regions) both from the bottom and top surfaces with an integration time of 10 s. Ten acquisitions were averaged for each geometrical point. Spectra of uncured RBCs were taken as reference. Post-processing and analysis of spectra was performed using the dedicated software LabSpec 5.0 (HORIBA Jobin Yvon S.A.S., Longjumeau Cedex, France). The ratio of double-bond content of monomer to polymer in the RBC was calculated according to the following equation (Equation 1):

$$\text{Equation 1. } DC\% = (1 - (R_{\text{cured}} / R_{\text{uncured}})) \times 100$$

where R is the ratio of peak intensities at 1639 cm^{-1} and 1609 cm^{-1} associated to the aliphatic and aromatic (unconjugated and conjugated) C=C bonds in cured and uncured RBCs, respectively.

III.1.5. Reversed-phase High-Performance Liquid Chromatography (RP-HPLC) measurements

Immediately after the irradiation, five samples of each material were immersed into 1.0 mL of the 75% ethanol/water storage medium in separate glass vials and stored in a 37°C incubator. As recommended by the ISO 10993-13 description, the ratio between the sample and the storage solution volume was greater than 1:10, thus the specimens were fully immersed in the medium. The storage solutions were collected for analysis after 72h. The RP-HPLC system (Dionex Ultimate 3000, Thermo Fisher Scientific Inc., Sunnyvale, CA, USA) consists of a Dionex LPG 3400 SD gradient pump, Rheodyne injector (Rheodyne, CA, USA), and a Dionex DAD 3000 RS UV-VIS detector (Dionex GmbH, Germering, Germany). Data acquisition was completed using Chromeleon software (version: 7.2.10). The separations were performed on a LiChrospher® 100 RP-18e (particle size: $5\text{ }\mu\text{m}$, pore size: 100 \AA) (Merck KGaA, Darmstadt, Germany) column ($250\text{ mm} \times 4.00\text{ mm}$) with gradient elution. The composition of Eluent “A” was 100% bidistilled water, whereas Mobile Phase “B” was 100% v/v acetonitrile (ACN) (VWR International, Radnor, PA, USA). During the 30-min chromatographic separation, the “B” eluent content increased from 30–95%. The flow rate was $1.2\text{ mL} \times \text{min}^{-1}$. For the regeneration of the stationary phase, the content of Mobile Phase B was decreased from 95% to 30% in 1 min, and after 31–46 min, the system was washed with 30% “A”. The detection of the eluted monomers was carried out at the following wavelengths: 205, 215, 227, and 254 nm. 205 nm were found to be optimal; therefore, the evaluation relied on the data collected at this wavelength [93]. The separations were undertaken at room temperature. The amounts of the eluted monomers (bisphenol A-diglycidil ether methacrylate, BisGMA; triethylene glycol dimethacrylate, TEGDMA; urethane dimethacrylate, UDMA; 1,12-dodecane dimethacrylate, DDMA) were calculated using the calibration curve with the areas under the curve of peaks produced by the monomers, respectively. The monomer release was counted to 1 mg RBC. The TEGDMA, UDMA,

BisGMA, and DDMA standard solutions had retention times of 12.2, 17.2, 19.1, and 27.2 min, respectively, whereas the peaks were well separated from each other.

III.1.6. Statistical Analysis

Pilot study results and sample size formula were used to estimate sample size.

$$\text{Sample size formula: } n = \frac{(z_{1-\frac{\alpha}{2}} + z_{1-\beta})^2 (s_1 + s_2)^2}{(M_1 + M_2)^2}$$

(z = standard score; α = probability of Type I error = 0.05; $z_{1-\alpha/2}$ = 1.96; β = probability of Type II error = 0.20; $1-\beta$ = the power of the test = 0.80; $z_{1-\beta}$ = 1.28, M_1 = 52, s_1 = 1.4, M_2 = 52, s_2 = 1.4). By adopting an alpha (α) level of 0.05 and a beta (β) level of 0.20 (power = 80%), the predicted sample size (n) was found to be a total of 3 samples per group. Instead of the calculated 3 samples, n = 5 per group sample size was selected.

The statistical analyses were performed with SPSS v. 26.0 (SPSS, Chicago, IL, USA). Levene's test was employed to test the equality of variance. This was followed by Paired Samples Test to analyze the differences in mean DC% between top and bottom surfaces and Two-tailed Independent Samples T-test to analyze the differences in mean DC% between the investigated materials polymerized at room temperature and with the application of pre-heating. The differences in monomer elution from the RBCs at the investigated temperatures were also compared with the Two-tailed Independent T-test. Multivariate analysis (General Linear Model) and Partial Eta-Squared statistics were used to test the influence and describe the relative effect size for *Material* and *Temperature* as independent factors. p values below 0.05 were considered statistically significant.

III.2. Investigation of the effect of ceramic and dentin thicknesses and the type of resin-based luting agents on intrapulpal temperature changes during the luting of ceramic inlays

III.2.1. The investigated resin-based luting agents

In this *in vitro* study, the effects of three resin-based luting agents — Variolink Esthetic LC (VE_LC) light-cured, Variolink Esthetic DC (VE_DC) dual-cured adhesive resin cement, and pre-heated sculptable submicron-filled restorative RBC, Estelite Sigma Quick (EQ_55 °C) — on pulpal temperatures were analyzed (Table 2).

Table 2. The brands, manufacturers, and chemical compositions of the luting agents						
<i>Material (shade)</i>	<i>Manufacturer</i>	<i>Classification</i>	<i>Resin System</i>	<i>Filler type</i>	<i>Filler loading</i>	<i>Code</i>
Variolink Esthetic LC (Light)	Ivoclar Vivadent, Schaan, Liechtenstein	Light-cured adhesive resin cement	UDMA; 1,10-DDMA	0.04–0.2 µm ytterbium trifluoride and spheroid mixed oxide	38 vol% 64 wt%	VE_LC
Variolink Esthetic DC (Light)	Ivoclar Vivadent, Schaan, Liechtenstein	Dual-cured adhesive resin cement	UDMA; 1,10-DDMA	0.04–0.2 µm ytterbium trifluoride and spheroid mixed oxide	38 vol% 64 wt%	VE_DC
Estelite Sigma Quick (A1 enamel)	Tokuyama Dental, Tokyo, Japan	Conventional submicron RBC pre-heated to 55 °C	BisGMA, TEGDMA	0.1–0.3 µm monodispersing spherical silica–zirconia filler; prepolymerized filler of silica-zirconia and copolymer	71 vol% 82 wt%	EQ_55 °C

III.2.2. The ceramic blocks used

Highly translucent A2 shade lithium disilicate ceramic blocks (6 × 6 mm) were fabricated from ceramic ingots (GC Initial LiSi Press; GC Europe, Leuven, Belgium) using the heat-pressed method and were then fired and glazed from one side according to the manufacturer's instructions. To achieve an even smoother surface, 220-, 400-, and 600-grit water-cooled sandpaper was used to finish the specimens, followed by polishing with a two-step rubber diamond polisher (fine, 8–32 µm grit size, Kenda Nobilis, Kenda AG, Vaduz, Liechtenstein; extra fine, 4–8 µm grit size, Kenda Unicus, Kenda AG, Vaduz, Liechtenstein). The fabricated ceramic blocks were intended to represent inlays with thicknesses of 2.0 mm, 2.5 mm, 3.0 mm, and 3.5 mm. The final dimensions of each ceramic block were determined using a digital caliper with an accuracy of 0.001 mm (Mitutoyo, Tokyo, Japan). To provide multiple measurements, the ceramic specimens were not acid-etched, silanated, or coated with an adhesive.

III.2.3. Cementation and radiant exposure

The ceramic blocks were cemented with light-cured adhesive cement, dual-cured adhesive cement mixed with an Automix syringe, and a pre-heated restorative RBC. A single-dose capsule of the latter RBC was pre-heated to 55 °C in an RBC warming device (Ena Heat Composite Heating Conditioner, Micerium, Avegno, Italy) for 15 min. Each capsule was heated once for the cementation of only one ceramic specimen. The resulting RBC temperature was measured using a non-contact infrared digital thermometer (TESTO 845, Testo Magyarország Kft., Budapest, Hungary). The infrared thermometer registered temperatures in an area as small as 1 mm² (optical resolution of 75:1), with a resolution of 0.1/1 °C. The data sampling frequency was 10 measurements/s. The ceramic blocks were pre-warmed in a composite warming device to reduce heat dissipation during cementation.

During each cementation a light-emitting diode (LED) light curing unit (LCU) (LED.D, Woodpecker, Guilin, China; $\lambda = 420\text{--}480$ nm; 8 mm exit diameter fiberglass light guide) was used in the standard mode for 40 s of exposure time. The LCU was powered by a line cord at room temperature (24 °C ± 1 °C). The position of the light guide tip was standardized to ensure that each sample received the same light beam character. A

CheckMARC radiometer (Bluelight Analytics, Halifax, NS, Canada) was used to monitor the radiant exitance (mW/cm^2) of the LCU before and after exposure. The tip of the LCU was placed at a standard distance of 1 mm from the radiometer sensor.

III.2.4. Sample preparation and pulpal temperature measurements

A caries-free, freshly extracted, human mandibular third molar for use in this study was cleaned and kept in physiological saline at room temperature. Ethical approval (No. PTE/3795) was obtained from the Regional Research Ethical Committee of University of Pécs. All thermal measurements were performed on a single-tooth model to limit any effects of structural differences in the dental hard tissues [20]. The apices of the roots were cut 5 mm from the furcation to expose the root canals, and all the pulpal residues were removed with an endodontic file, which was followed first by irrigation with 5.25 weight% sodium hypochlorite solution (Chloraxid, Cerkamed, Stalowa Wola, Poland) and then saline (NaCl 0.9%, B. Braun, Melsungen, Germany), and was finally dried with paper points (DiaDent, Burnaby, BC, VIEW 1 Canada). A hole was prepared on the mesial side of the tooth with a cylindrical diamond bur (836-012-FG cylinder diamond bur, 1.2 mm, medium; Meisinger USA, Centennial, CO, USA) to allow the insertion of the 0.5 mm diameter Cu/CuNi thermocouple probe (Type K thermocouple device; $\varnothing = 0.5$ mm; Cu/CuNi; TC Direct, Budapest, Hungary). The thermocouple sensor was positioned on the dentin at the top of the pulp chamber and assessed radiographically. To replicate the pulp tissue, the pulp chamber and root canal were injected with ECG gel (Aqua Sound Basic, Ultra-gel Hungary 2000, Budapest, Hungary). A flowable RBC (Filtek Supreme Flowable, 3M, St. Paul, MN, USA) was used to close the mesial hole and apical orifice, and the tooth was embedded in clear acrylic 1.0 mm below the cemento–enamel junction. The occlusal surface was prepared and polished flat, leaving dentin with a 2.5 mm thickness from the top of the pulp chamber. The occlusal thickness to be removed was estimated and controlled using digital intraoral radiography (Figure 9).

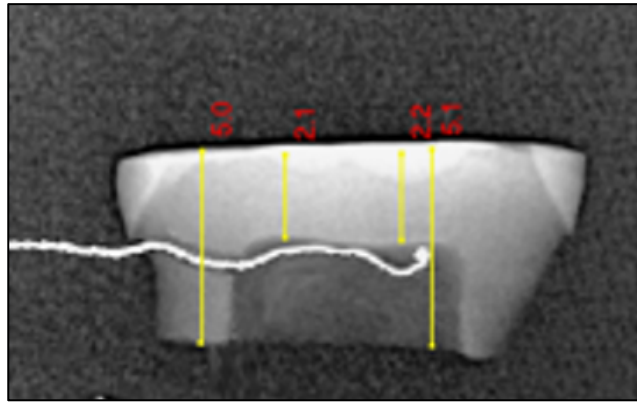


Figure 9. X-ray control of the K-type thermosensor location and the remaining occlusal dentin thickness (mm) [own editing]

To provide standard conditions during radiographic control of dentin reduction, the acrylic holder of the tooth was inserted in a poly-vinyl siloxane (Aquasil Ultra Plus, Dentsply Sirona, Charlotte, NC, USA) holder which was able to maintain the standard distance between the digital sensor and tube and provided correct position of the tooth for the parallel radiographic technique (Figure 10).

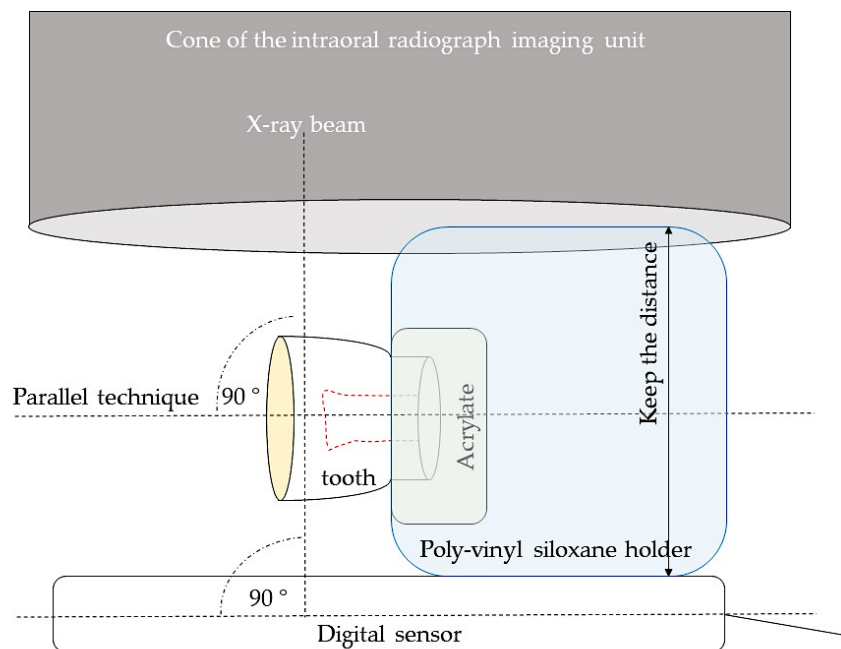


Figure 10. Schematic figure of the experimental set-up for radiograph taking during dentin thickness reduction [own editing]

After implementation of the first series of measurements with the different thicknesses of ceramic specimen and luting agent combinations, the dentin was reduced by 0.5 mm, resulting in a 2.0 mm dentin thickness. Following the conduction of the second series of measurements, the dentin thickness was further reduced by 0.5 mm, leaving behind 1.5 mm of dentin above the pulp chamber. For the last series of temperature registration, the dentin thickness was 1.0 mm thick after further reduction of 0.5 mm. The thickness of the remaining dentin was radiographically assessed. A cylindrical polytetrafluoroethylene (PTFE) mold with a crown diameter of ~12 mm and thicknesses of the four ceramic plates, with an inner hole of 6 × 6 mm, was fabricated to represent the axial walls of the cavity. The sample tooth was isolated with a rubber dam supported by a frame and immersed in a water bath at 36.0 ± 0.5 °C (Figure 11). Temperature measurements were recorded using a digital thermometer (El-EnviroPad-TC, Lascar Electronics Ltd., Salisbury, UK) attached to the above-described thermocouple, with a resolution of 0.1 °C and a frequency of one measurement per second.

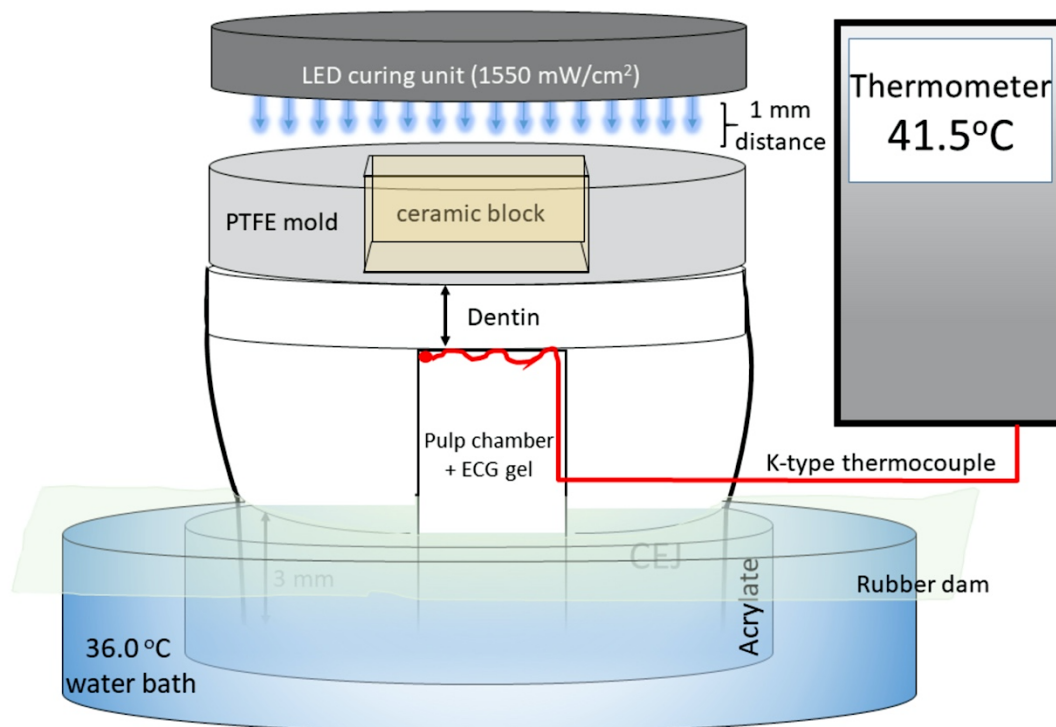


Figure 11. Schematic figure of the pulpal temperature measurement set-up [own editing]

First, the intrapulpal temperature changes during light exposure (40-s exposure) of dentin adhesive through the different dentin thicknesses using the empty molds were measured.

Prior to dentin adhesive (Adper Single Bond 2, 3M ESPE, St. Paul, MN, USA; without acid conditioning) application an agar/alcohol solution (3 wt% agar dissolved in 1:1 alcohol/water) was applied on the prepared dentin surface allowing the polymerized adhesive/luting agent to be removed without remnants. Thereafter, the thermal effect of the LCU through the eight combinations of different thicknesses of dentin and ceramic (Table 3) was recorded using the digital thermometer.

<i>D</i>	1.0	1.5	1.5	2.0	2.0	2.5	2.5	2.5
<i>C</i>	3.5	3.5	3.0	3.0	2.5	3.0	2.5	2.0

Temperature changes during cementation with the three resin-based luting agents were also measured.

A standard volume of the luting agent was applied to the center of the non-glazed ceramic surface, which was then centrally oriented to the tooth surface and manually (using the Optrasculpt instrument; Ivoclar Vivadent, Schaan, Liechtenstein) loaded with a 5-N load when resin adhesive cements were used and a 10-N load when the pre-heated restorative RBC was applied. Preliminary test results showed that manual placement of the ceramic specimen with a load of 5 or 10 N, depending on the type of luting agent, achieved a consistent layer thickness of $100 \pm 10 \mu\text{m}$. The load was measured using an algometer (Force Dial FDK 16, Wagner, Greenwich, USA). A microbrush was used to remove excess luting agent, which was photoactivated through the ceramic block for 40 s. After the cemented specimen was removed, its thickness was measured using a digital caliper. The film thickness of the luting agent was calculated from the difference between the thicknesses of the cemented specimen and ceramic block alone. There were 24 groups of the various dentin/ceramic/luting agent combinations, and the temperature measurements were recorded five times for each group ($n = 120$).

As separating solution was used on the tooth surface and no ceramic surface treatment was applied, the polymerized luting agent could be removed from both the tooth and ceramic block following the measurements, without leaving any deposits on either surface.

III.2.5. Differential scanning calorimetric measurements

A new series of the above-described ceramic blocks with thicknesses of 2.0 mm, 2.5 mm, 3.0 mm, and 3.5 mm were used for microcalorimetric measurements. Dentin slices were obtained from three third molars of a patient who was 19 years old. The teeth were sound, with no cracks or caries. The cleaned teeth were stored in saline at room temperature before testing, which took place within two days after extraction. To obtain dentin slices, the occlusal surface of the crown was horizontally removed using an orthodontic model trimmer (Gamberini, Bologna, Italy) under a water coolant. A minimum of 4 mm of dentin remained without exposing the pulp chamber. Dentin specimens with thicknesses of 1.0 mm, 1.5 mm, 2.0 mm, and 2.5 mm were sectioned using a precision cutter (IsoMet Low Speed Precision Cutter, Buehler, Lake Bluff, IL, USA) under a water coolant perpendicular to the long axis of the tooth. The enamel was removed to ensure that all sections contained only dentin, with final dimensions of 6×6 mm. The final dimensions of each dentin slice were measured using a digital caliper. Both ceramic and dentin specimens were weighed (ML-T Precision Balances, Mettler-Toledo, Greifensee, Switzerland).

Microcalorimetric measurements were carried out using a MicroSC microcalorimeter (Setaram Instrumentation, Caluire, France) in the differential scanning mode. Data were evaluated using the Calisto thermal analysis software (Setaram Instrumentation, Caluire, France). During the measurements, the samples were first equilibrated thermally for 5 min at 20 °C; then, the temperature was switched to 40 °C. The heat flow was recorded and plotted against time within 0–10 min. Considering that the sample holder itself needed 25 s to reach 40 °C after equilibration at 20 °C, data collected after 2 min were used for evaluation. Data evaluation was based on the following assumption: convective heat transfer between the sample holder of the microcalorimeter equipment and the samples occurred through the surface of the samples. The sample with higher heat conductivity requires less time to reach the thermal equilibrium at 40 °C. Accordingly, the data collected within the time range of 2–10 min were applied to estimate the heat conductivity, whereas the data collected within the time range of 0–10 min were applied to determine the heat capacities. The rate of the development of the thermal equilibrium at 40 °C was applied to compare the heat conductivities of the ceramic and dentin samples. The formation of thermal equilibrium was described by fitting the following equation (Equation 2) to the measured data:

$$\text{Equation 2. } \text{heat flow} = A * e^{\frac{-t}{\tau}}$$

where A is the pre-exponential factor (proportional to the heat capacity); t is the time required; τ is the time constant describing the rate of formation of thermal equilibrium; and e is the base of the natural logarithm. Measurements were performed in triplicate for each sample.

III.2.6. Statistical analysis

An earlier pilot study and a sample size formula were used to estimate the sample size [124].

$$\text{The sample size formula is as follows: } n = \frac{(z_{1-\frac{\alpha}{2}} + z_{1-\beta})^2 (s_1 + s_2)^2}{(M_1 - M_2)^2}$$

where z = standard score; α = probability of Type I error = 0.05; $z_{1-\alpha/2} = 1.96$ for 95% confidence; β = probability of Type II error = 0.20; $1-\beta$ = the power of the test = 0.80; $z_{1-\beta}$ = value of standard normal variate corresponding to 0.80 value of power = 0.84; s_1 = standard deviation of the outcome variable of group 1 = 0.64; s_2 = standard deviation of the outcome variable of group 2 = 0.36; M_1 = mean of the outcome variable of group 1; M_2 = mean of the outcome variable of group 2; $(M_1 - M_2) = 0.5$, if it is expected to detect 0.5 °C difference between two investigated groups as significant. Using the formula $N = \frac{2n}{1-0.1}$, the predicted sample size (n) was found to be 4.7 samples per group. According to the calculations, n = 5 per group sample size was indicated.

The SPSS v. 26.0 (SPSS, Chicago, IL, USA) software was used to perform statistical analyses. The normality of the data distribution was tested using the Kolmogorov–Smirnov test, followed by the application of parametric statistical tests. The differences in temperature changes were compared using one-way analysis of variance (ANOVA). Tukey’s post hoc adjustment was used for multiple comparisons for all the ANOVA models. To evaluate and explain the relative effect size on *dentin* and *ceramic thickness*, as well as *luting material* as independent variables, a General Linear Model (Multivariate analysis) and Partial Eta-Squared statistics were applied. Linear Regression was used to determine the correlation between dependent (*temperature*) and independent (*dentin thickness*, *ceramic thickness*, and *luting material*) variables. The statistical significance was set at $p < 0.05$.

IV. Results

IV.1. Investigation of the effect of pre-heating on monomer elution and degree of conversion of contemporary and thermoviscous bulk-fill resin-based dental composites

IV.1.1. Radiant exposure and temperature measurements

The measured maximum radiant exitance of the LED LCU was 1250 ± 15 mW/cm². The delivered radiant exposure was 25 J/cm². The LCU increased the temperature by an average of 7 °C when the thermocouple was irradiated through the empty 4 mm deep mold for 20 s.

Meanwhile, the Ena Heat Composite Heating Conditioner T2 setting stated preset temperature is 55 °C in 55 s, the real temperature of the FOB was 46 °C in the capsule after the recommended pre-warming period. The VisCalor Dispenser T1 setting provides 65 °C pre-warming in 30 s, however, the actual temperature of VCB was 60 °C after the recommended duration of pre-heating. Figure 12 shows the temperature change of RBC during the sample preparation from the material application into the mold until the end of the polymerization.

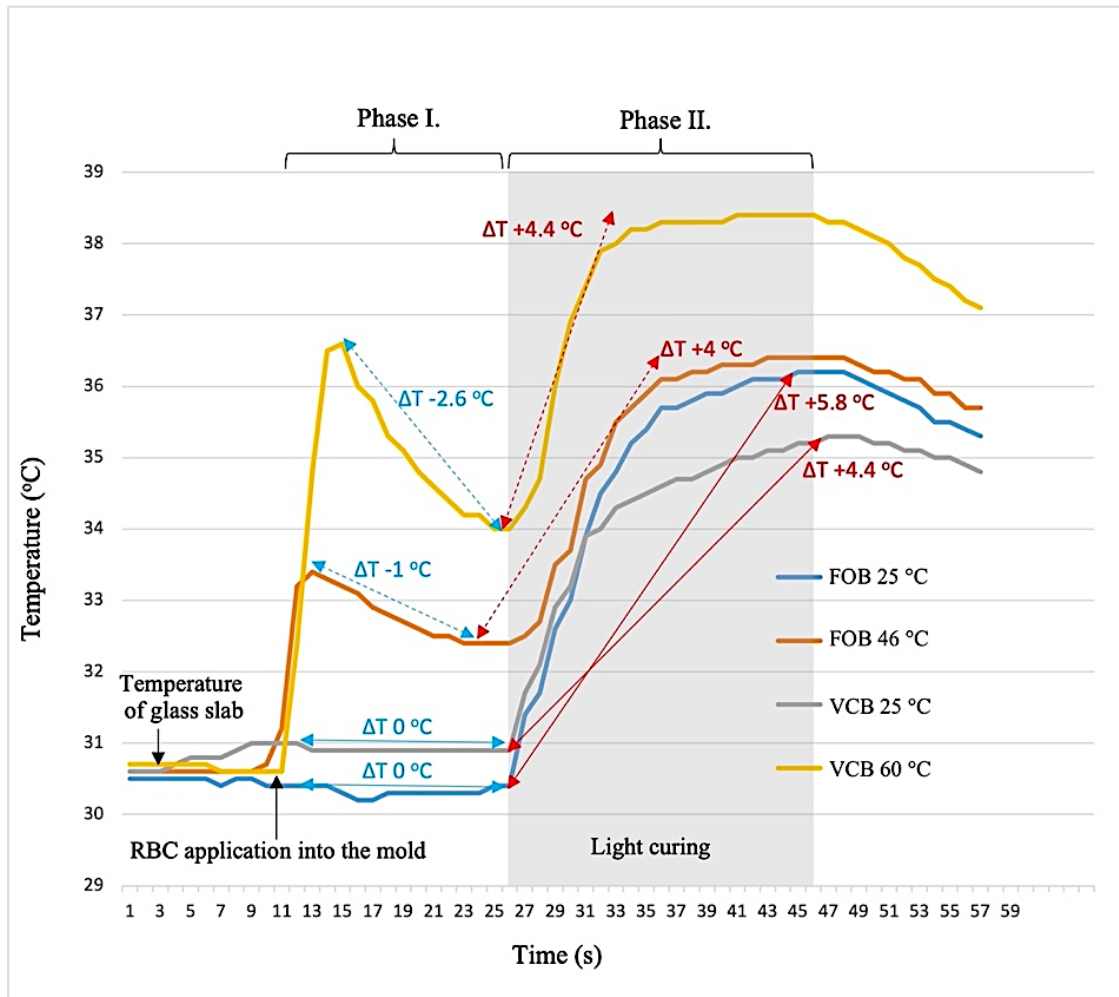


Figure 12. Temperature change of RBCs during the sample preparation

The temperature of the 46 °C pre-heated FOB decreased to 33.4 °C as it was removed from the warming device and started to be applied into the mold and showed further temperature drop of 1 °C during the condensation. The total temperature decrease from the pre-heating until the start of polymerization was 13.6 °C in approximately 20 s. In the second phase, during polymerization, the exothermic reaction and the heat released from the curing unit elevated the RBC's temperature by 4 °C. Regarding the thermoviscous VCB, its pre-heated temperature (60 °C) decreased to 36.6 °C during the initial phase of the application and continued to show a further drop of 2.6 °C during the condensation phase. The total drop of temperature for the pre-heated VisCalor Bulk from the pre-heating until the start of polymerization was 26 °C in approximately 20 s. The temperature rise caused by the light-curing and the exothermic reaction was 4.4 °C.

The consistency of the room temperature materials was highly viscous, especially of VisCalor making it difficult to squeeze out of the capsule. Compressing both room temperature RBCs was easy without sticking to the instruments. The pre-heating decreased the viscosity to a flowable consistency which allowed both materials to spread evenly throughout the template.

IV.1.2. Micro-Raman spectroscopy measurement

Considering the DC at the top and bottom surfaces in samples applied in 4 mm thickness, the mean percentages ranged between 54.2–64% and 45.0–51.8%, respectively (Table 4 and 5).

Table 4. Differences in mean DC% (S.D.) between top and bottom surfaces at room temperature						
	<i>Top</i>	<i>Bottom</i>	<i>t-value*</i>	<i>p-value*</i>	<i>95% CI</i>	
					<i>Lower</i>	<i>Upper</i>
<i>FOB</i>	63.0 (2.0)	51.8 (1.4)	31.7 (4)	<0.005	26.3	31.3
<i>VCB</i>	54.2 (2.9)	46.2 (1.4)	4.6 (4)	0.01	1.2	5.1

Table 5. Differences in mean DC% (S.D.) between top and bottom surfaces at pre-heated condition						
	<i>Top</i>	<i>Bottom</i>	<i>t-value*</i>	<i>p-value*</i>	<i>95% CI</i>	
					<i>Lower</i>	<i>Upper</i>
<i>FOB</i>	64.0 (1.4)	45.0 (1.9)	-17.2 (4)	<0.005	15.1	20.8
<i>VCB</i>	45.2 (4.1)	45.2 (4.1)	-25.1 (4)	<0.005	33.6	41.9

The DC values at the bottom of the specimens showed a statistically significant decrease for both materials at both temperatures compared to the DC values measured at the top. When room temperature specimens were applied, the DC values were very similar to that of pre-heated samples, except on the bottom surface of FOB which was significantly lower when applied after pre-heating. In a comparison of the two bulk-fill RBCs, VCB showed a statistically significantly lower DC (~10% less) both on the top and bottom when applied at room temperature. Samples applied following pre-heating showed a

significantly lower DC% only on the top. The lowest DC values were measured on the bottom surfaces of both investigated RBCs when they were applied with pre-heating as shown in the Figure 13.

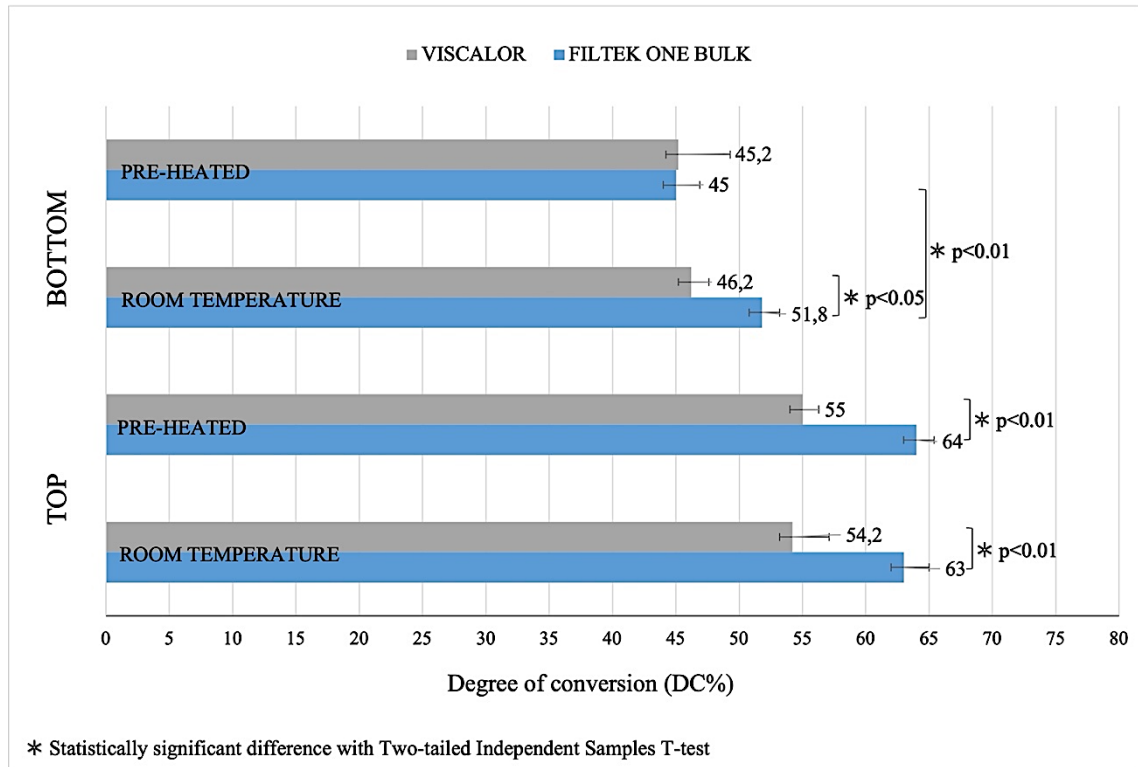


Figure 13. Comparison of degree of conversion between the two investigated RBCs

Table 6 shows the relative effect size of two independent variables, such as *Material* and *Temperature*, and additionally their interactions on the DC.

Table 6. The relative effect size of factors Material, Temperature and their interactions on degree of conversion of the top and bottom surfaces of the resin-based composites analyzed by General Linear Model and Partial Eta-Squared (η^2) statistics.				
Factor	DC			
	Top		Bottom	
	p-value	Partial η^2	p-value	Partial η^2
Material	< 0.001	0.86	0.028	0.27
Temperature	0.23	0.09	0.004	0.29
Material x Temperature	0.81	0.004	0.022	0.29

A 2 (*Material*) x 2 (*Temperature*) mixed-model ANOVA revealed that the main effect for *Material* on DC values measured on top surfaces was significant and the Partial Eta-

Squared was considered to be large. The effect of *Temperature* was found insignificant with medium effect size. The interaction (*Material x Temperature*) had no effect on the monomer conversion at the top. Regarding the DC values at the bottom surfaces, a significant effect for both the *Material* and *Temperature* factor was obtained. The interaction between the two variables (*Material x Temperature*) also significantly affected the monomer conversion at the bottom surfaces. The main effect for *Material* was significant at room temperature, meanwhile, the *Temperature* factor affected significantly only the FOB, but not the VCB.

IV.1.3. Reversed-phase HPLC measurement

During the HPLC measurements, in addition to the monomers specified by the manufacturers, other methacrylates, such as BisGMA from FOB and TEGDMA, DDMA from VCB were also detected with the help of standard monomers (Figure 14).

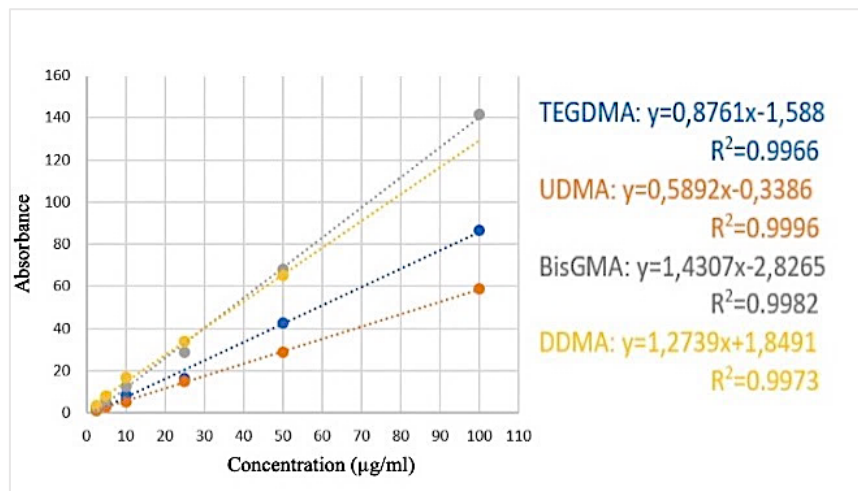


Figure 14. Calibration curves of the standard monomers

The differences in the monomer elution were also significant between FOB and VCB both when applied at room temperature and with pre-heating in the case of all the evaluated monomers, except for DDMA. This latter was released in similar (statistically insignificant) amounts from the pre-heated RBCs (Table 7 and Figure 15).

Table 7. Differences in monomer elution from Filtek One Bulk and VisCalor Bulk analyzed by Independent Samples T-test							
	Monomer	Resin composite	Mean (S.D.)	t-value (df)	p-value	95% CI	
						Lower	Upper
Room temperature	TEGDMA	FOB	0				
		VCB	0.1 (0.01)				
	UDMA	FOB	0.33 (0.34)	20.57 (8)	<0.001	0.29	0.36
		VCB	0.01 (0.01)				
	BisGMA	FOB	0.22 (0.01)	-19.43 (8)	<0.001	-2.33	-1.84
		VCB	2.3 (0.24)				
DDMA	FOB	0.26 (0.02)	13.08 (8)	<0.001	0.13	0,18	
	VCB	0.1 (0.02)					
Pre-heated	TEGDMA	FOB	0				
		VCB	0.08 (0.01)				
	UDMA	FOB	0.17 (0.11)	2.92 (8)	0,02	0.03	0.27
		VCB	0.02 (0.03)				
	BisGMA	FOB	0.08 (0.08)	-10.9 (8)	<0.001	-2.29	-1.49
		VCB	1.98 (0.38)				
	DDMA	FOB	0.15 (0.09)	1.19 (8)	0.27	-0.05	0.16
		VCB	0.09 (0.04)				

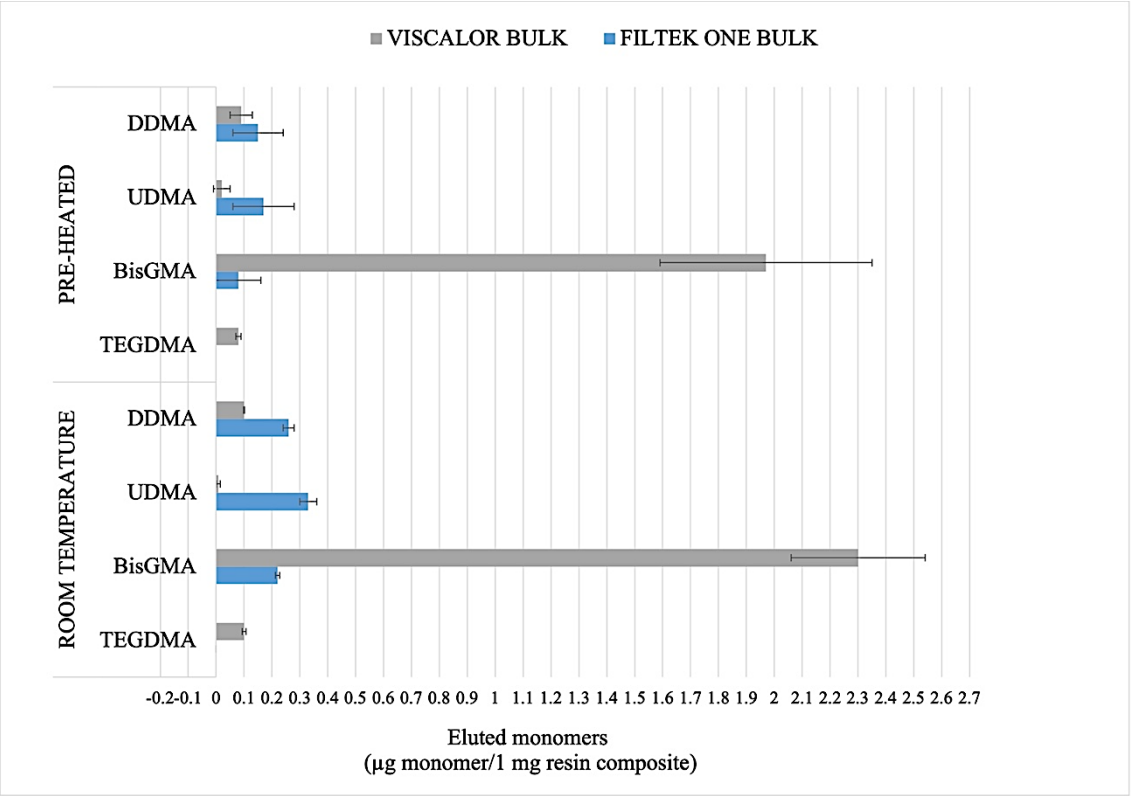


Figure 15. Monomer elution from Filtek One Bulk Fill Restorative and VisCalor Bulk when applied at room temperature and with pre-heating

At room temperature, 30 and 2.5 times as much UDMA and DDMA were released, respectively from FOB, while 10.5 times more BisGMA was eluted from VCB. The latter was the monomer released in the largest amount. With the utilization of pre-heating, 7.5 times as much UDMA was found to elute from FOB, while 25 times more BisGMA was released from VCB. For FOB, pre-heating significantly reduced the amount of eluted monomers, while for VCB, the temperature did not affect the dissolution (Table 8).

Table 8. Differences in monomer elution at room temperature and at pre-heated condition analyzed by Independent Samples T-test							
	<i>Monomer</i>	<i>Temperature</i>	<i>Mean (S.D.)</i>	<i>t-value (df)</i>	<i>p-value</i>	<i>95% CI</i>	
						<i>Lower</i>	<i>Upper</i>
<i>FOB</i>	<i>TEGDMA</i>	Room temperature	0				
		Pre-heated	0				
	<i>UDMA</i>	Room temperature	0.33 (0.34)	3.04 (8)	0.016	0.04	0.28
		Pre-heated	0.17 (0.11)				
	<i>BisGMA</i>	Room temperature	0.22 (0.01)	3.73 (8)	0.006	0.05	0.22
		Pre-heated	0.08 (0.08)				
	<i>DDMA</i>	Room temperature	0.26 (0.02)	2.6 (8)	0.03	0,01	0.21
		Pre-heated	0.15 (0.09)				
<i>VCB</i>	<i>TEGDMA</i>	Room temperature	0.1 (0.01)	3.12 (8)	0.014	0.01	0.04
		Pre-heated	0.08 (0.01)				
	<i>UDMA</i>	Room temperature	0.01 (0.01)	-0.87 (8)	0.41	-0.05	0.02
		Pre-heated	0.02 (0.03)				
	<i>BisGMA</i>	Room temperature	2.3 (0.24)	1.64 (8)	0.14	-0.13	0.79
		Pre-heated	1.98 (0.38)				
	<i>DDMA</i>	Room temperature	0.1 (0.02)	0.36 (8)	0.73	-0.04	0.05
		Pre-heated	0.09 (0.04)				

The following order of mean monomer elution was detected from FOB for both room temperature and pre-heated samples from highest to lowest: UDMA < DDMA < BisGMA, meanwhile the amount of leached monomers was roughly half (UDMA, DDMA) or one-third (BisGMA) for pre-heated specimens. Regarding VCB, both the order, as well as the amount of the released monomers were the same in the case of both the room temperature and the pre-heated samples (BisGMA < TEGDMA < DDMA < UDMA), except for TEGDMA, which showed a significantly lower elution from the pre-heated samples.

2 (*Material*) x 2 (*Temperature*) mixed-model ANOVA (Table 9) showed that the main effect for *Material* was significant on UDMA, BisGMA, DDMA release with a Partial

Eta-Squared value which was considered to be large. The *Temperature* factor also influenced significantly the monomer elution, however, its effect was slightly weaker compared to the *Material*'s effect. TEGDMA was released only from VCB. The effect of the *Temperature* factor was calculated to be significant on the elution of this monomer. The interaction between the two independent variables had a significant effect on UDMA and DDMA elution, while the elution of BisGMA was independent of the *Material x Temperature* interaction.

Table 9. The relative effect size of factors Material, Temperature, and their interactions on the monomer elution analyzed by General Linear Model and Partial Eta-Squared (η^2) statistics.

Factor	Monomer elution							
	BisGMA		UDMA		TEGDMA		DDMA	
	<i>p</i> -value	Partial η^2	<i>p</i> -value	Partial η^2	<i>p</i> -value	Partial η^2	<i>p</i> -value	Partial η^2
<i>Material</i>	< 0.001	0.96	< 0.001	0.75			< 0.001	0.49
<i>Temperature</i>	0.036	0.23	0.046	0.21	0.014	0.55	0.036	0.23
<i>Material x Temperature</i>	0.35	0.054	0.006	0.38			0.04	0.24

IV.2. Investigation of the effect of ceramic and dentin thicknesses and the type of resin-based luting agents on intrapulpal temperature changes during the luting of ceramic inlays

IV.2.1. Radiant exposure measurements

The maximum radiant exitance of the LED LCU was 1550 ± 15 mW/cm². The delivered maximum incident radiant exposure with a 40 s exposure duration was 62 ± 0.6 J/cm². The radiant exitance was reduced by 20% (1240 ± 12 mW/cm²) by the 6 × 6 mm orifice; thus, the radiant exposure with 40 s exposure duration was 49.6 ± 0.6 J/cm², which was delivered to the top of the ceramic specimens. The 2.0 mm, 2.5 mm, 3.0 mm, and 3.5 mm distances between the light guide tip and the radiometer sensor and the limited orifice of the mold significantly decreased the radiant exposure. Through the empty 2.0 mm, 2.5 mm, 3.0 mm, and 3.5 mm-deep molds, the radiant exposures decreased by 42% (36 ± 0.4

J/cm²), 45% (34 J/cm² ± 0.3), 48% (32.2 ± 0.3 J/cm²), and 51% (30.4 ± 0.3 J/cm²), respectively. The 2.0 mm, 2.5 mm, 3.0 mm, and 3.5 mm-thick ceramics further decreased the radiant exposures by 67% (20.5 ± 0.4 J/cm²), 70% (18.6 J/cm² ± 0.3), 73.2% (16.6 ± 0.3 J/cm²), and 76% (14.9 ± 0.3 J/cm²), respectively.

IV.2.2. Temperature measurements

The intrapulpal thermal changes induced by the 40 s light exposure of dentin adhesive through the 1.0 mm, 1.5 mm, 2.0 mm, and 2.5 mm dentin thicknesses, using the 2.0 mm, 2.5 mm, 3.0 mm, and 3.5 mm-deep empty molds are presented in Figure 16, representing the insulating effect of the dentin without the ceramic blocks and luting agents.

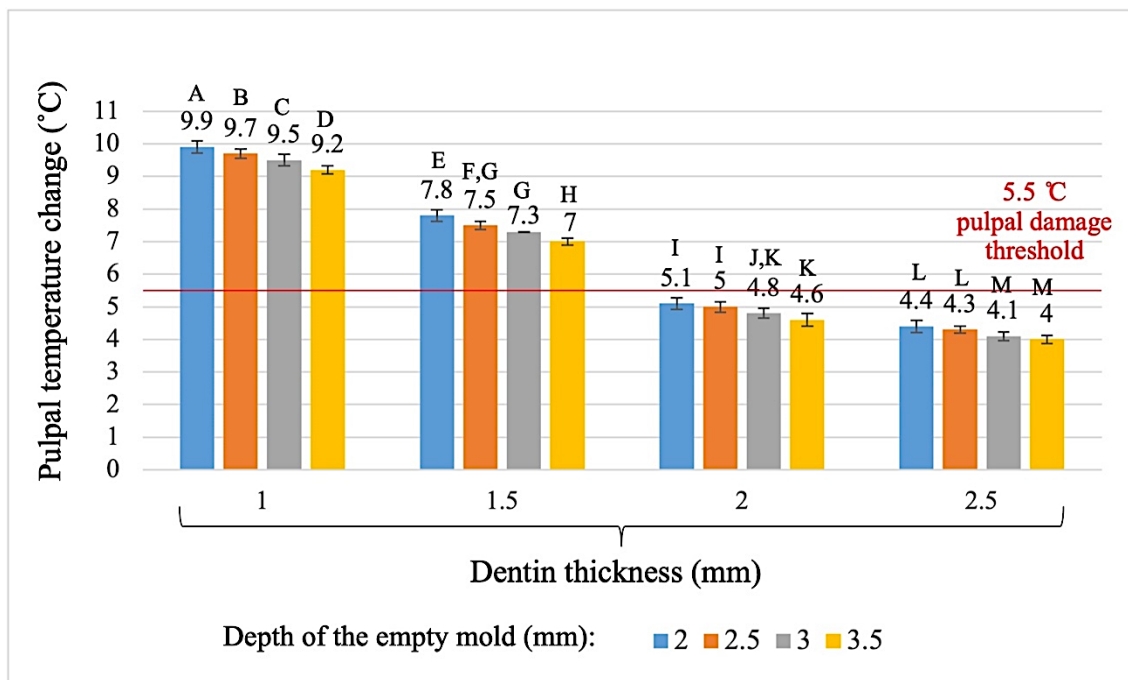


Figure 16. Intrapulpal temperature changes during light exposure of dentin adhesive through the different dentin thicknesses using the empty molds. Different capital letters (A–M) indicate statistically significant differences according to the one-way ANOVA and Tukey’s post-hoc test.

The thermal effect of the LCU through the eight combinations of different thicknesses of dentin and ceramic assemblies without luting agents is presented in Figure 17.

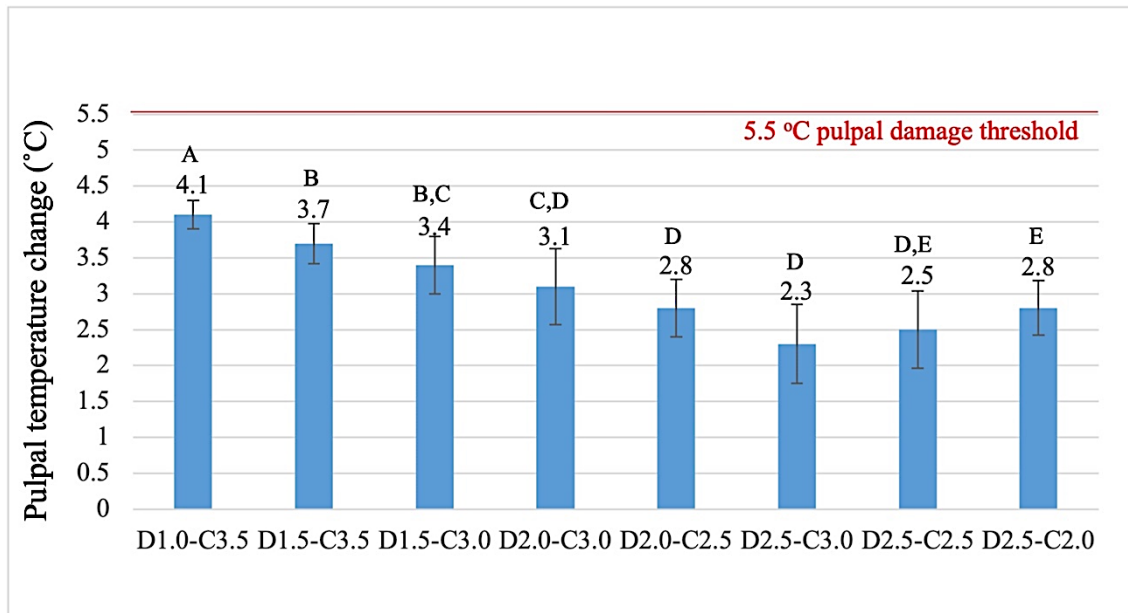


Figure 17. Pulpal temperature changes during light exposure through eight different combinations of dentin (D1.0, 1 mm; D1.5, 1.5 mm; D2.0, 2.0 mm; D2.5, 2.5 mm) and ceramic thicknesses (C2.0, 2.0 mm; C2.5, 2.5 mm; C3.0, 3.0 mm; C3.5, 3.5 mm) without luting cement. Different capital letters (A–E) indicate statistically significant differences according to the one-way ANOVA and Tukey’s post-hoc test.

None of the combinations approached the critical 5.5 °C threshold. Increased dentin thickness showed a more pronounced insulating effect than ceramic thickness.

Luting the ceramic blocks into cavities of different depths with light-cured and dual-cured adhesive cements, or with the restorative RBC pre-heated to 55 °C increased the pulpal temperature significantly (Figure 18).

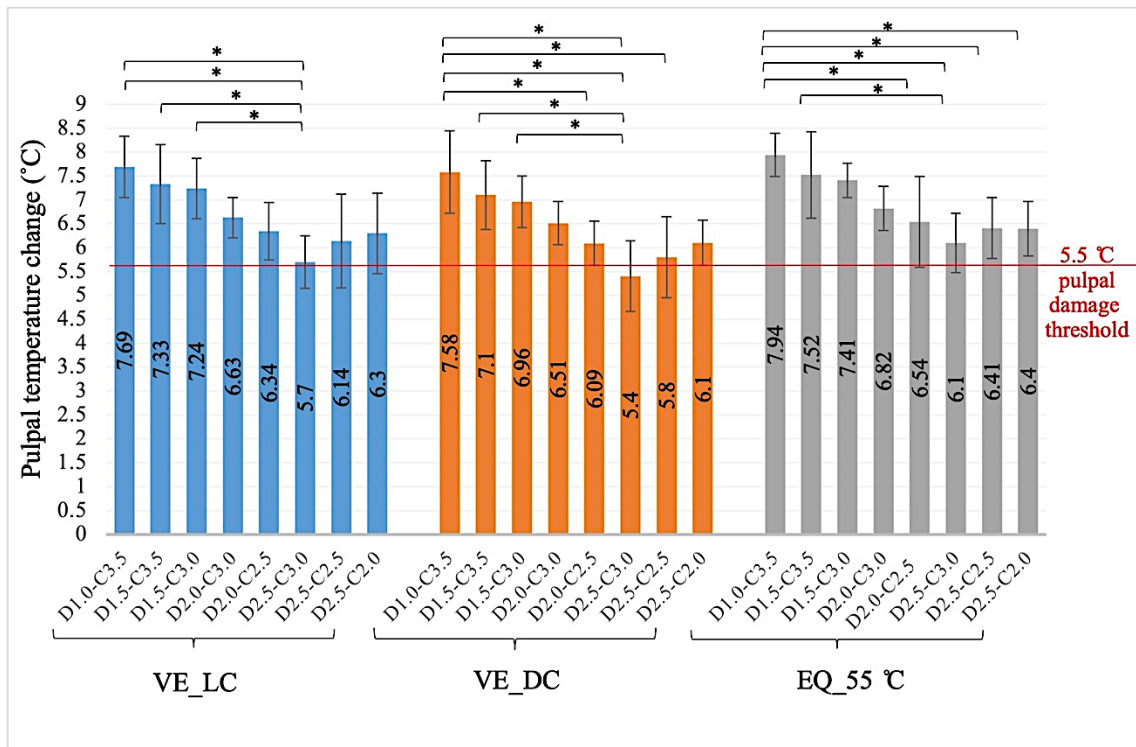


Figure 18. Pulpal temperature changes during cementation of eight different combinations of ceramic and dentin thicknesses cemented with light-cured and dual-cured adhesive cements and with the pre-heated restorative resin composite (*mark demonstrates statistically significant difference between groups according to the one-way ANOVA and Tukey's post-hoc test).

Subtracting the temperature rise caused by the LCU from the thermal change in the pulp chamber induced by the luting agent provides an estimation of the heat generated by the exothermic reaction. According to this calculation, which does not account for the thermal transfer between the thermodynamic system and its environment, the pre-heated RBC elevated the pulpal temperature to the highest value, although statistically significant difference was not detected between the luting materials (Figure 19).

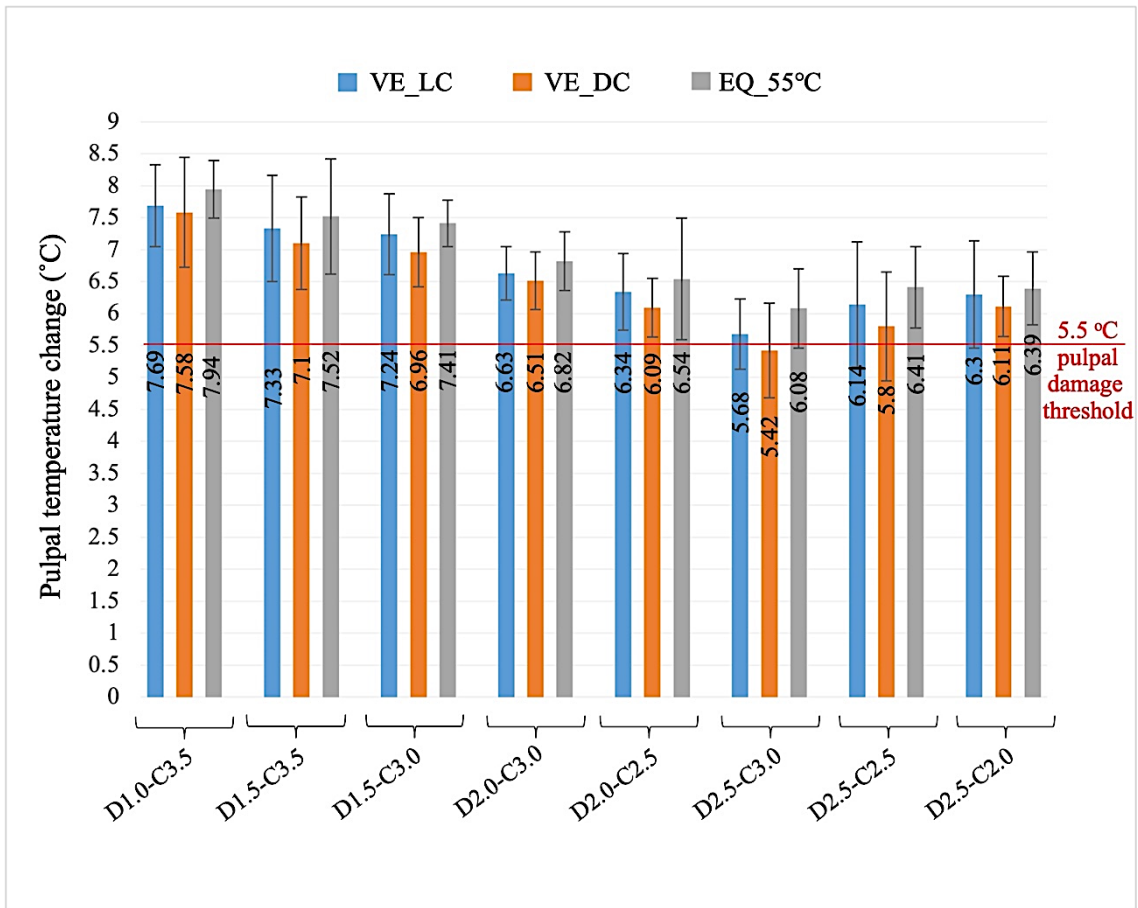


Figure 19. Comparisons of the pulpal temperature changes caused by the polymerization of light-cured and dual-cured adhesive cements and by the pre-heated restorative resin composite through eight different combinations of dentin and ceramic thicknesses (One-way ANOVA and Tukey's post-hoc test did not result in statistically significant differences between the tested groups.).

The mesh figure shows that dentin thickness below 1.5 mm is the most critical for the heat insulating effect (Figure 20).

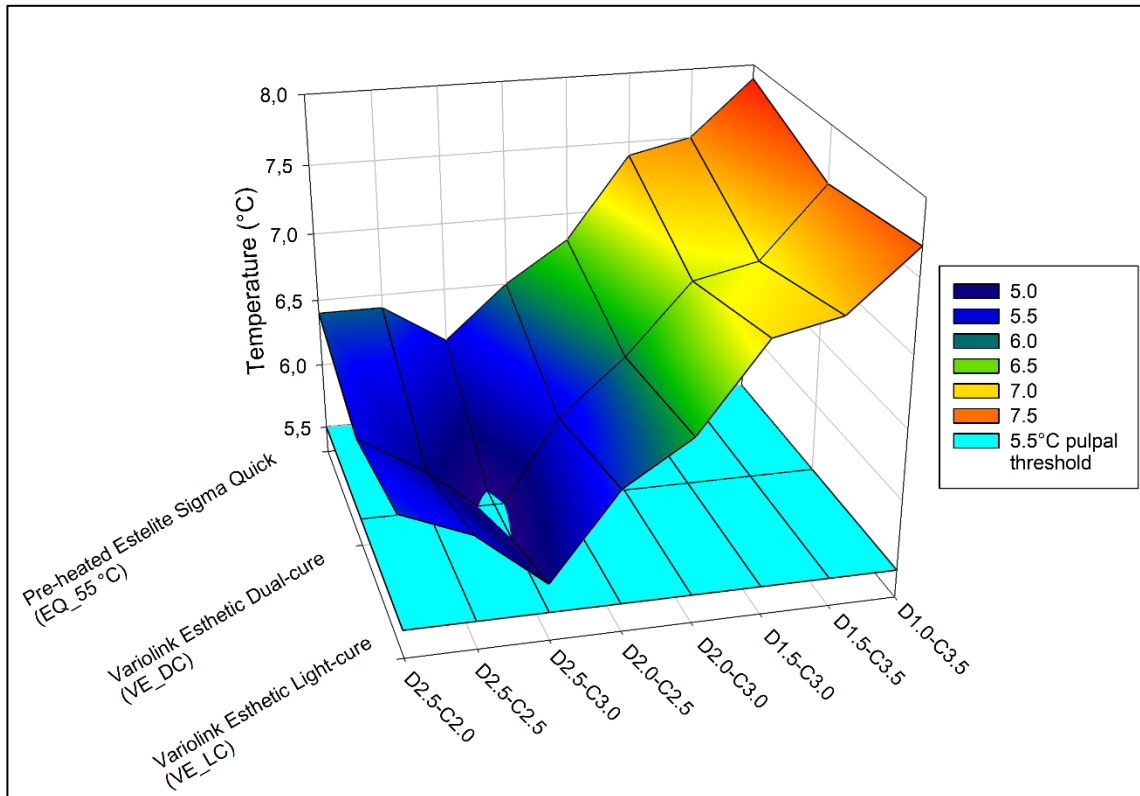


Figure 20. Changes in pulpal temperature during polymerization of the light-cured and dual-cured resin cements and pre-heated resin composite through different thicknesses of dentin and ceramic blocks representing an inlay. The light blue layer demonstrates the 5.5 °C temperature change thought to be associated with pulpal damage.

The Multivariate General Linear Model revealed that the *dentin thickness* had the main effect on the pulpal temperature changes ($F(3, 96) = 6.02, p = 0.001$), followed by the effect of *luting material* ($F(2, 96) = 4.29, p = 0.02$). The effect size was considered to be large for the *dentin thickness* (Partial $\eta^2 = 0.16$) and medium for the effect of *material* (Partial $\eta^2 = 0.08$). The effect of *ceramic thickness* on the pulpal temperature rise was considered to be insignificant according to the results of the General Linear Model ($F(3, 96) = 2.28, p = 0.09$), although the Eta-Squared indicated a medium effect (Partial η^2 was 0.07). However, there was no statistically significant three-way interaction between *luting material*, *dentin thickness*, and *ceramic thickness* [$F(14, 96) = 0.06, p = 1.0$; Partial $\eta^2 = 0.009$]. According to the linear curve fitting model, the data regarding the *dentin* and *ceramic thickness* and the tested *luting agents* allowed us to predict the behavior of the data series. The adjusted R-square statistics revealed higher values for *dentin* ($R^2 = 0.47$) and *ceramic thickness* ($R^2 = 0.41$), indicating a better fit; meanwhile, the R^2 value for the *luting materials* was 1%.

IV.2.3. Differential scanning calorimetric measurements

The thermal properties of the samples were evaluated by DSC measurements. The heat capacities were measured directly with the Calisto software by calculating the area of the curve, where the heat flow was plotted as a function of time until the curve reached the saturated region. Considering the mass of the samples and the area of the heating curves, the heat capacity of dentin was found to be 86% larger than the heat capacity of ceramic. To compare the heat conductivities, the time constant for the achievement of thermal equilibrium was applied. The average time constants of the dentin and ceramic samples were measured as 165.72 s and 133.33 s, respectively. The pre-exponential factors associated with the samples with increasing thickness and mass are $-1.35 \mu\text{W}$, $-4.08 \mu\text{W}$, $-6.08 \mu\text{W}$, and $-6.39 \mu\text{W}$, or $-7.13 \mu\text{W}$, $-9.13 \mu\text{W}$, $-11.38 \mu\text{W}$, and $-12.02 \mu\text{W}$ in respect to the dentin or ceramic samples. Calculating the thermal conductivity difference using the ratio of time constants, it was found to be approximately 24% lower in the case of dentin samples compared to ceramic specimens (Figure 21 and 22). Three scans of each sample were averaged then fitted using the exponential saturation function.

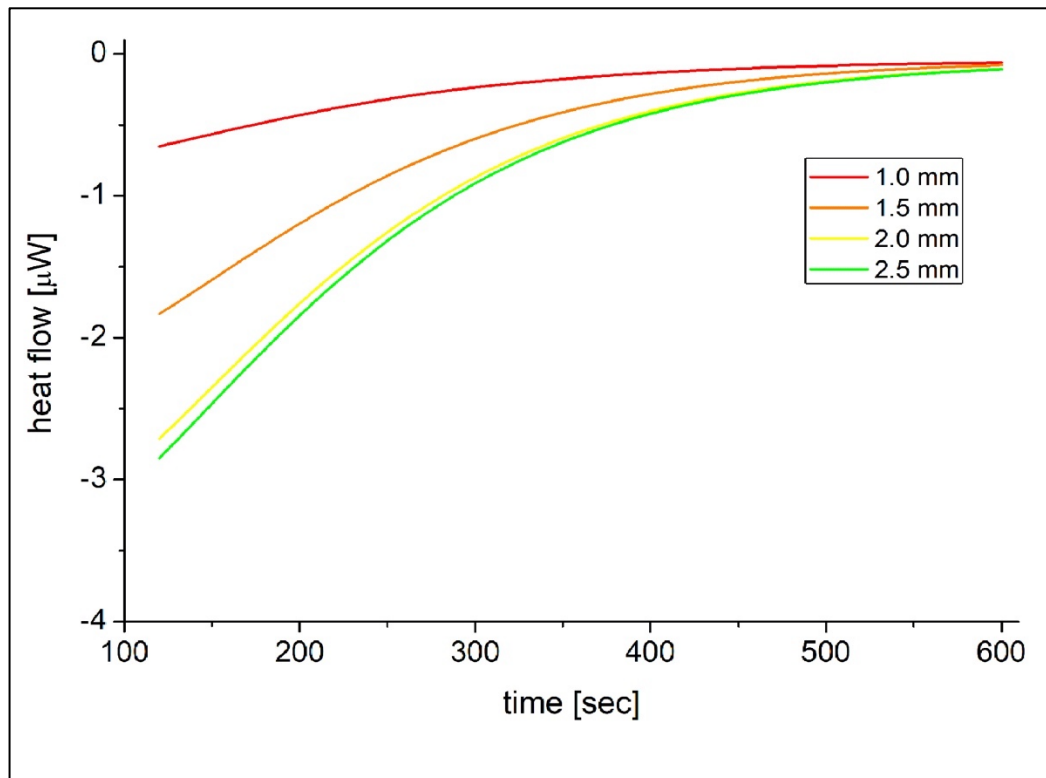


Figure 21. Heat flow associated with the dentin samples plotted as a function of the time

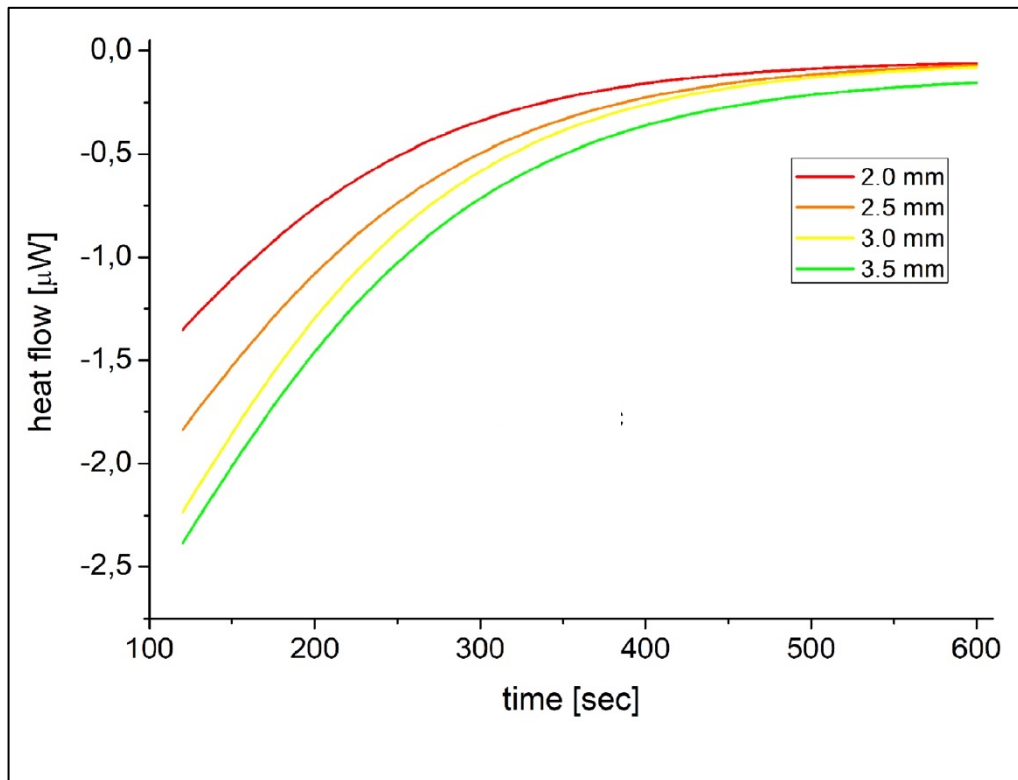


Figure 22. Heat flow associated with the ceramic samples plotted as a function of the time

V. Discussion

The purpose of pre-heating the RBCs is to eliminate the stickiness resulting from the viscosity, which allows good adaptation to the walls and margins of the cavity.

In addition, the pre-heated RBCs can also be used for adhesive cementation of indirect restorations, taking advantage of their low solubility, color stability and toughness resulting from their high filler loading.

During our investigations the aim was to test some properties of the pre-heated RBCs that may be indirectly related to the biocompatibility of the material, and its potentially harmful effect on dental pulp.

V.1. Investigation of the effect of pre-heating on monomer elution and degree of conversion of contemporary and thermoviscous bulk-fill resin-based dental composites

In our first *in vitro* study, the relation of DC and the elution of unreacted monomers of a thermoviscous and high-viscosity bulk-fill dental RBCs were assessed using micro-Raman spectroscopy and High-Performance Liquid Chromatography measurements. Additionally, the thermal change of the RBCs was also registered during the sample preparation with a K-type thermocouple to assess the temperature change of the room temperature and pre-heated RBCs during the manipulation and polymerization phases.

The setting reaction of RBCs has a major influence on their mechanical and biological properties [37]. RBC polymerization depends mainly on the chemical structure of the monomers, filler characteristics, the photoinitiator type and concentration, and the polymerization conditions [98]. The latter includes, among others, the volume and the layer thickness of the applied RBC, spectral characteristics of the curing unit, exposure time, and the pre-cure temperature of the material [94,63,72]. Since these were standardized in this study, except for pre-cure temperature, differences in the DC value and monomer elution of the bulk-fill RBCs can be attributed to the different compositions and temperatures of the materials before polymerization. Thus, the effect size of the material factor and pre-cure temperature factor became assessable on the degree of conversion and monomer elution from the investigated bulk-fill RBCs.

According to our results, the first null hypothesis, which stated that the pre-heating had no effect on post-cure DC% of VisCalor Bulk and Filtek One Bulk, was partially rejected,

since pre-warming of RBCs neither increased nor decreased the DC on the top of both materials and the bottom of VCB_65, however, the bottom DC was significantly decreased in the case of FOB_55. The second null hypothesis was also partially rejected, because the external heating of the investigated RBCs decreased the monomer elution in the case of FOB regarding all the investigated monomers and TEGDMA elution VCB, however, had no influence on the BisGMA, UDMA, and DDMA release from VCB.

It has been reported that increased pre-cure temperature of RBC may result in a greater extent of monomer to polymer conversion [40,150]. However, investigations, that have shown improvement in the degree of polymerization upon pre-warming generally maintained the RBC temperature constant during the experimentation [39,40]. On the other hand, there are also results that found increases in DC at non-isothermal conditions to be material composition-dependent [96,149].

Regarding the real-life clinical scenario, the RBC's temperature drops rapidly to the physiological level upon removal from the pre-heating device [102,108]. In contrast to the studies that have demonstrated optimized monomer conversion in the case of pre-heated RBCs under isothermal conditions, Yang et al. and Tauböck et al. reported that the pre-heated RBC's temperature dropped to ~35–36 °C during the handling phase before light-cure [158,149]. Additionally, the pre-warmed RBC can reach a lower internal temperature than the maximum stated preset temperature of the heating device [158,41]. To overcome this problem, a new warming device was developed, namely, VisCalor Dispenser. The capsule dispenser itself can provide homogeneous warming of the highly filled RBCs to 68 °C (only for VOCO products) with near-infrared technology. Thus, the RBC does not need to be removed from the heating device for dispensing into the prepared cavity. During application it is flowable and when it comes in contact with the tooth VCB reaches body temperature within a short time and thus returns to the high-viscosity, sculptable state.

In the present study, during the specimen preparation, the glass slab, holding the PTFE mold, was pre-set to a temperature of 30 ± 1 °C, representing a rubber dam isolated tooth [25]. In the case of both investigated materials, the temperature decreased during the extrusion from the capsule, irrespectively to the type of the heating device, and a further drop of temperature was observed during the condensation into the mold (Phase I on Figure 12). The measured temperatures for FOB_55 and VCB_65 were on average 32.5 °C and 34 °C, respectively, at the start of polymerization. The direct contact to the 30 °C molds and glass slab and also to the room temperature condensing instrument accelerated

the cooling of the RBCs. The equilibration of the ambient and the pre-heated RBC's temperature resulted in faster cooling of the warmer RBC. During the photopolymerization (Phase II on Figure 12), both exothermic reaction and the released heat from the curing device increased the RBC's temperature. The extent of temperature increase, however, seems to be influenced by the speed of the temperature drop in Phase I. Accelerated drop may hinder the exothermic temperature increase.

Adequate monomer to polymer conversion is crucial to the material's long-term clinical success [3]. While DC is the key parameter determining the effectiveness of monomer conversion, unfortunately, it cannot describe the microstructure of the resulted heterogenic polymer network, which has a major effect on the physical and chemical properties of the RBC [12]. To determine the DC, micro-Raman spectroscopy was used in our study. It offers the possibility of quantitative characterization of the polymerization extent in dimethacrylate-based RBCs [125]. Raman-spectra were taken after 24 h, since a significant increase in DC takes place during the 24 h post-irradiation [159]. Although the minimum DC% for clinically acceptable restoration has not yet been precisely defined [58,63], DC values below 55% may be inadequate for occlusal restorative layers [143,146]. Musanje and Darvell recommended that the depth of cure should be defined as the depth at which maximum conversion occurs for a given irradiance and exposure time [117]. The radiant exposure was defined at 16–24 J/cm² to reach an adequate polymerization degree for a 2 mm thick RBC layer [54,135], meanwhile, the minimum radiant exposure required to be delivered to different bulk-fill RBCs moves on a wider scale (14–23–47 J/cm²) [43,81]. In our study, the valid, portable radiometer measured a higher value of radiant exitance than the average light output given by the manufacturer. It was demonstrated that most of the LCUs – especially low-budget LCUs, like our LED.D – could have different light output characteristics [7]. Providing by the curing unit, the delivered radiant exposure was 25 J/cm², and the DC% on the top of FOB_25 and FOB_55 were 63% and 64%, respectively, which is a characteristic value for a well polymerized RBC [117]. In comparison to the FOB values, the DC% on the top of the VCB_25 and VCB_65 samples were significantly lower, 54% and 55%, respectively. The lower DC values are presumably due to the material composition. The monomer system has a major effect on the DC, which increases in the following order: BisGMA < BisEMA < UDMA < TEGDMA [142]. BisGMA is considered to be the most viscous monomer due to the strong intramolecular hydrogen bonding, resulting in limited rotational freedom, thus the reactivity and mobility of the monomer may decrease during the

polymerization process [22]. This might be one of the explanations for the significantly lower DC of the VCB, which is a BisGMA-based RBC. FOB is an UDMA-based bulk-fill RBC, containing both aliphatic and aromatic UDMA. Sideridou et al. found that UDMA, combining relatively high molecular weight with a high concentration of double bonds and low viscosity, was shown to reach higher final DC% values than BisGMA [142]. Although the viscosity of UDMA is much lower than that of BisGMA, when it is mixed with the high molecular weight BisGMA or BisEMA, it can significantly restrict the mobility of UDMA monomers and decrease their reactivity and conversion value [9,82]. In addition to the monomer system, the filler-matrix ratio is also decisive. VCB filler loading is higher (83 wt%) compared to the filler content of FOB (76.5 wt%), which may restrict the light penetration and the mobility of monomers and radicals. The DC of VCB was investigated by Yang et al., and their results showed similar values (~58 DC%) in a 2 mm thick sample, exposed either with 20 s (24 J/cm²) or 40 s (48 J/cm²) [159]. Similar to the above study, pre-heating did not influence the DC% values of our investigated materials at the top of the samples, assuming that the RBCs on the top reached their maximum conversion degree already at room temperature. Although Daronch et al. found that the increased pre-cure temperature significantly improved the DC compared to the room temperature, they also concluded, that at longer exposure (20 s, 40 s) top-surface composite conversion was equivalent and similar throughout the tested temperature range (22–60 °C) [39].

Contrary to the values measured at the top of the room temperature samples the DC% at the bottom of the 4 mm thick bulk-fill materials were lower by ~10% (FOB_25, 51.8%; VCB_25, 46.2%). While the depth of cure is improved in bulk-fills due to increased translucency, modified matrix composition, photoinitiator kinetics, and filler characteristics, not all of the commercial bulk-fill RBC are able to sustain a homogeneous conversion at a depth of 4 mm [65,93,148,161]. The present research examined two, so-called full-body bulk-fill RBCs that are often referred to as paste-like bulk-fills. These materials generally have a higher filler load which makes them highly viscous and therefore sculptable. The higher filler content renders the surface more wear-resistant without requiring any coverage. Decreased DOC from the surface to the bottom may be a result of the increased filler ratio which may hinder light penetration due to the nano-sized particles despite the increase in translucency. On the other hand, high molecular weight monomers, such as BisGMA (in VCB) and aromatic UDMA (in FOB), also help

to increase the viscosity, however, decreasing the reactive groups in the resin may negatively influence the DC [21,22].

Pre-heating makes highly filled, sculptable RBCs more flowable, adaptable, and easier to manipulate, without compromising the superior mechanical properties. Decreased viscosity has been shown to enhance marginal adaptation and reduce microleakage due to improved wetting of cavity walls [62,154]. Although, the increased pre-cure temperature has benefits through decreased system viscosity, enhanced radical mobility, and collision frequency of unreacted active groups resulting in additional polymerization and higher conversion [49], the diversity of study outcomes may result from different RBC composition and experimental set-ups. Isothermal conditions mostly favor the positive effect of pre-heating on monomer conversion resulting in more highly cross-linked polymer networking and improved mechanical and physical properties [39]. However, improved monomer to polymer conversion has a strong relation to polymerization shrinkage which may increase the shrinkage stress of the bonded restoration [49,53,143]. Despite the higher shrinkage which might be present, it may not be clinically significant, as it can be offset by the improved marginal adaptation [49].

Clinically relevant, non-isothermal circumstances enhance the strong effect of RBC composition on the results. Several studies found the effect of pre-heating to vary on DC (decrease, no change, increase) depending on the composition of the investigated RBC [62,96,149].

Confirming the above findings, our results also showed a dissimilar effect of pre-heating on the monomer conversion of the investigated RBCs. An increase in pre-cure temperature did not influence significantly the DC on the top surfaces neither for FOB_55 nor for VCB_65 and even did not affect the bottom DC of VCB_65 compared to the room temperature RBCs. The bottom DC values showed a significant decrease however in the case of FOB_55. The rapid temperature drop of pre-heated RBC during handling results in excess heat loss which may deprive energy of the system and might prevent a sufficient increase in polymerization reactivity and consequent enhancement in monomer conversion [96]. Considering the findings of the temperature measurements, it is visible, that the temperature increase during polymerization shows a direct correlation with the measured DC values. During polymerization (Phase II. on Figure 12), the temperature of both VCB_25 and VCB_65 increased by 4.4 °C and showed similar DC values at the top (54.2 °C and 55 °C, respectively) and as well on the bottom surfaces (46.2 °C and 45.2 °C, respectively). The mean differences between top and bottom DCs were around 10%.

Meanwhile, the temperature within FOB_25 during polymerization rose by 5.8 °C and showed a significantly higher DC both on the top (63 °C) and bottom (51.8 °C), compared to VCB_25. The mean difference on top vs. bottom DC was found to be 10% also. In contrast, pre-heating of FOB had a negative effect both on the exothermic reaction and on the kinetics of monomer conversion. During light-curing, the temperature rise within FOB_55 was 1.8 °C lower (4 °C) than in the case of FOB_25, and the bottom DC was 20% less (45%) compared to the top DC value, which kept its higher level (64 °C). Although the drop of temperature during the dispensing and condensation phase (Phase I. on Figure 12) of VCB_65 was rapid, its temperature at the initiation of light-curing was higher, compared to the FOB_55. It may have provided enough energy to the polymerizing system, assuming that even higher exothermic temperature rise and higher DC would have been achieved if the system temperature dropped slower. In contrast, the temperature of FOB_55 at the beginning of light-curing was lower, and the additional drop during the cooling phase may have deprived energy from the system, resulting in a weaker exothermic reaction and lower monomer conversion. Since the reaction behavior of multifunctional monomer systems is very complex and highly dependent on the reaction conditions and composition, other possible explanations for DC decrease may arise. The increased pre-cure temperature may induce thermal polymerization before irradiation. On one hand, thermal polymerization leads to the consumption of functional groups, and on the other hand, pre-polymerization of few monomers results in shrinkage, which decreases the system's initial free volume and restricts the diffusion of monomers during the progression of the photopolymerization [10]. This phenomenon in VisCalor Bulk was investigated by Yang et al. who concluded, that pre-heating did not cause adverse effects through premature polymerization [159]. Other factors which may have been responsible for the observed temperature behavior of the FOB_55 could be further thermal side effects including evaporation of the reactants and thermal degradation of the photoinitiator [24]. In the case of VCB_65, 30 s pre-heating time was enough in the special dispenser, however, for FOB_55 pre-warming took more time in the Ena Heat Composite Heating Conditioner (15 min), which may influence the chemical condition of the components. Among the above-mentioned thermal side effects, a higher significance should be attributed to oxygen inhibition in the case of elevated RBC temperatures. As the temperature increases, the decrease in viscosity promotes oxygen penetration into the RBC. Oxygen reduces the extent of polymerization by scavenging on free radicals resulting in less reactive peroxy radicals and/or quenching of the excited

triplet state of the initiator [12]. It is reasonable to assume a role also for a further FOB constituent, the so-called AFM, an addition-fragmentation chain transfer dimethacrylate monomer, which participates readily in network formation by copolymerizing with multifunctional methacrylates [11]. An AFM is a heteroatom (N or S or O) containing monomer with various vinyl activating groups which have been employed as chain transfer agents to reduce shrinkage stress [10,126]. However, chain-transfer reactions may also exert a retarding effect on the polymerization by increasing termination, especially at higher temperatures [10].

Although pre-heating did not increase monomer conversion in many cases, several studies have shown that the mechanical properties and marginal integrity of RBCs (including FOB and VCB as well) are satisfactory or better than those applied at room temperature [1,62,102,118]. In contrast, however, there are experiments concluding higher linear shrinkage of pre-heated RBCs and deterioration in marginal integrity [49,155], although, results of the available investigations show that the pre-heating has no significant impact on bond strength of RBC to dentin [67,88].

As our results confirmed, RBCs do not have a complete monomer to polymer conversion because of the condition-dependent kinetics of gelation, vitrification, immobilization, and steric isolation [113]. Incomplete conversion may result in the presence of unreacted monomer content within the polymer network which is partially or completely released short- or long-term [8,12]. Released monomers may depress the biocompatibility of the RBC by stimulating bacterial growth around the restoration leading to secondary caries development and may promote allergic reactions. Additionally, cytotoxic effects of monomers have been demonstrated [64]. Solubility and water sorption can accelerate the degradation and do harm to the mechanical/physical properties such as tensile-, flexural strength and wear [144]. To determine the quality and quantity of the residual monomers eluted from the investigated polymerized materials HPLC, as a generally applied investigative method, was used in our study [93,111,152]. Unreacted monomers can reduce the mechanical properties of the RBCs and their detection represents an important step for evaluating RBC biocompatibility [60].

During our experiment, aromatic (BisGMA) and aliphatic (TEGDMA, UDMA, and DDMA) dimethacrylate standard monomers were used to identify eluted monomers from the investigated RBCs.

In the present study, 75% ethanol/water solvent was used to extract most of the examined unreacted monomers from the polymerized RBC specimens to identify monomer

quantity. 75% ethanol–water solution is recommended by the United States Federal Drug Administration as a clinically relevant food–oral simulating liquid [28]. Besides the type of the solvent, the chemical nature of the matrix monomers and their combination, the degree of conversion, and the final network characteristics also play important roles in the quantity and quality of monomer elution from a certain RBC [8,93].

According to the manufacturer’s description, VCB is a BisGMA/aliphatic dimethacrylate based RBC but does not define in detail the aliphatic dimethacrylates. However, following HPLC measurements, TEGDMA, DDMA, and UDMA were detected as eluted aliphatic monomers from VCB. FOB is mainly an UDMA-based material, composed of both aliphatic and – as a BisGMA substitute – aromatic UDMA. However, released BisGMA was also detected during the HPLC measurements. Copolymers consisting of BisGMA and/or UDMA are crosslinked both chemically (C-C covalent bond) and physically (i.e., hydrogen bond). The latter determines the matrix viscosity, since the more numerous and stronger the hydrogen bonds are, the higher the viscosity of the system [22]. To create a sculptable RBC, the usage of these monomers is advantageous. Regarding the monomer release, our results showed elution to be strongly dependent on the material.

In this study, significantly (two-fold) more UDMA and DDMA were released from the room temperature FOB_25, meanwhile, VCB_25 samples leached almost three-fold more BisGMA. Pre-heating significantly decreased the monomer elution from FOB_55. There was no difference however in monomer elution between VCB_25 and VCB_65. The measured unreacted monomer release is in line with our results regarding the degree of monomer conversion in VCB_25 and VCB_65 since pre-heating did not change the DC on the top or bottom of VCB. On the other hand, the observed relationship between DC and monomer elution from FOB is contradictory. While the DC of the bottom surface decreased after pre-heating, the detected elution of unreacted monomers from FOB_55 samples was also lower. Although several studies have shown that the extent of leached unreacted monomer is correlated to the DC [52,94,141], the conversion degree does not necessarily correlate with the amount of free residual monomer, since the detected double bonds may remain as pendant groups bonded to the polymer structure and are not free to be released, however, may reduce the clinical success of the RBCs [22,56]. Probably, the above issue is the explanation for the lack of the expected relationship between the DC and monomer elution in the case of the pre-heated FOB_55.

While the number of monomer elution studies from bulk-fill RBCs is extensive, data regarding the effect of pre-heating on monomer release both from conventional and bulk-fill RBCs is limited in the literature, hence, the discussion of this issue and comparison to other results are also restricted. Elution from bulk-fills was found to be comparable to that of conventional RBCs despite their increased increment thickness [8,71]. The quality and quantity of released resins are strongly material dependent and the amount of most of the eluted monomers is increased with time [28,93]. The monomer detected to be eluted in the highest amount was BisGMA from both VCB_25 and VCB_65, with the latter showing a significantly lower quantity. As it was previously mentioned, the extremely high viscosity of BisGMA limits the DC, leaving behind more unreacted monomers, which may release into the oral cavity. Admixing low molecular weight monomers, such as TEGDMA and DDMA, to BisGMA, can lower its viscosity, and via their synergistic effect can increase the rate of polymerization [128]. The released quantity of the latter two was very small both from VCB_25 and VCB_65. FOB, on the other hand, is a UDMA-based RBC. At present, UDMA is the only commercial alternative to the bisphenol A-based dental methacrylates [22]. Although, UDMA viscosity is lower than BisGMA, still high enough to require the addition of a reactive diluent, such as DDMA. Due to UDMA's lower molecular weight in comparison to BisGMA, it is expected to show higher DC and lower unreacted monomer elution [140]. However, aside from aliphatic UDMA, FOB contains aromatic UDMA as well. Aromatic moiety and substitution symmetry play an important role in the steric hindrance, methacrylate group separation, limited conformational freedom, and increase of molecular stiffness. However, closer proximity of reactive groups facilitates the reaction-diffusion, which may lead to moderate DC and the planar geometry of benzene rings allows for building tighter structures [22]. Based on our results, the aliphatic UDMA release was moderate from FOB_25 and significantly lower from FOB_55, as a result of pre-heating.

However, in our study, neither eluted AUDMA nor AFM was detected in the absence of the standards, as their exact chemical structure is a trade secret.

To the best of our knowledge, only one published article deals with the monomer elution from three pre-heated RBCs and found no effect of pre-cure temperature (68 °C) on the amount of leached UDMA, TEGDMA, and BisGMA [30]. Few available pieces of research reported that pre-heating of both conventional and bulk-fill RBCs did not influence cell viability, however, polymerized samples were used to examine the cytotoxicity without determination of the eluted monomers [29,49,128]. de Castro et al.

investigated the sorption and solubility of RBC at higher pre-cure temperatures (60 °C) and concluded that longer curing times and higher temperatures led to lower values of sorption and solubility, but these differences were only significant for specific combinations of temperatures and curing times [46].

The main limitation of this study may be the *in vitro* nature of the investigation. Although during sample preparation a conscious effort was made to simulate an isolated tooth by adjusting the PTFE mold temperature, the thermal conductivity of a natural tooth, its position in the oral cavity, the cavity configuration, thus the contact surfaces with the RBC are just a few mentioned factors, which may influence the results *in vivo*. Furthermore, the analysis of the elution of selected unreacted monomers (BisGMA, UDMA, TEGDMA, DDMA) will not provide an absolute measure of the quality of released components, since, among others, various monomers, like AUDMA, AFM, degraded compounds, initiator molecules, and fillers may also leach and compromise the RBC biocompatibility. A further limitation may be the limited number of the investigated high-viscosity bulk-fill RBCs, especially considering the strongly material-dependent results. The results cannot be extrapolated to other room temperature and pre-heated RBCs, since the composition has a strong influence on both DC and monomer elution and can vary from RBC to RBC.

V.2. Investigation of the effect of ceramic and dentin thicknesses and the type of resin-based luting agents on intrapulpal temperature changes during the luting of ceramic inlays

To discover the thermal change in the pulp chamber using pre-heated restorative RBCs or light-cured and dual-cured adhesive resin cements a single tooth model was applied using different thicknesses of dentin and ceramic blocks imitating inlays. The results showed that in the case of investigated ceramic–dentin combinations, the cementing agents increased the intrapulpal temperature above the considered critical 5.5 °C. Furthermore, a qualitative comparison of thermal conductivity and capacity showed differences between dentin and ceramic. Therefore, all the tested null hypotheses were rejected. Our findings are consistent with the results of other studies regarding the shielding effect of the interposed ceramic and the remaining dentin thickness on pulpal temperature rise and the material-dependent temperature-increasing effect of the

polymerization of RBCs [20,90,91]. Thus, even though the interposed ceramic inlay and the remaining dentin thickness attenuate the light intensity of the curing unit and the delivered energy during the polymerization process, the exothermic temperature rise associated with the adhesive luting agent may jeopardize pulp health.

To eliminate any effects that may arise from structural differences and thermal properties of the tooth structure, this study was carried out on a representative permanent third molar without the use of acid conditioning of tooth and any ceramic surface treatment for all experimental groups. This provided the same tooth conditions for each measurement. However, the tooth differences regarding thermal changes are not accounted for in this model. Furthermore, this technique has limitations, as heat dissipation by pulpal, periodontal and osseous circulation is not reproduced [4,165]. Thus, the temperature changes measured in this study cannot be directly applied under *in vivo* conditions. Despite the absence of blood circulation in vital tissues, this study provides important information regarding the magnitude of temperature change in a model system when a ceramic inlay is cemented in cavities of different depths using different adhesive luting agents.

In this study, a thermocouple was used for the instantaneous observation of temperature changes during ceramic inlay cementation under laboratory conditions. The use of a thermocouple is a simple and well-known method for measuring differences in temperature during dental treatments [20,91]. Compared to more modern methods, such as infrared thermography, thermocouples can measure similar values with differences of less than 1 °C in favor of infrared measurements [23].

It has been reported that dental pulp is vulnerable to temperature changes despite its high vascularization, which is the main regulatory system for heat distribution and which is capable of dissipating external thermal stimuli transferred to the dentin–pulp complex [55]. The clinical relevance of increased intrapulpal temperature is that it is a potential risk factor for thermal pulp damage. According to Zach and Cohen, a 5.5 °C temperature rise is critical and may cause irreversible pulpal damage [164]. In line with the previous experiment, Pohto and Scheinin reported that the critical temperature for reversible pulp damage was between 42 °C and 42.5 °C [130]. Although the pathological threshold of pulpal temperature rise was determined to be approximately 5.5 °C, a more recent study did not find an average increase of 11.2 °C to compromise pulp health significantly [19]. However, a histomorphometric analysis using a clinically valid 3-dimensional organotypic *ex vivo* model showed an immediate reduction in cell number with a

temperature increase of 5.5 °C or greater, which was dependent on exposure time [106]. Additionally, immunohistochemical changes were observed at a temperature increase of 6 °C or higher [106].

Despite attempts to simulate *in vivo* conditions using several reliable methods, a wide range of intrapulpal temperature increases has been reported *in vitro* during photocuring [134]. To provide the blue light required for the polymerization of resin-based luting agents, a second-generation LED LCU was used in this study with a radiant exitance of $1550 \pm 10 \text{ mW/cm}^2$ in the wavelength range of 420–480 nm. According to previous studies, the intensity and duration of the applied light were the most crucial factors for the pulpal temperature rise [97,166]. The results of this study confirm the above statement because light curing without the interposition of a ceramic inlay and resin-based luting agent increased the pulpal temperature ($\Delta T = 4.0\text{--}9.9 \text{ }^\circ\text{C}$) by a significant degree, depending on the dentin thickness, with an inverse correlation. Although a thin layer of dentin adhesive was used during light curing without ceramic blocks and luting agents, the heat measured was predominantly from the curing unit. A pilot study was undertaken to assess the thermal effect of the dentin adhesive layer, and a negligible, insignificant (0.1–0.2 °C) temperature difference was detected during light curing with or without the adhesive.

Because the monomer-to-polymer conversion of an RBC is a function of the applied total energy during photocuring, it is advisable to increase the delivered radiant exposure for a higher degree of conversion [72]. This is highly relevant to adhesive luting of indirect restorations, where a high radiant exposure is needed for proper curing of the luting resin-based material underneath a certain thickness of ceramic [132]. The reciprocal relationship between the power output and exposure duration provides an opportunity to increase either irradiance or exposure time, resulting in a higher delivered radiant exposure [72]. It was affirmed that extended irradiation has a greater effect on the depth of cure than increasing the light irradiance of the curing unit [45]. While the increased delivered radiant exposure is indispensable to the acceptable polymerization of the adhesive luting agent, a strong positive correlation was found between the radiant exposure and the intrapulpal temperature rise [91,153]. In this study, an extended exposure time (40 s) with a light irradiance of 1550 mW/cm^2 resulted in 62 J/cm^2 of delivered radiant exposure. Onisor et al. [121] investigated the effect of extended exposure times on intrapulpal temperature increase through 1 mm of the remaining dentin thickness, interposing a 3 mm ceramic onlay and 0.3 mm of previously polymerized RBC

luting agent. They found a maximal temperature difference of 4.3 °C during the 3 × 20 s of extended irradiation, delivering ~60 J/cm² of total energy. Although the study design and the type of curing units are slightly different, these results are in line with our findings with a similar D1–C3.5 combination, where the intrapulpal temperature rise was 4.1 °C without the exothermic reaction of the luting agent and the delivered energy density was 62 J/cm². In addition to energy density, other characteristics of the photocuring unit affect the amount of heat generated within the pulp [4,115,121]. While the pulpal temperature in this study remained below the critical value of 5.5 °C without luting agents, the tested adhesive cements significantly increased it because of their exothermic reaction. Our study design ensured uniform conditions, allowing the comparison of exothermic thermal changes between different resin-based luting agents. The exothermic reaction was proportional to the amount of resin matrix, and it was found that the inorganic fillers have an impact on heat diffusion within the material by their capacity to absorb external and internal energy [17,91,156].

The Multivariate General Linear Model revealed that the material factor had a medium impact on the pulpal temperature rise. A tendency was observed when comparing the effects of the tested luting agents on temperature change, although there was no detectable statistically significant difference among the study groups. The highest temperature was measured with the pre-heated RBC in all the tested groups, followed by the light-cured adhesive cement, and the lowest temperature increase was observed with the dual-cured luting agent. Although the restorative RBC was pre-heated to 55 °C, it only increased the pre-polymerization temperature of the pulp by ~3 °C, and further cooling was detected during the cementation procedure before light curing. After removal from the warming device, the pre-heated RBC was reported to suffer from rapid cooling, which may compromise the degree of conversion [84]. Although the temperature-raising effect was less than expected owing to the rapid cooling and heat absorption by the ceramic and dentin, these results suggest the potential hazard to pulp health owing to their higher thermal effect. Regarding the composition of the investigated light- and dual-cured resin cements, they had the same resin matrix/filler ratio; however, the dual-cured cement showed a slightly, but not significantly, lower temperature rise in each tested group compared to that of the light-cured resin cement. Dual-cured resin cements are supposed to compensate for decreased light transmission and may be more efficient at monomer-to-polymer conversion, even with increased ceramic thicknesses [15,127]. In their systematic review, David-Pérez et al. found that dual-cured resin cements fail to achieve

the same degree of conversion as light-cured cements with up to 2 mm of interposed ceramic thickness [44]. Our results are indirectly in line with these findings because the polymerization process is proportional to the exothermic reaction, which results in a slightly lower pulpal temperature rise in the dual-cured resin cement [17].

Thermal transfer to the pulp is strongly dependent on the thickness of the remaining tooth structure [90]. The thermal conductivities of enamel and dentin are ~ 0.81 W/mK and ~ 0.48 W/mK, respectively, which are considered low [119]. Low thermal conductivity is equivalent to high insulating capability; thus, the pulp is protected from noxious thermal irritation if the tooth is intact [119]. The advantageous thermal conductivity may be explained by the porous tubular microstructure in the dentin layer, which is a mineralized connective tissue with an organic matrix of collagenous proteins [100]. However, during cavity preparation for direct or indirect restorations, the enamel and DEJ are partially removed, and the dentin thickness is reduced according to the extent of the caries or depending on the special cavity design. Hard tissue removal during cavity preparation and several steps of the adhesive restorative procedure (i.e., polymerization of the adhesive layer and the RBC/adhesive luting agent, and the polishing procedure) may cause thermal damage due to the weakened thermal insulation effect, especially in younger patients with wider dentinal tubules [92,101]. The present study investigated four thicknesses (1.0 mm, 1.5 mm, 2.0 mm, and 2.5 mm) of the remaining dentin layer. For dentin thickness below 2.0 mm, the heat transmitted by the curing unit increased the intrapulpal temperature above the 5.5 °C limit. These results are consistent with previous finding that showed a strong relationship between the thickness of the dentin and the intrapulpal temperature increase [163]. Similar to our findings, Kuo et al. concluded that there is a risk of damaging the pulp when the dentin thickness is less than 2.0 mm and the overall thickness of the dentin–ceramic assembly is less than 3.5 mm [90]. However, according to other studies, the dentin thickness (0.5 vs. 1.0 mm) had an insignificant role in the pulpal temperature increase compared to the curing unit type [70]. In addition to the dentin insulating effect, the already polymerized thin dentin adhesive may serve as a further protective layer during the cementing procedure. However, our pilot study showed that the thermal insulating effect of dentin–ceramic assemblies is not affected by the presence or absence of a polymerized adhesive layer. In the present study, Partial Eta-Squared statistics revealed that, among the investigated factors—such as dentin thickness, ceramic thickness, and the type of luting material—the remaining dentin layer had the most pronounced effect on the pulpal temperature change values, and the effect size was

considered to be large. The Linear Regression Model revealed a 47% value for the coefficient of determination and predicted a decreasing effect of thickness on temperature as the dentin thickness exceeded 2 mm. Even though the thinnest dentin was combined with the thickest ceramic, the highest intrapulpal temperature was detected in all measurements, regardless of the use of the luting agents.

In contrast, the effect of ceramic thickness on pulpal temperature rise was considered insignificant according to the results of the General Linear Model, although the Partial Eta-Squared indicated a medium effect. This result demonstrates that the shielding effect of the ceramic is not as great as that of dentin, although an inverse relationship between the ceramic thickness and temperature rise is evident, which is consistent with the findings of a previous study [90]. The regression curve fit for temperature as a function of different ceramic thicknesses showed a linear energy loss with increasing ceramic thickness, which reflects the light attenuation occurring through an absorptive/scattering medium. The value of the coefficient of determination for ceramic thickness was found to be 41% in our Linear Regression Model. The differences in temperature changes caused by the thermal shielding effect of dentin and ceramic can be explained by their distinct thermal conductivities. It is higher for silica-based ceramics, which is approximately 1.7 W/mK, compared to the thermal conductivity of the dentin (~0.48 W/mK) [147]. Increasing the inlay thickness may result in a proportional removal of tooth hard tissues by decreasing the thickness of the remaining enamel and dentin. Although the shielding effect is proportional to increasing ceramic thickness, a decreasing dentin thickness has a stronger inversely proportional effect on the temperature increase within the pulp chamber. These results are supported by the DSC measurements in this study, which revealed a 24% lower thermal conductivity of the dentin than that of the ceramic. However, the thermal capacity of the dentin was calculated to be 86% more compared to the investigated lithium disilicate ceramic. The importance of this result lies in the ability of dentin to store large amounts of heat and then dissipate it slowly, reducing the sudden thermal effects on the pulp. However, considering the multiple thermal effects during adhesive restorative treatment, the gradually increasing temperature of the dentin may conduct more heat towards the pulp during heat dissipation. According to these findings, it is advisable that more dentin should be preserved during cavity preparation to protect the pulp from undesirable temperature increases. Furthermore, it should provide more time for heat dissipation between treatment steps, which can have a thermal effect on the pulp, to avoid heat accumulation in the dentin.

VI. Conclusion of novel results

VI.1. Investigation of the effect of pre-heating on monomer elution and degree of conversion of contemporary and thermoviscous bulk-fill resin-based dental composites

Within the limitations of this *in vitro* study, the following conclusions can be stated:

- Significantly higher DC values were achieved on the top of the room temperature and pre-heated investigated bulk-fill RBCs than on the bottom.
- Room temperature VisCalor Bulk has lower DC% values both on the top and bottom compared to Filtek One Bulk Fill Restorative.
- Pre-heating did not influence the DC of VisCalor Bulk, however, significantly decreased the DC at the bottom of Filtek One Bulk.
- Pre-heating had no effect on the monomer elution from VisCalor Bulk, but significantly decreased the monomer release from Filtek One Bulk.
- *Material* factor had a significant effect on each investigated variable, while *Temperature* factor and its interaction with *Material* is surface- (top vs. bottom) and monomer-dependent.

Clinical significance:

While pre-heating had no beneficial effect on the degree of conversion neither of the thermoviscous VisCalor Bulk nor the contemporary bulk-fill RBC (Filtek One Bulk Fill Restorative) the increased pre-cure temperature may decrease the elution of unreacted monomers from the RBCs.

VI.2. Investigation of the effect of ceramic and dentin thicknesses and the type of resin-based luting agents on intrapulpal temperature changes during the luting of ceramic inlays

Within the limitations of this *ex vivo* study, the following statements can be declared:

- The intrapulpal temperature rise may exceed the critical 5.5 °C threshold during ceramic inlay cementation, regardless of the dentin and ceramic thicknesses and the type of resin-based adhesive luting material used.

- The temperature values were predominantly influenced by the remaining dentin thickness, followed by the applied resin-based adhesive luting materials, and were least influenced by the ceramic thickness.
- The thermal conductivity of dentin was 24% less compared to the ceramic, while the thermal capacity was 86% higher.

Clinical significance:

In order to provide safe dental treatment, a more conservative approach with more dentin preservation should be followed when it comes to the thermal effect applied to teeth during ceramic inlay cementations.

VII. References

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VIII. Publications

VIII.1. Publications related to the topic of the Ph.D. dissertation

Kincses D, Böddi K, Óri Z, Lovász BV, Jeges S, Szalma J, Kunsági-Máté S, Lempel E. Pre-Heating Effect on Monomer Elution and Degree of Conversion of Contemporary and Thermoviscous Bulk-Fill Resin-Based Dental Composites. *Polymers* **2021**, 13, 3599. <https://doi.org/10.3390/polym13203599>

Q1; *IF*₂₀₂₁: **4.967** *Citations*: 20 (*independent*: 17, *dependent*: 3)

Kincses D, Jordáki D, Szebeni D, Kunsági-Máté S, Szalma J, Lempel E. Effect of Ceramic and Dentin Thicknesses and Type of Resin-Based Luting Agents on Intrapulpal Temperature Changes during Luting of Ceramic Inlays. *Int J Mol Sci* **2023**, 24,5466. <https://doi.org/10.3390/ijms24065466>

Q1; *IF*₂₀₂₃: **5.6** *Citations*: 1 (*dependent*: 1)

Cumulative impact factor of dissertation related publications: **10.567**

VIII.2. Publications independent from the topic of the Ph.D. dissertation

Lempel E, Óri Z, **Kincses D**, Lovász BV, Kunsági-Máté S, Szalma J. Degree of conversion and in vitro temperature rise of pulp chamber during polymerization of flowable and sculptable conventional, bulk-fill and short-fibre reinforced resin composites. *Dent Mater* **2021** Jun;37(6):983-997. doi: 10.1016/j.dental.2021.02.013. Epub 2021 Mar 10. PMID: 33714623.

Q1; *IF*₂₀₂₁: **5.687** *Citations*: 29 (*independent*: 23, *dependent*: 6)

Lempel E, **Kincses D**, Szebeni D, Jordáki D, Lovász BV, Szalma J. Intrapulpal temperature changes during the cementation of ceramic veneers. *Sci Res* **2022** Jul 28;12(1):12919. doi: 10.1038/s41598-022-17285-x.

Q1; *IF*₂₀₂₂: **4.6** *Citations*: 3 (*independent*: 2, *dependent*: 1)

Cumulative impact factor of all publications: **20.854**

VIII.3. Conference poster presentations related to the Ph.D. dissertation

Kincses D, Böddi K, Óri Z, Lovász BV, Jeges S, Szalma J, Kunsági-Máté S, Lempel E. Pre-Heating Effect on Monomer Elution and Degree of Conversion of Contemporary and Thermoviscous Bulk-Fill Resin-Based Dental Composites. Hungarian Association of Aesthetic and Restorative Dentistry (HAARD) Annual Conference, Budapest, **2022**. - Original research poster presentation

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Article

Pre-Heating Effect on Monomer Elution and Degree of Conversion of Contemporary and Thermoviscous Bulk-Fill Resin-Based Dental Composites

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Abstract: Detection of unreacted monomers from pre-heated resin-based dental composites (RBC) is not a well-investigated topic so far. The objectives were to determine the temperature changes during the application and polymerization, the degree of conversion (DC) and unreacted monomer elution of room temperature (RT), and pre-heated thermoviscous [VisCalor Bulk(VCB)] and high-viscosity full-body contemporary [Filtek One Bulk(FOB)] bulk-fill RBCs. The RBCs' temperatures during the sample preparation were recorded with a K-type thermocouple. The DC at the top and bottom was measured with micro-Raman spectroscopy and the amounts of eluted BisGMA, UDMA, DDMA, and TEGDMA were assessed with High-Performance Liquid Chromatography. The temperatures of the pre-heated RBCs decreased rapidly during the manipulation phase. The temperature rise during photopolymerization reflects the bottom DCs. The differences in DC% between the top and the bottom were significant. RT VCB had a lower DC% compared to FOB. Pre-heating did not influence the DC, except on the bottom surface of FOB where a significant decrease was measured. Pre-heating significantly decreased the elution of BisGMA, UDMA, DDMA in the case of FOB, meanwhile, it had no effect on monomer release from VCB, except TEGDMA, which elution was decreased. In comparison, RBC composition had a stronger influence on DC and monomer elution, than pre-cure temperature.

Keywords: bulk-fill; pre-heating; degree of conversion; monomer elution

1. Introduction

During the restorative procedure, it is favorable to use an easy-to-handle, non-technique sensitive, durable and esthetic restorative material with quick and efficient polymerization. In the case of resin-based composite (RBC) restorations, expediting polymerization, increasing of maximum layer thickness, and the degree of monomer conversion can be considered as the main objectives [1]. To achieve a durable, successful composite restoration, the most important factors, among others, are mechanical properties, handling characteristics, polymerization stress, marginal adaptation, and degree of polymerization [2].

According to the literature data, there is a clear correlation between the degree of conversion (DC) and the physicochemical characteristics of RBCs [3,4]. Meanwhile, DC

is influenced by several factors, such as light exposure conditions, composition, shade, opacity, and thickness of the RBC, pre-cure temperature also plays an important role in the polymerization process [5,6]. As the success of RBC restorations depends on their polymerization and DC, the influence of temperature has become one of the central issues of several studies [7,8].

The polymerization of RBCs is an exothermic reaction and the released heat is mainly produced in the propagation phase [9]. The process of monomer conversion and the properties of the set polymer are influenced by the polymerization temperature [10]. An elevation in temperature promotes molecular mobility and increases the collision frequency of reactive radicals, resulting in higher conversion and delayed auto deceleration [6,9]. Not only the exothermic reaction but also the light energy absorbed contributes to the system temperature during the polymerization of light-cured RBCs [11]. While some authors attribute a greater significance to the heat emitted by the light-curing unit in the temperature rise, others regard the heat generated in the exothermic reaction as more important [12,13].

As reported by several investigations, pre-heating may have a beneficial impact on marginal adaptation, gap formation, and microleakage by reducing the viscosity of RBCs [7,14,15]. Improved handling properties, such as flowability can facilitate the application of the filling material, consequently making the procedure less time-consuming. Reduced viscosity also improves marginal seal thereby contributing further to the overall clinical success [16,17]. There may be a lack of efficiency associated with the use of conventional heating devices as some authors have shown a rapid decrease in RBC temperature after removal of the device, as well as during dispensing and handling [2,18]. Moreover, during the cooling phase, the system bears a loss of energy, so vitrification takes place earlier and causes decreased DC [19].

Several types of RBC dental materials have been developed over the years, including bulk-fill RBCs, which can be placed in larger increments to reduce operating time and technique sensitivity [20]. The primary advantage of bulk-fill RBCs over conventional ones is the increased depth of cure [21,22]. According to a literature review by Van Ende et al., the maximum layer thickness which still ensures adequate material characteristics as recommended by the manufacturers, is 4 mm or in some cases even 5 mm. Although most studies have confirmed the improved depth of cure for bulk-fill RBCs, some controversial data can still be found [23,24]. Besides the DC, polymerization shrinkage stress is another important issue addressing the clinical failures. Ausiello et al. demonstrated, that bulk-filling, especially in deep cavities induces higher shrinkage stress along the cavity walls compared to a multilayer technique (i.e., no shrinking glass-ionomer basing and shrinking bulk-fill RBC cover) [25].

Recently, a thermoviscous bulk-fill RBC (VisCalor Bulk) and a new heating device (VisCalor Dispenser) were introduced to combine the advantages of bulk-fills and pre-heating. This delivery system can warm up the filling material in seconds using near-infrared technology and allows immediate application without removal of the capsule from the heating device thereby maintaining its increased temperature [26]. Although VisCalor Bulk is a relatively new RBC, it has already been the subject of several investigations [27–31]. Yang et al. examined the effects of temperature on stickiness and packability, and the effect of pre-heating time on pre-cure properties. The study found pre-heating to lead to a reduced extrusion force and increased flowability without premature polymerization, while stickiness and packability remained within a clinically acceptable range [27].

The effect of pre-heating and exposure duration on other properties of VisCalor Bulk has been investigated in another study undertaken by Yang et al. This article reported a longer exposure duration not to have an effect on the degree of conversion, maximum rate of polymerization and polymerization shrinkage, however, it did lead to an increase top surface microhardness. The application of 3 min pre-heating and 20 s irradiation provided adequate hardness without unfavorable changes in polymerization shrinkage strain and polymerization kinetics [28]. The study of Marcondes et al. examined viscosity and thermal

kinetics of pre-heated RBCs including VisCalor Bulk as well as the effect of ultrasound energy on film thickness. VisCalor Bulk showed the greatest extent of viscosity reduction at 69 °C, while film thickness could not be reduced below 50 µm without the use of ultrasound. This study also claimed that to take full advantage of the pre-heated RBCs, the ideal working time is merely 10–15 s [29]. Demirel et al. investigated the effect of different insertion techniques on the internal void formation and found VisCalor Bulk to show the lowest void percentage with the utilization of the pre-heating technique [30]. Colombo et al. evaluated microhardness and depth of cure of four bulk-fill RBCs. According to the measured ratio of top to bottom hardness, all tested materials—including VisCalor Bulk—showed an adequate degree of polymerization. In addition, in the case of VisCalor Bulk, acid storage led to one of the highest mean percentage losses in micro-hardness of the external side [31].

Besides the physicochemical properties, the chemical characteristics are also important determinants of the clinical performance and biocompatibility of an RBC [32]. Although there is a strong inverse correlation between DC and monomer elution, the number of released monomers may be influenced further by other factors such as the quality of the monomer system, filler type, content, porosity as well as employed solvent [24,33]. Elution from bulk-fill RBCs was found to be comparable to that of conventional materials despite their increased increment thickness as monomer release is more dependent on the hydrophobicity of the base monomers and the final network characteristics of the resin-matrix [34]. Detection of unreacted monomers from pre-heated RBCs is not a well-investigated topic so far. To the best of our knowledge, the current literature has no information regarding the amount of eluted monomers from thermoviscous VisCalor Bulk.

Therefore, the purpose of the present study was to investigate the temperature changes during the application and polymerization of a new thermoviscous (VisCalor Bulk) and a high-viscosity full-body bulk-fill RBC (Filtek One Bulk-fill) in relation to different pre-cure temperatures. Further aims were to evaluate the effect of pre-heating on the degree of conversion (DC) and the number of released monomers using micro-Raman spectroscopy and Reversed-phase High-Performance Liquid Chromatography (RP-HPLC). The Null Hypotheses were: (1) Pre-heating had no effect on RBCs' post-cure DC%, and (2) pre-cure temperature did not affect the amount of released unreacted monomers.

2. Materials and Methods

2.1. The Bulk-Fill Resin-Based Composites and Sample Preparation

During this *in vitro* study two brands of high-viscosity bulk-fill RBCs (Viscalor Bulk and Filtek One Bulk-fill)—A2 shade for both—were investigated in 4 mm layer thickness. The specifications of the materials and their acronym codes are presented in Table 1.

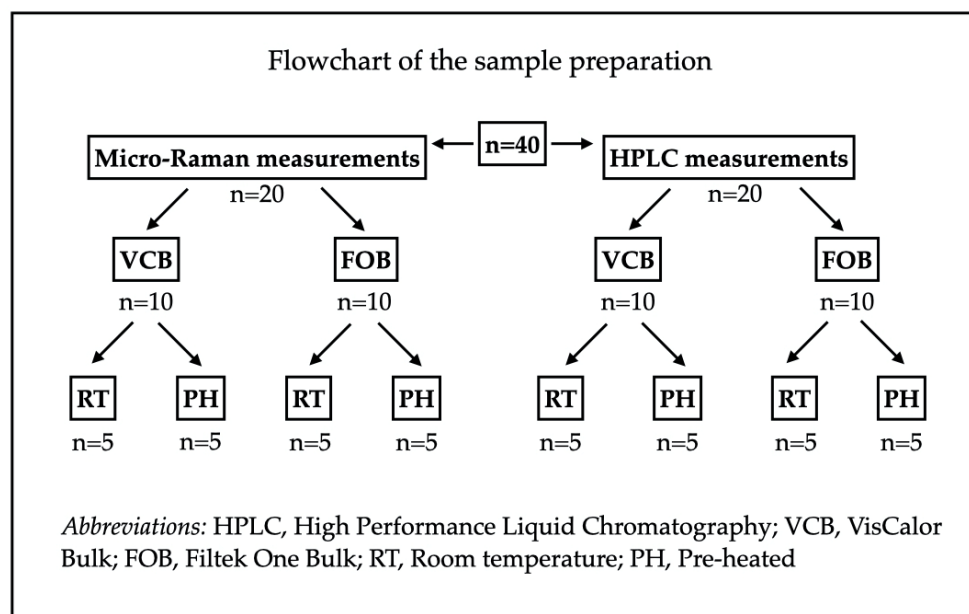
Table 1. Materials, manufactures, composition, and investigated pre-cure temperature of bulk-fill resin-based composites.

Material	Manufacturer	Pre-Cure Temperature	Code	Resin System	Filler	Filler Loading
VisCalor Bulk	Voco, Cuxhaven, Germany	Room temperature	VCB_RT	Bis-GMA, aliphatic dimethacrylates	Inorganic nanohybrid filler (not defined by the manufacturer)	83 wt%
		Pre-heated to 65 °C	VCB_65			
Filtek One Bulk-fill Restorative	3M ESPE, St. Paul, MN, USA	Room temperature	FOB_RT	AFM, UDMA, AUDMA, DDMA	20 nm silica, 4–11 nm zirconia, cluster Zr-silica, 0.1 µm ytterbium-trifluoride	58.5 vol% 76.5 wt%
		Pre-heated to 55 °C	FOB_55			

Abbreviations: BisGMA: bisphenol-A diglycidil ether dimethacrylate; AFM: addition-fragmentation monomer; UDMA: urethane dimethacrylate; AUDMA: aromatic urethane dimethacrylate; DDMA: 1,12-dodecane dimethacrylate; vol%: volume%; wt%: weight%.

According to the method of sample preparation, there were two experimental groups for each of the two investigated materials. The pre-cure temperature of the RBC samples in

the first group was 25 °C (room temperature—RT) (FOB_RT and VCB_RT), while RBCs in the second group were preheated before the sample preparation. In the case of VCB, pre-heating was performed by VisCalor Dispenser (VOCO, Cuxhaven, Germany) using T1 setting (VCB_65) (30 s pre-which warmed the device and RBC together to 65 °C). Pre-warming of FOB was undertaken by Ena Heat Composite Heating Conditioner (Micerium, Avegno, Italy) using T2 setting (FOB_55) (55 min pre-warming of the device to 55 °C and 15 min pre-warming of the RBC). Five specimens were prepared in each group, from each material for both the micro-Raman spectroscopy measurements as well as for the monomer elution measurements (Scheme 1).



Scheme 1. Flowchart of the sample preparation.

The samples were prepared in a cylindrical polytetrafluoroethylene (PTFE) mold with an internal diameter of 5 mm and a height of 4 mm, placed on a thermostatically controlled (30 ± 1 °C) glass slide to represent the isolated tooth. A polyester Mylar strip was positioned between the mold and the glass slide. A capsule dispenser gun was used to apply VCB_RT, FOB_RT, and FOB_55 materials into the mold. In the case of VCB_65, a VisCalor dispenser was used for both warming and application. The condensation of the RBCs was performed with a room temperature hand instrument. Before irradiation, the RBC sample was covered with a transparent polyester strip (Mylar, Dentamerica Inc., San Jose Ave, CA, USA) to avoid contact with oxygen. All specimens were irradiated with a Light Emitting Diode (LED) curing unit (LED.D, Woodpecker, Guilin, China; average light output given by the manufacturer $850\text{--}1000$ mW/cm²; $\lambda = 420\text{--}480$ nm; 8 mm exit diameter fiberglass light guide) in standard mode for 20 s, powered by a line cord at room temperature of 25 °C ± 1 °C, controlled by an air conditioner. The irradiance of the LED unit was monitored before and after polymerization with a radiometer (CheckMARC, Bluelight Analytics, Halifax, NS, Canada). The tip of the fiberglass light guide was in direct contact, centrally positioned, and parallel to the mold. All the samples were prepared by one operator.

2.2. Temperature Measurement

Temperature measurements during the application and the polymerization of RBCs' were recorded with a registration device (El-EnviroPad-TC, Lascar Electronics Ltd., Salisbury, UK) attached to 0.5 mm diameter Cu/CuNi thermocouple probes (K-type, TC Direct, Budapest, Hungary)—positioned at the bottom of the temperature regulated mold—with a frequency of one measurement per second and resolution of 0.1 °C (Figure 1).

schematic component). The quantity of heat emitted by the LED curing unit was also determined through the 4 mm empty mold.

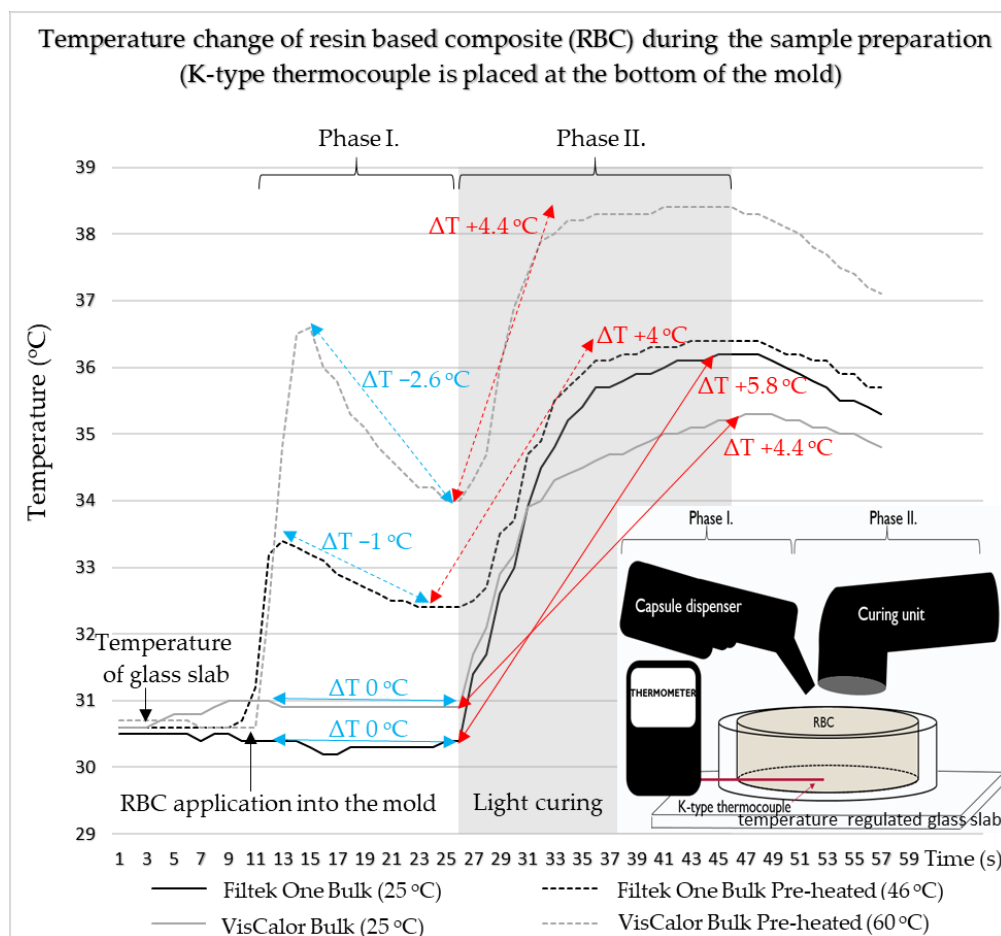


Figure 1. Temperature change of resin-based composite during the sample preparation.

2.3. Micro-Raman Spectroscopy Measurement

RBC samples made to estimate the temperature change of the composite during sample preparation were then used to measure the DC. Confocal Raman spectrometer (Labram HR 800, HORIBA Jobin Yvon S.A.S., Longjumeau Cedex, France) was used to evaluate the 24 h post-cure DC values of the polymerized RBC samples. Setting parameters for the measurements were the following: 20 mW He-Ne laser with 632.817 nm wavelength, magnification $\times 100$ (Olympus UK Ltd., London, UK), spatial resolution $\sim 15 \mu\text{m}$. The spectral resolution of $\sim 2.5 \text{ cm}^{-1}$ provided satisfactory results since the two peaks analyzed were $\sim 30 \text{ cm}^{-1}$ apart. Spectra were taken at three locations of the RBC samples (center, periphery, and between these two regions) both from the bottom and top surfaces with an integration time of 10 s. Ten acquisitions were averaged for each geometrical point. Spectra of uncured RBCs were taken as reference. Post-processing and analysis of spectra were performed using the dedicated software LabSpec 5.0 (HORIBA Jobin Yvon S.A.S., Longjumeau Cedex, France) [13]. The ratio of double-bond content of monomer to polymer in the RBC was calculated according to the following equation:

$$DC\% = (1 - (R_{\text{cured}}/R_{\text{uncured}})) \times 100$$

where R is the ratio of peak intensities at 1639 cm^{-1} and 1609 cm^{-1} associated with the aliphatic and aromatic (unconjugated and conjugated) C=C bonds in cured and uncured RBCs, respectively.

2.4. Reversed-Phase High-Performance Liquid Chromatography Measurement

Immediately after the irradiation, five samples of each material were immersed into 1.0 mL of the 75% ethanol/water storage medium in separate glass vials and stored in a 37 °C incubator. As recommended by the ISO 10993-13 description, the ratio between the sample and the storage solution volume was greater than 1:10, thus the specimens were fully immersed in the medium. The storage solutions were collected for analysis after 72 h. The RP-HPLC system (Dionex Ultimate 3000, Thermo Fisher Scientific Inc., Sunnyvale, CA, USA) consists of a Dionex LPG 3400 SD gradient pump, Rheodyne injector (Rheodyne, CA, USA), and a Dionex DAD 3000 RS UV-VIS detector (Dionex GmbH, Germering, Germany). Data acquisition was completed using Chromeleon software (version: 7.2.10). The separations were performed on a LiChrospher® 100 RP-18e (particle size: 5 µm, pore size: 100 Å) (Merck KGaA, Darmstadt, Germany) column (250 mm × 4.00 mm) with gradient elution. The composition of Eluent “A” was 100% bidistilled water, whereas Mobile Phase “B” was 100% *v/v* acetonitrile (ACN) (VWR International, Radnor, PA, USA). During the 30-min chromatographic separation, the “B” eluent content increased from 30–95%. The flow rate was 1.2 mL × min⁻¹. For the regeneration of the stationary phase, the content of Mobile Phase B was decreased from 95% to 30% in 1 min, and after 31–46 min, the system was washed with 30% “A”. The detection of the eluted monomers was carried out at the following wavelengths: 205, 215, 227, and 254 nm. 205 nm were found to be optimal; therefore, the evaluation relied on the data collected at this wavelength [24]. The separations were undertaken at room temperature. The amounts of the eluted monomers (Bisphenol A-glycidyl methacrylate, BisGMA; Triethylene-glycol-dimethacrylate, TEGDMA; urethane-dimethacrylate, UDMA; 1,12-dodecanediol-dimethacrylate, DDMA) were calculated using the calibration curve with the areas under the curve of peaks produced by the monomers, respectively. The monomer release was counted to 1 mg RBC. The TEGDMA, UDMA, BisGMA, and DDMA standard solutions had retention times of 12.2, 17.2, 19.1, and 27.2 min, respectively, whereas the peaks were well separated from each other.

2.5. Statistical Analysis

Pilot study results and sample size formula were used to estimate sample size.

$$\text{Sample size formula: } n = \frac{(z_{1-\frac{\alpha}{2}} + z_{1-\beta})^2 (s_1 + s_2)^2}{(M_1 + M_2)^2} = 3.$$

(*z* = standard score; α = probability of Type I error = 0.05; $z_{1-\alpha/2}$ = 1.96; β = probability of Type II error = 0.20; $1 - \beta$ = the power of the test = 0.80; $z_{1-\beta}$ = 1.28, $M_1 = 52$, $s_1 = 1.4$, $M_2 = 52$, $s_2 = 1.4$). By adopting an alpha (α) level of 0.05 and a beta (β) level of 0.20 (power = 80%), the predicted sample size (*n*) was found to be a total of 3 samples per group. Instead of the calculated 3 samples, $n = 5$ per group sample size was selected.

The statistical analyses were performed with SPSS v. 26.0 (SPSS, Chicago, IL, USA). Levene’s test was employed to test the equality of variance. This was followed by Paired Samples Test to analyze the differences in mean DC% between top and bottom surfaces and Two-tailed Independent Samples T-test to analyze the differences in mean DC% between the investigated materials polymerized at room temperature and with the application of pre-heating. The differences in monomer elution from the RBCs at the investigated temperatures were also compared with the Two-tailed Independent T-test.

Multivariate analysis (General Linear Model) and Partial Eta-Squared statistics were used to test the influence and describe the relative effect size for Material and Temperature as independent factors. *p* values below 0.05 were considered statistically significant.

3. Results

The measured maximum radiant exitance of the LED LCU was 1250 ± 15 mW/cm². The delivered radiant exposure was 25 J/cm². The LCU increased the temperature by an average of 7 °C when the thermocouple was irradiated through the empty 4 mm deep mold for 20 s.

Meanwhile, the Ena Heat Composite Heating Conditioner T2 setting stated preset temperature is 55 °C in 55 s, the real temperature of the FOB was 46 °C in the capsule after the recommended pre-warming period. The VisCalor Dispenser T1 setting provides 65 °C pre-warming in 30 s, however, the actual temperature of VCB was 60 °C after the recommended duration of pre-heating. Figure 1 shows the temperature change of RBC during the sample preparation from the material application into the mold until the end of the polymerization. The temperature of the 46 °C pre-heated FOB decreased to 33.4 °C as it was removed from the warming device and started to be applied into the mold and showed further temperature drop of −1 °C during the condensation. The total temperature decrease from the pre-heating until the start of polymerization was 13.6 °C in approximately 20 s. In the second phase, during polymerization, the exothermic reaction and the heat released from the curing unit elevated the RBC's temperature by 4 °C. Regarding the thermoviscous VCB, its pre-heated temperature (60 °C) decreased to 36.6 °C during the initial phase of the application and continued to show a further drop of (−2.6 °C) during the condensation phase. The total drop of temperature for the pre-heated VisCalor Bulk from the pre-heating until the start of polymerization was 26 °C in approximately 20 s. The temperature rise caused by the light-curing and the exothermic reaction was 4.4 °C. The consistency of the room temperature materials was highly viscous, especially of Viscalor making it difficult to squeeze out of the capsule. Compressing both room temperature RBCs was easy without sticking to the instruments. The pre-heating decreased the viscosity to a flowable consistency which allowed both materials to spread evenly throughout the template.

Considering the DC at the top and bottom surfaces in samples applied in 4 mm thickness, the mean percentages ranged between 54.2–64% and 45.0–51.8%, respectively (Table 2).

Table 2. Differences in mean DC% (S.D.) between the top and bottom surfaces of the investigated materials polymerized at room temperature and pre-heated condition.

	Room Temperature					Pre-Heated				
	Top	Bottom	t-Value *	p-Value *	95% CI Lower Upper	Top	Bottom	t-Value *	p-Value *	95% CI Lower Upper
FOB	63.0 (2.0)	51.8 (1.4)	31.7 (4)	<0.005	26.3 31.3	64.0 (1.4)	45.0 (1.9)	−17.2 (4)	<0.005	15.1 20.8
VCB	54.2 (2.9)	46.2 (1.4)	4.6 (4)	0.01	1.2 5.1	55.0 (1.3)	45.2 (4.1)	−25.1 (4)	<0.005	33.6 41.9

Abbreviations: DC, degree of conversion; FOB, Filtek One Bulk; VCB, Viscalor Bulk; CI, Confidence Interval.
* Paired Samples Test.

The DC values at the bottom of the specimens showed a statistically significant decrease for both materials at both temperatures in relation to the DC values measured at the top of the samples. When room temperature specimens were applied, the DC values were very similar to that of preheated samples, except on the bottom surface of FOB which was significantly lower when applied after pre-heating. In a comparison of the two bulk-fill RBCs, VCB showed a statistically significantly lower DC (~10% less) both on the top and bottom when applied at room temperature. Samples applied following pre-heating showed a significantly lower DC% only on the top (Figure 2). The lowest DC values were measured on the bottom surfaces of both investigated RBCs when they were applied with pre-heating.

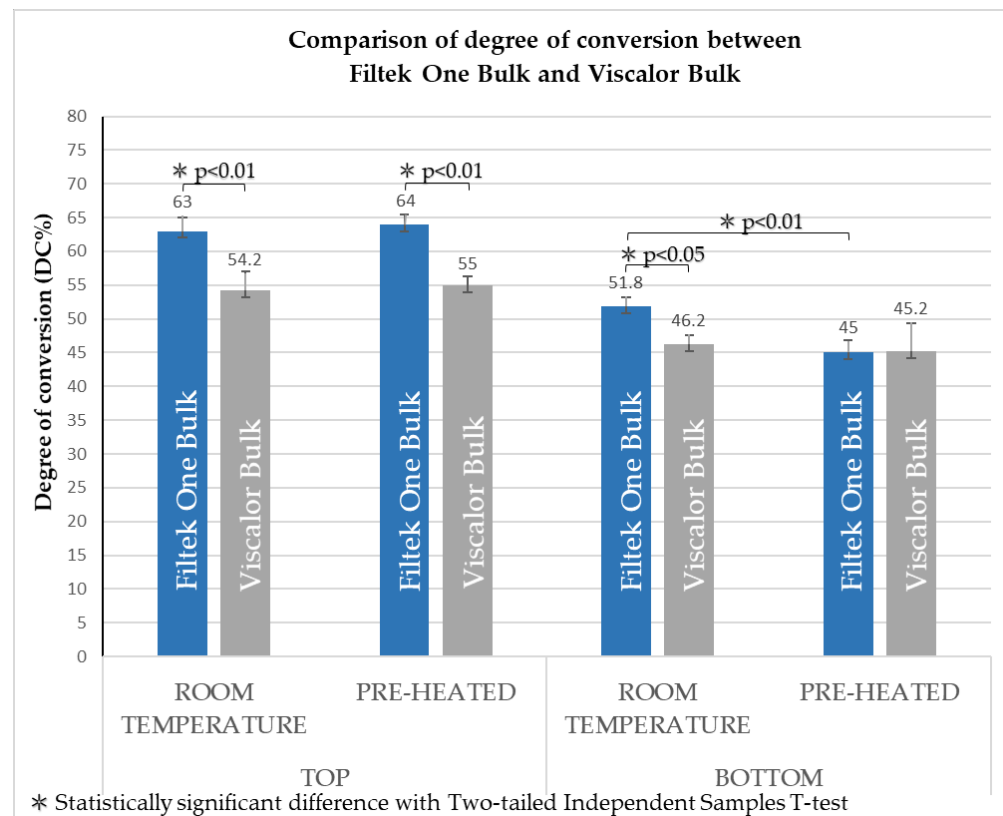


Figure 2. Comparison of the degree of conversion between Filtek One Bulk and Viscalar Bulk.

Table 3 presents the relative effect size of factor Material, Temperature, and their interactions on the degree of conversion of the top and bottom surfaces of the investigated resin-based composites.

Table 3. The relative effect size of factors Material, Temperature, and their interactions on the degree of conversion of the top and bottom surfaces of the investigated resin-based composites analyzed by General Linear Model and Partial Eta-Squared (η^2) statistics.

Factor	Degree of Conversion (DC)			
	Top Surface		Bottom Surface	
	<i>p</i> Value	Partial η^2	<i>p</i> Value	Partial η^2
Material	<0.001	0.86	0.028	0.27
Temperature	0.23	0.09	0.004	0.42
Material \times Temperature	0.81	0.004	0.022	0.29

A 2 (Material) \times 2 (Temperature) mixed-model ANOVA revealed that the main effect for Material on DC values measured on top surfaces was significant and the Partial Eta-squared was considered to be large, meanwhile not significant effect for Temperature was obtained with medium effect size. The interaction (Material \times Temperature) had no effect on the monomer conversion at the top.

Regarding the DC values at the bottom surfaces, the results showed a significant effect for both the Material and Temperature factor. The interaction between the two variables (Material \times Temperature) also significantly affected the monomer conversion at the bottom surfaces. The main effect for Material was significant at room temperature, meanwhile, the Temperature factor affected significantly only the FOB RBC.

In addition to the monomers specified by the manufacturers, other methacrylates were also detected from both FOB (BisGMA) and VCB (TEGDMA, DDMA) RBCs.

The differences in the monomer elutions were also significant between FOB and VCB both when applied at room temperature and with pre-heating in the case of all the evaluated monomers, except for DDMA which was released in similar (statistically insignificant) amounts from the pre-heated RBCs (Table 4 and Figure 3).

Table 4. Differences in monomer elution from Filtek Bulk One and Viscolor Bulk analyzed by Independent Samples T-test.

	Monomer	Resin Composite	Mean (S.D.)	t-Value (df)	p-Value	95% CI	
						Lower	Upper
Room temperature	TEGDMA	Filtek One Bulk Viscolor Bulk	0 0.1 (0.01)				
	UDMA	Filtek One Bulk Viscolor Bulk	0.33 (0.34) 0.01 (0.01)	20.57 (8)	<0.001	0.29	0.36
	BisGMA	Filtek One Bulk Viscolor Bulk	0.22 (0.01) 2.3 (0.24)	−19.43 (8)	<0.001	−2.33	−1.84
	DDMA	Filtek One Bulk Viscolor Bulk	0.26 (0.02) 0.1 (0.02)	13.08 (8)	<0.001	0.13	0.18
Pre-heated	TEGDMA	Filtek One Bulk Viscolor Bulk	0 0.08 (0.01)				
	UDMA	Filtek One Bulk Viscolor Bulk	0.17 (0.11) 0.02 (0.03)	2.92 (8)	0.02	0.03	0.27
	BisGMA	Filtek One Bulk Viscolor Bulk	0.08 (0.08) 1.98 (0.38)	−10.9 (8)	<0.001	−2.29	−1.49
	DDMA	Filtek One Bulk Viscolor Bulk	0.15 (0.09) 0.09 (0.04)	1.19 (8)	0.27	−0.05	0.16

Abbreviations: S.D., standard deviation; df, degree of freedom; CI, Confidence Interval; TEGDMA, triethylene glycol dimethacrylate; UDMA, urethane dimethacrylate; BisGMA, Bisphenol A diglycidil ether dimethacrylate; DDMA, dodecyl methacrylate.

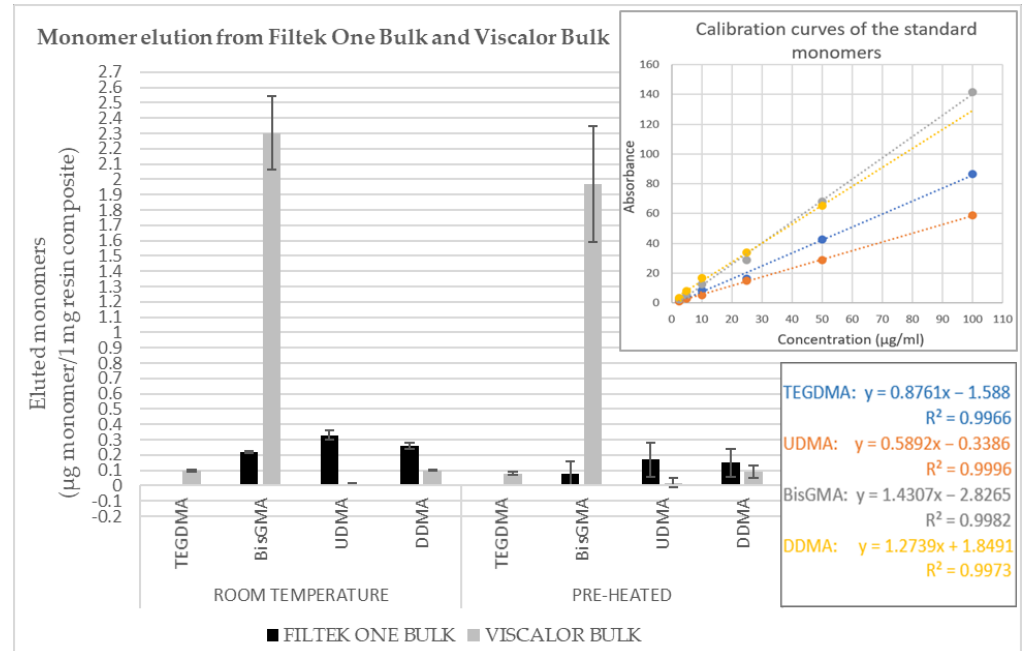


Figure 3. Monomer elution from Filtek One Bulk and Viscolor Bulk.

At room temperature, 30 and 2.5 times as much UDMA and DDMA were released, respectively from FOB, while 10.5 times more BisGMA was eluted from VCB. The latter was the monomer released in the largest amount. With the utilization of pre-heating, 7.5 times as much UDMA was found to elute from FOB, while 25 times more BisGMA was released from VCB. For FOB, preheating significantly reduced the amount of eluted monomers, while for VCB, the temperature did not affect the dissolution (Table 5).

Table 5. Differences in monomer elution at room temperature and pre-heated conditions were analyzed by Independent Samples T-test.

	Monomer	Temperature	Mean (S.D.)	t-Value (df)	p-Value	95% CI	
						Lower	Upper
Filtek One Bulk	TEGDMA	Room temperature	0				
		Pre-heated	0				
	UDMA	Room temperature	0.33 (0.34)	3.04 (8)	0.016	0.04	0.28
		Pre-heated	0.17 (0.11)				
Viscalor Bulk	BisGMA	Room temperature	0.22 (0.01)	3.73 (8)	0.006	0.05	0.22
		Pre-heated	0.08 (0.08)				
	DDMA	Room temperature	0.26 (0.02)	2.6 (8)	0.03	0.01	0.21
		Pre-heated	0.15 (0.09)				
Viscalor Bulk	TEGDMA	Room temperature	0.1 (0.01)	3.12 (8)	0.014	0.01	0.04
		Pre-heated	0.08 (0.01)				
	UDMA	Room temperature	0.01 (0.01)	−0.87 (8)	0.41	−0.05	0.02
		Pre-heated	0.02 (0.03)				
Viscalor Bulk	BisGMA	Room temperature	2.3 (0.24)	1.64 (8)	0.14	−0.13	0.79
		Pre-heated	1.98 (0.38)				
	DDMA	Room temperature	0.1 (0.02)	0.36 (8)	0.73	−0.04	0.05
		Pre-heated	0.09 (0.04)				

Abbreviations: S.D., standard deviation; df, degree of freedom; CI, Confidence Interval; TEGDMA, triethylene glycol dimethacrylate; UDMA, urethane dimethacrylate; BisGMA, Bisphenol A diglycidil ether dimethacrylate; DDMA, dodecyl methacrylate.

The following order of mean monomer elution was detected from FOB for both room temperature and pre-heated samples from highest to lowest: UDMA < DDMA < BisGMA, meanwhile the amount of leached monomers was roughly half (UDMA, DDMA) or one-third (BisGMA) for pre-heated specimens.

Regarding VCB, both the order, as well as the amount of the released monomers were the same in the case of both the room temperature and the pre-heated samples (BisGMA < TEGDMA < DDMA < UDMA), except for TEGDMA, which showed a significantly lower elution from the pre-heated samples.

Table 6 presents the relative effect size of factor Material, Temperature, and their interactions on the monomer elution from the investigated resin-based composites.

Table 6. The relative effect size of the factors Material, Temperature, and their interactions on the monomer elution from the investigated resin-based composites analyzed by General Linear Model and Partial Eta-Squared (η^2) statistics.

Factor	Monomer Elution							
	BisGMA		UDMA		TEGDMA		DDMA	
	p Value	Partial η^2	p Value	Partial η^2	p Value	Partial η^2	p Value	Partial η^2
Material	<0.001	0.96	<0.001	0.75			<0.001	0.49
Temperature	0.036	0.23	0.046	0.21	0.014	0.55	0.036	0.23
Material \times Temperature	0.35	0.054	0.006	0.38			0.04	0.24

Abbreviations: BisGMA: bisphenol-A diglycidil ether dimethacrylate; UDMA: urethane dimethacrylate; TEGDMA: triethylene glycol dimethacrylate; DDMA: 1,12-dodecane dimethacrylate.

A 2 (Material) \times 2 (Temperature) mixed-model ANOVA showed that the main effect for Material was significant on UDMA, BisGMA, DDMA release with a Partial Eta-squared value which was considered to be large. The Temperature factor also influenced significantly the monomer elution, however, its effect was slightly weaker compared to the Material's effect. TEGDMA was released only from VCB. The effect of the Temperature factor was calculated to be significant on the elution of this monomer. The interaction between the two factors (Material \times Temperature) had a significant effect on UDMA and DDMA elution, while the elution of BisGMA was independent of the Material \times Temperature interaction.

4. Discussion

In this *in vitro* study, the DC and the elution of unreacted monomers of a thermoviscous and high-viscosity bulk-fill dental resin composites were assessed using micro-Raman spectroscopy and High-Performance Liquid Chromatography measurements. Additionally, the thermal change of the RBCs was also registered during the sample preparation with a K-type thermocouple to assess the temperature change of the room temperature and pre-heated RBCs during the manipulation and polymerization phases.

The setting reaction of RBCs has a major influence on their mechanical and biological properties [35]. RBC polymerization depends mainly on the chemical structure of the monomers, filler characteristics, the photoinitiator type, and concentration, and the polymerization conditions [36]. The latter includes, among others, the volume and the layer thickness of the applied RBC, spectral characteristics of the curing unit, exposure time, and the pre-cure temperature of the material [4,14,37]. Since these were standardized in this study, except for pre-cure temperature, differences in the DC value and monomer elution of the bulk-fill RBCs can be attributed to the different compositions and temperatures of the materials before polymerization. Thus, the effect size of the material factor and pre-cure temperature factor became assessable on the degree of conversion and monomer elution from the investigated bulk-fill RBCs.

According to our results, the first null hypothesis, which stated that the pre-heating had no effect on post-cure DC% of VisCalor Bulk and Filtek One Bulk, was partially rejected, since pre-warming of RBCs neither increased nor decreased the DC on the top of both materials and the bottom of VCB_65, however, the bottom DC was significantly decreased in the case of FOB_65. The second null hypothesis was also partially rejected, because the external heating of the investigated RBCs decreased the monomer elution in the case of FOB regarding all the investigated monomers and TEGDMA elution VCB, however, had no influence on the BisGMA, UDMA, and DDMA release from VCB.

It has been reported that increased pre-cure temperature of RBC may result in a greater extent of monomer to polymer conversion [6,38]. However, investigations, that have shown improvement in the degree of polymerization upon pre-warming generally maintained the RBC temperature constant during the experimentation [6,10]. On the other hand, there are also results that found increases in DC at non-isothermal conditions to be material composition-dependent [19,39].

Regarding the real-life clinical scenario, the RBC's temperature drops rapidly to the physiological level upon removal from the pre-heating device [2,29]. In contrast to the studies that have demonstrated optimized monomer conversion in the case of pre-heated RBCs under isothermal conditions, Yang et al. and Tauböck et al. reported that the pre-heated RBC's temperature dropped to ~35–36 °C during the handling phase before light-cure [27,39]. Additionally, the pre-warmed RBC can reach a lower internal temperature than the maximum stated preset temperature of the heating device [27,40].

To overcome this problem, a new warming device was developed, namely, VisCalor Dispenser. The capsule dispenser itself can provide homogeneous warming of the highly filled RBCs to 68 °C (only for VOCO products) with near-infrared technology. Thus, the RBC does not need to be removed from the heating device for dispensing into the prepared cavity. During application it is flowable and when it comes in contact with the tooth VCB reaches body temperature within a short time and thus returns to the high-viscosity, sculptable state.

In the present study, during the specimen preparation, the glass slab, holding the PTFE mold, was pre-set to a temperature of 30 ± 1 °C, representing a rubber dam isolated tooth [41]. In the case of both investigated materials, the temperature decreased during the extrusion from the capsule, irrespectively to the type of the heating device, and a further drop of temperature was observed during the condensation into the mold (Phase I on Figure 1). The measured temperatures for FOB_55 and VCB_65 were on average 32.5 °C and 34 °C, respectively, at the start of polymerization. The direct contact to the 30 °C molds and glass slab and also to the room temperature condensing instrument accelerated the cooling

of the RBCs. The equilibration of the ambient and the pre-heated RBC's temperature resulted in faster cooling of the warmer RBC. During the photopolymerization (Phase II on Figure 1.), both exothermic reaction and the released heat from the curing device increased the RBCs' temperature. The extent of temperature increase, however, seems to be influenced by the speed of the temperature drop in Phase I. Accelerated drop may hinder the exothermic temperature increase.

Adequate monomer to polymer conversion is crucial to the material's long-term clinical success [42]. While DC is the key parameter determining the effectiveness of monomer conversion, unfortunately, it cannot describe the microstructure of the resulted heterogenic polymer network, which has a major effect on the physical and chemical properties of the RBC [43]. To determine the DC, micro-Raman spectroscopy was used in our study. It offers the possibility of quantitative characterization of the polymerization extent in dimethacrylate-based RBCs [44]. Raman-spectra were taken after 24 h, since a significant increase in DC takes place during the 24 h post-irradiation [28]. Although the minimum DC% for clinically acceptable restoration has not yet been precisely defined [45,46], DC values below 55% may be inadequate for occlusal restorative layers [47,48]. Musanje and Darvell recommended that the depth of cure should be defined as the depth at which maximum conversion occurs for a given irradiance and exposure time [49]. The radiant exposure was defined at 16–24 J/cm² to reach an adequate polymerization degree for a 2 mm thick RBC layer [50,51], meanwhile, the minimum radiant exposure required to be delivered to different bulk-fill RBCs moves on a wider scale (14–23–47 J/cm²) [52,53]. In our study, the valid, portable radiometer measured a higher value of radiant exitance than the average light output given by the manufacturer. It was demonstrated that most of the LCUs—especially low-budget LCUs, like our LED.D—could have different light output characteristics [54]. Providing by the curing unit, the delivered radiant exposure was 25 J/cm², and the DC% on the top of FOB_RT and FOB_55 were 63% and 64%, respectively, which is a characteristic value for a well polymerized RBC [49]. In comparison to the FOB values, the DC% on the top of the VCB_RT and VCB_65 samples were significantly lower, 54% and 55%, respectively. The lower DC values are presumably due to the material composition. The monomer system has a major effect on the DC, which increases in the following order: BisGMA < BisEMA < UDMA < TEGDMA [55]. BisGMA is considered to be the most viscous monomer due to the strong intramolecular hydrogen bonding, resulting in limited rotational freedom, thus the reactivity and mobility of the monomer may decrease during the polymerization process [56]. This might be one of the explanations for the significantly lower DC of the VCB, which is a BisGMA-based RBC. FOB is an UDMA-based bulk-fill RBC, containing both aliphatic and aromatic UDMA. Sideridou et al. found that UDMA, combining relatively high molecular weight with a high concentration of double bonds and low viscosity, was shown to reach higher final DC% values than BisGMA [55]. Although the viscosity of UDMA is much lower than that of BisGMA, when it is mixed with the high molecular weight BisGMA or BisEMA, it can significantly restrict the mobility of UDMA monomers and decrease their reactivity and conversion value [57,58]. In addition to the monomer system, the filler-matrix ratio is also decisive. VCB filler loading is higher (83 wt%) compared to the filler content of FOB (76.5 wt%), which may restrict the light penetration and the mobility of monomers and radicals. The DC of VCB was investigated by Yang et al., and their results showed similar values (~58 DC%) in a 2 mm thick sample, exposed either with 20 s (24 J/cm²) or 40 s (48 J/cm²) [28]. Similar to the above study, pre-heating did not influence the DC% values of our investigated materials at the top of the samples, assuming that the RBCs on the top reached their maximum conversion degree already at room temperature. Although Daronch et al. found that the increased pre-cure temperature significantly improved the DC compared to the room temperature, they also concluded, that at longer exposure (20 s, 40 s) top-surface composite conversion was equivalent and similar throughout the tested temperature range (22–60 °C) [10].

Contrary to the values measured at the top of the room temperature samples the DC% at the bottom of the 4 mm thick bulk-fill materials were lower by ~10% (FOB_RT,

51.8%; VCB_RT, 46.2%). While the depth of cure is improved in bulk-fills due to increased translucency, modified matrix composition, photoinitiator kinetics, and filler characteristics, not all of the commercial bulk-fill RBC are able to sustain a homogeneous conversion at a depth of 4 mm [24,59–61]. The present research examined two, so-called full-body bulk-fill RBCs that are often referred to as paste-like bulk-fills. These materials generally have a higher filler load which makes them highly viscous and therefore sculptable. The higher filler content renders the surface more wear-resistant without requiring any coverage. Decreased DOC from the surface to the bottom may be a result of the increased filler ratio which may hinder light penetration due to the nano-sized particles despite the increase in translucency. On the other hand, high molecular weight monomers, such as BisGMA (in VCB) and aromatic UDMA (in FOB), also help to increase the viscosity, however, decreasing the reactive groups in the resin may negatively influence the DC [56,62].

Pre-heating makes highly filled, sculptable RBCs more flowable, adaptable, and easier to manipulate, without compromising the superior mechanical properties. Decreased viscosity has been shown to enhance marginal adaptation and reduce microleakage due to improved wetting of cavity walls [14,15]. Although, the increased pre-cure temperature has benefits through decreased system viscosity, enhanced radical mobility, and collision frequency of unreacted active groups resulting in additional polymerization and higher conversion [63], the diversity of study outcomes may result from different RBC composition and experimental set-ups. Isothermal conditions mostly favor the positive effect of pre-heating on monomer conversion resulting in more highly cross-linked polymer networking and improved mechanical and physical properties [10]. However, improved monomer to polymer conversion has a strong relation to polymerization shrinkage which may increase the shrinkage stress of the bonded restoration [47,63,64]. Despite the higher shrinkage which might be present, it may not be clinically significant, as it can be offset by the improved marginal adaptation [63].

Clinically relevant, non-isothermal circumstances enhance the strong effect of RBC composition on the results. Several studies found the effect of pre-heating to vary on DC (decrease, no change, increase) depending on the composition of the investigated RBC [14,19,39].

Confirming the above findings, our results also showed a dissimilar effect of pre-heating on the monomer conversion of the investigated RBCs. An increase in pre-cure temperature did not influence significantly the DC on the top surfaces neither for FOB_55 nor for VCB_65 and even did not affect the bottom DC of VCB_65 compared to the room temperature RBCs. The bottom DC values showed a significant decrease however in the case of FOB_55. The rapid temperature drop of pre-heated RBC during handling results in excess heat loss which may deprive energy of the system and might prevent a sufficient increase in polymerization reactivity and consequent enhancement in monomer conversion [19]. Considering the findings of the temperature measurements, it is visible, that the temperature increase during polymerization shows a direct correlation with the measured DC values. During polymerization (Phase II. on Figure 1), the temperature of both VCB_RT and VCB_65 increased by 4.4 °C and showed similar DC values at the top (54.2 °C and 55 °C, respectively) and as well on the bottom surfaces (46.2 °C and 45.2 °C, respectively). The mean differences between top and bottom DCs were around 10%. Meanwhile, the temperature within FOB_RT during polymerization rose by 5.8 °C and showed a significantly higher DC both on the top (63 °C) and bottom (51.8 °C), compared to VCB_RT. The mean difference on top vs. bottom DC was found to be 10% also. In contrast, pre-heating of FOB had a negative effect both on the exothermic reaction and on the kinetics of monomer conversion. During light-curing, the temperature rise within FOB_55 was 1.8 °C lower (4 °C) than in the case of FOB_RT, and the bottom DC was 20% less (45%) compared to the top DC value, which kept its higher level (64 °C). Although the drop of temperature during the dispensing and condensation phase (Phase I. on Figure 1) of VCB_65 was rapid, its temperature at the initiation of light-curing was higher, compared to the FOB_55. It may have provided enough energy to the polymerizing system, assuming

that even higher exothermic temperature rise and higher DC would have been achieved if the system temperature dropped slower. In contrast, the temperature of FOB_55 at the beginning of light-curing was lower, and the additional drop during the cooling phase may have deprived energy from the system, resulting in a weaker exothermic reaction and lower monomer conversion. Since the reaction behavior of multifunctional monomer systems is very complex and highly dependent on the reaction conditions and composition, other possible explanations for DC decrease may arise. The increased pre-cure temperature may induce thermal polymerization before irradiation. On one hand, thermal polymerization leads to the consumption of functional groups, and on the other hand, pre-polymerization of few monomers results in shrinkage, which decreases the system's initial free volume and restricts the diffusion of monomers during the progression of the photopolymerization [65]. This phenomenon in VisCalor Bulk was investigated by Yang et al. who concluded, that pre-heating did not cause adverse effects through premature polymerization [28]. Other factors which may have been responsible for the observed temperature behavior of the FOB_55 could be further thermal side effects including evaporation of the reactants and thermal degradation of the photoinitiator [66]. In the case of VCB_65, 30 s pre-heating time was enough in the special dispenser, however, for FOB_55 pre-warming took more time in the EnaHeat Composite Heating Conditioner (15 min), which may influence the chemical condition of the components. Among the above-mentioned thermal side effects, a higher significance should be attributed to oxygen inhibition in the case of elevated RBC temperatures. As the temperature increases, the decrease in viscosity promotes oxygen penetration into the RBC. Oxygen reduces the extent of polymerization by scavenging on free radicals resulting in less reactive peroxy radicals and/or quenching of the excited triplet state of the initiator [43]. It is reasonable to assume a role also for a further FOB constituent, the so-called AFM, an addition-fragmentation chain transfer dimethacrylate monomer, which participates readily in network formation by copolymerizing with multifunctional methacrylates [67]. An AFM is a heteroatom (N or S or O) containing monomer with various vinyl activating groups which have been employed as chain transfer agents to reduce shrinkage stress [65,68]. However, chain-transfer reactions may also exert a retarding effect on the polymerization by increasing termination, especially at higher temperatures [65].

Although pre-heating did not increase monomer conversion in many cases, several studies have shown that the mechanical properties and marginal integrity of RBCs (including FOB and VCB as well) are satisfactory or better than those applied at room temperature [2,14,69,70]. In contrast, however, there are experiments concluding higher linear shrinkage of pre-heated RBCs and deterioration in marginal integrity [63,71], although, results of the available investigations show that the pre-heating has no significant impact on bond strength of RBC to dentin [72,73].

As our results confirmed, RBCs do not have a complete monomer to polymer conversion because of the condition-dependent kinetics of gelation, vitrification, immobilization, and steric isolation [74]. Incomplete conversion may result in the presence of unreacted monomer content within the polymer network which is partially or completely released short- or long-term [34,43]. Released monomers may depress the biocompatibility of the RBC by stimulating bacterial growth around the restoration leading to secondary caries development and may promote allergic reactions. Additionally, cytotoxic effects of monomers have been demonstrated [32]. Solubility and water sorption can accelerate the degradation and do harm to the mechanical/physical properties such as tensile-, flexural strength and wear [33]. To determine the quality and quantity of the residual monomers eluted from the investigated polymerized materials HPLC, as a generally applied investigative method, was used in our study [24,75,76]. Unreacted monomers can reduce the mechanical properties of the RBCs and their detection represents an important step for evaluating RBC biocompatibility [77].

During our experiment, aromatic (BisGMA) and aliphatic (TEGDMA, UDMA, and DDMA) dimethacrylate standard monomers were used to identify eluted monomers from the investigated RBCs.

In the present study, 75% ethanol/water solvent was used to extract most of the examined unreacted monomers from the polymerized RBC specimens to identify monomer quantity. Besides the type of the solvent, the chemical nature of the matrix monomers and their combination, the degree of conversion, and the final network characteristics also play important roles in the quantity and quality of monomer elution from a certain RBC [24,34].

According to the manufacturer's description, VCB is a BisGMA/aliphatic dimethacrylate based RBC but does not define in detail the aliphatic dimethacrylates. However, following HPLC measurements, TEGDMA, DDMA, and UDMA were detected as eluted aliphatic monomers from VCB. FOB is mainly an UDMA-based material, composed of both aliphatic and—as a BisGMA substitute—aromatic UDMA. However, released BisGMA was also detected during the HPLC measurements. Copolymers consisting of BisGMA and/or UDMA are crosslinked both chemically (C-C covalent bond) and physically (i.e., hydrogen bond). The latter determines the matrix viscosity, since the more numerous and stronger the hydrogen bonds are, the higher the viscosity of the system [56]. To create a sculptable RBC, the usage of these monomers is advantageous.

Regarding the monomer release, our results showed elution to be strongly dependent on the material.

In this study, significantly (two-fold) more UDMA and DDMA were released from the room temperature FOB_RT, meanwhile, VCB_RT samples leached almost three-fold more BisGMA. Pre-heating significantly decreased the monomer elution from FOB_55. There was no difference however in monomer elution between VCB_RT and VCB_65. The measured unreacted monomer release is in line with our results regarding the degree of monomer conversion in VCB_RT and VCB_65 since pre-heating did not change the DC on the top or bottom of VCB. On the other hand, the observed relationship between DC and monomer elution from FOB is contradictory. While the DC of the bottom surface decreased after pre-heating, the detected elution of unreacted monomers from FOB_55 samples was also lower. Although several studies have shown that the extent of leached unreacted monomer is correlated to the DC [4,78,79], the conversion degree does not necessarily correlate with the amount of free residual monomer, since the detected double bonds may remain as pendant groups bonded to the polymer structure and are not free to be released, however, may reduce the clinical success of the RBCs [56,80]. Probably, the above issue is the explanation for the lack of the expected relationship between the DC and monomer elution in the case of the pre-heated FOB_55.

While the number of monomer elution studies from bulk-fill RBCs is extensive, data regarding the effect of pre-heating on monomer release both from conventional and bulk-fill RBCs is limited in the literature, hence, the discussion of this issue and comparison to other results are also restricted. Elution from bulk-fills was found to be comparable to that of conventional RBCs despite their increased increment thickness [34,81]. The quality and quantity of released resins are strongly material dependent and the amount of most of the eluted monomers is increased with time [24,82]. The monomer detected to be eluted in the highest amount was BisGMA from both VCB_RT and VCB_65, with the latter showing a significantly lower quantity. As it was previously mentioned, the extremely high viscosity of BisGMA limits the DC, leaving behind more unreacted monomers, which may release into the oral cavity. Admixing low molecular weight monomers, such as TEGDMA and DDMA, to BisGMA, can lower its viscosity, and via their synergistic effect can increase the rate of polymerization [83]. The released quantity of the latter two was very small both from VCB_RT and VCB_65. FOB, on the other hand, is a UDMA-based RBC. At present, UDMA is the only commercial alternative to the bisphenol A-based dental methacrylates [56]. Although, UDMA viscosity is lower than BisGMA, still high enough to require the addition of a reactive diluent, such as DDMA. Due to UDMA's lower molecular weight in comparison to BisGMA, it is expected to show higher DC and lower unreacted monomer elution [84]. However, aside from aliphatic UDMA, FOB contains aromatic UDMA as well. Aromatic moiety and substitution symmetry play an important role in the steric hindrance, methacrylate group separation, limited conformational freedom, and

increase of molecular stiffness. However, closer proximity of reactive groups facilitates the reaction-diffusion, which may lead to moderate DC and the planar geometry of benzene rings allows for building tighter structures [56]. Based on our results, the aliphatic UDMA release was moderate from FOB_RT and significantly lower from FOB_55, as a result of pre-heating.

However, in our study, neither eluted AUDMA nor AFM was detected in the absence of the standards, as their exact chemical structure is a trade secret.

To the best of our knowledge, only one published article deals with the monomer elution from three pre-heated RBCs and found no effect of pre-cure temperature (68 °C) on the amount of leached UDMA, TEGDMA, and BisGMA [85]. Few available pieces of research reported that pre-heating of both conventional and bulk-fill RBCs did not influence cell viability, however, polymerized samples were used to examine the cytotoxicity without determination of the eluted monomers [63,83,86]. de Castro et al. investigated the sorption and solubility of RBC at higher pre-cure temperatures (60 °C) and concluded that longer curing times and higher temperatures led to lower values of sorption and solubility, but these differences were only significant for specific combinations of temperatures and curing times. [87].

The main limitation of this study may be the in vitro nature of the investigation. Although during sample preparation a conscious effort was made to simulate an isolated tooth by adjusting the PTFE mold temperature, the thermal conductivity of a natural tooth, its position in the oral cavity, the cavity configuration, thus the contact surfaces with the RBC are just a few mentioned factors, which may influence the results in vivo. Furthermore, the analysis of the elution of selected unreacted monomers (BisGMA, UDMA, TEGDMA, DDMA) will not provide an absolute measure of the quality of released components, since, among others, various monomers, like AUDMA, AFM, degraded compounds, initiator molecules, and fillers may also leach and compromise the RBC biocompatibility. A further limitation may be the limited number of the investigated high-viscosity bulk-fill RBCs, especially considering the strongly material-dependent results. The results cannot be extrapolated to other room temperature and pre-heated RBCs, since the composition has a strong influence on both DC and monomer elution and can vary from RBC to RBC.

5. Conclusions

Within the limitations of this in vitro study, the following conclusions can be stated:

- (1) Significantly higher DC values were achieved on the top of the room temperature and pre-heated investigated bulk-fill RBCs than on the bottom.
- (2) Room temperature VisCalor Bulk has lower DC% values both on the top and bottom compared to Filtek One Bulk.
- (3) Pre-heating did not influence the DC of VisCalor Bulk, however, significantly decreased the DC at the bottom of Filtek One Bulk.
- (4) Pre-heating had no effect on the monomer elution from VisCalor Bulk, but significantly decreased the monomer release from Filtek One Bulk.
- (5) *Material* factor had a significant effect on each investigated variable, while *Temperature* factor and its interaction with *Material* is surface- (top vs. bottom) and monomer-dependent.

Based on the results, the following clinical significance can be deduced: While pre-heating had no beneficial effect on the degree of conversion neither of the thermoviscous VisCalor Bulk nor the contemporary bulk-fill RBC (Filtek One Bulk) the increased pre-cure temperature may decrease the elution of unreacted monomers from the RBCs.

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Article

Effect of Ceramic and Dentin Thicknesses and Type of Resin-Based Luting Agents on Intrapulpal Temperature Changes during Luting of Ceramic Inlays

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Abstract: The adhesive cementation of ceramic inlays may increase pulpal temperature (PT) and induce pulpal damage due to heat generated by the curing unit and the exothermic reaction of the luting agent (LA). The aim was to measure the PT rise during ceramic inlay cementation by testing different combinations of dentin and ceramic thicknesses and LAs. The PT changes were detected using a thermocouple sensor positioned in the pulp chamber of a mandibular molar. Gradual occlusal reduction obtained dentin thicknesses of 2.5, 2.0, 1.5, and 1.0 mm. Light-cured (LC) and dual-cured (DC) adhesive cements and preheated restorative resin-based composite (RBC) were applied to luting of 2.0, 2.5, 3.0, and 3.5 mm lithium disilicate ceramic blocks. Differential scanning calorimetry was used to compare the thermal conductivity of dentin and ceramic slices. Although ceramic reduced heat delivered by the curing unit, the exothermic reaction of the LAs significantly increased it in each investigated combination (5.4–7.9 °C). Temperature changes were predominantly influenced by dentin thickness followed by LA and ceramic thickness. Thermal conductivity of dentin was 24% lower than that of ceramic, and its thermal capacity was 86% higher. Regardless of the ceramic thickness, adhesive inlay cementation can significantly increase the PT, especially when the remaining dentin thickness is <2 mm.

Keywords: pulpal temperature; indirect restoration; dentin thickness; ceramic thickness; preheated resin composite; adhesive cement



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1. Introduction

Posterior indirect partial restorations (inlay, onlay, overlay) are widely used in dental clinical practice to overcome issues resulting from the use of direct resin-based composites (RBCs) [1–3].

The adhesive cementation of ceramic inlays is recommended to improve the esthetic and mechanical properties of the restoration [4–8]. Resin-based adhesive cements can be classified as self-cured, dual-cured, or light-cured based on their polymerization mechanism [9]. An alternative innovation is the use of chairside preheated conventional restorative RBCs as a luting agent for indirect ceramic restorations. Owing to their reduced viscosity, low film thickness and good adaptation can be achieved [10]. In addition to color stability, favorable mechanical and physical properties are further benefits as a result of their high filler load [11–13]. An increased pre-polymerization temperature can improve the monomer-to-polymer conversion; however, this might be compromised during the luting procedure because of the rapid cooling of the luting RBC before it is light-cured [14–16].

The durability of adhesively bonded restorations depends on the degree of conversion of the adhesive cement [17,18]. However, indirect ceramic restorations attenuate the light passing through them [19]. The interposed material might impair the mechanical and esthetic properties of the luting agent caused by the reduced degree of conversion, thus compromising the durability of the indirect restoration [20,21]. To overcome or attempt to compensate for this drawback, the light intensity should be sufficiently high or the exposure time should be as long as possible, considering light attenuation as a function of the restoration thickness [20,22]. However, an increase in irradiance delivered from the curing unit or upon extending the exposure time as a strategy to enhance the polymerization degree might cause an unfavorable temperature rise within the pulp chamber [23]. The temperature rise during polymerization is due to the absorption of energy by the irradiated objects together with the heating of the curing unit itself [24]. However, the polymerization of resin-based materials is an exothermic reaction that leads to further heat generation [25,26]. Bouillaguet et al. [24] reported that, by using infrared imaging, the highest temperature increases were recorded inside the RBC material and not outside the tooth during photocuring. The exothermic reaction is a material-, consistency-, and thickness-dependent phenomenon [26]. Although the majority of the heat generated during RBC polymerization is dissipated, the increase in pulpal temperature may exceed the putative pulpal damage threshold [27]. Several studies have consistently reported that the remaining dentin thickness is a critical factor in relation to the intrapulpal temperature increase due to the heat dissipating effect [23,28,29]. According to the second law of thermodynamics, during light curing, heat flows from the external tooth surface or polymerizing material to the pulp chamber as the temperature difference is equalized by diffusion [30]. The thermal conductivity of human dentin was calculated at approximately 0.36–0.67 W/mK by de Magalhães et al. [31]. Although heat transfer or heat flux occurs at a lower rate in dentin, inducing a thermal insulating effect, the potential for pulpal damage is expected to be great in deep cavities where the tubular surface area increases and the light attenuation effect is weak [32–34]. Thus, clinically, it would be optimal for dentin preservation, or continuous high-energy output photo curing should be avoided to protect pulp tissues from thermal injury [35]. Onisor et al. [36] conducted active cooling to reduce the heat during prolonged polymerization employed for luting indirect adhesive restorations with light-cured materials. Additionally, the shielding effect of dentin resulted in a lower pulpal temperature increase than that of the interposed ceramic restoration during light curing [35]. In addition to the interposed materials and the distance between the cavity floor and the pulp, the effect of pulpal blood circulation, volume, and perfusion of the fluid in the dentinal tubules as well as in the surrounding tissues play important roles in heat conduction and protection against the rise in pulpal temperature [37,38]. The putative pulpal damage threshold was based on the study conducted by Zach and Cohen. A temperature rise in pulpal tissues of 5.5 °C may lead to irreversible changes [39,40]. In support of the above observation, a recent *in vivo* study showed that increased pulpal temperatures may induce inflammatory reactions, even if the temperature rise does not exceed the previously defined 5.5 °C threshold [41].

Although several investigations have been conducted on the effects of the light curing unit, resin composite type, and remaining dentin thickness on the pulpal temperature rise, data are lacking in the dental literature regarding the effects of different dentin thicknesses on pulpal temperature change during cementation of different thicknesses of indirect ceramic restorations with adhesive resin cements or preheated restorative RBCs [25,26,33,42–45]. Furthermore, reliable, comparative data on the thermophysical properties of dentin and ceramic are essential to obtain precise calculations of the thermal changes in teeth and provide safer dental procedures, such as ceramic inlay cementation.

Therefore, this study compared, *in vitro*, the intrapulpal thermal changes resulting from cementation of ceramic inlay with light- and dual-curing adhesive resin cements and preheated sculptable submicron restorative RBCs. The aim of this study was to assess

the influence of simultaneously variable dentin and ceramic layer thicknesses on pulpal temperature rise, supplemented by a qualitative comparison of the thermal properties between dentin and ceramics.

The null hypotheses of the study were threefold: (1) there is no difference in pulpal temperature change using different luting agents during ceramic inlay cementation; (2) there is no significant influence of ceramic and dentin layer thicknesses on pulpal temperature rise; and (3) there is no significant difference between the thermal conductivity and heat capacity of dentin and ceramics.

2. Results

The maximum radiant exitance of the LED LCU was 1550 ± 15 mW/cm². The delivered maximum incident radiant exposure with a 40 s exposure duration was 62 ± 0.6 J/cm². The radiant exitance was reduced by 20% (1240 ± 12 mW/cm²) by the 6×6 mm orifice; thus, the radiant exposure with 40 s exposure duration was 49.6 ± 0.6 J/cm², which was delivered to the top of the ceramic specimens. The 2.0 mm, 2.5 mm, 3.0 mm, and 3.5 mm distances between the light guide tip and the radiometer sensor and the limited orifice of the mold significantly decreased the radiant exposure. Through the empty 2.0 mm, 2.5 mm, 3.0 mm, and 3.5 mm deep molds, the radiant exposures decreased by 42% (36 ± 0.4 J/cm²), 45% (34 J/cm² ± 0.3), 48% (32.2 ± 0.3 J/cm²), and 51% (30.4 ± 0.3 J/cm²), respectively. The 2.0 mm, 2.5 mm, 3.0 mm, and 3.5 mm thick ceramics further decreased the radiant exposures by 67% (20.5 ± 0.4 J/cm²), 70% (18.6 J/cm² ± 0.3), 73.2% (16.6 ± 0.3 J/cm²), and 76% (14.9 ± 0.3 J/cm²), respectively. The intrapulpal thermal changes induced by the 40 s light exposure of dentin adhesive through the 1.0 mm, 1.5 mm, 2.0 mm, and 2.5 mm dentin thicknesses, using the 2.0 mm, 2.5 mm, 3.0 mm, and 3.5 mm deep empty molds, are presented in Figure 1, representing the insulating effect of the dentin without the ceramic blocks and luting agents. The thermal effect of the LCU through the eight combinations of different thicknesses of dentin and ceramic assemblies without luting agents is presented in Figure 2. None of the combinations approached the critical 5.5 °C threshold. Increased dentin thickness showed a more pronounced insulating effect than ceramic thickness.

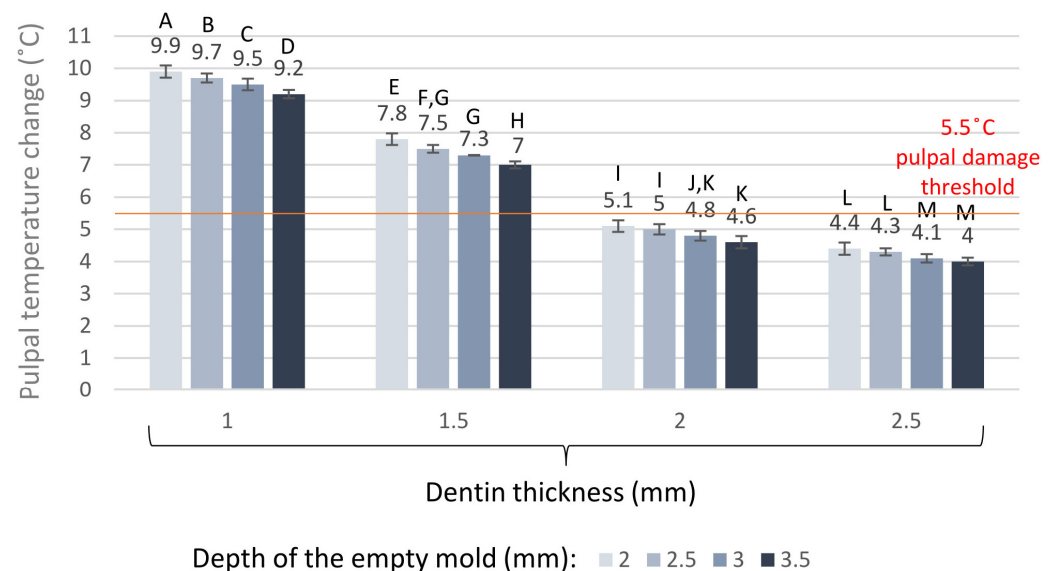


Figure 1. Intrapulpal temperature changes during light exposure of dentin adhesive through the different dentin thicknesses using the empty molds. Different capital letters (A–M) indicate statistically significant differences according to the one-way ANOVA and Tukey’s post-hoc test.

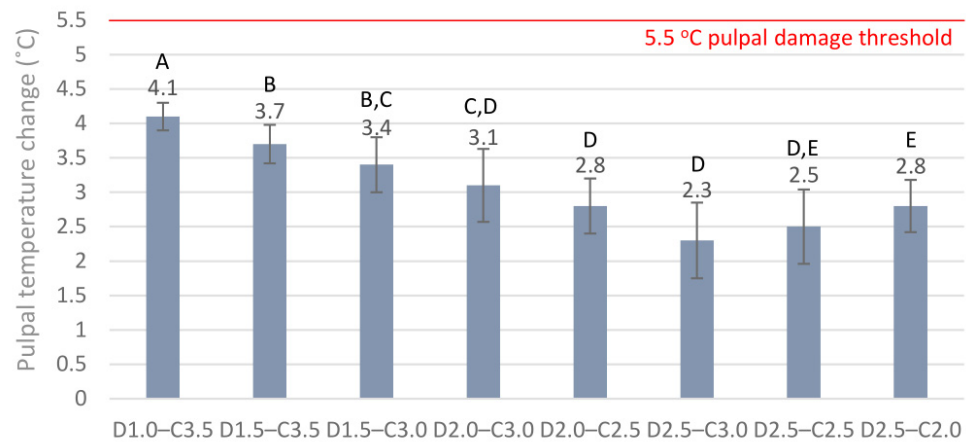


Figure 2. Pulpal temperature changes during light exposure through eight different combinations of dentin (D1.0, 1 mm; D1.5, 1.5 mm; D2.0, 2.0 mm; D2.5, 2.5 mm) and ceramic thicknesses (C2.0, 2.0 mm; C2.5, 2.5 mm; C3.0, 3.0 mm; C3.5, 3.5 mm) without luting cement. Different capital letters (A–E) indicate statistically significant differences according to the one-way ANOVA and Tukey’s post-hoc test.

Luting the ceramic blocks into cavities of different depths with light-cured and dual-cured adhesive cements, or with the restorative RBC preheated to 55 °C, increased the pulpal temperature significantly (Figure 3). Subtracting the temperature rise caused by the LCU from the thermal change in the pulp chamber induced by the luting agent provides an estimation of the heat generated by the exothermic reaction. According to this calculation, which does not account for the thermal transfer between the thermodynamic system and its environment, the preheated RBC elevated the pulpal temperature to the highest value, although a statistically significant difference was not detected between the luting materials (Figure 4).

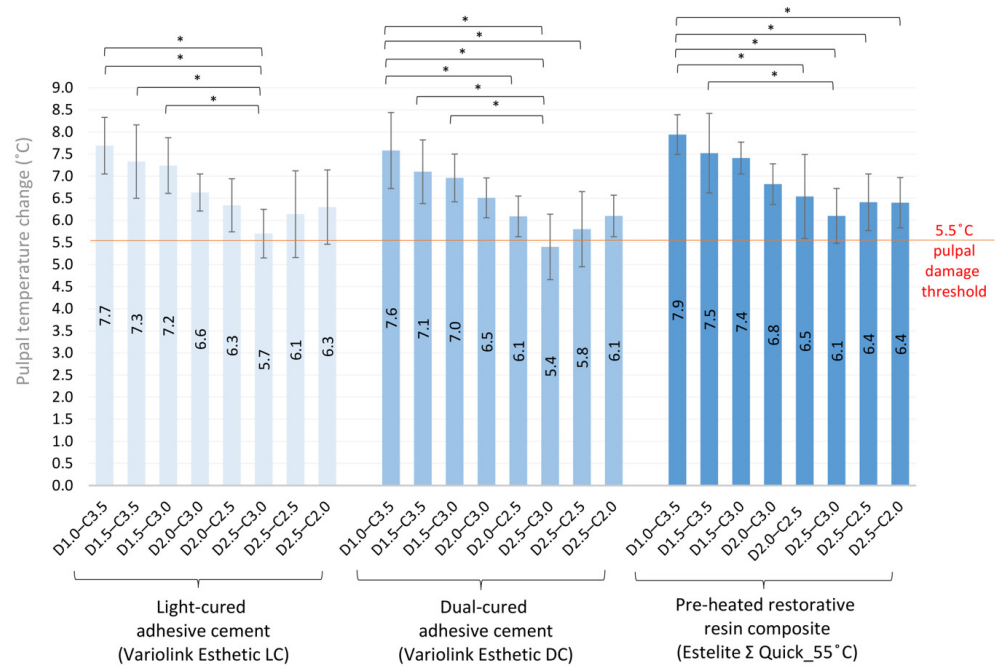


Figure 3. Pulpal temperature changes during cementation of eight different combinations of ceramic and dentin thicknesses cemented with light-cured and dual-cured adhesive cements and with the preheated restorative resin composite (* mark demonstrates statistically significant difference between groups according to the one-way ANOVA and Tukey’s post-hoc test).

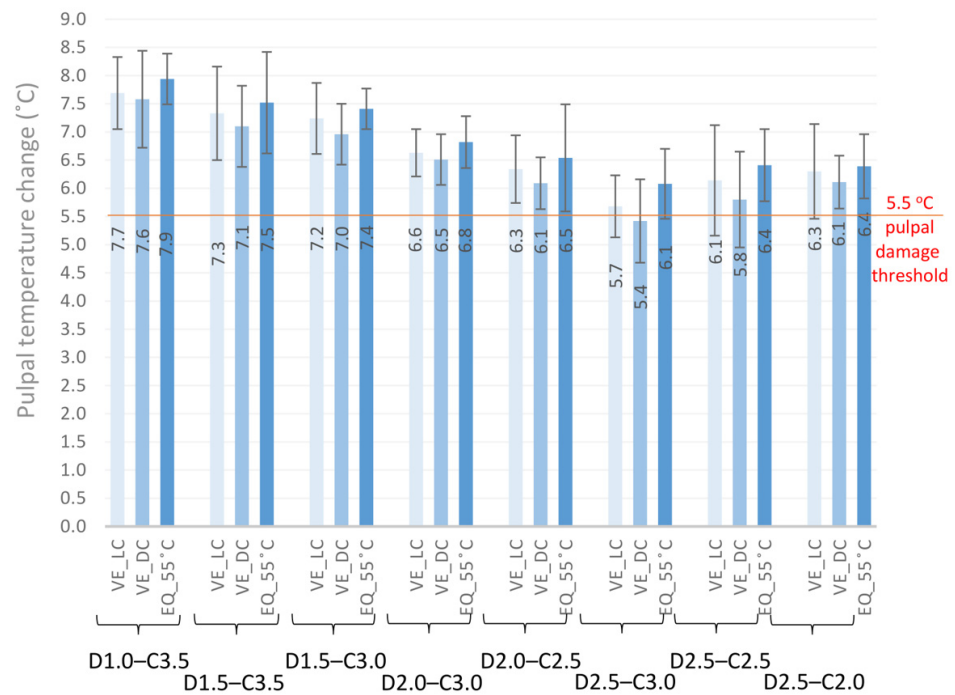


Figure 4. Comparisons of the pulpal temperature changes caused by the polymerization of light-cured and dual-cured adhesive cements and by the preheated restorative resin composite through eight different combinations of dentin and ceramic thicknesses (one-way ANOVA and Tukey’s post-hoc test did not result in statistically significant differences between the tested groups).

The mesh figure shows that the dentin thickness below 1.5 mm is the most critical for the heat insulating effect (Figure 5).

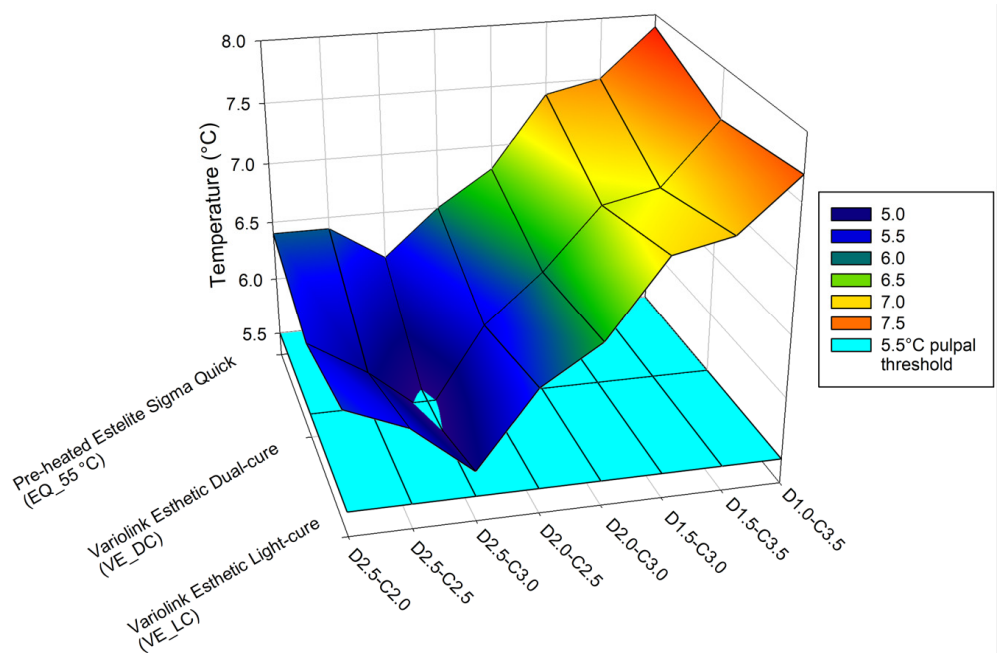


Figure 5. Changes in pulpal temperature during polymerization of the light-cured and dual-cured resin cements and preheated resin composite through different thicknesses of dentin and ceramic blocks representing an inlay. The light blue layer demonstrates the 5.5 °C temperature change thought to be associated with pulpal damage.

The multivariate general linear model revealed that the dentin thickness had the greatest effect on the pulpal temperature changes ($F(3, 96) = 6.02, p = 0.001$) followed by the effect of luting material ($F(2, 96) = 4.29, p = 0.02$). The effect size was considered to be large for the dentin thickness (partial $\eta^2 = 0.16$) and medium for the effect of material (partial $\eta^2 = 0.08$). The effect of ceramic thickness on the pulpal temperature rise was considered to be insignificant according to the results of the general linear model ($F(3, 96) = 2.28, p = 0.09$), although the eta-squared indicated a medium effect (partial η^2 was 0.07). However, there was no statistically significant three-way interaction between material, dentin thickness, and ceramic thickness [$F(14, 96) = 0.06, p = 1.0$; partial $\eta^2 = 0.009$]. According to the linear curve-fitting model, the data regarding the dentin and ceramic thicknesses and the tested luting agents allowed us to predict the behavior of the data series. The adjusted R-square statistics revealed higher values for dentin ($R^2 = 0.47$) and ceramic thickness ($R^2 = 0.41$), indicating a better fit; meanwhile, the R^2 value for the materials was 1%.

The thermal properties of the samples were evaluated by DSC measurements. The heat capacities were measured directly with the Calisto software by calculating the area of the curve, where the heat flow was plotted as a function of time until the curve reached the saturated region. Considering the mass of the samples and the area of the heating curves, the heat capacity of dentin was found to be 86% larger than the heat capacity of ceramic. To compare the heat conductivities, the time constant for the achievement of thermal equilibrium was applied. The average time constants of the dentin and ceramic samples were measured as 165.72 s and 133.33 s, respectively. The pre-exponential factors associated with the samples with increasing thickness and mass are $-1.35 \mu\text{W}$, $-4.08 \mu\text{W}$, $-6.08 \mu\text{W}$, and $-6.39 \mu\text{W}$, or $-7.13 \mu\text{W}$, $-9.13 \mu\text{W}$, $-11.38 \mu\text{W}$, and $-12.02 \mu\text{W}$ in respect to the dentin or ceramic samples. Calculating the thermal conductivity difference using the ratio of time constants, it was found to be approximately 24% lower in the case of dentin samples compared to ceramic specimens (Figure 6).

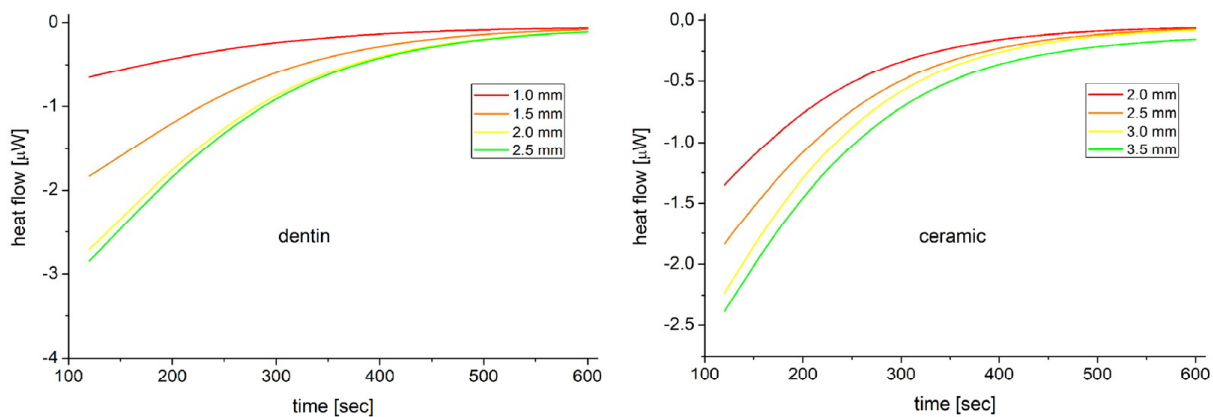


Figure 6. Heat flow associated with the dentin (left) or ceramic (right) samples plotted as a function of the time. Three scans of each sample were averaged then fitted using the exponential saturation function.

3. Discussion

In this *in vitro* study, the influence of the polymerization of light-cured and dual-cured adhesive resin cements and preheated restorative RBCs on the thermal change in the pulp chamber was investigated using different thicknesses of dentin and ceramic blocks imitating inlays. The results showed that, in the case of the investigated ceramic-dentin combinations, the cementing agents increased the intrapulpal temperature above the considered critical $5.5 \text{ }^\circ\text{C}$. Furthermore, a qualitative comparison of thermal conductivity and capacity showed differences between the dentin and the ceramic. Therefore, all the tested null hypotheses were rejected. Our findings are consistent with the results of other studies regarding the shielding effect of the interposed ceramic and the remaining dentin

thickness on pulpal temperature rise, and the material-dependent temperature-increasing effect of the polymerization of RBCs [25,26,35]. Thus, even though the interposed ceramic inlay and the remaining dentin thickness attenuate the light intensity of the curing unit and the delivered energy during the polymerization process, the exothermic temperature rise associated with the adhesive luting agent may jeopardize pulp health.

To eliminate any effects that may arise from structural differences and thermal properties of the tooth structure, this study was carried out on a representative permanent third molar without the use of acid conditioning of tooth and any ceramic surface treatment for all experimental groups. This provided the same tooth conditions for each measurement. However, the tooth differences regarding thermal changes are not accounted for in this model. Furthermore, this technique has limitations, as heat dissipation by pulpal, periodontal, and osseous circulation is not reproduced [46,47]. Thus, the temperature changes measured in this study cannot be directly applied under *in vivo* conditions. Despite the absence of blood circulation in vital tissues, this study provides important information regarding the magnitude of temperature change in a model system when a ceramic inlay is cemented in cavities of different depths using different adhesive luting agents.

In this study, a thermocouple was used for the instantaneous observation of temperature changes during ceramic inlay cementation under laboratory conditions. The use of a thermocouple is a simple and well-known method for measuring differences in temperature during dental treatments [25,26]. Compared to more modern methods, such as infrared thermography, thermocouples can measure similar values with differences of less than 1 °C in favor of infrared measurements [24].

It has been reported that dental pulp is vulnerable to temperature changes despite its high vascularization, which is the main regulatory system for heat distribution and which is capable of dissipating external thermal stimuli transferred to the dentin–pulp complex [48]. The clinical relevance of increased intrapulpal temperature is that it is a potential risk factor for thermal pulp damage. According to Zach and Cohen, a 5.5 °C temperature rise is critical and may cause irreversible pulpal damage [39]. In line with the previous experiment, Pohto and Scheinin reported that the critical temperature for reversible pulp damage was between 42 °C and 42.5 °C [49]. Although the pathological threshold of pulpal temperature rise was determined to be approximately 5.5 °C, a more recent study did not find an average increase of 11.2 °C to compromise pulp health significantly [50]. However, a histomorphometric analysis using a clinically valid 3-dimensional organotypic *ex vivo* model showed an immediate reduction in cell number with a temperature increase of 5.5 °C or greater, which was dependent on exposure time [40]. Additionally, immunohistochemical changes were observed at a temperature increase of 6 °C or higher [40].

Despite attempts to simulate *in vivo* conditions using several reliable methods, a wide range of intrapulpal temperature increases has been reported *in vitro* during photocuring [27]. To provide the blue light required for the polymerization of resin-based luting agents, a second-generation LED LCU was used in this study with a radiant exitance of 1550 ± 10 mW/cm² in the wavelength range of 420–480 nm. According to previous studies, the intensity and duration of the applied light were the most crucial factors for the pulpal temperature rise [51,52]. The results of this study confirm the above statement because light curing without the interposition of a ceramic inlay and resin-based luting agent increased the pulpal temperature ($\Delta T = 4.0$ – 9.9 °C) by a significant degree, depending on the dentin thickness, with an inverse correlation. Although a thin layer of dentin adhesive was used during light curing without ceramic blocks and luting agents, the heat measured was predominantly from the curing unit. A pilot study was undertaken to assess the thermal effect of the dentin adhesive layer, and a negligible, insignificant (0.1–0.2 °C) temperature difference was detected during light curing with or without the adhesive.

Because the monomer-to-polymer conversion of an RBC is a function of the applied total energy during photocuring, it is advisable to increase the delivered radiant exposure for a higher degree of conversion [53]. This is highly relevant to adhesive luting of indirect restorations, where a high radiant exposure is needed for proper curing of the luting resin-

based material underneath a certain thickness of ceramic [34]. The reciprocal relationship between the power output and exposure duration provides an opportunity to increase either irradiance or exposure time, resulting in a higher delivered radiant exposure [53]. It was affirmed that extended irradiation has a greater effect on the depth of cure than increasing the light irradiance of the curing unit [54]. While the increased delivered radiant exposure is indispensable to the acceptable polymerization of the adhesive luting agent, a strong positive correlation was found between the radiant exposure and the intrapulpal temperature rise [26,55]. In this study, an extended exposure time (40 s) with a light irradiance of 1550 mW/cm² resulted in 62 J/cm² of delivered radiant exposure. Onisor et al. [36] investigated the effect of extended exposure times on intrapulpal temperature increase through 1 mm of the remaining dentin thickness, interposing a 3 mm ceramic onlay and 0.3 mm of previously polymerized RBC luting agent. They found a maximal temperature difference of 4.3 °C during the 3 × 20 s of extended irradiation, delivering ~60 J/cm² of total energy. Although the study design and the type of curing units are slightly different, these results are in line with our findings with a similar D1–C3.5 combination, where the intrapulpal temperature rise was 4.1 °C without the exothermic reaction of the luting agent and the delivered energy density was 62 J/cm². In addition to energy density, other characteristics of the photocuring unit affect the amount of heat generated within the pulp [36,46,56]. While the pulpal temperature in this study remained below the critical value of 5.5 °C without luting agents, the tested adhesive cements significantly increased it because of their exothermic reaction. Our study design ensured uniform conditions, allowing the comparison of exothermic thermal changes between different resin-based luting agents. The exothermic reaction was proportional to the amount of resin matrix, and it was found that the inorganic fillers have an impact on heat diffusion within the material by their capacity to absorb external and internal energy [26,57,58]. The multivariate general linear model revealed that the material factor had a medium impact on the pulpal temperature rise. A tendency was observed when comparing the effects of the tested luting agents on temperature change, although there was no detectable statistically significant difference among the study groups. The highest temperature was measured with the preheated RBC in all the tested groups, followed by the light-cured adhesive cement, and the lowest temperature increase was observed with the dual-cured luting agent. Although the restorative RBC was preheated to 55 °C, it only increased the pre-polymerization temperature of the pulp by ~3 °C, and further cooling was detected during the cementation procedure before light curing. After removal from the warming device, the preheated RBC was reported to suffer from rapid cooling, which may compromise the degree of conversion [16]. Although the temperature-raising effect was less than expected owing to the rapid cooling and heat absorption by the ceramic and dentin, these results suggest the potential hazard to pulp health owing to their higher thermal effect. Regarding the composition of the investigated light- and dual-cured resin cements, they had the same resin matrix/filler ratio; however, the dual-cured cement showed a slightly, but not significantly, lower temperature rise in each tested group compared to that of the light-cured resin cement. Dual-cured resin cements are supposed to compensate for decreased light transmission and may be more efficient at monomer-to-polymer conversion, even with increased ceramic thicknesses [59,60]. In their systematic review, David-Pérez et al. found that dual-cured resin cements fail to achieve the same degree of conversion as light-cured cements with up to 2 mm of interposed ceramic thickness [61]. Our results are indirectly in line with these findings because the polymerization process is proportional to the exothermic reaction, which results in a slightly lower pulpal temperature rise in the dual-cured resin cement [57].

Thermal transfer to the pulp is strongly dependent on the thickness of the remaining tooth structure [35]. The thermal conductivities of enamel and dentin are ~0.81 W/mK and ~0.48 W/mK, respectively, which are considered low [62]. Low thermal conductivity is equivalent to high insulating capability; thus, the pulp is protected from noxious thermal irritation if the tooth is intact [62]. The advantageous thermal conductivity may be explained

by the porous tubular microstructure in the dentin layer, which is a mineralized connective tissue with an organic matrix of collagenous proteins [33]. However, during cavity preparation for direct or indirect restorations, the enamel and DEJ are partially removed, and the dentin thickness is reduced according to the extent of the caries or depending on the special cavity design. Hard tissue removal during cavity preparation and several steps of the adhesive restorative procedure (i.e., polymerization of the adhesive layer and the RBC/adhesive luting agent, and the polishing procedure) may cause thermal damage due to the weakened thermal insulation effect, especially in younger patients with wider dentinal tubules [63,64]. The present study investigated four thicknesses (1.0 mm, 1.5 mm, 2.0 mm, and 2.5 mm) of the remaining dentin layer. For dentin thickness below 2.0 mm, the heat transmitted by the curing unit increased the intrapulpal temperature above the 5.5 °C limit. These results are consistent with previous finding that showed a strong relationship between the thickness of the dentin and the intrapulpal temperature increase [29]. Similar to our findings, Kuo et al. concluded that there is a risk of damaging the pulp when the dentin thickness is less than 2.0 mm and the overall thickness of the dentin–ceramic assembly is less than 3.5 mm [35]. However, according to other studies, the dentin thickness (0.5 vs. 1.0 mm) had an insignificant role in the pulpal temperature increase compared to the curing unit type [28]. In addition to the dentin insulating effect, the already polymerized thin dentin adhesive may serve as a further protective layer during the cementing procedure. However, our pilot study showed that the thermal insulating effect of dentin–ceramic assemblies is not affected by the presence or absence of a polymerized adhesive layer. In the present study, partial eta-squared statistics revealed that, among the investigated factors—such as dentin thickness, ceramic thickness, and the type of luting material—the remaining dentin layer had the most pronounced effect on the pulpal temperature change values, and the effect size was considered to be large. The linear regression model revealed a 47% value for the coefficient of determination and predicted a decreasing effect of thickness on temperature as the dentin thickness exceeded 2 mm. Even though the thinnest dentin was combined with the thickest ceramic, the highest intrapulpal temperature was detected in all measurements, regardless of the use of the luting agents.

In contrast, the effect of ceramic thickness on pulpal temperature rise was considered insignificant according to the results of the general linear model, although the partial eta-squared indicated a medium effect. This result demonstrates that the shielding effect of the ceramic is not as great as that of dentin, although an inverse relationship between the ceramic thickness and temperature rise is evident, which is consistent with the findings of a previous study [35]. The regression curve fit for temperature as a function of different ceramic thicknesses showed a linear energy loss with increasing ceramic thickness, which reflects the light attenuation occurring through an absorptive/scattering medium. The value of the coefficient of determination for ceramic thickness was found to be 41% in our linear regression model. The differences in temperature changes caused by the thermal shielding effect of dentin and ceramic can be explained by their distinct thermal conductivities. It is higher for silica-based ceramics, which is approximately 1.7 W/mK, compared to the thermal conductivity of the dentin (~0.48 W/mK) [65]. Increasing the inlay thickness may result in a proportional removal of tooth hard tissues by decreasing the thickness of the remaining enamel and dentin. Although the shielding effect is proportional to increasing ceramic thickness, a decreasing dentin thickness has a stronger inversely proportional effect on the temperature increase within the pulp chamber. These results are supported by the DSC measurements in this study, which revealed a 24% lower thermal conductivity of the dentin than that of the ceramic. However, the thermal capacity of the dentin was calculated to be 86% more compared to the investigated lithium disilicate ceramic. The importance of this result lies in the ability of dentin to store large amounts of heat and then dissipate it slowly, reducing the sudden thermal effects on the pulp. However, considering the multiple thermal effects during adhesive restorative treatment, the gradually increasing temperature of the dentin may conduct more heat towards the pulp during heat dissipation. According to these findings, it is advisable that more dentin should be preserved during

cavity preparation to protect the pulp from undesirable temperature increases. Furthermore, it should provide more time for heat dissipation between treatment steps, which can have a thermal effect on the pulp, to avoid heat accumulation in the dentin.

4. Materials and Methods

4.1. Resin-Based Luting Agents, Ceramic Blocks, and Radiant Exposure

In this *in vitro* study, the effects of three resin-based luting agents—Variolink Esthetic LC (VE_LC) light curing, Variolink Esthetic DC (VE_DC) dual-curing adhesive resin cement, and preheated sculptable submicron filled restorative RBC, Estelite Sigma Quick (EQ_55 °C)—on pulpal temperatures were analyzed. The brands, manufacturers, and chemical compositions are listed in Table 1.

Table 1. Materials, manufacturers, classification, and composition of the investigated adhesive resin cements and preheated resin-based composites.

Material (Code)	Shade	Manufacturer	Classification	Resin System	Filler	Filler Loading
Variolink Esthetic LC (VE_LC)	Light	Ivoclar Vivadent, Schaan, Liechtenstein	Light-curing adhesive resin cement	UDMA; 1,10-DDMA	0.04–0.2 µm ytterbium trifluoride and spheroid mixed oxide	38 vol% 64 wt%
Variolink Esthetic DC (VE_DC)	Light	Ivoclar Vivadent, Schaan, Liechtenstein	Dual-curing adhesive resin cement	UDMA; 1,10-DDMA	0.04–0.2 µm ytterbium trifluoride and spheroid mixed oxide	38 vol% 64 wt%
Estelite Sigma Quick (EQ_55 °C)	A1 enamel	Tokuyama Dental, Tokyo, Japan	Conventional submicron RBC preheated to 55 °C	BisGMA, TEGDMA	0.1–0.3 µm monodispersing spherical silica–zirconia filler; prepolymerized filler of silica–zirconia and copolymer	71 vol% 82 wt%

Abbreviations: RBC: resin-based composite; BisGMA: bisphenol-A diglycidil ether dimethacrylate; UDMA: urethane dimethacrylate; 1,10-DDMA: 1,10-dodecane dimethacrylate; LC, light-cure; DC, dual-cure; vol%, volumetric %; wt%, weight %.

Highly translucent A2 shade lithium disilicate ceramic blocks (6 × 6 mm) were fabricated from ceramic ingots (GC Initial LiSi Press; GC Europe, Leuven, Belgium) using the heat-pressed method and were then fired and glazed from one side according to the manufacturer's instructions. To achieve an even smoother surface, 220-, 400-, and 600-grit water-cooled sandpaper was used to finish the specimens, followed by polishing with a two-step rubber diamond polisher (fine, 8–32 µm grit size, Kenda Nobilis, Kenda AG, Vaduz, Liechtenstein; extra fine, 4–8 µm grit size, Kenda Unicus, Kenda AG, Vaduz, Liechtenstein). The fabricated ceramic blocks were intended to represent inlays with thicknesses of 2.0 mm, 2.5 mm, 3.0 mm, and 3.5 mm. The final dimensions of each ceramic block were determined using a digital caliper with an accuracy of 0.001 mm (Mitutoyo, Tokyo, Japan). To provide multiple measurements, the ceramic specimens were not acid-etched, silanated, or coated with an adhesive.

The ceramic blocks were cemented with light-cured adhesive cement, dual-cured adhesive cement mixed with an Automix syringe, and a preheated restorative RBC. A single-dose capsule of the latter RBC was preheated to 55 °C in an RBC warming device (Ena Heat Composite Heating Conditioner, Micerium, Avegno, Italy) for 15 min. Each capsule was heated once for the cementation of only one ceramic specimen. The resulting RBC temperature was measured using a non-contact infrared digital thermometer (TESTO 845, Testo Magyarország Kft., Budapest, Hungary). The infrared thermometer registered temperatures in an area as small as 1 mm² (optical resolution of 75:1), with a resolution of 0.1/1 °C. The data sampling frequency was 10 measurements/s. The ceramic blocks were pre-warmed in a composite warming device to reduce heat dissipation during cementation.

During each cementation a light-emitting diode (LED) light curing unit (LCU) (LED.D, Woodpecker, Guilin, China; $\lambda = 420\text{--}480\text{ nm}$; 8 mm exit diameter fiberglass light guide) was used in the standard mode for 40 s of exposure time. The LCU was powered by a line cord at room temperature (24 °C ± 1 °C). The position of the light guide tip was standardized to

ensure that each sample received the same light beam character. A radiometer checkMARC radiometer (BlueLight Analytics, Halifax, NS, Canada) was used to monitor the radiant exitance (mW/cm^2) of the LCU before and after exposure. The tip of the LCU was placed at a standard distance of 1 mm from the radiometer sensor.

4.2. Sample Preparation for Pulpal Temperature Measurements

A caries-free, freshly extracted, human mandibular third molar for use in this study was cleaned and kept in physiological saline at room temperature. All thermal measurements were performed on a single-tooth model to limit any effects of structural differences in the dental hard tissues [25]. The apices of the roots were cut 5 mm from the furcation to expose the root canals, and all the pulpal residues were removed with an endodontic file, which was followed first by irrigation with 5.25 weight% sodium hypochlorite solution (Chloraxid, CerkaMed, Stalowa Wola, Poland) and then saline (NaCl 0.9%, B. Braun, Melsungen, Germany), and was finally dried with paper points (DiaDent, Burnaby, BC, Canada). A hole was prepared on the mesial side of the tooth with a cylindrical diamond bur (836-012-FG cylinder diamond bur, 1.2 mm, medium; Meisinger USA, Centennial, CO, USA) to allow the insertion of the 0.5 mm diameter Cu/CuNi thermocouple probe (Type K thermocouple device; $\varnothing = 0.5$ mm; Cu/CuNi; TC Direct, Budapest, Hungary). The thermocouple sensor was positioned on the dentin at the top of the pulp chamber and assessed radiographically. To replicate the pulp tissue, the pulp chamber and root canal were injected with ECG gel (Aqua Sound Basic, Ultra-gel Hungary 2000, Budapest, Hungary). A flowable RBC (Filtek Supreme Flowable, 3M, St. Paul, MN, USA) was used to close the mesial hole and apical orifice, and the tooth was embedded in clear acrylic 1.0 mm below the cemento-enamel junction. The occlusal surface was prepared and polished flat, leaving dentin with a 2.5 mm thickness from the top of the pulp chamber. The occlusal thickness to be removed was estimated and controlled using digital intraoral radiography. To provide standard conditions during radiographic control of dentin reduction, the acrylic holder of the tooth was inserted in a poly-vinyl siloxane (Aquasil Ultra Plus, Dentsply Sirona, Charlotte, NC, USA) holder which was able to maintain the standard distance between the digital sensor and tube and provided correct position of the tooth for the parallel radiographic positioning technique (Figure 7).

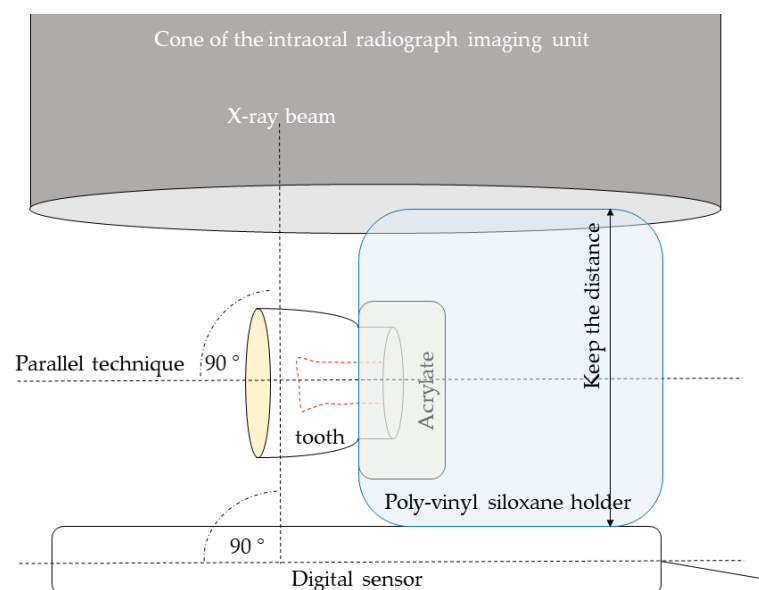


Figure 7. Schematic of the experimental set-up for radiograph taking during dentin thickness reduction.

After implementation of the first series of measurements with the different thicknesses of ceramic specimen and luting agent combinations, the dentin was reduced by 0.5 mm, resulting in a 2.0 mm dentin thickness. Following the conduction of the second series of measurements, the dentin thickness was further reduced by 0.5 mm, leaving behind 1.5 mm of dentin above the pulp chamber. For the last series of temperature registration, the dentin thickness was 1.0 mm thick after further reduction of 0.5 mm. The thickness of the remaining dentin was radiographically assessed. A cylindrical polytetrafluoroethylene (PTFE) mold with a crown diameter of ~12 mm and thicknesses of the four ceramic plates, with an inner hole of 6 × 6 mm, was fabricated to represent the axial walls of the cavity. The sample tooth was isolated with a rubber dam (Rubber Dam, Cerkamed, Stalowa Wola, Poland) supported by a frame and immersed in a water bath at 36.0 ± 0.5 °C. Temperature measurements were recorded using a digital thermometer (El-EnviroPad-TC, Lascar Electronics Ltd., Salisbury, UK) attached to the above-described thermocouple, with a resolution of 0.1 °C and a frequency of one measurement per second.

First, the intrapulpal temperature changes during light exposure (40-s exposure) of dentin adhesive through the different dentin thicknesses using the empty molds were measured. Prior to dentin adhesive (Adper Single Bond 2, 3M ESPE, St. Paul, MN, USA; without acid conditioning) application, an agar/alcohol solution (3 wt% agar dissolved in 1:1 alcohol/water) was applied on the prepared dentin surface, allowing the polymerized adhesive/luting agent to be removed without remnants. Thereafter, the thermal effect of the LCU through the eight combinations of different thicknesses of dentin and ceramic was recorded using the digital thermometer. Temperature changes during cementation with the three resin-based luting agents were also measured (Figure 8).

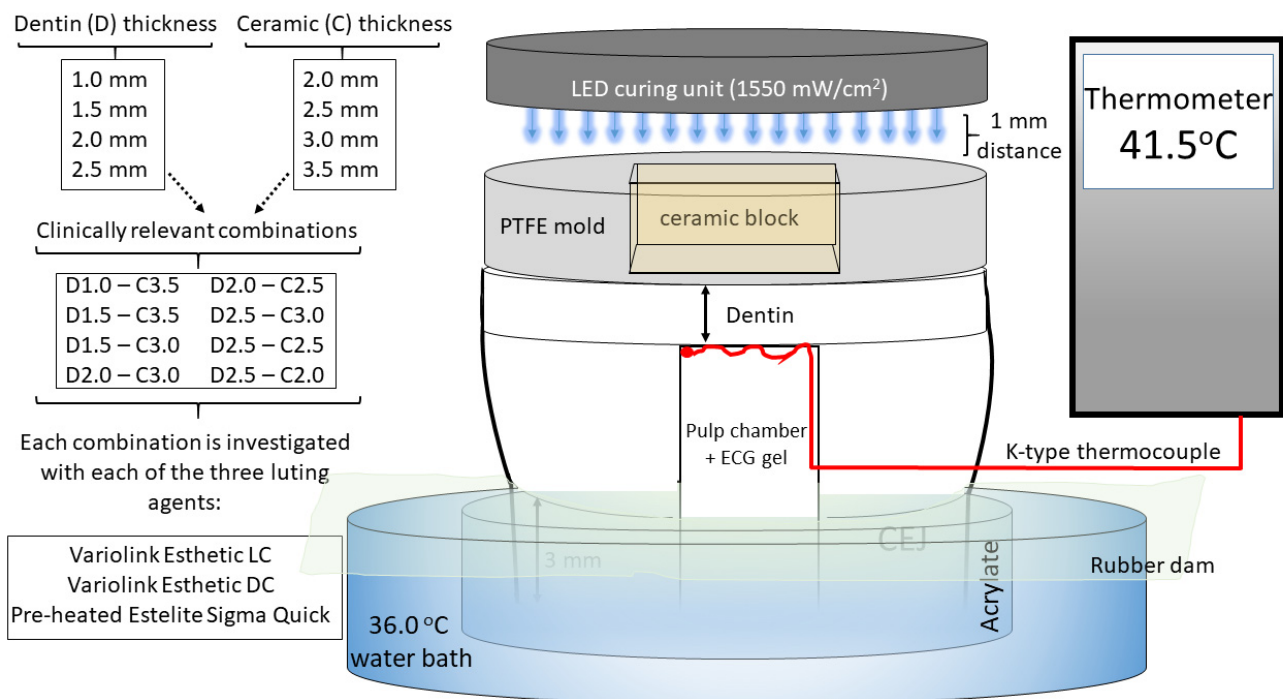


Figure 8. Schematic of the experimental set-up for pulpal temperature measurements.

A standard volume of the luting agent was applied to the center of the non-glazed ceramic surface, which was then centrally oriented to the tooth surface and manually (using the Optrasculpt instrument; Ivoclar Vivadent, Schaan, Liechtenstein) loaded with a 5-N load when resin adhesive cements were used and a 10-N load when the preheated restorative RBC was applied. Preliminary test results showed that manual placement of the ceramic specimen with a load of 5 or 10 N, depending on the type of luting agent, achieved a consistent layer thickness of 100 ± 10 μm. The load was measured using an

algometer (Force Dial FDK 16, Wagner, Greenwich, USA). A microbrush was used to remove excess luting agent, which was photoactivated through the ceramic block for 40 s. After the cemented specimen was removed, its thickness was measured using a digital caliper. The luting agent film thickness was calculated from the difference between the thicknesses of the cemented specimen and the ceramic block alone. There were 24 groups of the various dentin/ceramic/luting agent combinations, and the temperature measurements were recorded five times for each group ($n = 120$).

As separating solution was used on the tooth surface and no ceramic surface treatment was applied, the polymerized luting agent could be removed from both the tooth and the ceramic block following the measurements, without leaving any deposits on either surface.

4.3. Differential Scanning Calorimetric (DSC) Measurements

A new series of the above-described ceramic blocks with thicknesses of 2.0 mm, 2.5 mm, 3.0 mm, and 3.5 mm were used for microcalorimetric measurements. Dentin slices were obtained from three third molars of a patient who was 19 years old. The teeth were sound, with no cracks or caries. The cleaned teeth were stored in saline at room temperature before testing, which took place within two days after extraction. To obtain dentin slices, the occlusal surface of the crown was horizontally removed using an orthodontic model trimmer (Gamberini, Bologna, Italy) under a water coolant. A minimum of 4 mm of dentin remained without exposing the pulp chamber. Dentin specimens with thicknesses of 1.0 mm, 1.5 mm, 2.0 mm, and 2.5 mm were sectioned using a precision cutter (IsoMet Low Speed Precision Cutter, Buehler, Lake Bluff, IL, USA) under a water coolant perpendicular to the long axis of the tooth. The enamel was removed to ensure that all sections contained only dentin, with final dimensions of 6×6 mm. The final dimensions of each dentin slice were measured using a digital caliper. Both ceramic and dentin specimens were weighed (ML-T Precision Balances, Mettler-Toledo, Greifensee, Switzerland).

Microcalorimetric measurements were carried out using a MicroSC microcalorimeter (Setaram Instrumentation, Caluire, France) in the differential scanning mode. Data were evaluated using the Calisto thermal analysis software (Setaram Instrumentation, Caluire, France). During the measurements, the samples were first equilibrated thermally for 5 min at 20 °C; then, the temperature was switched to 40 °C. The heat flow was recorded and plotted against time within 0–10 min. Considering that the sample holder itself needed 25 s to reach 40 °C after equilibration at 20 °C, data collected after 2 min were used for evaluation. Data evaluation was based on the following assumption: convective heat transfer between the sample holder of the microcalorimeter equipment and the samples occurred through the surface of the samples. The sample with higher heat conductivity requires less time to reach the thermal equilibrium at 40 °C. Accordingly, the data collected within the time range of 2–10 min were applied to estimate the heat conductivity, whereas the data collected within the time range of 0–10 min were applied to determine the heat capacities. The rate of the development of the thermal equilibrium at 40 °C was applied to compare the heat conductivities of the ceramic and dentin samples. The formation of thermal equilibrium was described by fitting the following equation (Equation (1)) to the measured data:

$$\text{heat flow} = A \times e^{-\frac{t}{\tau}} \quad (1)$$

where A is the pre-exponential factor (proportional to the heat capacity); t is the time required; τ is the time constant describing the rate of formation of thermal equilibrium; and e is the base of the natural logarithm. Measurements were performed in triplicate for each sample.

4.4. Statistical Analysis

An earlier pilot study and a sample size formula were used to estimate the sample size [66].

$$\text{The sample size formula is as follows: } n = \frac{(z_{1-\frac{\alpha}{2}} + z_{1-\beta})^2 (s_1 + s_2)^2}{(M_1 + M_2)^2}$$

where z = standard score; α = probability of Type I error = 0.05; $z_{1-\alpha/2} = 1.96$ for 95% confidence; β = probability of Type II error = 0.20; $1 - \beta$ = the power of the test = 0.80; $z_{1-\beta}$ = value of standard normal variate corresponding to 0.80 value of power = 0.84; s_1 = standard deviation of the outcome variable of group 1 = 0.64; s_2 = standard deviation of the outcome variable of group 2 = 0.36; M_1 = mean of the outcome variable of group 1; M_2 = mean of the outcome variable of group 2; and $(M_1 - M_2) = 0.5$, if it is expected to detect a 0.5 °C difference between the two investigated groups as significant. Using the formula $N = \frac{2n}{1-0.1}$ the predicted sample size (n) was found to be 4.7 samples per group. According to the calculations, $n = 5$ per group sample size was indicated.

The SPSS v. 26.0 (SPSS, Chicago, IL, USA) software was used to perform statistical analyses. The normality of the data distribution was tested using the Kolmogorov–Smirnov test, followed by the application of parametric statistical tests. The differences in temperature changes were compared using one-way analysis of variance (ANOVA). Tukey's post hoc adjustment was used for multiple comparisons for all the ANOVA models. To evaluate and explain the relative effect size on dentin and ceramic thicknesses, as well as the luting material as the independent variable, a general linear model (multivariate analysis) and partial eta-squared statistics were applied. Linear regression was used to determine the correlation between the dependent (temperature) and independent (dentin thickness, ceramic thickness, and luting material) variables. The statistical significance was set at $p < 0.05$.

5. Conclusions

The intrapulpal temperature rise may exceed the critical 5.5 °C threshold during ceramic inlay cementation, regardless of the dentin and ceramic thicknesses and the type of resin-based adhesive luting material used. The temperature values were predominantly influenced by the remaining dentin thickness, followed by the applied resin-based adhesive luting materials, and were least influenced by the ceramic thickness. The thermal conductivity of the dentin was 24% less compared to that of the ceramic, while the thermal capacity was 86% higher.

In order to provide safe dental treatment, a more conservative approach with more dentin preservation should be followed when it comes to the thermal effect applied to teeth during ceramic inlay cementations.

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