

Effect of Polymerization and Type of Glass-Ionomer Cement on Thermal Behaviour

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Abstract

The purpose of this study was to evaluate the effect of type of glass-ionomer cement (GIC), extrinsic energy offer and light intensity of the curing device, on thermal behavior. Ketac Molar, 3M-ESPE, USA (KM), Diamond Rapid Set Capsules, Kemdent, Wiltshire, UK (D) and Ketac N100, 3M-ESPE, USA (KN), being all different types of GIC.

Measurements of coefficient of thermal expansion (CTE) were performed at 20-60°C and thermal behavior was evaluated in lower and higher temperatures. KM exhibits lower CTE values ($p < 0.001$). D exhibits significant differences ($p < 0.001$), showing greater CTE values. KN exhibits great differences ($p < 0.001$) in higher temperatures. Qualitative differences were also noted in CTE diagrams, diversifying between expansion and contraction, as temperature raised. Type of GIC and light intensity of curing device affect thermal behavior. Extrinsic energy offer had no important effect on CTE values.

Keywords: Coefficient of Thermal Expansion, Glass-Ionomer Cements, Dilatometer, Temperature, Polymerization.

1. Introduction

Wide thermal fluctuations in the oral environment are of dietary origin. During the consumption of hot food and drink, maximum tooth surface temperature is approximately 47°C. Temperature ranges between 0-67°C have also been reported, but they are considered unrealistic [6, 17, 18]. Such fluctuations imply corresponding volumetric increases and decreases, undermining mechanical properties [14]. Thermally induced loads continuously introduced to restored teeth are corresponded with marginal degradation and microleakage [2, 8, 12, 19, 25, 29]. The amount of deformation due to thermal loading is given by the coefficient of thermal expansion (CTE). The CTE is an inherent characteristic of each material at a specific temperature. When dealing with thermally induced volumetric changes, comparison of CTE values of the restorative material and the tooth substance is more important than the CTE value of the material itself [27]. When two materials expand or contract at a similar rate, gap formation at the interface is almost a nonissue, thus microleakage is negligible [2, 19].

1.1. CTE values

Many methods have been used to measure CTE values, such as using a thermomechanical analyzer, biomaterial strip, strain gauges, capillary mercury tubes, or dilatometers, and the research is done mainly on composite resins [7, 19, 21 – 23, 27]. The CTE of enamel is approximately $17 \times 10^{-6}/^{\circ}\text{C}$ and $11 \times 10^{-6}/^{\circ}\text{C}$ for dentine [8]. For composite resins, the CTE ranges from $20\text{-}80 \times 10^{-6}/^{\circ}\text{C}$ [25, 27], while for glass-ionomer cements (GIC), CTE values reported in the literature range from $11 \times 10^{-6}/^{\circ}\text{C}$ but this value may increase to -286 to $-165 \times 10^{-6}/^{\circ}\text{C}$ when temperatures exceed 50°C [9, 24]. Large variations of GIC CTE values, impose an issue of further research.

1.2. Glass-Ionomer Cements (GIC)

GICs are described as “smart materials” with respect to their thermal behaviour, since it is a desired feature, when restorative materials undergo thermally induced volumetric changes close to those of the tooth substance [27, 30]. GICs’ clinical indications are expanding [26] and they are mainly used in low load bearing situations. They can be either directly exposed to thermal conditions of the oral cavity as restoratives, or indirectly exposed when used as base materials. However, temperature changes also influence the thermal behaviour of base materials with the main restorative material being the thermal conductor [4]. In order to overcome brittleness and surface wear, considerable attention has been directed at improving GICs’ physical properties by introducing modified GICs [5, 15]. Modified GICs with altered acid compositions are designed as restorative material, indicated by the manufacturer for Class I and II restorations, build-up fillings and linings, core build-up, and retrograde root fillings. However, there is no previous research focussing on restorative GIC physical properties and only the orthodontic bonding of GICs with altered acid composition is evaluated in the literature [3, 11].

1.3. Purpose

The purpose of this study was to compare the CTE values of 3 types of GICs. Since polymerization of conventional GICs supposedly impacts microhardness values [16], the present study is also focused on the effect of extrinsic energy offerings through polymerization of conventional GICs on CTE values. The null hypothesis was that the same CTE values are exhibited by all types of GICs tested, in lower and higher temperatures, when polymerized with a low or high intensity light curing unit and with or without energy offering through polymerization.

2. Materials and Methods

A conventional GIC (Ketac Molar, 3M-ESPE, USA, LOT: 323615) (KM), a conventional GIC with altered acid composition (Diamond Rapid Set Capsules, Kement, Wiltshire, UK, LOT: 0948042) (D) and a resin-modified GIC (RMGIC) (Ketac N100, 3M-ESPE, USA, LOT: N166321) (KN) were used. Eight specimen 4mm thick were made from each material by placing the material into an open-ended stainless-steel mold and were then divided into two groups. Groups were: Ketac Molar not cured (KMNC), Ketac Molar cured (KMC), Diamond not cured (DNC), Diamond cured (DC), Ketac N100 polymerized with a lower light intensity LED curing device (KNL), Ketac N100 polymerized with a higher light intensity LED curing device (KNH).

2.1. Sample preparation

Each material was prepared according to manufacturer’s instructions and divided in groups mimicking their clinical use concerning variations in polymerization modes. Polymerization modes were cured and not cured for conventional GICs and cured with low and with high intensity curing unit, for RMGIC. GICs were allowed to set before removal from the mold. RMGIC specimens were polymerized over a thin cellulose strip. Higher light intensity LED was Flash max (CMS Dental, Copenhagen, Denmark) (3’’ curing time, $>4000\text{mW}/\text{cm}^2$) and lower light intensity LED was Radium plus (SDI, Bayswater, Australia) (20’’ curing time, $1500\text{mW}/\text{cm}^2$). Samples were stored in distilled water 37°C for 24h before CTE testing.

2.2. Measurements

Dilatometer DIL 402C (NETZSCH) was used to determine the values of CTE of composite resin materials at temperature range 20 – 60°C. Temperature range was divided into lower temperatures (21-37°C) and higher temperatures (37-55°C), regarding 37°C (body temperature) as division point. Thus thermal behavior was evaluated at two temperature groups. Extreme low and extreme high temperatures were excluded in order to attain unbiased results, by eliminating possible material shock. The probe of the dilatometer was calibrated with standard Al₂O₃ cylinders in the beginning. Each specimen was held horizontally in a chamber and volumetric changes were detected by a displacement transducer. Temperature raised at a slow rate of 2°C/min in order to obtain uniform distribution. CTE was calculated using internal software and diagrams were extracted which reproduce each material's thermal behavior. CTE was internally calculated using the following formula:

$$A = \Delta L / L_0 \times \Delta T,$$

where ΔL represents the probe displacement when temperature change is ΔT and L_0 corresponds to initial specimen length. $\Delta L / L_0$ represents linear shrinkage.

Thickness is measured for every specimen at 4mm and as a result volumetric changes were calculated by changes in specimen length. Statistical analysis was performed using Tukey-Kramer Multiple Comparisons test.

3. Results

Mean values ($\times 10^{-6}$) and standard deviations were as follows: for 21-37°C: KMNC (-8.8, 6.8), KMC (-10.7, 3.9), DNC (-11.7, 5.5), DC (-13.7, 3.9), KNL (17.3, 4.1), KNH (6.4, 4.1) and for 37-55°C: KMNC (-19.9, 7), KMC (-17.8, 12.6), DNC (-28.5, 11.1), DC (-55.9, 20.3), KNL (36.9, 5.3), KNH (29.2, 4.1). Values of CTE and % linear shrinkage are demonstrated in Figures 1-6 where qualitative differences can also be noted, expressed by mathematical signs (plus / minus) accompanying CTE values and diversifying between expansion and contraction. Quantitative differences issued from the statistical analysis, are shown in Table 1 for all material groups and for the two temperature groups.

4. Discussion

The null hypothesis of this study was partially accepted. Differences in CTE values were exhibited among different types of GICs, in lower and higher temperatures, when polymerized with a low or high intensity light curing unit. But energy offerings in conventional GICs through polymerization did not exhibit significant differences in CTE values (Table 1).

4.1. Why GIC contract?

Most materials expand with temperature increases. It was observed in this study that conventional GICs contract in a non-linear way (Figures 1 and 2) with temperature increases, as reported in the literature [23, 30]. This takes place in dry conditions [30] and could be attributed to volatile component removal (such as solvents and moisture) with heat [23] or to molecular or atomic re-arrangement in the solid material after heat energy absorption, finally resulting in material contraction. The GIC matrix is thought to contain "tightly" bound water and "loosely" bound water, the latter easily lost or regained through a reversible process [28]. shrinkage is noted when contraction due to water loss and thermal expansion due to temperature fluctuations occur, provided that the matrix structure of the material is flexible enough to allow these to take place [23]. However, the role of fluid in GIC porosities is not fully understood, as fluid lost from a rigid material does not always lead to significant dimensional changes [30]. Kim *et al.* 1998, on the other hand, showed that GICs expand with temperature increases.

4.2. Thermal behaviour of GIC with polyacrylic acid

We can also add, that even conventional GICs with altered acid compositions (from polyalkenoic acid to polyacrylic acid, groups DNC and DC) contract with temperature increases. This contraction is greater under higher temperatures; therefore, alterations in acid composition exhibit worse thermal behaviour when temperatures are greater.

However, Diamond Rapid Set Capsules, Kemdent, have never been tested with respect to their thermal behavior or compared to conventional GICs with polyalkenoic acid and thus we couldn't compare our results to previous literature. Thermal behavior of this material is shown in Figures 3 and 4.

4.3 Expansion of RMGIC

RMGIC showed expansion in the present study when temperature was increased (Figures 5 and 6). It is also been previously shown that RMGICs expand more under wet compared to dry conditions [9]. However, Yan *et al.* 2007, demonstrated that RMGICs contracted more than conventional GICs, possibly due to higher water content, which in turn leads to greater water loss. This disagreement with our results can be attributed to the different RMGIC chemical compositions. Incorporation of the resin component in GICs may significantly change their thermal characteristics [20]. The resinous phase expands with temperature increases, while the ionomer phase contracts [30]. Final thermal behaviour of the material may be determined by the amount of "loosely" bound water within its composition and the extent of counterbalance by polymerization shrinkage. It was shown, however, that the increase in volume due to thermal expansion is greater than the decrease in volume due to thermal contraction and cannot fully counteract polymerization shrinkage [13].

4.4. Effect of polymerization

It was previously demonstrated that higher light intensity of the curing device lead to lower CTE values in resinous materials [1], which complies with our results for KNL and KNH. It is therefore beneficial to polymerize with higher light intensity curing devices. Moreover, energy offering through polymerization is thought to improve the mechanical properties of chemically set GICs [16]. However CTE values were not significantly affected by extrinsic energy offerings, except for GICs with altered acid composition, which exhibited slightly increased CTE values when cured, especially in higher temperature ranges (Figure 3 and 4).

4.5. Understanding the significance of CTE

CTE changes with temperature are non-linear (Figures 1-6) and this is confirmed by Powers *et al.* 1979, and Sidhu *et al.* 2004. However, Versluis *et al.*, 1996, demonstrate results of good linearity. The mathematical sign of the CTE values has no correlation with CTE value increase or decrease. Increases in the CTE value demonstrate that the material expands (when $CTE > 0$) or contracts (when $CTE < 0$) at a higher rate, and vice versa. Published literature presents results as mean CTE values but fluctuations are considered the most influential factor in microleakage combined with the lack of adhesion [2]. Our results, as seen in the diagrams, show that CTE is defined at a specific temperature; for example, CTE is -12.5×10^{-6} at $40^{\circ}C$ (Figure 1). However in order to comply with the literature, we divided the temperature range $20-60^{\circ}C$ into two parts: $21-37^{\circ}C$ for the lower temperatures and $37-55^{\circ}C$ for the higher temperatures, as used by Sidhu *et al.* 2004. Differences were observed in the CTE values, depending on the temperature range examined, as shown in previously published studies [9, 22, 30]. The transition to higher temperatures, even if thermal equilibrium cannot be attained in a short time in the oral cavity, induces material fatigue [25] and this is why restorative materials are expected to have less predictable behavior at higher temperatures with respect to CTE.

4.6. Clinical significance and limitations

CTE measurement conditions in the present study were challenging for the materials tested. Data were collected during temperature increase [1, 22, 25], as it is thought to cause larger CTE variations because of material shock, compared to temperature decrease [13], when thermally induced volumetric changes are more even and milder. However great the challenge, oral conditions are much more complex and results of this study should be interpreted within certain limitations in mind. Higher intraoral temperatures last for a short time and are usually confined to the surface, depending on the thermal diffusivity of the tooth and on the action of the pulp as a heat sink with circulating blood supply [27]. Glass transition temperature of resinous materials [22] and moisture levels may also affect CTE measurements [9, 30]. In addition, tooth substance structures are not homogenous. An in vitro study showed that human dentine expands slightly on heating and contracts at higher temperatures [10].

5. Conclusions

- Type of GIC and light intensity of curing device affect thermal behavior.
- GICs contract in temperature raise, and RMGIC expands, but rate of volumetric change is not stable through temperature changes.
- Alterations in GIC typical composition play an important role in thermal behavior in higher temperatures, as polyacrylic acid probably induces greater contraction.
- It is beneficial to polymerize with a lower light intensity curing device.
- Extrinsic energy offer had no important effect on CTE values.

6. References

- Baek, C.J., Hyun, S.H., Lee, S.K., Seol, H.Y., Kim, H., Kwon, Y.H. (2008). The effects of light intensity and light-curing time on the degree of polymerization of dental composite resins. *Dental Materials*, 27(4), 523-33.
- Bullard, R.H., Leinfelder, K.F., Russell, C.M. (1988). Effect of coefficient of thermal expansion on microleakage. *Journal of American Dental Association*, 116, 871-4.
- Clark, J.R., Ireland, A.J., Sherriff, A.M. (2003). An in vivo and ex vivo study to evaluate the use of a glass polyphosphonate cement in orthodontic banding. *European Journal of Orthodontics*, 25(3), 319-23.
- Drummond, J.L., Robledo, J., Garcia, L., Toepke, T.R. (1993). Thermal conductivity of cement base materials. *Dental Materials*, 9(1), 68-71.
- Ferracane, J.L. (1995). *Materials in dentistry. Principles and applications.* (pp. 75-81, 102-109). J. B. Lippincott Company, Philadelphia.
- Gale, M.S., Darvell, B.W. (1999). Thermal cycling procedures for laboratory testing of dental restorations. *Journal of Dentistry*, 27, 89-99.
- Hashinger, D.T., Fairhurst, C.W. (1984). Thermal expansion and filler content of composite resins. *Journal of Prosthetic Dentistry*, 52, 506-10.
- Henchang, X., Wenyi, L., Tong, W. (1989). Measurement of thermal expansion coefficient of human teeth. *Australian Dentistry Journal*, 34, 530-5.
- Kim, Y.G., Hirano, S., Tadashi, H. (1998). Physical properties f resin-modified glass-ionomers. *Dental Materials Journal*, 17(1), 68-76.
- Kishen, A., Asundi, A. (2001). Investigation of thermal property gradients in the human dentine. *Journal of Biomedical Materials Research*, 55, 121-30.
- Knox, J., Chye, K.Y., Durning, P. (2004). An ex vivo evaluation of resin-modified glass polyalkenoates and polyacid-modified composite resins as orthodontic band cements. *Journal of Orthodontics*, 31(4), 323-8;discussion 301-2.
- Lambrechts, P., Ameye, C., Vanherle, G. (1982). Conventional and microfilled composite resins. Part II: Chip fractures. *Journal of Prosthetic Dentistry*, 48, 527-38.
- Mucci, V., Arenas, G., Duchowicz, R., Cook, W.D., Vallo, C. (2009). Influence of thermal expansion on shrinkage during photopolymerization of dental resins based on bis-GMA/TEGDMA. *Dental Materials*, 25, 103-14.
- Musanje, L., Darvell, B.W. (2004). Effects of strain rate and temperature on the mechanical properties of resin composites. *Dental Materials*, 20, 750-65.
- Naasan, M.A., Watson, T.F. (1998). Conventional glass-ionomers as posterior restorations. A status report for the American Journal of Dentistry. *American Journal of Dentistry*, 11(1), 36-45.
- O'Brien, T., Assadi, F.S., Lea, S.C., Burke, T., Palin, W.M. (2010). Extrinsic energy sources affect hardness through depth during set of a glass-ionomer cement. *Journal of Dentistry*, 38, 490-5.
- Palmer, D.C., Barco, M.T., Billy, E.J. (1992). Temperature extremes produced orally by hot and cold liquids. *Journal of Prosthetic Dentistry*, 67, 325-7.
- Plant, C.G., Jones, D.W., Darvell, B.W. (1974). The heat evolved and temperature attained during setting of restorative materials. *British Dental Journal*, 137, 233-8.
- Powers, J.M., Hostetler, R.W., Dennison, J.B. (1979). Thermal expansion of composite resins and sealants. *Journal of Dental Research*, 58, 584-7.

- Puckett, A.D., Fichie, J.G., Bennett, B., Hembree, J.H. (1995). Microleakage and thermal properties of hybrid ionomer restoratives. *Quintessence International*, 26, 577-81.
- Rose, E.E., Lal, J., Green, R., Cornell, J. (1955). Direct resin filling materials: coefficient of thermal expansion and water sorption of polymer methacrylate. *Journal of Dental Research*, 34, 589-96.
- Sideridou, I., Achilias, D., Kyrikou, E. (2004). Thermal expansion characteristics of light-cured dental resins and resin composites. *Biomaterials*, 25, 3087-97.
- Sidhu, S.K., Carrick, T.E., McCabe, J.F. (2004). Temperature mediated coefficient of dimensional change of dental tooth-colored restorative materials. *Dental Materials*, 20(5), 435-40.
- Sidhu, S.K., Sherriff, M., Watson, T.F. (1977). The effects of maturity and dehydration shrinkage on resin-modified glass-ionomer restorations. *Journal of Dental Research*, 76, 1495-501.
- Soderholm, K.J.M. (1984). Influence of silane treatment and filler fraction on thermal expansion of composite resins. *Journal of Dental Research*, 63, 1321-6.
- Strassler, H.E (2011). Glass ionomers for direct-placement restorations. [Online]. Available: http://www.ineedce.com/courses/2052/PDF/1104cei_glassionomer_web.pdf (Feb, 2011).
- Versluis, A., Douglas, W.H., Sakaguchi, R.L. (1996). Thermal expansion coefficient of dental composites measured with strain gauges. *Dental Materials*, 12, 290-4.
- Wilson, A.D., McLean, J.W. (1988). *Glass ionomer cement*. Chicago: Quintessence, (pp.43-56).
- Yamaguchi, R., Powers, J.M., Dennison, J.B. (1989). Thermal expansion of visible-light-cured composite resins. *Operative Dentistry*, 14, 64-7.
- Yan, Z., Sidhu, S.K., Carrick, T., McCabe, J.F. (2007). Response to thermal stimuli of glass ionomer cements. *Dental Materials*, 23, 597-600.

Comment for Figures 1-6:

Each line corresponds to a different specimen of each group. Arrow pointing right corresponds to CTE values, while arrow pointing left corresponds to linear shrinkage measurements throughout whole temperature range. Diagram lines were smoothed. Slopes of CTE measurements represent the rate, at which CTE values are altered throughout temperature fluctuations.

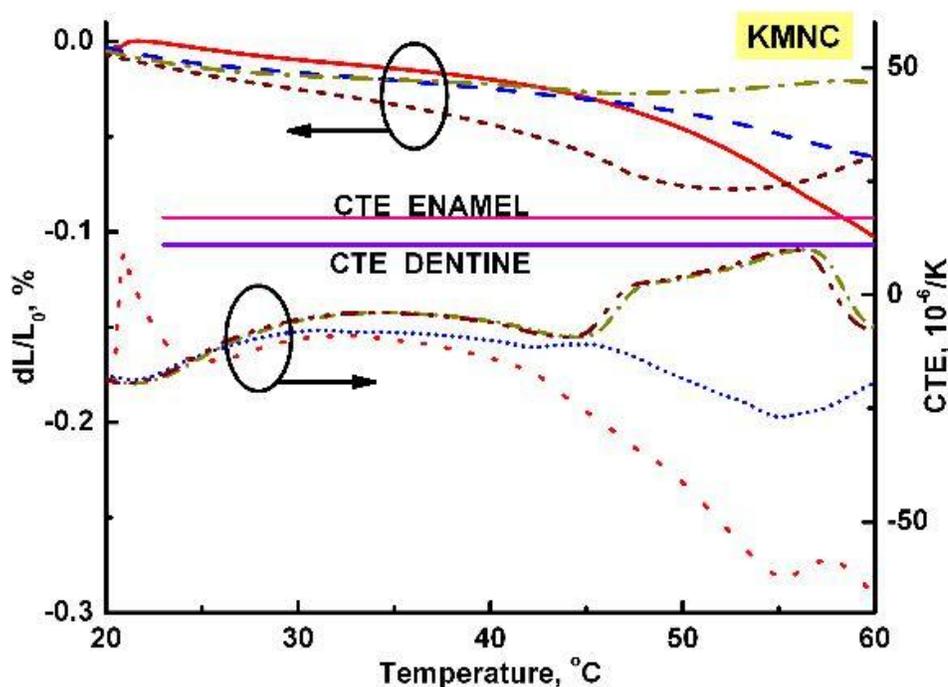


Figure 1

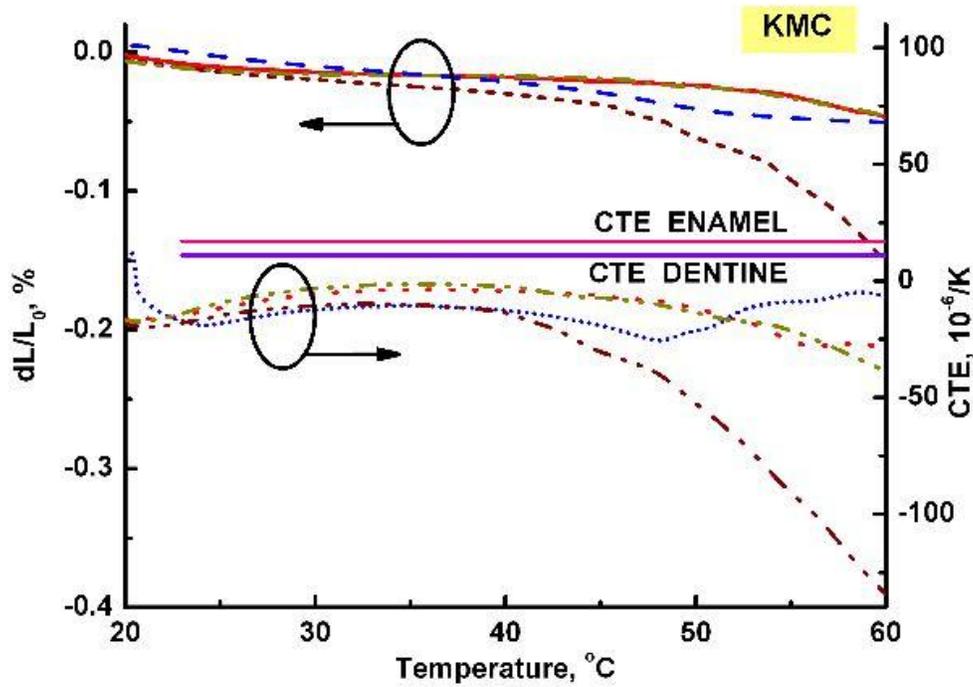


Figure 1 and Figure 2

Title: Diagrams of conventional GIC (groups KMNC and KMC): Slope of % linear shrinkage measurements shows that GIC contracts with temperature raise, either in cured or in not cured mode. CTE values are lower than enamel and dentine CTE values, for both groups.

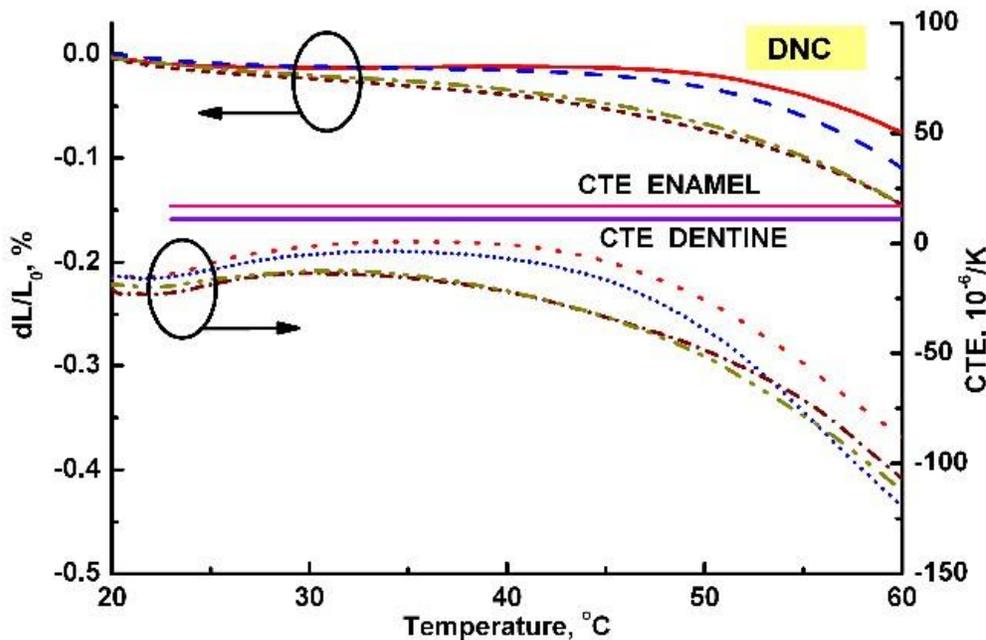


Figure 3

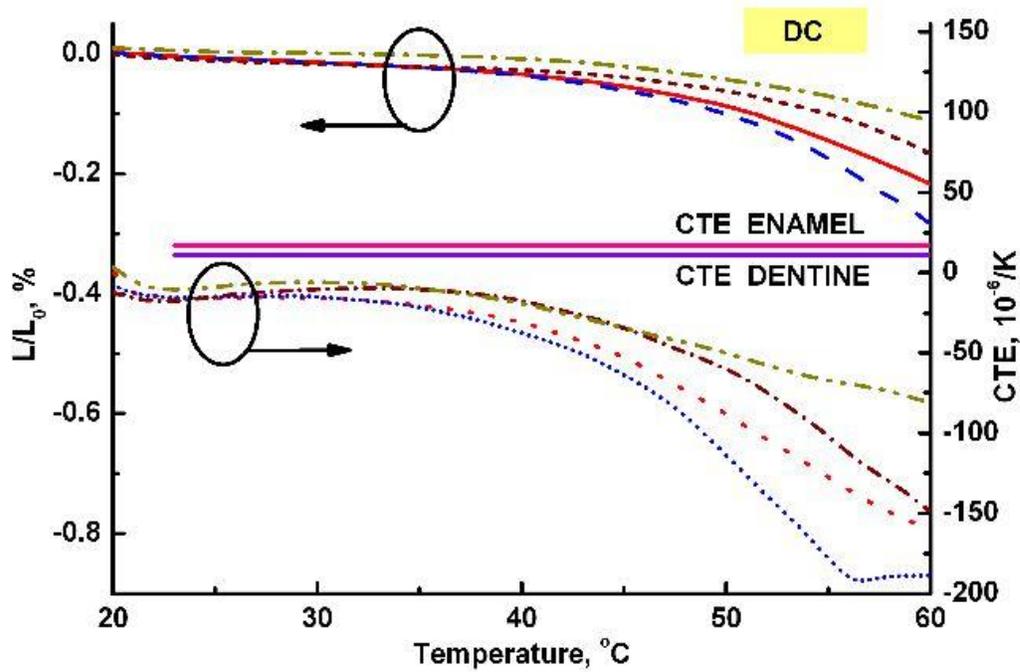


Figure 4

Figure 3 and Figure 4

Title: Diagrams of conventional GIC with altered acid composition (groups DNC and DC): Slope of % linear shrinkage measurements shows that GIC contracts with temperature raise, either in cured or in not cured mode. But values of CTE change more smoothly in these groups than in conventional GIC. Modulus of CTE values increases as temperature increases, meaning that the rate at which contraction takes place, increases. CTE values are lower than enamel and dentine CTE values, for both groups.

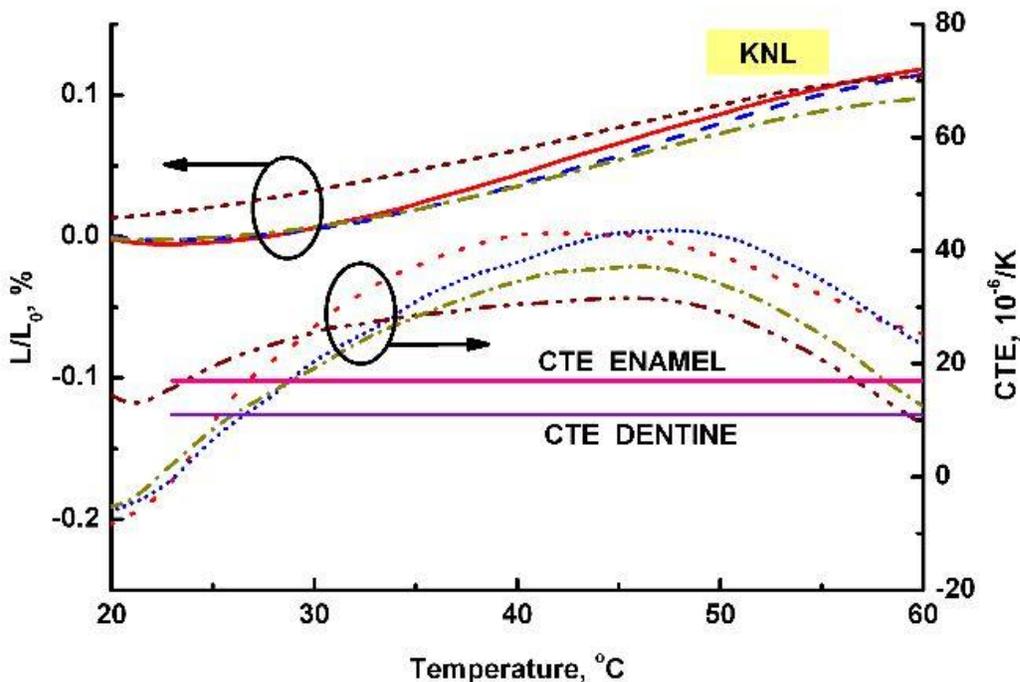


Figure 5

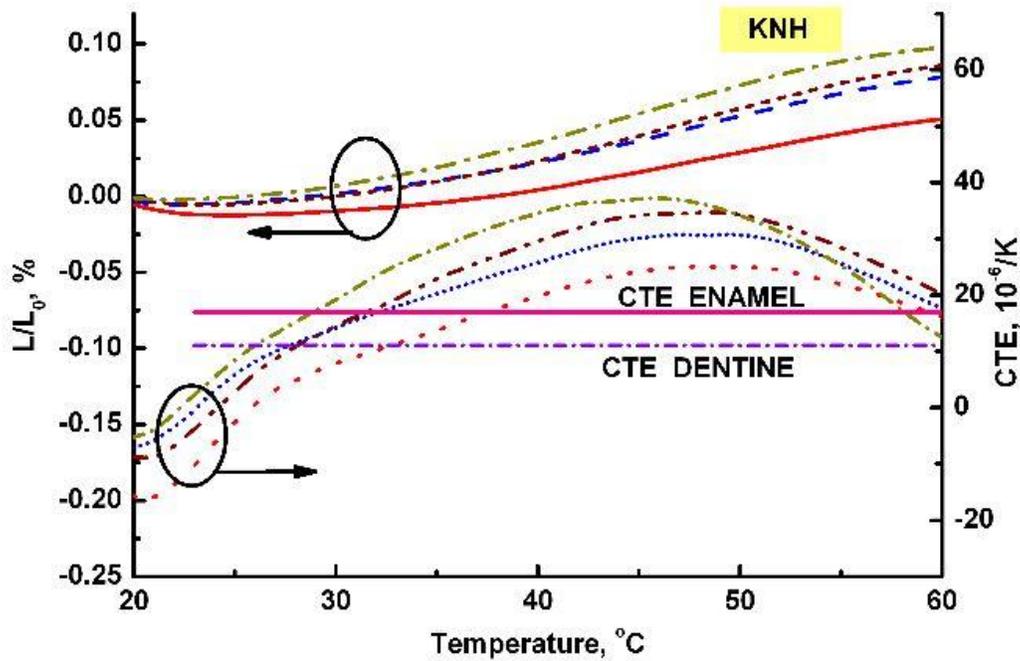


Figure 5 and Figure 6

Title: Diagram of RMGIC (groups KNL and KNH): Note that CTE values increase until a certain temperature value and then decrease. CTE values are not lower than enamel and dentine CTE values, in every temperature. Slope of % linear shrinkage measurements exhibits that this material expands with temperature raise.