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Effect of silver-coated silica nanoparticles on the thermal conductivity of thermally activated acrylic resin

Efeito das nanopartículas de sílica revestidas por prata na condutividade térmica da resina acrílica ativada termicamente

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ABSTRACT

Objective: Thermally activated acrylic resins (RAATs) are widely used in dentures as a base material due to their good dimensional stability and biocompatibility. However, their low thermal conductivity is a disadvantage, as it affects acceptance when using dental prostheses. Thus, the objective of this work was to measure the conduction heat in RAATs with and without incorporation of silica and silver nanoparticles (NP) and rigid reline (RR). **Material and Methods:** For this, samples were made and divided into 10 groups (n = 6). The first five groups were 2-mm-thick samples: G1 (RAAT control), G2 (RAAT + RR control), G3 (RAAT and NP + RR), G4 (RAAT + RR and NP), and G5 (RAAT and RR modified by NP). In the other five groups, 8-mm-thick samples were made: G6 (RAAT control), G7 (RAAT + RR control), G8 (RAAT and NP + RR), G9 (RAAT + RR and NP), and G10 (RAAT and RR modified by NP). The heat that cross the surface of the specimens was quantified using a wireless device. The data were submitted to two-factor ANOVA statistical analysis and Tukey´s test with a 5% significance level. **Results:** After measuring the temperature variation as a function of time, it can be observed that there was a statistically significant difference for thermal conduction between the control groups and those modified with NP. **Conclusion:** Thus, it was possible to conclude that the NP improved the heat conduction in RAAT and in the RR because the nanoparticles have a higher thermal conductivity.

KEYWORDS

Thermal conductivity; Acrylic resin; Nanoparticle; Silver.

RESUMO

Objetivo: As resinas acrílicas termicamente ativadas (RAATs) são amplamente utilizada em próteses dentárias como material de base, pois possuem uma boa estabilidade dimensional e biocompatibilidade. Porém, como desvantagem, possuem baixa condutividade térmica, o que prejudica a aceitação do uso de próteses dentárias. Assim, o objetivo deste trabalho foi medir a condução de calor em RAAT com e sem incorporação de nanopartículas de sílica e prata (NP) e reembasador rígido (RR). **Material e Métodos:** Para isso, foram confeccionadas amostras que foram divididas em 10 grupos (n=6). Os primeiros cinco grupos eram amostras de 2 mm de espessura: G1 (RAAT controle), G2 (RAAT + RR controle), G3 (RAAT e NP + RR), G4 (RAAT + RR e NP) e G5 (RAAT e RR modificados por NP). E nos outros cinco grupos foram feitas amostras com espessura de 8 mm: G6 (RAAT controle), G7 (RAAT + RR controle), G8 (RAAT e NP + RR), G9 (RAAT + RR e NP) e G10 (RAAT e RR modificados por NP). O calor percorrido pela superfície dos corpos – de prova foi quantificado por meio de um dispositivo sem fio. Os dados foram submetidos à análise estatística ANOVA dois fatores e teste de Tukey com 5% de significância. **Resultados:** Após medir a variação da temperatura em função do tempo, pode-se observar que houve diferença estatisticamente significante para a condução térmica entre os grupos controle e os modificados com NP. **Conclusão:** Assim, foi possível concluir que a NP melhorou a condução de calor na RAAT e no RR, pois as nanopartículas apresentam maior condutividade térmica.

PALAVRAS-CHAVE

Condutividade térmica; Resina acrílica; Nanopartículas; Prata.

INTRODUCTION

Acrylic resins are polymeric materials widely used in dentistry. Some of its applications include total and partial prostheses, myorelaxant splints, individual trays, casting patterns, complete dentures, temporary crowns, artificial teeth, total prostheses rebase, and the acrylization of orthodontic appliances. This versatility is possible because acrylic resins are tasteless, odorless, non-toxic, non-irritating to tissue, insoluble in oral environments, and disinfectable, in addition to their suitable dimensions and color stability [1-3].

These materials can reproduce the oral surface in detail and can be easily relined. However, in contrast to these advantages, large difficulties are encountered with their use due to undesirable properties, such as a high coefficient of thermal expansion, low thermal conductivity, low elasticity coefficient, low impact, and fatigue strength [4-7]. In particular, the distinct disadvantage of polymethylmethacrylate (PMMA) is its low thermal conductivity [8,9].

Thermal conductivity is an important characteristic of acrylic resins, as it affects the acceptance of prostheses by patients. This is mainly a factor in upper total prostheses, in which the palate of the patient is fully covered with a layer of acrylic resin. Some patients claim to lose their sense of taste when using complete dentures because the mucosa is covered and has no direct contact with food [9]. For this reason, studies have proposed implementing additional methods to provide better comfort to patients, such as adding silver nanoparticles (AgNPs). These nanoparticles can provide an antimicrobial effect, especially in high concentrations [10], and increase the thermal conductivity of the acrylic resin used in the confection of prostheses. These findings are supported by a study in which the antimicrobial effect of incorporating AgNPs was demonstrated [11].

Silica is one of the most popular material for enhancing the properties of AgNPs and facilitating it application in health-related areas [12]. The attributes of high stability, chemical inertia, controllable porosity, processability, and optical transparency make silica an ideal auxiliary material to compensate for the disadvantages arising from employing solely AgNPs [13,14]. Silica can be combined with silver to reduce the effects of toxicity by coating silica particles with silver. Using this method, it is possible to limit the release of free silver ions and minimize cytotoxicity [15]. Based on these results, the clinical use of AgNPs seems to be beneficial. Importantly, measuring the heat conduction can reveal if there is a real advantage in combining these particles with the pure acrylic resin.

Hamedi-Rad et al. (2014) [7], analyzed the thermal conductivity, compressive strength, and tensile strength of PMMA reinforced with AgNPs. The study showed that the thermal conductivity and compressive resistance of the acrylic resin increased with the incorporation of the NPs. These findings support the integration of NPs in PMMA for areas such as the palate of total prostheses. However, the addition of 5 vol% NPs in the PMMA decreased the tensile strength, indicating that further studies are needed to clarify the relationship between the material ratios and corresponding properties.

The objective of this study was to quantitatively evaluate the thermal conductivity of an acrylic resin (RAAT) used in prostheses and reline materials when it is combined with silica and silver NPs by using a wireless thermal sensor. The following variables were considered: the utilization of a rigid reline (RR), proportions of silica and silver NPs in the RAAT and in reline, and thickness of the RAAT.

The null hypothesis is that the RAAT will not exhibit significant thermal conductivity changes when varying the thickness, use of reline, or addition of NPs. The study hypothesis is that the RAAT and RR modified with silica and silver NPs will have higher heat conduction when compared to the control groups.

METHODS AND MATERIALS

Confection of acrylic resin specimens

Specimens were confectioned of RAAT with different thicknesses (2 mm and 8 mm) and were divided into 10 groups (n = 6) (Figure 1 and 2).

The samples were fabricated from two types of stainless-steel patterns with dimensions of 2.0 cm × 2.0 cm × 0.8 cm and 2.0 cm × 2.0 cm × 0.2 cm. These patterns were molded with the same type of laboratory silicone-based impression material (Rhodorsil–VWL, Prolabor). From these molds, replicas were obtained of the samples in wax nº 7 (Clássico, Artigos Odontológicos Clássico Ltda., São Paulo, Brazil).

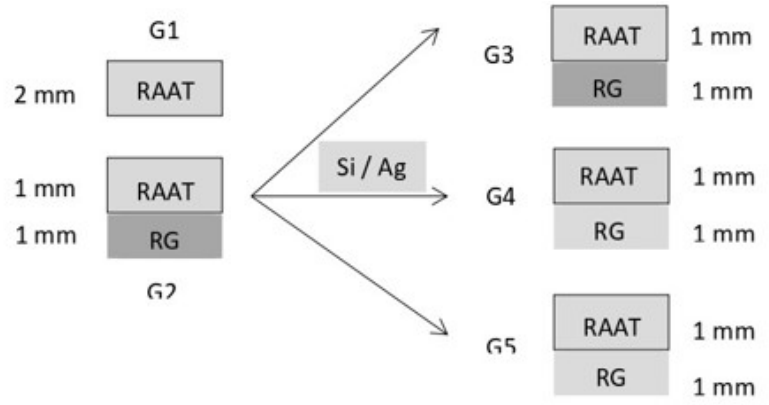

Figure 1 - Experimental groups (thickness=2 mm). RG – Rigid reline.

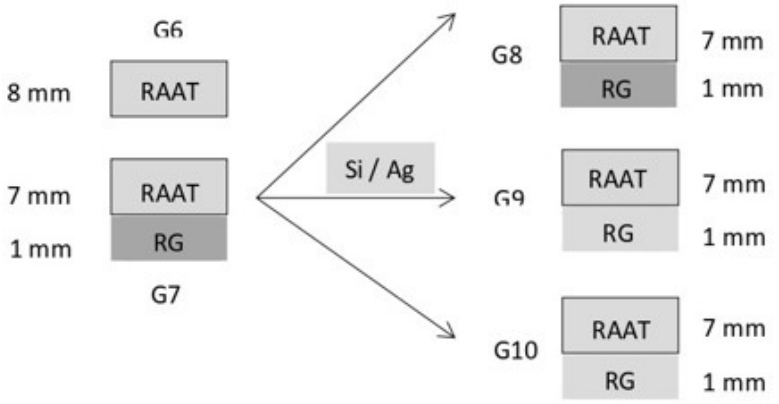

Figure 2 - Experimental groups (thickness=8 mm). RG – Rigid reline.

To make the specimens in RAAT, the samples in wax were used together with metallic flasks and type II plaster. These plasters were manually mixed, and the six wax patterns were positioned. Only the side with the largest area of the patterns was in intimate contact with the plaster surface (Figure 3).

After the final plaster setting, a thin layer of plaster and acrylic resin (Al–Cote, Dentsply Sirona India Pvt. Ltd.) was applied, and the plaster was added again to fill the flask. Approximately 1 h after the final inclusion, the phase was completed by opening the flask and removing the wax patterns with a hot water bath. For the pressing procedure, two portions of colorless RAAT (Vipi Cril Dental Vipi Ltd. Brazil) were agglutinated in the plastic phase and were placed into the flask.

For the modified sample groups, NPs were dispersed in the RAAT monomer, and the solution was stirred on a magnetic stirrer for 15 min for complete dispersion. The nanoparticle concentration used was 2 wt% of the resin mass, with 1.2 g of NPs for 60 g of resin. The polymer was added to the modified monomer and agglutinated in a glass pot covered with a lid. The mixture was allowed to rest until the plastic phase of the resin was obtained so that it could be accommodated in the spaces left by the wax patterns.

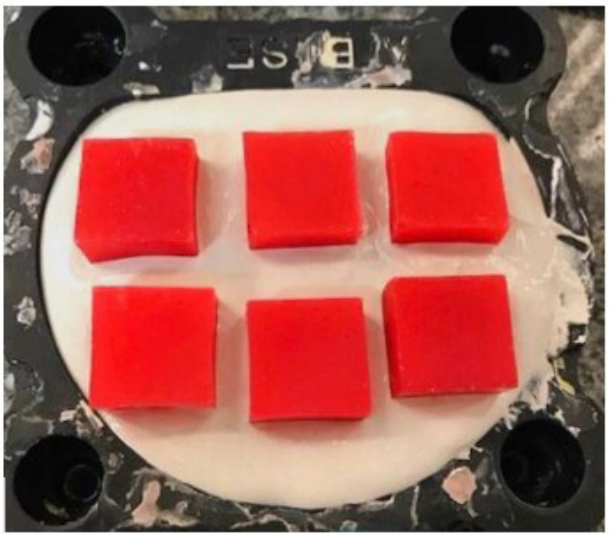

Figure 3 - Molds preparation for specimen manufacturing.

The flasks with RAAT were closed and positioned in a hydraulic press (Techno Máquinas,

Campinas, São Paulo, Brazil), and a discrete increasing pressure was applied until reaching 1250 kgf, where no additional flow of resin was visible. A long polymerization cycle was used: 74 °C for 9 h in a hot water bath.

After completing the polymerization cycle, the flasks were maintained for 2 h at ambient temperature (24ºC), on a bench to cool down. Then, the flasks were opened, and the samples were conditioned in distilled water.

For the finishing process, the samples were thinned and polished (EcoMet 250, Buehler) under constant water-cooling using aluminum oxide sandpaper (Norton) with progressively smaller grit sizes (400, 600, and 1200). Subsequently, the samples were held in a vise and polished with pumice stone and whip mix (Dazzle PS), with constant cooling. To determine the final dimensions of the samples, the samples were first cleaned in an ultrasonic bath (Thorton) with distilled water for 2 min to remove residues accumulated during material wear. Afterward, the sample measurements were performed using a digital caliper (Figure 4).

Silver-coated silica nanoparticle synthesis

The silica NPs were synthesized by hydrolysis and controlled condensation of tetraethylorthosilicate (TEOS) in alcohol, which is known as the Stober method (Stober et al., 1968) [16]. The following reagents and solutions were used to synthesize the SiO_2: TEOS 98% (LS Chemical, São Paulo, Brazil, lot MKBP8202), ammonium hydroxide P.A. (LS Chemical, São Paulo, Brazil, lot 71008), absolute ethyl alcohol 99.5%. (Neon Comercial, São Paulo, Brazil, lot 20930), silver nitrate P.A. (LS Chemical, São Paulo, Brazil, lot 119308/14), anhydrous dextrose (LS Chemical, São Paulo, Brazil, DA11.3), and anhydrous soda ash P.A. (Labsynth, São Paulo, Brazil, lote18610). Two solutions were prepared: the first was a mixture of 9 mL TEOS with 55 mL ethyl alcohol and the second solution contained 5 mL ammonium hydroxide, 55 mL ethyl alcohol, and 30 mL deionized water. The solutions were then mixed and maintained under constant agitation in a magnetic agitator for 1 h.

Once the solution of SiO_2 was obtained, the Si particles were coated with silver, using a method similar to the one reported by Nischala et al. (2011) [17]. For the first step of the coating process, 50 mL of the solution obtained in the previous stage was mixed with 12.5 mL deionized water, 1.70 g silver nitrate (10 mmol), and 2 g glucose. The mixture was stirred in a magnetic agitator for 30 min at ambient temperature. Then, 25 mL of a sodium carbonate solution (25 mL deionized water + 2 g sodium carbonate) was added dropwise to the mixture, and agitation was maintained for over 1 h. The particles were separated using a centrifuge (9000 rpm, 10 °C, for 5 min) and washed with distilled water three times to remove the silver supernatant particles. The precipitate was dried at 37 °C and it was ground to a fine powder using an agate mortar.

Reline application

A RAAT original sample was molded with condensation silicone and subsequently reduced to 1 mm of its original thickness by polishing with sandpaper (Politriz Ecomet 250, Buheler). On the surface of the thinned sample, a chemically activated RR was applied (Soft Confort Dura Kit - Dencril). Using a silicone mold, the rebase application was continued until the original thickness of the sample was reestablished. When the rebase was added to the base resin, a glass plaque was positioned with continuous pressure on the sample until total polymerization was achieved. The silver and silica NPs were added to the reline using the same method as for the RAAT. These samples with reline were also finished and polished (Figure 5).

Figure 4 - Finished samples. Left to right, the groups G1, G2, G3, G4, G5.

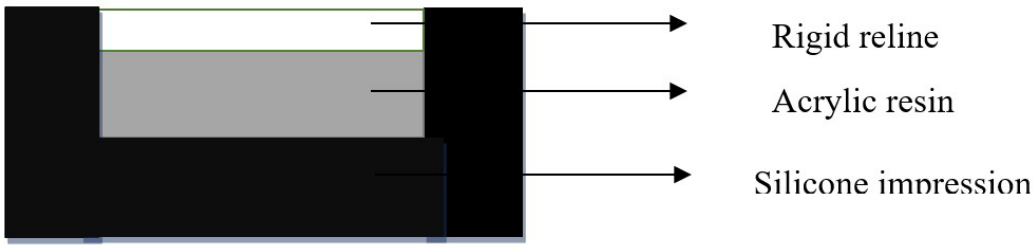

Figure 5 - Illustration of the rebase in the specimens.

Thermal conductivity analysis

iButton technology was used for temperature analysis. The sensors, iButtons DS1922E, and DS1925 (Maxim Integrated, San Jose, CA, USA), are resistant and self-sufficient systems that measure temperature between –40 °C and +85 °C. Readings were recorded in a protected memory section, with measurements taken once per second. The diameter and thickness of the sensors were 17.35 mm and 6.4 mm, respectively. These sensors were configured to communicate with a computer using a serial 1-Wire protocol connected through a USB. The temperature conduction analysis was performed three consecutive times on each sample, under the same conditions.

Application of temperature sources

The thermic sources applied were of temperatures similar to those present in oral environments, for both hot (60 °C) and cold (1 °C) conditions [18,19].

For hot sources, a heating magnetic stirrer was used (752A, Fisatom, Brazil, 60 Hz, 650 W, 230 V, Series 1138488), regulated at temperatures between 50 °C and 60 °C. To first graduate the temperature, the iButton dispositive was applied directly on the magnetic stirrer to check the temperature-time relation and determine the maximum thermic value. The minimum time to reach that value was determined as a reference to compare with the sample tests. Based on the test result, a duration of 17 min was chosen for the measurement of temperature variation as a function of time, and the measurements were performed every 1 s. To conduct the measurements, each sample was positioned directly on the flat surface of the heat source. A thin layer of thermal paste (Implastec) was applied to the sample, and the thermal sensor was positioned on the opposite side of the heated surface and in direct contact with the thermal paste (Figure 6 and 7).

For the cold-temperature measurements, a small ball of polystyrene was prepared with an interior space removed with dimensions close to those of the sample, to minimize heat dissipation in the ball. Distilled water was inserted into this space, and the ball was placed in a freezer for 48 h before the first measurement for the water to be completely frozen. Directly before the cold-temperature measurements began, the polystyrene with ice was removed from the freezer, and the iButton was immediately applied on the surface, similar to the method described in the heated condition until a minimum stable temperature rate was verified. Based on this observation, the time for analysis was chosen to be 30 min. The temperature variation measurement was determined to be equal to that of the heat source.

After obtaining the data, the following formula was applied to calculate the degree of thermal conductivity:

$$\frac{\Delta Q}{\Delta t} \bullet \frac{L}{A} \equiv K \bullet \Delta T \quad (1)$$

The thermal conductivity coefficient corresponds to the amount of energy, in the form of heat, that passes through 1 m^2 of surface in a second, when the difference in temperature between the two sides of the material is 1 °C. In the formula, the variables are the heat quantity (ΔQ), time interval (Δt), thickness of the material (L), area of the material (A), thermal conductivity (K), and applied temperature difference (ΔT). The results were analyzed according to the values of ΔQ (J/s) obtained for an applied ΔT.

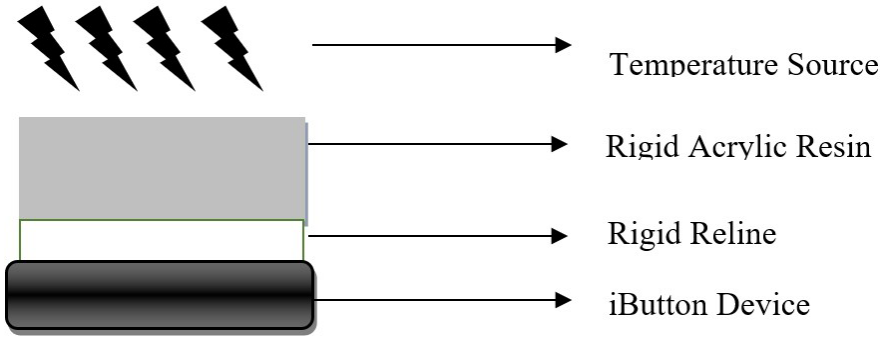

Figure 6 - Illustration on how to apply the thermal source and the recording device.

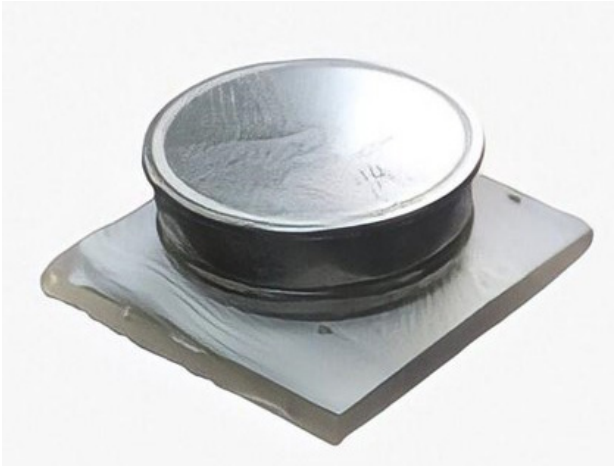

Figure 7 - Mode of application of the thermal source and the recording device.

Statistical analysis

Descriptive analyses of the values obtained were considered, in addition to the inferential statistics. The factors of NP presence, type of material, and thickness were the independent variables and thermal conductivity were taken as the dependent variables. Two-way ANOVA and Tukey's test with a 5% significance level were also used.

RESULTS

After measuring the temperature variation as a function of time, a statistically significant difference was observed between the means of the groups according to the two-way ANOVA results ($p = 0.000$).

For the temperature variation test with the heat source, groups G1 (RAAT), G2 (RAAT + RR), G6 (RAAT), and G7 (RAAT + RR), exhibited the lowest heat conduction values as a function of time, because the RAAT and RR have low thermal conductivities. Groups G3 (NP RAAT), G4 (NP RR), G5 (NP RAAT + RR), G8 (NP RAAT), G9 (NP RR), and G10 (NP RAAT + RR) showed higher values of heat conduction as a function of time since they were modified by NPs (Tables 1 and 2).

For the temperature variation test with the cold source, groups G1 (RAAT control), G2 (RAAT + RR control), G6 (RAAT control), and G7 (RAAT + RR control), presented the lowest heat conduction values as a function of time, again because the RAAT and the RR have low thermal conductivities. Accordingly, groups G3 (RAAT with NP + RR), G4 (RAAT + RR with NP), G5 (RAAT and RR with NP), G8 (RAAT with NP + RR), G9 (RAAT + RR with NP), and G10 (RAAT and RR with NP) also exhibited higher values of heat conduction as a function of time since they were modified by NPs (Table 3 and 4).

Table 1 - Two -factor ANOVA and Tukeys´s test. 2 -mm-thick samples in heat source,p=0.000; α=0,05

Groups	Mean ± standard deviation (j/s)
G1	0.63±0.06 A
G2	0.74±0.07 A
G3	12.92±2.15 B
G4	12.65±1.94 B
G5	12.87±1.05 B

Table 2 - Two -factor ANOVA and Tukeys´s test. 8 -mm-thick samples in heat source,p=0.000; α=0,05

Groups	Mean ± standard deviation (j/s)
G6	0.44 ± 0.1 A
G7	0.5 ± 0.06 A
G8	8.04 ± 1.01 B
G9	8.35 ± 1.21 B
G10	6.84 ± 1.53 B

Table 3 - Two -factor ANOVA and Tukeys´s test. 2 -mm-thick samples in cold source,p=0.000; α=0,05

Groups	Mean ± standard deviation (j/s)
G1	0.56 ± 0.05 A
G2	0.61 ± 0.07 A
G3	10.24 ± 0.7 B
G4	9.6 ± 0.56 B
G5	9.79 ± 0.6 B

Table 4 - Two -factor ANOVA and Tukeys´s test. 8 -mm-thick samples in cold source,p=0.000; α=0,05

Groups	Mean ± standard deviation (j/s)
G6	0.42 ± 0.02 A
G7	0.52 ± 0.04 A
G8	9.19 ± 0.93 B
G9	9.41 ± 0.51 B
G10	8.24 ± 0.66 C

DISCUSSION

RAAT is known to be an excellent material for use in denture bases. However, the principal disadvantage of RAAT is its low thermal conductivity, which leads to the displeasure of many patients [7,9]. According to the findings of the present study, the null hypothesis was rejected, since there was an increase in thermal conduction of the samples modified with the silver and silica NPs. The positive hypothesis of the study was accepted because there were significant changes in the heat conduction of the samples modified with the NPs.

Other studies have presented similar results to the current study. For example, a control group with acrylic resins presented a lower thermal conductivity when compared to sample groups modified with 0.2 wt% or 2 wt% AgNPs. The group with 2 wt% NPs presented the highest thermal conductivity. In addition, it was possible to perceive a statistically significant difference between the groups [20].

Another study reported that the addition of nanosilver to acrylic resin increases its thermal conductivity and that this depends on the percentage of nanoparticles [21].

Palmer et al. (1992) [20] conducted tests to measure the maximum and minimum intraoral temperatures with hot and cold liquids. The maximum temperature measured in the posterior region of the jaw was 53.1 °C and the minimum temperature was 1 °C, while the maximum temperature of 58.5 °C was observed in the anterior region of the maxilla

Hamedi-Rad et al. (2014) [7] cited the importance of thermal conductivity to determine a gustatory response to food, as acrylic-based dentures possess a low thermal conductivity in comparison to metal-based dentures.

However, other studies showed that metallic or acrylic resin-based palate prostheses inhibited the gustatory response of acids under heat stimulus, whereas they increased the gustatory sensitive response cold stimulus. Therefore, the higher thermal conductivity demonstrated by metallic palate bases might not be a great advantage in terms of the gustatory sensitive response [9].

Nevertheless, the thermal conductivity of denture bases plays an important role in activating the secretion of the parotid glands, which is the digestive response to the temperature variation and taste of foods. The functional secretion of the parotid glands increases as the temperature of the palate tissue increases [9]. Therefore, the greater the thermal conductivity of denture bases, the better the transmission of the temperature of the intraoral environment to the palate, increasing the parotid gland secretion and improving the digestive response and sensitivity to the flavor of the food.

In this study, the incorporation of silver successfully improved the thermal conductivity of the acrylic resin. Upon analyzing different sample groups, we observed an increase in thermal conduction with the addition of 2 wt% silica and silver NPs in the 2 mm and 8 mm samples, independent of the material into which the NPs were added. Samples of 8 mm width are important for use in obliterating palate prostheses or in cases of a ridge with greater bone resorption. Hamedi-Rad et al. (2014) [7] also obtained an increase in the thermal conductivity of PMMA by the addition of 5 wt% AgNPs.

Although this is not the focus of this study, it is worth mentioning that certain mechanical properties can also be affected by particle incorporation, as the flexural strength of PMMA can change depending on the concentration of AgNPs used. Samples with a lower concentration of AgNPs show higher

flexural resistance than those with higher AgNP concentrations [22].

It has also been reported that at low concentrations, AgNPs do not negatively affect the mechanical properties of acrylic resin. Moreover, an alteration in the thermal properties of the acrylic resin was reported after the addition of AgNPs [23]. Monteiro et al. (2012) [24] showed that AgNPs are efficient against *Candida albicans*, especially in the case of PMMA containing 5% AgNPs. From these studies, there appears to be a connection between the concentration of AgNPs in acrylic resin and its effectiveness as an antimicrobial agent.

A disadvantage of adding AgNPs to acrylic resin is that the NPs can darken the resin coloring, limiting its use in esthetic areas. However, small concentrations can be added to the palate region of dentures to improve the perception of flavor and patient satisfaction [25]. Such color alterations were also observed in this study. Therefore, future studies are necessary to analyze the effects of adding silica and silver NPs at other concentrations to achieve better optical properties while maintaining the beneficial thermal conductivity improvement presented in this initial study.

The results of this study demonstrate that the incorporation of 2 wt% silica and silver NPs increased the thermal conductivities of RAAT and RR. However, better homogeneity and a wider variety of NP concentrations in the materials must be explored in future studies to improve the optical aspect of the materials and extrapolate these observations for clinical use of prosthetic parts.

CONCLUSION

According to the results obtained in this study, the addition of silver and silica NPs improved the heat conduction in RAAT and RR, regardless of how they were added to the materials or the sample thickness.

Authors' Contributions

JFGS: Methodology, formal analysis, software, investigation and writing-original draft preparation. NRR: Methodology, formal analysis, software. BRCM: Methodology. GPT: Writing-review and editing, supervision and project administration. TAPJ: Conceptualization, writing-review and editing, supervision, project administration and funding acquisition. All authors have read and agreed to the published version of the manuscript.

Conflict of Interest

The authors declare no conflict of interest.

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Regulatory Statement

The authors declare that this in vitro study did not require application to the institution's ethics committee.

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