Comparison of residual monomer content and flexural strength of two auto-polymerizing reline resins subjected to microwave post-polymerization treatment

Original Article

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ABSTRACT

Statement of Problem: Microwave post-polymerization has been suggested as a method to improve the flexural strength of auto-polymerizing denture relines resin. However, the effect of microwave post-polymerization on residual monomer content and its influence on flexural strength has not been investigated. **Purpose:** This study analyzed the effect of microwave post-polymerization on the residual monomer content and its influence on the flexural strength of two auto-polymerizing reline resins (Kooliner and Denture Liner) and compared its flexural strength. **Materials and Methods:** For each material, 70 specimens (64x10x3.3 mm) were polymerized according to manufacturer's instructions and divided into seven groups (n is equal to 10). Control group specimens remained as processed. Before testing, the specimens were subjected to post-polymerization in a microwave oven using different power (550 and 650W) and time (three, four and five minutes) settings. The specimens of each group were then manually ground into fine powder and samples extracted from the specimens using reflux method. The samples were then subjected to gas chromatography for residual monomer determination in area%. The specimens were subjected to three-point bending device with a span of 50mm and crosshead speed of 5mm/min and flexural strength was determined in MPa. Data analysis included Student 't' Test and One-way analysis of variance. **Results:** For Kooliner and Denture Liner reline resin, the residual monomer content decreased and the flexural strength increased significantly with the application of microwave irradiation using different time/power combinations. The specimens with the lowest residual monomer content were the similar specimens which presented with the highest flexural strength. **Conclusion:** Microwave post-polymerization irradiation can be an effective method to increase the flexural strength of Kooliner (at 550 W for 5 mins) and Denture Liner (at 650 W for 5 mins) by reducing the residual monomer content by further polymerization at free radical sites. The Kooliner reline resin had superior flexural strength compared to Denture Liner reline resin.

KEY WORDS: Autopolymerizing reline resin, flexural strength, residual monomer

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INTRODUCTION

Complete denture adaptation to the oral tissue is one of the important factors to achieve good support, stability and retention. However, alveolar resorption is a continuous process, with resulting loss of it in local areas of the denture base,[1-3] requiring reline of the complete denture to re-establish the fit and improve the supportive capability of the denture base.

Many commercially available hard auto-polymerizing reline resins[4-6] allow the dentist to reline removable prostheses directly, intra-orally and also have the advantage of reproducing the morphological features of oral soft tissue directly on the denture base when compared to the heat cure reline resin. The heat cure

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reline resins have the added disadvantage of extra patient visit, as well as laboratory fee, and the patient must be without dentures for a period of time.[7] Complete denture relined with auto-polymerizing hard reline resins has decreased mechanical strength due to the presence of higher residual monomer content.[8-9] which can also elicit irritation, inflammation and sometimes allergic response in oral mucosa.[10-12]

It has been demonstrated that the residual monomer in an auto-polymerizing acrylic resin may be reduced by further polymerization at the free radical sites which could be achieved following a period of immersion in hot water.[13-16] Microwave energy has been used for polymerization, having the advantage of reduced time for curing, a smaller time for obtaining of the plastic phase, a bigger homogeneity of the mixture and the achievement of a prosthetic material with excellent mechanical strength.[17] These microwaves are electromagnetic waves generated by a magnetron. It has been reported that microwave polymerization involves heating the acrylic resin monomer only, and not the polymer. This allows a relatively low processing temperature around the material, resulting in little residual monomer and good dimensional accuracy. And also that microwave irradiation of an auto-polymerizing acrylic resin soon after polymerization decreases the residual monomer content by 25%, with an increase in the impact strength and glass transition temperature.[18] Similarly, it has been demonstrated that microwave post-polymerization resulted in a high degree of conversion and higher flexural strength of an auto-polymerizing denture reline resin repair material.[19] With appropriate combination of power and time for the polymerization of heat polymerized denture base acrylic resin using microwave irradiation, it is possible to minimize the level of residual monomer and possibly reduce toxicity.[17]

The influence of different power and time settings of microwave post-polymerization on flexural strength has been investigated. But the same on residual monomer content and also the influence of decrease in residual monomer content on the flexural strength of auto-polymerzing denture reline resin has not been investigated. Therefore, the purpose of the investigation was to determine, quantitatively, the effect of different microwave oven power/time combinations on the residual monomer content and its influence on the flexural strength of two auto-polymerizing hard denture reline material and to compare their flexural strength.

MATERIALS AND METHODS

Two commercially available auto-polymerizing hard denture reline resin was selected for this study [Table 1]. 70 samples were fabricated in a stainless steel mold of dimension 64x10x3.3 mm as per ISO/FDI 1567 standards.[20] The material was proportioned and manipulated following the manufacturer's instructions [Table 1] and packed within the mold in a temperature maintained chamber. The samples were finished with 400 grit silicon carbide paper to remove the irregularities. The accuracy of the dimensions was verified with a digital vernier caliper, at three locations of each dimension to within 0.2mm tolerance. The final sample dimensions were measuring as - length 64 plus/minus 0.2mm; width 10 plus/minus 0.2mm; height 3.3 plus/minus 0.2mm. All samples were stored in a thermostatically controlled water bath at 35^{0}C for 48 hours before testing.[20]

The 70 samples were divided into seven groups (n is equal to 10). Control Group C remained as finished. Six experimental groups (Group 3A to Group 5B) of samples were subjected to different post polymerization irradiation procedures in a domestic wattage adjustable microwave oven by altering the power (550 W and 650 W) and the time settings (three, four and five minutes) [Table 2]. The samples were placed in the microwave oven and exposed to microwave energy directly.

Gas chromatography test

The specimens of each group were then manually ground into fine powder and samples extracted from the specimens using reflux method. Gas chromatography was employed to know the residual monomer content, using a glass column of 2mm long and 3.5 internal diameters, filled with 10% carbowax 20 M (polyethylene glycol 20,000).The oven temperature was 85^o C, the injection temperature was 200^o C, and the flame ionization detector temperature 150^{o}C. Nitrogen was used as a carrier gas at 30ml/min. The detector output was linked to a chart recorder, with the chart speed set at 2 min/cm. An amount of 2µl of the methanolic extract was injected into the gas-liquid chromatography and the calibration of the instrument was checked using a known amount of monomer in methanol before and

Table 1: Materials used

Product	Manufacturer	Polymer/monomer ratio	Polymer	Monomer	Polymerization time
Kooliner	GC America, Alsip, III	2.1g/1.5 ml	PEMA	IBMA	10 min at room temperature
Denture liner	Shofu, Kyoto Japan	1.7g/ 1 ml	PEMA	MAATE	13 min at room temperature

PEMA-Poly (ethylmethacrylate); IBMA-isobutyl methacrylate; MAATE- Methacrylicacidtriester.

Table 2: Conditions of microwave postpolymerization

Time	Power (W)	Group code
3 min	550	550/3
	650	650/3
4 min	550	550/4
	650	650/4
5 min	550	550/5
	650	650/5
Time	**Power (W)**	**Group code**
3 min	550	550/3
	650	650/3
4 min	550	550/4
	650	650/4
5 min	550	550/5
	650	650/5

Table 3: Comparison of various combination of power and time i.e. with in the groups of Kooliner reline resin using Student 't' unpaired test

	550X3	550X4	550X5	650X3	650X4	650X5
550X3	_	0.5425	5.8416*	0.2996	1.7820	1.5813
550X4	_	_	3.5687*	0.6585	0.5628	1.2790
550X5	_	_	_	2.3689*	7.3208*	4.0779*
650X3	_	_	_	_	1.2462	1.7869
650X4	_	_	_	_	_	1.6930

t- tabulated value is 2.101 at 5% level, *indicates calculated > tabulated. Among all combinations, the combination 550/5 is the best since it has significant difference among all other combinations

Table 4: Comparison of various combination of power and time i.e. with in the Groups of Denture Liner reline resin using Student `t` unpaired test

	550X3	550X4	550X5	650X3	650X4	650X5
550X3	_	2.0172	2.0584	1.3089	0.4798	5.3664*
550X4	_	_	0.4347	1.3266	2.0411	2.1660*
550X5	_	_	_	2.0904	2.0676	4.9704*
650X3	_	_	_	_	1.2549	4.3028*
650X4	_	_	_	_	_	9.9679*

t- tabulated value is 2.101 at 5% level, *indicates calculated > tabulated. Among all combinations, the combination 650/5 is the best since it has significant difference among all other combinations

after each set of monomer determinations.

Flexural strength test

All the samples were subjected to flexural strength test in a servo-hydraulic Instron testing machine (ITM), using three-point loading. A crosshead speed of 5 mm/min was used and the distance between the supports was 50mm. Load was applied until fracture, and the fracture load was recorded in newtons (N) by reading the peak of the graph provided by the ITM software. The Flexural Strength (MPa) was calculated using the formula: $FS = 3\ WL/2\ (bd)^2$.

Statistical analysis

The data were evaluated statistically using student's t test and one-way analysis of variance. The data of the material were analyzed using student t test to study the significance of flexural strength differences resulting from the various power and time combination with in the group and also between the two companies One way analysis of variance was used to evaluate the significance of flexural strength keeping the three different time factors as constant and varying the power for each.

Table 5: Comparison of Mean flexural strength between Kooliner and Denture Liner reline resin using students 't' test

Various combinations	t values
550/3	4.04*
550/4	1. 08
550/5	4.69*
650/3	2.13*
650/4	9.89*
650/5	0.65

t- tabulated value is 2.101 at 5% level, *indicates calculated > tabulated. Since Kooliner gave better average flexural strength values than Denture Liner for more combinations, it can be concluded that Kooliner offers better flexural strength.

Table 6: Analysis of variance (ANOVA) test has been used to compare the groups with the same time variable for Kooliner reline resin

Groups	f-value
550X3 / 650X3	3.6714
550X4/ 650X4	0.3168
550X5/ 650X5	16.6278*

f tabulated value is 4.41 at 5% level of significance. *indicates f calculated > f tabulated. There is a significant difference with 550/5 being the best combination.

Table 7: Analysis of variance (ANOVA) test has been used to compare the groups with the same time variable for Denture Liner reline resin

Groups	f-value
650X3 / 550X3	1.7131
650X4/ 550X4	3.0714
650X5/ 550X5	7.6592*

f tabulated value is 4.41 at 5% level of significance. *indicates f calculated > f tabulated. There is a significant difference with 650/5 being the best combination.

RESULTS

Statistically analyzing [Figures 1-4] a comparison between the control group and the other samples subjected to microwave post-polymerization treatment, showed that the control group had the highest residual monomer content and the least flexural strength. The group 550/5 and 650/5 for Kooliner and Denture Liner reline resin had the least amount of residual monomer content and highest flexural strength respectively.

The statistical analysis were performed with student 't' test and values were tabulated in Table 3 for Kooliner

and Table 4 for Denture Liner. Table 5 shows the comparative values between the two companies. The best combination was 550/5 for denture liner and 650/5 for Denture liner. Since Kooliner gave better average flexural strength values than Denture Liner for more combinations, it can be concluded that Kooliner offers better flexural strength.

The analysis of variance (ANOVA) was performed to compare the groups with the same time variable and 'f' value were recorded and tabulated in Table 6 for Kooliner and Table 7 for Denture Liner as 'f' calculated values. Significant 'f' values are designated as * as it is greater than 'f' tabulated values i.e., 4.41 at 5% level. Result showed that there is a significant difference between the 650/5. 650/5 is the best combination for maximal flexural output and minimal residual monomer level for Denture Liner reline resin.

DISCUSSION

During the last few years, curing process have been modified in order to improve the physical and mechanical properties of these materials. Among the several curing methods microwave energy[15,16,21,22] has been with the advantage of reduced time for curing requires a smaller time to obtain the plastic phase, a bigger homogeneity of the mixture and the achievement of a prosthetic material with excellent adaptation.

The flexural strength testing conducted in this investigation is relevant since it reflects the loading arrangement in the clinical situation.[23] The samples were subjected to microwave energy in dry conditions because uptake of water by the acrylic resin would lead to plasticization of the resin, making it more flexible and resilient.[24]

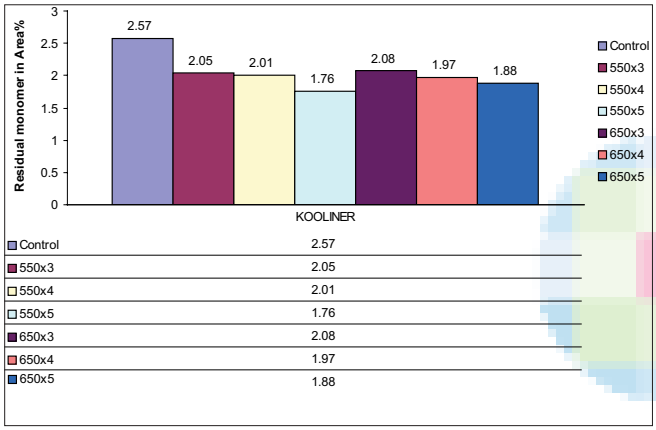

Figure 1: Comparison of residual monomer content of control group with other groups of Koolier reline resin subjected to various power and time cominations after microwave postpoymerization treatment

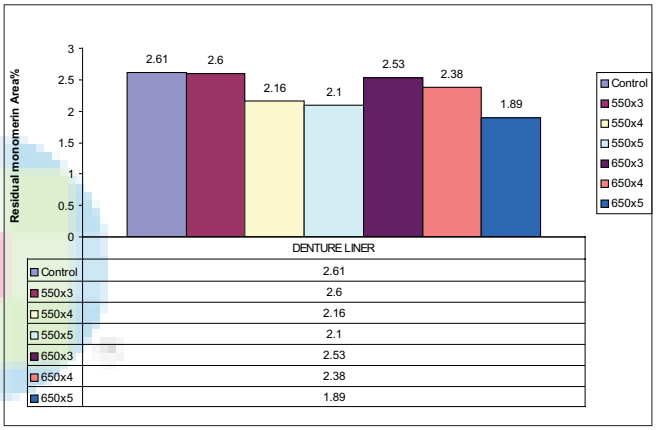

Figure 2: Comparison of residual monomer content of control group with other groups of Denture Liner reline resin subjected to various power and time combination after microwave postpolymerization treatment

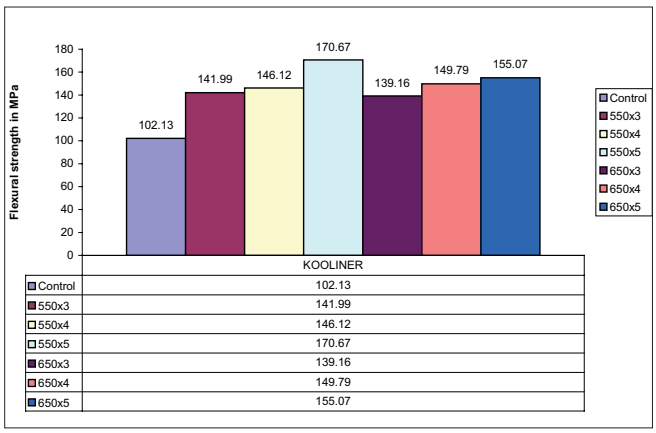

Figure 3: Comparison of mean flexural strength of control group with the other groups of Kooliner reline resin subjected to various power and time combination after microwave postpoymerization treatment

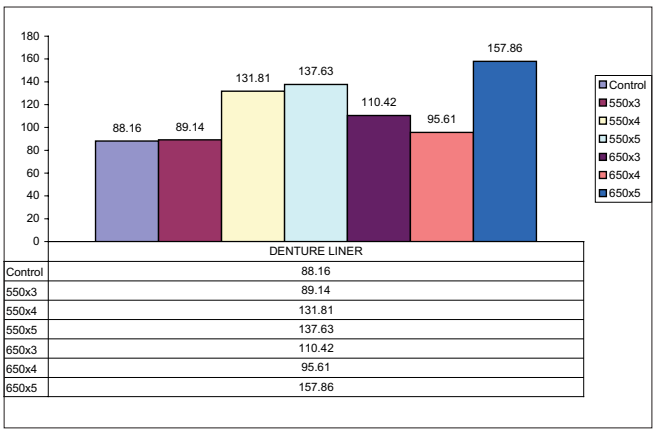

Figure 4: Comparison of mean flexural strength of control group with the other groups of Denture Liner reline resin subjected to various power and time combination after microwave postpolymerization treatment

For the microwave-cured acrylic auto-polymerized reline resins, it has been demonstrated that the temperature developed during the reaction is not constant. It increases quickly at the beginning, goes through a maximum and then decays, being able to reach peaks of the order of 150-200˚C, depending on the working conditions.[25,26] Hence, both the power of the microwave and time of exposure can be regulated to control, in these systems, the rate of polymerization and the degree of conversion.

On evaluation the 550/5 Kooliner samples and 650/5 Denture liner samples revealed the least amount of residual monomer and highest flexural strength. (P less than 0.001) which was in accordance with the study conducted by Vergani *et al*.[27] No further beneficial effect appeared to be gained on extending the polymerization time beyond this as the residual monomer levels remained constant thereafter. In fact, too long and irradiation time caused wrapping, distortion and eventually discoloration, as evident by the specimens irradiated for 9 mins or more than that.[17]

Dogan *et al*,[28] showed that a longer curing period at 100^{0}C decreased the level of residual monomer. Harrison and Huggett[19] reported that a terminal boiling stage assured an optimum in terms of minimal residual monomer. The results from this study corroborate their reports, when comparing the curing process of hot water bath heat and microwave energy used in this study. These findings are in agreement with those reported by Blagojevich and Murphy,[18] who observed that microwave irradiation of an auto-polymerizing acrylic resin, increased the degree of conversion, the impact strength and the glass transition of an auto-polymerizing resin. Similar results have been found by Neisser and Novaes Olivieri[29] when contrasting impact resistance and Knoop hardness of several resins with different polymerization cycles and microwave cured ones. The results of the present study were in contrast with the findings of study conducted by Azzarri *et al*, to evaluate the effect of the different conditions of curing on the residual monomer levels, hardness and impact strength of a microwave polymerized acrylic resin in which no difference were found among the different groups. The difference in results may be due to the method of grouping the samples, where, low power samples were coupled with longer duration of time and vice versa.

The highest flexural strength of the microwave polymerized KRR and DLRR could be related to lowest residual monomer level obtained as a result of higher degree of conversion in accordance to the findings of Harrison *et al*,[30] who showed that the highest level of residual monomer was found when the material was at its weakest.

Thus the present study supports the statement that the highest flexural strength of the microwave polymerized specimens could be related to the lowest residual monomer obtained as a result of a higher degree of conversion. The reason for variation of flexural strength with power and time combination among the samples could be supported by following points:

Since microwave heating is independent of thermal conductivity, this method of di-electric heating raises the temperature rapidly and the inside and outside of the material are equally heated. Hence, additional heat could not prove beneficial as seen in the case of heat cure resins.[19]

The presence of cross-linking agents may be an important factor in the flexural strength observed in the present study. The cross-linking agents of denture base polymers may also affect the residual monomer content of the polymer. The final conversion of Methylmethacrylate (MMA) with an Ethyleneglycol dimethacrylate (EGDMA) cross linking agent decreases with increasing content of the cross-linking agent.[31] This is due to the cross-linked main chain segments which are bound together via cross-linking agent. A rigid polymer structure thus hinders the conversion of MMA monomers especially at curing temperature lower than glass transition temperature. Hence, once this glass transition temperature is reached, further conversion of MMA doesn't take place, thereby limiting the content of residual monomer with no effect of heat thereafter.

Limitations of the study

The methodology used in this study such as microwave curing of the samples with increase in power and time have given the values of flexural strength and residual monomer in exponential range but if the procedure would have been in the fashion of increase in power and decrease in time unit and vice versa then the results could have proved a much better interpretation with respect to importance of curing with microwave energy.

Masticatory load applies to the denture reline material and to the denture base as a whole, rather than to reline material alone. This study did not simulate the intra-oral environment to evaluate the mechanical response of the samples and properties at the reline resin/ denture base resin interface.

CONCLUSION

The mechanical property of the Reline Resins that are microwave post-polymerized depends on the exposition time and microwave power. These two parameters play a pivotal role in achieving best mechanical performance of the material. The polymerization reaction never

reaches 100% conversion and monomer remains free within the material. Modifying one or both parameters could reduce the undesirable effect of free residual monomer.

Within the limitations of the study the following conclusions can be drawn:

For KRR, maximum flexural strength and minimal residual monomer content was offered with the application of microwave irradiation at 550 W for five minutes.

For DLRR, maximum flexural strength and minimal residual monomer content was offered with the application of microwave irradiation at 650 W for five minutes.

The least amount of residual monomer achieved with microwave post-polymerization is related to the highest flexural strength which is inversely proportional.

Kooliner reline resin offered a better flexural strength compared to Denture Liner reline resin.

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