Mechanical Properties of Soft Liner–Poly(methyl methacrylate)-Based Denture Material

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ABSTRACT: In this study, the mechanical properties of two different permanent soft lining materials and their bonding to poly(methyl methacrylate) (PMMA) were compared. Both of the soft liners were heat-cured commercial materials. The polymerization was carried out by conventional methods suggested by manufacturer, and the curing was done at the temperature of boiling water for 5, 15, 25, and 35 min. The sample groups were tested in the computer-aided tensile-testing machine at a rate of 2 mm/min. The slow rate helps the collection of more and more reliable data. At this time, the stress–strain curves were used to calculate ultimate tensile strength, elastic modulus, resilience, and toughness. The measurements were carried for PMMA, Molloplast B, Flexor, and a combination of PMMA/soft liner. After introducing the soft lining material on PMMA of the same thickness, the new material structure was more elastic than the original PMMA. Flexor showed adhesive failure at studied curing periods, but Molloplast B gave larger tear strength values and cohesive rather than adhesive failure at the 25-min and 35-min curing times. © 2002 Wiley Periodicals, Inc. J Appl Polym Sci 85: 467–474, 2002

Key words: adhesive; dental polymers; mechanical properties; poly(methyl methacrylate)

INTRODUCTION

Resilient denture materials are commonly used on dental prosthesis to distribute applied stresses in the patient's mouth more evenly and to have a cushioning effect as a result of their viscoelastic behavior. Plasticized acrylic resin and silicones are the most common materials that have been proposed as denture liners.[1] Their desirable properties are well defined,[2–3] and many studies have reported them.[1,4–6] The clinical application showed that their physical and mechanical properties[1,4,5] are quite suited to being resilient lining materials. The reported data show that the information on these properties is useful in characterizing the performances of soft liners.[5] However, the selection of a particular liner cannot be based on any single property.[4] None of the resilient lining materials has been found to be entirely satisfactory, as a result of their clinic failure. These failures are related to the poor physical and mechanical properties that foul the lining material

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Table I List Of Test Material

Group Number	Description
1	PMMA (QC20)
2	PMMA + Primo Adhesive + Molloplast B
3	PMMA + Adhesive (Flexor Bonding Agent) + Flexor
4	Silicon-based soft liner (Molloplast B)
5	Copolymer-based soft liner (Flexor)

PMMA = poly(methyl methacrylate).

with fungal and bacterial growth and to the poor adhesion of the lining to the denture base.[2] The silicone-based resilient lining materials mostly used in clinical application are commonly found to separate from the acrylic base material.[7] The bond strength between liners and poly(methyl methacrylate) (PMMA) has been studied by peel,[4,8–12] tensile,[8,10–11,13–17] and shear[7,10,18] tests. It has been reported that the bond strength can be improved by modifying the surface topography of PMMA.[8] However, more work has to be done using different parameters and test methods to get information related to the morphology and the surface modification of denture base material in combination with different commercial soft liners.

In this study, PMMA samples that will be compatible with the resilient lining material were prepared. The mechanical properties of PMMA base material, two different resilient lining materials separately, and PMMA combined with each of the two different liner materials using an adhesive[19] for attachment were tested. The properties studied were ultimate tensile stress, percentage elongation, elasticity moduli, resilience, and toughness. The bond (tear) strength between PMMA and resilient liners was measured by the peel test. The distribution of adhesive on the surface of PMMA and the resilient lining was observed by stereomicroscope.

EXPERIMENTAL

Materials and Methods

The prosthetic base materials used were heat-cured type QC 20 (DeTrey Dentsply Ltd., Wegbridge, Surrey, UK) methyl methacrylate, MMA powder polymer, and liquid monomer. These materials were used as received. Liner materials used were heat-polymerized elastomeric silicon (Molloplast-B, Deyax, Karl Huber GmBH and Co KG. Ettlinger, Germany) and copolymer silicon (Flexor, Schutz Dental, Rosbach, Germany).

The stress–strain tests were carried on the Lloyd LS 500K test machine. For the stress–strain tests, the dumbbell-shaped test specimens 65 × 10 × 3 mm in size were prepared according to ASTM 0638M. There were five groups (Table I) of test material, with four samples for each test group for statistical estimations, and four measurements for each sample of a group were carried out. Thus, the total number of samples prepared was 80. For the first group, the 3 : 1 mixture of powder/liquid PMMA was mixed into a dough and heated at 60°C for 30 min. It was then placed completely in a previously made mold of proper size (4 × 4 cm for each group) and adjusted to a thickness of 3 mm by placing a brass cover of a thickness equal to the thickness of the liner (both Molloplast and Flexor). For the next two groups, the thickness of the PMMA was adjusted to 3 mm and the rest of the mold filled with a liner of

Table II Mechanical Test Data for QC20 Samples

Curing (min)	UTS (MPa)		Elongation (%)		Elastic Modulus (MPa)		Resilience (mMN/m^2)		Toughness (mMN/m^2)	
n = 4	Average	SD	Average	SD	Average	SD	Average	SD	Average	SD
5	13.913	1.869	0.145	0.025	122.403	24.716	0.211	0.093	2.411	0.164
15	25.243	8.225	0.078	0.021	363.775	79.900	0.384	0.346	0.945	0.621
25	41.910	8.678	0.105	0.010	546.745	45.833	0.437	0.250	0.906	0.190
35	47.145	3.743	0.122	0.029	529.670	110.540	0.737	0.104	3.824	1.805

SD = standard deviation.

Table III Mechanical Test Data for QC20+Molloplast B

Curing Time (min) $n = 4$	UTS (MPa)		Elongation (%)		Elastic Modulus (MPa)		Resilience (mMN/m^2)		Toughness (mMN/m^2)	
	Average	SD	Average	SD	Average	SD	Average	SD	Average	SD
5	14.875	1.348	0.700	0.151	63.854	26.206	0.711	0.366	1.359	0.144
15	12.397	2.017	1.087	0.297	42.706	8.339	1.301	0.691	1.977	0.781
25	20.740	5.618	0.645	0.057	69.355	12.652	2.276	1.884	3.549	0.558
35	14.862	1.219	1.035	0.752	61.365	12.551	0.437	0.232	7.486	5.432

SD = standard deviation.

desired thickness (3 mm). The last two group samples were made in similar mold, using both Molloplast and Flexor as only material with same thickness (3 mm) as in the one combined to PMMA. Thus, the thickness of PMMA and the liner when combined was the same as their thicknesses when studied alone. The materials tested are summarized in Table I. After being pressed for 10 min under high pressure,[20] a set of four of each group was cured at the temperature of boiling water for 5-, 15-, 25-, and 35-min periods. The finishing of the samples was done before measuring, as suggested by the manufacture of the raw materials (producing a smooth surface).[21–24] The stresses applied were 10, 500, and 2500 N. The rate of elongation was 2 mm/min. A computer program processed the data to calculate elastic modulus, and ultimate tensile strength (UTS). The area under the stress–strain curves in the region of elastic material was determined as resilience in mMN/m^3. The curve-fit applications were used on a fourth-degree polynomial to find the functional curve equation, and the integral of the area under that curve gave toughness in mMN/m^3. The Matched 7.0 program was used for the calculations.

The peel tests were done on samples of dimensions $10 \times 25 \times 6$ mm. The sample was placed on a Lloyd's tensile-testing machine with the PMMA layer in the upper clamp and the soft liner material in the lower one. This gives an angle of 180° for peeling the liner, which was done at a rate of 2 mm/min. The load, F, was read from the computer output. The peel bonding (tear) strength P (in N/mm) were calculated from

$$P = 2F/W,$$

where W is the width of the bonded surface (in mm). The average and standard deviations were calculated for four samples in each group. After determination of the tear strength between PMMA and the soft liner, the stereomicroscopic (Nikon, SM2-2T) photographs of the material surface were taken to observe the adhesive bonding and distribution between them.

RESULTS AND DISCUSSION

The stress–strain results for QC20 samples cured at different periods are given in Table II. The UTS

Table IV Mechanical Test Data For QC20+Flexor

Curing (min), $n = 4$	UTS (MPa)		Elongation (%)		Elastic Modulus (MPa)		Resilience (mMN/m^2)		Toughness (mMN/m^2)	
	Average	SD	Average	SD	Average	SD	Average	SD	Average	SD
5	7.910	2.268	1.232	0.433	62.075	0.186	0.283	0.212	2.211	1.524
15	12.697	4.179	0.745	0.172	54.218	32.598	0.994	0.972	1.755	0.880
25	20.120	1.370	1.287	0.481	49.520	12.059	2.908	1.668	6.315	5.974
35	24.167	5.228	1.287	0.257	51.078	12.271	2.462	2.462	7.002	1.723

SD = standard deviation; UTS = ultimate tensile strength.

Table V Mechanical Test Data for Molloplast B

Curing (min)	UTS (MPa)		Elongation (%)		Elastic Modulus (MPa)		Resilience (mMN/m^2)		Toughness (mMN/m^2)	
$n = 4$	Average	SD	Average	SD	Average	SD	Average	SD	Average	SD
5	. . .	. . .	. . .	. . .	. . .	. . .	. . .	. . .	. . .	. . .
15	1.963	0.405	6.732	1.442	1.219	0.162	0.045	0.011	20.589	14.593
25	1.459	0.341	2.395	0.716	1.664	0.275	0.033	0.017	1.9663	0.714
35	1.732	0.232	2.985	0.538	1.613	0.134	0.025	0.019	2.301	0.146

SD = standard deviation; UTS = ultimate tensile strength.

increases with curing time. The reported values[2] for PMMA are in the range of 48.3–62.1 MPa. Therefore, the UTS value of 47 MPa for a 35-min curing period is close to the range, but the crosslinking of the samples is still not completed and higher curing periods are needed. However, it is in good agreement with our previously reported values.[25] The elongation percentage of uncrosslinked PMMA reported[2] is in the range of 1–2%, which is almost 10 times larger than the values obtained in this work. This is because the samples in this work are highly crosslinked and have no tendency toward elongation. The elongation percentage shows fluctuation with the curing period, which shows the different percentages and types of crosslinking. The elastic modulus increased with curing time. These values are larger than that of the values we have reported before[25] but smaller than some other reported values.[2] Thus, depending on preparation conditions, the properties of the material might be different. The resilience increased with curing time. The material became tougher and more brittle when cured for about 35 min.

The data for the mechanical properties of the combination of prosthetic base material and PMMA (QC20)/laminated lining material together (Molloplast B) are given in Table III. The ultimate tensile stress did not change much with the curing period, but the values are much smaller compared to those of PMMA without liner (Table II). In this case, the elongation percentages are much larger and the elastic modulus of PMMA/laminated lining material smaller than that of PMMA alone. Both the elongation percentages and the elastic modulus did not change much with an increase in curing time. However, the values of resilience and toughness are relatively higher for this material, and both values increased with curing time with the exception of resilience for the 35-min curing period, which can be explained simply as the material becoming more brittle by further curing. The toughness decreased during the 15- and 25-min curing periods. There was probably further polymerization, giving less crosslinking at this stage. However, with a longer curing period (35 min), the crosslinking predominated and toughness increased.

The mechanical test results for the prosthetic base material PMMA (QC20)/Flexor together (laminated) are given in Table IV. For this mate-

Table VI Mechanical Test Data for Flexor

Curing (min)	UTS (MPa)		Elongation (%)		Elastic Modulus (MPa)		Resilience (mMN/m^2)		Toughness (mMN/m^2)	
$n = 4$	Average	SD	Average	SD	Average	SD	Average	SD	Average	SD
5	. . .	. . .	. . .	. . .	. . .	. . .	. . .	. . .	. . .	. . .
15	0.027	0.025	0.790	0.739	0.064	0.067	0.001	0.001	0.054	0.104
25	5.000	1.633	11.350	1.000	0.781	0.197	0.060	0.014	27.503	10.655
35	5.712	2.618	9.125	2.018	1.658	0.035	0.035	0.019	14.357	7.452

SD = standard deviation; UTS = ultimate tensile strength.

Table VII Tear Strength of Resilient Liners

Curing Time (min) $n = 4$	Load (N)		Tear Strength (N/mm)			
			OC20+Mollopsat		QC20+Flexor	
	Average	SD	Average	SD	Average	SD
5			No measurements			
15	3.445	2.144	0.688	0.428	—	
15	2.559	0.201			0.511	0.040
25	2.703	2.200			0.540	0.439
35	2.727	0.345			0.545	0.068

SD = Standard deviation.

rial, the ultimate tensile stress, resilience, and toughness increased with curing time, but not systematic changes were observed in the elongation percentage and the elastic modulus. The values are not much different when Molloplast B and Flexor are used as the soft liner material.

The mechanical test results for Molloplast B are given in Table V. The material obtained after the 5-min curing was very soft, and mechanical test measurements could not be made for this sample. After the 15-min curing period, the material became strong enough to take the data. However, the high values of elongation percentage and toughness at the 15-min curing period decreased with further curing. The UTS and elastic modulus did not change much with curing time. Similar results were also reported by other workers.[4–6,26–30] The elastic modulus for Molloplast B measured by this method has not been reported before. The manufacturer's suggestion for the curing time of this type material is 150 min. However, similar properties were obtained after curing it for 35 min in this work.

The mechanical test results for Flexor are given in Table VI. In this case, the measurements could not be taken for material cured for 5 min, and very small values were observed for the 15-min curing. The values observed for UTS, elongation percentage, resilience, and toughness are much larger for Flexor than for Molloplast B. Elastic moduli are similar after the 35-min curing period.

The Peel tests results for the tear strength of QC20+Molloplast B and QC20+Flexor are given in Table VII. For both soft liner materials, the measurements after the 5-min curing period could not be taken because of material failure. The tear strength for Flexor did not change much with curing time. Similar results with slightly larger values were obtained for Mollo-

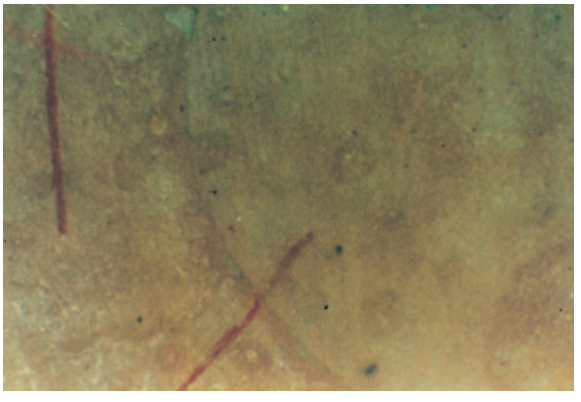

Figure 1 Stereomicrograph of poly(methyl methacrylate) surface after it was peeled from Molloplast B cured for 15 min.

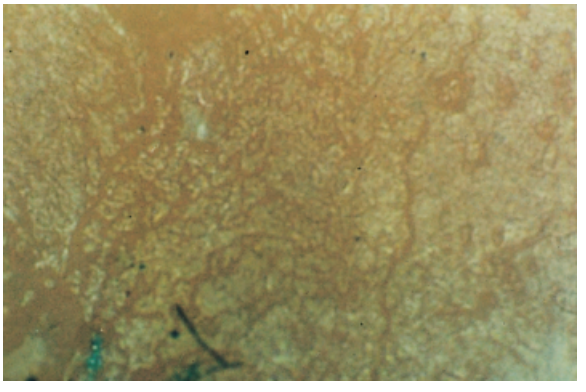

Figure 2 Stereomicrograph of Molloplast B surface after it was peeled from poly(methyl methacrylate) cured for 15 min.

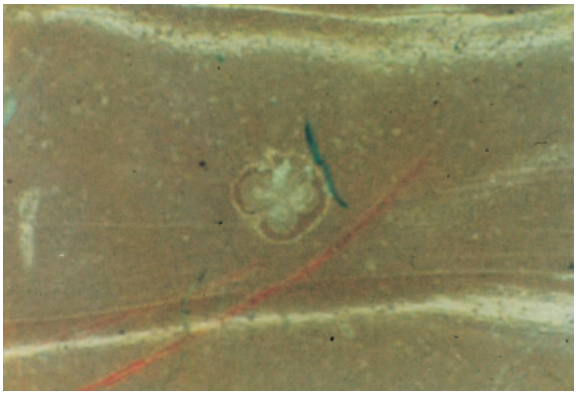

Figure 3 Stereomicrograph of poly(methyl methacrylate) surface after it was peeled from Molloplast B cured for 25 min.

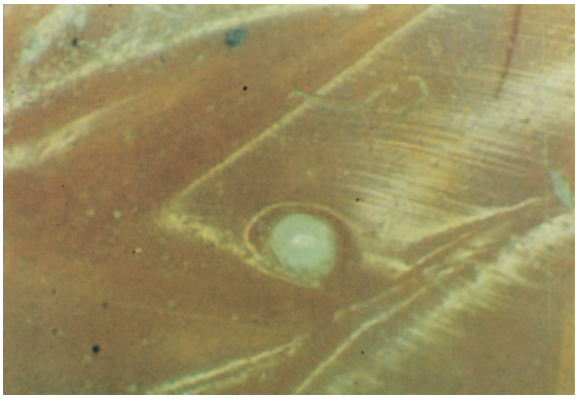

Figure 5 Stereomicrograph of poly(methyl methacrylate) surface after it was peeled from Molloplast B cured for 35 min.

plast B. Adhesive failure between the liner and the denture base material results in bacterial growth that causes the prosthesis to deteriorate. In this work, Primo adhesive and Flexor bonding agent were used as adhesive for Molloplast B and Flexor, respectively, as suggested by the manufacturers. The magnitude of the tear strength indicates how easily the liner will tear away.[4,19] The observed values in this work after a 35-min curing period are smaller than some of the reported[4,7,11,29] values, which were measured after 2 h of curing.

The distribution of adhesive and the surface morphology of the denture base material are important for an improved soft liner attachment material. To see how the adhesive is attached to the soft liner and the base material, surface stereomicrographs of the peeled soft liner and the denture base material were taken and can be seen in Figures 1–8. The adhesive seen in the photographs as a in the second phase, and the surface of the material gives information about any surface deformation caused by tearing the two surfaces (PMMA and soft liner) away from each other. The distribution of adhesive is independent of the dimensional orientation. For QC20+ Molloplast B, the adhesive is tightly attached and distributed on the PMMA (QC20) surface but not on the soft liner (Molloplast B) for the 15-min curing (Figs. 1–2). However, for the 25-min (Figs. 3–4) and 35-min (Figs. 5–6) curings, the adhesive is evenly attached to the surface of both soft liner and PMMA. In the case of Flexor attached to PMMA (QC20), the attachment of the adhesive to the surfaces of both the soft liner (Flexor) and PMMA (QC20) is very poor and uneven for all

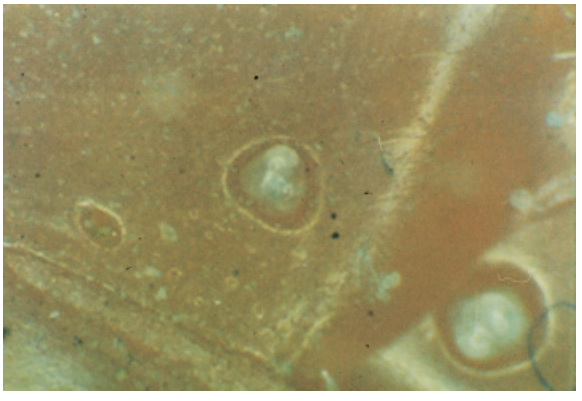

Figure 4 Stereomicrograph of Molloplast B surface after it was peeled from poly(methyl methacrylate) cured for 25 min.

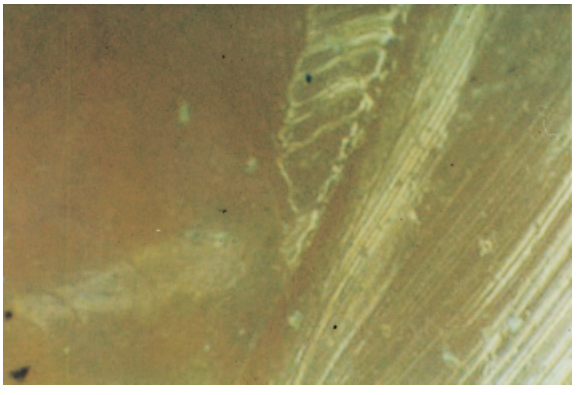

Figure 6 Stereomicrograph of Molloplast B surface after it was peeled from poly(methyl methacrylate) cured for 35 min.

curing periods (Figs. 7–8). Therefore, the failure of this adhesive may create an environment for potential bacterial growth and a deteriorating prosthesis when PMMA+Flexor are used.

CONCLUSION

The results obtained that can be concluded are as follows: First, the UTS, elastic modulus, and resilience for PMMA used as dental base material and cured at different time intervals showed an increase with curing time, but the elongation percentage remained almost the same and toughness first decreased then increased with curing time. The material became more brittle with an increasing in curing time.

Second, the UTS of silicon-based Molloplast B did not change much with curing time and it showed the highest resilience after the 15-min curing. The UTS for copolymer-based Flexor was highest for the 25- and 35-min curings and was better than that of Molloplast B. Flexor's highest resilience was obtained after the 25-min curing.

Third, the mechanical properties of the PMMA/soft liner combination have not been reported before. Their values are in between those of PMMA and the liner. This makes the PMMA more elastic.

Finally, adhesive compatibility between PMMA and Flexor is not good in any studied curing period but is much better between PMMA and Molloplast B for the 25- and 35-min curings.

Figure 7 Stereomicrograph of poly(methyl methacrylate) surface after it was peeled from Flexor cured for 35 min.

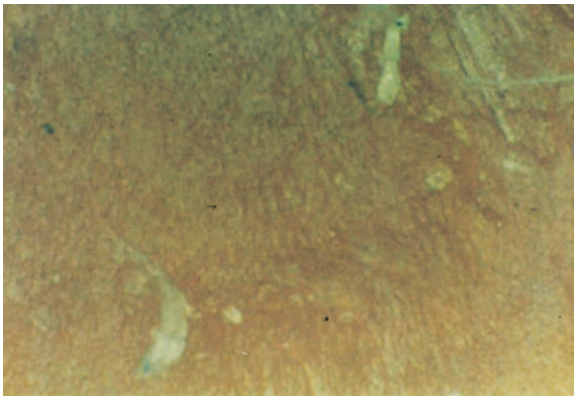

Figure 8 Stereomicrograph of Flexor surface after it was peeled from poly(methyl methacrylate) cured for 35 min.

REFERENCES

1. Van Fraunhofer, J. A.; Sichina, W. J. Int J Prosthodont 1994, 7, 120.
2. Craig, R. G. Restoration Dental Materials, 9th ed.; Mosby: St. Louis, MO, 1993.
3. Phillips, R. W. Skinner's Science of Dental Materials, 9th ed.; W.B. Saunders: Philadelphia, 1991.
4. Craig, R. G.; Gibbons, P. J. Am Dental Assoc 1961, 63, 382.
5. Dootz, E. R.; Koran, A.; Craig, R. G. J Prosthet Dent 1992, 65, 707.
6. Dootz, E. R.; Koran, A.; Craig, R. G. J Prosthet Dent 1993, 69, 114.
7. Al Athel, M. S.; Jagger, R. G.; Jerolimov, V. Int J Prosthdont 1996, 9, 167.
8. Al Athel, M. S.; Jagger, R. G. J Prosthet Dent 1996, 76, 535.
9. Jacobsen, N. L.; Mitchell, D. L.; Johnson, D. L.; Holt, R. A. J Prosthet Dent 1997, 78, 153.
10. Amin, W. M.; Fletcher, A. M.; Ritchi, G. M. J Dent 1981, 9, 336.
11. Kutay, O. J Prosthet Dent 1994, 71, 525.
12. Wright, P. S. J Dent Res 1982, 61, 1002.
13. Kutay, O.; Bilgin, T.; Sahos, O.; Beyli, M. Eur J Prosthodont Res 1994, 2, 123.
14. Bates, J. P.; Smith, D. C. J Am Dent Assoc 1965, 70, 344.
15. Emmer, T. J.; Vaidynathes, J.; Vaidynathes, T. K. J Prosthet Dent 1995, 74, 595.
16. Khan, Z.; Martin, J.; Collard, S. J Prosthet Dent 1989, 62, 196.
17. El-Hadary, A.; Drummond, J. L. J Prosthet Dent 2000, 83, 356.
18. Polyzois, G. L. J Prosthet Dent 1992, 68, 854.
19. Baysan, A.; Parker, S.; Wright, P. S. J Prosthet Dent 1998, 79, 182.

20. Muhtaroğulları, I. Y.; Doğan, A.; Muhtaroğulları, M.; Usanmaz, A. J. Appl Polym Sci 1999, 74, 2971.
21. Radford, D. R.; Watson, T. F.; Walter, J. D.; Challacombe, S. J. J Prosthet Dent 1997, 77, 200.
22. Jacobsen, N. L.; Mitchell, D. A.; Johnson, D. L.; Holt, R. A. J Prosthet Dent 1997, 78, 153.
23. Reesen, M. G.; Jepson, N. J. A. J Prosthet Dent 1998, 79, 355.
24. Dogan, A.; Bek, B.; Çevik, N. N.; Usanmaz, A. J Dent 1995, 25, 313.
25. Usanmaz, A.; Türker, F.; Doğan, A.; Akkaş, N. J Appl Polym Sci 1998, 69, 1409.
26. Honorez, P.; Catalan, A.; Angnes, U.; Grimonster, J. J Prosthet Dent 1989, 61, 510.
27. Robinson, J. G.; McCabe, J. F. J Prosthet Dent 1982, 48, 135.
28. Kawano, F.; Dootz, E. R.; Craig, R. G. Int J Prosthodont 1997, 10, 178.
29. Kawano, F.; Dootz, E. R.; Koran, A.; Craig, R. G. J Prosthet Dent 1992, 68, 368.
30. Von Fraunhofer, J. A.; Sichina, W. J. Int J Prosthodont 1994, 7, 120.