Effect of Overall Powder-to-Liquid Ratios on Surface Hardness and Initial Viscoelasticity of A Short-Term Soft Liner Incorporated with *Litsea* Cubeba Oil

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Abstract

The aim of this study was to evaluate surface hardness and initial viscoelasticity of short-term denture soft liner incorporated with Litsea cubeba essential oil. The nine groups with different compositions in overall powder-to-liquid ratios (P/L = 0.9, 1.2, and 1.5) and L. cubeba oil concentrations (0, 10, and 20 %w/w) were investigated. The material hardness was determined at 2 hours and 7 days by Shore A0 durometer. The initial viscoelastic properties, including gelation time, storage (or elastic) modulus and loss (or viscous) modulus were investigated by an oscillating plate rheometer until 30 minutes after mixing the materials. Five independent experiments were done thorough the study. The surface hardness of the soft liner with and without L. cubeba oil was mainly influenced by overall P/L ratio. The soft liners with higher P/L ratio significantly showed greater surface hardness at both 2 hours and 7 days (p<0.05). The addition of L. cubeba oil at 10 %w/w did not affect the material hardness. Whereas, high concentration (20 %w/w) of L. cubeba oil significantly decreased Shore A0 hardness at 2 hours (p<0.05). The gelation time, initial elastic and viscous moduli of the soft liners were also contributed by the overall P/L ratios. The soft liner compositions with higher overall P/L ratio showed the shorter gelation times and higher initial moduli (p<0.05). No statistical effect of the L. cubeba oil concentration on the initial viscoelastic properties was observed. In conclusion, the surface hardness and initial viscoelasticity of the short-term soft liner were dominantly influenced by the overall P/L ratios. The incorporation of L. cubeba oil concentration did not dramatically affect the material hardness and initial viscoelastic properties.

***Keywords:** Soft liner/ Hardness/ Viscoelastic/ Litsea cubeba/ Essential oil*

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Introduction

Denture soft liner has been used in prosthodontic fields to treat abused tissues, improve the fit of ill-fitting dentures, prevent trauma from a sharp ridge or atrophic mucosa, and distribute the masticatory force.[1,2] By duration of usage, the materials could be categorized in two types, short-term and long-term soft liners.[1] For the short-term soft liners, the materials are commonly used as tissue conditioners or as temporary soft lining materials for around 7 days.[1]

Basic chemical compositions of the short-term soft liners are plasticized acrylic resins. The materials are applied beneath the denture base to improve fitness of dentures and absorb masticatory force. This would make the mucosa return to a healthy condition.[1,3] However, the materials are easily degradable and susceptible to microbial colonization.[4,5] One attractive method is an incorporation of antimicrobial agents into short-term soft liners to extend their clinical longevity and prevent microorganism proliferation.[6] Many kinds of additives have been applied including well-known polyene or azole drugs such as Nystatin, Amphotericin B, miconazole, ketoconazole,[7-9] metallic oxides,[10,11] and natural products.[12-14]

Recently, several essential oils derived from plants and fruits could improve the anti-microbial properties of soft lining materials.[12-14] However, some studies found that the incorporation of antimicrobial additives into the soft lining materials could also

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influence other material properties such as surface hardness, surface roughness, tensile strength, setting time, and viscoelasticity.[14-16] Such alteration effects from the additive could be caused by two main reasons. First, the essential oils would decrease the overall powder-to-liquid (P/L) ratio of the soft lining material because of their liquid-like nature. Another reason might be a plasticization or softening effect of the essential oil on polymer networks of the soft lining materials.[16]

Herein, an essential oil from *Litsea cubeba* was introduced as an additive into a short-term soft liner. *L. cubeba* or mountain pepper is a plant distributed in large Asiatic areas. It has been used as herbal medicines or aromatherapies. Its essential oil has been reported about various pharmacological effects including anti-fungal activity, anti-bacterial activity, and anti-inflammation, etc.[17-20] Although benefits of *L. cubeba* oil on denture soft liner could be expected, an influence of this additive on the material properties has to be examined.

In this research, a surface hardness and initial viscoelasticity of a short-term soft liner after incorporating with *L. cubeba* oil were evaluated. Two variables including the essential oil concentration and overall P/L ratio were investigated. For the null hypothesis, there is no difference in surface hardness and initial viscoelastic properties of the short-term soft liner with different oil concentrations and overall P/L ratios.

Materials and methods

The essential oil and soft lining material

L. cubeba essential oil was purchased from Thai-China Flavours and Fragrances Industry Co. Ltd., Nonthaburi, Thailand. It was extracted by hydrodistillation technique from its fruits. Main chemical constituents of *L. cubeba* oil analyzed by the product supplier were citral and *D*-limonene with 60% and 17%, approximately.

The short-term acrylic soft liner, GC Soft Liner™ (GC corporation, Bunkyo-ku, Tokyo, Japan), was used in this study. Its powder contains poly(ethyl methacrylate) (PEMA) with an average molecular weight of 2.34×10^5 g/mol.[21] Major compositions of GC soft liner liquid are butylpthalylbutyl glycolate (BPBG;~81%) and ethanol (~15%). Density of the liquid at 25°C is 0.9 g/mL.[22]

There were nine experimental groups with different compositions in overall P/L ratios and oil concentrations. The liquid of GC soft liner was priorly mixed with a certain amount of *L. cubeba* oil. The powder was manually mixed with prepared liquid. The amount of all compositions for initial viscoelastic testing was shown in Table 1. For hardness testing, all experimental groups with similar proportions as the viscoelastic testing were also investigated. The concentrations of *L. cubeba* oil to total liquid were assigned as percentages by weight.

Table 1 Various compositions of GC soft liner incorporated with *L. cubeba* oil at different oil concentrations and overall powder-to-liquid (P/L) ratios for initial viscoelastic testing.

Group	*L. cubeba* oil (mg)	GC soft liner Liquid (mg)	GC soft liner Powder (mg)	% *L. cubeba* oil to total liquid	Overall P/L (w/w)
P/L0.9_control	0	500	450	0% w/w	0.9 : 1
P/L0.9_LC-10	55	500	499.5	10% w/w	0.9 : 1
P/L0.9_LC-20	125	500	562.5	20% w/w	0.9 : 1
P/L1.2_control	0	500	600	0% w/w	1.2 : 1
P/L1.2_LC-10	55	500	666.7	10% w/w	1.2 : 1
P/L1.2_LC-20	125	500	750	20% w/w	1.2 : 1
P/L1.5_control	0	500	750	0% w/w	1.5 : 1
P/L1.5_LC-10	55	500	832.5	10% w/w	1.5 : 1
P/L1.5_LC-20	125	500	937.5	20% w/w	1.5 : 1

Shore A0 surface hardness testing

The surface hardness of the soft liners with all compositions was determined by Shore A0 hardness durometer according to ISO 10139-1:2018.[23] All soft liner compositions were prepared as mentioned previously at room temperature. The samples were then loaded into the stainless steel mold to fabricate the specimens with 55 mm diameter and 8 mm depth.[15] After 15 minutes, the test specimens were immersed into the 100 mL water-filled water bath at 37°C for 2 hours. Then, the specimens were removed from the mold and determined the Shore A0 hardness. After that, the specimens were re-immersed into the water bath for 7 days and then measured the hardness again but on the opposite side. Five specimens (n=5) of each group were tested.

Evaluation of the initial viscoelasticity

The initial viscoelastic property of all groups of the soft liners was evaluated by an oscillating rheometer (Bohlin gemini HR nano rheometer, Marvern, UK). The parallel plate geometry was 20 mm in diameter. An oscillating frequency was set at 1 Hz. The stress was controlled at 40 Pa and the temperature was set at 37°C. All samples were manually mixed at room temperature. The material was placed on the lower geometry of the rheometer, then the upper plate was lowered on its guide into the position with 1 mm gap between the plates. Shear stress and shear strain were monitored for 30 minutes.

The complex shear modulus (G*) is calculated from the shear stress (τ) and shear strain (γ). The phase lag between stress represents as δ. The shear storage modulus (G'), shear loss modulus (G"), and loss tangent or damping coefficient (tan δ) can be determined as the following equation;[24]

$$|G^*| = \tau/\gamma$$

$$G^* = G' + iG'' \text{ (where } i^2 = -1)$$

$$G' = |G^*| \cdot \cos\delta$$

$$G'' = |G^*| \cdot \sin\delta$$

$$\tan\delta = G''/G'$$

G' and G" could also be called as elastic and viscous moduli. Gelation time or 'gel point' can be defined as the time point when G' and G'' are equal (tan δ = 1) as shown in Figure 1.[24] The initial elastic and viscous moduli at different time points (5 to 30 minutes) after mixing were also evaluated. Five independent tests (n=5) were carried out for each group.

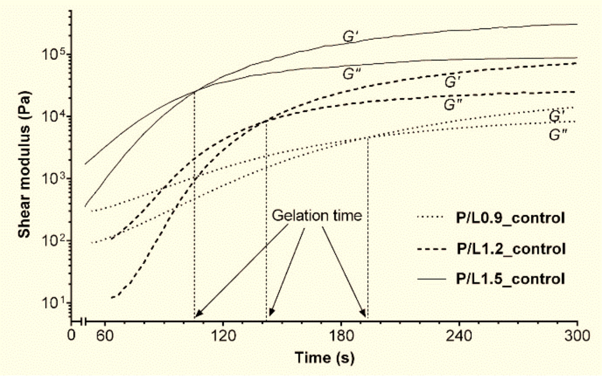

Figure 1 The initial viscoelasticity of GC soft liner as a function of time after mixing. G' and G" represented the shear elastic and viscous moduli. The gelation times of the soft liners with different compositions could be defined as the time at the intersect point (G' = G").

Statistical analysis

All statistical computations were performed by SPSS software (IBM corp. released 2013, IBM SPSS statistics for Windows, version 22.0, IBM, Armonk, NY, USA). The significant level was set at α=0.05.

Shapiro-Wilk test was performed to validate normality of the data and Levene's test for equality of variances between group data. An influence of the oil concentration and overall P/L ratio on the gelation time and surface hardness and interaction between these two variables were analyzed by two-way analysis of variance (ANOVA). The multiple comparisons between groups were performed by Tukey's test when the data met the homogeneity of variances assumption or by Games-Howell *post-hoc* test for unequal variance situation. In addition, an alteration in the material hardness from 2 hours to 7 days was analyzed by dependent *t*-test. The significance level was set at α=0.05.

Results

Surface hardness of different compositions of GC soft liner incorporated with *L. cubeba* oil

Shore A0 hardness of all groups of the soft liners was measured at the initial stage (2 hours) and late stage (7 days) as shown in Table 2 and 3. All samples showed a statistically significant increase in the hardness after aging process ($p<0.05$; dependent *t*-test). From the two-way ANOVA, there was no interaction between the oil concentration and overall P/L ratio on the material hardness ($p>0.05$). The overall P/L ratio significantly affected the surface hardness of GC soft liner with and without *L. cubeba* oil ($p<0.05$). While, the oil concentration significantly influenced only on the hardness at 2 hours but not affect the hardness at 7 days.

Table 2 Shore A0 hardness at 2 hours of GC soft liner with different compositions in overall powder-to-liquid (P/L) ratios and concentrations of *L. cubeba* oil.

Overall P/L	Shore A0 hardness (Mean ± SD) at 2 hours*		
	No additive	10%w/w *L. cubeba* oil	20%w/w *L. cubeba* oil
0.9	$9.88 \pm 1.50^{A; a}$	$9.04 \pm 0.86^{D; a}$	$6.28 \pm 0.87^{G; b}$
1.2	$19.96 \pm 1.09^{B; c}$	$18.68 \pm 2.32^{E; c}$	$16.36 \pm 0.52^{H; d}$
1.5	$28.36 \pm 1.35^{C; e}$	$26.52 \pm 1.10^{F; e,f}$	$26.24 \pm 0.52^{I; f}$

*Mean pairs with different superscripts represented statistical differences ($p < 0.05$).
Capital letters indicate statistical differences between groups in same columns (same P/L ratios).
Lower case letters indicate statistical differences between groups in same rows (same oil concentrations).

Table 3 Shore A0 hardness at 7 days of GC soft liner with different compositions in overall powder-to-liquid (P/L) ratios and concentrations of L. cubeba oil.

Overall P/L	Shore A0 hardness (Mean ± SD) at 7 days*		
	No additive	10%w/w *L. cubeba* oil	20%w/w *L. cubeba* oil
0.9	17.64 ± 1.55^{A}	17.64 ± 1.02^{D}	17.08 ± 1.31^{G}
1.2	26.32 ± 0.97^{B}	25.96 ± 0.57^{E}	25.84 ± 1.18^{H}
1.5	34.32 ± 0.91^{C}	34.88 ± 0.92^{F}	33.76 ± 0.80^{I}

*Mean pairs with different superscripts represented statistical differences among various P/L ratios ($p<0.05$). There were no statistical differences in comparisons between groups in same rows.

At 2 hours, the Shore A0 hardness values of GC soft liner control with overall P/L ratios at 0.9, 1.2, and 1.5 were about 10, 20, and 28, respectively. The addition of only 10% w/w *L. cubeba* oil into GC soft liner with all P/L ratios did not significantly alter the hardness at 2 hours ($p>0.05$). Nevertheless, the incorporation of 20% w/w *L. cubeba* oil statistic significantly diminished the Shore A0 hardness at 2 hours of the soft liner with all P/L ratios ($p<0.05$).

Whereas, the incorporation of *L. cubeba* oil at both 10 and 20% w/w did not influence the surface hardness at 7 days of the soft liner with all P/L ratios ($p>0.05$). Shore A0 hardness values at 7 days of the soft liners were about 17, 26, and 34 for the overall P/L ratios at 0.9, 1.2, and 1.5, respectively.

Initial viscoelasticity of different compositions of GC soft liner incorporated with *L. cubeba* oil

Three initial viscoelastic properties including gelation time, shear elastic, and viscous moduli were evaluated in this study. The alteration by the time of G' and G" after mixing GC soft liner control with different P/L ratios was illustrated in Figure 1. The gelation time of the soft lining materials could be defined as the time at the intersect point (G'=G").

The soft liner control with overall P/L ratios at 0.9, 1.2, and 1.5 displayed the gelation times about 200, 150, and 100 seconds, respectively (Table 4). From the two-way ANOVA, there was no significant interaction between the overall P/L ratio and oil concentration on the gelation time (p>0.05). The addition of *L. cubeba* oil slightly decreased the gelation time of GC soft liner but there was, however, no statistical difference from the soft liner without additive. An increase in the overall P/L ratios statistic significantly shortened the gelation time of GC soft liner without additive. For GC soft liner with 10% and 20% oil content, the gelation time was significantly reduced when the overall P/L ratio increased from 0.9 to 1.2 (p<0.05).

Table 4 Gelation time of GC soft liner with different compositions in overall powder-to-liquid (P/L) ratios and concentrations of *L. cubeba* oil.

Overall P/L	Gelation time (s)*		
	No additive	10%w/w *L. cubeba* oil	20%w/w *L. cubeba* oil
0.9	205.4 ± 27.65^{A}	203.0 ± 43.39^{D}	180.8 ± 7.43^{F}
1.2	151.2 ± 45.48^{B}	123.0 ± 14.39^{E}	120.2 ± 13.61^{G}
1.5	100.2 ± 16.16^{C}	102.2 ± 9.20^{E}	98.6 ± 11.65^{G}

*Mean pairs with different superscripts represented statistical differences among various P/L ratios (*p*<0.05). There were no statistical differences in comparisons of gelation times between groups in same rows.

For initial shear moduli, both G' and G" of all samples dramatically increased during the first 2-4 minutes and then gradually increased until almost stable values. From the two-way ANOVA of each time point, there was no significant interaction between the overall P/L ratio and oil concentration on the shear moduli for all time points (p>0.05). The soft liners with a higher P/L ratio showed significantly greater elastic moduli at all time points (300 to 1800 seconds) after mixing (Table 5, p<0.05; multiple comparisons of each time point). The higher P/L ratio also led to higher viscous moduli especially at 300 and 600 seconds (Table 6). The addition of *L. cubeba* oil slightly influenced G' and G" of the soft liner but no statistical difference was observed.

Table 5 Shear elastic modulus (G') at 300 to 1800 seconds after mixing of GC soft liner with different compositions in overall powder-to-liquid (P/L) ratios and concentrations of L. cubeba oil.

Group	Elastic modulus, G' (×10^4 Pa)*					
	at 300 s	at 600 s	at 900 s	at 1200 s	at 1500 s	at 1800 s
P/L0.9_control	1.79 ± 1.09^{A}	4.53 ± 2.20^{D}	5.60 ± 2.63^{G}	6.09 ± 2.83^{J}	6.32 ± 2.94^{M}	6.49 ± 3.02^{P}
P/L0.9_LC-10	2.90 ± 1.93^{A}	6.84 ± 3.48^{D}	8.31 ± 3.99^{G}	8.98 ± 4.25^{J}	9.28 ± 4.27^{M}	9.58 ± 4.54^{P}
P/L0.9_LC-20	2.33 ± 0.97^{A}	5.05 ± 2.07^{D}	6.06 ± 2.42^{G}	6.59 ± 2.63^{J}	6.82 ± 2.73^{M}	7.00 ± 2.82^{P}
P/L1.2_control	7.04 ± 4.15^{B}	12.32 ± 5.94^{E}	13.98 ± 5.98^{H}	14.87 ± 6.30^{K}	15.58 ± 6.69^{N}	15.42 ± 6.02^{Q}
P/L1.2_LC-10	10.24 ± 6.35^{B}	16.37 ± 9.70^{E}	18.62 ± 11.02^{H}	19.39 ± 11.38^{K}	19.81 ± 11.55^{N}	20.05 ± 11.60^{Q}
P/L1.2_LC-20	7.31 ± 2.06^{B}	11.70 ± 3.33^{E}	13.24 ± 3.92^{H}	14.10 ± 4.26^{K}	14.39 ± 4.26^{N}	14.47 ± 4.24^{Q}
P/L1.5_control	19.31 ± 10.62^{C}	27.27 ± 15.11^{F}	30.61 ± 17.35^{I}	30.26 ± 15.59^{L}	31.92 ±17.48^{O}	31.30 ± 16.29^{R}
P/L1.5_LC-10	14.77 ± 5.21^{C}	20.87 ± 7.56^{F}	23.56 ± 8.29^{I}	25.47 ± 10.97^{L}	25.68 ± 9.24^{O}	25.23 ± 8.15R
P/L1.5_LC-20	15.30 ± 7.84^{C}	21.51 ± 10.94^{F}	23.75 ± 12.70^{I}	24.99 ± 13.52^{L}	24.66 ± 12.74^{O}	26.20 ± 14.27^{R}

* Mean pairs with different superscripts represented statistical differences in column comparison (p<0.05).

Table 6 Shear viscous modulus (G”) at 300 to 1800 seconds after mixing of GC soft liner with different compositions in overall powder-to-liquid (P/L) ratios and concentrations of L. cubeba oil.

Group	Viscous modulus, G” (×10^4 Pa)*					
	at 300 s	at 600 s	at 900 s	at 1200 s	at 1500 s	at 1800 s
P/L0.9_control	1.02 ± 0.50^{A}	1.53 ± 0.62^{D}	1.64 ± 0.67^{G}	1.68 ± 0.69^{I}	1.67 ± 0.67^{L}	1.68 ± 0.69^{N}
P/L0.9_LC-10	1.57 ± 0.85^{A}	2.28 ± 1.07^{D}	2.48 ± 1.08^{G}	2.52 ± 1.11^{I}	2.52 ± 1.18^{L}	2.58 ± 1.16^{N}
P/L0.9_LC-20	1.24 ± 0.51^{A}	1.72 ± 0.66^{D}	1.84 ± 0.75^{G}	1.86 ± 0.70^{I}	1.92 ± 0.76^{L}	1.94 ± 0.79^{N}
P/L1.2_control	2.61 ± 1.22^{B}	3.12 ± 1.26^{E}	3.16 ± 1.17^{G}	3.09 ± 1.04^{J}	3.14 ± 0.99^{L}	3.16 ± 1.49^{N}
P/L1.2_LC-10	3.45 ± 1.96^{B}	4.13 ± 2.26^{E}	4.15 ± 2.44^{G}	4.20 ± 2.63^{J}	3.94 ± 2.22^{L}	4.07 ± 2.31^{N}
P/L1.2_LC-20	2.57 ± 0.69^{B}	3.02 ± 0.96^{E}	2.98 ± 0.80^{G}	2.89 ± 0.90^{J}	3.11 ± 0.89^{L}	3.09 ± 0.93^{N}
P/L1.5_control	5.29 ± 2.69^{C}	5.03 ± 1.89^{F}	6.20 ± 3.16^{H}	5.56 ± 2.72^{K}	5.59 ± 2.85^{M}	6.39 ± 3.58^{O}
P/L1.5_LC-10	4.18 ± 1.42^{C}	4.79 ± 2.40^{F}	5.50 ± 3.20^{H}	4.31 ± 1.50^{K}	5.12 ± 2.40^{M}	5.03 ± 2.28^{O}
P/L1.5_LC-20	4.28 ± 2.10^{C}	4.60 ± 2.21^{F}	5.09 ± 2.81^{H}	4.64 ± 2.74^{K}	5.45 ± 4.21^{M}	5.55 ± 3.32^{O}

* Mean pairs with different superscripts represented statistical differences in column comparison ($p<0.05$).

Discussion

In the present study, *L. cubeba* essential oil that has been reported the anti-fungal properties was introduced as an additive into the short-term soft liner.[18-20] Since *L. cubeba* oil is one of the essential oils, it might affect the initial viscoelastic properties of the soft liner in the same manner as the previous study.[16] In such research, the diminished effect of three essential oils on the initial moduli of GC soft liner was mainly contributed by the overall P/L ratio.

From the preliminary experiment, 1.25g of *L. cubeba* oil was incorporated into GC soft liner with 5g of the liquid and 6g of the powder. This mixture contained 20% w/w of *L. cubeba* oil into total liquid content. The Shore A0 hardness values of such composition were about 12 and 19 at 2 hours and a week, respectively. Whereas the same soft liner without additive showed the surface hardness about 20 and 26 at 2 hours and a week, respectively. It would be hypothesized that the addition of the essential oil increased total liquid component and, therefore, lowered the overall P/L ratio of the soft liner. The decrease in P/L ratio of soft liners could lead to a lesser modulus of the materials.[25]

This preliminary finding created an interesting idea in an alteration of the soft liner composition. To compensate the increase in the total liquid after adding the essential oil, more amount of the powder should be added. For instance, 7.5g of GC soft liner powder was used if 1.25g of *L.cubeba* oil was incorporated into the composition to maintain the overall P/L ratio of 1.2 as illustrated in Figure 2.

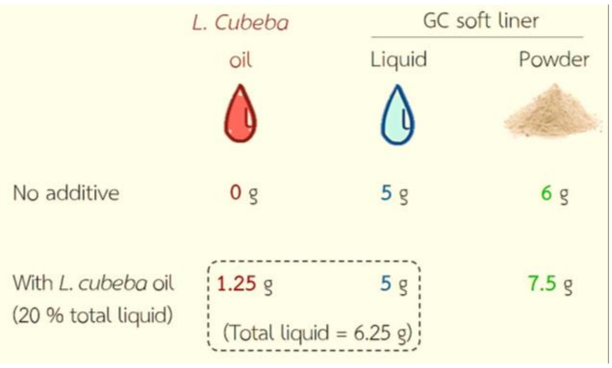

Figure 2 Demonstration for a proposed formulation of GC soft liner containing 20% w/w *L. cubeba* oil with overall powder-to-liquid ratio at 1.2.

The results from the surface hardness testing (Table 2 and 3) strongly supported this proposed idea. The Shore A0 hardness of the soft liners was mainly affected by the overall P/L ratio. Therefore, compensation of P/L ratio by adding more powder into the soft liner mixture could minimize the softening effect from *L. cubeba* oil. In comparison to GC soft liner control with the same overall P/L ratios, the Shore A0 hardness of the soft liner incorporated with 10% w/w *L. cubeba* oil did not significantly lower than those of the control.

However, the addition of 20% w/w *L. cubeba* oil significantly decreased the material hardness at the 2 hours for all P/L ratios. The softening effect at a high concentration of *L. cubeba* oil could be due to the plasticization of the polymer network in the soft liner by the additive molecules. One relevant parameter that influences the softening process or so-called ‘plasticization’ is a solubility parameter (δ). If the solubility parameters of additives are close to that of polymers, the softening of such polymers could be occurred.[26] Herein, the solubility parameters of main compounds in GC soft liner and *L. cubeba* oil were shown in Table 7.[27-29] Polymer in the soft liner powder was PEMA and major composition in the liquid component was BPBG (~81%). The solubility parameters of those compounds were around 19 and 21 $MPa^{1/2}$ that were close together.[27,28] Therefore, BPBG could act as a plasticizer to make the polymer softer. While, *L. cubeba* oil contained citral up to 60%. Its solubility parameter was 19 $MPa^{1/2}$, approximately.[29] In the same manner as BPBG, *L. cubeba* oil could soften PEMA and then decrease the hardness of the soft liner. In addition, because the solubility parameters of main compounds in both GC soft liner liquid (BPBG) and *L. cubeba* oil (citral) were close together, the essential oil could be well dispersed in the soft liner liquid.[27]

Table 7 Solubility parameters (δ) of main chemical constituents in GC soft liner and *L. cubeba* oil.[27-29]

Materials	Main compositions	δ ($MPa^{1/2}$)
GC soft liner powder	Poly(ethyl methacrylate) (PEMA)	19.30
GC soft liner liquid	Butylpthalylbutyl glycolate (BPBG) Ethanol	20.65 26.00
L. cubeba oil	Citral	19.10

The incorporation of *L. cubeba* oil did not only influence the material hardness but also the initial viscoelastic properties. In this study, time-dependent shear elastic and shear viscous moduli during the initial setting of the soft liners were investigated by the oscillating plate rheometer (Figure 1). The alteration in both moduli by time can reflect the gel-forming process of short-term soft liners.[24,30] In the beginning, immediately after mixing, the materials are still in a fluid-like state and be dominated by the viscous modulus (G”).[24,29,30] Afterwards, polymeric molecules of the soft lining materials would form physical or pseudo crosslinked gel that leads to a dramatic increase in the elastic modulus (G’).[24,29] At the time in which the elastic modulus exceeds the viscous modulus, it indicates the fluid-gel transition and can be defined as the gelation time.[16,24,31] The gelation time could be related to the setting time and working time of the soft liners.[25,32]

In the present study, the gelation times of the soft liners with different compositions were mainly contributed by the overall P/L ratio (Table 4). Whereas, the influence of the oil concentration on the material gelation times was not remarkable. As mention previously, the gel formation of the short-term soft liner can occur *via* pseudo crosslink consisting of polymer chain entanglements.[29] For lower P/L ratios, the polymer chains in the soft liner system would be surrounded by abundant liquid molecules. These could retard the interaction or intermolecular force between the polymer chain entanglements. Consequently, the gelation time would be prolonged to form sufficient pseudo crosslinks of the soft liner polymer.[29,33]

Besides the gelation time, the initial elastic and viscous moduli could provide essential information relating to clinical procedures including material rigidity and viscosity.[24,32] In this study, the elastic and viscous moduli were interpreted at every five minutes until thirty minutes after mixing. The intraoral working time of GC soft liner according to the manufacturer’s instruction is about 4-5 minutes. So, the moduli at five minutes could relate to the material properties after the initial setting during a chairside manipulation. While the moduli at 20-30 minutes could represent the material after a clinical visit.

From the results in Table 5-6, and Figure 3, all compositions of the soft liners showed a regular pattern in an alteration of the elastic and viscous moduli. The shear elastic moduli (G') of all samples considerably raised until 15 minutes approximately and then a slight increase or plateau phase was observed. While their shear viscous moduli (G") from 5 to 30 minutes were almost steady. The continuous increase of the elastic moduli after the fluid-gel transition point would remind the crosslinking process of the soft liners is still incomplete. Thus, the materials require at least 15 minutes to complete the gel formation process.

Additionally, the elastic modulus could reflect the material hardness or rigidity.[24,32] In this study, the initial elastic moduli of the soft liners were significantly affected by the overall P/L ratio in the almost similar pattern as the surface hardness experiment. The higher overall P/L ratio caused the higher initial elastic modulus (Table 5). Theoretically, an increase in polymer powder content would lead to more polymer chain entanglement. This resulted in more physical crosslinks and thereby greater elastic modulus.[33]

Although there was no statistical difference in the effect of the oil concentration, some distinguish trends in the initial elastic moduli were observed as shown in Figure 3. At excess powder content (P/L ratio=1.5), the incorporation of *L. cubeba* oil diminished the initial elastic modulus of the soft liner. For the compositions with lower P/L ratios (0.9 and 1.2), the soft liners with *L. cubeba* oil exhibited higher elastic moduli than the compositions without the essential oil. Interestingly, the incorporation of 10 %w/w *L. cubeba* oil into the soft liners with such P/L ratios showed the elastic moduli higher than the compositions with 20% w/w *L. cubeba* oil.

In general, most soft lining materials require optimal formulations of solvent mixtures and suitable amount of powder.[29] An alteration of each component concentration and/or using improper ratio could diminished material properties. One published work found that formulated short-term soft liners with the same P/L ratio at 1.2 appeared atypical trend in the effect of ethanol content on shear elastic moduli.[24] Such materials with 5 and 10% w/w ethanol obtained higher elastic moduli than the short-term soft liner with 20% w/w ethanol. Nevertheless, this hypothesis should be further investigated.

From the result in this study, it could be helpful to establish an optimal composition of GC soft liner with *L. cubeba* oil that exhibits an anti-microbial effect without alteration on the mechanical and physical properties.

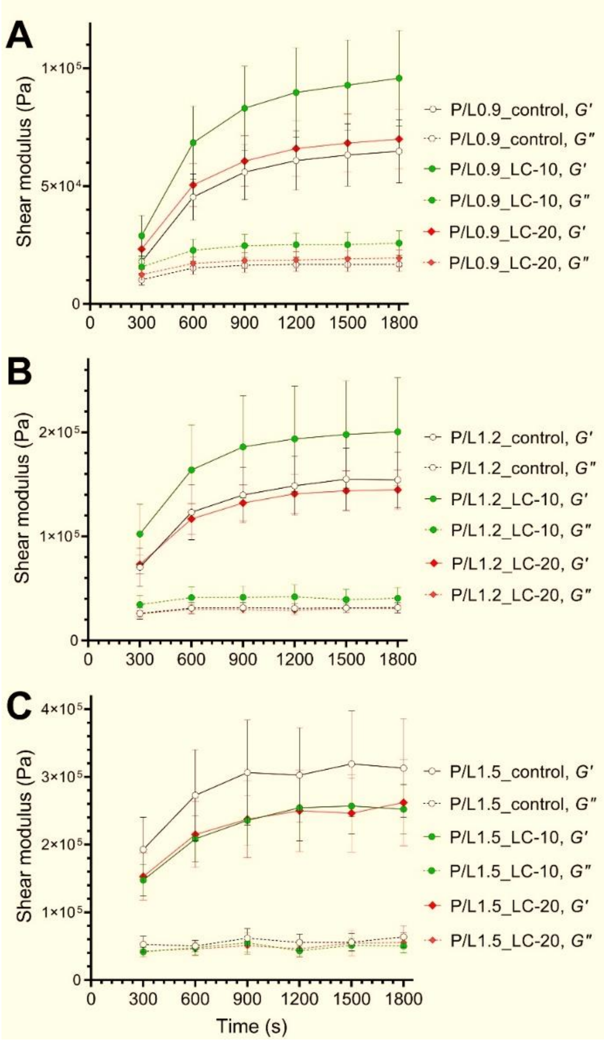

Figure 3 Shear elastic moduli (G'; plotted as straight lines) and viscous moduli (G", plotted as dash lines) of GC soft liner with different compositions. The powder-to-liquid (P/L) ratios of 0.9, 1.2, and 1.5 were illustrated in Figure 3A to 3C, respectively. The concentrations of L. cubeba oil were 0% w/w (hollow circle), 10% w/w (green circle), and 20% w/w (red diamond).

Conclusion

The deteriorating effects in the material hardness and initial viscoelastic properties of the soft liner after incorporating with *L. cubeba* oil could be minimized or compensated by an adjustment of P/L ratios. The compositions with the same overall P/L ratios exhibited almost similar surface hardness and initial viscoelastic properties. At higher P/L ratios, the materials significantly showed the greater hardness, faster gelation time, and higher initial shear moduli. There was less effect of *L. cubeba* concentration on the surface hardness of the soft liner. While the gelation time and the initial moduli were not statistically influenced by the oil concentration.

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ผลของอัตราส่วนผงต่อน้ำโดยรวมต่อค่าความแข็งผิวและสมบัติวิสโคอิลาสติกช่วงแรกของวัสดุเสริมฐานฟันเทียมชั่วคราวชนิดอ่อนที่ผสมน้ำมันตะไคร้ภูเขา

นันท์มนัส ย่างสุก มนตรี รัตนจันทร์ฉาย** วิชญ กาญจนะวสิต***

บทคัดย่อ

การศึกษานี้มีวัตถุประสงค์เพื่อศึกษาความแข็งผิวและสมบัติวิสโคอิลาสติกช่วงแรกของวัสดุเสริมฐานฟันเทียมชนิดอ่อนที่ผสมด้วยน้ำมันตะไคร้ภูเขา โดยประกอบด้วย 9 กลุ่มการทดลอง ที่ส่วนผสมมีความแตกต่างในอัตราส่วนของผงต่อน้ำ (0.9, 1.2, และ 1.5) และความเข้มข้นของน้ำมันตะไคร้ภูเขา (ร้อยละ 0, 10, และ 20 โดยน้ำหนัก) ทั้งนี้วัสดุแต่ละกลุ่มถูกเตรียมเพื่อนำมาวัดความแข็งผิวที่ 2 ชั่วโมง และ 7 วันด้วยเครื่องวัดความแข็งแบบชอร์และมีการทดสอบสมบัติวิสโคอิลาสติกช่วงแรกของวัสดุ ด้วยเครื่องวิเคราะห์การไหลแบบจานแกว่งตั้งแต่เริ่มผสมวัสดุจนครบ 30 นาที ซึ่งประกอบไปด้วยการหาระยะเวลาการก่อวุ้น ค่ามอดูลัสสะสมและมอดูลัสสูญเสีย โดยแต่ละการทดลองมีการทำซ้ำ 5 ครั้ง จากผลการศึกษาพบว่าอัตราส่วนของผงต่อน้ำของวัสดุเสริมฐานฟันเทียมชนิดอ่อนเป็นปัจจัยหลักต่อค่าความแข็งผิวของวัสดุ โดยกลุ่มที่ใช้อัตราส่วนของผงต่อน้ำมากจะมีค่าความแข็งผิวที่เวลา 2 ชั่วโมง และ 7 วันสูงกว่าอย่างมีนัยสำคัญ ($p<0.05$) ทั้งนี้การใส่น้ำมันตะไคร้ภูเขาร้อยละ10 ลงในวัสดุเสริมฐานฟันเทียมชนิดอ่อนไม่ส่งผลต่อความแข็งผิวของวัสดุ อย่างไรก็ตามการผสมน้ำมันตะไคร้ภูเขาที่ความเข้มข้นร้อยละ 20 ทำให้ความแข็งผิวที่ 2 ชั่วโมงของวัสดุ ต่ำกว่ากลุ่มอื่นอย่างมีนัยสำคัญ ($p<0.05$) นอกจากนั้นพบว่าอัตราส่วนของผงต่อน้ำยังเป็นปัจจัยหลักที่ส่งผลต่อสมบัติวิสโคอิลาสติกช่วงแรกของวัสดุเสริมฐานฟันเทียมชนิดอ่อน โดยกลุ่มที่ใช้อัตราส่วนของผงต่อน้ำมากกว่าจะมีระยะเวลาการก่อวุ้นสั้นลง และค่ามอดูลัสสะสมและสูญเสียสูงขึ้นอย่างมีนัยสำคัญ ($p<0.05$) ทั้งนี้ไม่พบว่าความเข้มข้นของน้ำมันตะไคร้ภูเขาส่งผลต่อสมบัติวิสโคอิลาสติกช่วงแรกของวัสดุ โดยสรุปจากการศึกษานี้ พบว่าความแข็งผิวและสมบัติวิสโคอิลาสติกช่วงแรกของวัสดุเสริมฐานฟันเทียมชนิดอ่อนที่ผสมน้ำมันตะไคร้ภู เขาขึ้นกับปัจจัยอัตราส่วนผงต่อน้ำเป็นหลัก โดยที่ความเข้มข้นของน้ำมันตะไคร้ภูเขาไม่ได้มีผลเด่นชัดต่อสมบัติดังกล่าวของวัสดุ

คำไขรหัส: *วัสดุเสริมฐานฟันเทียมชนิดอ่อน/ ความแข็งผิว/ วิสโคอิลาสติก/ น้ำมันตะไคร้ภูเขา/ น้ำมันหอมระเหย*

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