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Dynamic viscoelastic properties of vinyl polysiloxane denture soft lining materials

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SUMMARY The aim of this study was to investigate the dynamic viscoelastic properties of seven commercially available vinyl polysiloxane denture soft lining materials. Five rectangular specimens (2 × 10 × 30 mm) were prepared from each material. The complex modulus E^* (MPa) and loss tangent (tan δ) of each specimen were determined with a non-resonance forced vibration method using an automatic dynamic viscoelastometer at 1 Hz after 1 day of dry storage, and after 1, 30, 60, 90 and 180 days of wet storage at 37 °C. All data were analysed using one-way ANOVA and Bonferroni/Dunn's test for multiple comparisons with a significance level of $P < 0.01$. All materials varied widely in terms of viscoelasticities and showed both an increase in E^* and a decrease in tan δ at 1 Hz after the 1-day wet storage. After 60 days of wet storage, both E^* and tan δ did not change significantly. The stiffer materials (>30% filler content) with high E^* values (>2·00 MPa) showed elastic behaviour with tan δ values of around 0·03. The softer materials (6% filler content) with high tan δ values (initial value > 0·10) showed viscous behaviour and were easily affected by water absorption after the 1-day wet storage. It can be concluded that for the proper selection of vinyl polysiloxane denture soft lining materials, it is very important to evaluate the viscoelastic properties after 60 days of wet storage.

KEYWORDS: dynamic viscoelastic properties, vinyl polysiloxane, auto- and addition-curing silicone, soft lining materials, soft liners, complex modulus, loss tangent

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Introduction

Denture soft lining materials are used for patients who have high ridge resorption, a thin mucosa coating or are unable to tolerate hard-based acrylic dentures because of pain while chewing (1). The soft materials mostly consist of acrylic, silicone and polyolefin. Silicone materials are classified into auto-curing and heat-curing types. The auto-curing type has been widely used in clinical cases because it can be used both for direct and indirect methods. Recently, vinyl polysiloxane materials were proposed as replacements for a heat-cured silicone (Molloplast B*) because of their similar viscoelasticities and better handling properties (1). However, because the silicone materials degrade relatively faster than the hard materials and it is difficult to predict their durability, regular post-treatment maintenance is required.

Making silicone soft lining materials with the required properties is more challenging than making hard lining materials with similar properties. This means that no product to date has resolved all the following problems associated with soft lining materials: long-term retainment of viscoelasticity, bond strength to base material, presence of bacteria and food residues and suitability for cutting and polishing (2–7). The main reason for the development of silicone soft lining materials is to have a sufficient cushioning effect for distribution and absorption of functional stress. Recently, three different types of silicone materials (stiff, medium and soft according to ISO 10139-2) in terms of viscoelasticities have become available. We

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Table 1. Vinyl polysiloxane denture soft lining materials

Brand name	Code	Manufacturer	Batch no.	Filler fraction (wt%)
Mucopren soft	MPS	Kettenbach, Eschenburg, Germany	70171	30*
Sofreliner tough medium	STM	Tokuyama Dental Corp., Tokyo, Japan	48	20*
Sofreliner medium soft	SMS	Tokuyama Dental Corp., Tokyo, Japan	566	6*
Sofreliner super soft	SSS	Tokuyama Dental Corp., Tokyo, Japan	261	6*
GC reline soft	GS	GC Corp., Tokyo, Japan	801292	37†
GC reline extra soft	GES	GC Corp., Tokyo, Japan	803121	30†
GC reline ultra soft	GUS	GC Corp., Tokyo, Japan	803131	18†

Basic composition of all materials: vinyl polysiloxane, silicon dioxide, platinum catalyst and others.

*Data taken from the manufacturer's information.

†Data taken from McCabe *et al.* (2002)[2].

should select the materials possessing the best characteristic in terms of long-term retainment of viscoelasticity and high bond strength to base material, but almost all manufacturers have developed products that address only viscoelasticity and not the other requirements. The reasons for this include the following: (i) The clinical criteria to differentiate the types, such as thickness of patient's mucosa coating and viscoelasticity, has not been clearly determined; (ii) The softer the materials are, the faster they degrade, which induces the growth of bacteria and residues of rotting food and (iii) The softer the materials are, the thicker they should be; but even if they are thick enough, the softness can reduce denture stability, leading to pain from wrong occlusion. Therefore, under a situation where it is difficult to select the materials with appropriate viscoelasticities by taking each patient's mucosa condition into consideration, we would have no choice but to select the products which can be used for a long time with as low a degradation rate as possible while emphasizing the elastic property.

The purpose of this study was, therefore, to investigate the dynamic viscoelastic properties of seven commercially available vinyl polysiloxane denture soft lining materials, and to provide information that might enable the proper selection of auto- and addition-curing silicone.

Materials and methods

Vinyl polysiloxane denture soft lining materials

Seven commercially available vinyl polysiloxane denture soft lining materials: Mucopren soft (MPS), Sofreliner tough medium (STM), Sofreliner medium soft (SMS), Sofreliner super soft (SSS), GC reline soft (GS), GC reline extra soft (GES) and GC reline ultra soft (GUS) were selected for investigation (Table 1).

Specimen preparation

All materials were supplied in the form of a two-paste cartridge and auto- and addition-curing silicones. The cartridge was set into a dispensing gun, and these pastes were automatically mixed during extrusion. Five rectangular specimens (thickness, 2 mm; width, 10 mm and length, 30 mm) of each material were prepared according to the manufacturer's instructions. The specimens were dried at 37 °C for 1 day and were successively stored in distilled water at 37 °C for 1, 30, 60, 90 and 180 days except during the measuring period.

Measurements of dynamic viscoelastic properties

The dynamic viscoelastic properties of five specimens of each material were measured using an automatic dynamic viscoelastometer (RHEOVIBRON DDV-25FP†). Specimens were set in a tensile jig and tested at a frequency of 1 Hz and a temperature of 37 °C. The ratio of amplitude and phase difference between applied sinusoidal strain and resulting stress through the specimen was measured by a non-resonance forced vibration method.

The complex modulus E^* (MPa) was then calculated by the expression

$$E^* = \{\text{storage modulus } (E')^2 + \text{loss modulus } (E'')^2\}^{1/2}$$

Loss tangent (tan δ) was calculated by the equation

$$\tan \delta = E''/E'$$

†Orientec Inc., Tokyo, Japan.

where E' is the elastic stiffness of the material and E'' describes its viscous behaviour. High E^* generally indicates the characteristic of being hard to deform. The scale of energy loss is indicated by tan δ; materials that absorb energy as a result of deformation show a high tan δ value.

Statistical analysis

One-way ANOVA and Bonferroni/Dunn's test for multiple comparisons were used to statistically analyse the mean values of E^* (MPa) and tan δ for each storage period at 0·01 probability level.

Results

The E^* and tan δ values at 1 Hz for the one-day dry and wet conditions for each material are shown in Fig. 1. All materials showed both an increase in E^* and a decrease in tan δ after the 1-day wet storage, and in particular, SMS and SSS exhibited significant changes of those parameters ($P < 0·01$).

Variations of E^* at 1 Hz under wet conditions as function of time for 180 days are shown in Fig. 2. All materials except SMS showed significant increases in E^* between one and 30 days of wet storage ($P < 0·01$). After 60 days of wet storage, the E^* values did not show significant differences ($P > 0·05$). The decreases in tan δ were coincident with the increases in E^*.

Figure 3 shows the relationship between E^* and tan δ for all materials at 1 Hz after the 90-day wet storage. No significant difference was found between the E^* values of GUS and SMS ($P > 0·05$). The 95% confidence interval of the E^* value for GUS, SMS and SSS was 1·27–1·59 MPa, and the viscous behaviour of these materials widely changed within the range of the E^* value. As for tan δ, there were no significant differences among GS, GBS and MPS, and no significant difference between GS and STM ($P > 0·05$). The 95% confidence interval of tan δ values for GS, GBS, MPS and STM was 0·028–0·033, and these materials had a tendency to show elastic behaviour at the tan δ value of around 0·03.

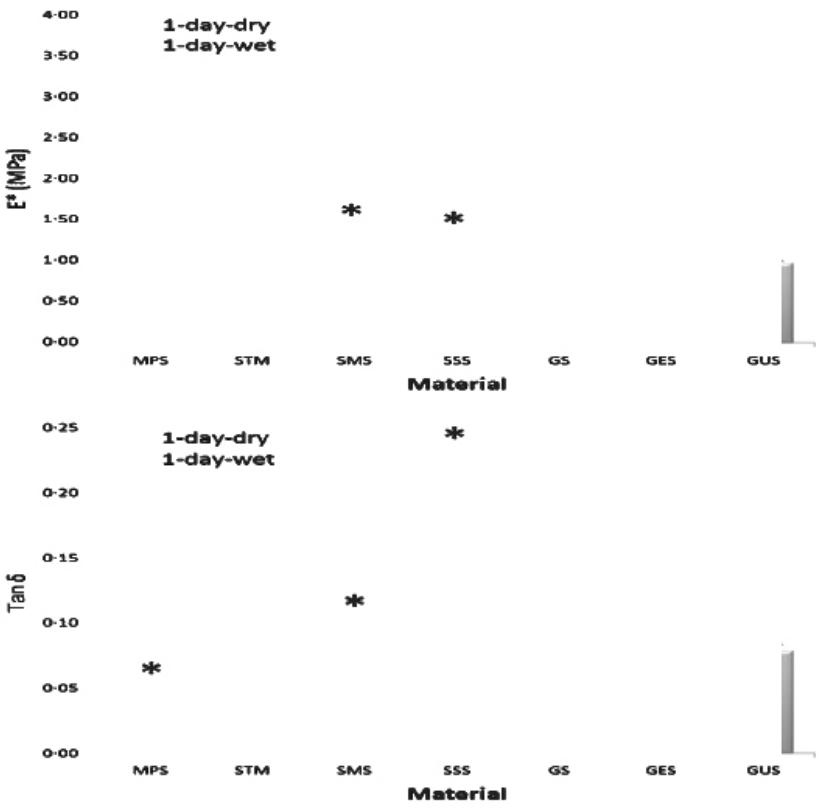

Fig. 1. Mean complex modulus E^* (MPa) and loss tangent (tan δ) values at 1 Hz for the 1-day dry and wet conditions for seven vinyl polysiloxane denture soft lining materials. * indicates significant differences between dry and wet conditions ($P < 0·01$). Abbreviations are as in Table 1.

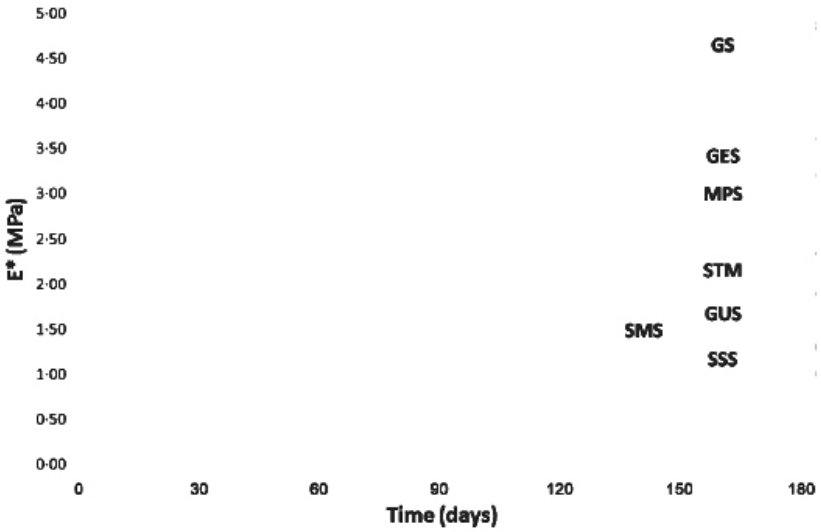

Fig. 2. Complex modulus E^* (MPa) variations at 1 Hz under wet condition as function of time for 180 days. Abbreviations are as in Table 1.

Discussion

The non-resonance forced vibration method used in this study, which is commonly used in the study of polymeric material development, subjects the material to various frequencies and temperatures and evaluates the effects of these conditions (7). This testing characterizes the periodic deformation of the tested materials which occurs in clinical use. Therefore, the dynamic viscoelastic testing in this study was conducted at a frequency of 1 Hz and a temperature of 37 °C because these conditions closely approximate the oral environment.

The complex modulus E^* is resolved into two components, i.e. storage modulus E' and loss modulus E''. E' represents the elastic component of material behaviour, and it is directly proportional to the energy storage in a cycle of deformation. E'' represents the viscous component of material behaviour, and it is directly proportional to the average dissipation or loss of energy as heat in a cycle of deformation. Loss tangent (tan δ) is a useful parameter and a measure of the ratio of energy lost to energy stored during cyclic deformation. Because the material with the higher E^* value indicates the lower tan δ value and has the characteristic of being hard to deform, such material cannot have

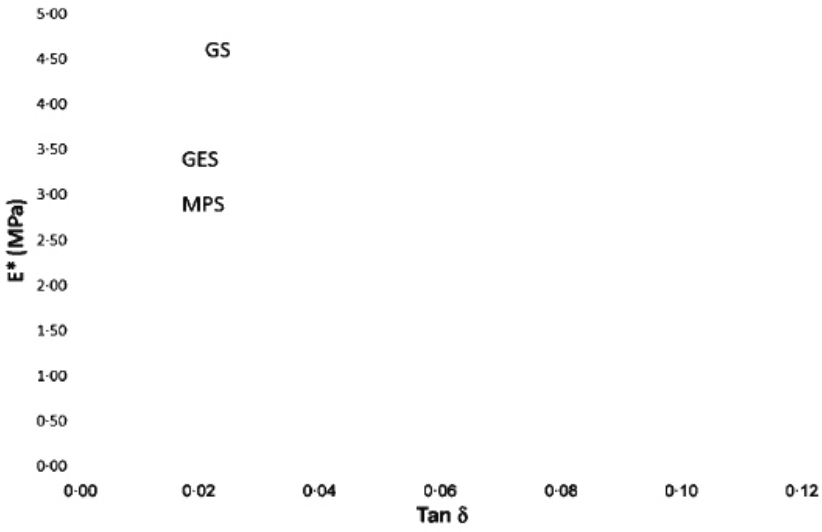

Fig. 3. Relationship between complex modulus E^* (MPa) and loss tangent (tan δ) at 1 Hz after the 90-day wet storage for the seven materials investigated. The 95% confidence interval of the E^* value for GUS, SMS and SSS was 1·27-1·59 MPa. The 95% confidence interval of tan δ values for GS, GES, MPS and STM was 0·028-0·033. Abbreviations are as in Table 1.

viscous behaviour in order to distribute and absorb functional stress (Fig. 3). Conversely, the material with the higher tan δ value can absorb energy as a result of deformation. Murata *et al.* (8) illustrated that the ideal materials have relatively high values of E^* and tan δ though it would seem to be impossible to develop the materials with both the high E^* and the high tan δ values. In general, the silicone materials are more elastic than the acrylic materials. Therefore, in this study, in order to properly evaluate the silicone materials in view of their viscoelastic properties, the two parameters E^* and tan δ were selected.

The long-term soft lining materials tested in this study consist of silicone such as vinyl polysiloxane. These materials based on vinyl polysiloxane have siloxane linkage, $-Si(CH_3)_2-O-Si(CH_3)_2-$, and small amount of functional vinyl groups in their molecular structure. Murata *et al.* (7) reported that the viscoelastic properties of silicone materials remained unchanged after 3 years of storage while those of the acrylic materials underwent significant change. Hence, Murata's results using the same method as used in this study confirm that the silicone materials rather than the acrylic materials should be selected for durability.

E^* values at 1 Hz under wet condition were measured as a function of time for 180 days to evaluate the durability of the tested materials (Fig. 2). For the first 30 days, the E^* values of all materials except SMS changed significantly. Although the E^* values of GES in particular showed a large variation in the first 30 days, the viscoelastic behaviour of all tested materials remained stable after 60 days of wet storage. Hence, the selection of materials, with reference to their viscoelastic properties, should be based on these properties attained after the 60-day period.

The dynamic viscoelastic properties and parameters such as E^* and tan δ varied widely amongst the tested materials. The viscoelastic properties of the tested materials appear to be influenced by different compositional parameters such as filler fraction. The filler content of MPS was about 30% (Table 1) according to information supplied by the manufacturer. The filler contents of STM, SMS and SSS, according to information from the manufacturer, were 20%, 6% and 6%, respectively; and SMS and SSS contained 28% silicone resin powder in order to improve their bonding strength to base material. McCabe *et al.* (2) have reported that the filler contents of GS, GES and GUS were 37%, 30% and 18%, respectively. According to the classification outlined in ISO 1039-2, MPS, STM and GS are classified as stiff, SMS and GES as medium and SSS and GUS as soft. However, on the basis of results of this study, GES could be classified as being just as stiff as GS and MPS because the E^* value of GES was found to be between those of GS and MPS after the 30-day wet storage, and these materials contained greater than 30% filler.

GS, GES and GUS, produced by the same manufacturer, had different values of E^*, and STM, SMS and SSS had different values of tan δ. Each manufacturer had a different concept for the viscoelastic behaviour of soft lining materials and developed these products with different viscoelasticities. Therefore, from the viewpoint of the relationship between E^* and tan δ, the viscoelastic properties of all tested materials were analysed. For GS, GES, MPS and STM, the great diversity in the E^* values was because of different filler fractions, while the tan δ value remained around 0·03. On the other hand, for GUS, SMS and SSS, wide changes in the tan δ values were found below the E^* value of 2·00 MPa. Therefore, the materials based on vinyl polysiloxane have a tendency to converge to the tan δ value of approximately 0·03 as an index of the viscous property, for E^* values higher than 2·00 MPa. Moreover, the observed small changes in E^* below 2·00 MPa and the great diversity in the viscous behaviour were because of the intrinsic viscoelasticity of vinyl polysiloxane. As for filler fractions, STM (20% filler) and GUS (18% filler) with the E^* value of around 2·00 MPa could be classified as medium stiff in this study, and these materials have very little influence on clinical handling.

The auto- and addition-curing silicone materials remained the most stable over time compared to the acrylic materials. This can be explained by the assumption of the low water absorption and solubility of these materials (7). However, the tested materials showed both an increase in E^* and a decrease in tan δ at 1 Hz after the 1-day wet storage, and the changes in both parameters can be attributed to water sorption effects (Fig. 1). For SMS and SSS, because the significant large differences between the E^* values of the 1-day dry and wet were 47% and 189%, respectively, the differences between their tan δ values also were 71% and 114%, respectively. Because SMS and SSS contained only as little as 6% filler and 28% silicone resin powder, much lower than the other materials, they can be expected to be easily affected by water absorption. In that case, it

would be difficult to control their viscoelastic properties and reline the materials onto the dentures under clinical situations. Hence, SMS and SSS could be classified as soft in this study. However, even if these materials would demonstrate viscous behaviour when compared to the other tested materials, tan δ values (<0·10) of these materials were smaller than those (>1·0) of the acrylic material reported by Murata *et al.* (7). The previous study for relationship between dynamic viscoelasticity and masticatory function (8) indicated that the difference in the patient's subjective assessments of satisfaction between the silicone and the acrylic materials was very small.

Although the different problem of bond strength to base material was not evaluated in this study, it can be noted that the viscoelastic properties can marginally affect the bond strength. McCabe *et al.* (1) have reported that stiffer materials produced the greatest tensile bond strength, but the lowest peel bond strength. Mutluay and Ruyter (3) have also demonstrated that the tensile bond strength to base material of the soft lining materials was similar to their tensile strength results. However, under the same bonding system, the peel bond strength to base material of GUS, GES and GS after the 30-day wet storage was 1·44, 2·16 and 2·47 N/mm, respectively, and there was no significant difference between these values for GES and GS (4). Hence, the viscoelastic behaviour could be related to the bond strength, however, the different tests exhibited the different tendencies in their results. In particular, SMS and SSS tested in this study were softer materials and contained silicone resin powder to compensate for their lower bond strengths. On the other hand, it has been reported that the bonding to base material is obtained by softening of the surface layer with a solvent and partially impregnating the layer with a polymer solution (3). Therefore, in the development of improved adhesives for the bonding of vinyl polysiloxane and base materials, the nature of the primer is a major factor (1).

The elastic moduli of the oral mucosa have ranged from approximately 0·4 to 4·4 MPa (9). Although the elastic moduli reported there were not exactly equal to the E^* values in this study, the E^* values of the tested materials mostly existed within the range of the elastic moduli for the mucosa. The viscoelastic properties of the materials should be similar to those of the oral mucosa, but we should realize that it would be difficult to assess the viscoelasticities of the mucosa by numerical values and we should rely on our subjective in-clinic estimation. Therefore, for clinical application, stiffer silicone materials should be selected first, because of the ease of handling, but also for their bonding strength to base material.

Conclusions

Within the limitations of this study, the following conclusions were reached:

1 vinyl polysiloxane denture soft lining materials varied widely in terms of dynamic viscoelastic properties represented by the parameters such as complex modulus E^* (MPa) and loss tangent (tan δ). The values for materials ranged between those for GS (E^*, 4·59 MPa; tan δ, 0·03) and SSS (E^*, 1·14 MPa; tan δ, 0·10);

2 the stiffer materials (>30% filler content) with high E^* values (>2·00 MPa) such as GS, GES and MPS showed the elastic behaviour with tan δ values of around 0·03;

3 materials with about 20% filler content and with the E^* values of around 2·00 MPa such as STM and GUS should to be classified as medium stiff;

4 the softer materials (6% filler content) with high tan δ values (initial value > 0·10) such as SMS and SSS showed the viscous behaviour and were easily affected by water absorption, especially on the first day of application;

5 regarding the long-term measurements reported in this study, it is very important that for the proper selection of materials, the viscoelastic properties should be evaluated in tests of longer than 60 days.

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