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INFLUENCE OF CHEMICAL COMPOSITION ON THE PROPERTIES OF EXPERIMENTAL SILICONE-BASED SOFT LINING COMPOSITES

The paper analyses the influence of chemical composition of silicone-based composites on their properties in the aspect of using them as long-term soft denture lining materials. Different concentrations of filler and methylhydrosiloxane-dimethylsiloxane copolymer were used. The filler was introduced into the composite with mechanical mixing combined with ultrasonic homogenization. Scanning electron microscopy was used to investigate the quality of filler dispersion. Shore A hardness, tensile strength, sorption, solubility and tensile bond strength to poly(methyl methacrylate) were measured. Tests show satisfactory results for some experimental composites, which met all the requirements for such materials.

Keywords: dental material, soft lining, silicone, silica filler, mechanical properties

1. Introduction

Studies related to dental materials and dental engineering are currently one of the major areas of engineering research for medicine. Virtually all people have a direct contact with dental materials. In Poland, almost every person has a permanent dental filling or a denture [1]. The area of interest of this specialty encompasses engineering research in the field of materials, numerical modeling and design [2-5]. The continuous research progress in the field of metallic, ceramic, polymeric and composite materials is observed [6-8], but polymers and polymeric matrix based composites seem to be particularly promising. Among these, soft denture linings play an important role. Soft denture lining materials are used the most often in the so-called troublesome cases, i.e. in patients with atrophied or sharp alveolar ridges, atrophic mucosa, low tolerance of oral mucosa to the pressure exerted by hard denture bases or in the cases of lesions or lacerations appearing on oral mucosa [9]. They are also used for lining in implantology during implant healing to keep dentures on implants or to create post-surgical obturators [10-12]. It has been unambiguously proven that using such materials helps to increase the satisfaction of patients and aids wound healing [13-15]. The main advantage of soft linings is more even distribution of loads transferred by a denture to the bearing area. This group of materials also plays an important role in compensation of impact of manufacturing inaccuracies of hard denture bases being in contact with soft tissues, what prevents their damage [9]. This result is achieved due to soft lining materials show a low modulus of elasticity and ability to dissipate energy.

Dental prosthetics market offers soft lining materials for short-term (up to four weeks) and long-term uses [16]. They are acrylate- and silicone-based materials, but acrylic materials exhibit lower stability of physical and chemical properties during their period of use. Currently a relatively wide range of silicone materials is available for long-term lining. The range of properties expected for this group of materials is defined in the ISO 10139-2:2016 standard [17]. However, all materials of this type show similar imperfections in clinical and laboratory conditions. The most important are low microbiological resistance and tendency of the lining material to debonding from the acrylic denture base [9]. While debonding of linings from dentures depends on the properties of the bonding agent, in the case of low microbiological resistance, which is very important from the point of view of medical applications, the problem lies in the material and it has not been satisfactorily solved so far [18]. Currently, research in this direction is aimed at creating composite materials with antimicrobial properties. These works are usually focused on the modification of commercially available denture materials [19,20]. Unfortunately, the composition of the basic material, which constitutes the manufacturer's "know how", is known only in general terms, what limits the possibility of interpretation of the obtained results. Thus the first step to obtaining more complex composites should be the development of a reference composite material with certain biofunctional features, which would be the starting point for

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more complex studies. Therefore, the aim of this study was to investigate the impact of crosslinker (trimethylsiloxy terminated methylhydrosiloxane-dimethylsiloxane copolymer) and silica-filler concentrations on the selected mechanical and performance properties of poly(dimethylsiloxane)-based materials in view of the application of the developed composites in long-term denture linings.

2. Materials and methods

2.1. Materials preparation

In the presented investigations the following substrates were used to prepare silicone based composites: vinyl terminated polydimethylsiloxane (PDMS), trimethylsiloxy terminated methylhydrosiloxane-dimethylsiloxane copolymer (HMS) as crosslinker, platinum -divinyltetramethyldisiloxane complex as catalyst (CAT) and hexamethyldisilazane-treated amorphous silicon dioxide as a filler (FI), all purchased from Gelest, USA.

Two components of self-curing addition silicone material, traditionally called "base" and "catalyst" paste, were prepared. The substrates concentrations in both of them were calculated in such a way that they had to be mixed together at a mass ratio of 1:1 during curing.

It was assumed that components of materials will be prepared in 30 g portions. The filler was introduced into silicone matrix at concentrations of 15 wt.% and 30 wt.%. Filler concentrations were chosen based on preliminary tests. The material with filler concentration of 10% presented very low tensile strength, materials with filler concentration of 35% (or higher) were very difficult to manipulate during the crosslinking process, due to the high initial viscosity of the components. First, mixtures without FI were prepared. For the "catalyst" component the mixture of 0.36 wt.% of CAT with PDMS was prepared. For the "base" components mixtures of PDMS and HMS polymers were prepared. Five concentrations of HMS in materials ("base" and "catalyst" together) were tested: 2, 3 ,4 ,5 and 6 wt.%. The masses of HMS and PDMS necessary to prepare "base" components were calculated according to the following equations:

$$m_{HMS} = c_{HMS} \times (m_B + m_C) \quad (1)$$

$$m_{PDMSB} = m_B - m_{HMS} - c_{FI} \times m_B \quad (2)$$

where m_{HMS} was the HMS mass [g], c_{HMS} was the HMS concentration in the material after mixing components [wt.%] m_B and m_C were the mass of "base" an "catalyst" components with filler, respectively (both 30 g), m_{PDMSB} was the mass of PDMS in the "base" component [g] and c_{FI} was the filler concentration (15 or 30 wt.%).

All mixtures were prepared in 50 mL Griffin form beaker by stirring with a magnetic stirrer at room temperature for 24 h.

Next the FI was compounded into the previously prepared mixtures successively, in portions of 0.5 ±0.1 g or lower, when necessary. After addition of each portion the material was mechanically ground (shear forces) with a spatula on the wall of the beaker until an optically homogeneous consistency was achieved. When the whole FI was compounded, the compositions were ultrasonically homogenized (Ultrasonic Homogenizer UP200St, Hielscher Ultrasonics GmbH, Germany) in five series, each 90 ±10 s, with ten-minute intervals to cool the composition in water at a temperature of 15 ±2°C. Finally, the compositions were placed in a modified vacuum stirrer (Twister evolution, Renfert, Germany) under the pressure of 80 mbar for 20 min to remove air bubbles generated during the compounding process.

All samples were cured by mixing the "base" and "catalyst" with a spatula in a mass ratio of 1:1. For most investigations (excluding bond strength test) after mixing materials were packed into stainless steel or dental stone molds and loaded on a hydraulic press with the pressure of 0.3 MPa until the end of setting time. The following parameters of the curing process were applied: working time with materials was approx. 1 minute, initial setting time was 3 minutes and finally samples were cured in distilled water at 50°C under the pressure of 0.2 MPa for 10 min.

2.2. Methods

Samples for the Scanning Electron Microscopy (SEM) investigations of composites' morphology were cured in a stainless steel mold measuring 40×10×6 mm. After crosslinking, a standard, freeze-fracturing process was used. Samples were individually immersed in liquid nitrogen. After approximately 3 minutes each sample was broken with tongs and removed from the container with liquid nitrogen. The obtained fractures were sputtered with gold. The morphologies of failures obtained during tensile bond strength test were observed after sputtering with gold. The bonding layer between PMMA resin and silicone composite was observed on the samples after tensile bond strength test, but cross-sections were obtained with the previously described freeze-fracturing process. Samples were sputtered with gold. All samples were observed using a Zeiss SUPRA 35 scanning electron microscope at accelerating voltages of 5 to 10 kV.

Hardness after 5 s of loading was investigated with a method presented in the ISO standard [17], but in the present study additional storage times were used. Samples measuring 6 mm in thickness and 40 mm in diameter were cured in a stainless steel mold. Three samples were manufactured for every material ($n = 15$). Their hardness was measured with a Shore A digital durometer (Bareiss HPE II-A, Bariess, Germany) after 24 h, 7 days, 14 days and 28 days of storing in distilled water at 37 ±1°C. The hardness of every sample after each storing time was measured at five points. After measurements samples were immediately reimmersed in distilled water until the next measurement.

For tensile strength tests dumbbell-shaped samples [21] were prepared with a manual cutting press (ZCP 020, Zwick, Germany). The specimens were cut off from 2 mm thick cured plates and only one sample for a specific storing time was made from one plate. Three storing times in distilled water at 37 ±1°C were used (24 h, 7 days, 28 days). Thirty samples

were manufactured for every material (n = 150). Tensile testing was performed at a cross-head speed of 10 mm/min [22] using a universal testing machine (Zwick Z020, Zwick GmbH & Com, Germany). Ultimate tensile strength was calculated according to the following equation:

$$TS = \frac{F}{A} \tag{3}$$

where: TS is ultimate tensile strength in MPa, F is force at rupture in N, A is the initial cross-sectional area of specimen in mm^2.

The extension was measured continuously using a video extensometer [21] and the maximal elongation at break was calculated according to the following equation:

$$E_b = \frac{L - L_0}{L_0} \times 100\% \tag{4}$$

where: E_b is ultimate elongation in percent, L is initial distance between the markers, L_0 is the distance between the markers at the point of specimen rupture.

Sorption and solubility were determined using a method described in the ISO standard [17]. Test samples measuring 50 mm in diameter and 0.5 mm in thickness were cured in stainless steel molds. Five samples of each material were made (n = 25). The samples were weighed on an AS 110/C/2 analytic scale (Radwag, Poland) with an accuracy of 0.1 mg and they were dried inside desiccators with freshly dried silica gel in a dryer at 37 ±1°C, until daily changes in mass were no higher than 0.2 mg. Stable mass values were recorded as m_1. At this point also the diameter and thickness of samples were measured with a digital caliper with an accuracy of 0.01 mm. The samples were stored in a chamber with distilled water at 37 ±1°C. After 7 days they were removed from water, visible moisture was removed using filter paper and the samples were carefully air-dried (approximately 10-20 s) and weighed. The obtained mass was recorded as m_2. Finally, the samples were placed again in desiccators and dried as described above and stable masses were recorded as m_3. Sorption and solubility of each sample were calculated using the following equations:

$$w_{sp} = \frac{m_2 - m_3}{V} \tag{5}$$

$$w_{sl} = \frac{m_1 - m_3}{V} \tag{6}$$

where w_{sp} is sorption, w_{sl} is solubility, m_1 is the initial mass of dried samples in µg, m_2 is the mass after aging in µg, and m_3 is the mass after the final drying in µg and V is the volume of the samples in mm^3.

The tensile bond strength (TSB) of the obtained materials to the denture base resin was investigated with a method presented in the ISO standard [17]. Square pieces of polymethyl methacrylate resin Vertex Rapid Simplified (Vertex-Dental B.V., The Netherlands) measuring 25 mm on the side and 3.2 ±0.2 mm in thickness were produced with a standard flasking technique used in prosthetic dentistry and polymerized in accordance with the manufacturer's instructions. The pieces were preliminarily wet-ground on abrasive paper, first with grit size P220 (Struers A/S, Denmark) to standardize the plates. The PMMA samples were rinsed, and their working surfaces were wet-ground again with P500-grit abrasive paper to remove the scratches made during the previous grinding. Samples were conditioned in distilled water at 37 ±1°C for 28 days. After conditioning the surfaces of the PMMA pieces were dried with filter paper, and the bonding agent Sofreliner Tough M – bonding (Tokuyama, Japan) was applied with a brush. A polyethylene ring with the thickness of 3 ±0.1 mm and the internal diameter of 11 mm was placed in the center of the PMMA piece, the manually mixed material was injected into the ring, a second acrylic sample was placed over the silicone-based material and compressed. Ten samples were made from each material (n = 50). When the curing procedure was finished, the samples were stored in distilled water at 37 ±1°C for 24 ±1 h. Two hours before the end of storing, the handles (M4 screws) were fixed to specimens by the cold-curing PMMA resin (Vertex Castapress, Vertex-Dental B.V., The Netherlands) with specially prepared reducers mounted in the jaws of the universal testing machine. The specimens with handles were placed in distilled water to finish the storing procedure. Finally, after storing, each sample was mounted in reducers again, placed in the jaws of the testing machine and tensile testing was performed at a cross-head speed of 10 mm/min. Tensile bond strength TSB (MPa) was calculated according to the following equation:

$$TBS = \frac{F_{\max}}{A} \tag{7}$$

where: TSB – tensile bond strength, MPa; $F_{\max}$ – maximal force, N; A – cross-sectional area of the internal section of the polyethylene ring, mm^2.

Statistical analysis of the results was performed with the use of the Statistica 12.5 software. The distributions of the residuals were tested with Shapiro-Wilk test and the equality of variances was tested with Levene test. When the distribution of the residuals was normal and the variances were equal, the one-way or two-way ANOVA with Tukey HSD *post-hoc* tests were used (α = 0.05). If the distributions of the residuals were not normal and/or the variances were not equal, the non-parametric Kruskal-Wallis test (α = 0.05) was used.

3. Results and discussion

Representative SEM images of the morphology of sample failures of the obtained composites were presented in Fig. 1. The observations revealed the presence of filler particles and their aggregation, measuring on average about 20-100 nm, but aggregations of about 200 nm were also detected. The obtained dispersion quality was similar or better than for other materials developed for experimental purposes or commercially available, that are intended for this application [23,24]. Obtaining proper dispersion quality of nanofillers in matrix during the process of making nanocomposites is particularly important [25], because

inhomogeneous distribution of the filler and its aggregation has a negative impact on the properties of the obtained materials, including their mechanical properties [27]. On an industrial scale fillers are introduced by mixing machines which use vacuum and usually high shear action [28]. In the case of laboratory scale, which is characterized by a limited quantity of substrates, the technology is developed individually, usually by using the method of mechanical mixing [27], ultrasonic homogenisation [29], sometimes with simultaneous inclusion of dissolution of matrix material in order to lower viscosity of the composition [20]. In the presented study the use of mechanical mixing and ultrasonic homogenization made it possible to obtain the expected results, which is a good prognosis before studies of mechanical and performance characteristics.

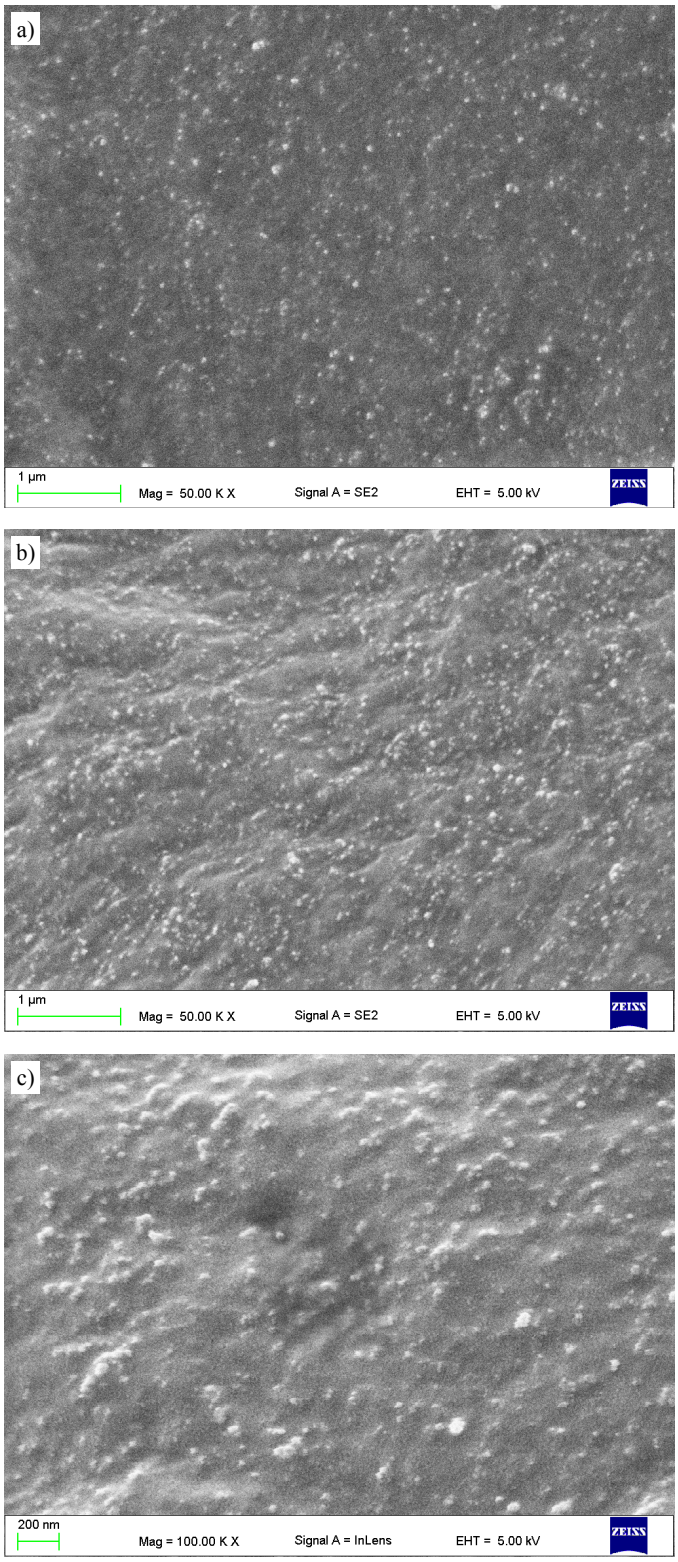

Fig. 1. SEM images presenting the morphologies of the fractured, polymerized materials with 4 wt.% of HMS copolymer and filler concentrations of 15% (a) and 50% (b,c)

The results of hardness tests were presented in Fig. 2. Hardness of materials with a higher FI concentration was statistically significantly higher ($p < 0.01$). This was in accordance with the expectations, because increasing filler concentrations is one of the ways of increasing hardness of silicone materials [26]. Effectiveness of this process depends on many factors like particles' size, their specific surface area or the method of FI functionalization, therefore it is difficult to foresee without experimental studies. Statistically significant impact of HMS copolymer concentration on hardness values ($p < 0.01$) has been proven. The lowest hardness was obtained with an HMS concentration of 2%, what indicates at a too low number of SI-H groups in relation to vinyl groups. Among materials with FI concentration of 15% the highest hardness was achieved at an HMS concentration of 3%. In the case of materials with 30% FI concentration, similar hardness values were recorded after the introduction of 3 to 5% of HMS. Although the increase in hardness of materials for soft denture linings is undesired [10], the most promising materials with a determined FI concentration were the ones with the highest Shore A hardness values after 24 h, because for these composites the proportion of the reacted Si-H groups from HMS and vinyl function groups from PDMS or FI was the most favorable. Earlier precise definition of respective proportions of components on the basis of theoretical considerations was not justified because it is difficult to estimate the number of FI functional groups taking part in the reaction, e.g. due to aggregation of silica filler particles, what was presented by Jia et al. [30] and Maxson [31].

A statistically significant impact of the time of storing in distilled water on hardness values has been registered ($p < 0.01$) but in the case of materials containing 15% of FI hardness stabilized after 14 days from sampling. In the case of 30% of FI after seven days of storing in water the samples still hardened, but hardness changes were slower than during the first week. Material hardening during storing in distilled water should be associated with the continuation of the curing process [9]. This is typical for most materials of this type and it is responsible for e.g. increasing hardness of soft lining materials, e.g. as a result of application of thermal cycles [32]. Simultaneously, the ongoing curing process and the accompanying increase in hardness were for a longer period visible in materials with higher FI concentrations due to a larger number of functional groups from FI that could take part in the reaction. Shore A hardness values as well as their changes in time were in accordance with the ISO 10139-2:2016 standard [17]. Moreover, these values were similar or more favorable than in the case of commercially available silicone based soft lining materials, because after 24 hours and

28 days of storing at distilled water Kim et al. [33] registered 28.9 and 42.8 Shore A units for the softest material, and 50.1 and 57.2 Shore A units for the hardest one, respectively.

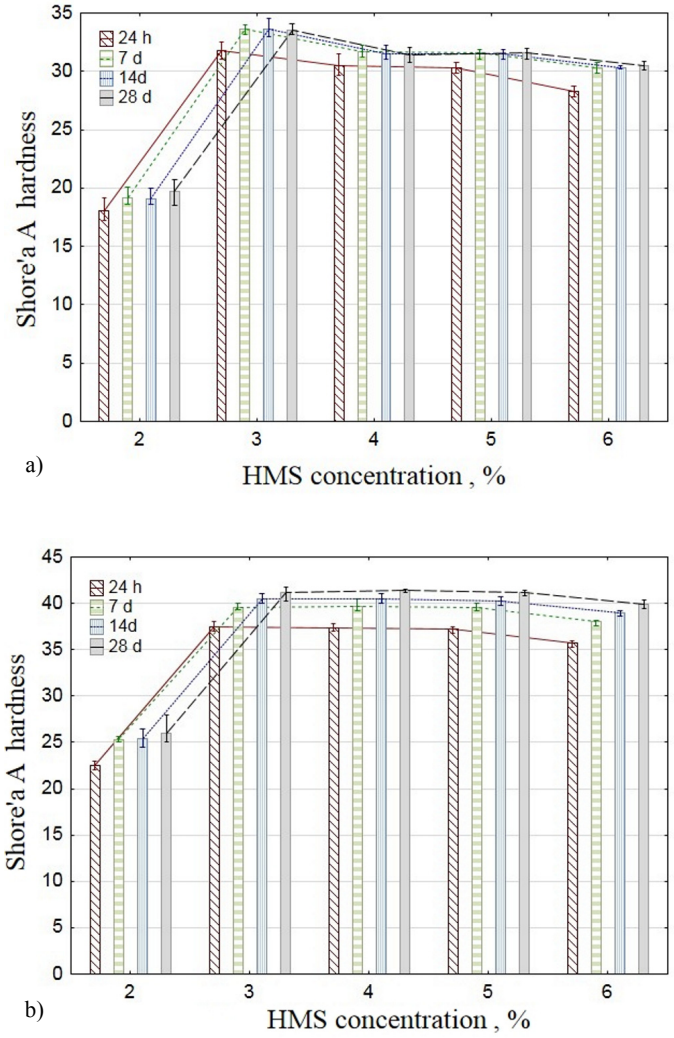

Fig. 2. Medians with minimal and maximal hardness values in Shore A units for materials compounded with 15% (a) and 30% (b) of silica filler

Tensile strength test results were presented in Table 1. Materials with a higher filler concentration were characterized by about two times higher TS values ($P < 0.01$). The use of a higher filler concentration also made it possible to obtain statistically higher ($p < 0.01$) ultimate elongation mean values, which in the case of materials containing 15% of FI were from 250 to 280%, and for 30% from 350 to 420%. This indicates at the expected effectiveness of increasing filler concentration in matrix [34]. The time of sample storage in distilled water at a temperature of 37ºC did not statistically significantly affect the tensile strength of materials containing 15% and 30% of filler ($p = 0.13$ and $p = 0.18$ respectively), what was favorable from the point of view of the planned application. HMS concentration affected TS values in the case of both filler concentrations ($p < 0.01$). For 2% of HMS lower statistically significant TS values were obtained. For materials containing 15% of FI the highest TS values were obtained for 3% of HMS, but the *post hoc* test did not show any differences in comparison to materials with higher HMS concentrations, except for the material containing 6% of HMS. A similar tendency was noted for materials containing 30% of FI, but there were no statistically significant differences starting from 3% of HMS concentration. Both the HMS concentration and water storage time did not statistically significantly influence ultimate elongation values. TS values were similar or better than the ones for currently available commercial polysiloxane-based lining materials, for which these values range from 1.7 MPa to 3.9 MPa [22]. Additionally, TS and elongation values were also comparable to the other experimental materials [24,35].

Sorption and solubility test results for the developed materials were presented in Table 2. Statistical analyses did not show significant differences in sorption and solubility between materials differing in filler concentration ($p = 0.06$ and $p = 0.71$ respectively). HMS concentration did not statistically significantly influence sorption values of materials containing 15% and 30% of filler ($p = 0.06$ and $p = 0.71$ respectively). These results were in accordance with the expectations due to the fact that both the used matrix and the functionalized FI are hydrophobic materials [34]. The obtained sorption and solubility values were a few times lower than boundary values required by the ISO standard (20 μg/mm^3 and 3 μg/mm^3, respectively).

The results of tensile bond strength tests were presented in Table 3. They showed statistically significantly higher ($P < 0.01$) TBS values for materials with a higher filler concentration. All samples were subject to cohesive failures, so there was no debonding of the composite material from denture base material. An example of failures and bonding layer were presented in Fig. 3. In the case of cohesive failures TSB values of liners correlate with TS values of the material, what was demonstrated

TABLE 1

Mean ±standard deviation tensile strength (MPa) for particular materials stored in distilled water*

HMS concentration, wt.%	Filler concentration 15 wt.%			Filler concentration 30 wt.%		
	Storing time			Storing time		
	24 hours	7 days	28 days	24 hours	7 days	28 days
2	1.15±0.1^{A}	1.16±0.03^{A}	1.30±0.08^{A}	2.06±0.28^{A}	2.29±0.31^{A}	2.10±0.38^{A}
3	2.04±0.26^{B}	2.15±0.22^{B}	1.91±0.19^{B}	3.54±0.65^{B}	3.60±0.31^{B}	3.51±0.08^{B}
4	1.86±0.33^{B}	1.93±0.11^{B,C}	1.81±0.27^{A,B}	4.33±0.44^{B}	4.14±0.22^{B}	4.05±0.16^{B}
5	1.87±0.13^{B}	1.93±0.19^{B,C}	1.69±0.27^{A,B}	3.49±0.23^{B}	3.95±0.41^{B}	4.04±0.25^{B}
6	1.73±0.22^{B}	1.55±0.15^{A,C}	1.41±0.26$^{A.B}$	3.50±0.6^{B}	4.02±0.46^{B}	3.82±0.17^{B}

* Groups with the same uppercase superscript letters; (A–C) for each column are not significantly different at the $p > 0.05$ level.

TABLE 2

Mean ±standard deviation sorption and solubility for investigated materials after 7-days' storing in distilled water

HMS concentration, wt.%	Filler concentration			
	15 wt.%		30 wt.%	
	Sorption, μg/mm^3	Solubility, μg/mm^3	Sorption, μg/mm^3	Solubility, μg/mm^3
2	2.55±0.47	0.69±0.28	2.75±0.57	0.59±0.36
3	2.51±1.17	0.61±0.28	2.81±0.47	0.59±0.29
4	2.02±1.01	0.75±0.37	2.61±0.43	0.75±0.29
5	1.88±0.64	0.71±0.41	2.83±0.76	0.71±0.36
6	2.18±0.51	0.53±0.20	2.51±0.61	0.47±0.41

by Mutluay et al. [22]. TSB values are in such a case determined by the properties of the lining material, what in practice means that the strength of bonding area is higher than the strength properties of the analyzed material. Such a situation is desirable, because with time there occurs liquid migration to bonding area [36], what conduces debonding of material from denture base, while strength properties of the silicone material itself remain stable or increase [9]. TSB values for materials containing 15% of FI and materials containing 30% of FI and 2% of HMS were lower than the ones required by the ISO standard (min. 1 MPa). The remaining materials containing 30% of FI were characterized with TBS values conforming to the ISO standard, but statistically highest values were exhibited by the material with 4% of HMS (1.75 MPa). The obtained values were comparable to the results obtained with the application of a similar study protocol for materials used in dentures nowadays, for which mean TSB values were from 0.84 MPa to 2.74 MPa, but usually they did not exceed 2 MPa [22,33].

TABLE 3

Mean ±standard deviation tensile bond strength (MPa) for materials after 24 h in distilled water*

HMS concentration, wt.%	Filler concentration	
	15 wt.%	30 wt.%
2	0.39±0.06 ^{A}	0.75±0.08 ^{A}
3	0.56±0.10 ^{B}	1.35±0.16 ^{B}
4	0.57±0.10 ^{B}	1.75±0.13 ^{C}
5	0.55±0.10 ^{B}	1.42±0.22 ^{B}
6	0.46±0.13 ^{A,B}	1.48±0.27 ^{B}

* Groups with the same uppercase superscript letters; (A–C) for each column are not significantly different at the $p > 0.05$ level.

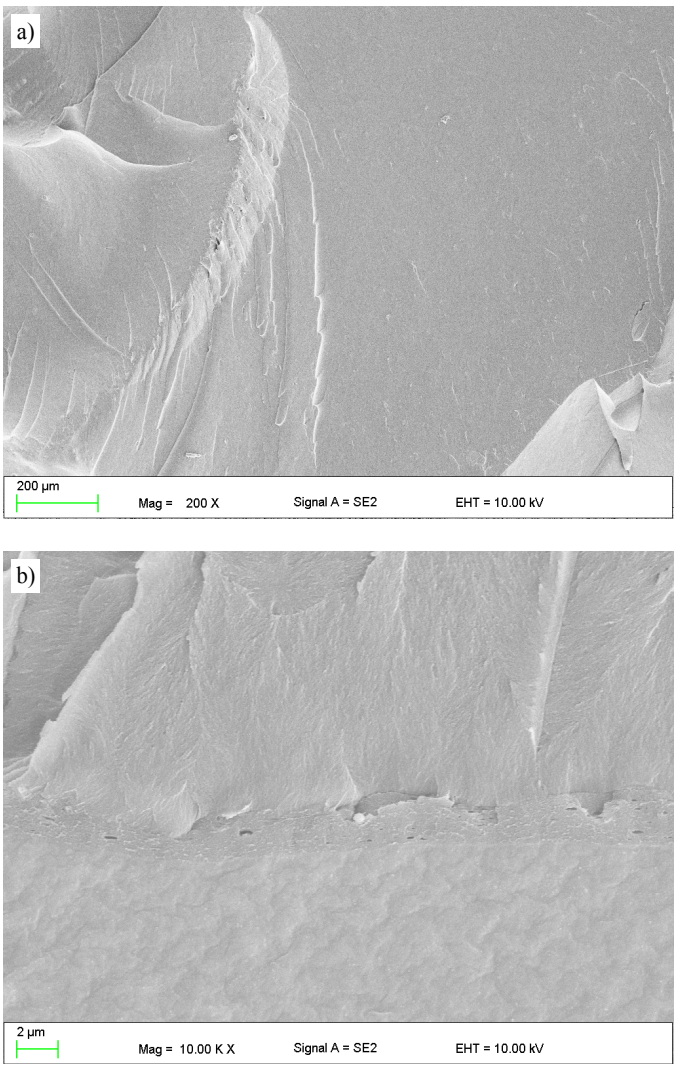

Fig. 3. SEM images presenting the morphology of typical cohesive failures observed during tensile bond strength test (a) and the bonding layer between PMMA resin and silicone composite (b)

4. Conclusions

The investigations of the impact of crosslinker and functionalized silica filler concentrations on the selected mechanical and performance properties of poly(dimethylsiloxane)-based composites allowed to select the optimum composition of the matrix and concentration of filler in view of the application of the developed composites in long-term denture linings. The proposed technology of introducing filler into the matrix has made it possible to obtain good FI distribution and, in consequence, to develop materials characterized by properties suitable for soft denture lining materials for long-term use. These materials can be used in future investigations to develop more complex hybrid composites, containing micro- and nanofillers that would provide materials with specific new performance characteristics. Additionally, relations between particular properties and methylhydrosiloxane-dimethylsiloxane copolymer and hydrophobic silica filler concentrations have been shown.

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