

ORIGINAL ARTICLE

Effects of Tellurium Oxide (TeO₂) Particles on the Thermal Conductivity, Hardness, and Roughness of Maxillofacial Silicone Elastomers

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Abstract

Purpose: The purpose of this research was to evaluate how different concentrations of TeO₂ filler particles influence the thermal conductivity, hardness, and surface roughness of room-temperature-vulcanizing VerSilTal 50 silicone elastomeric materials.

Materials and Methods: A total of 90 samples were prepared by varying the weight percentages of TeO₂ powder (0, 3, and 5 wt. %). Thirty samples from each group were used in the analysis. Two experimental groups with 3 and 5 wt. % of TeO₂ fillers were prepared and subjected to thermal conductivity, surface hardness, and surface roughness tests. The data were analyzed using descriptive statistics and analysis of variance with multiple comparison tests, and a P value <0.05 indicated significance.

Results: Thermal conductivity and hardness improved as the percentage of TeO₂ increased from 3 wt. % to 5 wt. %, compared with those in the control group, whereas surface roughness decreased.

Conclusion: The means for thermal conductivity and surface hardness of the 3 and 5 wt. % TeO₂ experimental groups increased significantly relative to those in the control group.

Keywords: Tellurium Oxide; Silicone Elastomer; Room-Temperature Vulcanizing; VerSilTal 50; Thermal Conductivity; Physical Properties.

1. Introduction

Maxillofacial prostheses were made from a variety of resources, including poly (methyl methacrylate), poly (vinyl chloride), polyurethane, chlorinated polyethylene, and silicone [1]. Silicone elastomers are considered excellent materials for fabricating facial prostheses because of their good biocompatibility, suitable mechanical properties, chemical inertness, ease of handling, and optical transparency. These materials have specific properties that meet the requirements of facial prosthesis manufacturing but still need to be enhanced because of some shortcomings [2]. The structures and properties of silicone elastomers used in maxillofacial restoration have received considerable interest [1]. Biocompatible restorative materials must have sufficient tensile strength to withstand external forces, sufficient hardness and elasticity, tear strength to withstand daily wear and tear, and low surface energy [1].

Nanotechnology has been used in fabricating materials with improved mechanical and physical properties. Advanced and tailored materials with unique physical and mechanical properties not possible in base materials can be created by incorporating nanoparticles into base materials. This advanced technology is used to meet the requirements of basic material applications [3-5].

The thermal conductivity of pure silicone rubber is usually extremely low (0.165 W/m·K). However, silicone rubber with good thermal conductivity can be made by filling it with thermally conductive fillers, such as metal powder, graphite, Al₂O₃, SiC, BN, and ZnO. Fillers improve the thermal conductivity, mechanical properties, and thermal stability of elastomers [6].

Tellurium oxide (TeO₂) has a wide range of application prospects because of its special properties, including high chemical stability, mechanical resistance, high refractive index, good optical nonlinearity, and thermal conductivity [7, 8].

The current study aimed to determine how adding different weight percentages (3 and 5 wt.%) of TeO₂ filler particles affect the thermal conductivity, surface hardness, and surface roughness of VerSilTal 50 (VST-50) room-temperature-vulcanizing (RTV) silicone elastomers.

The null hypothesis (H₀) is that the incorporation of TeO₂ filler particles does not improve physical properties,

and the alternative hypothesis (H₁) is that the addition of TeO₂ filler atoms improves physical properties.

2. Materials and Methods

This study compared thermal conductivity, surface hardness, and surface roughness. Molds were fabricated by custom cutting 2 and 4 mm-thick acrylic sheets (PT. Margacipta Wirasentosa, Indonesia) with a laser cutting machine (JL-1612, Jinan Link Manufacturing Trading Co., Ltd., China). The upper and lower parts are made of plates 4 mm thick (sample thickness required in some tests) rather than 2 mm thick [9]. VST-50 (Factor II, lakeside, USA, cas no. F 15U138R06) is a two-part platinum RTV silicone elastomer. Part A represents the silicone base and part B represents the catalyst.

TeO₂ nanoparticles (Sigma, Germany, cas no. 99 7446-07-3) have been used as filler particles. The particle size analyzer verified that the effective diameter of TeO₂ powder after milling was equal to 137.9 nm as shown in Figure 1. To prevent the dispersion of the TeO₂ filler, silicone was added after weighing the TeO₂ filler with an electronic digital balance (0.000 digit; China; part A). The modified silicon was mixed for 10 min in a vacuum mixer (Multivac 3, Degussa, Germany). To prevent the package from being sucked in, the vacuum was turned off for 3 min and then turned on again for 7 min at 360 rpm and -10 bar vacuum. To produce a homogeneous and bubble-free mixture, the silicon base (part A) or modified silicon (part A and TeO₂) with the silicon catalyst (Part B) for 5 min using a vacuum mixer [10]. A mold was coated with an insulating medium and allowed to dry. The silicone mixture was poured into the mold, which was then sealed with screws and clamps [2]. According to the manufacturer, silicone should be left on the bench for 24 h at 23.2 °C and 50% humidity.

After the silicone material was polymerized at room temperature (23 °C) for 24 h, the mold was opened, and the samples were carefully removed [11]. The samples were rinsed thoroughly with running water, dried with paper towels, and cleaned with a #11 scalpel blade. They were then stored under ideal conditions in a storage container (Polarbag, China) for at least 16 h before testing [11-13].

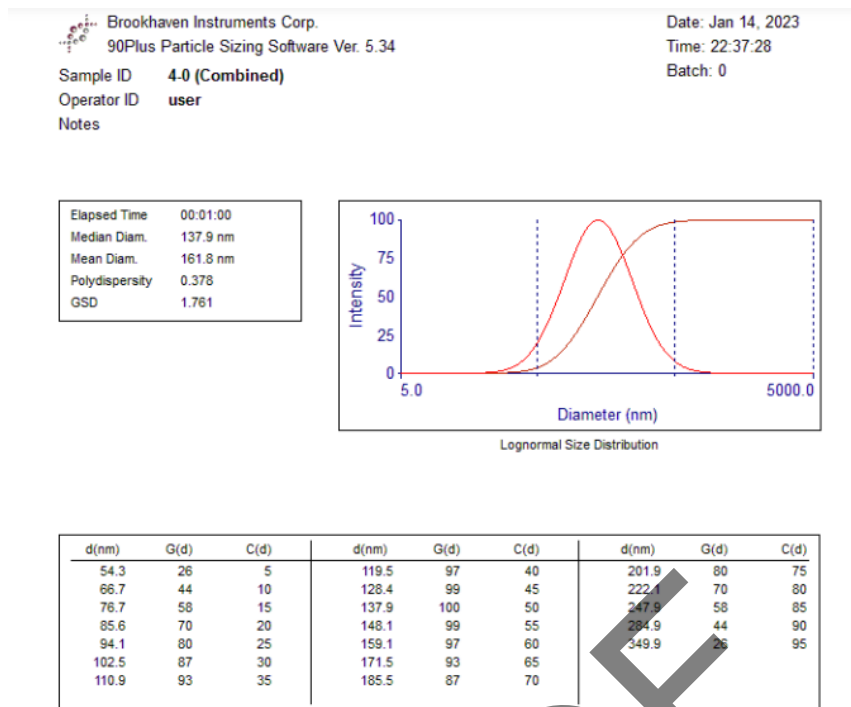


Figure 1. Particle size analyzer result of TeO_2

2.1. Scanning Electron Microscopy (SEM)

To determine the distribution pattern of TeO_2 particles within silicone material; Scanning Electron Microscopy (SEM) was conducted. Three samples were tested: one control sample and two experimental samples containing 3% or 5% TeO_2 particles.

2.2. Thermal Conductivity Test

The Lee disk method has been used to study the thermal conductivity of various materials [14–15]. Thirty samples were prepared using Lee's Disc fixture instructions from the Materials Engineering Department, Technical University (Figure 2). The disc samples had 40 mm radii and were 6 mm thick.

The device was composed of three copper discs (A, B, and C), and each disc had a hole where a thermometer would be inserted. A 60 W electrical plate heater was sandwiched between discs B and C, and the specimen was placed between copper discs A and B. After the clamp screw was tightened to hold all the discs together, the heater was switched on. The whole assembly was placed in an enclosure to minimize the effect of environment temperature. A fourth thermometer was placed within the enclosure,

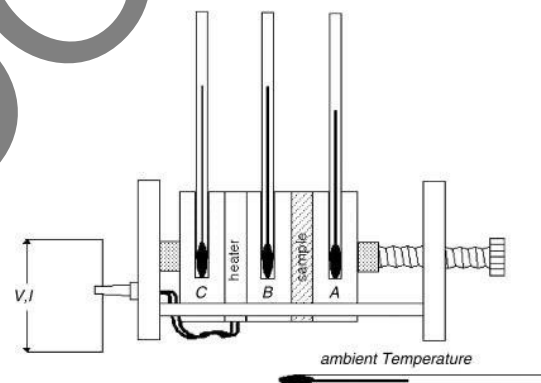


Figure 2. Lee's disk device [16]

close to the apparatus to measure the ambient temperature. When the heater was switched on, discs B and C started to have higher temperatures than disc A because the specimen acted as an isolator. The temperatures in discs C and B were initially different. Readings were obtained at 10 min intervals until equilibrium was reached. These were the mean values when temperature at discs B and C were equal and when the temperatures of all the apparatus's components were stable within ± 0.1 °C for 30 min [10]. At this time, readings were recorded, and thermal conductivity was calculated with Equation 1.

$$e = \frac{V1}{a_A T_A + a_s \frac{T_A + T_B}{2} + 2a_H \frac{T_B + T_C}{2} + a_B T_B + a_C T_C} \quad (1)$$

e: Loss in temperature per time in seconds and difference in temperature between discs and surrounding

V: Voltage within the heater (volt).

a_A, a_B, a_C: surface areas of disc A, B, and C, respectively, in m²

a_s: Surface area of the specimen in m²

a_H: Surface area of the heater in m²

T: Temperatures of discs.

a: Surface area

T_A, T_B, and T_C: temperatures of discs A, B, and C, respectively, in °C; after we obtained the value of e, we calculated the thermal conductivity with Equation 2.

$$k = \frac{ed_s}{2\pi r_s^2(T_B + T_A)} \left[a_s \frac{T_A - T_B}{2} + 2a_A T_A \right] \quad (2)$$

K: thermal conductivity in w/m·°C

d: thickness of the specimen in m

r: diameter of the specimen in m

T_A, T_B, and T_C: temperatures in discs A, B, and C measured in °C.

2.3. Surface Hardness Test

Thirty samples were fabricated according to the specifications of ISO 7619-1:2010 [17]. The sample size was 25 mm × 25 mm, and the minimum thickness was 6 mm.

In this study, surface hardness was measured using a digital Shore A durometer (HS-A, Ezitown, China) with a 1.25 mm diameter blunt indenter. The surface of each sample was marked with five dotted areas. Marking points were 6 mm from the center and spaced apart. The surface hardness for each sample was calculated by averaging five readings.

2.4. Surface Roughness Test

Thirty samples were fabricated according to the specifications of ISO 7619-1:2010 [17]. The dimensions of the specimens were the same as those of the surface hardness specimens.

A profilometer (TR200, China) with an accuracy of 0.001 μm was used to measure the surface roughness. The device was equipped with a sensitive diamond tip (surface analyzer) to follow the contours of surface irregularities. The device was adjusted so that the stylus touched the sample surface at only three different points to obtain three readings per sample, which was placed on a stable and hard surface. The stylus moved along the specified surface (11 mm) to touch the first point. The reading was displayed on the digital scale of Ra. This parameter represented a series of single measurement values of the peak and valley of the average value of the surface [18]. The average of the three measurements was then obtained as the roughness value [19].

2.5. Statistical Analysis

Data for this study were evaluated using the Statistical Package for the Social Sciences (version 23) program. The following statistical tests were performed:

Box plots were used for descriptive statistics.

The inferential analysis included an analysis of variance (ANOVA) table to compare all group means, and Levene's test was used for homogeneity of variance, and Bonferroni multiple-comparison test was performed to determine the significant differences between groups.

Statistically, a P-value ≥0.05 is not significant, a P-value <0.05 is significant, and a P-value ≤0.01 is highly significant.

3. Results

3.1. Scanning Electron Microscopy (SEM)

SEM results of VST-50 silicone and before and after the addition of 3% and 5% TeO₂ powder are shown in Figures 3-5, respectively. This test showed a good dispersion of the micro fillers and incorporation within the polymeric matrix of silicone and showed that there was a slight little agglomeration of micro filler as filler loading increased.

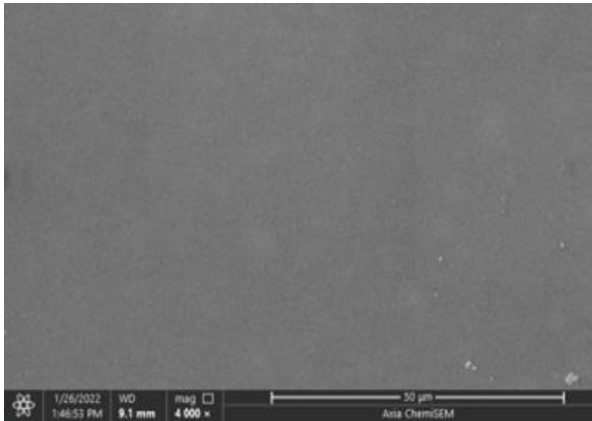


Figure 3. Scanning electron microscopy image of control specimens; 4000 magnification

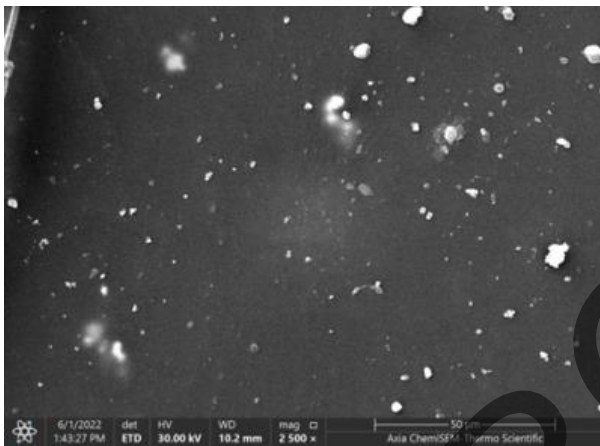


Figure 4. Scanning electron microscopy image of 3wt. % TeO₂ specimens; 2500 magnification

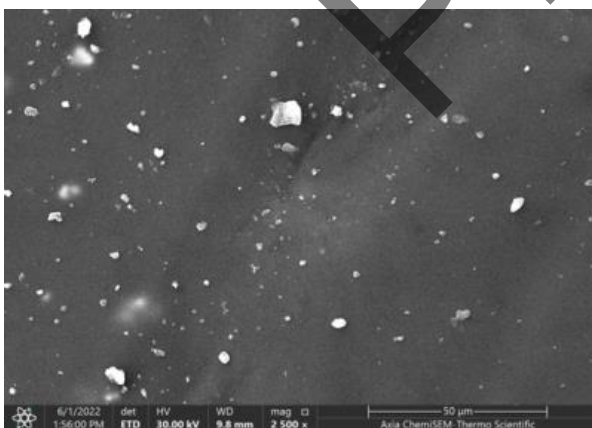


Figure 5. Scanning electron microscopy image of 5 wt. % TeO₂ specimens; 2500 magnification

3.2. Thermal Conductivity Test

The test group (5 wt. %) had the highest mean (0.288 W/m·°C), followed by the 3 wt. % group with

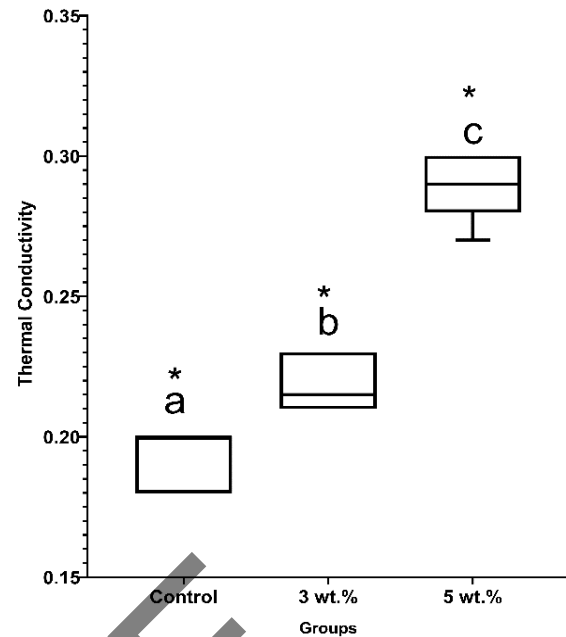


Figure 6. Box plot of thermal conductivity showing the means and standard deviations for all groups in W/m·°C; values with different letters showed a significant difference at P value <0.001

a mean of 0.2180 W/m·°C. The control group had the lowest mean (0.1940 W/m·°C; Figure 6). One-way ANOVA results showed highly significant differences ($P < 0.01$) among all test groups. Levene's test was used to assess the homogeneity of variances and select the type of multiple comparison. Bonferroni test was used to examine the mean of the study group. The difference between the control group and the 3 wt. % or 5 wt. % group was extremely significant, and the mean difference among the groups was highly significant ($P < 0.01$).

3.3. Surface Hardness Test

The test group (5 wt. %) had the highest mean (35.30 A), followed by the 3 wt. % group with a mean of (35.12 A) and the control group with a mean value of 33, 14 A (Figure 7). One-way ANOVA results showed highly significant differences (P value < 0.01) among the test groups. Levene's test was used to assess the homogeneity of variances and select the type of multiple comparison. Bonferroni test was used to check the mean difference of the study groups. P value < 0.01 indicated a highly significant difference between the control group and each group (3 and 5 wt. %), whereas a nonsignificant mean difference was found between the 3 and 5 wt. % groups ($P > 0.05$).

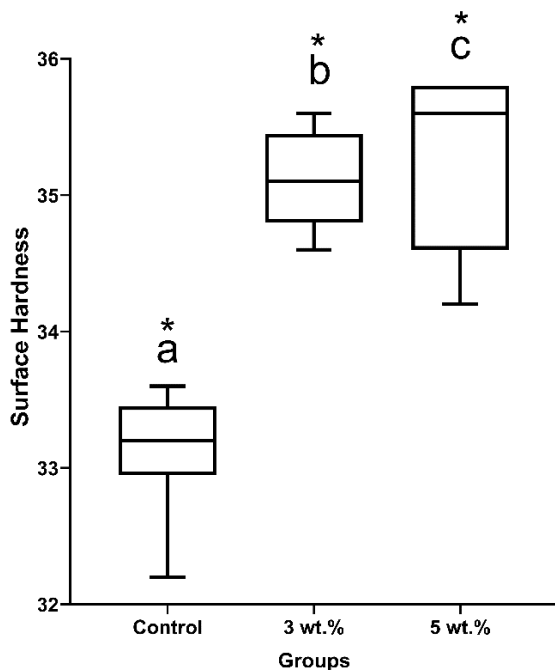


Figure 7. Box plot of shore A hardness represents the means and standard deviations from the mean for all study groups; values with different letters showed significant differences at P value <0.001

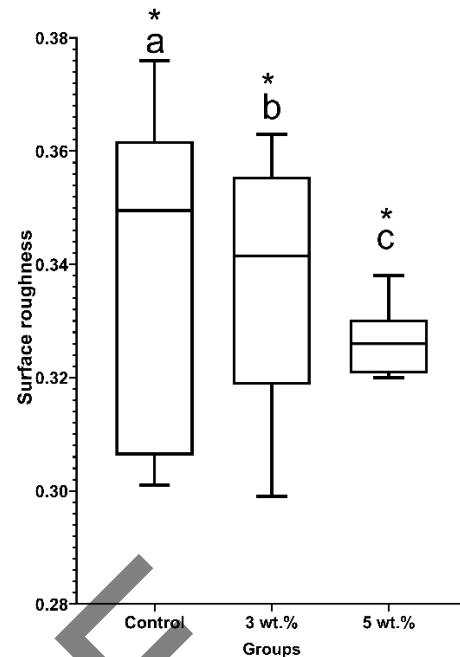


Figure 8. Box plot of surface roughness test represents the means and standard deviations from the mean for all study groups; values with different letters showed significant differences at P value <0.001

3.4. Surface Roughness Test

The control group had the highest mean (0.3381 Ra), followed by the 3 wt. % group (0.3364 Ra) and 5 wt. % group (0.3262 Ra) as shown in Figure 8. The results of one-way ANOVA showed no significant difference between the test groups ($P > 0.05$). Levene's test was used to determine the homogeneity of variance and the type of post hoc test to use when the P value shows a significant difference. Gameshowell multiple comparison test selected.

4. Discussion

The rate at which heat may be conducted through a material cross-section at a specific period is known as thermal conductivity and is one of the main thermal qualities of dental materials [20]. The thermal conductivity of pristine silicone elastomer is 0.2 W/m·K [21]. The thermal conductivity of composite materials depends on the composition, shapes, sizes, and loading levels of fillers and the intrinsic thermal conductivity of fillers and matrices [22–24].

Compared with the control group, the 5 wt. % group shows the highest increase in thermal conductivity, followed by the 3 wt. % group.

As concentration increases, nanoparticles start to develop dense structures inside matrices, which are firm and thick and provide heat conductive pathways because of the huge volume proportion of nanoparticles at identical mass fractions. However, as filler loading increases, contact among nanoparticles increases. The condition of filler diffusion is crucial. Filler units must be cohesive to develop continuous heat conduction paths and enhance thermal properties. At low concentrations, stuffed silicone rubber demonstrates relatively low thermal conductivity compared with what is expected probably because of the failure of nanoparticles to develop perfect thermally conductive pathways. However, the superfine sizes and large surface energy of nanoparticles inhibit them to scatter uniformly in silicone rubber. Consequently, most nanoparticles are disintegrated in silicon rubber and cannot produce conductive pathways in matrices [25-26].

The results agree with those of Mu *et al.* (2007), who studied the thermal conductivity of silicone rubber filled with ZnO. As the content of ZnO particles in silicone rubber, the amount of formed conductive chains

increases and conductive chains tend linearly to increase the thermal conductivity of composites [6].

The results also agree with those of Zhou *et al.* (2007), who studied the effect of the particle size of Al_2O_3 on the properties of filled heat-conductive silicone rubber and found that particles begin to make contact with one another and form compact packing structures as filler concentration increases. Therefore, the thickness of matrix resin layers among adjacent particles decreases, and thermal conductivity is considerably enhanced because of decreased thermal contact resistance [27].

A material used in a maxillofacial prosthesis should have the same hardness as the missing facial part to be replaced and should be relatively soft. On a shore A scale, the hardness of commercially available silicone ranges from 16 A to 45 A, but a good material should be in the 25–35 A range, which is the resiliency required for the prosthesis to respond to facial motion [28–29].

The results show a significant increase in hardness (3 wt. % TeO_2) after the addition of 5 wt. % TeO_2 . Increasing TeO_2 content correlates with increasing hardness values. The increase in hardness values after the addition of 3 and 5 wt. % TeO_2 is likely due to the dispersion of filler particles and the formation of interfiller networks within silicone matrices and between polymer chains. This effect increases the penetration force and penetration resistance [30–31]. The TeO_2 filler particles have strong ionic atomic bonds, resulting in the desirable material properties of hardness and strength [32].

Surface roughness is an important indicator of materials' mechanical properties because surface irregularities can act as crack nucleation sites [33].

The surface roughness results show that the average roughness of the experimental group decreases compared with that of the control group. The lowest surface roughness was observed in the 5 wt. % group. Reduction in surface roughness may be due to sequential polymerization that promotes the assembly and complementarity of polymer chains and produces fine and smooth silicone surfaces. Due to polymerization kinetics, the presence of fillers can influence the rate at which polymerization occurs. TeO_2 fillers may act as nucleating agents, providing sites for polymer chain initiation and accelerating the reaction [34]. This result can be also due to the extremely small and well-dispersed TeO_2 particles (Figures 2, 3, and 4). Moreover, the

surface roughness test refers to the outer surface of a composite rather than its inner surface. Therefore, if only a small amount of TeO_2 particles are added to a silicone elastomer, the number of particles on the sample surface would be negligible [23].

5. Conclusion

Under the conditions set in this study, the thermal conductivity, surface hardness, and roughness improved after the addition of TeO_2 filler particles to RTV VST-50 maxillofacial silicone elastomers as the concentration of TeO_2 increased.

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