Effect of 3-methacryloxypropyltrimethoxysilane on diametral tensile strength of rice husk silica-based dental composite

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ABSTRACT

Introduction: Rice husk silica has been studied as a filler in dental composite, however, the mechanical properties of the resulting composites are below the commercial due to the poor surface modification between silica and resin matrix. 3-methacryloxyprpyltrimethoxysilane (MPTS) is one of the coupling agents that are effective to modify the silica surface. The purpose of this study to analyze the effect of MPTS on the diametral tensile strength (DTS) of the self-made composite using rice husk silica. **Methods:** The research type of this study was an experimental research laboratory. The samples (26 samples) were divided into two groups, namely the test group (using MPTS-modified silica) and the control group (using non-modified silica). The samples were cylindrical in shape with a diameter of 6.0 ± 0.1 mm and a height of 3.0 ± 0.1 mm according to ANSI/ADAS No. 27, 1993 and ISO 4049, 1988. The Fourier Transform Infrared (FTIR), HORIBA, was used to characterize the MPTS grafting on the silica surface. The DTS value is measured by Universal Testing Machine (UTM), Lloyd LRX Plus. **Results:** FTIR presents the additional peak of carboxyl and methacryloyl group at 1716, 1555, and 1410 cm-1 wavenumber, respectively in MPTS-modified silica. The DTS value of the test group was 43.40 ± 4.43 MPa and the control group was 25.80 ± 2.63 MPa. The test group was significantly higher than the control group (p-value = 0.001). **Conclusion:** The MPTS is effective to enhance the DTS value of rice husk silica-based composite.

Keywords: Rice husk silica; dental composite; diametral tensile strength; 3-methacryloxypropyltrimethoxysilane.

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INTRODUCTION

Composite resin is direct restorative materials that are widely used today to restore damaged tooth tissue because it has adequate aesthetic and mechanical properties as a restorative material.[1,2] Currently, due to most of the commercial composite resins used in Indonesia being imported from other countries, the costs involved are quite expensive. Various efforts have been made to develop and produce composites by acting on Indonesia's natural resources, such as using zircon sand as a filler.[3,4] Filler is one of the components of composite resin that is widely studied due to its role in the mechanical properties of composite resin.[5,6,7]

Diametral tensile strength (DTS) is the important mechanical properties of composite resins as restorative materials. The diametral tensile strength of composites is strongly influenced by the filler characteristics such as composition, surface properties, etc.[8,9,10] Based on composition aspect, silica is one of the most frequently used composite fillers today.1[1,12] Currently, rice husks, one of the natural materials that can be used as a source for silica, have been developed as natural-based composites resin. Rice husk is obtained from waste rice mills that are usually left piled up and underutilized. The utilization of rice husks as a source of silica can reduce agricultural waste and increase the sustainability of the environment.[13,14,15] Unfortunately, the use of rice husk silica as a self-made composite in Indonesia has never been done.

Another characteristic of filler composite that affects the DTS is the surface properties.[9] 3-methacryloxyprpyltrimethoxysilane (MPTS) is one of the most widely used coupling agents to modify the surface properties of filler and gain chemical bonding between filler and resin matrix.[16] Research by Kleczewska et al. stated that MPTS is effectively used to modify the silica surface as a composite filler.

MPTS has a highly reactive methacryloyl group at the end of its chain, which will provide good chemical reactivity with methacrylate-based composite matrices, and a methoxy group that will bind to inorganic fillers.[17] In a previous study, rice husk silica-based dental composite have lower mechanical properties compared to commercial composite. Rice husk silica that incorporates resin matrix is not modified by MPTS.[18] This study aims to analyzed the effect of adding the MPTS to rice husk silica on the DTS value of self-made composites.

METHODS

The type of research used in this study was an experimental research laboratory. The materials used in this research were rice husk silica as filler material for making self-made composite and MPTS as coupling agents. The rice husk silica was obtained from the Chemical Inorganic Laboratory of the Department of Chemistry Universitas Padjadjaran. The Diurethane dimethacrylate (UDMA), 2-Dimethylaminoethyl methacrylate (DMAEMA) and Chomporquinone (CQ) were obtained from Sigma-Aldrich®.

The research procedure consisted of 3 stages. The first stage was to modify the surface of rice husk silica using MPTS. The second stage was making the self-made composite by combining rice husk silica with resin matrix. The third stage was to test the diametral tensile strength. Self-made composite samples tested for diametral tensile strength were divided into two groups, namely the test group (MPTS-modified filler, n= 13), and the control group (non-modified filler, n= 13).

The first stage began by modifying the surface of the rice husk silica by silanization using MPTS via the calculation of Joana et al.17 In this study, the weight of the filler used was 9 grams. The surface area of the filler obtained from the BET test was 38.032 m2/g and The minimum coverage area of the MPTS was 314 m2/g. Therefore, the amount of silane required was 1.09 grams (1.04 mL). Subsequently, a 3% MPTS solution was made by dissolving 1.04 mL of MPTS into 33.6 mL of the solvent containing water and ethanol with a ratio of 1:3 and 0.35 mL of acetic acid. After that, the filler was dried at 60ºC in the incubator for 3 days. Non-modified filler and MPTS-modified filler were characterized by Fourier Transform Infrared (FTIR) from HORIBA.

The second stage was making the self-made composite samples (the test group and the control group). The resin matrix consisted of the following components: UDMA (100%), CQ (0.05%), and

DMAEMA (0.1%). The Procedure began by mixing and stirring the resin matrix components for one hour on a magnetic stirrer. Later, the filler was gradually poured into the matrix until the paste consistency was obtained. Weight percentage of filler/resin (WP F/M) was achieved by preliminary study for consistency. The paste consistency was achieved in a WP F/M of 53 wt%: 47 wt% for the control group. On the other hand, the test group achieved the same consistency in an WP F/M of 65 wt%: 35 wt%. Furthermore, the composite paste was taken gradually and placed into the mold (4 x 6 mm) followed by the process of incrementally curing using DENTSPLY Caulk's SmartLite® Focus® LED with a light intensity of 1000 mW/cm2.

The light cure device should be placed as close to the surface as possible, approximately 1 mm without touching the surface of the self-made composite. After that, the samples were polished and placed into a container containing artificial saliva and then stored in an incubator at 37ºC for 24 hours. In the third stage, the self-made composite was tested for diametral tensile strength (DTS). The DTS was carried out using a Universal Testing Machine (UTM), Shimadzu, with a crosshead speed of 1.0 mm/min until the samples were broken.

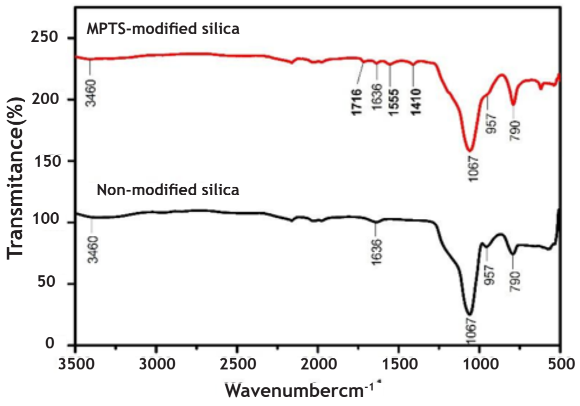
Statistical analysis

Acquired data were analyzed using an independent t-test via software of SPSS statistics 23.0 to analyze the significant difference in DTS value between the test and control groups.

RESULTS

FTIR characterization aims to identify the MPTS grafting on the rice husk silica surface. The FTIR spectrum in figure 1 shows the difference in functional groups from the non-modified silica and MPTS-modified silica. The MPTS-modified silica showed the addition of carboxyl functional groups (C=O) at 1716 cm^{-1} wavenumber and methacryloyl group (C-H) at 1555 and 1410 cm^{-1} wavenumber.

Both non-modified and MPTS-modified silica show similar peaks at 3460 and 1636 cm^{-1} that are attributed to the stretching modes of silanol (Si-OH) and hydroxyl groups (-OH). Transmittance band regions in 1067 and 790 cm^{-1} are assigned to the vibration of Si-O-Si groups in



The FTIR spectrum shows the MPTS-modified silica having some additional peaks at 1716, 1555, and 1410 cm^{-1} wavenumber. Non-modified silica shows the characteristic of silica at 3460, 1636, 1067, 957 and 790 cm^{-1}.

the silica structure. The presence of a 957 cm^{-1} peak is related to unhydrolyzed Si-OC2H5 groups. The DTS results with a sample of 13 self-made composites using MPTS-modified silica and 13 self-made composites using non-modified silica can be seen in Table 1. It shows that the test group has a

Table 1. Diametral tensile strength of two groups of self-made dental composite samples (control and test groups)

No	Diametral tensile strength (MPa)w	
	Control Group	Test Group
1	22.40	45.50
2	27.10	47.80
3	27.40	48.60
4	30.10	37.70
5	21.10	48.00
6	24.20	34.30
7	25.10	39.50
8	29.40	42.40
9	24.40	43.50
10	24.10	42.00
11	27.60	43.30
12	27.30	48.90
13	25.30	43.20
Means	25.80	43.40
Standart Deviation	2.63	4.43

highest DTS value of 48.90 MPa while the control group has a highest DTS value 0f 30.10 MPa.

In Table 1, the test group possesses the highest values (43.40 MPa) compared to the control group (25.80 MPa). Figure 2 shows the p-value lower than

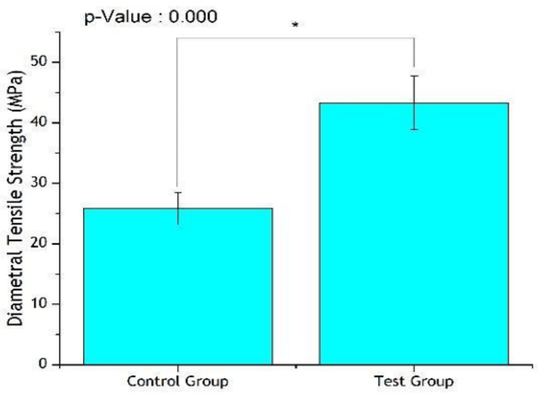


Figure 2. The test group shows a statistically higher DTS value compared to the control group with a p-Value 0.001.

0.005. It means there is a statistically difference in DTS value between the control and test groups.

DISCUSSION

Silica has hydrophilic properties so that it is difficult to disperse in a hydrophobic resin matrix. Surface modification was carried out to improve the mechanical properties of dental composite by increasing the compatibility of the inorganic filler and methacrylate-based resin. Silane coupling agent is the most frequently used to modify the silica by reducing the tendency of silica fillers to agglomerate. Monolayer silane that is grafted on a filler surface is more desirable after silanization. Thicker layer will reduce the share of covalent bonds in filler-matrix interphase and the stress-transfering ability of the dental composite.[17,19,20]

The FTIR result (Figure 1) shows that hydrolyzed MPTS was grafted on the surface of silica. The stretching vibration of the carboxyl group (C=O) at a wavenumber of 1716 cm-1 is the characteristic of MPTS. The peak intensity is low indicating the monolayer of the MPTS attachment. The methacryloyl groups indicate the silica already has a reactive functional group. It means that the silica is ready to bind to the methacrylate-based resin. Compared with the FTIR of non-modified silica, the other showed lower hydroxyl (-OH) group peaks. This indicates that fewer free -OH groups are unbounded to MPTS.[17,21]

MPTS is a type of coupling agent that is often used to modify the surface of silica. The MPTS has a methoxy group that can undergo hydrolysis and reacts with the hydroxyl group belonging to the filler. In addition, the MPTS also has a methacryloyl group which binds to a methacrylate-based matrix, so that the MPTS chemically combines the filler material with the matrix in a composite. Silane modification of the surface of silica will increase the mechanical strength by increasing the compatibility between filler-matrix and weakening the interaction between fillers – fillers.[19,21,22]

The DTS result shows the test group has a higher average value than the control group (Table 1 and Figure 2). The further explanation can be stated that the MPTS increases the wettability of the resin matrix toward the silica. Wettability is the ability of a liquid matrix to spread evenly over the surface of a solid. As the wettability of the matrix increase, the amount of resin matrix required to reach the wet point of a filler is reduced, thus allowing a composite containing more filler to be wetted by a certain amount of resin matrix.[19,20] In this study, the test group can reach 65% by weight of the filler in the composite, while the control group contains 53% by weight of filler.

On the other hand, non-modified silica fillers are hydrophilic on the surface. This property causes the filler to tend to agglomerate and be less dispersed in the matrix thereby decreasing the amount of content. Another reason for the high DTS value of the test group is the increased stress transfer received by the resin matrix to MPTS-modified silica.[9] As the thory stated, Silane coupling agent alows the flexible resin matrix to transfer stress to the hogher-modulus filler particles.[23]

The DTS of commercial composite resins ranges from 34 to 45 MPa. For instance, dental composite Vit-l-escence that uses silica as a filler has a DTS value of 34.45 MPa. Meanwhile, the self-made composites have been carried out in previous studies using 3-mercaptopropyltrimethoxysilane (MPTS) and 3 -aminopropyltrie hoxysilane (APTS). They have the DTS value of 13.78 and 8.90 MPa, respectively. The lower DTS value indicates a poor bond between the matrix and the filler.24 In this study, the self-made composite made from rice husk silica filler using MPTS has a DTS with an average of 43.40 MPa. It can be stated that the MPTS plays a role in producing a composite restorative material with adequate DTS. This also shows that the self-made composite is adequate when compared to the DTS value of commercial composite resins. Although, it is difficult to compare with commercial composite because the specification of the test equipment may differ.

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