Tribological Evaluation of Dental Composite Resins Containing Prepolymerized Particle Fillers

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The Tribological evaluation of commercial dental composite resins containing prepolymerized particle fillers was investigated. Composite resins such as Metafil, Silux Plus, Heliomolar, and Palfique Estelite were selected as specimens. In the wear tests, a ball-on-flat wear test method was used. The friction coefficient of Metafil was quite high. The wear resistance of Silux Plus and Palfique Estelite was better than that of Metafil and Heliomolar under the same experimental conditions. The main wear mechanism of the composite resins containing prepolymerized particle fillers was abrasive wear caused by the brittle fracture of the prepolymerized particles and the debonding of the filler and the matrix.

Key Words : Composite Resin, Dental Materials, Pre-Polymerized, Wear Mechanism, Specific Wear Rate

1. Introduction

The dental composite resins, extensively used in restoring anterior and filling cavities of posterior teeth. Many clinical reports have their superior performance in restoration compared with amalgam, which has the disadvantage of mercury releasing(Lutz et al., 1984 and Bayne et al., 1994). However, composite resins also have certain drawbacks such as shrinkage during polymerization, low compressive strength, low tensile strength, and wear resistance(Bowen et al., 1983, Goldman, 1983, Gibson et al., 1982 and Hendriks et al., 1987). Therefore, there is still a need for concentrated research in this area.

Recent studies have focused on fillers, resin monomers, polymerization initiator, and silane-coupling agents in order to improve their tensile strength, wear resistance, grinding, and esthetic characteristics(Suzuki et al., 1991, Degani, 1992, Taira et al., 1988 and Mohsen et al., 1995). Of these studies were particularly focused the development of micro filler to improve its grinding properties and wear resistance. Since a micro filler has a larger surface, this decreases its content in resin matrix. To solve this problem, micro fillers were pre-mixed with a resin monomer and then polymerized. After being crushed, the crushed particles, prepolymerized particle fillers, were then re-mixed with the micofillers and resin monomer to increase their content. The failures of brittle materials, such as porcelain and composite resins in dentistry, usually result from the fracture

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of the surface or substructure because of their brittleness. Accordingly, an assessment of their critical tensile strength is also important(Higo et al., 1991). The size and shape of the filler have an effect on both the initiation and propagation of the cracks in the composite resins. Similarly, the size and shape of a prepolymerized particle fillers also have a significant effect on the fracture mechanisms of composite resins containing prepolymerized particle fillers. The friction and wear characteristics are closely related to the size, shape, distribution, density of the filler, and resin matrix, etc. (Suzuki et al., 1991, Wassell et al., 1994 and Li et al., 1985).

In this study, fracture toughness, and wear tests were conducted to understand the fracture and wear characteristics of composite resins containing pre-polymerized particle fillers.

2. Materials and Experimental Method

2.1 Materials and specimen preparation for ground surface observations

For commercial composite resins containing prepolymerized particle fillers used as experimental materials, such as Metafil (Sun-Medical Co., Osaka, Japan), Silux Plus (3M Co., St. Paul. MN, USA), Heliomolar (Vivadent Co., Schaan, Lulechlenstein) and Palfique Estelite (Tokuyama Co., Tokuyama, Japan). Table 1 shows the chemical composition of fillers of these composite resins.

Specimens for SEM observation of ground surface texture made by filling composite resins into a mold (5mm width, 2.5mm thickness) and light curing for 1 min were embedded in a chemical cured epoxy resin and stored in water at 37℃ for 24 hrs. The surface of the composite resins was polished with water/proof silicon-carbide paper in sequence of #800, #1000 and # 1200 and then polished with diamond pastes (6μm, 3μm, 1μm, 0.25μm, 0.1μm). After vapor coating with carbon, the surface were examined with a field emission scanning electron microscope (S-4200, Hitachi Co., Japan).

2.2 Determination of weight filler contents

The weight filler contents of the composite resins were determined by the standard ash method of ISO NO. 4049. The weight of a rectangular specimen (2.5×5×10mm) (W_0) was measured with an electric balance, and after heating the specimen in an electric furnace at 600℃ for about 30 min to burn out the organic matrix, the weight of the specimen (W_1) which consisted only of inorganic filler was calculated. The weight percentage of filler content of composite resins was determined from the weight difference between W_0 and W_1. The filler volume fraction of inorganic filler was calculated by the equation reported elsewhere(Kim et al., 1991).

2.3 Fracture toughness test

Specimens for fracture toughness were made in a split mold (30mm(L)×5mm(W)×2.5mm(B)) containing a razor blade notch fixed into a slit in the mold (single edge notch specimen). The resulting notch/width ratio of the specimen was controlled in the range of 0.45-0.55.

After packing the resin into the mold, the specimens were illuminated in five 30s illumination steps along each side with a light curing machine (Powerlite 100, USA). The light-cured specimens were removed from the mold, trimmed and stored in distilled water at 37℃ for 24 hrs before testing.

The specimens were tested by three-point bending in an Instron testing machine (4202, Instron Corp., Canton, MA, USA) at a cross-head speed of 0.1mm/min. The stress intensity factor, K_Q (MPa·m$^{0.5}$) was obtained from the peak load (P_Q) and specimen configuration by AETM-E399.

2.4 Hardness test

Specimens for hardness test were made in a metal mold size of 5 mm diameter, 1 mm thickness. After packing the resins into the mold, the specimens were illuminated for 30 secs with light curing machine(Powerlite 100, U.S. A.). Then the specimens were stored in distilled water at 37℃ for 24 hrs.

The Vickers hardness test under 200 gf for 15

Table 1 Chemical compositions of fillers of composite resins examined

Materials \ Elements	O	Na	F	Al	Si	Zr	Yb
Metafil	52.72			1.17	46.11		
Silux Plus	50.23			1.15	48.63		
Heliomolar	28.05		1.03	1.45	35.78		33.68
Palfique Estelite	44.17	2.03		1.28	35.55	16.97	

Table 2 Filler fractions, Vickers hardness, and fracture toughness of composite resins containing prepolymerized particle fillers

Brand Name	Filler[vol%]	Hardness[Hv]	K_{IC}*[$MPam^{1/2}$]
Metafil	25.7	48.18	0.63±0.01
Silux Plus	31.7	63.18	0.81±0.07
Heliomolar	41.2	54.31	0.84±0.02
Palfique Estelite	51.1	71.74	0.85±0.01
Si_3N_4		1562	

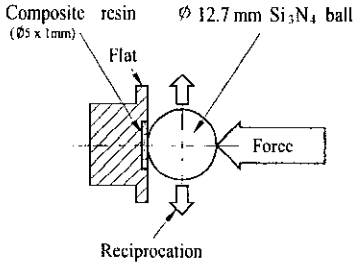


Fig. 1 Schematic illustration of ball in flat wear test

seconds(Future-Tech, FM-7, Matsuzawa Co., Japan) was conducted three times on the surface of one specimen.

2.5 Wear test specimen preparation and wear test

After packing the resins into the mold(Fig. 1), the specimens were light-cured for 30 secs, and stored in distilled water at 37℃ for 7 days. Then the surface of the specimens was polished with water-proof silicon-carbide paper in sequence of #800, #1000, #1200 and #1500. Surface roughness of polished specimen was detected about 0.1μm Ra.

Wear test machine used in this experiment is a self-made linear repetitive assembling one. Dead weight from 1N to 100N can applied on the specimen. Characteristics of this wear test machine is that the sliding velocity and sliding distance can be controlled freely, respectively from 0.1 to 1.0mm/sec of sliding velocity and from 0.2 to 10mm of sliding distance. Friction force detected during wear test was amplified and recorded into computer. Wear test was conducted by ball-on-flat tribometer method, as illustrated in Fig. 1, which is the method that the ball, applied constant load, run linearly on the flat specimen. The test was conducted in room temperature and dry condition. The test conditions were 50mm/min of siliding speed of ball and 9.8 N of applied load.

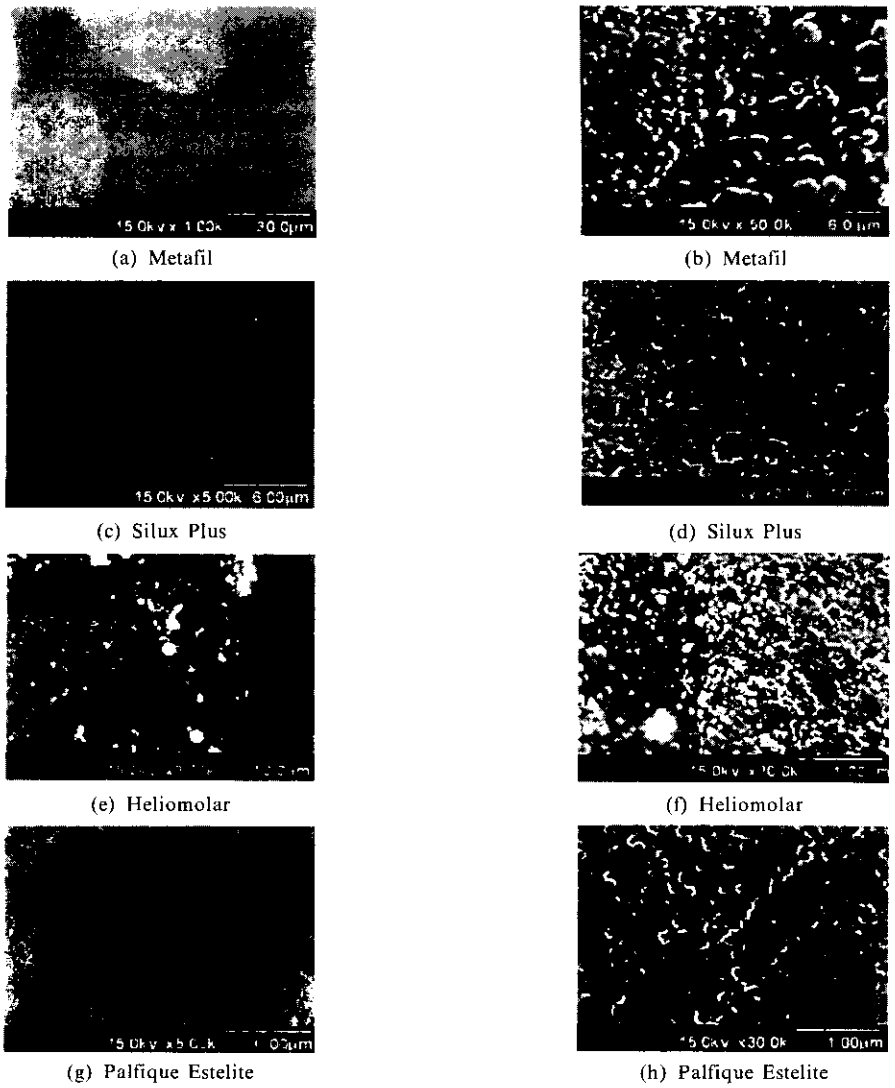
Antagonist was the silicon nitride(Si_3N_4) of 12.7mm diameter and to exclude the effect of ball itself, the position of ball was changed at every 1000 cycles.

Friction coefficient(μ=F/P) was calculated by detected friction force(F) on the load cell and perpendicular load(P) applied on the specimen. Wear volume was calculated by detecting the linear distance of wear track by surface roughness tester. Specific wear rate($Ws=W/P\cdot L$) was calculated by wear volume(W), applied load(P), and total siliding distance.

3. Results and Discussion

3.1 Ground surface, filler fractions, hardness, and fracture toughness

SEM photographs of composite resin containing prepolymerized filler particles are shown in Fig. 2. Round shaped prepolymerized filler particles with sizes of 30-60μm, and irregular shaped prepolymerized filler particles with



(a) Metafil (b) Metafil

(c) Silux Plus (d) Silux Plus

(e) Heliomolar (f) Heliomolar

(g) Palfique Estelite (h) Palfique Estelite

Fig. 2 SEM photographs of ground surfaces of specimens

sizes of 5-10μm, are shown in Metafil. Round fillers with sizes of 0.1-0.2μm are included in the resin matrix and round fillers smaller than 0.05μm are seen in irregular shaped prepolymerized particle filler. Prepolymerized filler particles of various shapes and sizes, ranging from 5 to 30μm, are seen in Silux Plus. Round filler of 0.1-0.2μm are included in the prepolymerized particle fillers and the resin matrix.

Heliomolar includes round and irregular shaped prepolymerized filler particles, sized 30μm and smaller than 10μm respectively. These prepolymerized filler particles and matrix consisted of round fillers, sized 0.05-0.1μm. In Palfique Estelite irregular shaped prepolymerized particle fillers of various sizes, less than 30μm (d_1) and round fillers of 0.05-0.1μm, are included in prepolymerized filler particles and the resin matrix.

Filler volume fraction, Vickers hardness, and fracture toughness is listed in Table 2. The filler fraction ranged from 25.7 vol% to 51.1 vol%. Metalfil shows the lowest filler content, next comes Silux Plus of 37.1 vol% and Heliomolar of 4.2 vol%. Palfilque Estelite shows the highest filler content.

In the hardness values, Palfique Estelite shows the highest hardness value, second is Silux Plus, next is Heliomolar, Metafil shows the lowest hardness value. The hardness value is influenced by filler volume fractions and the composition of fillers. As shown in Table 1, Metafil and silux Plus is mainly consisted of SiO_2 and small portion of Al_2O_3. In Heliomolar, YbF_3 is contained, which perhaps decrease the hardness, in spite of higher filler fraction. Palfique Estelite also shows the rather lower hardness value compared to that of Heliomolar, though it has higher filler content. Because it contains the filler consisting of Na and Zr. The fracture toughness showed values in the range of 0.03 to 0.85 MPa $m^{1/2}$. Palfique Estelite shows the highest fracture toughness of 0.85 MPa $m^{1/2}$, next is Heliomolar, third is Silux Plus, and Metafil has the lowest value of fracture toughness value. Generally, it is known that fracture toughness is an increasing function of filler content in dental composite resins. However, Kim et al. reported that in dental composite resins, fracture toughness increases until it reaches a certain limiting filler content, then the fracture toughness decreases with the increasing of filler content in dental composite resins(Kim et al., 1991). The composite resins examined here showed a volume fraction in the range of 25 to 51%, so the fracture toughness of the composite resins tested in this experiment was increased with increasing filler volume fraction (Fig. 3).

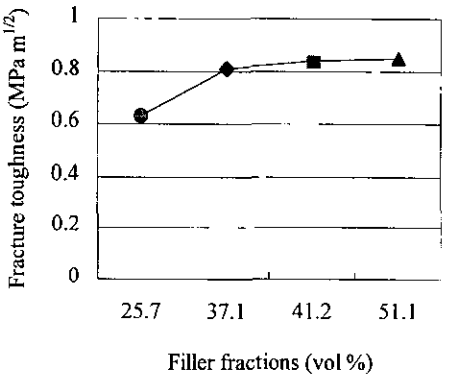
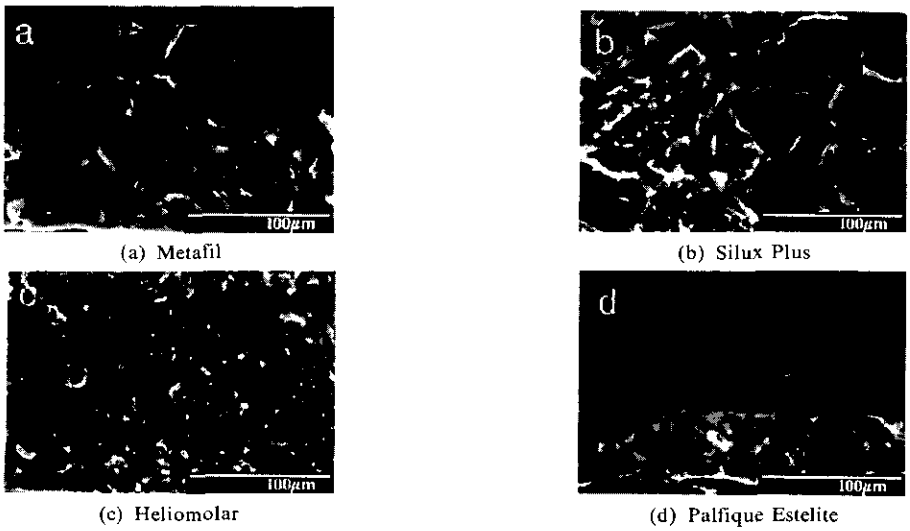


Fig. 3 Filler fractions relative to fracture toughnes of specimens ○Metafil; ◇Silux Plus □ Heliomolar; △Palfique Estelite

3.2 SEM observation of fracture surfaces

SEM fractographs of the fracture surfaces of composite resins containing prepolymerized filler particles are shown in Fig. 4. Metafil and Silux Plus reveal very rough fracture surfaces while Heliomolar and Palfique Estelite show rather flat fracture surfaces. In Metafil, cleavage planes sized 30–60μm were observed, and these cleavage planes were linked by the interface of irregular shape filler and matrix. A brittle fracture surface was also observed in Silux Plus. The cleavage



(a) Metafil (b) Silux Plus

(c) Heliomolar (d) Palfique Estelite

Fig. 4 Scanning electron micrograph of fractured surface of composite resin containing Pre-polymerized fillers

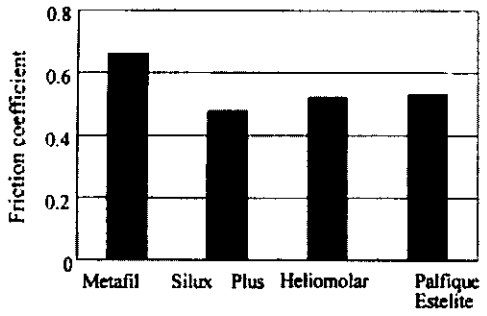


Fig. 5 Average friction coefficient of specimen under 9.8N

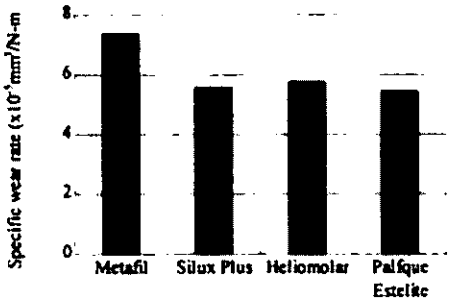


Fig. 6 Variation of specific wear rate for specimen under 9.8N

planes, sized 30μm, and rough fracture surface in the cleavage planes were observed. In Heliomolar, fracture units of 30μm were locally seen, and the greatest portion of the fracture surface was composed of the interface of fractured fillers and matrix. In Palfique Estelite, a different fracture surface, being very flat, was observed.

Palfique Estelite had smoother fracture surfaces than those of the above three composite resins. This indicates that the cracks went straight through the pre-polymerized particle filler.

3.3 Frictional and wear characteristics

Figure 5 presents the average friction coefficient of each specimen. The highest was Metafil at 0.67. This was because Metafil had the lowest hardness and fracture toughness, and, therefore, fretted heavily in the abrasive process with the Si_3N_4 ball, which had a much higher hardness. As a result, a higher wear volume was generated. In contrast, Silux Plus, Helimolar and Palfique Estlite had coefficients of 0.48, 0.52 and 0.53, respectively.

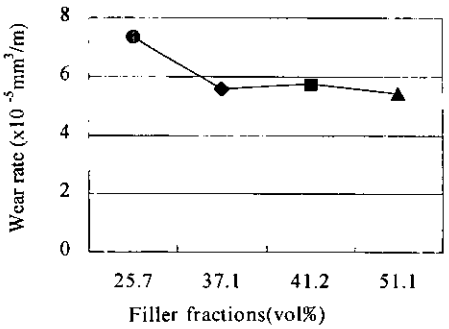


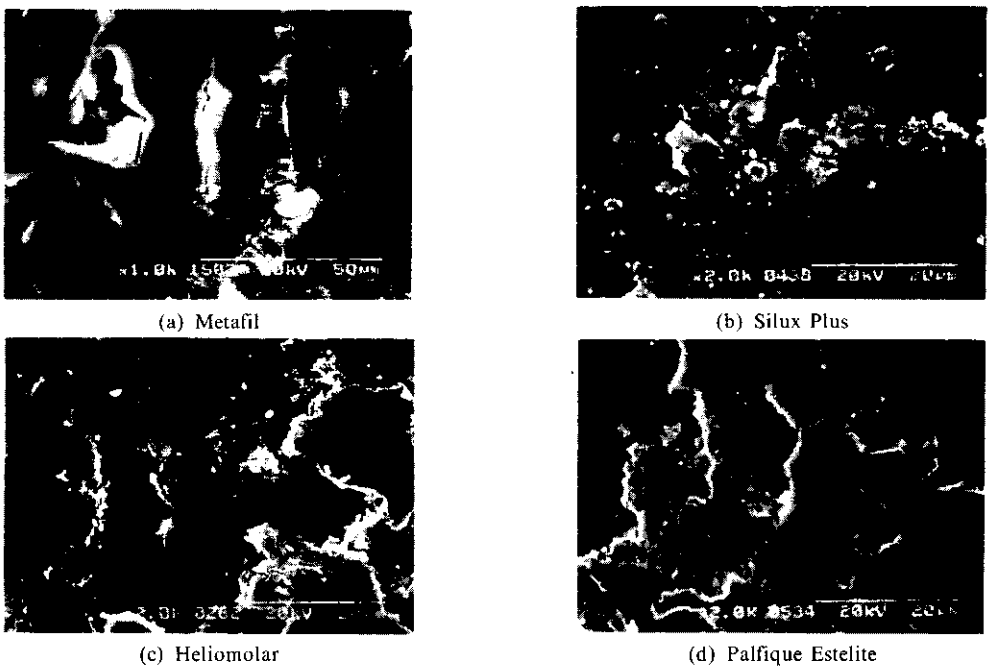
Fig. 7 Relationship of filler fractions realtove to wear rate of specimens ○Metafil; ◇Silux Plus □Heliomolar; △Palfique Estelite

Figure 6 presents the specific wear rates of the various specimens, in which Silux Plus and Palfique Estelite demonstrated a superior wear resistance. This indicates that the filler fraction, hardness, and fracture toughness have a significant effect on the wear resistance. A higher fracture toughness produced a higher wear resistance. Larger prepolymerized particle filler showed an easily fractured surface due to the cleavages generated at the interface between the filler, the prepolymerized particle fillers and the resin matrix. The wear volume was also increased. Fig. 7 shows the relationship of the filler fraction with the wear rate of the specimens.

3.4 SEM observations of worn surfaces

Figure 8 shows SEM photographs of wear specimen after 9000 cycles (sliding distance: 36 m). Figure 8(a) shows the worn surface of Metafil where there were many surface cracks basically in the sliding direction along with filler debris. This indicates that with regard to the abrasions, the worn surface stood the repetitive tensile strength and generated fracture cracks, the propagation of cracks and fracture of the prepolymerized particle filler. The weak interface strength dislodged the filler from the resin matrix and caused wearing.

Figure 8(b) shows the worn surface of Silux Plus. The filler separated from the matrix and the debris of prepolymerized particle filler were the main causes of the wear. This indicates that a good compatibility between the filler and the



(a) Metafil (b) Silux Plus

(c) Heliomolar (d) Palfique Estelite

Fig. 8 SEM photographs of worn surface for specimens

matrix, plus the prepolymerized particle filler and the matrix produces a superior fracture toughness, hardness, and wear resistance. The amount of filler debris was also decreased.

Figure 8(c) shows the worn surface of Heliomolar where the surface cracks were due to surface fracture, caused by damage to the pre-polymerized particle filler. The filler debris produced severe wearing. This indicates that the stress concentration at the interface between the matrix and the pre-polymerized particle filler under pressure and shear stress caused the dislodgment and impairment of the pre-polymerized particle filler.

Figure 8(d) shows the wear surface of Palfique Estelite. In this case, the laminated debris of the pre-polymerized particle filler caused severe wearing. This indicates that higher filler fractions produce superior compatibility, interfacial strength and fracture toughness between the matrix and the filler.

On the contacted surfaces, the repetitive tensile strength caused fracture, wearing, and the generation of debris from the pre-polymerized particle filler.

Accordingly, an increase in the 30~60μm spherical pre-polymerized particle filler and super micro filler improved the wear resistance of the composite resins containing pre-polymerized particle fillers by decreasing its friction coefficient and increasing its fracture toughness.

4. Conclusions

This study measured the fracture toughness and friction and wear of composite resins containing prepolymerized particle fillers. The purpose was to explain the effect of the shape, size, and filler-matrix interface strength of a prepolymerized particle filler on the fracture strength and wear resistance of a composite resin in dentistry. Each specimen was investigated using SEM, which helped to determine the micro fabric and worn surface in detail. The conclusions were as follows:

- Metafil, with the lowest filler fractions, had the highest friction coefficient, while Palfique Estelite, Helimolar and Silux Plus had relatively lower ones.
- Under the same experimental conditions, Palfique Estelite, and Silux Plus, with higher

filler fractions, exhibited a superior fracture toughness, Vickers hardness, and wear resistance.

• The pre-polymerized particle filler with a 30 ~60μm spherical shape was the most effective in improving the fracture toughness. Its increased density produced an increase in the fracture toughness.

• The microstructure of the specimen surfaces and microscopic observations of the worn surfaces showed that the surfaces had many surface cracks, and the wear particles of the fillers were generated by plastic deformation and micro-fractures. It was also found that the main wear mechanism was abrasive wear caused by the brittle fracture of the pre-polymerized particle fillers.

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