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SURFACE MODIFICATION OF ALUMINA-SILICA BY ADDITIVE AGENT USING SOL-GEL METHOD AS FILLER DENTAL COMPOSITE

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KEYWORDS

Dental composite
Chitosan
Polyethylene glycol
Sol gel method
Inorganic filler

ABSTRACT

Background. *Macro-sized and inhomogeneous distribution of ceramic filer particles make it difficult to obtain smooth surfaces of dental composite after polishing.* ***Objective.*** *This study aims to synthesize alumina silica (Al_2O_3-SiO_2) fillers using sol gel method with the additives agent chitosan 5%, 10% and polyethylene glycol (PEG) 5% to produce micro particle size and evenly distributed particle as filler dental composite.* ***Methods.*** *The type of research is descriptive explorative. The sol-gel process was utilized to synthesize filler particles of Al_2O_3-SiO_2. TEOS was dissolved in ethanol and mixed with acetic acid as catalyts. $Al_2(NO_3)_3$ was added into the solution and mixed homogenously. Subsequently, the additive agents (Chitosan 5%, chitosan 10% and PEG) were mixed into the mixture for 30 minutes. Drying the samples for 48 hours at 60°C in the oven. The Dynamic light scattering (DLS) SZ-1 was used to evaluate the size, particle distribution and zeta potential.* ***Result.*** *The results showed that the addition of chitosan 10% produced a smaller size of Al_2O_3-SiO_2 compared to other samples. A homogeneous particle distribution is shown in the sample with PEG 5%. Meanwhile, zeta potential values of the filler particles Al_2O_3-SiO_2 with the addition of chitosan 10% shows the biggest value.* ***Conclusion.*** *The additive agent of chitosan 10% can modify the surface of filler Al_2O_3-SiO_2 in order to inhibit particle growth more effectively but better particle distribution is shown in samples with PEG 5% due to the lower viscosity than chitosan thus it is easily homogenized in the solution*

INTRODUCTION

Since the 1960s, dental composites have received a lot of attention for the repair of decaying teeth, mostly because of their better biocompatibility and aesthetics.[1] These composites consist of a mixture of resin matrix and inorganic fillers.[2,3] The investigation of filler formulas and monomer structures has yielded significant results, however the primary reasons for repair failure in dental composites remain secondary caries and restorative fractures.[4–6] In other words, the functional and expected reinforcing effect for dental composites cannot be achieved by the fillers currently on the market.[1] Consequently, a lot of research was done to create fillers with a structure and functionality comparable to that of actual human teeth.[7]

Fillers usually constitute about 78–85% weight percent of resin dental composites. They are basically come in three types: conventional, microfine and hybrid. Conventional or macrofillers are amorphous particles that range in size from 0.1 to 100 μm and are produced by grinding or crushing bigger fragments. Ceramics, metal oxides, heavy metal glasses (such as Sr or Ba glasses), and quartz are the materials used as traditional fillers. It is challenging to polish the resin to a smooth surface in dental composites due to the macrofillers and uneven distribution of particle size. It will make the restoration more vulnerable to stain buildup and plaque.[8]

The most common types of microfine fillers are pyrogenic silica (0.01 μm - 0.1 μm) and colloidal silica, which are both chemically manufactured. The optical characteristics of composite resins are enhanced by microfine filler particles. Aluminosilicate glasses, barium silicate glasses, barium borosilicate glasses, strontium borosilicate glasses, zirconium/silica (0.5 μm–10 μm), and pyrogenic silica (0.01 μm – 0.1 μm) are examples of glass-ceramic filler particles that are hybrid fillers. These days, researchers are creating fillers with very fine diameters in order to enhance the mechanical performance, optical qualities, and finishing of dental composites.[8–13]

The industry uses conventional ways to generate glass fillers, but these procedures are not environmentally friendly because they require high temperatures for synthesis, extended grinding times to produce silicate particles, and significant water and HCl usage.[8] Since the synthesis may be done at a low temperature, the sol-gel approach is appealing and uses less energy. Furthermore, during synthesis, this approach allows for the regulation of stoichiometry and molecular homogeneity of precursors. Controlling the polymerization reactions, condensation, hydrolysis, and additional of additive agents are all possible ways to modify the homogeneity of particles size. The homogeneity will result in the production of filler particles that are small and evenly distributed.[14]

Additive agents such as chitosan and polyethylene glycol are known to be used to control the growth of particles of metal oxide compounds via sol-gel methods.[15,16] Faza et al observed chitosan 0-20% as an additive to control the growth of ZrO_2-Al_2O_3 and SiO_2 particles. Chitosan 10% was known to produce the best particle distribution.[15] Meanwhile, Gorbunova et al. used PEGs 5% to control the growth of silica particles.[16]

The study aims to synthesize alumina silica (Al_2O_3-SiO_2) fillers using sol gel method with the addition of additives agents chitosan 5%, 10% and polyethylene glycol (PEG) 5% to produce micro particle size and evenly distributed particle as filler dental composite. In this investigation, a particle size analyzer will be used to assess the size and distribution of the filler particles. Zeta potential of the particles is also observed.

METHODS

The type of research is descriptive explorative. The research was conducted in August-November 2023 in the integrated laboratory of the Dental Faculty and Finder CoE laboratory Universitas Padjadjaran. Aluminum nitrate ($Al_2(NO_3)_3$) and Tetraethyl Orthosilicate (TEOS) were used as the precursor to produce Al_2O_3-SiO_2. Chitosan and PEG were purchased from the local chemical store.

This study had three groups of samples, each group replicated three times (n=3). Groups 1 and 2 used 5% and 10% chitosan additives, while group 3 used 5% PEG additive. The sol-gel process was used to synthesize filler particles of Al_2O_3-SiO_2. After dissolving TEOS in ethanol, agitate for half an hour. Acetic acid was used to bring the pH down to 4, add $Al_2(NO_3)_3$, and stir for one additional hour. Then, add the additive agent (chitosan 5%, chitosan 10% and PEG 5%) to the mixture and mixed for 30 minutes. Drying the sample for 48 hours at 60°C in the oven. Later, Al_2O_3-SiO_2 filler particle was obtained.

The size, particle distribution and zeta potential are evaluated using Dynamic light scattering (DLS) SZ-100

RESULTS

Figure 1 shows the size and distribution graph of the particles of the filler Al_2O_3-SiO_2 with the addition of PEG 5%, chitosan 5% and 10%. Filler particles with PEG 5% produce the particle size of 1546 nm and monodispersed particle. Filler particles with the addition of chitosan 5% produce the largest particle size, 2892 nm. The particle distribution in this group is seen as polydispersed with a long distance between the peaks on the graph. The filler particles with 10% chitosan appear to produce the smallest size , 803 nm. A picture of the particle distribution in this group appears to be polydispersed with a close distance between two peak on the graph.

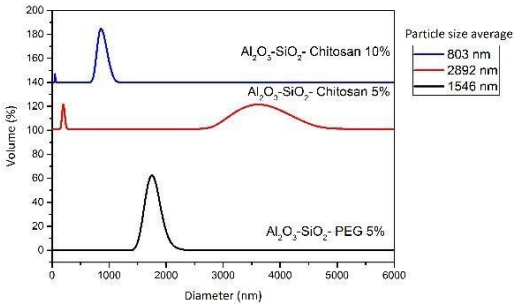

Figure 1. Size and distribution of the particles of the filler Al_2O_3-SiO_2 with the addition of PEG 5%, Chitosan 5% and 10%.

Figure 2 shows the zeta potential values of the filler particles Al_2O_3-SiO_2 with PEG 5%, chitosan 5%, and 10%. They show the charge values in sequence of 19, 17 and 29 mV.

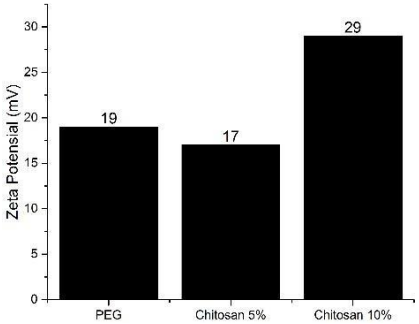

Figure 2. zeta potential values of the filler particles Al_2O_3-SiO_2 with the addition of PEG 5%, chitosan 5%, and 10%.

DISCUSSION

DLS is the most popular technology in determining the size of particles by utilizing the time variation of light dispersed on the particles within the suspension through brownian motion to obtain the distribution of the hydrodynamic size of the particle. DLS measures the diffusion of the group of particles when the concentration increases and/or for relatively smaller particles.[17,18] In Figure 1, the smallest particle size of Al_2O_3-SiO_2 is characterized using DLS at 803 nm and the largest at 1546 nm. DLS is also used in determining the polydispersity index (PI) to measure the homogeneity of the particle size through distribution graphic.[19]

In sol-gel techniques, hydrolysis and condensation reactions begin immediately after the catalyst (acetic acid) is added to the solution. At this stage, the atoms starts to form and then the aggregation reaches the colloidal dimension until it produces sol. At this stage, additive agent plays a role in preventing larger particle growth and in homogenizing the growth of particles in the solution through the hydroxyl groups found in the additive compound.[20,21]

Figure 1 shows the size of the particle Al_2O_3-SiO_2 with the addition of 10% chitosan is smaller than the other group. However, the distribution of particles is not so homogeneous. Different things are seen on the sample with the addition of PEG.The particle size appears to be larger but the particle-size distribution is more homogeneous (monodispersed). This is supposed to be due to a lower PEG viscosity that facilitates the homogeneity of the solution with additives.[22] Meanwhile, chitosan with a higher viscosities requires longer mixing time to be perfectly homogenous in the solution.[23]

The electrical charge on the surface of a particle is generally known through the determination of electrical potential that is located far from the surface of particles. This layer relates to the movement of a particle in a fluid and is known as a sliding or slipping field. The electrical potential measured in this field is called a zeta potential which is a very important parameter for a colloid or a fine particle within a suspension. This value is closely related to the stability of the suspension or the morphology of the particle surface.[24]

Based on the DLVO theory that the greater the zeta potential value, the more stable a particle will be in the suspension. The theory states that the zeta potential values are grouped as follows: highly unstable (±0-10 MV), relatively stable (±10-20 mV), moderate stability (±20-30mV) and high stability (21,22).[18] The results of the study, in Figure 2, show the greatest zeta potential values in sampel with the addition of 10% chitosan. The predominant deposits of amino groups (cations) on the outer surface of the particle, which do not bind to the particles, make the zeta potential charge positive and quite effective in inhibiting particle growth.[19] This is consistent with the results in Figure 1.

Chitosan 10% is able to inhibit particle growth better than chitosan 5% and PEG 5%.

CONCLUSION

The additive agent of chitosan 10% can modify the surface of filler Al_2O_3-SiO_2 in order to inhibit particle growth more effectively but better particle distribution is shown in samples with PEG 5% due to the lower viscosity than chitosan thus it is easily homogenized in the solution.

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