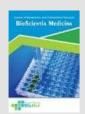
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Utilization of Silica Nanoparticles from Rice Husks for Improving the Mechanical

Properties of Dental Materials: A Literature Review

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ABSTRACT

Rice husk is a waste product whose utilization is not optimal in Indonesia. Rice husks contain organic materials in the form of cellulose, hemicellulose, and lignin; and minerals in the form of silica, alkali, and trace elements. The silica content in rice husks is very high, ranging from 87-97% to other minerals. Silica nanoparticles from rice husks were synthesized through various methods, including the sol-gel method, the combustion method, and the combination method, to obtain the purity and size of the silica particles up to the manometer scale. Mixing silica nanoparticles from rice husk to dental material at a certain concentration is effective in producing better mechanical properties. Silica nanoparticles synthesized from rice husks have the potential as fillers and reinforcements that can improve the mechanical properties of dental materials. Besides that, the price is more economical because the raw materials come from Indonesian natural waste. This literature review aims to describe the method of synthesizing silica nanoparticles from rice husks and their effectiveness in improving the mechanical properties of dental materials.

1. Introduction

The discovery of silica in various biological systems has been known previously and has been investigated¹, especially the biomineralization of silica in the form of diatoms and unicellular photosynthetic organisms, which show the formation of several structures of silica nanoparticles. Another example of silica biomineralization is the formation of silica cell walls in rice husks.² Although the silica structure of rice husk looks simpler and unattractive compared to its diatomic structure, the large quantity of rice husk biomass is capable of producing structural silica nanoparticles for industrial applications.³ Rice husk has low nutrient properties and high ash content, but so far, the use of rice husk in Indonesia is still limited. In some countries, discarded rice husks cause additional costs and environmental pollution problems. Isolating silica from rice husks not only benefits from the biomass value of rice husks but also minimizes environmental problems associated with rice husk waste.⁴

Rice husk is the outermost part of rice that will be separated through the milling process, producing about 20-22% of the total weight of rice⁵, containing 65-75 organic materials including lignin, cellulose, hemicellulose and 15-20% inorganic materials such as SiO₂, alkalis, and trace elements.^{6,7} Rice husks have been investigated in various applications for bio-fuel production, heating in boilers, power generation, and blending in animal feed.8 Rice husk production in Indonesia is abundant, but its utilization is not optimal. If it is burned, it can cause air pollution. The chemical composition of rice husks varies in various regions, influenced by climate and geographical conditions where rice is produced. The silica content in rice husk is about 87-97% dry weight in addition to other minerals after undergoing complete combustion.⁹ The high silica content of rice husk is of great interest among researchers because of its potential to produce various silicon materials, including silicon carbide, silica, silica nitride, silicon tetrachloride, pure silicon, and zeolites.¹⁰

Silica nanoparticles in various fields of science are highly developed due to their superiority in improving mechanical properties. The first research to isolate silica nanoparticles from rice husk was synthesized in 2012 by using the HCl treatment method and through the pyrolysis process, which resulted in particle size of 25-30 nm.³ In the next study, silica nanoparticles were synthesized using the high-energy ball-milling method at room temperature, capable of producing a particle size of 70 nm.11 In addition, the sol-gel method has also been used to synthesize silica nanoparticles with an average particle size of 3 nm through the addition of sodium hydroxide solution to produce silicate solutions, and silica precipitation was obtained by sulfuric acid droplets.¹² The high-pressure method using an autoclave to produce silica nanoparticles from rice husks is able to reach nano sizes of 10-30 nm with the addition of hydrochloric acid.13 In addition, the use of the fungus Fusarium oxysporum to synthesize silica nanoparticles from rice husks has been successfully carried out with particle sizes of 2 to 8 nm¹⁴ and another method using internal circulation rotating packed beds.15

The advantage of silica nanoparticles is that they have a high surface area (porosity and small particle size), so they are used in other chemical applications such as absorption, insulation, and catalysis.¹⁶ In addition, porous silica is also used in biomedical applications, namely biosensors, and is applied in the field of dentistry as a filler material.¹⁷⁻²⁰ Utilization of silica nanoparticles from husks synthesized by several methods developed in the field of dentistry, such as their effectiveness as fillers and reinforcements in composite resins, acrylic resins, and ceramics, obtained satisfactory results for improving mechanical properties.¹⁹⁻²³ This literature review aims to describe the method of synthesizing silica nanoparticles from rice husks and their benefits in improving the mechanical properties of dental materials.

Synthesis of silica nanoparticles from rice husk

Silica or silicon dioxide (SiO₂) is a chemical compound formed from the reaction of silicon and oxygen atoms. Silicon and oxygen are the most abundant elements found on earth in the form of minerals, vegetables, and crystal synthesis. Mineral silica is found in mining materials in the form of quartz sand, granite, and feldspar, which contain silica crystals. Vegetable silica has an amorphous structure found in rice husks, bamboo, and corn cobs. The vegetable silica commonly used today is silica from rice husk.²⁴

Silica nanoparticles synthesized from rice husks have several advantages compared to mineral silica, including having finer grains and being more reactive. The synthesis procedure is simpler and relatively inexpensive because rice husk raw materials are widely available in nature.²⁵ In general, the synthesis of silica nanoparticles is classified into three methods, including the chemical method, combustion method, and combination method.⁴

Chemical method

Silica nanoparticles synthesized from rice husks can be carried out chemically using strong oxidizing agents such as hydrogen peroxide, sulfuric acid, acetic acid, and hydrochloric acid²⁶, which is referred to as the sol-gel method. These acids are generally used to remove metals from rice husks, but these substances are quite harmful to the environment and humans. Pure chemical methods are rarely used because the process of removing impurities usually takes a relatively long time, so it becomes less economical than the combustion method. Chemical processes are energy intensive and require high temperatures and pressures, as well as strong acids that can have an effect on the environment. Taking into account the increasing demand for silica particles at the nanometer scale, an alternative approach that is economical and environmentally friendly is needed.^{4,27}

The sol-gel process involves hydrolysis, condensation, and hot decomposition of the metal solution precursors. The precursor dissolves the rice husk to form a stable solution called a sol, and then the sol undergoes hydrolysis and condensation to form a gel.28 Water, alcohol, acid, or base can be used to control the reaction kinetics. Changes in the concentration, temperature, and pH of the precursor will cause a change in particle size. The gel will experience maturation in some time, and the gel will be processed at high temperatures to decompose organic compounds to produce nanoparticles.28 The sol-gel method is the process of changing the sol into a gel through the stages of hydrolysis, gelation, aging, and drying.^{11,29,30} The sol-gel method is widely used by various researchers in the field of Dentistry because the procedure is simpler and can produce pure silica nanoparticles.28,31,32

Methods of burning

Rice husks are usually burned in a heating furnace with a temperature of 500-800°C and a controlled time, produces ash in the range of 20%, and more than 90% contains silica. A well-controlled temperature and a suitable environment during combustion can produce a better quality rice husk ash because the particle size and specific surface area are affected by the combustion conditions. Completely burnt rice husks will be ash to white in color, while rice husk ash that is not completely burned will be black.^{4,33}

Combination method

The chemical method using acid is used as a pretreatment of rice husk to remove impurities and help produce high-purity silica.²⁹ These inorganic compounds must be removed before combustion or sonication because they can inhibit the formation of silica which has an amorphous structure. Some of the combined methods include the sol-gel method with sonication, called sonochemistry, and the sol-gel method is usually carried out to obtain a smaller particle size of silica nanoparticles by means of vibration and variations in sonication time. The procedure is simpler and environmentally friendly because it uses distilled water as a solvent.³⁵

Benefits of silica nanoparticles from rice husks in dental materials

Various studies have been conducted to examine the potential of silica nanoparticles from rice husks to improve mechanical properties, such as flexural strength, compression, shear, and hardness in dental materials.

Composite resin restorative materials

Zulkifly et al. synthesized silica nanoparticles from husks using the sol-gel method using 1M H₃PO₄ and calcined at a temperature of 550°C. The characterization test with SEM and TEM showed the shape of mesoporous silica nanoparticles and agglomerated, with an average size of 2-50 nm silica particles. XRD results show that the sharpest peak of amorphous silica is at 2 theta = 22°. The FTIR results show the presence of silica at the peak with a wave number range of 4000–400 cm⁻¹, while silica with the most Si–O–Si chains has the sharpest peak at 1107 cm^{-1,31}

Research Noushad et al. synthesized silica nanoparticles from rice husks using the sol-gel method using 10% NaOH and calcined at a temperature of 500°C. The characterization test with the Particle Size Analyzer (PSA) showed a particle size of 48-534 nm with an average particle size of 261 nm. The results of the Fourier Transform InfraRed (FTIR) showed that the nanohybrid particles synthesized from rice husks showed a silica structure with the sharpest peaks at wave numbers 1084 and 1111 cm⁻¹. The results of the transmission electron microscope (TEM) show the size distribution of the spherical nanohybrid silica particles. The results showed that the higher the ratio of filler concentration of silica nanoparticles (40 and 50 wt%) of rice husk to the matrix resulted in an increase in compressive strength (181.4 and 190.6 MPa), flexural (92.4 and 106.6 MPa) and hardness (33.44). and 38.66 VHN) composite resin. Thus, silica nanoparticles from rice husks can be used as fillers that meet the requirements of restorative materials in anterior and posterior teeth.²¹

The research of Yusoff et al. also synthesized silica nanoparticles from rice husks using the sol-gel method and calcination at a temperature of 500°C. The PSA test produced a particle size of 48-448 nm with an average value of 218 nm. Furthermore, the manufacture of flowable silica nanoparticle composite resin from rice husks with various filler concentrations of 40 and 50% on the matrix. The SEM results showed that all surface textures were homogeneous and smooth, with a surface roughness of $<0.2 \mu m$, which indicated that there was no increase in the accumulation of bacteria on the surface of the resin material. Surface roughness is one of the factors that affect the aesthetic value, wear resistance, and quality of composite resin materials. The smooth flowable composite resin surface provides patient comfort. The results of this study showed that there was no significant difference in surface roughness between the flowable silica nanoparticles composite resin from rice husks and commercial ones (p<0.05). The hardness test results showed the hardness value of flowable silica nanoparticles from rice husk 29-31 HV. This indicated that there was no significant difference with commercial flowable (p<0.05), as well as the flexural value and compression modulus. The results of this study indicate that silica nanoparticles from rice husks have good physical and mechanical strength, so they can be used as fillers in flowable composite resins.³⁶

Acids such as HCl, HF, HNO₃, and H₃PO₄ which are used in the synthesis of silica nanoparticles, are strong oxidizing agents whose role is to break the chain bonds between sodium and silica to produce white pure silica precipitates.^{31,37,38}

Acrylic resin base material

Research by Wongwitthayakool et al. synthesized silica from rice husks by acid treatment and calcination at a temperature of 700°C. XRF results showed that the chemical composition of silica compounds from rice husk ash was the highest compared to other compounds (93.8%), and XRD results showed a pattern of crystallinity and amorphous silica form at the peak of $2\theta = 21.7^{\circ}$. Mechanical test results after mixing silica from rice husk ash with a concentration variation of 0; 0.25; 0.5; and 1% of Poly Methyl Methacrylate Acrylic (PMMA) acrylic resin base powder, including the most optimal base hardness value with the addition of 0.5% silica; and the most optimal value of flexural strength in the addition of 0.25%. Rice husk is potentially effective as a source of silica that can improve the mechanical properties of PMMA base material.23

Ceramic fixed partial dentures

Research by Santos et al. mixed silica powder with lithium silicate ceramic powder in a balanced ratio of SiO₂ commercial, SiO₂ from rice husk, and LiO₂ by 33% to produce lithium silicate ceramic powder at burning 1550°C for 4 hours. The XRD results show the sharpness of the crystallinity peaks that are almost the same between silica from rice husks and commercial silica. Scanning Electron Microscope (SEM) results show that there is no morphological difference between silica from rice husk and commercial silica. Changes in silica morphology from rod shape to needle shape after burning ceramic material showed an increase in ceramic fracture resistance. The results of this study stated that silica from rice husks could be recommended as a filler material for ceramic denture materials based on lithium disilicate, which can increase fracture resistance.²² In addition, a study conducted by Zain et al. synthesized silica nanoparticles from rice husks by mixing them with commercial opaque porcelain powder with a mixing variation of 0.25%; 0.5%; 0.75%, and 1% obtained the results that mixing silica nanoparticles 1% resulted in the most significant flexural strength (p<0.05) in ceramic metal samples.³⁹

Rice husk is a waste product whose use is not optimal in Indonesia. The growing interest of researchers in synthesizing silica nanoparticles from rice husks in the field of dentistry will increase the value of rice husks into useful products.

2. Conclusion

Silica nanoparticles synthesized from rice husks have the potential as fillers and reinforcements that can improve the mechanical properties of dental materials.

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