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Research Paper

Silica from Natural Sources: a Review on the Extraction and Potential Application as a Supporting Photocatalytic Material for Antibacterial Activity

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Abstract

Silica has become a popular material due to its high abundance and many advantages in various fields. This material can be produced synthetically and extracted from nature with resultant advantages in the application of green production. Therefore, this article deals with the form of silica extracted from quartz sand, leaves, and agricultural wastes found in nature. The extraction process from various sources would be described using thermal, biological, and chemical methods. This review also highlights the potential application of silica as a photocatalytic antibacterial-supporting material and discusses its role in increasing the effectiveness of the process. The discussion was continued with research on this procedure, where synthetic auxiliary materials were compared to the extracted silica. Furthermore, results obtained indicated that the extracted material had very good potential as a photocatalyst adjunct in its application in the antibacterial field.

Keywords

Antibacterial, Extraction Method, Silica, Support Material, Photocatalyst

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1. INTRODUCTION

Silica (SiO_2) which is a constituent of silica sand and mining products, is the most occuring oxide on the planet (Lutgens and Tarbuck, 2000) after oxygen in terms of its composition (Matichenkov and Calvert, 2002). The formation of silica sand is due to the weathering process of rocks containing quartz and feldspar (Bernstein and Carpi, 2015). In addition, silicones can be produced in amorphous and crystalline forms. The center of this structure is developed from atoms containing four oxygen fused to a tetrahedral angle site close to the silicon molecule by covalent bonds (Bernstein and Carpi, 2015; Julia, 2002). Consequently, this tetrahedral structure bonds with each other and forms a large silica matrix (Salh, 2011).

The bond angle around O-Si-O is 109.5°, which is a tetrahedral angle and varies in length between 1.54-1.69 Å. Meanwhile, the oxygen bridge (Si-O-Si) gives silica its unique properties resulting in a very wide industrial application due to the different shapes and compositions of silica (Rayner-Canham and T., 2015).

Most silica is currently obtained from the extraction of natural materials such as waste from agriculture which is usually disposed of or burned without prior treatment and can cause pollution (Ferronato and Torretta, 2019). The large amount of waste produced each year from harvesting due to the increase of agricultural products to fulfill human needs is cause for great concern. Therefore, to reduce this problem, researchers have worked to put a use value on agricultural waste. This is widely used as a source of silica extract for further application (Rayner-Canham and T., 2015; Liang et al., 2020). The increase in waste is influenced by increased production of agricultural materials. FAO statistics show a significant increase in agricultural products each year.

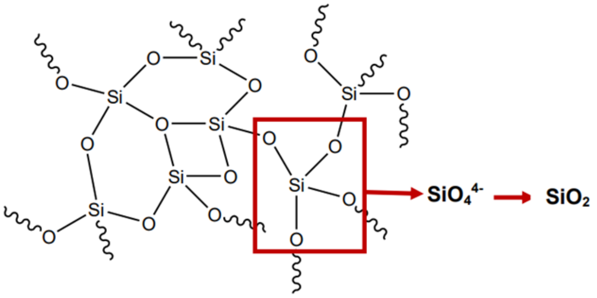

Figure 1. Structure of Bonded SiO_4 to Form SiO_2

There have been many discussions regarding the function-

Table 1. Quantity of Agricultural Production in the World (2016-2019) (FAOSTAT, 2021)

Item	Production quantity (Million Ton)			
	2016	2017	2018	2019
Cassava	290.65	286.71	295.05	303.57
Coconut	58.46	57.43	63.76	62.45
Maize	1,127.35	1,138.65	1,124.72	1,148.49
Maize, green	11.46	11.41	9.01	8.3
oil palm	330.03	400.29	403.58	410.7
Olive	20.03	21.05	21.88	19.46
Rice, paddy	739.52	751.73	762.84	755.47
Sugar cane	1,881.08	1,835.46	1,930.51	1,949.31

alization of silica, especially in the field of renewable energy. In the industrial world, they are found in tires, rubber, glass, cement, concrete, ceramics, textiles, paper, cosmetics, electronics, paints, films, toothpaste, health, and other industries are widely used (Setyoningrum et al., 2020). This review article will discuss more intensely the function of silica as a support material in the field of photocatalysis which has been proven to improve its performance and potential as antibacterial agents. Furthermore, this article discusses extraction methods and sources of silica from waste and its characteristics as a support material. The discussion functions to improve the study of silica sources and their use as a support material. Also, it summarizes the optimal results from the research on silica producers obtained from the nature of fabrication and its relevance in various fields.

2. RESULTS AND DISCUSSION

2.1 Source of Silica

Silica can be manufactured synthetically and extracted from the abundant amounts found in nature. This compound is also a part of the prospective minerals selected for development and application in various industrial fields (Kim et al., 2016; Lee and Yoo, 2016; Elma et al., 2020). Various studies have been conducted using natural materials, such as beach sand and agricultural waste to produce high purity silica extracts. This beach sand has a more dominant silica concentration than other oxides (Ismail et al., 2020; Eddy et al., 2015; Okereafor et al., 2020; Rattanaudom et al., 2020). Several studies have shown the results of high purity silica extraction in different sand extraction areas.

2.1.1 Sand

Mining products, including silica sand, are widely used in various industries, and this material is commonly known as quartz or white sand is usually found in the lowlands. It is produced from the weathering process of rocks which contain the main minerals, such as quartz and feldspar (Bernstein and Carpi, 2015). The availability of silica sand in Indonesia is very abundant (Ishmah et al., 2020), reaching 55.30-99.87 % in 2017. Therefore, this provides benefits from various sectors in increasing its use-value (Sumarno, 2015). This material is not found in pure form but as a mixture, hence necessitating an extraction process first to obtain pure silica. Extraction is a process of separating substances from the mixture (Eddy et al., 2015). This substance has various uses in the industrial sector and usually acts as a doping or composite support or additional raw material for the manufacture of various materials. These include cement, ceramics, tiles, cast/precast, paint, cosmetics, refractory bricks, petroleum or mining, hardener in the rubber industry, and others (Madina et al., 2017).

Ishmah, 2019 succeeded in extracting silica from the sand on the beaches of Bengkulu, Indonesia. This research produced amorphous silica with a very high purity of 97.3 % and was applied as a supporting photocatalyst material to remediate phenol waste. Setyoningrum et al., 2020 also succeeded in adding to the economic importance of determining silica based on its purity and the extract used was obtained from the Kokap area, Kulonprogo. According to the experimental results, the particle size reduced with an increased conversion while the increase in the amount of extracted silica was directly proportional to the NaOH concentration. Consequently, silica gel was the product generated from this research.

2.1.2 Agricultural Waste

- Rice Husk

Silica exists as nanoparticles and is the major inorganic constituent of rice husks (85- 95 %) which is a derivative obtained from rice milling and husk ash through burning in a distinct boiler (Hossain et al., 2018). The ash form is an agricultural waste, hence various studies including the extraction of trapped silica, have been conducted to promote its economic value.

Klankaw et al., 2012 extracted SiO_2 from rice husks coated together with TiO_2 on a thin film, which was prepared on a sliding glass by using the dipping method. The best function for the photocatalytic decolorization of MB dye with a total efficiency of 81 % was delivered by a thin film that consists of SiO_2:TiO_2, with values of 20:80. Furthermore, biogenic SiO_2 with high purity and surface area have been previously extracted from rice husks to prepare SnO_2/SiO_2 composites. The resulting silica has an amorphous form and is obtained from the acid pathway method, where inefficient SiO_2 extraction produces high purity biogenic SiO_2 on the nanometer scale (Ferreira et al., 2015).

The chemical method for synthesizing amorphous silica nanoparticles from burnt rice straw was conducted by hydrolysis with acid-base treatment. In this case, the resulted silica particles ranged in size from 60–90nm and obtained a total yield of 76.43 % through physiological observations (Uda et al., 2021). Furthermore, research involving the extraction of silica has been conducted through an environmentally friendly chemical treatment approach and those with a yield of 93.08 ± 0.11 % were extracted from RHA. The characterization studies indicated that the difference between the commercial-grade silica (19.49 ± 13.03 nm) and mean particle size of the extracted SiO_2-NPs (17.71 ± 7.53 nm) was insignificant (Nayak and Datta, 2021). According to Motlagh et al., 2020, extract silica and activated carbon simultaneously formed the two value-added chemical products from rice waste, where the results indicated that more silica was obtained from the straw (83 %) than from the husks (66 %).

- Fly Ash Waste

Fly ash is an intricate derivative obtained from burning a variety of mineral coals (Yao et al., 2015), with an SiO_2 content of 45–60 % (Wang et al., 2020). This material is considered to be less dense than cement and to contain spherical vitreous particles with sizes ranging from 8 to 20 mm. Besides, the synthesis of mesoporous silica, fly ash has attracted attention because of the characteristics of its resulting material (Miricioiu and Niculescu, 2020; Mehmood et al., 2017) which is rich in metal oxides, mainly silica. This substance can harm the environment Estevez et al., 2009 by interacting with water and soil which causes groundwater pollution with heavy metals, such as Cr, V, Ni, Cd, and Pb (Miricioiu and Niculescu, 2020). In addition, silica-rich fly ash acted as renewable source material for silica synthesis (Yadav et al., 2020).

Furthermore, nanoporous substances, such as zeolite or mesoporous silica are produced from fly ash which is used as a possible precursor due to its high silica content. These mesoporous substances, such as MCM-41 or SBA-15, are obtained from materials with a great deal of silica content and used to remove or capture CO_2 from emissions or for wastewater treatment (Miricioiu and Niculescu, 2020).

Research conducted by Yadav et al., 2020 used a simple, efficient, and cost-effective alkaline fusion process for the production of nano-silica which measured 10 to 60 nm in spherical and aggregate shapes from fly ash-based tiles. Several studies aimed to minimize the danger of environmental ash stockpiles by reusing fly ash from biomass power plants to produce silica material. According to a previous study, the synthesized amorphous silica was successful with a purity of 44.41 % to 93.63 % and a yield of 20.45 %. The size of the agglomerate particles ranged between 380.9 nm to 178.8 nm, when the ash was converted into spherical silica (Liang et al., 2020).

- Cassava Waste

Multiple studies have been performed to increase the economic value of cassava waste because the result obtained is usually disposed of carelessly or burned (Adebisi et al., 2018; Adebisi et al., 2019; Farirai et al., 2021). Studies on the amorphous silica nanoparticles from the cassava periderm were successfully conducted using the modified sol-gel method. These results showed that the ethylene glycol-modified silica was less agglomerated with a higher yield and a lower particle size. In this study, silica nanoparticles were used as a precursor for the synthesis of silicon nanoparticles (Adebisi et al., 2017).

- Palm Ash Waste

Also, oil palm ash and other agricultural wastes are used in the synthesis of silica (SiO_2) as a renewable source of energy (Imoisili et al., 2020; Razak et al., 2019; Faizul et al., 2014; Faizul et al., 2013; Pa et al., 2016). The result, in this research, showed that silica with a purity of more than 90 % could be extracted from palm ash (Pa et al., 2016).

- Palm shells

Silica nanoparticles obtained through the use of the modified sol-gel extraction technique have been successfully formed from palm shell ash. Furthermore, the microstructural analysis showed that the unit size of the extracted materials was between 50–98 nm, with a very high specific surface area of 438 m^2g^{-1} (Imoisili et al., 2020).

- Coconut Husk Ash

Generally, coconut husks are disposed of as waste material, either by combustion or in a waste disposal site while its fruit is used globally as a source of nutrition, drink, or other products. Therefore, this material causes various environmental and health problems without proper processing or disposal methods (Anuar et al., 2020).

Research has been conducted to explain the synthesis, optical and physical properties of silica obtained from coconut coir waste, and its possible use in optical applications. Furthermore, prior to the utilization of the green synthesis method in the extraction of silica from ash after treatment with sulfuric acid, this material was burned at a temperature between 500– 700 ^{o}C to produce coconut coir ash (CHA). Subsequently, the weight of the coconut husk particle size distribution was reduced from 200–750 nm to 200–410 nm at temperatures ranging between 221–360 ^{o}C (Anuar et al., 2020).

- Olive Seed

Investigation of silica extract from olive seeds using the alkaline leaching extraction method conducted by Naddaf et al., 2020 was successful. According to the results, the extracted powder consisted of 15-68 nm porous nanoscale silica with several hundreds of nanometer-sized particles. The resulting material which has been used for biological applications was amorphous silica which turned into a crystalline phase known as cristobalite, after sintering at 900 ^{o}C.

Silica extraction from natural materials and agricultural waste byproducts are much in demand because it provides advantages compared to other conventional methods, the advantages of this method include simple, lower costs, higher safety margins and lack of pollution produced during the synthesis process (Mor et al., 2017).

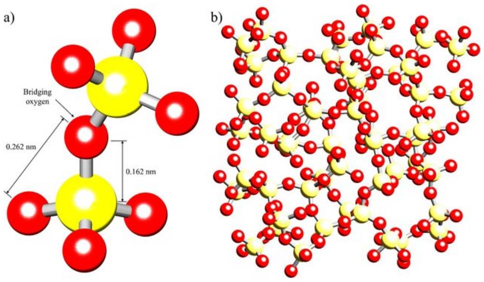

Figure 2. Atomic structure of amorphous silica. (a) Schematic representation of two tetrahedra showing nominal distances between neighbouring Si-O and O-O atoms, and a bridging oxygen between two neighbouring tetrahedra. (b) Subsection of an amorphous silica MD simulation showing a random arrangement of silica tetrahedra, Adapted from Lunt et al., 2018

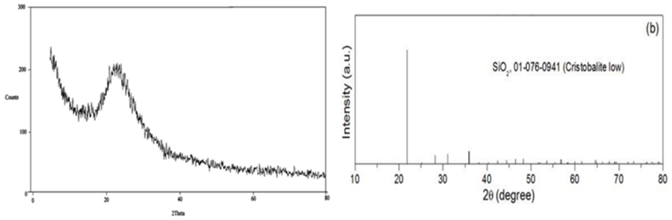

Figure 3. X-ray diffraction (XRD) spectra of SiO2, (a) Amorphous Adapted from Waseem et al., 2009; (b) Crystallite, Adapted from Joni et al., 2018

2.2 Characteristic of Silica

Silicon dioxide or silica (Figure 2) is the most abundant oxide compound form of silicon on earth with the chemical formula SiO_2. The center of this structure is developed from atoms containing four oxygen fused to a tetrahedral angle site close to the silicon molecule by covalent bonds (Bernstein and Carpi, 2015; Julia, 2002). Consequently, this tetrahedral structure bonds with each other and forms a large silica matrix (Salh, 2011).

Silica material has a pore size between 5-3000 Å and can either be amorphous or quartz, which is more stable as shown in Figure 3. The bond angle around O-Si-O is $109.5\ ^o$ which is a tetrahedral angle and varies in length between 1.54-1.69 Å. The angle of the Si-O-Si (siloxane) bond varies between 120-180^o which is influenced by changes in bond energy, hence allowing it to rotate freely and easily and to form amorphous or irregular structures (Julia, 2002; Yao et al., 2015; Sun et al., 2017).

This oxygen bridge (Si-O-Si) and its different shapes and compositions give silicon dioxide its unique properties. In addition, the industrial application of silica is very wide (Rayner-Canham and T., 2015) due to these physical properties (Haynes, 2011):

Chemical formula : SiO_2
Molar mass : 60.08 g/mol
Density : 2.648 (α-quartz), 2.196 (amorphous) $g{\cdot}cm^{-3}$
Melting point : 1,713 ^{o}C (amorphous)
Boiling point : 2,950 ^{o}C
Magnetic susceptibility : $-29.6{\cdot}10^{-6}\ cm^3/mol$

Silica is naturally crystalline, hence it is necessary to synthesize it by certain methods to obtain an amorphous form (Julia, 2002; Yao et al., 2015). The crystalline form of silica is quite diverse and the shapes are known as polymorphs.

2.3 Extraction Methodologies

The silica extraction method is concerned with removing impurities or unexpected substances from the natural materials used. This substance can be extracted using several methods, including thermal and microbial processing (Uda et al., 2021).

2.3.1 Thermal Method

The thermal method involves heating in the form of calcination or pyrolysis. According to Venkateswaran et al., 2013, the extraction of silicon from rice husks was conducted through a thermal method that functions to remove most of the organic substances. In this study, the heating temperature was varied to see its effect on the percent Si obtained. Furthermore, after dehydration with sodium hydroxide solution, the acid-treated rice husks are used for the direct extraction of silica. Subsequently, silica is obtained from the resultant sodium silicate through the addition of an appropriate amount of mineral acid for 5–6 hours to acquire the rice husk ash (RHA) by RH pyrolysis at temperatures between 500 °C to 850 °C in a muffle furnace. The results showed that the percentage of silicon obtained increases with the calcination temperature, where absorption peaks and wavelengths of around 300–310 nm are present at 850 °C. The results in this case, where the silicon powder had a pure phase formation, high purity, and good absorption peak were better than other investigations.

2.3.2 Biological methods

This method makes use of living things such as animals, bacteria, and fungi. Estevez et al., 2009 reported the use of California worms to extract silica from rice husks which were moistened and digested in their mouth using enzymatic fluids containing lipase, amylase, trypsinogen, etc. However, the structure of silica, which can only be completely dissolved with hydrogen hydrofluoric acid, makes the enzymes unable to react with silica. The rice husk enters the pharynx which is located in ring 6, crushing and sucking the food and sending it to the oesophagus where the pH is neutralized using $CaCO_3$. In-ring 20, the intestine begins the process of digestion and absorption, during which the strong action of the muscles in the intestinal wall destroys food while the endocrine, pharyngeal and calciferous glands provide enzymes, such as amylases and proteases that produce degradation of organic matter. However, the mechanical work is produced by the movement of the muscles in the intestines of the worms grinding the fine silica. The humus is dried at room temperature, pulverized to 210 μm, and stored for a week in a secure plastic bag. Subsequently, this is calcined at different temperatures and the undigested sample rinsed with distilled water until a neutral pH was obtained and dried. This research produced high purity silica with nano-size distribution and spherical shape.

2.3.3 Chemical method

This method involves chemicals such as acids, bases, and other agents which are used to remove impurities and increase the purity of the silica contained in it. Apart from this, acid leaching can convert impurities into ions that are capable of dissolution (Vaibhav et al., 2015). The effect of acids in removing metal impurities from rice husks was demonstrated through the use of HCl, which was more effective when compared to H_2SO_4, and HNO_3 (Chakraverty et al., 1988).

According to Gao et al., 2019, the silica extraction from rice husks was conducted through chemical methods, where HFA was dissolved with 15 % (1: 4.5) sodium hydroxide solution by weight and stored at 100 °C for 2 hours, then filtered. Furthermore, NaOH was added to separate the silica from impurities and to form a sodium silicate solution, which was transferred to hydrosol silica by adjusting the pH to 6 or 7 with the HAc solution through an ultrasonication assisted process. Consequently, the effect of pH on silica purity was produced and the powdered SiO_2 was obtained after dehydrogenation gelation and drying. Kamath and Proctor, 1998 reported the solubility of amorphous silica to be low at pH <10, hence a more alkaline pH is excellent in producing a material of high purity. The recent extensive application of this form of nano-silica in areas, including chromatography, pharmaceuticals, adsorbent materials, electronic components, drug delivery systems, catalysts, and dyes has resulted in its increasingly high demand.

Several studies were conducted using chemical methods which are widely used due to their simplicity when compared with other processes. Adebisi et al., 2017 also used this method to obtain nano-sized silica from the cassava periderm using three different routes. The acid treatment pre and post calcination are effective in reducing or removing soluble metal impurities. Furthermore, silica obtained from the pre-treatment, post-calcination process, and the ethylene glycol-modified sol-gel method was discovered to produce particles of higher purity that were in the nano range from 3.12 to 50.75 nm (Adebisi et al., 2017). Table 1 shows current research in silica extraction from natural materials.

The extraction of silica from natural materials produces different particle sizes. Particle size and size distribution are the most important characteristics of the nanoparticle system due to its influence on the properties of the material and the stability of particles. Nanoparticle-sized silica (1-100 nm) provides better activity. This is because nanoparticles have a relatively higher intracellular absorption compared to microparticles and are available for a wider range of biological targets due to their small size and relative mobility. In addition, smaller particles have a larger surface area and nanoparticle surface characteristics can be easily modified to produce certain properties (Li et al., 2020). Based on this, extraction methods that take into account the size of nanoparticles, eco-friendly extraction and can be widely used in most natural materials or waste is a chemical method. This method is quite simple and relatively inexpensive as well as produces good results. Nonetheless, this method needs to be adapted to the 12 principles of green chemistry to support eco-friendly and harmless for the surrounding environment.

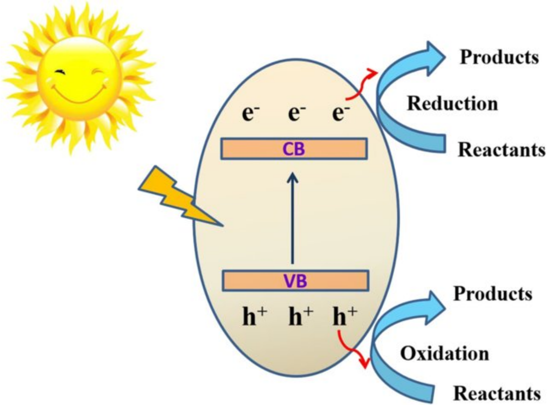

Figure 4. Photocatalytic Process in Semiconductor Materials, Adapted from He and Zhang, 2019

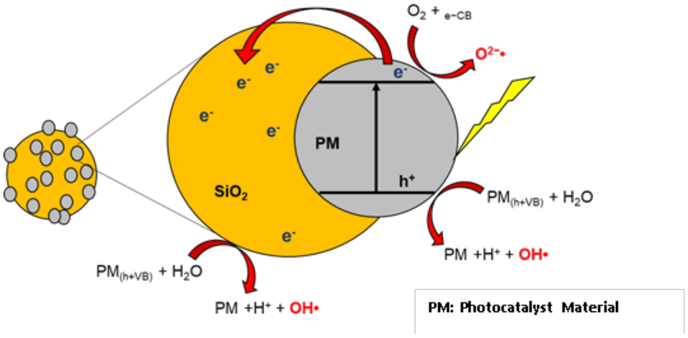

Figure 5. The Role of Silica in Photocatalyst

2.4 Extract Silica as a Supporting Material in Antibacterial Photocatalysis

Silica has various applications, which include its role as a supporting material for a photocatalyst in the photocatalytic process. This procedure is a combination of chemical reactions that requires light elements and photocatalysts to accelerate chemical transformations (Effendy, 2010). This compound is generally a semiconductor because it has a bonding and an antibonding band separated by bandgap energy (Jal et al., 2004; Holleman A. and Wiberg, 2001). Figure 4 demonstrates this phenomenon in semiconductor materials.

Table 2 shows the difference particle size in the extraction product. In photocatalyst, particle size become an important factor that is able to influence the activity of photocatalysts. The surface structure and shape of crystals can be affected by

Table 2. Current Research in Silica Extraction

Starting material	Method	Products	Particle size (nm)	References
Sand	Chemical	Silica	-	(Triwikantoro D and Zainuri, 2015)
	Chemical	Silica	Pore Size 15-68 nm	(Vaibhav et al., 2015)
	Chemical	Silica	-	(Eddy et al., 2015)
	Chemical	SiO_2	Macrometer	(Ishmah, 2019)
	Sol-gel	Silica	~200 nm	(Ismail et al., 2020)
	Chemical	Silica Gel	150 mesh	(Setyoningrum et al., 2020)
Rice Husk	Biotransformation	Silica	152-254 nm	(Espindola-Gonzalez et al., 2010)
	-	SiO_2 in Thin Film	-	(Klankaw et al., 2012)
	Acid Pre-treatment	Silica	6 nm	(Rafiee et al., 2012)
	Hydrolysis	Amorphous Silica	50-200 nm	(Zemnukhova et al., 2015)
	Leaching Calcination	Silica	181.2-294.7 nm	(Carmona et al., 2013)
	Pyrolysis	Silicon	70-100 nm	(Venkateswaran et al., 2013)
	Acid Leaching	SiO_2 Biogenic Amorphous	Nanometer	(Ferreira et al., 2015)
	-	Silica	-	(Motlagh et al., 2020)
	-	Nanoparticle Silica Amorphous	17.71 ± 7.53 nm	(Nayak and Datta, 2021)
Palm Ashvia	Acid Leaching	SiO_2	-	(Pa et al., 2016)
	Sol-gel	Nanoparticle Silica	50–98 nm	(Imoisili et al., 2020)
Fly Ash Waste	Thermal	SiO_2 Mesopore	8-20 mm	(Permatasari et al., 2016)
	Alkaline	Nanoparticle Silica Amorphous	10-60 nm	(Yadav et al., 2020)
	-	Nanoparticle Silica	Nanosize	(Wang et al., 2020)
	Alkaline	Silica Amorphous	380.9 nm-178.8 nm	(Liang et al., 2020)
	Acid-base	Nanoparticle Silica Amorphous	60–90 nm	(Uda et al., 2021)
	-	Silica Nano-sorbent	~27.176 nm	(Chatterjee et al., 2020)
	-	Silica Mesopore	-	(Miricioiu and Niculescu, 2020)
	Alkaline	Silica Fume (SF)	-	(Sevinç and Durgun, 2020)
Coconut Husk Ash	Green Synthesis	Orthorhombic Tridymite (silica)	200–750 nm	(Anuar et al., 2020)
Maize Stalk	Green Production	Silica Nanoparticles	-	(Adebisi et al., 2019)
Olive Stones	Alkali Leaching Process	Nano-silica	Nanosize	(Naddaf et al., 2020)
Traditional Chinese Joss Paper	Alkaline Treatment	Silica	Nanosize	(Ramanathan et al., 2020)
Sugarcane Bagasse Ash	Alkali Leaching Process	Silica Microparticles	Microsize	(Adebisi et al., 2017)
Cassava Waste	Sol-gel	Silica Amorphous	Nanosize	(Adebisi et al., 2017)
Bamboo Leaves	The Box Behnken Design	Amorphous Nano-silica	-	(Olawale, 2020)
Corn Cobs Husks	Biotransformation	Silica Nanoparticles	~40 and ~70 nm	(Piela et al., 2020)

particle size (Cao et al., 2016; Hwang et al., 2017). Research conducted by Lin et al., 2006 successfully synthesized TiO_2 nanoparticles of different particle sizes (12–29 nm) and found their influence on ribbon gaps indicating that particle size is capable of affecting the electro-optical properties of photo. The increase in particle size causes a decrease in the rate of photocatalytic, and vice versa (Lin et al., 2006). Then the increase in photocatalyst activity is also influenced by the area of the active side (surface) of the material. The increase in particle size causes a decrease in surface area. This is because the dynamics of the charge carrier and the efficiency of the particle adsorbs change thus providing different photocatalytic activity (Cheng et al., 2013). As an antibacterial, size becomes important because the infiltration of particles in the cells in the bacteria will be easier if it has a small size (nanoparticles) so that the degradation or destruction of bacteria will be more optimal (Qi et al., 2017).The mechanisms that occur include (Sirimahachai et al., 2010; Castellote and Bengtsson, 2011; Sellapan, 2013):

- The subjection of an n-type semiconductor to light (γ) with the appropriate energy, causing electrons (e-) in the valence band to be excited towards the conduction band and leaving a positive hole which is abbreviated as h+, in the valence band.

- The e^- and h^+ pairs will undergo recombination, either on the surface or in the bulk particles. Subsequently, the hole (h^+) will initiate an oxidation reaction and the electron (e^-) will initiate a chemical reduction reaction around the semiconductor surface.

- Products resulting from a redox reaction with the environment will form radicals that have high oxidation-reduction power.

Compounds that primarily kill or retard the development of bacteria, without generally poisoning the adjacent tissue possess antibacterial action (Hajipour et al., 2012). Its mechanisms of action include inhibiting cell wall growth, resulting in changes in cell membrane permeability, inhibiting protein synthesis, and inhibiting cell nucleic acid synthesis (Ferronato and Torretta, 2019; Hajipour et al., 2012; Kim et al., 2007; Khan et al., 2020). The exact mechanism of nanoparticle toxicity to various bacteria is not fully understood (Hajipour et al., 2012) and is generally activated by the introduction of oxidative stress through the formation of free radicals, such as ROS, after administration (Sellapan, 2013; Khan et al., 2020; Yang et al., 2016). Figure 6 shows a schematic of nanoparticle toxicity.

The mechanism behind nanoscale activity in bacteria has

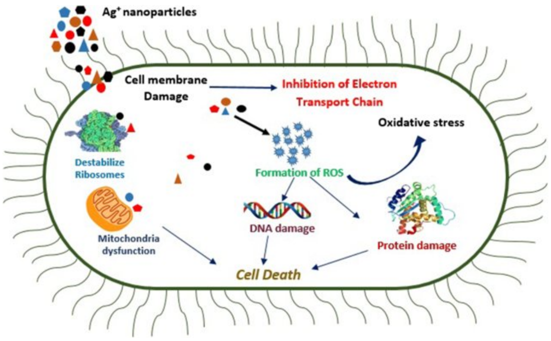

Figure 6. Proposed Mechanism of Action of Silver Nanoparticles Against Bacterial Growth/Proliferation, Adapted from Rahman et al., 2019

not been fully elucidated. The three most common mechanisms of toxicity proposed to date include the (Asharani et al., 2008):

- DNA replication and disruption of ATP production prior to the absorption of free ions
- Nanoparticles and ion generation ROS, and
- Nanoparticles direct damage to cell membranes.

Asharani et al., 2008 suggested that the concentration of h+ is important in the reactions that occur within the mitochondria of eukaryotic cells. Likewise, when there is a proton motive force in the cell membrane, a similar mechanism can occur. ROS works against bacteria through multiple methods, including the interaction with the thiol group of enzymes and proteins which are essential for bacterial respiration. Also, by relating with the transport of essential substances across cell membranes (Cho et al., 2005), and binding to bacterial cell walls and, altering its cell membrane functions (Percival et al., 2005).

Overall, the potential of silica as supporting material for antibacterial photocatalysts can be proven by its role in increasing the produced ROS. Therefore, the review of silica application in the field of photocatalyst as a supporting material is necessary. There have been many studies discussing the effectiveness of photocatalyst performance after adding silica.

Klankaw et al., 2012 succeeded in making TiO_2-SiO_2 thin film with the extracted silica for dye degradation. This thin film is made through the self-assembly method by mixing a solution of SiO_2 with titanium precursor and allowed to stand for precipitation together with silica and titanium crystals. The results showed the photocatalytic decolorization of methylene blue dye (MB) reached 81 %, where the advantages of SiO_2 include; (1) the increase of the film absorbency and (2) the provision of hydroxyl radicals to promote the photocatalytic reaction. The photocatalytic reactivity of thin films for decolorization of MB dye depends on the increase in the specific surface area and chemical structure of the photocatalyst (Figure 7).

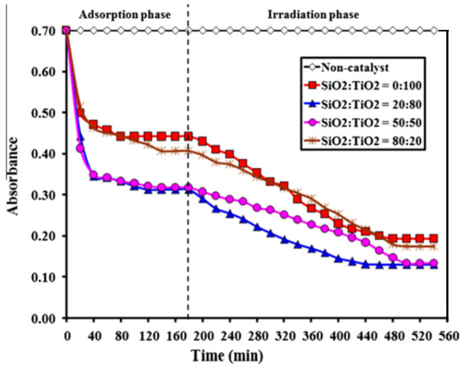

Figure 7. Decolorization of MB Dye of TiO_2–MCM-41 Thin Film at Various Molar Ratios (Klankaw et al., 2012)

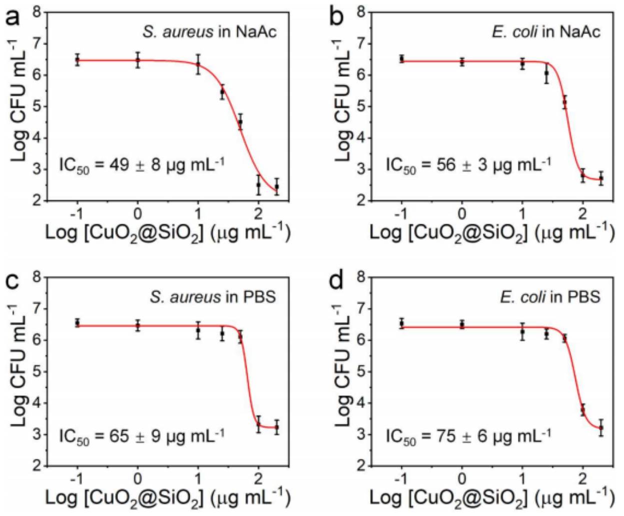

Figure 8. IC50 of CuO_2@SiO_2. *S. aureus* in NaAc buffer (a) and PBS (c) and *E. coli* in NaAc buffer (b) and PBS (d). Red lines are the sigmoid fitting curves to obtain IC50 values, Adapted from Li et al., 2021 with American Chemical Society

Mohamed et al., 2019 reported biogenic silica as the supporting material for nano polyacrylonitrile in the degradation of malachite green (MG) dye. An investigation was conducted on the multiple factors affecting the degradation of this color, including solution pH, dye concentration, and irradiation time and on its photocatalytic performance under visible light in an aqueous solution. The results from the studied nanocomposite fiber demonstrated an exceptional photodegradation performance with a 98 % maximum efficacy in less than 10 minutes for Malachite green. Furthermore, the fabricated fiber is used in continuous operating modes, such as fixed-bed columns because of its flexibility.

Sarkar et al., 2017 explained that doping or composite manufacturing is an active method of diffusing light absorption into the visible region. The study describes the extraction procedures for nano-silica from rice husks and the sol-gel preparation of titania-nano-silica composites for photocatalytic and photovoltaic applications. Furthermore, the extracted nano-silica acts as a green filler to increase the surface area of titania photocatalysts and inhibit recombination. The general plan is anticipated to increase the semiconductor activity of TiO_2 by passive composites. The titania-nanosilica hybrid composites showed significant results in the application of solar cell and photocatalyst fields.

These studies prove that adding SiO_2 to a photocatalyst can improve its performance. Therefore, silica can increase the adsorption power, provide hydroxyl radicals, promote photocatalytic reactions, minimize recombination, prevent agglomeration, increase thermal resistance, and increase stability against high temperatures (Bapat et al., 2016; Cho et al., 2005; Percival et al., 2005; Sarkar et al., 2017; Otero-González et al., 2014). The advantage of this silica is that it can enhance the process by improving the performance of the photocatalyst (Nilchi et al., 2010; Hou et al., 2018; Shaban et al., 2020). This also encourages the assumption that more ROS will be produced, which has a very important role in the inactivation of bacteria as previously described (Marambio-Jones and Hoek, 2010; Kim et al., 2011; Hou et al., 2018).

So far, a great deal of research have been conducted on silica as a supporting material for antibacterial photocatalysis (Zemnukhova et al., 2015; Carmona et al., 2013; Permatasari et al., 2016; Ramanathan et al., 2020; Olawale, 2020; Piela et al., 2020). However, the silica used was synthetic as was conducted by Hou et al., 2018, in making SiO_2 mesoporous spherical particles that were coated separately with Ag_2O, Cu_2O, CeO_2, and NiO nanoparticles and used for 26 antibacterial applications. Synthesis of CuO_2@SiO_2 as an antibacterial is also performed by Li et al., (2018) to achieve percent inactivation bacteria reached 99.9 %.

The bactericidal effect of the composites was tested against 27 Staphylococcus aureus in dark or LED lighting conditions. The results showed that the growth of 28 Staphylococcus aureus was severely stunted within 3 hours. Research on SiO_2 produced from chemical synthetic materials has been conducted and proven to be able to be a good supporting material. Research conducted by Tan et al., (2016) showed antibacterial activity of AgCl and AgCl@SiO_2 nanoparticles on *E. Coli* and explain about the role of SiO_2. Researchers have successfully synthesized AgCl@SiO_2 nanoparticles with a porous shell-shaped SiO_2 that show improved stability compared to Ag(I) samples. Silica shell protection makes nanoparticles resistant to light irradiation and can be used repeatedly. The application of SiO_2 as supporting material TiO_2 for antibacterial in textiles application has been done by Kartini et al., 2010. The result of this study is the increased antibacterial activity of TiO_2/SiO_2 in layered cotton which explains that the presence of SiO_2 is able to increase antibacterial activity. Researchers explained the possibility of increased antibacterial activity due to the increase in coating sole adhesive to cotton fibers.

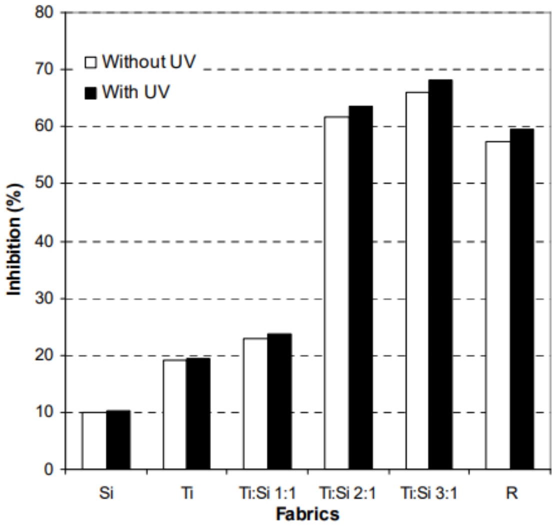

Figure 9. Antibacterial Activity of the TiO_2-SiO_2 Sols Coated Cotton Fabrics and Commercial Antibacterial Product (R) Against *Escherichia coli*, Adapted from Kartini et al., 2010

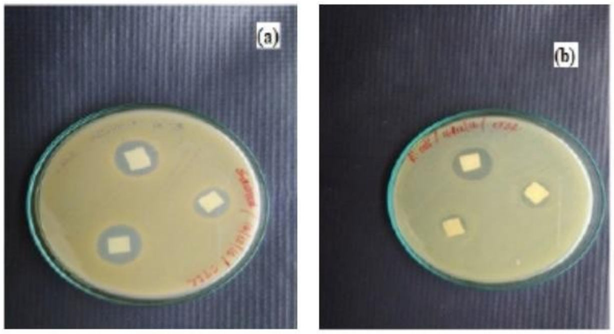

Figure 10. Antibacterial Activity of ZTS Nanocomposite Coated Cotton Fabric Against a) *S. aureus* area b) *E. coli*, Adapted from Manoharan et al., 2018

Manoharan et al., 2018 also successfully synthesized nanocomposite ZnO/TiO_2/SiO_2 as antibacterial agents in cotton fabrics against *E. coli* bacteria (Gram negative) and *Staphylococcus aureus* (Gram positive) are very significant. Strong bactericidal effects observed for nanocomposite-coated clothing for both bacteria strains had an inhibitory zone of 19 mm observed for *S. aureus* and 18 mm for *E. coli*.

The advantages of the hydrophobic properties of SiO_2 in its antibacterial activity are explained in Zhao et al., 2020 research. In this study the fabrication of MoO_3-SiO_2-Ag_2O was done by distributing a small amount of Ag_2O separately in the amorphous SiO_2 matrix. The desired antimicrobial activity to inactivate *Escherichia coli, Salmonella Typhimurium* and *Staphylococcus aureus* can be achieved due to the presence of this layer of surface hydrophobicity in SiO_2, and other reasons such as the release of Ag^+ ions, surface acid reactions and photocatalytic activity, which can be used to reduce hospital-acquired infections.

Hoang et al., 2016 showed *E.coli* inactivation under UV-C irradiation using photocatalysts TiO_2 (Degussa-P25), TiO_2-SiO_2 or 1 % Ag-TiO_2-SiO_2 for 30 minutes. SiO_2 is signifi-

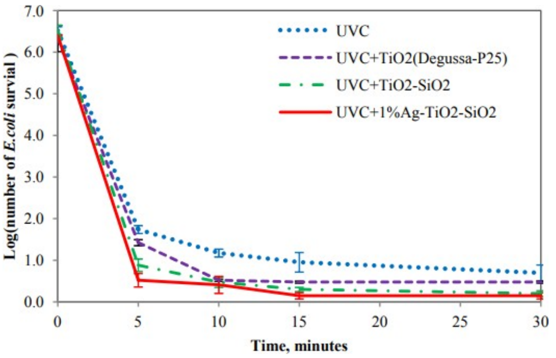

Figure 11. Inactivation Effect Against *E.coli* by Ag-TiO_2-SiO_2 Photocatalysts under UV-C Irradiation Compared to TiO_2 (Degussa-P25) and TiO_2-SiO_2, Adapted from Hoang et al., 2016

cantly capable of increasing specific surface areas compared to commercial TiO_2 (P25). The higher the specific surface area of the photocatalyst the higher the area that opens between UV-C rays and the catalyst so as increase photo-active of the catalyst. Therefore TiO_2-SiO_2 antibacterial activity is superior to commercial TiO_2 (P25) under UV-C irradiation.

This proves that synthetic silica can be used as supporting material for antibacterial photocatalysts unlike those extracted from natural ingredients which are rarely used. The utilization of extracted silica is very important as a green production effort. Therefore, it can be concluded that the material has the potential to improve the performance of antibacterial photocatalysts as a species supporting material (Hou et al., 2018).

3. CONCLUSIONS

Silica can be produced from the extraction of natural materials such as sand, agricultural waste, and leaves. This material contains strong tetrahedral bonds which form amorphous and crystalline structures and can be extracted by several methods such as thermal, biological, or chemical methods or their combination. Silica as a photocatalyst support material is excellent for use with an increased degradation function and has the potential as supporting material for antibacterial photocatalysts.

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