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Methods for the Preparation of Silica and Its Nanoparticles from Different Natural Sources

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Abstract

Silica (SiO_2), a component of the earth's crust, has been in use for many nanotechnological applications. This review presents one of the newest methods for safer, more affordable, and more ecologically friendly production of silica and its nanoparticles from the ashes of agricultural wastes. The production of SiO_2 nanoparticles (SiO_2NPs) from different agricultural wastes, including rice husk, rice straw, maize cobs, and bagasse, was systematically and critically discussed. The review also emphasizes current issues and possibilities linked with contemporary technology to raise awareness and stimulate scholars' insight. Furthermore, the processes involved in isolating silica from agricultural wastes were explored in this work.

Keywords Silica · Agricultural waste · Nanotechnology · Silica nanoparticles · Renewable · Synthesis

Introduction

Silicon dioxide (SiO_2), sometimes referred to as silica white color, is a useful inorganic compound with wide applications [1]. Around 75% of the earth's crust is made up of this substance and can be extracted or synthesized from a variety of natural resources, including beach sand and other non-biological sources [2], as well as coal wastes, such as flying ash sludges [3], with over 91% recovery. Each type of SiO_2, in the form of gels, as well as crystalline or amorphous states [4], has various important physical and chemical properties. (part 1). Most industrial silica is made from natural sources. Quartz and quartzite are the most stable and pure polymorphs of silica found in almost all mineralogical rocks. These rocks are heated to generate sodium silicate [5]. In addition to sodium silicate, tetraethyl orthosilicate (TEOS), and tetramethyl orthosilicate, commercially available alkoxysilane compounds can be used as silica precursors. A prolonged acute exposure to TEOS can lead to death [6]. To synthesize silica particles in the presence of a catalyst, the

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previously stated sources are frequently employed [7]. In order to produce them, quartz sand and sodium carbonate are melted together at 1300 °C. This process is associated with the discharge of wastewater and the production of significant amounts of carbon dioxide, which is a primary concern in regard to the greenhouse gas effect [8]. These are not encouraging prospects for the production of commercial silica [9]. Because of this, there is a pressing need to obtain a silica precursor that is not only less expensive but also more friendly to the environment [10]. Soltani and his colleagues studied the utilization of the technology for extracting silica and activated carbon from rice husks [11].

Indeed, silica can also be obtained from agricultural wastes, such as rice husks [12], coconut pulps [13], coffee husks [14], wheat husks [15], corn cobs [16, 17], and bagasse ashes [18, 19], with almost a 99% acquisition rate, depending on the natural source. It is possible to obtain biosilica, also known as reactive silica by using a variety of biomass extraction techniques, including hydrothermal [20], sol–gel [21], chemical vapor deposition [22], combustion synthesis [23], microbial hydrolysis processing [24], and other methods, e.g., the leaching and mixing alkaline treatment followed by precipitation with acid [25–28]. Particularly, the thermal approach to the preparation of bio-silica from biomass is relatively easy and cost-effective, and it also requires lesser time than chemical processes.

In general, the thermal breakdown of biomass consists of three stages: the removal of moisture and drying (up to 150 °C), the thermal decay of organic compounds (between 215 and 350 °C), and the thermal decay of carbonaceous components (between 350 and 690 °C) [29–31]. These three stages can be adopted in many applications, including the production of glass [32, 33], biomedical applications [34, 35], the production of bio-cements [36], and adsorbents [22], including those used as fillers in composites, electrical parts, catalysts, drug delivery devices, thermal insulators, rubbers, chromatographic packings, and ceramics [37, 38]. They can also be applied as anticorrosion agents to strengthen polymeric composites [39].

The development of nanotechnology has allowed for the production of silica nanoparticles (SiO_2NPs). In recent years, SiO_2NPs have come into the spotlight as a result of their beneficial use in numerous industrial applications. SiO_2NPs can be obtained using many methods, such as the vapor phase reaction or the sol–gel process, among others [40–43]. Additionally, this list includes solution precipitation and sol–gel processes [44, 45]. The sol–gel method is one of the most important ones for the production of nano SiO_2. This is due to the possibility of methodical monitoring of reaction conditions with this method, which allows to control the particle size, the size distribution, and the form of the resulting nanomaterial. Mohanraj and his colleagues produced nano SiO_2 from corn cob ash using a precipitation method [16]. The sol–gel approach offers the advantage of the production of SiO_2NPs at much reduced costs. Worms are also used to generate nano SiO_2 using the bio-digestion process of rice husks [46]. In addition to their usage as a filler in composite materials, SiO_2NPs are also used in drug delivery systems, catalysts, biomedicine applications, biological imaging, chromatographic packings, and sensors. Liquid armor is another application for SiO_2NPs [38, 47–49].

This review discusses both SiO_2 recovered from agricultural wastes and suitable extraction procedures/processes and methods applied for its production. A variety of SiO_2 sources that are derived from agricultural wastes as well as the extraction procedures/processes used to obtain SiO_2 from these sources are discussed. SiO_2NP synthesis that utilized agricultural wastes is also presented. This review presents a summary of the research that was carried out regarding the valorization of natural resources used in the investigation and development of value-added SiO_2-based materials (Fig. 1). Meanwhile, the most significant applications of SiO_2 in different fields are highlighted.

Extraction Methods

Production of SiO_2 from agricultural wastes can be accomplished in three different ways: chemical treatment, thermal treatment, or microbiological treatment [10] (Fig. 2). In the chemical treatment, also known as the acid leaching, SiO_2 is removed from agricultural wastes by means of the chemical procedure [34, 50–56]. The acid leaching is used to enhance the quality of SiO_2 found in agricultural wastes by removing any unnecessary contaminants. A straightforward acid treatment is required to decompose the organic compounds that are present in the agricultural wastes, eliminating them in this way and transforming other types of contaminants into the solution in their dissolved forms, i.e., ions [57]. Procedures for SiO_2 leaching usually involve the utilization of solutions of sulfuric acid (H_2SO_4), hydrochloric acid (HCl), and nitric acid (HNO_3) [58]. Many researchers concluded that the preliminary leaching of rice husks (RH) with a solution containing HCl, HNO_3, H_2SO_4, NaOH, and NH_4OH, followed by boiling the mixture prior to the heat processing at temperatures ranging from 500 to 1400 °C for varying amounts of time, is the most effective method to remove the majority of metallic impurities and produce ash-silica that is white and has a high specific area [29].

Many researchers also used a severe acid-leaching procedure to remove metallic contaminants and organics from RH [58–60]. The reflux boiling in a 3% (v/v) HCl and 10% (v/v) H_2SO_4 solution was used in several studies to perform the mentioned acid leaching Riveros and Garza employed a 3% HCl solution for that process [61].

Fig. 1 The major agricultural waste sources of silica

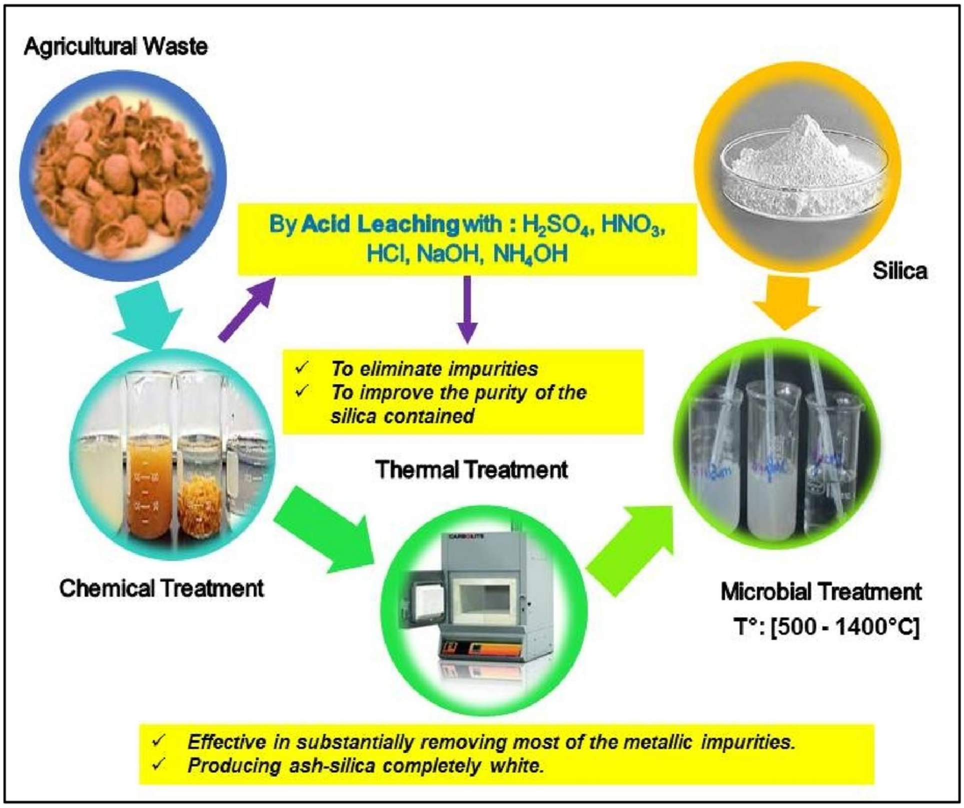


Fig. 2 Production of silica from agricultural wastes

Conradt et al. also used the reflux boiling in 2.4 mol/l HCl or 3.6 mol/l H_2SO_4 solutions to perform the acid leaching [62]. The work of Kalapathy et al. on the extraction of SiO_2 from rice husk ash (RHA) is noteworthy [63] (Fig. 3).

In alkaline conditions, SiO_2 is extracted as silicates, and the subsequent processing with acid enables it to precipitate as a gel. In this study, the resulting SiO_2 gel was heated at 80 °C for 12 h in order to produce amorphous SiO_2 Xerogels

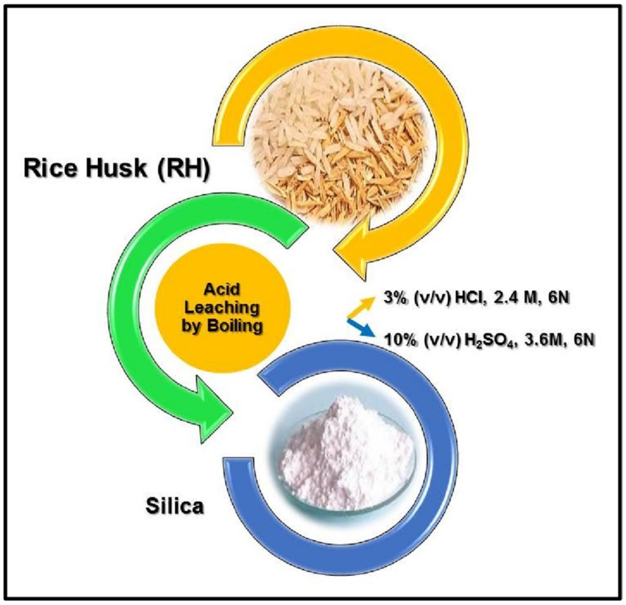


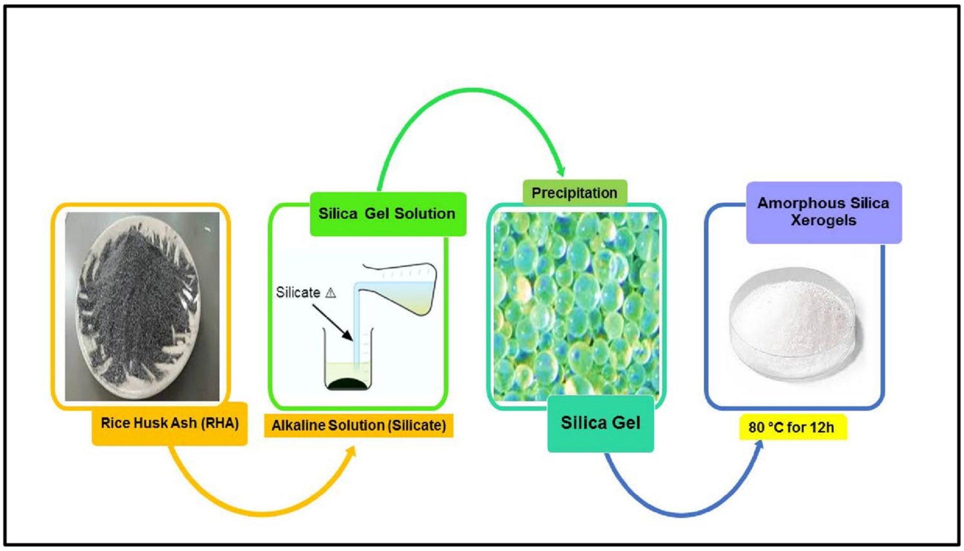
Fig. 3 Extraction of silica from rice husk ash (RHA)

[63]. In another study [29], the same researchers extracted SiO_2 from RHA using a procedure that was only marginally distinct from the one reported by Kalapathy et al. [63]. They utilized this method to acidify solutions of RHA-extracted silicate by adding various acids to the mixture. It was found that the acid composition and the subsequent washing process both had an effect on the elemental content of gelatinous precipitated SiO_2 [63]. There were several attempts made by Della et al. to extract SiO_2 from RHA as well [29].

Furthermore, silica has been effectively generated from RHA, as reported in a study. RHA was pretreated with several acids in the study (1, 3, 5, or 7 using 6 N HCl, HNO_3, and H_2SO_4). From bio-waste, a variety of chemical processes yield silica material (Fig. 4). Hu et al. used three efficient procedures to make amorphous silica and activated carbon: toluene/ethanol, $NaClO_2$, and KOH. Calcination was used to make nanoscopic silica nano-spheres (100–120 nm). In order to manufacture amorphous silica, organic acid leaching was utilized as an alternative to the use of a strong acid [50, 64]. According to Chen et al., ionic liquid was also utilized in the manufacture of silica from agricultural waste [65].

Supitcha et al. reported a yield of 54.95 percent by silica extraction with a 1:6 ratio of RHA and 0.5 N NaOH (analytical grade) at 60 °C. According to Handayani et al., the exact ratio of RHA and 1 N NaOH (analytical grade) at 80 °C yielded a value of 91.59% (Fig. 5). NaOH and extraction temperature can be used to determine how much silica is extracted from a given amount of NaOH [66, 67]. By using a simple acid treatment, metallic impurities can be converted to soluble ions [68, 69]. As a result of the utilization of volatile hydrochloric acid, the reaction equipment can be harmed by the high temperature and the acidity present during the process. Additionally, costs go up and the environment becomes polluted. In light of this, sulfuric acid treatment can be helpful as acid treatment, with purification done in two processes. The first stage is to remove soluble metallic impurities with an acid solution, and the second is to sinter the leached sample to reduce the overall carbon content. The overall purity of the silica content will be improved by these two stages. Paya et al. [70] made a glyceosilicate solution by treating RHA with glycerol. This solution was then compared to a $Ba(OH)_2$ aqueous glycerol solution. Their research produced a white powder that turned out to be amorphous silica. Yalc et al. [20] worked on extracting silica from RH that was relatively

Fig. 4 Silica extract in alkaline solution



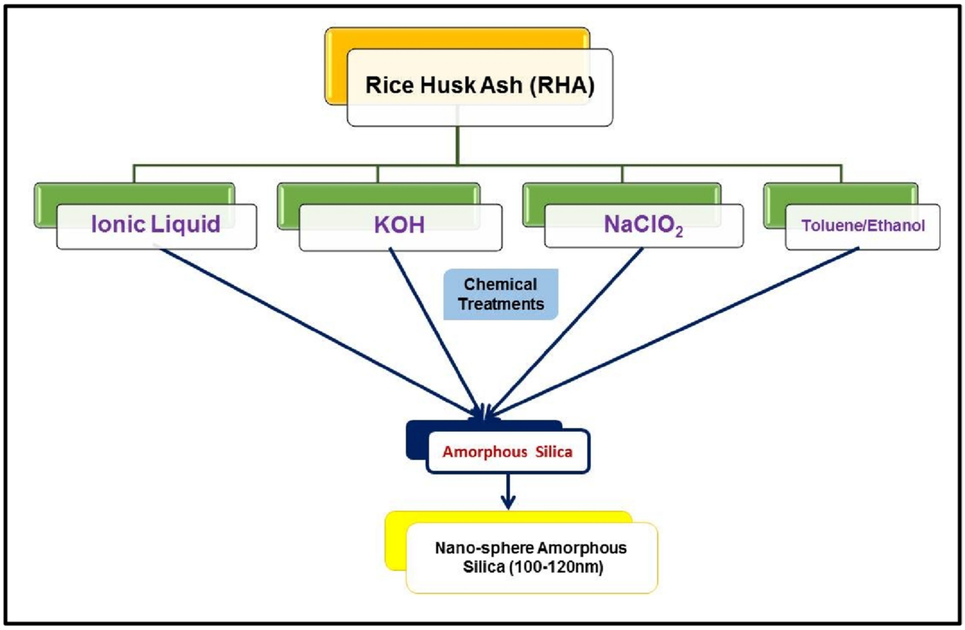


Fig. 5 Preparation of amorphous silica

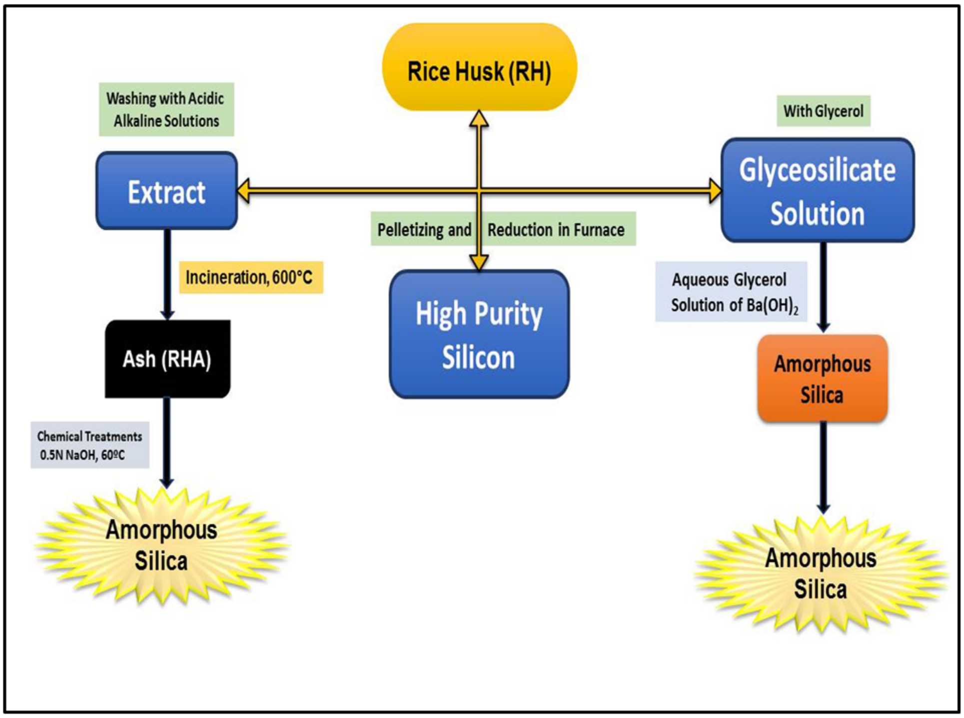


Fig. 6 Extraction of silica from RH

pure (Fig. 6). RH were cleaned in acidic and alkaline solutions before being burned at 600 degrees Celsius in a static air atmosphere to convert them to ash. After that, silica was extracted from the ash. This approach yielded amorphous silica with a purity of more than 99%. Many techniques have been used to extract silica from RHA. Hunt et al. [71] used a modified electric arc furnace to pelletize, reduce RH, and produce a high-purity silicon. Two studies reported that acid leaching and gasification can be used to recover silica from the hull of the RH [30, 72]. Real et al. [73] reported that the process of acid leaching in RH before they are burned produces silica powder with a large surface area. If the acid leaching stage is done after the combustion process, the amount of pollution caused by the process is reduced. Figure 4 shows the method used to extract silica from RH.

Acid Leaching

In order to prevent the melting of biosilica during the thermal treatment process, metallic impurities such as (Na, K, Mg, Ca, Al, etc.), as well as their oxides, must be eliminated early into the process. This approach is utilized to accomplish this goal. It has been reported that HCl has a significant leaching capability for these particular pollutants, and that explains why it was selected [29, 63]. In order to get silica of a high purity, the husk must first be pre-treated with mineral acids, followed by regulated ashing [83–85]. The use of strong acid poses an economic challenge due to the high cost of acid. To help address this, some studies employed acids that are cost-effective, environmental friendly, safe, and harmless [59]. For example, cassava periderm was prepared by Adebisi et al. [2017b]; the cassava periderm was soaked in 0.1 M HCl for 90 min at 45.5 °C to remove soluble compounds [86], there are also many examples from previous studies shown in Table 1.

Thermal Processing

Usman et al. [87], investigated the silica concentration of bagasse using thermal treatment with temperature variations of 500, 600, and 700 °C, which yielded ash content of 12.65, 10.89, and 9.95 percent, respectively. Mehta [88] reported that temperatures exceeding 700 °C yield reactive amorphous ash.

Alkaline Treatment (Sol–Gel)

The alkaline extraction sol–gel method was used in several studies [89–91]. Ash can be transformed into silica gel via a number of processes; one of the most prevalent of which is this approach. In the beginning, silica was produced from ash by heating sodium silicate sol, silicon alkoxide gels, or halide gels until they turned into a gelatin polymer. The process can be repeated until the gels achieved their desired solid state [92]. According to Kalapathy et al., amorphous silica can be

Table 1 Different methods of extracting silica from natural products

Raw material	Average silica particle size	Acid washing	Extraction methods	Temperature	Purity index	Products	References
Rice husk ash	0.50–0.70 µm	HCl washed prior to extraction	Acid precipitation method	60 °C	99.2%	Amorphous silica	[12]
Sugarcane waste ash	> 20 nm	HCl washing	Surfactant mediated synthesis	60 °C	99.08%	SiO_2NPs	[74]
Paddy straw	15–20 nm	Acid wash	Acid precipitation	37 °C	NA	Nano-silica	[75]
Olive stone	15–68 nm	10% HCl wash	Alkali leaching process	Ambient temp	NA	Crystalline silica	[76]
Pine cone	37 nm	3 M sulfuric acid	Thermal decomposition	600 °C	NA	Silica nanoparticles	[77]
Wheat straw	75–320 nm	4:1(v:v) mixture of nitric and sulfuric acid	Leaching in a 10% (v/v) HNO_3 and calcination	400–700 °C	NA	Amorphous hydrated silica	[78]
Coconut husk ash	NA	NA	5 N Sulfuric acid treatment	50 °C	High purity	Crystalline silica	[13]
Coconut husk ash	NA	NA	2.5 N NaOH treatment	100 °C	High purity	Amorphous silica	[13]
Rice husk ash	10–15 nm	No washing with acid	Acid precipitation technique	80 °C	98.9%	Amorphous silica	[79]
Cassava periderm	62.69 nm	0.1 M HCl	Sol–gel method	700 °C	NA	Silica nanoparticles	[80]
Palm kernel shell ash	50–98 nm	No washing with acid	Sol–gel method	750 °C	Na	Amorphous silica nanoparticles	[17]
Olive stones	15–68 nm	Acid wash	Alkali leaching extraction method	900 °C	–	Crystalline silica	[81]
Banana leaves	1–2 µm	NA	Dissolution and precipitation process	650 °C	–	Silica powders amorphous structure	[82]
Teff straw	–	Acid wash	Sol–Gel method	900 °C	> 99%	Biosilica	[51]

obtained from RHA through the use of alkaline extraction combined with alkaline solvents at low temperatures. The solubility of amorphous silica is relatively very low at pH levels below 10, but it rapidly increases once the pH level is raised above 10 [93]. Potassium hydroxide (KOH) and sodium hydroxide are two common alkaline solutions (NaOH) used. The use of alkaline solubilization of amorphous silica as a low-energy strategy has the potential to reduce cost compared to the conventional process of smelting.

Nano-Silica Synthesis

Several studies have demonstrated the extraction of nanostructured silica from rice husk, coconut shells, and wheat straw [94–96]. The acid leaching procedure was used to make the silica nano-powder [97]. Attempts to make nanosilica powder from RH have been made in recent years [62, 73, 91, 94, 98, 99]. In line with this, Haiharan et al. [100] produced nano-silica by burning RH at 650 °C for 2 h, yielding a virtually spherical, homogeneous, and agglomerated material of 90 nm in size and 99 percent purity. In addition to other studies that employed the sol–gel process, Singh et al. [101] formulated amorphous silica nanoparticles by hydrolyzing tetraethoxyorthosilane (TEOS) in ethanol. The alkaline extraction process was also used to create nanostructured amorphous silica from RHA using 2.0, 2.5, and 3.0 N NaOH solutions. The treatment of 2.5N NaOH resulted in 90.44% silica concentration for RHA [102]. Furthermore, by employing the alkaline sol–gel technique with NaOH, silica nanoparticles were produced from Vietnamese RH in another study. H_2SO_4/water/butanol at pH = 4 was used to precipitate silica. The process was used to successfully produce silica nanoparticles with a high specific surface area [40]. Awizar et al. [103] employed an alkaline extraction procedure to make silica nanoparticles, which were used as green corrosion inhibitors. Using nitric acid (65%) and sodium hydroxide, Adem et al. produced spherical silica nanoparticles from RH. The ash generation in this technique did not require any calcination [82, 103, 104].

Applications

Many industries, such as the building and agricultural industries (Fig. 7), use silica as a precursor ingredient in their production processes. Amorphous silica is utilized in a variety of applications, including but not limited to food additives, absorbents, catalysis, bioactive implants, drug delivery systems, nano-electronics, and sensing systems (Table 2).

The following is a list of some of the uses that silica nanoparticles can be put to:

Adsorption

Adsorption is a common procedure for treating colored effluents including methylene blue (MB); silica-based materials are commonly utilized in adsorption processes [105–107]. For protein adsorption and separation, silica nanoparticles are excellent. The creation of hydrogen bonds and mild electrostatic repulsion forces are the primary factors that influence the adsorption of nucleic acid (DNA) onto the surface of silica nanoparticles. This technique is considered to be cost-effective and is used for the detection of nucleic acid (DNA).

Fig. 7 A field of application of silica

Table 2 Different domains of silica and its nanoparticles application

Domain application	Uses	References
Medical field	- Toothpaste to remove tooth plaque - Bactericide field - Bio-imaging application - Biomedical application - Pharmaceutical industry for making tablets - Food additive	[119–124]
Chemical engineering	- Adsorption	[105–108]
	- Catalysis	[117, 118]
Agriculture	- Pesticides - Agriculture and food industry - Fertilizers - Water purification	[121–135]
Building material	- Building industries - Ceramic use - Adhesive and sealer - Material packing - Textile industry - Produce concrete - Production of glass	[136–142]
Nano-electronics	- Electronic applications - Semiconductors	[143–147]

Application of Silica in Catalysis

The current chemical, pharmaceutical, and petrochemical industries heavily rely on catalysts [108, 109]. Catalysis has become a strategic subject of attention over the last few decades. Consequently, a lot of effort has gone into researching and developing new catalytic materials that are more efficient and stable [110–113]. The subject of catalysis has been extended by significant advances in nanotechnology, particularly in the synthesis of NPs [114–116]. Various high-reactive nano-catalysts have been developed. Over 90% of the chemical, biological, and energy industries rely on catalysts [117, 118]. It is better to produce long-lasting and highly active catalysts utilizing elements that are easily available on the planet. This is because these catalysts can be put to use in both industrial and everyday settings. Silica has found good use in catalysis.

Silica Applications in the Medical Field

Silica-based ceramics are currently attracting a lot of attention in the biomedical field. Many compositions of silica-based glasses were the first materials to be considered bioactive, resulting in a burgeoning field of research into novel silica materials for biomedical applications [119]. They can also be used as carriers for delivering genes and medications. The holes of nanoparticles serve as storage areas for medicinal compounds. In addition, in the field of medical imaging, particles of contrast chemicals are encapsulated, such as iron oxide, quantum dots, organic dyes, and gold nanoparticles [120].

Applications of Nano-Silica in Agriculture

Nano-scale silicon particles have unique physicochemical features with wide beneficial uses in many fields, including agricultural applications. Abiotic and/or climate change-induced agricultural damage can be mitigated by Si-NPs because of their unique properties [121]. In the agricultural sector, Si-NPs were applied as a weapon against heavy metal toxicity [122], UVB stress [121], salinity stress [123], and dehydration [124], among other applications. Si-NPs can also be used as fertilizers, insecticides, and herbicides. This means that Si-NPs have the potential to enhance crops in order to ensure long-term agribusiness viability and stability.

Si-NPs as Pesticides

In the recent decade, nanotechnology has contributed to the production of disease-free industrial crops [125]. Si-NPs have been suggested for use as nano-pesticides in several studies [126, 127]. Two ways in which Si-NPs can be used are as field pesticides that kill insects and larvae and as nanocarriers that release industrial pesticides to increase their efficacy [128–130].

Si-NPs Fertilizers

Because of their unique physical and chemical features, silicon nanoparticles (Si NPs) can swiftly penetrate plant cells and alter their metabolism, influencing plant growth and production through a variety of interactions. Plant productivity can be increased by combining nano-silicon dioxide with organic fertilizer [131]. Mesoporous silica nanoparticles (MSNs) with a specified pore size (2–10 nm) were found to be an effective delivery system for urea, boron, and nitrogenous fertilizers [132, 133]. Si-NPs can thus be used as a stand-alone fertilizer for specific crops as well as a delivery system for herbicides and fertilizers in plants.

Application of Silica Nanoparticles in Water Purification

In most cases, the removal of biomaterials is accomplished by utilizing silica (SiO_2) nanoparticles [134]. Traditional designs that do not include nanoparticles are not very effective. The antiviral activity of silver nanoparticles combined with silica nanoparticles was superior to that of MS2 in deionized, ground, surface, and tap water. The antibacterial qualities of particles made of Ag_3O-SiO_2 are affected by both temperature and the amount of organic matter present in the environment. It is possible to modify these particles such that they operate as a filter for water, removing viruses that have a detrimental effect on the environment [135].

Application in Building Material

As silica is present in the crust of the planet, it can be found in a significant amount in many building materials. There are many different types of materials that include silica; some of these include asphalt, brick, cement, concrete, drywall, grout, mortar, stone, sand, and tile. Silica nanomaterials, also known as silica nanoparticles, are utilized as remarkable building blocks in several applications in material science. These applications include the production of a number of valuable materials [136–138]. There has been a great deal of research done on silica nanomaterials and their use in a wide range of applications due to the unique properties they possess, which include low toxicity in combination with high biocompatibility, ease of surface modification, stability, and cost-effectiveness [139–142].

Conclusions

Silica is found in many agricultural by-products, with some having a significant amount of silica in their matrix. Burning the material, followed by treating it with a chemical or acid-based solution, can be used to separate the silica from the matrix. The rice husk ionic liquid can be used to extract silica nanoparticles as well. Rice husk, maize cob, wheat straw, grasses, and sugarcane waste are examples of agricultural by-products that can serve as renewable sources of silica. Silica nanoparticles can be used to make polymer nanocomposites, biosensors, and catalysts, as well as agricultural, environmental, and coating applications. Silica is abundant in many agricultural by-products. This study provides the essential basis for research into developing sustainable novel technologies and applications of silica in science and technology.

We anticipate that green silica-polymer nanocomposites will soon find a unique application because of their great characteristics. Due to the optical characteristics of the metalcore, silica-polymer nanocomposites, for instance, may be employed for a variety of diagnostic approaches. Due to the carrying capacity of silica and the special characteristics of a metal core, silica-polymer nanocomposites have a promising future in the development of cutting-edge imaging-guiding treatments. Theranostics, which may be a future trend in medicine, may be made more successful by combining many modalities into a single system thanks to the silica layer-metalcore nanostructure. Silica-polymer nanocomposites can be combined with other natural polymers to build innovative nanostructures with exceptional and brilliant characteristics for certain biological applications.

In contrast to those made using chemical methods, the biomedical use of silica-polymer nanocomposites generated using green methods has not received as much attention. Particularly, the majority of investigations have ended in in vitro studies, and just a small number have continued with in vivo trials. Further investigations on large animal experiments and clinical trials are advised to assess the efficacy of silica-polymer nanocomposites made using a green technique.

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Data Availability Data are available in the manuscript.

Declarations

Conflict of Interest The authors declare no competing interests.

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Highlights - Silica has distinct physiological properties in nanotechnology applications.

- Some novel methods for the formulation, safer, more affordable, and more ecologically friendly silica.
- Innovative methods for obtaining silica (SiO2) and its nanoparticles from the ashes of agricultural waste for use in nanotechnology.
- Processes involved in isolating silica from agricultural waste.