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Recent Progress of Rice Husk Derived Silica for Industrial Applications

Komrojee Haripriya Gaayathri [1], Ranjit Debnath [1], Manas Roy [1], Mitali Saha [1,*]

[1] Department of Chemistry, National Institute of Technology Agartala, Tripura – 799046, India
* Correspondence: mitalichem71@gmail.com (M.S.);

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Abstract: In the last few years, the valorization of rice husk (RH) to produce inorganic silica has drawn considerable attention as an important research topic. However, precise morphological control, manufacturing processes, complex extraction, and the use of toxic chemicals for production require comprehensive solutions for the real use of RH in industries. The technology for producing high-value-added silica for application-oriented processes or the eco-friendly technique for producing silica or silicon derivatives needs to be more facile for their practical industrial applications, which is yet to be explored. In this competitive market, RH and its ash (RHA) can benefit industrial sectors in various ways if utilized efficiently. This review paper was mainly focused on the extraction of silica from rice husks and the utilization of this rice husk-derived silica for different kinds of commercial applications. The systematic findings of their synthesis and the possible applications can contribute to the development of rice husk-derived silica, potentially transforming this agricultural waste material into a value-added material.

Keywords: rice husk; rice husk ash; silica; synthesis; applications.

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1. Introduction

Rice husk (RH) is one of the major agricultural wastes containing large quantities of silicate. It is a source of various silicon compounds, such as silicon nitride[1], silicon tetrachloride[2], silicon carbide[3], zeolite[4], and pure silicon[5]. It produces an unusually high amount of ash compared to other biomasses, which causes a great threat to the surrounding environment, causing damage to the land. During rice refining, its removal creates disposal, handling, and transportation problems due to its low density. Therefore, using RH and its ash (RHA) commercially and industrially may be the only solution to the disposal problem. Despite the low cost and easy availability of RH and RHA worldwide, their application is very limited, so a tremendous scope is observed to use these materials for upcoming industrial processing and technology. A systematic and sustainable approach can give birth to a new commercial and industrial sector of rice husk. Looking at this aspect, this review article has shown the research depth and directions of the synthesis methods of rice husk-derived silica and their commercial utilization during these intervening years. It has presented a critical review of the current production techniques of silica from RH, which can solve the global problem of disposal and waste management.

2. Chemistry of Silica to the Modern World

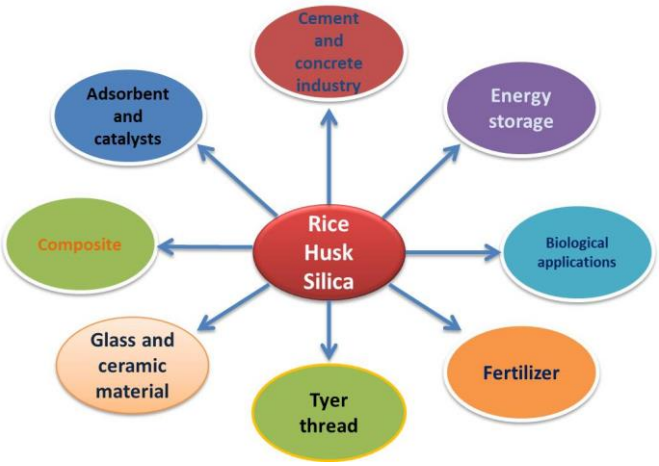
Nowadays, the world is facing a major problem of the disappearance of natural resources and the management of excess waste produced. With the perspectives of the need for energy and the pollution-free environment of developing and developed nations, the world economy must eliminate these problems by utilizing the wastes during the main streams of production[6,11]. With this realization, different industrial sectors were identified to utilize waste materials in their productions. It is known that around 120 to 130 million tons of rice husk (RH) are generated worldwide annually, which finds no useful applications and hence is often dumped into rivers or burned in open piles on fields. On the one hand, RH can be used as a raw material for producing xylitol, furfural, ethanol, and acetic acid, as cleaning or polishing agents in the metal/machine industry.

On the other hand, rice husk ash (RHA) produced from the combustion of RH possesses high silica (>92%), making it with high economic applicability in industries including electronics, construction, chemicals, ceramics, etc. Interestingly, silica produced from RH has more advantages than silica obtained from quartz sand, as RH-derived silica has fine grains, is more reactive, and can be obtained easier and at a relatively low cost. Since the end of the 20th century, silica was proved to be a major constituent of the modern industrial base. Still, the quality required for high-specification products occurs in a limited number of viable deposits. Regarding its dispersity, it can be available as silica sols, soluble silica, xerogels, aerogels, hydrogels, precipitated silica, etc. In the present era, silica's chemistry/physics/metallurgy must be understood so well that this incredibly useful material can be utilized in multiple industries.

3. Rice Husk Derived Silica for Industrial Applications

The interest in the natural biogenic silica nanoparticles produced from rice husks has rapidly grown in the last decade, so knowing their preparation techniques, biocompatibility, and potentiality in different sectors has become important. In this respect, many studies have targeted this research area in recent years, and interestingly, RH and RHA have been considered progressive raw materials for obtaining advanced materials nowadays. In the year 2020, Swapnil Dharaskar *et al*. prepared silica nano absorbent by keeping crushed RH in a muffle furnace for 5 hr at 700 °C, which was found to be a cost-effective absorbent for the removal of fluorine from water [6]. H. K. Tchakouté and coworkers prepared micro and nanocomposites from rice husk ash, eggshells, and nano-silica (CaO/SiO_2) and used them as additives in the production of geopolymer cement [7]. Silica nanoparticles were synthesized from RH by wet milling technique and were applied in oil recovery techniques to increase the amount of crude oil extracted from an oil field [8]. Muhammad NasirAmin *et al*. developed highly reactive silica from RHA to produce cement mortar having high performance and sustainability compared to control mortar [9]. Ismail Abdulazeez *et al*. prepared aminated silica ($ASiO_2$) from RHA and functionalized it with aminopropyl-triethoxysilane (APTES). The $ASiO_2$ exhibited good performance in separating binary gas mixtures like CO_2/CH_4, CO_2 /H_2, and CO_2 /N_2 and was also used in capturing CO_2 from the atmosphere. It was considered a green source of energy in households [10]. Cornelius Tsamo *et al*. extracted silica from RH by treating it with HCl followed by calcination at 540^oC in a muffle furnace. The efficiency of the silica was very high in removing Rhodamine B, a heavy molecular dye [11]. Durgalakshmi Dhinasekaran *et al.* synthesized silica nanomaterial from RH of sizes 20 and 40 nm to develop a chemotherapeutic agent. 5-Fluorouracil was functionalized with silica nanoparticles intermediated with chitosan

and showed good anticancer activity [12]. The RH-extracted silica played an important role in the alumina matrix composite, where silica was reinforced in the alumina matrix hybrid composite. Hybrid composites' chemical and physical properties increased significantly to be used in various advanced applications, including engine pistons, microelectronic devices, and other automobile components [13]. Venkata Rao Madduluri and coworkers reported that RH-derived silica and carbon composites were used to support Nickel (Ni) catalysts to achieve good activity in the hydrogenation of furfural and levulinic acid [14]. Sang Mun Jeong developed silicon nanowires from RH for their application as an anode in high-performance lithium-ion batteries [15]. Mohamed S. Hasanin extracted porous silica from rice husk to prepare an eco-friendly material based on polyvinyl alcohol as a polymer binder and functionalized it with nicotinic acid. This acted as an absorbent for wastewater treatment from the textile industry [16]. Scheme 1 showed the various applications of Rice Husk silica.



Scheme 1. Applications of Rice Husk silica.

G. Suresh Kumar *et al*. synthesized tin oxide-functionalized with amorphous silica nanostructures (SnO_2@RH-SiO_2) where RH template was used as a silica source and found to have potential applications in energy storage devices and electrochemical supercapacitors [17]. Silica nanoparticles derived from RH and doping with calcium peroxide acted as a good absorbent of fluorine in groundwater [18]. Ekrachan Chaichana and coworkers reported RH-derived silica as a good supporter of zirconocene for ethylene polymerization, and it contains lower amounts of impurities compared to commercial silica [19]. D P Ubit *et al*. synthesized a mesoporous silica composite with activated carbon derived from RH and coconut shells, which were utilized as an absorbent of methyl orange dyes [20]. K. R. Sunaja Devi *et al*. synthesized a catalyst by impregnating CeO_2 and Sm_2O_3 on RH-derived silica to convert lignocellulosic biomass levulinic acid to n-butyl levulinate [21]. Suresh K.Bhargava *et al*. developed a SiO_2-Cu-Al-Mg catalyst from RH-derived silica for one-pot synthesis of furfural and furfuryl acetate [22]. Manaswini Behera *et al*. fabricated low-cost ceramic membranes from silica by blending them with soil for their applications in cation exchange membranes in microbial fuel cells (MFCs) [23]. Kasumba A.Buyondo and coworkers reported that the Geopolymers/geopolymer mortar formulated with RH-derived silica and metakaolin in the presence of an alkaline activator is expected to have a high compressive strength [24].

In 2021, Jalil Zulkarnain *et al.* synthesized a dual catalyst, namely natural silica extracted from RH, and nickel for hydrogen storage [25]. Woon ChanChong and coworkers have modified the polysulfone membrane by fusing carbon dots and using silica as additives

for dye removal. Both SiO_2 and carbon dots were extracted from RH to form CD/ SiO_2 composite [26]. Soran Kamari *et al*. extracted pure silica to produce magnetic mesoporous MCM-41, which was used as a dye removal agent and an absorbent [27]. Qing Liu *et al*. prepared nickel phyllosilicate using the hydrothermal reaction of RH-derived silica and nickel nitrate to be used as a catalyst for CO_2 methanation [28]. K. Ganesan *et al*. first developed nitrile butyl rubber-based hybrid composites, then increased the tensile strength and hardness of the composites using RH-derived silica and cotton fabric as filler materials [29]. B.M.Faroldi and others synthesized Ni and Ni-Ru catalysts by wet impregnation method over SiO_2 derived from RH; these catalysts achieved a good methane production rate with the lowest metal loading [30]. Silica derived from RH coated with chitosan was used for Cr(VI) adsorption [31]. Charitha Thambiliya-godage *et al*. successfully synthesized RH-derived silica functionalized with 3- aminopropyltriethoxysilane for effective adsorption of methylene blue dye from the aqueous solutions [32]. Sonchai Intachai and coworkers synthesized phosphorous-adsorbed silica from rice husk for its application in controlled-release fertilizers [33]. Yishan Song *et al.* fabricated a novel sedimentary rock (SR-Fe_3O_4) with RH-derived mesoporous silica for its application in the removal of Arsenic (III) [34]. It was observed that RH-derived silica possessed the highest catalytic rate for methane decomposition, and it was found to be an interesting route for hydrogen production, free from CO_2 emissions [35]. Youqu Zheng *et al.* extracted high specific area nano silica from RH by simple heat treatment and applied it to prepare acetic acid steam reforming catalyst (Ni/RH-SiO_2) [36].

Streptozotocin (STZ) damages the pancreatic β cells, resulting in hypoinsulinemia and hyperglycemia. It was found that RH-derived silica liquid helped in protecting the pancreatic cells from oxidative damage [37]. Violeta-Carolina Niculescu and Maria Simona Raboaca developed an MCM-41 catalyst from RH-derived silica for its application in removing hazardous organic dyes from water in the dye industry [38]. Mohammad Hossein Sayadi *et al*. synthesized thiol-modified magnetic mesoporous silica (mag-MCM-41) to remove herbicides [39]. RH-silica can be used as inorganic support to prepare heterogeneous catalysts like iron oxide (α-Fe_2O_3/RH-Silica) [40]. Jinfeng Wang *et al*. synthesized adsorbents like silica carbon composite (RH-SiO_2-C), activated carbon (RH-Ac), and silicon dioxide (RH-SiO_2) to control air pollution by adsorbing hazardous compounds like ammonia [41]. Arsenic has a low concentrated gradient, so it becomes difficult to remove it, but RH-silica loaded with iron oxide was efficiently used as an adsorbent for removing As(III) from water [42]. Both As(III) and As(V) were removed from water by iron-manganese oxide combined with active silica [43]. P.W.Olupot and coworkers reported that RH-derived silica could be a promising agent for producing geopolymers at room temperature [44]. Silica nanoparticles derived from RH acted as a supporting agent for nickel catalyst to improve the yield of H_2 production during methane cracking [45]. Azimi *et al*. synthesized sodium silicate to replace the commercial sodium silicate in a geopolymer mixture and was further used as an activator to produce epoxy-geopolymer cement [46]. Hanif Amrulloh and coworkers prepared a ZSM-5 catalyst from silica for the transesterification reaction of Ricinus communis oil (castor oil). It showed good catalytic activity and converted 96% of Ricinus communis oil to fuel [47]. Phuong LanTran-Nguyen *et al.* synthesized NaX zeolite using RHA without a calcination process, and the results showed high crystallinity and purity [48]. Syed M.Al-Amsyar developed a sulfonated-silica/carbon composite using RH, which acted as a heterogeneous catalyst in converting fructose to methyl levulinate [49]. Siavash Iravani *et al.* studied that RH-derived silicon, as well as silica nanoparticles, possessed diverse applications in the fields of drug delivery, tissue

engineering, regenerative medicine, lightweight aggregates, cancer diagnosis, energy storage, and catalytic applications [50]. M. Nasikin *et al*. produced silica nanoparticles from RH using the sol-gel method, and it showed great potential for industrial applications as superhydrophobic material on the bitumen [51].

In 2022, Zhengshun Wu *et al*. extracted silica using the sol-gel method to synthesize SiO_2–Hac catalyst for hydrogen production in methane decomposition. This SiO_2–Hac showed good catalytic activity, and the conversion of methane increased from 17.01% to 38.83% [52]. Dong Quy Hoang *et al.* synthesized silica, lignin, hemicellulose, and cellulose by a simple biorefining process in the presence of alkaline peroxide followed by acid precipitation and ethanol extraction of RH [53]. Kwanruethai Boonsong *et al*. fabricated anionic waterborne polyurethane (aWPU) using RH-Silica and hydoxytelechelic epoxidized natural rubber followed by modification by poly(diallyl dimethyl ammonium chloride), a cationic polymer [54]. In the petrochemical sector, high-concentration sulfide is generally formed, which can lead to massive fish killings and irritating odors in the atmosphere. It was seen that RH-derived silica has the potential to remove sulfide in the form of sodium sulfide from wastewater [55]. Narendra Pal Singh Chauhan and coworkers replaced carbon black with RH-derived silica and used it as a filler in the tire industry which drastically improved the physical properties of the product [56]. Medyan Riza *et al*. reported that adding SiO_2 to biocomposite material increased the thickness from 0.178 to 0.0205 mm [57].

Nadir Dizge *et al*. extracted silica and optimized the extraction conditions using the response surface methodology. The extracted silica was used in Safranin dye adsorption [58]. Similarly, Amin Salem *et al.* used RH-derived silica nanoparticles to treat tannery wastewater to remove Cr(III) from it [59]. Abdullah Mohamed and coworkers reported the incorporation of RHA and silica fumes as a partial replacement for cement in concrete to cut down the negative impact of alkali-silica reactions [60]. RHA-extracted biogenic nano-silica and commercial silica were used to prepare Al_2O_3 –SiO_2 inks using an aqueous binder, and the in situ mullite foams were fabricated by 3D printing [61]. Nik Akmar Rejab *et al*. extracted nano-sized silica from RH through acid leaching and thermal treatment for its application as an additive/stabilizing agent for capturing CO_2 [62]. Anderson J.Schwanke *et al*. synthesized ITQ-2 and MCM-22 layered zeolites impregnated with cerium from RH-derived silica for total oxidation of benzene [63]. Ahmed H. Sadek and coworkers developed nanofillers from mesoporous silica for its application in optoelectronics. The films exhibited different optoelectronic properties based on silica content. An increase in the silica content resulted in a decrease in film transmittance, leading to an enhanced adsorption coefficient in the UV region [64]. Biplob Kumar Biswas *et al*. synthesized zeolite faujasite using RH-derived silica, which was used as an adsorbent for the removal of Chromium (VI) and Lead (II) [65]. Yusuf Wibisono and coworkers developed inorganic-organic composite membranes from RH-based silica along with a biopolymer chitosan matrix to purify crude biodiesel [66]. V.Sairam *et al.* reported that using nano-silica extracted from RH in Portland, cement can strengthen the mechanical properties and durability [67]. Aflatoxin B_1 is known to be a cancer-causing agent, so amino-grafted magnetic mesoporous silica was prepared using RH-derived mesoporous silica, which was then magnetized, followed by amino-modification with 3-aminopropyltriethixysilane and then used for the removal of Aflatoxin B_1 [68]. Shurong Wang *et al.* synthesized sodium alginate-silica (SAS) composite aerogels from RHA through the sol-gel method and freeze-drying. It showed good adsorption capacity for organic pollutants [69]. Sima Askari and coworkers developed a molecular sieve. SAPO-34 using RH-silica obtained

by calcination and leaching methods and applied as a catalyst for converting methanol to olefin [70]. Betina M. Faroldi *et al.* developed a catalyst Ni5/MCF for CO_2 to CH_4 conversion and a sorbent lithium silicate for capturing CO_2 at high temperatures using RH-derived silica [71]. Vinay Kumar Singh *et al.* synthesized silica-doped tricalcium phosphate from rice husk and animal bone waste for its application in tissue engineering [72]. RH-silica was also used as a silica source to immobilize lipase from Candida Antarctica B (CALB) by sol-gel technique [73]. A simple preparation of CuO.Fe_3O_4/silica composite was carried out using RH-based silica to improve the Fenton-like catalytic degradation of tartrazine in a wide pH range [74]. Rini Dharmastiti *et al.* extracted silica from rice husks using NaOH. Silica was then used to prepare white mineral trioxide aggregate (WMTA) [75]. Juarez Benigno Paes and coworkers reported the silica nanoparticles extraction from RH by acid leaching with hydrochloric acid followed by thermal treatment at 500ºC, 550ºC, and 600ºC. The impregnation of these silica particles on Pinus Elliotti wood was found to increase the biological resistance against xylophagous fungi and termites [76]. RH-based silica prepared through calcination at 700ºC for 2 hr was used to formulate tyer thread and carbon black [77].

Prof. Masaru Tanaka *et al.* synthesized silica gel from rice husks and used them in encapsulating the lipase enzyme. Due to its high thermal stability, the encapsulating enzyme activity test was carried out for the transesterification reaction of palm oil under various conditions. The results indicated the highest conversion of oil into FAME [78]. Thapanee Sarakonsri *et al*. synthesized polyaniline-coated nanosilica@reduced graphene oxide using RH-derived silica, which showed excellent electrochemical performance when used in lithium-ion batteries [79]. RH-silica was also used to develop PVC-coated lignin/silica composites and as an anode in lithium-ion batteries [80]. Harry Agusnar *et al.* synthesized sodium silicate from RH to synthesize silica nanoparticles using the sol-gel method for the adsorption of methamphetamine in water [81]. Zeolites like NaA (RA) and NaX (RX) were synthesized from RH-silica for adsorbing the CO_2 from the atmosphere to control global warming [82]. W.L.Da Silva *et al.* synthesized nanobioglass by sol-gel process using RH-silica and magnesium nanoxide with nanobioglass (MgNPs-BV) for bone prostheses application [83]. Ken-ichi Saitow *et al*. developed orange-red electroluminescence silicon quantum dots from RH, which were used in LEDs development [84]. Amri Amin *et al*. synthesized an ultrafiltration membrane from RH-silica for purification and separating small solutes below 0.01 µm to 1 nm [85]. Piyada Theerakulpisut *et al*. studied the effect of RH- silica on the physiology of rice under salt stress [86]. Dr. Anil Kumar Sahu and co-workers synthesized RHA and Silica fumes for their application in performance-based concrete [87]. Hideya Kawasaki *et al*. prepared rose bengal-impregnated rice husk nanoparticles (RB-RH NPs) for their potential photosensitization and O_2 generation as antimicrobial photodynamic inactivation [88]. Yogalakshmi Kadapakkam Nandabalan *et al*. demonstrated an anode based on rice husk-derived silicon nanoparticles (RH-Si) and recorded the maximum power and current density of 190.5 mW m^{-2} and 1.5 A $m^{-2,}$ respectively [89]. Kuen-Song Lin and coworkers extracted silica nanoparticles from rice husk and effectively investigated the inhibition of liver cancer cell proliferation [90]. Overall, it was found that RH-derived silica has shown great potential for different applications in the recent few years, and this has to be further explored at real scales, i.e., for industrial applications in the future.

4. Conclusions

As agricultural waste, RH is abundantly available worldwide and hence requires exploration of how to manage waste pollution. Since the disposal systems were not efficient and environmentally friendly, the research focused on sustainable RH waste management strategies to produce silica or silicon products from it to enhance the socio-economic conditions and industrial developmental growth. The present chapter highlighted a systematic study of the scientists and researchers regarding the important aspects of silica production from RH. This review was focused mainly on the different silica extraction techniques from RH and potential commercial applications for the upcoming industrial sectors.

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Conflicts of Interest

The authors declare no conflict of interest.

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