Rice husk/rice husk ash as an alternative source of silica in ceramics: A review

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ABSTRACT
Use of waste or by-products from different industries and the agricultural sector has received increasing attention in the scientific, technology, ecological, economic and social spheres in recent years. Rice husk (RH) is a by-product of rice milling and rice husk ash (RHA) is generated by combustion in a separate boiler. Both RH and RHA are abundantly accessible in rice growing countries such as China, India, Brazil, the USA, and Southeast Asia. RH has therefore been recycled by burning it for energy production. This generates RHA, which contains a huge quantity (85–95%) of amorphous silica. Over the past two decades, RHA has been used extensively in numerous fields for manufacturing of different silicates, zeolites, catalysts, nanocomposite, cement, lightweight construction materials, insulators, and adsorbents. This paper presents a comprehensive overview on the processing of nano-silica from RH/RHA. It tries at the same time, to present a critical review of the application of RHA as an ingredient for the production of various ceramic materials, e.g. refractory, glass, whiteware, oxide and non-oxide ceramics, silica aerogel and SiO₂/C composites. In summary, amorphous silica derived from RHA or RH provides a potential alternative to conventional silica sources (e.g. quartz) for the manufacture of value-added ceramics for practical applications.

1. Introduction
Today, the whole world is suffering from two types of problem, i.e. disappearance of virgin resources and the production of excess waste. From the perspectives of the economies, energy needs, and environments of developing as well as developed nation, the only way to eliminate these problems is to utilize these wastes in the main streams of production. In accordance with this realization, every production sector is looking to raise its income by using waste as a resource in its products. Similarly, the ceramics industry is also seeking to utilize waste in its productions. Much research has been conducted in last few decades to achieve this goal of the ceramics industry [1,2]. As a result, some waste has been identified as potential ingredients for use in the production of ceramics, e.g. fly ash [3,4], bottom ash [5], blast furnace slag [6], glass waste [7], petroleum waste [8], polished tile waste [9], paper-processing residues [10], waste marble powder [11], water treatment sludge [12] and oil production waste [13].

Ceramics are solid compounds of inorganic, non-metallic and metalloid atoms bonded primarily by a mixed type of bonding comprising of ionic and covalent bonds. Richerson et al. [14] defined ceramics as “most solid materials that aren’t metal, plastic, or derived from plants or animals.” Ceramics have some distinctive properties not offered by metals, including good chemical inertness, high-temperature stability, brittleness, a high melting point, and an electrical insulation capability. Due to these properties, ceramics offer a wide range of applications for modern society. In a broad sense, ceramics can be regarded as refractories, glass, tiles, sanitary ware, tableware, and ceramics for electrical applications [15]. In all these industries, silica (SiO₂) is the most important ingredient for production. Due to the massive use of silica in ceramic products, silica is believed to be the backbone of the ceramics industry [16]. Most manufacturers use silica sand, gravel, sandstone, granite, quartz, and quartzite as a silica source for the manufacture of ceramics [17]. All these sources of silica are found in nature. With the use of these raw materials, our problem remains the same, i.e. a loss of virgin raw materials. Therefore, the whole world is looking toward waste utilization to obtain silica. RHA is found to be the most promising waste to serve as a potential silica source.

Rice is the second most widely consumed food item globally, with rice paddy production registering about ~ 758 million metric tons in 2017, a number that will increase gradually due to the projected demand of the world population [18]. The statistics for rice paddy production in different countries according to production volume in 2017 is shown in Figure 1 [19]. China and India are the leading countries for rice paddy production, accounting for around 48.87 wt.% of the total volume of rice paddy production. Rice husk (RH) is a by-product of the industrial processing of rice and approximately 20 wt.% of bulk grain weight. The main constituents of...
RH are 70–80% organic substances such as cellulose, lignin, etc., and the remaining 20–30% comprise mineralogical components such as silica, alkalis and trace elements [20]. Due to its high calorific value (16,720 kJ/kg), most can be used as fuel in boilers for energy production through direct combustion or by gasification [21]. This burning generates new waste, designated as rice husk ash (RHA), which is roughly 25% of the initial husk weight and which causes environmental pollution as well as disposal problems [22]. Therefore, 1000 kg of paddy grain produces about 200 kg (20%) of RH, and when it is burned to generate energy, about 50 kg (25%) of RHA is generated, a volume containing around 45 kg (85–95%) of amorphous silica. The properties of RHA depend on the ecological circumstances of its origin as well as the process applied for burning the husk [23]. It has a high silica content in the amorphous form, which has broad applicability in industries including ceramics, construction, chemicals and electronics among others [24]. Consequently, numerous investigations have been carried out to benefit from this aspect of RHA’s applications.

Prasara-A and Gheewala [25] discussed sustainable use of RHA on a “triple bottom line” basis, i.e. economic, social and environmental. However, the technical viability or performance of RHA in different sectors is not covered by this study. Pode [26] extensively discussed RHA generation in different countries and its applications for advanced materials fabrication in several fields. Soltani et al. [27] spotlighted the applications of RH in different fields and discussed the uses of RH silica to synthesize some advanced non-oxide ceramics, silicon, and nano SiO₂, and their different fabrication processes as well as the process parameters. Liu et al. [28] elaborated the pozzolanic and adsorbent characteristics of RHA in a study. Shen (China) [29] reviewed current growth in the preparation of RH silica and its applications in environmental and energy functional materials. Shen et al. (Japan) [30] extensively discussed RH-derived porous silica and carbon and their application in the synthesis of zeolite materials, non-oxide ceramics, silica aerogels (SAs) and mesoporous or hierarchical carbon. As per our survey of the literature, no review articles were found that contain a wide discussion of “RHA as a replacement for silica in different areas of ceramics.” This study presents a comprehensive summary of recent progress in the use of RHA as an alternative for silica in the manufacture of different ceramic materials, including both traditional and advanced ceramics.

2. Extraction of silica from RH/RHA

Silica is an important raw material for different industries including ceramics. It has a wide range of applications as a raw material in different fields of the ceramics industry. Quartz is a crystalline form of silica, which is mainly used in industry. Recently, the use of amorphous silica is gradually increasing due to some properties distinct from those of crystalline silica, i.e. the greater reactivity of amorphous silica. Fused silica, the main source of amorphous silica in industry, is expensive because it is an industrial product. RH/RHA can solve this difficulty, however due to its enormous active silica content and abundant availability. Therefore, several researchers are investigating economical, eco-friendly, easy ways of extracting high-purity silica from RH/RHA.
Broadly speaking, two types of synthesis methods were reported for extraction of silica, i.e. the chemical method and combustion method. The present review article considers only these extraction methods.

2.1 Combustion method

Direct combustion is the oldest and most widely used method of obtaining energy from biomasses or agriculture residues. This process is used in both domestic and industrial activities; it can be conducted in open fire stoves or different type of boilers, such as fluidized bed, stoker and suspension-fired boilers [31]. Oxygen present in the biomass acts as an oxidizing agent during the oxidation reaction of burning, which is an exothermic reaction. In the burning process, biomass is oxidized by the combustion, resulting in heat energy and a combustion product (ash). Heat produced in this burning is used to generate steam or reused later for drying operations through heat exchangers. The high-temperature and high-pressure steam drive the blades of a turbine that produces electricity. 90 wt.% of RH is used as a fuel to generate energy through a combustion process [32,33], moreover, due to the high energy potential of RH of around 15 to 18 MJ/Kg [34]. After combustion of RH, about ~ 20 wt. % of ash known as RHA is generated. RHA contains more than 85 wt.% of silica in its composition. The chemical composition of RHA, which was analyzed in different studies, is shown in Table 1 [35–37]. The composition variations of RHA depend mainly on the type of soil, weather conditions, agronomic handling and other parameters [37,38]. Typically, RHA containing silica is the amorphous form found in nature as verified by the XRD diagram, i.e. the absence of any crystalline peak, shown in Figure 2 [35].

Gomes et al. [36] discussed bubbling fluidized-bed combustion (BFBC) for RHA generation and the different parameters such as fluidization velocity, combustion temperature and elutriation behavior, which influence the characteristics of silica as well as its purity. A schematic diagram of RHA generation through BFBC is presented in Figure 3. The particle diameters are improved with increasing combustion temperature and fluidization velocity from 0.30 to 0.50 m/s. Bakar et al. [39] synthesized high-purity amorphous silica by combustion after acid treatment of RH. The purity of silica increased due to acid leaching of RH from 95% to around 99%. Subsequently, the specific surface area improved significantly with acid treatment. Sankar et al. [40] used the same process for the extraction of nano-silica powder from RH. They used acid treatment after the formation of RHA; however, it annealed again at 700°C for 2 h. Another researcher also performed acid treatment of RH but in a controlled atmosphere, i.e. high pressure (2 kgf-cm$^{-2}$) and temperature (150°C) [41]. Gu et al. [42] discussed the kinetics of various pretreatments of RH before calcination deeply, i.e. water soaking, acid leaching and grinding. These pretreatment processes greatly influence the removal of metallic impurities and decomposition of organic matter in the

| References          | Bhardwaj et al. [35] | Gomes et al. [36] | Van et al. [37] |
|---------------------|----------------------|------------------|-----------------|
| SiO$_2$             | 92.810               | 86.0             | 87.40           |
| Na$_2$O             | 2.658                | 0.05             | 0.04            |
| Al$_2$O$_3$         | -                    | 5.12             | 0.40            |
| P$_2$O$_5$          | 1.071                | 0.48             | -               |
| K$_2$O              | 1.021                | 1.82             | 3.39            |
| CaO                 | 0.417                | 1.26             | 0.90            |
| Fe$_2$O$_3$         | 0.312                | 1.12             | 0.30            |
| MgO                 | 0.212                | 0.48             | 0.60            |
| SO$_2$              | 0.132                | 2.79             | 3.39            |
| TiO$_2$             | 0.112                | 0.17             | -               |
| Others              | 1.255                | 0.71             | 3.58            |

Figure 2. XRD image of RHA.
preparation of amorphous silica. The characteristics of powders (purity, surface area, and particle size) prepared by different researchers using different techniques are demonstrated in Table 2.

### 2.2 Chemical method

High-purity nano-silica has recently found wide-ranging applications in different fields such as pharmaceuticals, dyes, chromatography, drug delivery systems, electronic components, catalysts and adsorbent materials [43,44]. As a result, demand for high-purity silica is increasing. Combustion-derived RHA (without acid or alkali treatment) contain less than 95 wt.% of SiO$_2$, and the remaining part comprises different alkali oxides and impurities. With appropriate acid or alkali treatment of RH/RHA, however, the SiO$_2$ content can be increased in the system to more than 99 wt.%. Several researchers have therefore adopted different chemical routes to derive high-purity nano-silica.

Zulkifli et al. [45] extracted silica particles from RH by the alkali extraction method using the process shown in Figure 4. Initially, RHA was treated with HCl in a water bath at 75°C for 4 h to remove metallic impurities. Filtration was done by repeatedly washing it with DI water until the pH reached 7, and then drying it at 110°C for 12 h. The sample was reacted with NaOH to prepare sodium silicate solution with constant stirring for 1 h at 90°C. The sodium silicate solution was then reacted with ethanol, and water was added with constant stirring for 10 min. The whole mixture was titrated with 3 M H$_3$PO$_4$ till gel formation occurred. Centrifugation of the yellowish gel was conducted, and the product was washed with DI water to remove any excess sodium silicate and phosphate, followed by calcination to obtain silica particles. The following formulas can represent the overall chemical reaction:

$$\text{SiO}_2(s) + 2\text{NaOH}(l) \rightarrow \text{Na}_2\text{SiO}_3(l) + \text{H}_2\text{O}(l) \quad (1)$$

$$3\text{Na}_2\text{SiO}_3(l) + 2\text{H}_3\text{PO}_4(l)$$

$$\rightarrow \text{SiO}_2(s) + 2\text{Na}_2\text{PO}_4(l) + 3\text{H}_2\text{O}(l) \quad (2)$$

Santana and Paranhos [46], and Zulfiqar et al. [47] extracted silica by the same method with some modifications. They extracted silica from RH in two steps: (i) conversion of RH into RHA; and (ii) formation of amorphous silica. They used concentrated H$_2$SO$_4$ and H$_3$PO$_4$ for precipitation of SiO$_2$ particles from silicate solution. The properties of the silica particles are tabulated in Table 2.

Song et al. [48] used the Taguchi method for extraction of silica from RH; it consists of two steps: the first comprising the formation of ash from RH, i.e. RHA, and the second corresponding to production of silica particle form this RHA, i.e. the Taguchi method. Flow charts of the two steps are shown in Figure 5(a) and (b), respectively. To complete the first step, the whole RH was washed and leached to remove impurities followed by screening with one 820 µm sieve. The sieved mass was then washed with deionized water with continuous stirring for 1 h and dried at 70°C to remove adhering dust. The dried mass was then leached with 1 M HCl for 2 h at 90°C. Now the whole sample was rinsed repetitively

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**Table 2. Properties of extracted silica by different method.**

| Process                            | BET surface area (m$^2$/g) | Particle size | SiO$_2$ (%) | Ref.               |
|-----------------------------------|---------------------------|---------------|-------------|--------------------|
| Fluidized bed combustion          | Not mention               | 750 µm        | 86.00       | Gomes et al. [36]   |
| Acid treatment before combustion  | 208–218                   | 0.50–0.70 µm  | > 99        | Bakar et al. [39]  |
| Acid treatment after combustion   | 200–247                   | 10–20 nm      | Not mention | Sankar et al. [40] |
| Acid treatment (control atmosphere) before combustion | Not mention | 181–294 nm | Not mention | Carmona et al. [41] |
| Alkali extraction                 | 364                       | 75–252 nm     | Not mention | Zulkifli et al. [45] |
| Alkali extraction                 | 173–294                   | Not mention   | 96.35–98.34 | Costa and Paranhos [46] |
| Alkali extraction                 | 200–339                   | 63–170 nm     | 95.9        | Zulfiqar et al. [47] |
| Alkali extraction                 | 7.93–740.77               | 24–87 nm      | 95.68–99.89 | Song et al. [48] |
| Hydrothermal                      | Not mention               | 10–50 nm      | Not mention | Tolba et al. [50]   |
| Hydrothermal                      | Not mention               | 15–35 nm      | Not mention | Bathla et al. [51]  |
| Carbonation                       | Not mention               | Not mention   | 99.97       | An et al. [52]      |
with DI water and calcined at 700°C for 2 h. This dried mass was marked as RHA. The second step, extraction of silica by the Taguchi method, is shown in Figure 5(b). RHA was used as an ingredient and mixed with 1.5 M NaOH and heat-treated at 90°C for 1 h. This sodium silicate solution was diluted to 1 M by adding DI water and titrated against 1 M HCl up to pH 7 to neutralize the solution. The sol was stirred until a gel was formed and then dried at 70°C for seven days for aging, followed by centrifugation at 10,000 rpm.

Liou and Yang [49] studied the different parameters of silica production from RHA through the alkali-extracted route. The acid and alkali concentrations, gelation pH, aging time and temperature were optimized for preparation of SiO$_2$ nanoparticles from RHA. The effects of different acids on the surface area and particle size of silica were also evaluated.

Tolba et al. [50] introduced a new technique of synthesizing nano-silica from RH, i.e. the hydrothermal route. A clean 2 g of RH was mixed with 10 mL of nitric acid and 10 mL of distilled water, and the mixture was placed in an autoclave for complete reaction at different temperatures, pressures and times. The resultant product was filtered and washed several times with distilled water to remove excessive acid and dried at 60°C to obtain white nano-silica. Bathla et al. [51] also used a different type of hydrothermal synthesis process to fabricate nano-silica from RH. First, organic acid leach RH was used to produce RHA at 700°C. Then the RHA was mixed with ferric nitrate [Fe(NO$_3$)$_3$·9H$_2$O] in a ratio of 1:6

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**Figure 4.** Extraction method of silica from RHA [45].

**Figure 5.** (a and b). Taguchi method for extraction of silica from rice husk [48].
and calcined at 420°C to obtain a composite powder. 2.5 gm of the composite powder was mixed with 25 mL of water and 40 mL of edamine \([C_2H_8NH_2]\) and autoclaved for 5 days at 200°C. After completion of the reaction, centrifugation was used to separate the product from the solution, and the product was then washed several times with DI water. \(Fe_2O_3\) was removed from the system by repeating the acid-leaching process, and white amorphous \(SiO_2\) was finally obtained. Subsequently, An et al. [52] also developed a new route for preparation of silica from RHA via a carbonation process. Here, \(Na_2CO_3\) was used as a reagent for \(SiO_2\) extraction. However, this route is complicated due to the multiple processing steps, and efficiency is low at around ~72%.

Silica was extracted by different methods, which are included in Figure 6. In conclusion, it was found that silica extraction directly from RH is not economical or efficient (due to its high volume). For economical utilization, RH must first be used as a fuel for boilers, where it produces RHA as waste. Silica extraction from this waste can be done by two routes, which differ from each other in the purity of their end products. If RHA is used directly without any treatment, it gives 83–95% pure silica, while treatment of RHA with acid yields more than 95% pure silica. For high-purity silica, alkali extraction or other chemical methods which give greater than 99% pure nano-silica should be used.

3. Application of RH silica

In past few decades, many researchers have shown an interest in applying RH silica in several fields. The present article has attempted a review of their work with reference to ceramics. Application areas of RH silica in different ceramic fields are illustrated in Figure 7.

3.1 Refractories

Because RHA has low thermal conductivity (\(\kappa\)), it can be used as an ingredient in the manufacture of insulation refractories. The \(\kappa\) of refractory materials is ascribed to vibrating atoms caused by phonons (low temperature) and photon conductivity (high temperature) in crystalline materials. RHA containing silica is amorphous, however, which causes it to act as a non-heat conducting ingredient in refractories [53]. RHA insulation refractories are fabricated by mixing of different flux contents (as binder), plasticizers (due to the lack of plasticity of RHA) and pore-forming agents (to increase the porosity of final products) [23,35,54]. Air is entrapped in the pores of refractories, a characteristic which is attributed to lower values of \(\kappa\), because entrapped air acts as a barrier to the flow of heat. These insulating refractories may be used as a second lining of furnaces or kilns in ferrous or non-ferrous industries. RHA is also used in ferrous industries for the casting of steel and for molten metal insulation in ladles and tundishes [55,56]. This insulation of molten metal prevents cracks in the structure of refractories that can occur due to temperature drops of 200°C when molten metal is transferred from a ladle to a tundish. The low thermal conductivity of RHA also leads to a gradual decrease in the temperature of steel, and moreover to its uniform solidification. In addition, several researchers are trying to use RHA in the production of advanced refractories, such as cordierite [57,58], forsterite [59] and mullite refractories [60].

![Figure 6. Conclusion of different methods.](image-url)
3.2 Glass

Glass is a non-crystalline supercooled liquid. Generally, glass is amorphous; however, “glass ceramics” are a crystalline form of glass. Present-day glass is one of the most widely used materials for innumerable purposes, and the number is increasing tremendously. Silica ($\text{SiO}_2$) is the major ingredient for glass making, acting as a “network-former” in glass networks. Huge silica resources are therefore required for glass industries. For this reason, researchers are trying to find alternative sources of silica. RHA has tremendous potential to fulfill this requirement because it contains more than 90 wt.% of amorphous silica. Lee et al. [61] used RH silica to fabricate photoluminescent (PL) glass by two different routes, i.e. the mixing technique and the layering technique. For the mixing technique, PL pigment and molten glass were mixed directly; for the layering technique, however, PL pigment was sandwiched between two layers of RH glass. This waste-derived glass is analogous to ordinary silica glasses, which should see their applications expand. Glass ceramics have some unique properties lacking in glass, such as mechanical, thermal, optical and electrical properties; these make them useful in other fields besides glass. Andreola et al. [62] and Sharma et al. [63] prepared glass ceramics using RHA as a silica source. Chen et al. [64] synthesized mesoporous bioactive glass (MBG) from RHA using the sol–gel method for application in drug delivery and bone regeneration. The obtained results exhibit excellent biocompatibility with no cytotoxicity in normal cells and MBG-folic acid with camptothecin (a water-insoluble anticancer drug) that destroys the cancer cells efficiently and selectively. It can be concluded that RHA-derived MBG can potentially be used in medical applications. Other researchers have therefore also prepared bio-glass from RHA by the sol–gel method [65].

3.3 Whiteware

Silva and Surangi [66] studied the effects of RHA addition for partial replacement of clay in the composition of clay roof tiles used for roofing buildings. Their study concludes that 10 wt.% RHA addition shows a ~ 45.97% increment in the breaking load, whereas density is reduced by ~ 3.04% and the thermal properties are also improved (indoor temperature decreases ~ 4°C). In whiteware bodies, the substitution of a conventional RHA silica source influences the vitrification temperature of the bodies, which is around 50–100°C [67,68]. Consequently, thermal expansion is reduced and mechanical strength improved at lower temperatures in mature bodies [69]. Bondioli et al. [70,71] analyzed the feasibility of using RHA in place of quartz in pigment and frit development. In the case of pigments, RHA was used to replace silica in Pr–ZrSiO$_4$ solid solution for synthesis of yellow-colored ceramic pigments. The obtained pigment shows the development of a stable, intense yellow color, which is similar to that of a composition containing pure quartz. Similarly, RHA is also applicable for frit manufacturing for use in tiles and glazes.
3.4 Oxide ceramics

3.4.1 Mullite ceramics
Mullite is an aluminosilicate refractory material with various attractive properties, such as low thermal expansion, a high melting point, high thermal stability, high thermal shock resistance and chemical stability [72,73]. Due to these properties, mullite is a potential material for use in traditional as well as advanced ceramics. It is the only stable crystalline phase in $\text{Al}_2\text{O}_3$–$\text{SiO}_2$ systems with the chemical formula $3\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$ [60]. The method of synthesis drastically affects mullite phase development, the mullitization temperature and its properties, because it controls the degree of mixing of raw materials. As mullite is a mixture of alumina and silica, so RHA can be a potential raw material for use as a silica source in its development. For this reason, many researchers have sought to use RHA as a raw material for mullite over the past few decades. The contribution of RHA to mullite development is a two-step process: the first step is extraction of silica from RHA and the second is application of the silica. To accomplish the first step, different methods have been used viz. alkali extraction, as discussed in Section 2.2. The second step was accomplished by different methods. Sembiring et al. [74] used a gel-derived method for development of the mullite phase. They prepared silica sol by treating extracted silica with 5% KOH solution, followed by the addition of this sol to aluminium sol in an appropriate molar ratio of 3:2. HCl was added to the mixture slowly to produce gel, and the gel was then aged for 3 days followed by rinsing with distilled water to remove excess alkali and acid content. The rinsed mass was dried at 110°C for 12 h and fired at 800, 950, 1050, 1150, 1250, and 1350°C. Serra et al. [60] used a reaction-sintering method for development of mullite. They mixed RHA and an alumina source in adequate amounts and ball-milled the mixture to achieve proper mixing of the raw materials. This mixed mass production was followed by pellitization and sintering of the pellets at 1100–1600°C. Other researchers also used the reaction-sintering method moreover, for development of the mullite phase with RHA and other wastes [75]. In both approaches, i.e. the gel-derived and reaction-sintering methods, mullite phase formation increased with the firing temperature. With the gel-derived method, mullite was formed at 1350°C with a low amount of secondary phases compared to lower temperatures, i.e. 950–1250°C. Similarly, for the reaction-sintering method, secondary phases such as cristobalite and pure alumina phase were found for almost all temperatures but their intensities decreased with temperature. This can be attributed to incomplete reaction between silica and alumina at lower temperatures [60,74].

3.4.2 Cordierite ceramics
Cordierite is a magnesium aluminosilicate ($\text{Mg}_2\text{Al}_4\text{Si}_2\text{O}_{10}$) material with excellent thermal, electrical and chemical properties. It has a very low thermal expansion coefficient, i.e. $< 4 \times 10^{-6}/°\text{C}$ [76–79]. Cordierite also has a very low dielectric constant and high melting point; moreover, i.e. 1460°C. Due to these properties, researchers have found cordierite to be a potential material for use in gas turbines, industrial and lab-scale furnaces, and electronics packing materials, etc. Other uses for cordierite include integrated circuits due to its low dielectric constant [80,81]. As stated above, cordierite is a magnesium aluminosilicate system, and RHA can therefore be used as a silica source for cordierite. A few researchers have conducted studies in this area in the past two decades. Simbering et al. [57] used an alkali extraction method for extraction of silica from RHA followed by solid-state mixing of the raw materials for development of cordierite. For the later step, i.e. development of cordierite, they mixed all the raw materials in an alcohol medium in an appropriate ratio of $\text{MgO}$: $\text{Al}_2\text{O}_3$: $\text{SiO}_2$ i.e. 2:2:5. Mixing was conducted using a magnetic stirrer for 6 h in an alcohol medium. Upon completion of the mixing process, the mixed mass was filtered and dried at 110°C for 8 h to absorb residual alcohol. The dried mass was palletized using uniaxial hydraulic pressing and sintered at temperatures of 1050–1350°C. Similarly, Kurana et al. [78] used solid-state mixing of raw materials for development of the cordierite phase. They used RHA in place of kaolinite for development of cordierite. The milling was conducted in a planetary ball mill, however, and the medium was water. A sample was palletized at 1.96 MPa and sintered at 950–1350°C. The ramp rate for temperatures up to 1000°C was 5°C/min and 2.5°C/min for higher temperatures with a holding time of 1 h. Naskar et al. [82] used the sol–gel method for development of cordierite. They first prepared separate sols of silica and other ingredients and then mixed them. The mixtures were dried at 90°C, followed by calcination at 400–1400°C. In all three studies, formation of α-cordierite occurred at temperatures above 1200°C. μ-cordierite was formed at lower temperatures, i.e. below 1200°C, but the transition from μ-cordierite to α-cordierite occurred only at above 1200°C. The formation of μ-cordierite at lower temperatures indicates that a diffusion reaction between cristobalite and spinel ($\text{MgAl}_2\text{O}_4$) starts at this temperature [83]. Moreover, At high temperatures > 1200°C this μ-cordierite starts converting into α-cordierite, moreover, due to the formation of an Mg–O–Al–O–Si bond during phase transformation [57,82,83]. It can therefore be observed that active silica present in RHA promotes conversion of μ-cordierite to α-cordierite at low temperatures (1300°C) compared
to other conventional sources (1400°C) due to its high surface area and fluxing properties. Besides this, application of RHA leads to a decrease in the activation energy required for crystallization of α-cordierite [78].

### 3.4.3 Lithium alumino-silicate

Lithium alumino-silicate (LAS) is a class of glass ceramics with extraordinary thermal and chemical properties. It has a negligible thermal expansion coefficient and high thermal and chemical durability [84,85]. It is very low thermal expansion makes it a suitable candidate for use in equipment requiring structural stability, i.e. no change in dimensions such as gas turbines. Its application fields also include telescope mirrors, cookware, cooktops, etc. In general, LAS has one of two types of crystalline form, i.e. β-eucryptite (Li₂O. Al₂O₃.2SiO₂) or β-spodumene (Li₂O. Al₂O₃.4SiO₂). Chatterjee and Naskar [86] studied the effects of various sources of silica on the properties of LAS by the sol–gel method. They prepared LAS successfully using RHA as one of the sources of silica.

### 3.4.4 Forsterite

Crystalline magnesium silicate, which is known as forsterite, has the chemical formula Mg₂SiO₄. It has a high melting point (1890°C), high chemical durability, good insulation properties, a low dielectric constant, low electrical conductivity and very low thermal conductivity, which make it a potential material for use in dielectric substrates [87], pigments [88], refractory materials [89], etc. Since forsterite is a part of the MgO-SiO₂ system, RHA can be used as a source of silica for its preparation. Mathur et al. [90] prepared nanocrystalline forsterite using RHA as a silica source by a solid-state method. In their study, they observed that some secondary phase of forsterite occurs at below 1000°C but that the pure forsterite phase is obtained at 1000°C. This formation of forsterite at 1000°C can be attributed to the diffusion of magnesia in the enstatite or clinoenstatite phases, which are formed at low temperatures.

### 3.4.5 Wollastonite

Wollastonite is a calcium silicate (CaSiO₃) mineral generally found in two polymorphic forms, i.e. α-wollastonite (pseudo-wollastonite) and β-wollastonite. It has low dielectric loss, high strength, low shrinkage, low volatile constituents, good bioactivity and biocompatibility [91]. Therefore, it has a wide range of applications, such as glasses, electrical insulators, paper, cement, paints, and biomaterials [92,93]. Ismail et al. [94] successfully derived β-wollastonite from RH straw as a source of silica and limestone as a source of lime (CaO). They investigated its bioactivity and biocompatibility for applications as biomaterials and dental implants. An alternative of the sol–gel method was used in their work. Both the ingredients were mixed at an appropriate ratio, i.e. a CaO:SiO₂ ratio of 4:5:5, followed by soaking in distilled water. The mixture was then autoclaved at 135°C for 8 h. Precipitated masses obtained from this step were then dried and sintered at 90°C and 950°C, respectively. Mansha et al. [95] developed acid-resistant calcium silicate with RHA as a source of silica. First, all the amorphous silica was extracted from RHA, and this was then allowed to react with CaO in an excess water medium. Heat treatment was conducted for the temperature range of 500°C to 1100°C. The Si/ Ca molar ratio varied from 1.4 to 2.4. Molar ratios > 2.1 showed high acid resistivity compared to lower ratios. β-wollastonite was obtained at a molar ratio 1.8.

### 3.5 Non-oxide ceramics

#### 3.5.1 Silicon carbide

Silicon carbide (SiC) is a promising non-oxide ceramic for numerous engineering applications, including reinforcement of ceramic and metal matrix composites, catalyst support materials, optic devices, electronic devices and grinding media, due to its unique characteristics such as high hardness and mechanical strength, wear and corrosion resistance, excellent thermal stability, wide band gap, chemical inertness and unique optical properties [96–98]. Conventional SiC powder is fabricated by the Acheson method based on carbothermal reduction of a quartz sand and coal mixture at high temperatures (~ 2400°C) [99]. The major problem with the carbothermal process is the high fabrication temperature, long reaction time and bulky grain size of SiC. Large particles reduce the sinterability of the product and require an extra milling process. The particle size of SiC powder can also be controlled by regulating the carbon and silica particle size in the mixture. Finding a more economical and simple fabrication approach is therefore a major challenge for synthesizing SiC. During the past few decades; RH acquired a great deal of attention as an ingredient for preparation of SiC (whiskers and particles). Cutler et al. [100,101] first used RH for the production of SiC. Many modifications have since been made for industrialization of this route through different studies. Recently, microwave heating is utilized to fabricate SiC [102–104]. Different catalyst activities for the formation of SiC have also been investigated in several studies [105]. Some metallic catalysts (Fe, Ni, Cr, Co and Pd) greatly influence the reaction rate between C and SiO₂ in the temperature range of 1200–1600°C, which is attributed to the high production rate with a desirable particle morphology. The RH route seems more promising, however due to low-temperature synthesis process and cheap ingredients. Almost all studies focus on fabrication of SiC from RH in two processing steps: (i) removal of volatile matter from RH (cooking) under a controlled
atmosphere (400–800°C); and (ii) high-temperature heat treatment (> 1300°C) for formation of SiC by reacting between cooked RH containing carbon and silica [106]. The chemical reaction for the formation of SiC from RH can be expressed as follows [99]:

\[
\begin{align*}
\text{SiO}_2(s) + C(s) & \rightarrow \text{SiO}(g) + CO(g) \\
\text{SiO}(g) + 3\text{CO}(g) & \rightarrow \text{SiC}(s) + 2\text{CO}_2(g) \\
\text{SiO}(g) + 2\text{C}(s) & \rightarrow \text{SiC}(s) + CO(g) \\
\text{CO}_2(g) + C(s) & \rightarrow 2\text{CO}(g)
\end{align*}
\]

Most recently, Li et al. [107] developed a new process for synthesizing SiC nanowires from RH silica with no catalysts or protective atmosphere. RHA and phenolic resin were mixed in a high-speed planetary mill with a stoichiometry of SiC. The mixed mass was then placed in a graphite crucible with a graphite cover, and the crucible was placed in an alumina container. The residual space in the alumina container was packed with graphite dust, and the container was locked with an alumina plate. This atmosphere stopped any oxidation of the mass. Calcination of the mixed mass was conducted at 1600°C for 3 h without a protective atmosphere. Afterwards, light-green β-SiC nanowires were acquired from the graphite crucible.

3.5.2 Silicon nitride

Silicon nitride (Si₃N₄) is a non-oxide high-temperature applicable ceramic material. It has some excellent characteristics such as high corrosion resistance, a low thermal expansion coefficient, high strength at high temperatures, higher thermal shock resistance than other ceramics and high creep resistance, which makes it a most promising structural ceramic for high temperatures [108,109]. Many processes have therefore been developed to produce Si₃N₄ powder. Carbothermal-nitridation of silica is the most common and economical method for preparation of Si₃N₄ powder [110]. High-purity fine silica and carbon, which are commercial available are required for this route. The uniform distribution and high reactivity of carbon and silica accelerate the nitridation reaction. The agricultural byproduct RH consequently contains high reactive silica and carbon with good distribution. Thus, RH is a potential ingredient for Si₃N₄ manufacture [111]. Many researchers have therefore investigated carbothermal-nitridation of RH for Si₃N₄ synthesis [112–114]. Pavarajarn et al. [115] deeply investigated the mechanism of the carbothermal-nitridation reaction of RH for Si₃N₄ fiber and whisker fabrication. RH was first pyrolyzed in a horizontal tube furnace with a continuous flow of argon gas (36L/h) at 600°C for 3h. For the carbothermal-nitridation reaction, the pyrolyzed powder was placed on an alumina tray with a 2 mm depth of powder, and the tray was placed in the tube furnace. Heat treatment was conducted for 3 to 10 h in the temperature range of 1400–1470°C with a reaction gas mixture (90% N₂ + 10% H₂) atmosphere at a gas flow rate of 50 L/h. The differently shaped Si₃N₄ and unreacted powder were found in separate locations on the tray. Fibers were formed outside the cavity of the tray and blade-like whiskers formed on top of the unreacted powder. Whisker-shaped Si₃N₄ powder contains a single crystalline α form phase and the fibers contain a mixture of α and β phases. The reaction mechanism of this process is described by the following reaction formulas:

\[
\begin{align*}
\text{SiO}(g) + 3\text{C}(s) + 2\text{N}_2(g) & \rightarrow \text{Si}_3\text{N}_4(s) + 3\text{CO}(g) \quad \text{(xii)} \\
\text{SiO}_2(s) + \text{CO}(g) & \rightarrow \text{SiO}(g) + \text{CO}_2(g) \\
\text{CO}_2(g) + \text{C}(s) & \rightarrow 2\text{CO}(g) \\
\text{SiO}_2(s) + \text{H}_2(g) & \rightarrow \text{SiO}(g) + \text{H}_2\text{O}(g) \\
\text{SiO}_2(s) + 3\text{CO}(g) + 2\text{N}_2(g) & \rightarrow \text{Si}_3\text{N}_4(s) + 3\text{CO}_2(g) \\
\text{SiO}(g) + 3\text{C}(s) + 2\text{N}_2(g) & \rightarrow \text{Si}_3\text{N}_4(s) + 3\text{CO}(g)
\end{align*}
\]

3.6 Silica aerogel

SA is a synthetic nano-porous, ultra-light, structure-controllable, solid material, which possesses excellent characteristics such as super-low bulk density (0.03–0.5 g/cm³), huge porosity (80–99%), a high specific surface area (500–1500 m²/g), low thermal conductivity, catalysis and a low dielectric constant [116–118]. It has a wide range of technological applications, therefore such as in insulation materials for buildings, membranes, pollutant absorbents, microelectronics, catalytic supports, drug carriers and dielectric materials [119,120]. Many reports were found in which SA was successfully prepared from RHA through the sol–gel route [121–125]. In every case, SiO₂ was first extracted from RHA in the form of a sodium silicate (water glass) solution using a sodium hydroxide solution. The sodium ions were then removed by the cation exchange resin method [124,125] or by neutralizing the water glass solution using acid [121–123] to form silica hydroxide. A small quantity of tetraethyl orthosilicate (TEOS) was added to form a gel, and washing was conducted with water and ethanol. The pretreated gel was dried at atmospheric pressure [121–123] or supercritical drying [125] to attain SA. RHA-derived SA exhibits a porosity of about 80–85%, a pore volume of about 0.7–3 cm³/g, a specific surface area of about 950–270 m²/g and bulk density of about 0.3–0.7 g/cm³.
3.7 C/\textit{SiO}_2 composite

RH-derived C/\textit{SiO}_2 composite material is the primary choice as an electrode (anode) material for next-generation high-performance lithium ion batteries (LIBs). This porous composite exhibits superior cycling stability, a good discharge specific capacity and a high rate capability for use in LIB anodes. A sustainable, facile and low-cost composite material, it may have huge potential to replace traditional graphite anodes in LIBs. Wang et al. [126] synthesized the C/\textit{SiO}_2 composite from RH through a one-step heat-treatment process at 900°C in an inert atmosphere and used it as an LIB anode. It exhibits a good initial discharge capacity of 325 mAh.g\(^{-1}\) and this value gradually increases with a number of cycles (485 mAh.g\(^{-1}\) after 84 cycles) due to electrochemical activation of the composite. Another author prepared this composite from RH by a two-stage method [127]. The first step was carbonization, which occurred at 450°C under an N\(_2\) gas flow. After that, the carbonized powdered precursor was calcined at 900°C under an argon/hydrogen mixed atmosphere. Cui et al. [128] fabricated the micro-porous C/\textit{SiO}_2 composite by a conventional carbonization process with the addition of ZnCl\(_2\) as an activating agent in the RH system. As an LIB anode, this porous composite shows 1105 mAh.g\(^{-1}\) of discharge specific capacity with no degradation after 360 charge/discharge cycles at 0.1 Ag\(^{-1}\). Watari et al. [129] synthesized a highly porous C/\textit{SiO}_2 composite by heating RH pellets at 800–1150°C. The pellets were composed of RH powders with different particle sizes and different molding pressures. The results obtained from RH-derived C/\textit{SiO}_2 composite possess a specific surface area of 450 m\(^2\)g\(^{-1}\), a pore size of ~2 nm and porosity of around 87%.

4. Conclusions

Today, by-product and waste management for the development of new products has gained immense interest. In this framework, RH by-product and RHA waste are being considered as renewable and sustainable silica sources because they contain large quantities of amorphous silica. Thus, RHA has high potential as a replacement for conventional silica sources for the making of different ceramics such as refractories, glasses, whiteware, Si\(_C\), Si\(_3\)N\(_4\), mullite, cordierite, wollastonite, SA, and C/\textit{SiO}_2 composite. This review considers the numerous studies that have focused on the synthesis of silica from RH/RHA and its application for production of ceramics. Within the parameters of the objectives of this review, the following significant facts can be drawn:

- The purity, particle size and surface area of extracted silica depend on the extraction route, acid treatment and annealing temperature. The purity of silica obtained from different routes increases with increases in the number of chemical treatments.
- Utilization of RH for extraction of silica is not economically beneficial; however, its conversion to RHA adds a step of energy generation which contributes positively to economic viability.
- RHA can be used for preparation of insulating refractories due to its low thermal conductivity.
- As RHA is a huge source of amorphous silica, it has potential applications in glass formation. Silica acts as a “network former” in glass, with network formation easily achieved in the case of amorphous silica due to its high reactivity. This reason also makes it suitable for use in the preparation of SA.
- RHA can reduce the vitrification temperature of ceramic bodies a capability allowing its use in whiteware bodies up to a certain percent.
- The activation energy required for crystallization has been reduced by application of RHA, which assists in formation of Mullite and cordierite.
- SiC conventionally is prepared by the Acheson method in which quartz and coal are mixed, followed by heat treatment at a very high temperature (2400°C). The addition of RH decreases the formation temperature of SiC, however, since it already contains a homogeneous mixture of carbon and silica. Similarly, RH has tremendous potential for using as an ingredient for preparation of Si\(_3\)N\(_4\) by the carbothermal-nitridation method, as it contains a homogeneous mixture of carbon and silica at the atomic level.

**Highlights**

1. Extractions of amorphous silica from rice husk (RH) or rice husk ash (RHA).
2. Overview about utilization of RH/RHA in ceramic industries for traditional as well as advanced ceramic product manufacturing.

To study the feasibility of RH silica to replace conventional source of silica.

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