Journal of Applied Organometallic Chemistry
2024, Volume 4, Issue 2

Original Article

A Cheaper and Greener Approach for the Synthesis of Highly Pure Silicon Carbide Composites Using Coffee Husk Wastes

Samuel N. Ndung'u[a,*] | Esther W. Nthiga[b] | Ruth N. Wanjau[a] | Anthony M. Ndiritu[c]

[a]*Department of Chemistry, Kenyatta University, P.O Box 43844-00100, Nairobi, Kenya*
[b]*Department of Chemistry, Dedan Kimathi University of Technology, P.O Box 657-10100, Nyeri, Kenya*
[c]*National Phytotherapeutics Research Centre, Kenyatta University, P.O Box 43844-00100, Nairobi, Kenya*

Use your device to scan and read the article online

Citation S. N. Ndung'u, E. W. Nthiga, R. N. Wanjau, A. M. Ndiritu **A Cheaper and Greener Approach for the Synthesis of Highly Pure Silicon Carbide Composites Using Coffee Husk Wastes.** *J. Appl. Organomet. Chem.*, **2024**, *4*(2), 135-144.

https://doi.org/10.48309/JAOC.2024.446126.1171

Article info:
Submitted: 28 February 2024
Revised: 02 April 2024
Accepted: 28 April 2024
ID: JAOC-2402-1171
Checked for Plagiarism: Yes
Language Editor Checked: Yes

Keywords:
Silicon carbide (SiC), Carbothermal reduction, Extracted silica, Biochar, Alkali fusion

ABSTRACT

The extensive use of silicon carbide in many fields such as in abrasives, structural materials, refractories, electronic components, and nuclear reactors amongst others demands for an innovative, cheaper, and an environmentally friendly approach for its synthesis. The aim of the study was to use coffee husk wastes as silica and carbon sources in SiC synthesis. This was done via carbothermal reduction synthetic route using the extracted silica and biochar materials at a temperature of 300 °C for 12 hours. The silica, biochar, and SiC composites were characterized using XRF, FT-IR, XRD, SEM, and EDX. The results for silica showed spherical-shaped granules with Si and O components. The biochar showed a highly amorphous carbon structure with silica and carbon contents. The FT-IR, SEM, and EDX results revealed macron sized SiC composites with Si (59.3 %) and C (28.7 %) components. This cheaper and greener approach makes coffee husk wastes a novel material for the synthesis of highly pure Silicon Carbide (SiC) composites for application in various industrial fields.

Introduction

Silicon carbide is an important ceramic material with vast industrial applications [1]. This is due to its excellent thermal and electrical conductivity, hardness, excellent corrosion and resistance to thermal shock [2]. Because of its unique qualities, it is widely used in several fields such as abrasives, structural materials, refractories, electronic components, and nuclear reactors [3]. With the emerging technological inventions, silicon carbide materials have gained demand in making high-power, high-frequency, high-temperature, anti-radiation, and high-density integrated electronic devices [4]. Some exhibit photoluminescence in the near-Ultra Violet to the visible blue spectral region making them attractive for light-emitting devices [5]. This is because of its unique bandgap width of 2.20-3.26 eV [6]. This has made its synthesis methods greatly explored in the field of materials science. Various methods such as sol gel [7], solvothermal [8], chemical vapor deposition [9], arc discharge [10], and

*Corresponding Authors: Samuel N. Ndung'u (samuelndungu530@gmail.com)

carbothermal reduction [11,12] among others are reported for the synthesis of SiC composites. Carbothermal reduction has however remained the most promising technique due to its cost-effective, efficient and large-scale preparation [13]. In addition, the method is unreliable on hazardous precursors unlike other techniques [14]. In carbon thermal reduction process, raw materials of carbon and silicon sources are reacted at a certain temperature to form SiC. This is as shown in Equation 1.

$$SiO_2(s) + 3C(s) \longrightarrow SiC(s) + 2CO(g) \quad (1)$$

The SiC composite formation occurs even at very low temperatures via carbothermal reduction compared to the other reported methods [3]. Various agricultural wastes such as corn cob [15,16], macadamia nuts [17], rice husk [3] amongst others are reported in the SiC synthesis. The study sought to explore the use of coffee husk wastes as carbon and silica sources in the synthesis of SiC composites.

Experimental

Materials

The chemicals (all of anal grade) used were Hydrochloric acid (HCl) and Sodium hydroxide (NaOH), all sourced from Kobian limited (outlet of Sigma Aldrich), Nairobi, Kenya. Distilled water obtained from Kenyatta University chemistry laboratories. The coffee husk wastes were collected randomly at Othaya Constituency, Nyeri County, Kenya. Afterwards, they were cleaned using distilled water, chopped and oven-dried at 105 ^{o}C for 1 day to remove moisture, and then ground into a fine powder.

Instrumentation

The instruments and equipment used were Fourier Transform Infrared Spectrophotometer (IR Tracer-100, Japan), Automated X-Ray Fluorescence Spectrometer (Bruker S1 Titan 600, Tracer 5/ CTX), Distiller (WSB 14), Field Emission Scanning Electron Microscope (FEI ESEM, Vega3 Tescan LMH), Diffractometer (XRD, Rigaku MiniFlex II; Tokyo, Japan), Grinding mill (Retsch SR 200), Drying oven (WTC binder FD53), Magnetic stirrer with hot plate (WH240-HT), Test sieve (BK-TS 200), Thermostat-controlled muffle furnace (MC5-12 Biobase), and Analytical weighing balance (ATX224 Shimadzu).

Biochar and ash preparation

A 20 g of the raw powder was placed in a ceramic crucible and pyrolyzed in a thermostat-controlled muffle furnace at a frequency of 50 Hz, 10 °C/min (heat rate), voltage of 220 V and power output of 3.0 kW at 300 ℃ for 5 hours. The biochar sample was allowed to cool, ground and weighed. The obtained biochar was acid leached using 3 M HCl acid for 3 hours to remove any inorganic ash components. The ash material was prepared by pyrolysis of the raw biomass at 300 ℃ for 12 hours. Both the coffee husk biochar and ash materials were stored in airtight bottles awaiting subsequent experiments.

Alkali fusion process

The silica was extracted using a method described by [18] with some modifications. A 100.000 g of the coffee husk ash powder was soaked in 6 N HCl solution in a glass beaker for 12 hours. The mixture was then filtrated, the residue washed with distilled water and then dried at 105 °C to a constant weight. A 400 mL of 10 N NaOH solution was then added to the ash residue and heated at 150 ^{o}C while stirring for 4 hours. The filtrate (sodium silicate) was separated from the unreacted ash residues using Whatmann filter paper No. 1 and 6 N HCl solution slowly added to form a white gel (pH 7). The reaction mixture was left overnight at room temperature, filtered and rinsed with distilled water. The gel was then dried at 105 °C to a constant weight to obtain amorphous silica.

Silicon carbide (SiC) synthesis

The Silicon Carbide (SiC) composites were prepared by carbothermal reduction using a method described by [19]. The process was performed using a digestion bomb locally made at Kenyatta University mechanical engineering

laboratories, school of engineering. The bomb was made of a stainless steel (SS-316) whose cylindrical body was 105 mm in total length, 38 mm in external diameter, 23.5 mm in internal diameter. The top lid had a total length of 39 mm and thread length of 23.5×1.5×22 mm. The biochar powder was mixed with the extracted silica in the ratio of 3 to 2 for two hours, and then placed in a digestion bomb. The bomb was tightly closed and placed in a thermostat-controlled muffle furnace and heated at 300 ^{o}C for 12 hours and the final product allowed to cool and stored awaiting characterization.

Results and Discussion

Characterization of silica

The XRF analysis

The results for XRF analysis of the extracted silica are presented in Table 1. The results showed that silica was the main component (> 91%) with minimal inorganic contents (< 1%). This showed the effectiveness of the alkali fusion extraction process. The findings showed a higher silica content than those reported by [18] on the facile extraction and characterization of silica nanopowder from marine national park beach sand via alkali fusion route.

Table 1. XRF analysis of the extracted silica

Oxide (%)			
SiO_2	Fe_2O_3	Al_2O_3	CaO
91.38 ± 0.09	0.54 ± 0.13	0.04 ± 0.07	0.69 ± 0.08

The FT-IR analysis

The Figure 1 shows the FT-IR spectrum for the extracted silica The FT-IR spectrum showed a broad band at 3421.78 cm^{-1} which was ascribed to –OH stretching vibration of the water molecules attached to the silica surface [18]. This was confirmed by its bending vibration at 1638.84 cm^{-1}. The peaks at 1028.01 cm^{-1} and 737.93 cm^{-1} exhibited asymmetrical and symmetrical stretch modes for Si-O-Si groups [20]. The absorbance of peak at 497.98 cm^{-1} corresponds to the Si-O-Si bend vibrations [21]. These results are coherent to findings reported by [22] during their research study on the synthesis of silica nanopowder from Algerian river sand respectively via alkali fusion route.

The EDX analysis

The EDX analysis of the extracted silica is depicted in Figure 2.
The EDX spectrum (as displayed in Figure 2) showed that Silicon and Oxygen as the main components of the extracted silica with percentage composition of 52.7% and 46.8%,

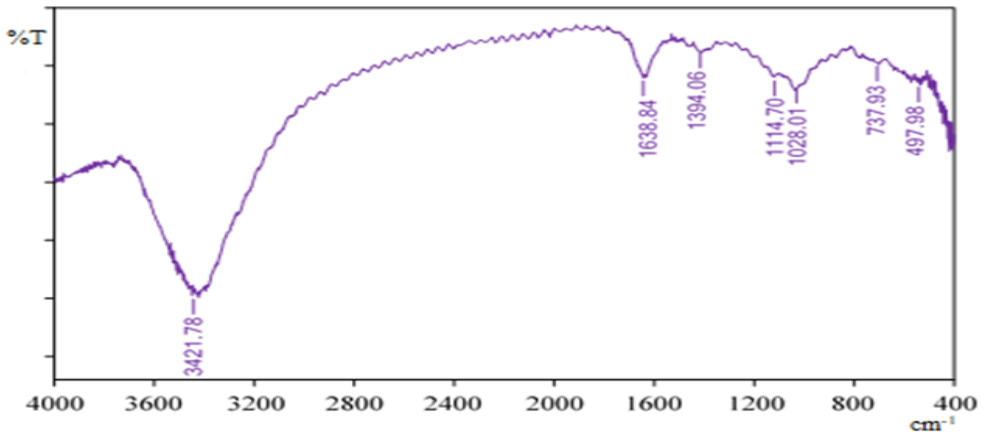

Figure 1. FT-IR spectrum of extracted silica

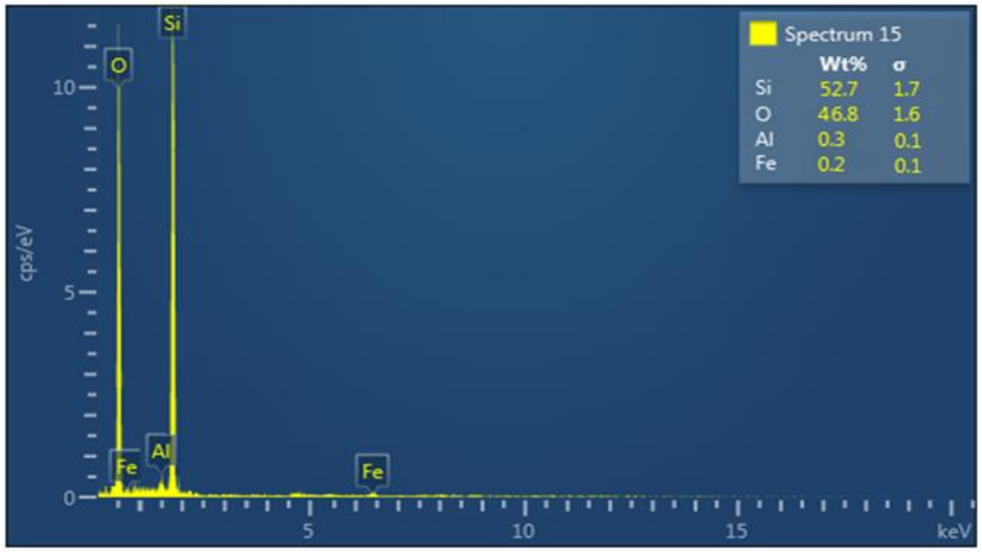

Figure 2. EDX spectrum of the extracted silica

respectively. The lower composition of Al (0.3%) and Fe (0.2%) showed that alkali fusion lowered the metal oxide impurities to < 1%). The findings are in agreement with the XRF results.

Characterization of biochar

The coffee husk biochar material pyrolyzed at 500 °C for 5 hours was characterized by XRD, SEM, EDX, and FT-IR.

The XRD analysis

The XRD results of the coffee husk biochar are demonstrated in Figure 3. The results for the XRD diffractogram showed two major peaks at $2\theta = 23^{o}$ and 45^{o} which corresponded to the highly amorphous and diffuse graphite peaks (JCPDS-ICDD file No. 41-1487). This characterized a predominantly amorphous carbon structure. There was no evidence of any inorganic phases in the XRD patterns showing that the prepared biochar samples had negligible ash content [19]. The findings of the study showed a highly amorphous carbon structure of the biochar materials.

The SEM analysis

The SEM micrograph of the biochar material is demonstrated in Figure 4.

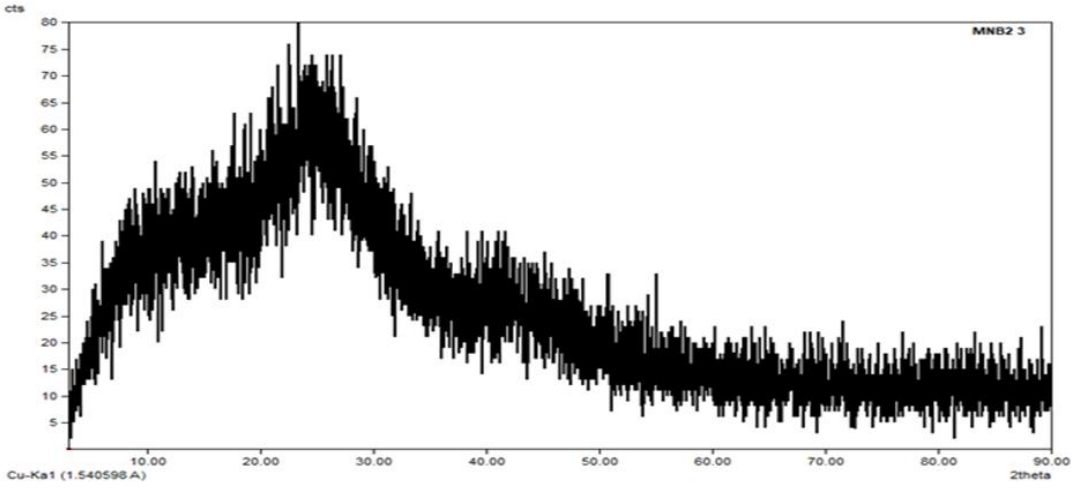

Figure 3. The XRD pattern of the coffee husk biochar

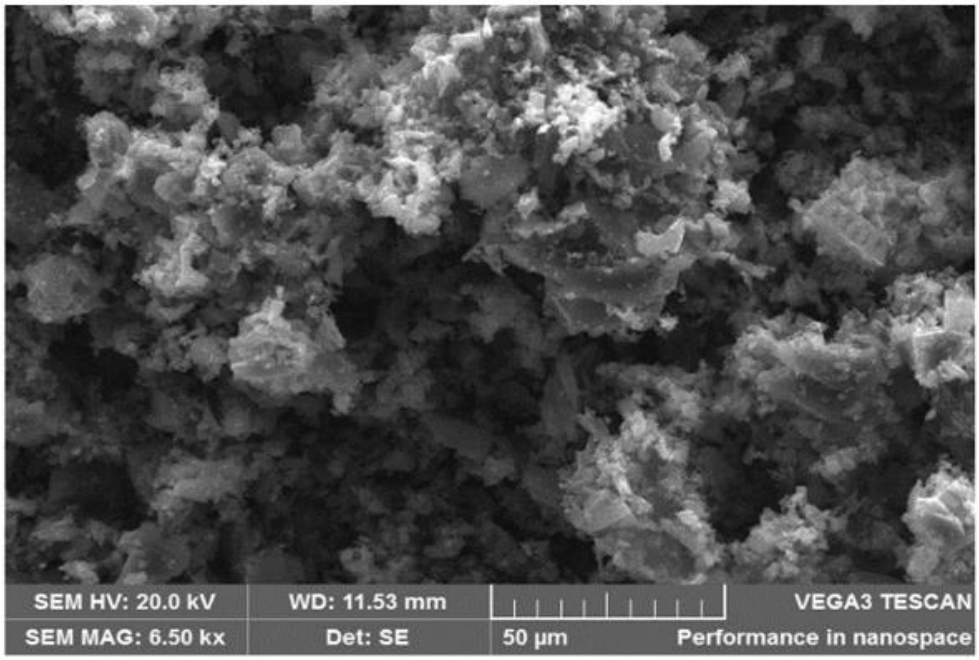

Figure 4. SEM micrograph of coffee husk biochar

The SEM image (Figure 4) showed a rugged and more porous biochar surface. This could be due to decomposition of volatile compounds during pyrolytic treatment of the biomass material [19]. This porous nature of the biochar carbon structure enhanced its surface area which could improve its usability for carbothermal reduction of silica materials during SiC synthesis.

The EDX analysis

The EDX spectrum of the coffee husk biochar is illustrated in Figure 5.

The EDX spectrum showed elemental composition of 73.4% (C), 21.8% (O), 3.6% (Si), 0.6% (Al), and 0.5% (Cl). The results showed carbon and silica as the main components of the biochar material with negligible ash contents.

The FT-IR Analysis

The FT-IR Analysis of the biochar material is depicted in Figure 6.

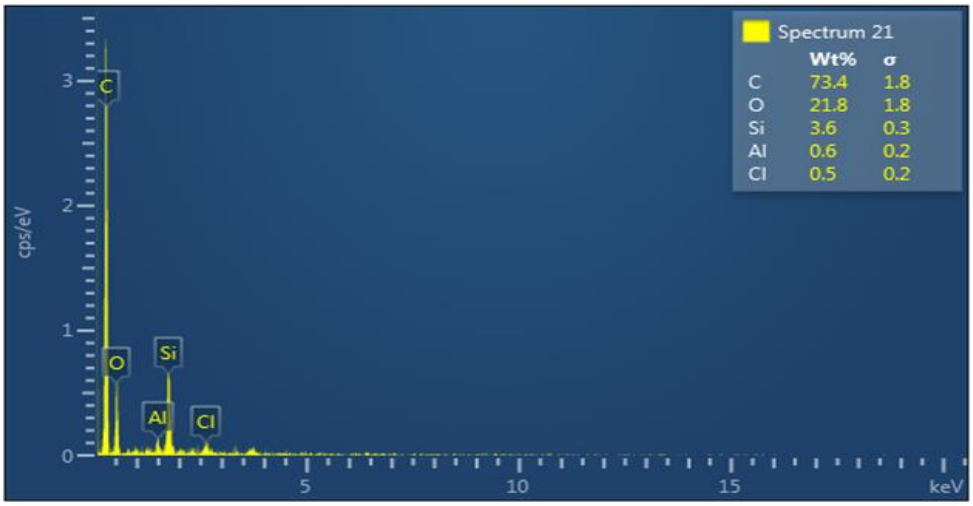

Figure 5. The EDX spectrum of coffee husk biochar

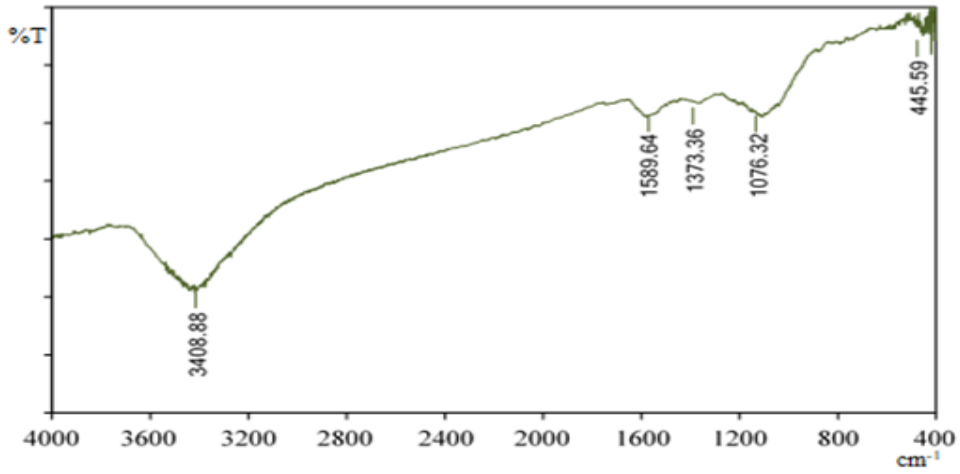

Figure 6. The FT-IR spectrum of coffee husk biochar

The FT-IR spectrum (Figure 6) showed a characteristic peak at 1589.64 cm^{-1} which could be due to aromatic C=C groups [22]. The peaks at 1076.32 cm^{-1} and 445.59 cm^{-1} confirmed the presence of silica contents in the biochar material. A broad peak at 3408.88 cm^{-1} can be ascribed to the stretching vibration of O-H of adsorbed water molecules on the biochar material [24].

Characterization of SiC composites

The synthesized SiC composites were characterized by FT-IR, XRD, SEM, and EDX. The results are presented in the following sub-sections.

The FT-IR analysis

The FT-IR spectrum for the Silicon carbide (SiC) composite material is shown in Figure 7.
The FT-IR results showed a characteristic peak at 849.23 cm^{-1} attributed to Si-C stretch band [17]. A peak at 3408.88 cm^{-1} could be due to adsorbed water molecule on the SiC surface. This is confirmed by a flexion band at 1643.68 cm^{-1}.

The XRD analysis

The XRD pattern of SiC composite is presented in Figure 8.

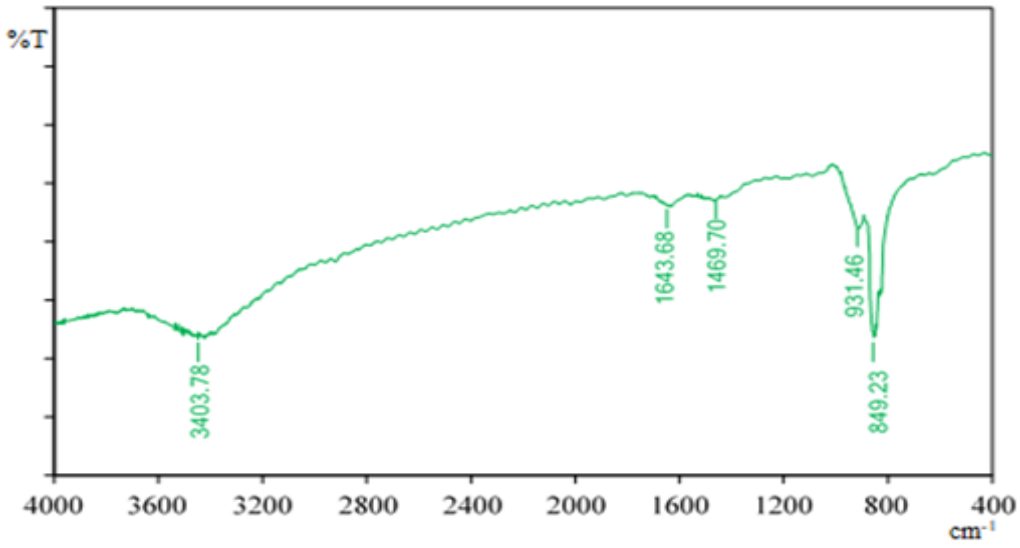

Figure 7. The FT-IR spectrum of SiC composite

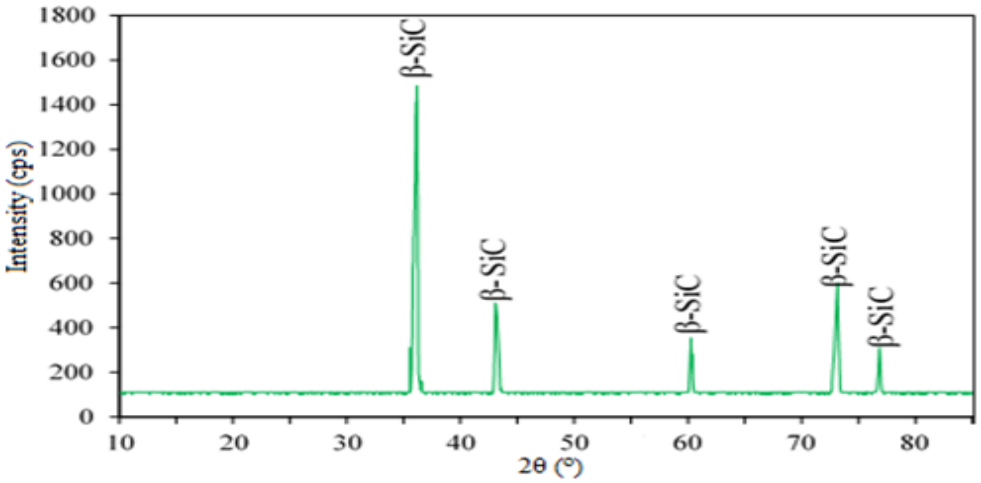

Figure 8. XRD pattern for SiC composite

The XRD diffraction pattern exhibited β-SiC (JCPDS-ICDD file No. 75-0254) confirming the formation of SiC. The noise level and the absence of any other diffraction peaks showed that the synthesized β-SiC was highly pure. This also revealed absence of silica and carbon residues in the SiC composite material.

The SEM analysis

The SEM results for SiC composites are demonstrated in Figure 9. From the SEM micrograph (Figure 9), the synthesized SiC composites consist of sphere-shaped and macron sized composites. The SEM images showed that the particle size of β-SiC was less than 5 μm.

The EDX analysis

The EDX spectrum for SiC composite is illustrated in Figure 10.

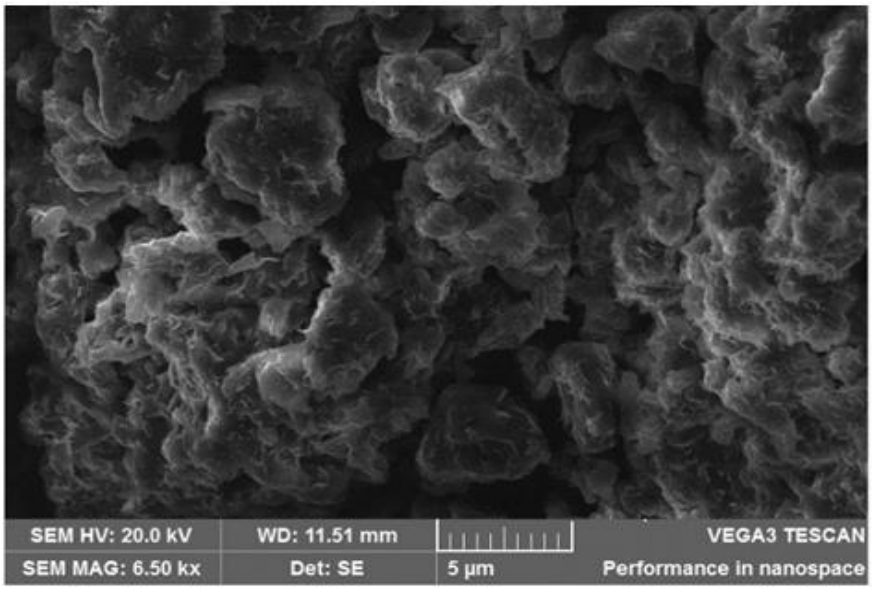

Figure 9. SEM micrograph for SiC composite

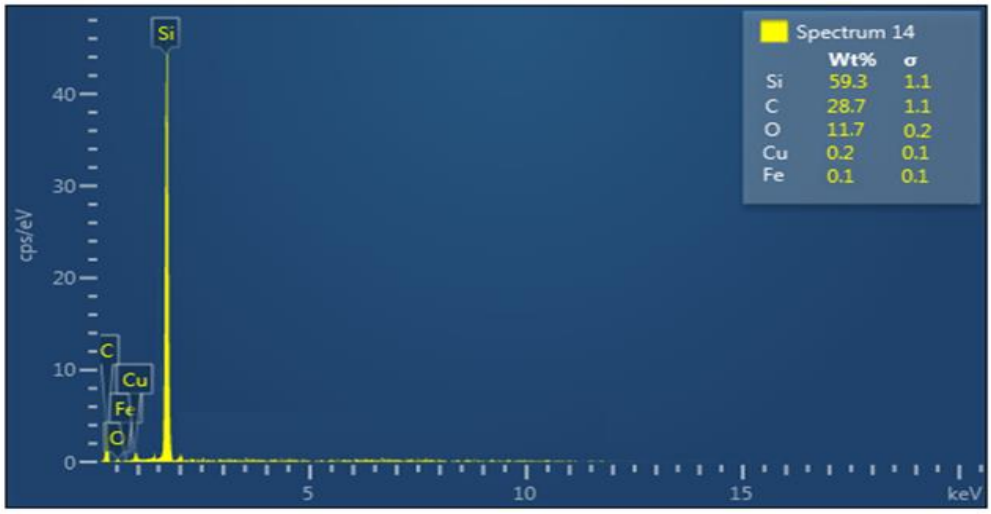

Figure 10. EDX spectrum of SiC composite

The main chemical composition (EDX spectrum) was found to be Si (59.3%) and C (28.7%). It can be seen that the sample compose of silicon and carbon. Minor peak for copper observed could be due to contamination from the corundum tube during the ball milling and/or firing process and the copper grid sample holder [25].

Conclusion

The results for the extracted silica showed a highly amorphous silica with hydroxyl in silanol (Si-OH) and siloxane (Si-O-Si) functional groups. The results for the coffee husk biochar showed a highly amorphous, porous carbon structure with a high carbon/silica contents as carbon and silica precursors for the synthesis of SiC composites. The carbothermal reduction process yielded a highly pure and spherical shaped SiC materials. The findings of the study revealed the use of coffee husk wastes novel materials (with a higher silica and carbon contents) for the synthesis of SiC bioceramic composite using a cheaper and greener synthetic approach for vast applications in various fields.

Orcid

Samuel N. Ndung'u
https://orcid.org/0000-0002-3935-9593

Esther W. Nthiga
https://orcid.org/0000-0001-7923-8465

Ruth N. Wanjau
https://orcid.org/0000-0002-3108-7466

Anthony M. Ndiritu
https://orcid.org/0009-0003-2130-9445

References

[1]. Y. Gong, S. Qu, Y. Yang, C. Liang, P. Li, Y. She, Some observations in grinding SiC and silicon carbide ceramic matrix composite material, *The International Journal of Advanced Manufacturing Technology*, **2019**, *103*, 3175-3186. [Crossref], [Google Scholar], [Publisher]

[2]. K.W. Mas'udah, M. Diantoro, A. Fuad, Synthesis and structural analysis of silicon carbide from silica rice husk and activated carbon using solid-state reaction, *Journal of Physics: Conference Series, IOP Publishing*, **2018**, 012033. [Crossref], [Google Scholar], [Publisher]

[3]. M. Pereira, D. Figueira, B. Girolamo, F. Vernilli, Synthesis of silicon carbide from rice husk, *Cerâmica*, **2020**, *66*, 256-261. [Crossref], [Google Scholar], [Publisher]

[4]. Y. Xie, Z. Huang, Q. Ni, S. Wang, L. Ban, T. Yun, J. Dai, L. Wang, X. Song, Synthesis of silicon carbide nanoplates by using laboratory wastes

as resource at low temperature, *Journal of the Ceramic Society of Japan*, **2022**, *130*, 943-947. [Crossref], [Google Scholar], [Publisher]

[5]. M. Dasog, L.F. Smith, T.K. Purkait, J.G. Veinot, Low temperature synthesis of silicon carbide nanomaterials using a solid-state method, *Chemical Communications*, **2013**, *49*, 7004-7006. [Crossref], [Google Scholar], [Publisher]

[6]. L. Carassiti, A. Jones, P. Harrison, P.S. Dobson, S. Kingman, I. MacLaren, D.H. Gregory, Ultra-rapid, sustainable and selective synthesis of silicon carbide powders and nanomaterials via microwave heating, *Energy & Environmental Science*, **2011**, *4*, 1503-1510. [Crossref], [Google Scholar], [Publisher]

[7]. A. Najafi, F.G. Fard, H. Rezaie, N. Ehsani, Synthesis and characterization of SiC nano powder with low residual carbon processed by sol–gel method, *Powder Technology*, **2012**, *219*, 202-210. [Crossref], [Google Scholar], [Publisher]

[8]. H.X. Li, G.Y. Li, T.J. Hu, X.D. Li, Y.Q. Yang, Preparation of amorphous silicon carbide nanostructures via solvothermal method, *Applied Mechanics and Materials*, **2014**, *597*, 49-52. [Crossref], [Google Scholar], [Publisher]

[9]. Q. Wu, S. Yu, H. Zhong, X. Li, T. Xiao, L. Liu, X. Guo, Y. Li, Y. Nie, Preparation of silicon carbide coating by chemical vapor deposition by using hexamethyldisilylamine precursor, *Surface and Coatings Technology*, **2018**, *334*, 78-83. [Crossref], [Google Scholar], [Publisher]

[10]. A.Y. Pak, K. Larionov, A. Korchagina, T.Y. Yakich, A.Y. Nalivaiko, A. Gromov, Silicon carbide obtaining with DC arc-discharge plasma: synthesis, product characterization and purification, *Materials Chemistry and Physics*, **2021**, *271*, 124938. [Crossref], [Google Scholar], [Publisher]

[11]. S. Maroufi, M. Mayyas, V. Sahajwalla, Novel synthesis of silicon carbide nanowires from e-waste, *ACS Sustainable Chemistry & Engineering*, **2017**, *5*, 4171-4178. [Crossref], [Google Scholar], [Publisher]

[12]. S. Ndung'u, R. Wanjau, E. Nthiga, Efficacy of adsorption of congo red dyes from an aqueous media using silicon nitride (Si_3N_4) adsorbent derived from sand and coffee husk wastes. Eurasian, *Eurasian Journal of Science and Technology*, **2024**, *4*, 253. [Crossref], [Google Scholar], [Publisher]

[13]. R. Rajarao, R. Ferreira, S.H.F. Sadi, R. Khanna, V. Sahajwalla, Synthesis of silicon carbide nanoparticles by using electronic waste as a carbon source, *Materials Letters*, **2014**, *120*, 65-68. [Crossref], [Google Scholar], [Publisher]

[14]. S. Maroufi, M. Mayyas, R.K. Nekouei, M. Assefi, V. Sahajwalla, Thermal nanowiring of e-waste: a sustainable route for synthesizing green Si_3N_4 nanowires, *ACS Sustainable Chemistry & Engineering*, **2018**, *6*, 3765-3772. [Crossref], [Google Scholar], [Publisher]

[15]. S. Qadri, E. Gorzkowski, B. Rath, J. Feng, S. Qadri, H. Kim, J. Caldwell, M. Imam, Nanoparticles and nanorods of silicon carbide from the residues of corn, *Journal of Applied Physics*, **2015**, *117*. [Crossref], [Google Scholar], [Publisher]

[16]. Z. An, J. Xue, H. Cao, C. Zhu, H. Wang, A facile synthesis of silicon carbide nanoparticles with high specific surface area by using corn cob, *Advanced Powder Technology*, **2019**, *30*, 164-169. [Crossref], [Google Scholar], [Publisher]

[17]. R.R. Ravindra Rajarao, V.S. Veena Sahajwalla, A cleaner, sustainable approach for synthesising high purity silicon carbide and silicon nitride nanopowders using macadamia shell waste, *Journal of Cleaner Production*, **2016,** *133*, 1277-1282. [Crossref], [Google Scholar], [Publisher]

[18]. S. Ndung'u, E.W. Nthiga, R. Wanjau, Facile extraction and characterization of silica nanopowder from marine national park beach sand via alkali fusion route, *International Journal of Technology,* **2023**, *13*, 1-6. [Crossref], [Google Scholar], [Publisher]

[19]. S.N. Ndung'u, E.W. Nthiga, R. Wanjau, Characterization of amorphous silicon nitride prepared from sand and coffee husk wastes by carbothermal reduction-nitridation, *African Journal of Pure and Applied Sciences*, **2023**, *4*, 24-34. [Crossref], [Google Scholar], [Publisher]

[20]. A. Andriayani, Y. Muis, D.Y. Nasution, Chemical reduction of silica into silicon from extracted quartz sand using sodium hydroxide and hydrochloric acid solutions, *AIP Conference Proceedings, AIP Publishing*, **2021**. [Crossref], [Google Scholar], [Publisher]

[21]. M. Zainuri, Synthesis of SiO nanopowders containing quartz and cristobalite phases from silica sands, *Materials Science-Poland*, **2015**, *33*, 47-55. [Crossref], [Google Scholar], [Publisher]

[22]. A. Boualem, L. Leontie, S.A.G. Lopera, S. Hamzaoui, Synthesis and characterization of mesoporous silica from Algerian river sand for solar grade silicon: Effect of alkaline concentration on the porosity and purity of silica powder, *Silicon*, **2022**, *14*, 5231-5240. [Crossref], [Google Scholar], [Publisher]

[23]. J.N. Njeri, E.W. Nthiga, G.K. Muthakia, The efficacy of coffee husks biochar in the adsorption of methyl red from textile dyeing wastewater, *International Journal of Scientific Research in Chemical Sciences*, **2023**, *10*, 1-9. [Crossref], [Google Scholar], [Publisher]

[24]. K. Philippou, I. Anastopoulos, C. Dosche, I. Pashalidis, Synthesis and characterization of a novel Fe_3O_4-loaded oxidized biochar from pine needles and its application for uranium removal. Kinetic, thermodynamic, and mechanistic analysis, *Journal of Environmental Management*, **2019**, *252*, 109677. [Crossref], [Google Scholar], [Publisher]

[25]. J. Huang, Z. Huang, S. Yi, Y.g. Liu, M. Fang, S. Zhang, Fe-catalyzed growth of one-dimensional α-Si_3N_4 nanostructures and their cathodoluminescence properties, *Scientific Reports*, **2013**, *3*, 3504. [Crossref], [Google Scholar], [Publisher]