Green Synthesis and Characterization of Antibiotic Amorphous Nano Silicon Oxide Powder Extracted from Rice Husk Ash

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ABSTRACT

Introduction: Green synthesis of nano silicon oxide powder has been done using rice husk as raw material, which contains more than 91% of silica oxide and other mineral compositions.

Aim: To synthesise pure nano-silica oxide powder and utilize pure silica oxide for the production of wound dressings work as an antibiotic.

Materials and Method: Rice husks were burned for three hours in a Muffle furnace at 700°C, then treated with sodium hydroxide solution using a magnetic stirrer at 100°C for four hours to extract the silica present in the ash to generate sodium silicate (Na_2SiO_3). The pure silica powder is obtained by the titration method with 5N solution of sulfuric acid purification of nano-silica powder is employed through the refluxing method.

Results: X-ray diffraction, energy-dispersive x-ray spectroscopy (EDX), field emission scanning electron microscopy (FE-SEM), and FT-IR techniques were used to characterize nano-silica powder. According to Bragg's equation of X-ray diffraction, nano silicon oxide crystals have orthorhombic amorphous forms with average crystal sizes of 33.406, 53.135, and 10.10362 nm. FE-SEM show the partial size in nano size. FT-IR technic shows that the absorbance band at 1072.46 cm^{-1} refers to asymmetric vibration Si-O-Si (siloxane group), 808.20 cm^{-1} refers to Si-O-Si bond symmetric vibration.

Conclusion: The formation of pure nano Silica oxide from rice husk ash, has been demonstrated to exhibit antibacterial efficacy against Gram-negative bacteria (*Escherichia coli*), with a concentration of 62.5 $\mu\text{g/ml}$ as well as utilizing pure silica oxide for the production of wound dressings work as an antibiotic.

Key Words: Antibiotic, Nanoparticles, Rice husk, Green synthesis, Wound dressings and silicon oxide

INTRODUCTION

The green synthesis approach of nanomaterials has a large potential for industrial application due to cheap, safe and less waste of environmental contamination than other methods. Conditions can be under control for the processing of nanomaterials such as acidity degree, the concentration of reactants mixing temperature.^{1,2} The structural properties of nanocrystal material depend on a specific preparing approach. Several physical and chemical methods of preparation have been used for the latter one has been much interested, due to relies on the use of natural raw materials instead of chemicals that are more toxic and expensive.^{3,4} Biological methods are depending on natural materials that involve production and developing ways to synthesis new nanomaterials that being environmentally friendly.⁵ Physical approach techniques

have been especially advantageous on the chemical method that use toxic materials but consider more expensive if compare with other methods therefore the biological method is more appropriate.^{6,7} Silica nanoparticle materials are prepared in different ways, such as the sol-gel method and precipitation method. Rice husk is contained 80-90% silica and other material impurities like Alumina Al_2O_3 and Potassium oxide (K_2O).^{8,9} Rice husk can obtain from agricultural waste material; around 80 million tons of rice husk ash (RHA) are produced annual^{10,11}. It has lightweight according to reports, nearly 0.23 tons of husk a reproduced for every ton of rice consumed.¹² Because rice ash is available and free of cost that makes valuable material for researchers, especially this is continent, many of composition.¹³ Amorphous silica nanoparticles appear to be particularly interesting candidates for

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this application not only will they deliver Orthosilicic acid ($\text{Si}(\text{OH})_4$) up to its solubility limit, but they will also help with wound healing.¹⁴ Infected wounds are a complicated, non-trivial condition that puts patients' health and lives in jeopardy. A chronic ulcer with deep tissue damage in diabetic patients is one of the most common disorders, which is linked to aberrant fibroblast activity, reduced cell migration, and decreased angiogenesis, all of which can lead to vascularization impairment, diabetes causes wound contraction to be delayed, resulting in non-healing diabetic wounds.¹⁵ The ratio of biofilm-forming to non-biofilm-forming bacteria in a lesion is affected by the type and duration of treatment.¹⁶ As a result because a wide range of bacteria dwells in wounds, there is a generally recognized the need for new antibacterial medications to combat the global rise in resistance.¹⁷ Many recent studies show Silica supplementation or increased dietary Silica intakes, have been linked to improvements in bone and skin health in people.¹⁸ In the present work, nano-silica oxide will be synthesised from rice husk by an adjustable condition method and the product is characterised with different instrumental techniques. Then measure antibiotic activity against gram-negative bacteria (*E. Coli*).

Experimental Material

Rice husk is locally agricultural Iraq(RH), Hydrochloric acid is from British Drug purity 98.5%, Sodium Hydroxide is about Reagent World purity 97%, USA. Sulphuric acid is from British Drug House purity 95-97% and tribal distilled water. Muller Hinton agar powder (Microexpress)

Preparation of silica oxide method

Rice husks are mechanically purified from impurities such as soil, then washed with distilled water. Dried is done at 100 °C for 3 hours. Burning has been done in a furnace (Nabertherm model HTCT 08/16) Germany at 700 °C for 2 hours to get on white ash. Into 10 g of burning rice husk ash (RHA) sample, added 80 ml of (2.5N) sodium hydroxide solution with a magnetic stirrer under 100 °C for 4 hours to dissolve the silica existing in rice husk ash by using 250 ml of an Erlenmeyer flask. The resulting is filtered out and washed with warm distilled water. In this step, have a yield with transparent viscous, colourless, it is a sodium silicate solution as in the following reaction equation

Extraction of pure silicon dioxide by titration the yield with (5N) of sulphuric acid solution H_2SO_4 , (CAS 7664-93-9) gradually adding to a sodium silicate solution with a magnetic stirrer under 100-90 °C. Complete precipitation of silica at this step and form sodium sulphate as in the following reaction equation

The gel product is washed many times with warm distilled water to remove the sodium sulphate impurities. To extract the pure Nano silicon dioxide through the refluxing method with 100 ml of (6N) Hydrochloric acid solution for 6 hours under 80-90 °C, in presence of boiling stone to uniform temperature distribution of reaction vessel. The product solution is filtered out and the precipitate is washed with warm distilled water to remove impurity then dried in an oven at 100 °C for 24 hours. X-ray diffraction high resolution (2700 AB HAO YUAN), FT-IR technic (IR Affinity-1S SHIMADZU), EDX and SEM measurements were employed to characterize nanopowder (silicon oxide). EDX and FE-SEM (MAG 400 Kx) Germany measures the sample after and before purification by the refluxing method and calcination for 700 °C. (Figure 1) represents the sequence of four steps of rice husk to conversion into nanopowder of SiO_2 .

Antibiotic activity test

Preparation of solutions, by dissolving 38 g of the miller Hinton agar powder in 1000 ml of triple distilled water, then sterilized by using an autoclave (Hirayama SN 30613075947) for 30 min at 121° C, the solution cooling to 37 °C, then poured the solution into sterilized Petri dishes, after adding inoculum. It is preparation by taking an appropriate amount of gram-negative bacteria (*Escherichia coli*), dilution by the sterilized saline solution to get on 0.5 McFarland (1.2×10^8 CFU/ml). Microbial colonies (*Escherichia coli*) were taken from the wound and burn patients after being isolated and identified by biochemical tests.¹⁹ Investigation antibiotic activity of pure nano-silica oxide powder that preparation with concentration (control = 0,62.5, 31.25, and 3.125) µg/ml, SiO_2 dissolve in DMSO solvent. Petri dishes were incubated at 37°C for 24 hours after applying pure nano-silica oxide powder on wound dresses.

RESULTS

Synthesis of nano-silica oxide powder after passing many stages such as shown in (Figure 1). The product nanopowder characterization by X-ray diffraction (2700 AB HAO YUAN) such as (Figure 1.), Energy-dispersive X-ray spectroscopy (EDX), FE-SEM that show percentage weight of the element in the sample and partials size in nanoscale such as shown in (Table 2,3) and (Figure 3,4). FT-IR spectra of nano-silica powder are represented in (Figure5). Pure silica oxide powder extraction from rice husk ash gives perfect antibiotic activity at a concentration of 62.5 µg/ml such as shown in (Table 4).

DISCUSSION

Refluxing process of the reaction solution is aimed at heating the reaction for a long time without losing solution volume.

As a complete melting of the sample is achieved, which is thermodynamically preferred, as it increases the speed of the reaction, regulates the crystals, and obtains nanoparticles. Nano silica powder is detected by X-ray diffraction technique (2700 AB HAO YUAN), Cu K α ($\lambda = 1.54 \text{ \AA}$), since (XRD) used the sample attended as homogenous powder put on a slice of an X-ray path. The scanning rate 10 /min with a two theta (2θ) range is limited from 10 to 80 degrees. (Figure 2) shows the patterns of orthorhombic amorphous nano silicon dioxide. The average size crystal of silica powder calculates from the Debye-Scherrer equation such as shown in (Table 1) $D = K \lambda / \beta \cos \theta$, D represents the average particle size, K is the dimension spherical shape factor 0.94, λ is the X-ray wavelength Cu- K α (0.154) nm and β , is the liner broadening.^{20,21,22} Energy-dispersive X-ray spectroscopy (EDX) is a technique for measuring the composition percentage in the sample, that give information about the purity of pattern, such as shown in (Table 2,3) that explain the percentage weight of elements found in silica oxide powder we are prepared from rice husk ash that shows content carbon, oxygen, sodium, silica, and sulphate with percentage weight (5.31, 53.39, 2.98, 35.69 and 4.38 %) respectively. (Table 3) and (Figure 4) shows the results of the sample powder prepared after purification with more time 24 hours by reflexing method, washing sample many times by warm distilled water, ethanol and calcination at 700 °C for 3 hours, we notice from the results of the EDX technique that the percentage weight of impurity be zero and appear the high-intensity peak for silica element with high percentage weight 40.81%. These results show we are gate on silica oxide with high purity from rice husk ash. (Figures 3,4) shows FE-SEM image a regulation of morphology sample, the shape of nanoparticles of silica oxide is spherical and average particle size smaller than 100 nm nanoscale. Results agree with results of x-ray diffraction that show particle size of sample powder in nanoscale.²³ FT-IR spectra of nano-silica powder are represented in (figure 5) since appear absorbance band at wavelength 1072.46 cm⁻¹ due to asymmetric vibration Si-O-Si (siloxane group), symmetric vibration at 808.20 cm⁻¹ by bond Si-O-Si and 484.14 cm⁻¹ attributed to the bending vibration of Si-O.^{24,25} Antibiotic activity the results of inhibition growth of microbial for *Escherichia coli* show in the table that indication nano-silica oxide extraction from rice husk ash has high sensitivity against gram-negative *E.coli*, the antibacterial activity for different doses of pure silica oxide applied on wound dresses in this work. A Silica oxide dose of 62.5 $\mu\text{g/ml}$ pluperfect thereby a 25 mm such as in (Table.4) shows the inhibition growth in Petri dishes.

CONCLUSIONS

We must confirm that green chemical products and processes are clean and safe. Green chemistry Future trends

to chemists are using their innovative and creative skills from all over the world to build up new processes, reaction conditions, synthetic methods, catalysts etc. About 91% of amorphous pure silicon oxide powder has been obtained from rice husk as agriculture waste product through green synthesis route. Characterization by X-ray, EDX, FE- SEM and FT-IR technique which shows revelation crystal size by Debye – Sherrer equation in the nanoscale ($D = 42.862, 28.624, 28.7335, 57.74055, 43.457, 9.159$ and 12.663 nm). FE-SEM show the morphology of the surface and Patricia size in nanoscale. FT-IR peak appears (Si-O-Si) silicone groups at wavelength 1072.46 cm⁻¹, 808.20 cm⁻¹ for asymmetric vibration and symmetric vibration respectively. This work provides a cheap natural source and direct method to produce amorphous nano-silica with high purity after treatment with reflexing method and calcination at 700 °C for 3 hours. Successful results of antibiotic activity show nano-silica oxide represent a promising nanomaterial to eliminate microbial infections and thereby wounds, by using a natural source more friendly for environmentally.

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All authors helped with data analysis, authoring, and revising the manuscript, and they agreed to take responsibility for all areas of the project.

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Table 1: The average practical size for pure Nano of silicon dioxide SiO₂.

NO.	2θ [Theta]	Practical size nm
1	19.8980	42.862
2	21.8158	28.624
3	24.149	28.733
4	26.4091	57.740
5	28.0972	43.457
6	30.109	58.208
7	32.3190	58.477
8	35.734	9.159
9	39.608	9.159
10	48.9902	12.663

Table 2: Explain the intensity and percentage weight of Nano of silica powder.

Element	Intensity	Weight%
C	40.2	5.13
O	252.6	53.39
Na	61.0	2.98
Si	370.8	35.69
S	105.3	2.80

Table 3: Explain the intensity and percentage weight of pure Nano silica powder.

Element	Intensity	Weight %
O	289.6	56.78
Si	984.0	40.81
C	30.96	2.41

Table 4: The effect of dose of pure Nanosilica oxide that applied on wound dresses on the growth microbial (*Escherichia coli*).

Zone inhibition/mm	Concentration / $\mu\text{g/ml}$	
5.2	3.125	
16.5	31.25	
25	62.5	

Figure 1: Preparation steps of Nano silicate dioxide. A. Rice husk, B. Rice husk ash at 700 °C treatment, C. silica gel, and D. Nano silicate dioxide powder.

Figure 2: Spectra of X-ray diffraction of silicon oxide.

Figure 3: EDX of Nano silica oxide powder with FE-SEM surface morphology.

Figure 5: FT-IR spectra of pure Nano silica powder.

Figure 4: EDX of pure Nano silica oxide powder with FE-SEM surface morphology.