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Facile synthesis, characterization and antibacterial activity of nanostructured palladium loaded silicon carbide

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ABSTRACT

In this work, noble metal (Palladium) loaded silicon carbide (SiC) nanoparticles have been successfully synthesized using a single step synthetic route and its antibacterial action against gram-negative (E. coli) and gram-positive (S. aureus) bacteria have been investigated. The structural and morphological characterizations of pure SiC and Palladium (Pd) loaded SiC nanoparticles were carried out by x-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), Field emission scanning electron microscopy (FE-SEM), Energy dispersive x-ray spectroscopy (EDX), Elemental mapping and Transmission electron microscopy (TEM). The characterizations results offer substantial proof that the SiC surface was successfully decorated by Pd. Furthermore the EDS analysis reveals that the product contained Pd as well as W and O, thus reaffirming the production of Pd loaded SiC nanoparticles. The MICs and MBCs values examined by standard agar dilution methods show that MICs and MBCs values of pure SiC were > 16 and > 32 mg/ml, respectively against E. coli and S. aureus, whereas Pd loaded SiC nanoparticles exhibited MIC and MBC value of 4 mg/ml and 8 mg/ml, respectively. The morphological and structural alterations caused by SiC and Pd loaded SiC nanoparticles on *E. coli* and S. aureus cells were further investigated by SEM analysis. A noteworthy improvement in antibacterial performance was observed, when *E. coli* and *S. aureus* cells were exposed to Palladium (Pd) nanoparticles (NPs) loaded silicon carbide (SiC). The results obtained show a significant impact by loading Pd on SiC in the deactivation of microorganisms in vitro.

1. Introduction

Recently, carbon based nanomaterials such as fullerene (C60 and C70), carbon nanotubes (single walled carbon nanotubes and multiwall carbon nanotubes), graphene (graphene oxide, reduced graphene oxide and graphene) and diamond like carbon have been widely utilized for biological applications [1–4]. Several scientist focus has been at present demonstrating an increasing enthusiasm to study the impact of carbon-based nanomaterials on antibacterial performance [5]. The greater part of the accessible articles represents the impact of such carbon-based nanostructures on bacteria and cells [6]. It is hard to find reports depicting the interaction between other carbons based nanomaterials such as silicon carbide nanostructures and microorganisms.

Silicon carbide (SiC) is a carbon-based ceramic material that has recently been examined by numerous specialists for various applications such as sensor, photo-catalysis, photovoltaics, energy storage devices, water splitting, conversion of CO_2 into value added fuels, electrode material for supercapacitor [7–9]. There are a multitude of factors, which account for its various applications; for instance, its high chemical, mechanical and thermal stability and many other distinct features. Moreover, microcrystalline SiC have been utilized for studying the impact on mammalian cells [10]. It has been demonstrated that micro SiC does not initiate destructive impacts on tissues [11].

On the other hand, noble metal such as silver (Ag), platinum (Pt) and palladium (Pd) have been widely utilized as doping or loading material to enhance the performance of nanomaterials for biological applications [12–14]. Among the noble metals, pallidum is gaining interest due to its significant properties [15,16]. Palladium nanoparticles are highly thermally and mechanically stable, and during interaction between microorganism and palladium, it can eliminate

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microorganisms and pathogens [17]. Consequently, the utilization of palladium as a loading material for the improvement of antibacterial action of the silicon carbide or other nanomaterials would be profoundly encouraging. Owing to the advantageous properties of silicon carbide and palladium, it would be interesting to synthesize nanostructured pallidum loaded silicon carbide for the evolution of antibacterial studies. To the best of our knowledge, there exist no report on noble metal (Palladium) loaded silicon carbide being used for antibacterial studies on gram-positive and gram-negative bacteria.

In this research work, carbon-based ceramic material (silicon carbide) and noble metal (Palladium) loaded silicon carbide for antibacterial studies on gram-positive and gram-negative bacterial are proposed. Palladium loaded silicon carbide have successfully been prepared using single step synthetic route and tested for antibacterial action on gram-positive and gram-negative bacteria. In addition, the structural, optical and morphological studies like XRD, FTIR, FE-SEM and TEM analysis of using pure silicon carbide and Palladium loaded silicon carbide were carried out. The antibacterial activity of pure silicon carbide and Palladium loaded silicon carbide against gram-positive and gram-negative bacteria was also carried out by determining MIC/MBC using standard agar dilution methods. Furthermore, the morphological alteration caused by theses nanoparticles against *E. coli* and *S. aureus* was investigated by SEM analysis.

2. Experimental

2.1. Reagents and chemicals

Silicon carbide (SiC) nanoparticles and $Pd(NO_3)_2.xH_2O$ were purchased from Sigma Aldrich, Inc. USA. All other required chemicals were purchased from Sigma Aldrich, Inc. USA.

2.2. Synthesis of nanostructured palladium loaded silicon carbide

The palladium loaded nano silicon carbide was synthesized by wet incipient technique. For the synthesis of Pd loaded SiC, concentrated palladium nitrate solution in deionized water (1 wt% palladium) was poured drop wise on 1 g fine SiC powder using a micropipette to make a paste and this paste is made homogeneous and initially dried at 80 °C. This dried material, under the pure hydrogen gas environment was annealed at 400 °C for 3 h to obtain the final Pd loaded SiC.

2.3. Characterization

The surface morphology and structure of silicon carbide (SiC) powder before and after loading of palladium (Pd) was examined using SEM and TEM. SEM (*FEI, ISPECT S50, Czech Republic*) was carried out at an acceleration voltage of 20 kV and TEM (*FEI, Morgagni 268, Czech Republic*) at 80 kV. SEM images of both the specimens, SiC and Pd loaded SiC were taken at 25 and 50 kX magnifications. Furthermore, energy dispersive X-ray spectroscopy (EDX) and quantitative elemental mapping were performed to evaluate the chemical composition of pure SiC and Pd loaded SiC. EDX equipment Apollo x SDD (silicon Drift Detector) and quantitative elemental analysis was performed using FE-SEM (FE-SEM, TESCAN FERA3). The samples were mounted on a SEM stub with a double-sided adhesive tape and analyzed for SEM analysis. For TEM analysis, the powder was dispersed in ethanol, sonicated for 10 min and deposited onto TEM grid having carbon support film. The grids were dried before mounting onto the TEM. TEM images were acquired in bright-field mode and electron diffraction analysis was performed by selecting the area of interest to confirm the crystalline nature and XRD data. The pure SiC and Pd loaded SiC nanoparticles were also characterized by powder X-ray diffractometery (Bruker AXS Diffractometer D8) and Fourier transform infrared spectroscopy in ATR mode at room temperature (Nicolet 6700 FT-IR spectrophotometer).

2.4. Evaluation of antibacterial activity

E. coli and *S. aureus* (Gram-negative and Gram-positive bacteria) were selected to evaluate the antibacterial properties of SiC and Pd loaded SiC nanoparticles. Before experiments, *E. coli* and *S. aureus* were grown overnight at 37 °C in nutrient broth medium in a shaking incubator (150 rpm). The culture was washed with phosphate buffer to remove the media from the culture and then *E. coli* and *S. aureus* suspensions were diluted with 0.9% NaCl solution to reach concentrations of approximately 10^7 CFU/ml.

2.4.1. Minimal inhibitory concentration (MIC)

The antibacterial properties of SiC and Pd loaded SiC nanoparticles was assessed on MHA plates using agar dilution method as previously described [18,19]. The MIC was then determined using serial dilutions of SiC and Pd loaded SiC nanoparticles in concentrations varying from 32 mg/ml to 0.25 mg/ml. The MIC is the lowest concentration of SiC and Pd loaded SiC nanoparticles at which no visible growth was seen [18,19].

2.4.2. Minimal bactericidal concentration (MBC)

The MIC plates without any bacterial growth were further selected for MBC assessment [18]. Briefly, 100 µl 0.9% normal saline were poured onto the MIC plates and then transferred to another freshly prepared MHA plates and incubated again for overnight at 37 °C [18]. The lowest concentration of pure SiC and Pd loaded SiC nanoparticles at which no growth of bacterial cells has been found or less than three CFUs were present, were recorded as MBC [18,19].

2.4.3. SEM analysis: Effect of NPs on the morphology of E. coli and S. aureus

Further, the effects of pure SiC and Pd loaded SiC nanoparticles on the morphology of *E. coli* and *S. aureus* cells investigated by scanning electron microscope as previously reported [18]. Briefly, ~10^6 CFU/ml of both *E. coli* and *S. aureus* cells were treated with sub-MIC concentration of SiC and Pd loaded SiC nanoparticles at 37 °C. The treated and untreated bacterial cultures were then washed at 12000 rpm for 10 min and the collected pellets were then again washed at least two to three times with PBS and fixed with primary fixation i.e., 2.5% glutaraldehyde and then fixed with secondary fixation i.e., 1% osmium tetroxide. After washing, all the samples were dehydrated by a series of ethanol i.e., 30%, 50%, 70%, 95% and 100% [18]. The cells were then fixed on the aluminum stubs, dried in a desecrator and coated with gold. Finally, samples were examined at an accelerating voltage of 20 kV by scanning electron microscope.

3. Results and discussions

3.1. Morphological and structural studies

Scanning electron microscopy equipped with energy dispersive X-ray spectroscopy (SEM-EDX) was used to evaluate the morphology and chemical composition of pure SiC and Pd loaded SiC. Figs. 1 and 2 show the results of SEM imaging, EDX and elemental mapping. The SEM micrographs of the pure silicon carbide (SiC) are shown in Fig. 1(a, b) which revealed the formation of well dispersed nanoparticles of SiC under 100 nm. A similar morphology was seen when added the Pd particles in the SiC matrix (Fig. 1(c, d)). The presence of Pd nanoparticles on SiC were obvious as some small particles brighter in contrast were seen attached to SiC (highlighted by white arrows), which can be attributed to aggregation or overlapping of smaller particles with size lesser than SiC particles as judged by high magnification electronic images.

SEM-EDX spectra and elemental mapping images of the SiC and Pd loaded SiC nanoparticles are displayed in Fig. 2. SEM images from where EDX spectra were obtained are also shown along with spectra