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Electrochemical and antimicrobial activity of tellurium oxide nanoparticles

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ABSTRACT

Thin film of tellurium oxide (TeO_2) has been synthesized by chemical vapour deposition method onto indium tin oxide (ITO) coated glass substrate without using any catalyst. XRD pattern of TeO_2 thin film suggests that the structure of TeO_2 changes from amorphous to crystalline (paratellurite) on dispersing into deionized water. Zeta potential measurement reveals a positive surface potential of 28.8 mV. TEM images shows spherical shaped TeO_2 nanoparticles having average particle size of 65 nm. Electrochemical studies of TeO_2/ITO electrode exhibit improved electron transfer owing to its inherent electron transfer property at pH 7.0 of phosphate buffer. Antimicrobial activity of TeO_2 has been studied for gram-positive (*Staphylococcus aureus* and *Streptococcus pyogenes*) and gram negative (*Escherichia coli* and *Klebsiella pneumoniae*) bacterial and fungal strains (*Aspergillus nizer* and *Candida albicans*). These studies suggest that the TeO_2 NPs inhibit the growth of *E. coli*, *K. pneumoniae* and *S. aureus* bacteria, whereas the same particles enhance the growth of *S. pyogenes* bacteria.

1. Introduction

Tellurium oxide (TeO_2) is a wide band (3.75 eV) p-type chalcogenide material available in both crystalline and amorphous phases [1–3]. In the crystalline form, TeO_2 exists in paratellurite (tetragonal) and tellurite (orthorhombic) phases [4,5]. TeO_2 has remarkable properties including high chemical stability, mechanical durability, high refractive index, good optical non-linearity, electrical conductivity and piezoelectricity, which makes it suitable for various applications [6,7]. Several methods, including RF sputtering, thermal evaporation, sol-gel, dip-coating and chemical vapour deposition (CVD) have been used to fabricate TeO_2 films [8–11]. However, CVD techniques provide uniform, highly purified and reproducible films. Therefore, we have synthesized TeO_2 thin film on ITO substrate using CVD technique and monitored its electrochemical activity using electrochemical techniques. This technique has advantage of fabrication of TeO_2 film is simple and cost-efficient and can be used for wide applications in electronics devices including sensors/biosensor and solar cell systems. Beside this, it is also observed that the TeO_2 was a nearly 'forgotten' material in the field of biology, but during the last few years, several studies have fuelled a renewed research interest in this material [3]. The nano-bio interaction is an important aspect for the safe use of nanoparticles (NPs) to any biological system. Recently, Arakha et al. explored the interaction of negatively charged iron oxide NP n(IONP) and chitosan modified positive charge p(IONP) with *Bacillus subtilis* and *E. coli* and reported that chitosan modified p(IONP) have significantly increases the antimicrobial propensity [12]. Jones et al., reported significantly higher antibacterial effects of ZnO NPs on *Staphylococcus aureus* (*S. aureus*) than other metal oxide NPs that depend on the size and presence of normal visible light [13]. Copper and silver NPs have also been used to study the antimicrobial activity against *Escherichia coli* (*E. coli*) and *Bacillus subtilis* (*B. subtilis*) [14]. A higher susceptibility of copper NPs having size 100 nm with *B. subtilis* and silver NPs (40 nm) with *E. coli* has reported [14]. Ruparelia et al. [15] also reported the antimicrobial properties of silver and copper NPs against *E. coli*, *B. subtilis* and *S. aureus*, and they found that the bacterial sensitivity to NPs depends on the microbial species.

Here, we have explored the interaction study of some microorganisms such as gram negative (*Escherichia coli* and *Klebsiella pneumoniae*), gram-positive (*Staphylococcus aureus* and *Streptococcus pyogenes*) bacterial and fungal strains (*Aspergillus nizer* and *Candida albicans*) towards TeO_2 NPs. It has been observed that the size, stability and the concentration of TeO_2 in the growth medium affect the antibacterial activity. Some reports suggest that the growth medium amended with NPs may inhibit the bacterial

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population growth due to specific interaction [16]. Normally, the outer cell membrane of bacteria has nano size pore with overall bacteria size in the micrometer range. Due to nanosize, the particles can easily cross the cell membrane and may inhibit the microbial growth. Zhong et al. [17] reported the antimicrobial effect of tellurium dioxide using three gram positive bacterial strains (*S. aureus*, *Bacillus subtilis* and *β-Streptococcus*) and four gram negative bacterial strains (*E. coli*, *Pseudomonas aeruginosa*. *Bacillus subtilis* and *Salmonella enteritidis*) by agar diffusion bioassay method. They have used the TeO_2 sol, prepared by utilizing acetic acid (AA) and gallic acid (GA) separately. The drawback of this work is that the antibacterial activity may not be only due to TeO_2 but also due to AA and GA used for TeO_2 sol preparation. This AA and GA have their own antimicrobial activity, which might influence the results. The synthesis of TeO_2 using catalyst may result in additional catalyst particle impurity in the as-prepared sample, which may require additional chemical treatment to remove the catalyst impurities. The present work represents the synthesis of pure sample without using any catalyst which is more reliable approach as compared to a catalyst based grown sample. However, catalytic particles may also play an important role for the improvement in reaction yield, reduction in temperature in the chemical process and promotion of scientific enantioselectivity in an asymmetric synthesis [18].

In present work, we have synthesized pure TeO_2 without any catalyst to provide more reliable, cost-effective, one-step synthesis with purity for investigating antimicrobial activity against bacterial strains such as gram positive (*S. aureus* and *S. pyogenes*) and gram negative (*E. coli* and *K. pneumoniae*). Also, we have used two fungus *A. nizer* and *C. albicans* to study the fungicidal activity of TeO_2. The microbial strains have been selected by their association with skin infection [19,20]. Simultaneously, the electrochemical behavior of TeO_2 is studied with a pH variation of different buffer solutions used as the electrolyte. Various workers [21–23] studied the electrochemical behavior of TeO_2 in the acidic medium using H_2SO_4, HCl, HNO_3 and $HClO_4$ of lower pH values and observed that the peak current increases with a decrease in pH value, and a maximum was observed in between the pH 1–2. However, in the present work, we have used phosphate, acetate and citrate buffer solutions of pH 3–8 and the redox current increases with the increase in pH and the maximum current obtained at pH 7. The advantage of this maximum redox current of TeO_2/ITO film is to enhance the sensitivity of the devices. On the basis of these studies, it is suggested that TeO_2 particles may be used for the fabrication of biosensors. From the literature survey, it is found that very limited studies on the interaction of TeO_2 particles with microorganisms have been reported [17]. Therefore, the present results based on the interaction of TeO_2 particles grown without a catalyst with microorganisms may be interesting and may find application for biomedical application.

2. Materials and methods

Tellurium (Te) metal powder (99.99%) and indium tin oxide (ITO) coated glass plates were procured Sigma-Aldrich and Balzers (UK) respectively. All other chemicals such as citric acid, sodium citrate solution, acidic acid, sodium acetate, HCl, and NaOH were procured from Fisher Scientific Ltd. To examine antimicrobial activity, the bacterial species such as *Staphylococcus aureus* (*S. aureus*), *Streptococcus pyogenes* (*S. pyogenes*), *Escherichia coli* (*E. coli*), *Klebsiella pneumoniae* (*K. pneumoniae*), *Aspergillus nizer* (*A. nizer*), *Candida albicans* (*C. albicans*) were gifted from a local hospital. Nutrient broth, nutrient agar, sabouraud agar and sabouraud were procured from Himedia. All these chemicals were used to prepare solutions in deionized water without further purification. The buffer solutions of phosphate, sodium acetate and citrate buffer were prepared at 0.1 M concentration with variable pH values in the range of 3–8, which were adjusted using diluted HCl and NaOH solution.

2.1. Synthesis of TeO_2

TeO_2 were synthesized on ITO glass substrate using CVD method [Fig. 1]. During this process, the tellurium metal powder (0.5 g) was dispersed in graphite powder and kept in a ceramic boat, and then placed inside the tubular furnace. Here, the graphite powder is used as supporting material that provides localized and uniform heating. The temperature was raised to 410 °C with a heating rate of 10 °C min^{-1}. A continuous flow of oxygen gas at a rate 60 mL min^{-1} was maintained throughout the process, and the tube pressure was kept at 60 Torr. Finally, TeO_2 was deposited onto ITO surface for 1 h at 410 °C.

2.2. Antimicrobial activity and growth kinetics

We employed disc method to study the antimicrobial activities of TeO_2 against pathogenic bacterial (*E. coli*, *K. pneumoniae*, *S. aureus* and *S. pyogenes*) and fungal strains (*A. nizer & C. albicans*) with two dilutions (10 μg 10 μL^{-1} and 1 μg 10 μL^{-1}) of TeO_2 in Dimethyl sulfoxide (DMSO). Discs were prepared from Whatman filter paper and sterilized in an autoclave. Each disc was impregnated with 10 μL of each dilution and was kept overnight for drying. A certain volume (200 μL) of bacterial and fungal cultures broth of age 36 and 48 h, respectively were transferred to their respective media plates and spread. The growth of the microorganisms was determined visually after the incubation for 24 h at 37 °C (bacteria) or 48 h at 30 °C (fungi). For microbial growth kinetics, bacterial culture broth of all four bacterial species was prepared in nutrient broth [24]. Whereas, TeO_2 suspension of 100 μg mL^{-1} was prepared by adding 1 mg TeO_2 into 9.8 mL nutrient broth and 200 μL of each bacterial cultures separately. The growth kinetics was studied using UV–Visible spectroscopy at a wave length of 580 nm. All the experiments were repeated three times under the same experimental conditions to check the reliability and reproducibility of the results.

2.3. Characterizations

The crystallinity of synthesized TeO_2 was investigated by using X-ray diffractometer (PANalyticX'pert PRO diffractometer with CuKα radiation at λ = 1.5406 Å). Morphology of TeO_2 was investigated by using Field Emission Scanning Electron Microscopy (FE-SEM, NOVA NANOSEM 450). To study the microstructure of as-prepared thin film, we used Transmission Electron Microscopy (TEM Tecnai G2 30S Twin, USA). For the preparation of TEM

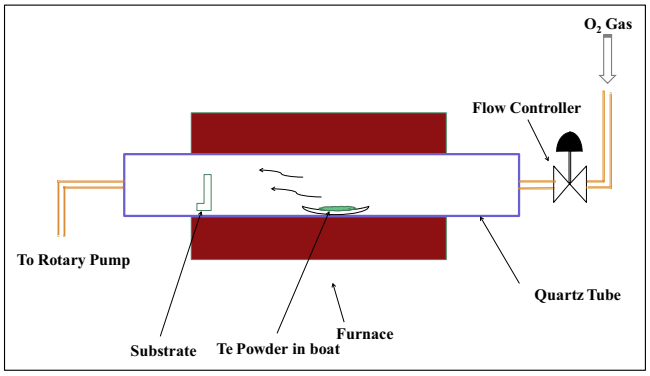

Fig. 1. Schematic diagram of synthesis of TeO_2.