



## Environmental Nanotechnology, Monitoring & Management

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# Control synthesis of metallic gold nanoparticles homogeneously distributed on hexagonal ZnO nanoparticles for photocatalytic degradation of methylene blue dye

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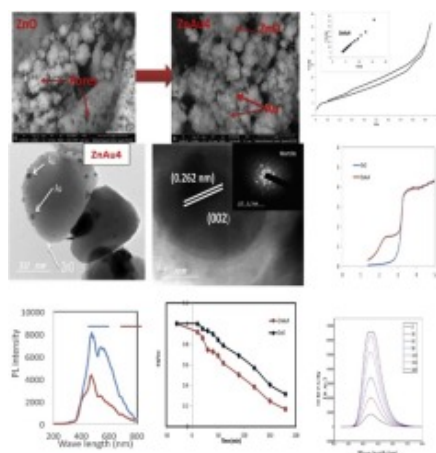
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### Abstract

The incorporation of an appropriate amount of metallic gold nanoparticles on ZnO surface has been demonstrated to be one of the effective way in improving the photocatalytic performance of ZnO. In this novel research work, we report a facile and economical sol-gel route for synthesis ZnO doped gold nanoparticles using PVP as pore and structure directing agent. The physicochemical properties of the prepared nanoparticles were investigated by (XRD), N<sub>2</sub>-adsorption-desorption isotherm, diffuse reflectance spectra (DRS), Energy dispersive X-ray (EDX), Photoluminescence (PL), Field emission electron microscope (FESEM) and high resolution transmission electron microscope (HRTEM). Homogeneous distribution of metallic gold nanoparticles of dimension 2–3 nm on the surface of mesoporous ZnO with hexagonal bipods structure was accompanied by exceptional degradation of MB dye. A pronounced reduction of band gap energy from 3.3 to 3.08 eV upon incorporation of Au nanoparticles on ZnO surface and the reduction in the photoluminescence intensity reflects a positive influence of metallic gold nanoparticles in shifting the photocatalytic response to visible region and reducing the electron-hole recombination. The enhanced photocatalytic performance of gold doped ZnO can be attributed to the greater ability of Au to accept electron from ZnO conduction band, which in turn reducing the recombination of photoinduced electron-hole couples and increasing in the life time of reactive species. Conclusively, it can be emphasized without doubt that these Au/ZnO nanoparticles would have considerable impact on future development of hybrid photocatalysts for efficient photocatalytic degradation of various organic pollutants.

## Graphical abstract

The incorporation of gold nanoparticles on ZnO surface enhances the textural feature and shift the photocatalytic response to the visible region



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## Introduction

Photocatalytic degradation of organic pollutants under solar energy is considered a recent challenge for wastewater treatment (Ahmed, 2012; Ahmed et al., 2013a; Ahmed et al., 2013b; Zhang et al., 2012; Xiang et al., 2015; Elbanna et al., 2016; Wang et al., 2015; Ismail et al., 2015; Narayanan and El-Sayed, 2005; Torrell et al., 2010). ZnO nanoparticles is a potential low-cost, non-toxic and stable photocatalyst for complete destruction of organic pollutants (Faisal et al., 2015; Zhou et al., 2013; Gomez-Solis et al., 2014; Klubnuan et al., 2016; Zhang et al., 2015; Chang et al., 2016). However, the low surface area, electron-hole recombination, wide band gap, and the photo-corrosion are the primary cause for limiting its industrialization. The incorporation of noble metals as Pt, Ag, or Au enhance the photocatalytic performance of ZnO by improving the electron-hole separation through formation of Schottky barrier (Ren et al., 2010; Chen et al., 2016; Lee et al., 2015; Senthilraja et al., 2015). Au is a promising nanoparticles for newer technologies in the twenty-first century owing to its ability to generate surface plasmon at desired wavelength. The proper design of Au-ZnO nanocomposite is expected to extend the photoexcitation response to visible region and boost the hot electron injection between gold nanoparticles and ZnO conduction band (Lu et al., 2016a; Ismail et al., 2016a; Lee et al., 2015; Manna et al., 2015; Chen et al., 2014; Yu et al., 2016). Many authors incorporate metallic gold nanoparticles on ZnO surface by impregnating gold salt solution onto ZnO surface followed by reducing  $\text{Au}^{3+}$  into metallic Au nanoparticles using  $\text{NaBH}_4$ , ascorbic acid or glucose as reducing agent. This rapid reduction of gold cations is accompanied by formation of various agglomerates of metallic gold nanoparticles on ZnO surface which preventing the light penetration on the photocatalyst surface and decrease the catalyst reactivity. The mild reduction of  $\text{Au}^{3+}$  as proposed in our research by thermal treatment of the sample at  $200^\circ\text{C}$  is a prime factor in controlling the dispersion of metallic gold nanoparticles on the active sites on ZnO surface that is responsible for enhancing the samples reactivity. In this research work, we make an attempts for synthesis mesoporous ZnO nanoparticles through sol-gel

route following by homogeneous incorporation of metallic gold nanoparticles on ZnO surface and on the pore mouth without using reducing agent. The influence of incorporation of appropriate amount of metallic Au nanoparticles on the structural, morphology, crystalline, and photocatalytic features of the ZnO toward degradation of methylene blue as cationic organic pollutant model dye was investigated. The nature of the reactive species that responsible for dye degradation was investigated using various scavengers.

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## Section snippets

### Materials

Zinc nitrate hexahydrate [ $\text{Zn}(\text{NO}_3)_6 \cdot 6 \text{H}_2\text{O}$  (99%)], Hydrogen tetrachloroaurate [ $\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$  (25% Au)], ammonia solution (25%), polyvinyl pyrrolidone were purchased from Sigma-Aldrich company...

### Methods

ZnO nanoparticles were prepared by adjusted sol-gel method adopting micelle template approach. About 30 g of zinc nitrate hexahydrate were dissolved in 100 ml distilled water with constant stirring for one hour. To the above solution, 5 ml of PVP (2 w/v %) was added with vigorous stirring for one hour. Then, a...

### X-ray diffraction

Fig. 1 depicts the diffraction pattern of the prepared nanoparticles that illustrates several sharp crystalline peaks at  $2\theta = 31.7, 34.4, 36.2, 47.5, 56.6, 62.8, 66.3, 67.9,$  and  $69.1$ , marked by their miller indices [(100), (002), (101), (102), (110), (103), (200), (112) and (201)] which reflects the existence of ZnO nanoparticles with Wurtzite structure (JCPDS no. 36-1451). On careful examination Fig. 1, one can notice that the diffraction pattern of Au/ZnO nanoparticles resembles the...

### Conclusions

In summary, novel hexagonal Au/ZnO nanoparticles were successfully prepared by sol-gel method using PVP as structure and pore directing agent. Homogeneous distribution of metallic Au nanoparticles on ZnO surface enhances the photocatalytic response in removal of methylene blue dye. The incorporation of gold nanoparticles on ZnO surface is associated with reduction in band gap energy and improve the efficiency of hole-electron recombination. The photocatalytic activity of ZnAu4 is nearly twice...

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