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Semiconductor Synthesis and Application for the Treatment of Ciprofloxacin Antibiotic by Solar Heterogeneous Phocatalysis

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Abstract: The objective of this study is to examine the relevance of treatment with the antibiotic ciprofloxacin by heterogeneous solar photocatalysis. The efficiency of the catalyst (ZnO) synthesized by the precipitation method is also determined. The solid- visible UV analysis showed that the spectrum reveals an absorption peak at 364 nm, which can be attributed to the band gap with of ZnO, in the case of the electron transition from the band of valence towards the conduction band (2p 3d) [3] and [4]. Under optimal conditions, initial ciprofloxacin concentration 10 ppm, ZnO dose 0.1 g/L and free pH (6) ; treatment by solar photocatalysis in the presence of ZnO showed a ciprofloxacin elimination yield of 87% is obtained after 240 min of treatment. The total degradation of ciprofloxacin is observed, by the disappearance of the ciprofloxacin absorption peak during the treatment time. Analysis by infrared spectroscopy showed that only two groups are obtained O-H and C=O at 3500 cm^{-1} and 1600 cm^{-1}, respectively after 240 min of oxidation. The BOD_5/COD ratio increased from 0.005 initially to 2.13 after 240 min of ciprofloxacin treatment by solar photocatalysis. The relevance of the solar photocatalytic process is shown. At the end of these results, the relevance of the treatment of ciprofloxacin antibiotic by solar photocatalysis in the presence of the semiconductor ZnO is shown.

Keywords: Semicoductor sythesis, Ciprofloxacin, Heterogeneous photocatalysis, Biodegradability, Depollution

Introduction

Thousands of tons of antibiotics are used in human and veterinary medicine to treat diseases, bacterial infections, etc.. However, their use is often partially metabolized by the body, so these substances or their metabolizes are continuously discharged into sewage treatment plants. The latter are the main sources of dispersion of antibiotics in the environment. (Balakrishna et al., 2017; Madikizel et al., 2017).

Thus, the presence and accumulation of antibiotics on natural waters constitute an emerging pollution leading to the disruption of ecosystems. In order to remove antibiotics from wastewaters, advanced oxidation processes has

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attracted great attention through generation of reactive oxygen species (ROS) (Miklos et al. 2018; Lado Ribeiro et al., 2019).

Among these processes, photocatalysis has been widely used in order to remove organic compounds in contaminated waters. This process involves irradiation of semiconductor, such as TIO_2 and ZnO, when energy photons are greater than the width of semiconductor band gap, electronic gaps (commonly called holes and noted h+ and electron overload noted e^-) are created (Zaviska et al., 2009; Miklos et al., 2018). These charges migrate to the surface, act as electron donors or electron acceptors and initiate the redox reactions, generating radical species (Zaviska et al., 2009). ZnO is considered as an environmentally friendly catalyst due to its non-toxic nature coupled with low cost (Sordello et al., 2019).

The aim of this study is to examine the relevance of treatment with the antibiotic ciprofloxacin by heterogeneous solar photocatalysis. The efficiency of the catalyst (ZnO) synthesized by the precipitation method is also determined.

Method

The zinc oxide (ZnO) used in this study was synthesized chemically (Figure 1). The semiconductor obtained is considered in order to depollute the solution loaded with ciprofloxacin antibiotic. Several analytical techniques were carried out in order to characterize and identify the semiconductor produced, namely scanning electron microscopy (SEM), UV-visible spectroscopy and Fourier transform infrared spectroscopy (FTIR).

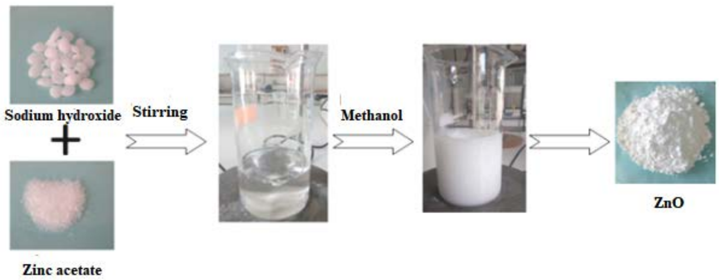

Figure 1. Zinc oxide synthesis steps

The degradation of ciprofloxacin was carried out in a photocatalytic reactor in Batch mode. A solution of ciprofloxacin with a dose of ZnO at free pH is treated in the presence of solar irradiation with magnetic stirring. Several analytical techniques were used to determine and follow the evolution of the residual concentration, the chemical oxygen demand, the biological oxygen demand over 5 days of the antibiotic ciprofloxacin during the treatment by heterogeneous photocatalysis.

Results and Discussion

Characterization of the Photocatalyst

The main physicochemical characteristics of the photocatalyst used in this study are determined by UV-Visible spectrophotometry of the solid, by infrared and by scanning electron microscopy (SEM).

Characterization by UV-Visible Spectroscopy and Infrared Spectrophotometry of Zinc Oxide

The Figure 2 shows the UV-Visible absorption spectrum of ZnO nanoparticles. The spectrum reveals an absorption peak at 364 nm, which can be attributed to the band gap width of ZnO, in the case of electron transition from valence band to conduction band (2p $\rightarrow$ 3d) (Chen et al., 2002; Fujita et al.,2014). The figure 3 show the infrared spectrum recorded for ZnO powder. It appears that the ZnO spectrum presents a band at λ= 420 cm^{-1} which is attributed to the stretching vibrations of the Zn-O bonds.

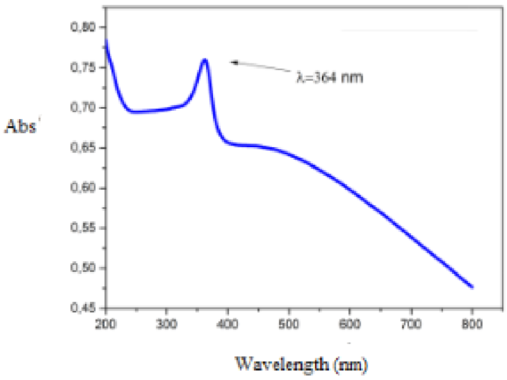

Figure 2. UV-Visible absorption spectrum of ZnO

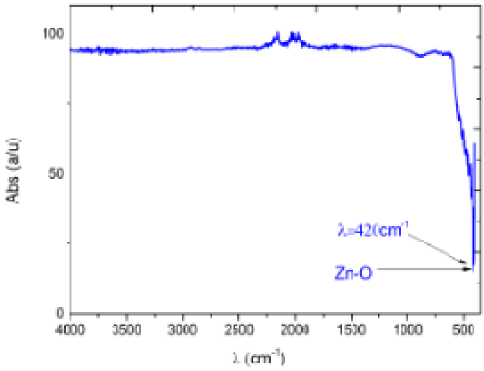

Figure 3. Infrared spectroscopy of ZnO

Characterization by Scanning Electron Microscopy (SEM)

The SEM image shows that the powder is composed of particles of spherical shape and small sizes of the nanometric order.

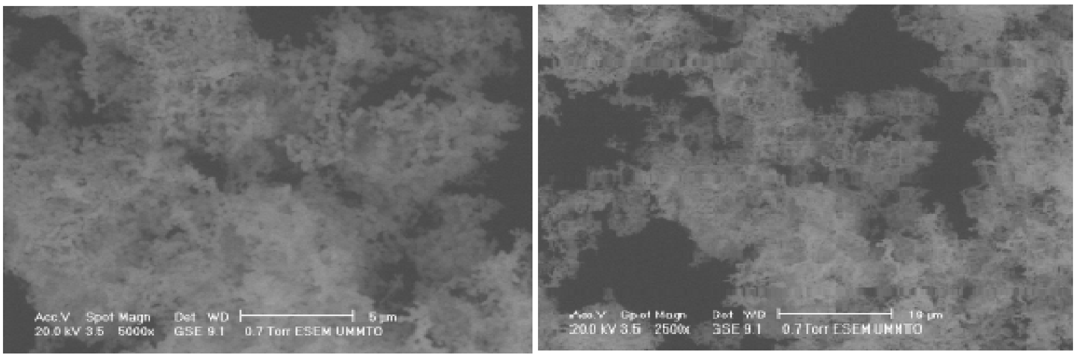

Figure 4. SEM micrograph of ZnO powder

Treatment by Photocatalysis of the Antibiotic Ciprofloxacin in the Presence of ZnO

Figures 5 and 6 show the photodegradation of ciprofloxacin under solar radiation in the presence of ZnO catalyst. Under optimal conditions (C_0=10 ppm, free pH (6) and the dose of ZnO=0.1g/L). An elimination yield of 87.44% is observed for ciprofloxacin, after 240 minutes of treatment.

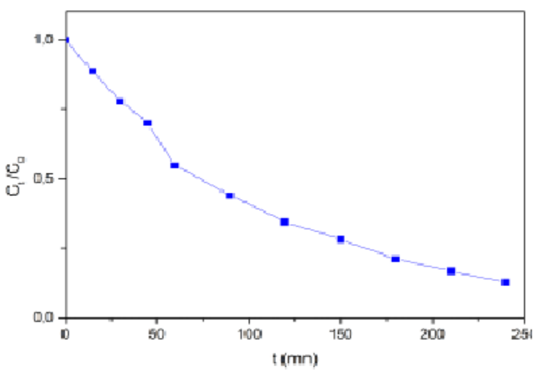

Figure 5. Degradation of ciprofloxacin by photocatalysis under optimal condition

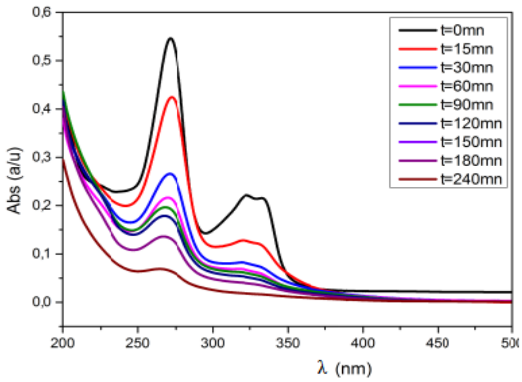

Figure 6. Spectral evolution of ciprofloxacin during treatment under optimal conditions

The degradation of ciprofloxacin showed the disappearance of the absorption peak of the antibiotic during the treatment. The relevance of the treatment by solar photocatalysis in the presence of ZnO is confirmed. Before treatment, the ciprofloxacin solution has several functional groups such as –C-H,=C-H, O-H, C=C, C=O, N-H, Figure 7. After treatment only two groups were observed during analysis by infrared spectroscopy.

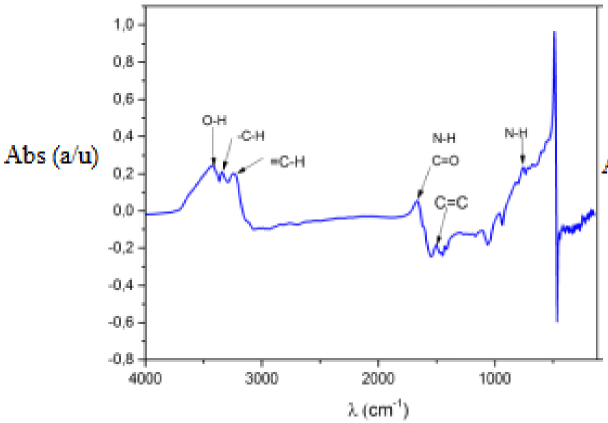

Figure 7. The infrared spectrum of solution ciprofloxacin before treatment

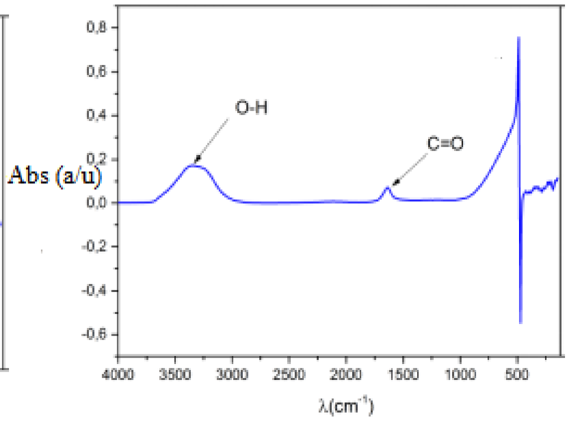

Figure 8. The infrared spectrum of solution ciprofloxacin after treatment

These O-H and C=O groups are obtained at 3500 cm^{-1} and 1600 cm^{-1}, respectively figure 8. This can probably be explained by the fact that the antibiotic ciprofloxacin was completely mineralized into H_2O and CO_2 during the treatment by solar photocatalysis in the presence of the ZnO catalyst.

Biodegradability Test

The BOD_5/COD ratio increased from 0.005 initially to 2.13 after 240 min of ciprofloxacin treatment by solar photocatalysis. The relevance of the solar photocatalytic process is shown.

Conclusion

At the end of these results obtained during this study, the treatment of recalcitrant pollutants by solar photocatalysis proves to be very effective and therefore a very useful technique for reducing water pollution while reducing the energy cost of treatment.

Recommendations

In this context, the exploitation of solar radiation is very interesting, particularly in a country like Algeria where the solar potential is very important, so it is interesting to apply the results obtained on a large scale and this, through the design a pilot fixed-bed photoreactor for the treatment of biorecalcitrant pharmaceutical effluents.

Scientific Ethics Declaration

The authors declare that the scientific ethical and legal responsibility of this article published in EPSTEM journal belongs to the authors.

Acknowledgements or Notes

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