Scientific Journal of Impact Factor(SJIF): 3.134
e-ISSN(O): 2348-4470
p-ISSN(P): 2348-6406

International Journal of Advance Engineering and Research Development

Volume 2,Issue 2, February -2015

Photocatalytic Degradation of Pharmaceutical Compounds Using Titanium Dioxide Nano Particles

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***Abstract** - The presence of drugs in the aquatic media has emerged in the last decade as a new environmental pollution. The main objective of the study was to evaluate the photocatalytic degradation of an aqueous solution containing antibiotic using TiO2 for the degradation of the Azithromycin and Esomeprazole. First of all TiO2 nano particles was successfully synthesized by the sol-gel method and characterization by the X-ray Diffraction (XRD) and Transmission Electron Microscope (TEM).The Photocatalytic degradation of the Esomeprazole and Azithromycin antibiotics in aqueous solution was studied under UV irradiation and TiO2 nano powder. Percentage Degradation of this antibiotics was improved when Titanium dioxide concentration was increased, the degradation was found maximum when TiO2 was 0.5 -1.0 gm/L. At a room temperature and PH 6, 60% COD reduction was achieved with a 150 min. in case of Esomeprazole and 67% COD reduction was achieved in a case of Azithromycin. The result show that COD removal was increased with increasing TiO2 concentration and decreasing initial concentration of antibiotics.*

***Key-Words:** Photocatalysis, Sol-Gel Process, TiO2 nano particles,Characterization, Antibiotics.*

I. INTRODUCTION

Water is basic requirement in all industrial processes, domestic and commercial activities, so the wastewater generated from different activities contains various contaminants which are harmful for human and environment.

Over the past few years, pharmaceuticals are considered as a prominent environmental problem due to their continuous input and persistence into the aquatic ecosystem. These compounds have been observed in surface water, ground water, sewage effluent and even in drinking water. After their use, pharmaceuticals waste or their metabolites are excreted into the effluents and reach the sewage treatment plants (STP). Among all the pharmaceutical compounds that cause contamination of the environment, antibiotics occupy an important place due to their high consumption rate in both veterinary and human medicine.

These substances are nature biologically active, and they have a limited biodegradability. Drugs can reach wastewaters by various routes: via industrial effluents (i.e. drugs industries residues) and via effluents from hospitals and via domestic effluents (waste material may contain both these compounds and their metabolites). The extended usage of hazardous pharmaceuticals is accompanied by an increased pollution of surface, ground and drinking water. The risk associated with pharmaceuticals in the environment is a rising issue of global concern as significant amounts have been detected. After their use, these substances are usually discharge only partially metabolized and end up in the sewage system, and these dangerous and hazardous contamination present in low concentration in drinking water is still a challenge. This number of drugs and their concentrations significant increase day by day and achieved concentrations in the ppb to low ppm range. They are different type of method is used for the Degradation of these non-biodegradable organic compounds, and degradation of this compounds is not possible by conventional biological treatment processes, so there has been a lot of interest in application of the advanced oxidation processes (AOP's) for the removal of such organic compounds. Many processes such as Photolysis, Photocatalytic oxidation, ozonation, Fenton oxidation, Electrochemical treatment, Biological treatment, Wet air oxidation and Membrane separation has been proposed for the degradation of these compounds even at low concentration.

Several of them are antibiotics that were demonstrated to lead to increased antibiotic-resistant pathogens in wastewaterand potentially in receiving streams. The semiconductor heterogeneous photocatalytic is a popular technique that possesses great potential for application in solving the increasingly serious problems of environmental pollution. Among several oxide semiconductor, Titanium oxide (TiO2) is a suitable material due to its strong oxidizing, biological and chemical inertness, and non-toxicity. Titanium dioxide, also known as Titanium oxide or Titania, is the naturally occurring oxide of Titanium. The photocatalytic properties of TiO2 depend strongly on the sample preparation conditions, crystal phase, surface area, size distribution, porosity, and the presence of additional compounds such as metal particles that are used to enhance the catalytic response.

II. EXPERIMENTAL

A. Materials

Aqueous solution ofEsomeprazole and Azithromycin were prepared in distill water. Titanium Tetrachloride ($TiCl_4$) was used as precursor and Ethanol (C_2H_5OH) was used as solvent.

B. Nanoparticles Synthesis Procedure

For synthesis of 4 volume% $TiCl_4$ in ethanol, 2 ml of $TiCl_4$ was slowly added drop wise, into 48 ml of ethanol, at room temperature (250C).A light yellow solution was obtained after adding all the $TiCl_4$.The solution was then gelatinized for time periods 4h in magnetic stirrer and each prepared solution was subjected to an aging process for 3 h. Finally, the gel solution was calcined a 400 ◦C for 1 h to form TiO_2 nano powder.

C. Characterization of Nano Particles

The particle size and size distribution ofnano particles can be determined using numerouscommercially available instruments. Instruments can be used for the analysis of dry powders andpowders dispersed in suspension.The morphology, size and purity of the nano particles are investigated by transmission electron microscope (TEM), Dynamic Light Scattering (DLS) and X-ray Diffraction (XRD), Brunauer-Emmett-Teller (BET), Scanning Electronic Microscope (SEM).
X-ray diffraction is one of the most important characterization tools used in solid state chemistry and materials science. We can determine the size and the shape of the unit cell for any compound most easily using X-ray diffraction. X-ray diffraction (XRD) measurement was performed to examine the structure of TiO_2 nanoparticles.

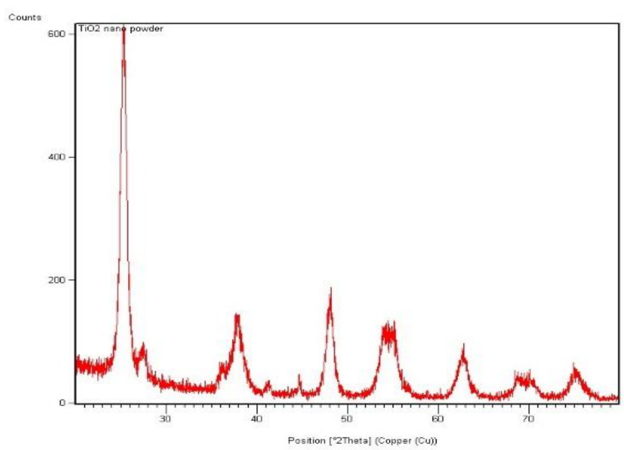

Fig. 1 XRD pattern of TiO2 nanoparticles

Figure (2) show the XRD patterns of TiO_2 nanoparticles synthesized at calcinations temperature of 400 °C, respectively. It can be observed that TiO_2 nanoparticles formed are amorphous as well as crystalline. Constant drop-wise slow addition of $TiCl_4$ can result in sharp crystal formation of TiO_2 nanoparticles.As seen from figure (1), a characteristic peak for TiO_2 particles ($2\theta = 25.5°$) is observed, which represents plane of anatase phase of TiO_2. By Debye-Scherre formula which is given below we can find the particle size.

$D = k\lambda/\beta\cos\theta$;
Where;
K (shape factor) =0.8-1.39(usually close to unity)
λ = wavelength of the radiation,
β= FWHM (full width at half maximum) in radians,

β= half-width (degree) π/180

$\boldsymbol{\theta}$= the position of the maximum of diffraction

With help of Debye-Scherre formula we can find a particle size, and by calculation we obtained a 8-12 nm size of the TiO2 nano particles.

TEM forms a major analysis method in a range of scientific fields, in both physical and biological sciences. TEM find application in cancer research, materials science as well as nanotechnology and semiconductor research. TEM image of TiO2 nano particles is given below:

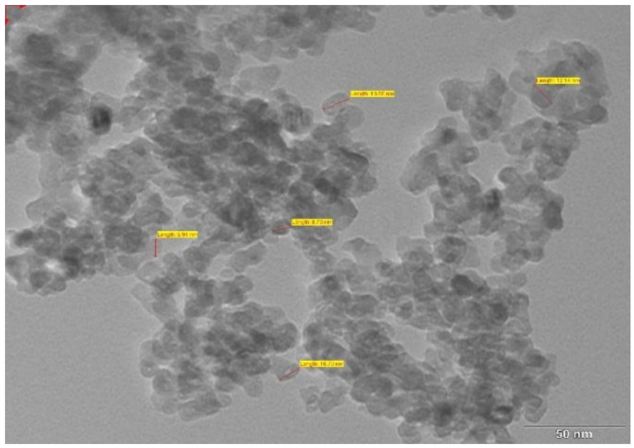

Fig.2 TEM image of TiO2 nanoparticles

D. Photocatalytic Degradation Experiment

The photocatalytic degradation of Esomeprazole was conducted in a glass beaker.Antibiotic aqueous solution was prepared by dissolving 40 mg of Esomeprazole in a 1000 ml distilled water. Specific amount of TiO2 catalyst was suspended in the aqueous solution of Esomeprazole. The reactor (Fig.3) was equipped with 6W UV lamp,protected by a quartz jacket.The batch reactor was placed on a magnetic stirrer for proper mixing of catalyst with the sample solution.

Same procedure for Azithromycin at initial concentration 150 gm in 1000 ml distilled water but catalyst loading is 1 gm/L.in aqueous solution. At a certain time interval sample is taken from glass beaker and COD of that sample was measured on a COD analysis apparatus.

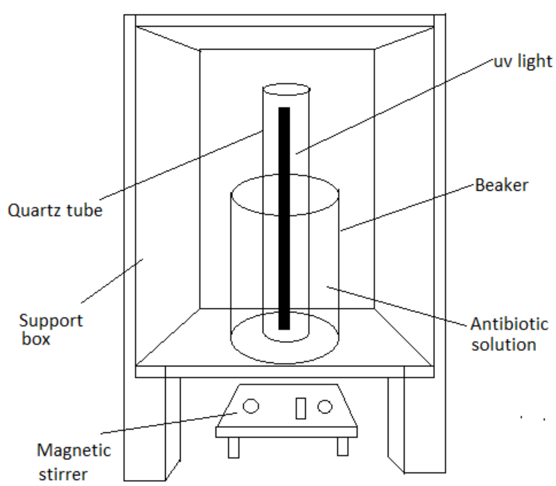

Fig. 3 Photocatalytic reactor

III. RESULT

To evaluate the degradation of antibiotics aqueous solution of Azithromycin and Esomeprazole was run under UV light. From the given graph, it is clear that degradation of these antibiotic aqueous was increased when TiO2 nano particles concentration is increased. The first graph is indicate the complete degradation of Esomeprazole at a 0.5 gm/L catalyst TiO2 nano particles with in a 150 min., when second graph indicate the complete degradation of Azithromycin at a 1 gm/L catalyst TiO2 nano particles with in a 150 min.

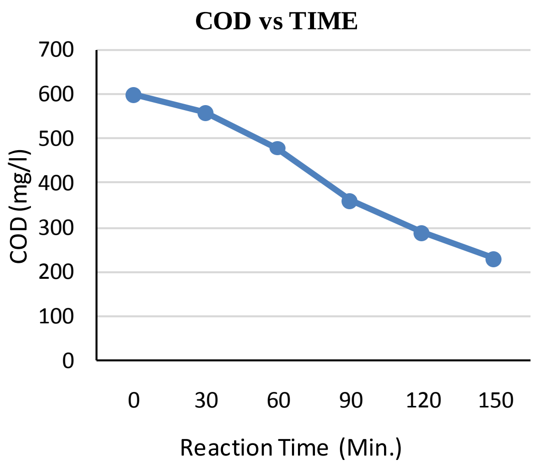

Fig.4 Effect of Irradiation time on Esomeprazole degradation

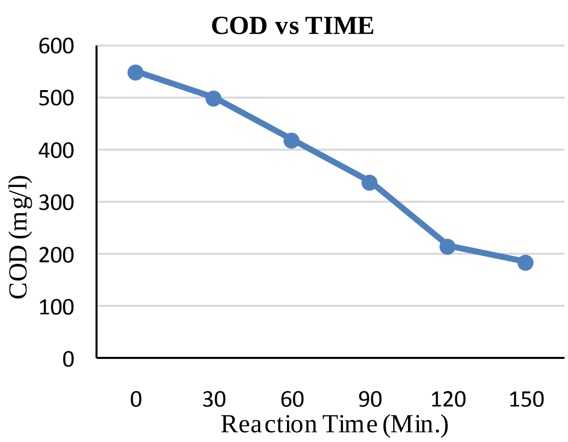

Fig.5 Effect of Irradiation time on Azithromycin degradation

IV. CONCLUSION

Photo-catalysis enhanced by UV irradiation appears to be a promising technology for the degradation of pharmaceuticals in wastewater.The advanced oxidation processes (AOP) appear as interesting tools in comparison with other techniques such as activated carbon adsorption, Fenton oxidation, Electrochemical treatment, Biological treatment, Wet air oxidation and Membrane separation and reverse osmosis.The main advantages of the process are lack of mass transfer limitations and operation at ambient conditions, and the catalyst is inexpensive, commercially available, non-toxic and photo chemically stable. In this experiment TiO2 catalyst used and prepared by sol-gel techniques .In the present study the photocatalytic treatment applied to the degradation ofAzithromycin and Esomeprazole antibiotics in the aqueous solution. The amount of catalyst is very important for the degradation of antibiotics.At a room temperature, 60% COD reduction was achieved with a

150 min. in case of Esomeprazole and 67% COD reduction was achieved in a case of Azithromycin.The result show that COD removal was increased with increasing TiO2 concentration.

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