Asian Journal of Green Chemistry 8 (2024) 336-348

Asian Journal of Green Chemistry

Original Research Article

View Article Online | View Journal

Tetracycline Drug Degradation Using Binary Hybrid Advanced Oxidation Processes of Photocatalytic (UV/ TiO_2 and UV/ZnO) in Aqueous Solutions

Haider Falih Shamikh Al-Saedi[a], Ola Hamad Salah[b], Russul Reidh Abass[c], Manal Morad Karim[d], Salima B. Alsaadi[e], Salam Ahjel[f], Zainab Jamal Hamoodah[g], Rathab Ali Ahmed[h,*]

[a]*Faculty of Pharmacy, Department of Pharmaceutics, University of Al-Ameed, Iraq*
[b]*Department of Medical Engineering Al-Manara College for Medical Sciences, Maysan, Iraq*
[c]*Al-Farahidi University, College of Medical Technology, Medical Lab, Techniques Department, Baghdad, Iraq*
[d]*College of Pharmacy, National University of Science and Technology, Dhi Qar, Iraq*
[e]*Department of Medical Engineering, Al-Hadi University College, Baghdad, Iraq*
[f]*Department of Pharmacy, Al-Zahrawi University College, Karbala, Iraq*
[g]*Department of Medical Engineering, Mazaya University College, Iraq*
[h]*Department of Medical Engineering, Al-Nisour University College, Baghdad, Iraq*

ARTICLE INFORMATION

Submitted: 2024-01-15
Revised: 2024-03-23
Accepted: 2024-03-24
Manuscript ID: AJGC-2403-1488
Checked for Plagiarism: Yes
Language Editor Checked: Yes

DOI: 10.48309/AJGC.2024.449148.1488

KEYWORDS

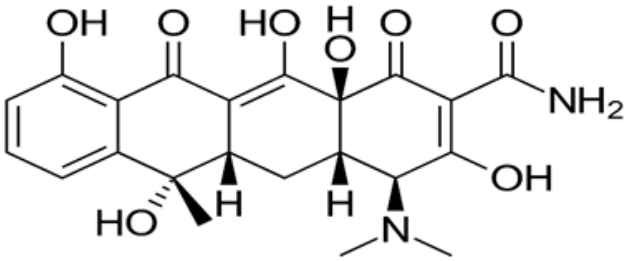
Tetracycline
Drug
Advanced oxidation processes
ZnO NPs
TiO_2 NPs

ABSTRACT

In this study, we examined the capacity of photocatalytic degradation catalyzed by TiO_2 NPs and ZnO NPs to remove the tetracycline TC drug in different solution pHs (3–10). TiO_2 NPs and ZnO NPs powders were prepared using the hydrothermal method and characterized via FESEM, TEM, TGA, and XRD. The effects of several method factors, such as weight of photocatalyst (0.1-0.4 g), concentration of TC drug (25-100 mg/L), different pH values (3-10), and irradiation time, on the removal of TC drug were studied. The photocatalytic degradation capacity of TC drug was fast in the present optimum condition (pH 10) and slower in (pH 3) of TiO_2 NPs and ZnO NPs; the degradation capacity of the TC drug under UV light decreased with an increasing concentration of TC drug. The photocatalytic degradation capacity of TiO_2 NPs is 81.33%, while the photocatalytic degradation capacity of ZnO NPs is 92.45 %. These data underline the use of effective, eco-friendly, easily available, and low-cost TiO_2 NPs and ZnO NPs as photocatalysts for degrading TC drugs in aqueous solutions. Therefore, as shown by the results, the photocatalytic degradation of the TC drug using zinc oxide nanoparticles is greater than that using TiO_2 nanoparticles.

Corresponding authors, email: hiv.nursing2022@gmail.com (R.A. Ahmed).
Tel.: 9647800744220.

Graphical Abstract



Introduction

The environmental risks posed by the effluents generated by the wastewater industry are classified as the main source of groundwater pollution [1, 2]. Wastewater resulting from hospital waste, including drugs and pharmaceuticals, constitutes one of the most hazardous types of waste, causing numerous diseases in animals [3-5]. Many semiconductors have been used as photo catalysts to extract various liquid wastes from their aqueous solutions. Pharmaceutical pollutants in water contaminated with high temperature, acidity and other soluble substances are affected [6, 7]. The main pollution in wastewater comes from waste from pharmaceutical factories and hospitals. There are many techniques to get rid of these dangerous pollutants from wastewater, as photo-oxidation is considered as a very important technology that is used in many fields and has high efficiency in removing toxic liquid pollutants in Water environment. Some of the common ways utilized to remediate pharmaceuticals pollutants like Tetracycline TC and its degradation products like: coagulation, membrane filtration adsorption, sedimentation and photo degradation [8, 9]. The use of Advanced Oxidation Processes (AOPs) as a potential solution. AOPs are driven via generation of highly oxidizing free radicals which are talented of mineralizing pharmaceuticals and therefore removal the formation of pollutions. Photo-catalysis is one of the Advanced Oxidation Processes that has received substantial attention due to its potential for being economically useful at a commercial scale [10]. TiO_2 NPs has been the photo catalyst of choice due to its eco-friendly, chemical stability, and high degradation capacity when illuminated with UV light .There are three natural poly-morphs of TiO_2, brookite, rutile, and anatase [11-13].

Zinc oxide (ZnO) is a wide band gap (Eg = 3.37 eV at 25 °C, near-UV spectral region) semi-conductor with high exaction binding energy (60 meV) enabling persistence of excitonic emission processes at or above room temperature. It has great potential for various applications, such as UV light emitters, photo catalysts, surface acoustic wave devices, piezoelectric transducers, optical waveguides [14, 15]. Among metal oxides, ZnO is a prototypical n-type material with numerous applications, including catalysts, gas sensors, visitors, transparent electrodes, etc. Renewed interest has recently emerged for its ultraviolet light emission capabilities [16-19] .

Titanium dioxide is a robust semiconductor material with a band gap of 3.2 eV which has been extensively explored for heterogeneous photo catalysis due to its non-toxic nature and easy availability. Nevertheless, wide band gap of

TiO_2 restricts its operational range to only UV region that is less than 5% of whole solar spectrum, relatively fast recombination of photo-exactions (e^-, h^+) is another drawback associated with TiO_2 that lowers its photo catalytic performance. One constraint associated with doping approach is that it can sometimes also reduce the redox potential of charge carrier species that results in decline of photo catalytic efficiency [20-26].

Tetracycline, which has different brand names, is named for its four ("tetra-") hydrocarbon rings ("-cycl-") derivation ("-ine") [27] is an antibiotic in the tetracycline's family of drugs, utilized to treat a number of infections, like plague, cholera, acne, brucellosis, malaria, and syphilis. Its chemical formula is $C_{22}H_{24}N_2O_8$ with molar mass of 444.440 g·mol^{-1}, and chemical structure of a drug, as displayed in Figure 1 including side effects have diarrhea, poor tooth, sun burning, vomiting, kidney problems rash, and loss of appetite [1-3, 27, 28].

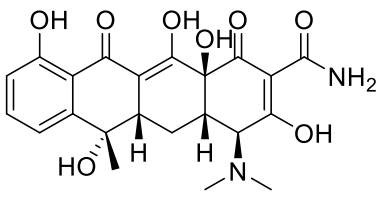


Figure 1. Chemical structure of tetracycline drug [27]

Experimental

Materials and Methods

All of the chemical reagents used (Sigma-Aldrich), all chemicals used in the research that degree analytical and used directly without purification (i.e. Titanium Dioxide Purity (99.0%), Zinc Oxide Purity (99.0%)). Tetracycline TC drug was purchased from factory Samara-Iraq, the spectra of UV–Vis absorption of solution 100 mg/L of TC drug was analyzed, at 235 nm was chosen as indicator for removal rates of TC drug. Titanium dioxide nanoparticles and zinc Oxide were previously prepared by the hydrothermal method [4, 29].

Photo catalytic degradation experiments

The UV-A light source (wavelength: 366 nm), was placed at a distance from solution surface in the reactor. The source of UV-A light open after on 10 min started of the reaction; 100 mL of the solution TC drug with concentration 50 mg/L was placed in a glass beaker, an appropriate 0.3 g from catalyst TiO_2 NPs or ZnO NPs was added to it, and light source, magnetic stirrer, were turned on for photocatalytic experiment. The absorbance was measured, and the concentration after photocatalytic degradation Ct mg/L was calculated according in calibration curve. The PDE% was selected as the evaluation index for the photo degradation of TC drug calculated in Equation (1).

$$\text{PDE}\ (\%) = (C_0 - C_t)/C_0 \times 100 \qquad (1)$$

Where, PDE% is percentage of removal; C_0 is the initial concentration; and C_t is the residual concentration after a selective time of degradation of drug under studying.

Results and Discussion

FE-SEM image of different structures are displayed ZnO NPS and TiO_2 NPs it was conducted to reveal the morphological characteristics of different samples. The results are depicted in Figure 2a, in which a zinc oxide nanoparticle was observed in the form of a white spherical structure stacked in regular layers, one above the other and in Figure 2b titanium oxide nanoparticles appeared in the form of a spherical structure that was brown color and was not arranged in organized layers,

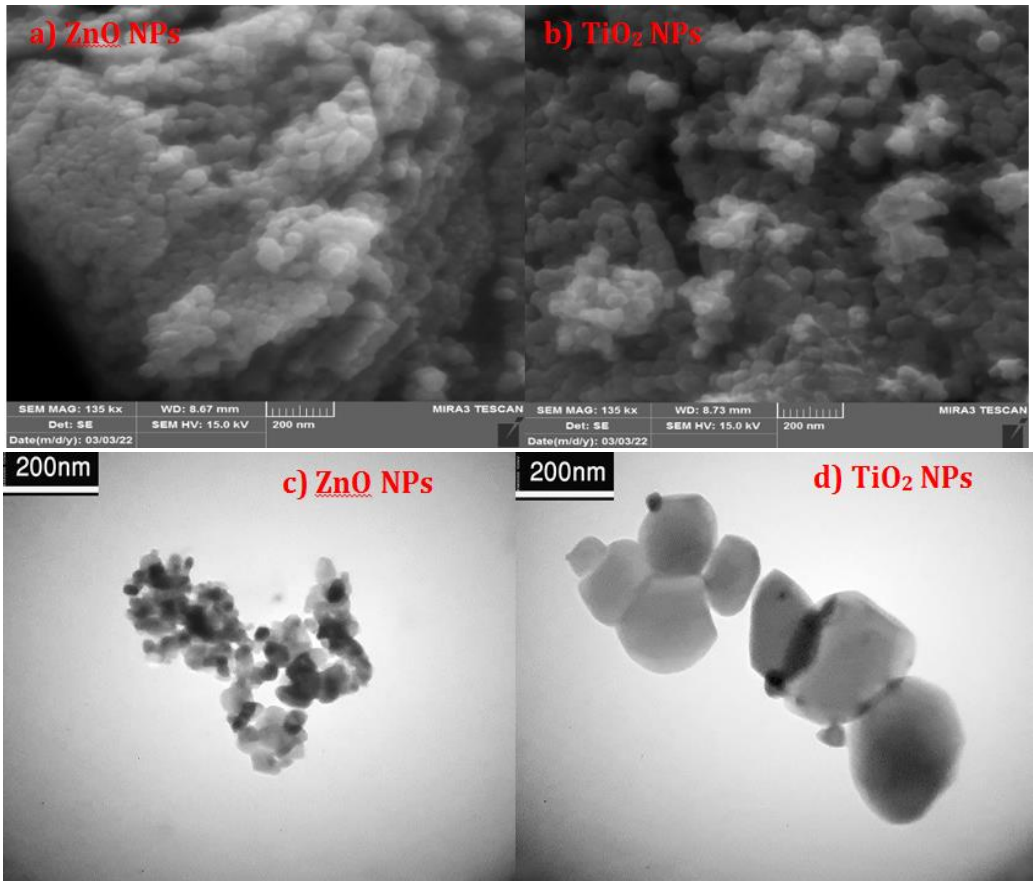


Figure 2. EF-SEM image a) TiO_2 NPs, b) ZnO NPs and TEM image, and c) TiO_2 NPs , d) ZnO NPs

but rather in the form of a random and disorganized spherical structure [6, 30].

TEM image was used to determine the particle size, morphology and crystal structure of TiO_2 and ZnO nanoparticles. Zinc oxide appears in the form a white, spherical crystalline structure. This is evidence of the purity of the prepared surface and also of thecalcination at 500 °C that increased the purity of zinc oxide nanoparticles [13, 31, 32]. The particle structure of TiO_2 NPs is presented in a spherical stacked structure and is uniformly indicated as a dense small black spherical packed together closely, as shown in Figure 2d.

X-Ray diffraction patterns of TiO_2 NPs

The diffraction peaks were observed at 25.3°, 37.9°, 48.2°, 54.5°, 55.3°, 62.02°, and 68.7°, which correspond to the (101), (108), (004), (112), (211), (200), (106), and (212) lattice planes of anatase TiO_2 the crystal shape of the diffraction peaks appear clear and sharp, as depicted in Figure 3a [33, 34]. X-Ray diffraction patterns of zinc oxide nanoparticles are observed in Figure 3 b.

The diffraction pattern

Nine peaks appear at 31.7°, 34.4°, 36.2°, 47.5°, 56.5°, 62.8°, 66.4°, 67.9°, and 69.1, which correspond to the (100), (002), (101), (102), (110), (103), (200), (112), and (201), reflection planes, at the same order. The ZnO PNs better crystalline structure and pure phase with highly crystalline produced due to very sharp and intense peaks [35, 36].

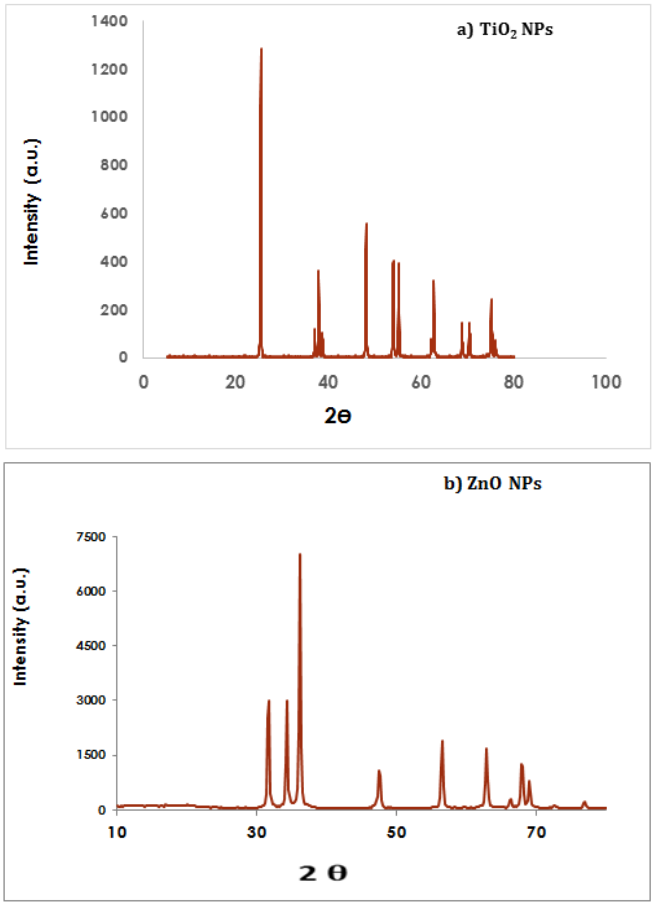


Figure 3. X-ray diffraction of a) TiO_2 NPs and b) ZnO NPs

The most important technique used to determine the thermal stability of the preparation surface is the thermogravimetric analysis (TGA) technique, where the mass of the sample is determined versus temperature or time, to study the stability and thermal stability of the nanopartical, and also indicate the purity of the nanocomposites, the TGA curve for (TiO_2 NPs and ZnO NPs) in the temperature range of 10-560 °C. The TGA curve of TiO_2 NPs and ZnO NPs appears to show no thermal decomposition or weight loss [5, 37]. It appears as a straight line due to the high thermal stability of the nanocomposite. Therefore, TiO_2 NPs and ZnO NPs exhibit higher resistance to thermal degradation compared to other nanoparticles, as shown in Figure 4.

Effect of TC drug concentration

To evaluate the suitability of the ZnO NPs and TiO_2 NPs in solution as a photo catalyst for degradation of TC drug, the initial concentration of TC drug solution is crucial in determining the photocatalytic degradation of TC drug solution. The effect of concentration of TC drug on degradation capacity was studied via altering the concentration TC drug (25-100 mg/L) at weight of each ZnO NPs and TiO_2 NPs 0.3 g. The degradation capacity of TC drug under UV- light decreased with increasing concentration of TC drug. As a result, the capacity quantity of OH radical available for TC drug photocatalytic degradation decreases, causing in a drop in TC drug degradation [13, 37], as demonstrated in Figure 5.

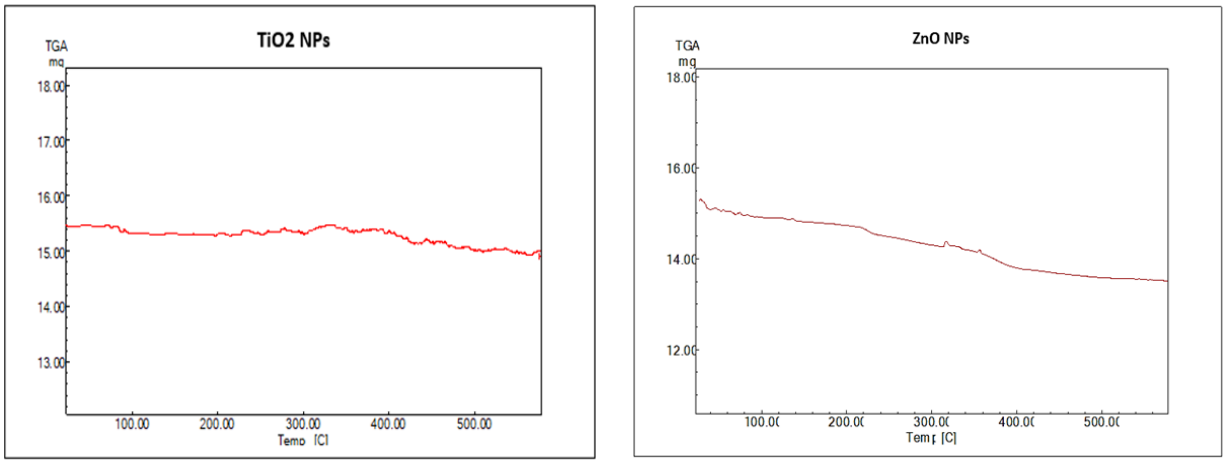


Figure 4. Thermogravimetric analysis (TGA) technique of ZnO NPs and TiO_2 NPs

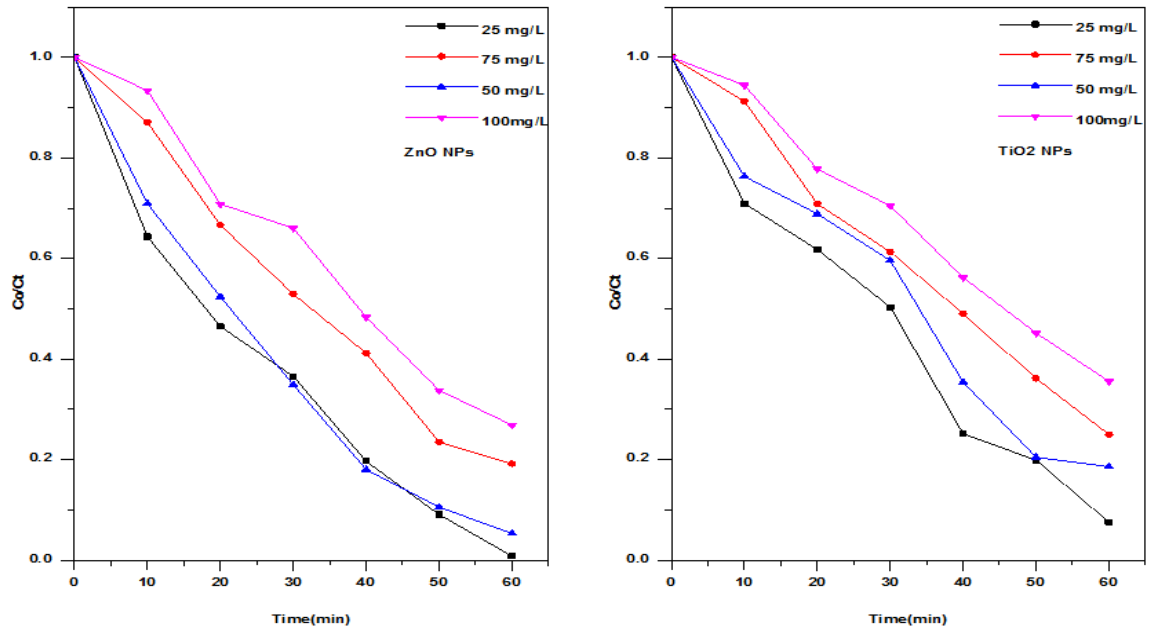


Figure 5. Effect of photocatalytic degradation of TC drug at different concentration by use two surface ZnO NPs and TiO_2NPs

Effect of mass catalyst

To investigate the effect of the catalyst on the photocatalytic efficiency of stimulation, different weights of each of ZnO NPs and TiO_2 NPs were used, ranging from 0.1 to 0.4 g and 50 mg/L concentration of TC drug and at 25 °C, as demonstrated in Figure 6. From Figure 6, it is obvious that when the amount of the catalyst for two surface ZnO NPs and TiO_2 NPs increase, the photocatalytic degradation increases first and remains constant at a given scale. However, when the amount of the catalyst exceeds 0.4 g/L, the degradation does not change. This may be due to the additional free catalyst particles that form in the aggregation process, and also due to the phenomenon known as screening [2, 5, 38].

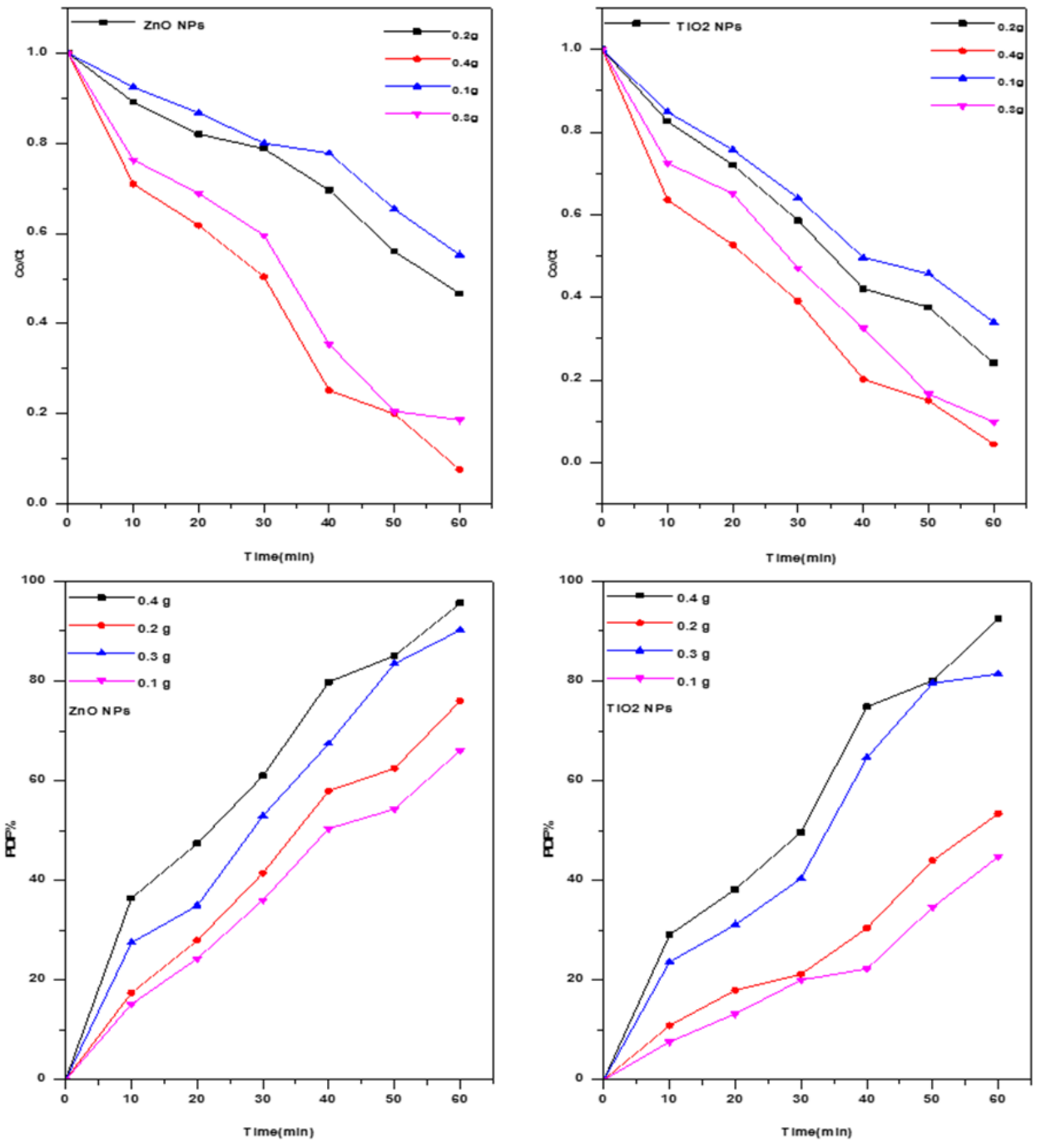


Figure 6. Effect of photocatalytic degradation of TC drug at different masses of a) ZnO NPs and b) TiO_2 NPs

Effect of pH

pH is important factor in the photo degradation of pharmaceutical. The pH can greatly affect the TC drug solution for the photocatalytic process as well as the properties of the TC drug and its surface zinc oxide and titanium dioxide nanoparticles. Figure 7 shows the effect of pH on the photocatalytic degradation process of TC drug solution using 0.3 g of each one (i.e. zinc oxide and titanium dioxide nanoparticles). It has been observed

that the activity of the photocatalytic process is low in acidic media and increases with increasing the pH of TC drug solution until it reaches the basic pH [13]. It was found that the decomposition efficiency in alkaline media is almost constant at pH 10. Thus, the pH of the solution affects the absorption of TC drug solution and the separation of photo generated charge carriers on the surface of the photo catalyst zinc oxide and titanium dioxide nanoparticles. According to the zero charge point, the surface of TiO_2 NPs and ZnO NPs are supposed to be positively charged in acidic solution and negatively charged in alkaline solution. Accordingly, pH variation can affect the efficiency of photo degradation of TC drug solution [23, 39].

Photo catalytic degradation: A Comparison among TiO_2NPs and ZnO NPs

A solution of Tetracycline TC drug 50 mg/L was degraded under UV-light at time of 15 min in the dark (adsorption). After that in every solution, ZnO NPs or TiO_2 NPs 0.3 g/200 ml was added. Photo degradations have been carried out for 60 min. After this, photo catalytic activity percentages. Photocatalytic degradation and comparison between TiO_2 NPs and ZnO NPs shows that ZnO NPs degrade active principles in solution faster than TiO_2 NPs [26]. Thus, degradation percentages were evaluated with the dark (adsorption) associated no any photocatalytic degradation of solution drug but give best degradation of TC drug solution after 10 min and a comparison of photocatalytic degradation at time of 60 min shows that difference between TiO_2 NPs and ZnO NPs for TC drug is outstanding and specially for ZnO NPs give the best photocatalytic degradation, because ZnO NPs has a large surface area and good dispersion in the solution while TiO_2 NPs has a small specific surface area, compared to ZnO NPs, and requires a longer time than zinc oxide to decompose TC drug 100% [8,9] as shown in Figure 8.

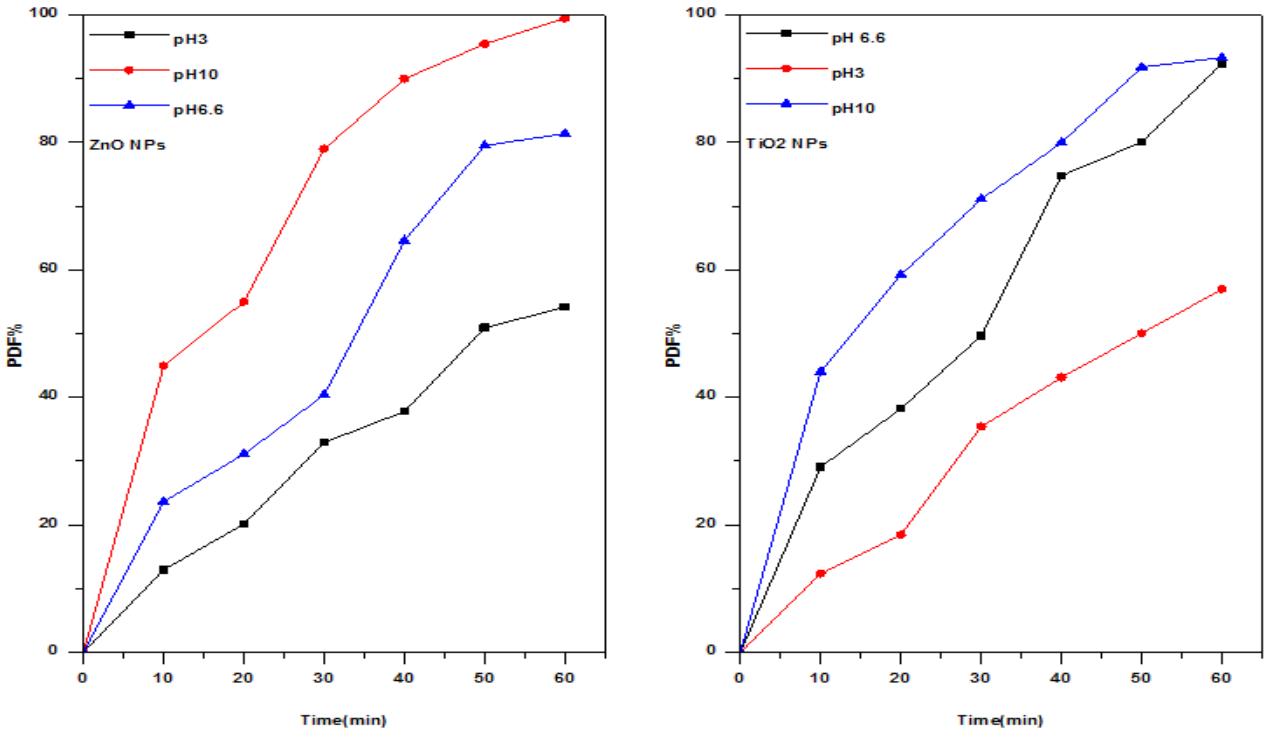


Figure 7. Effect of photocatalytic degradation of TC drug at solution pH a) ZnO NPs and b) TiO_2 NPs

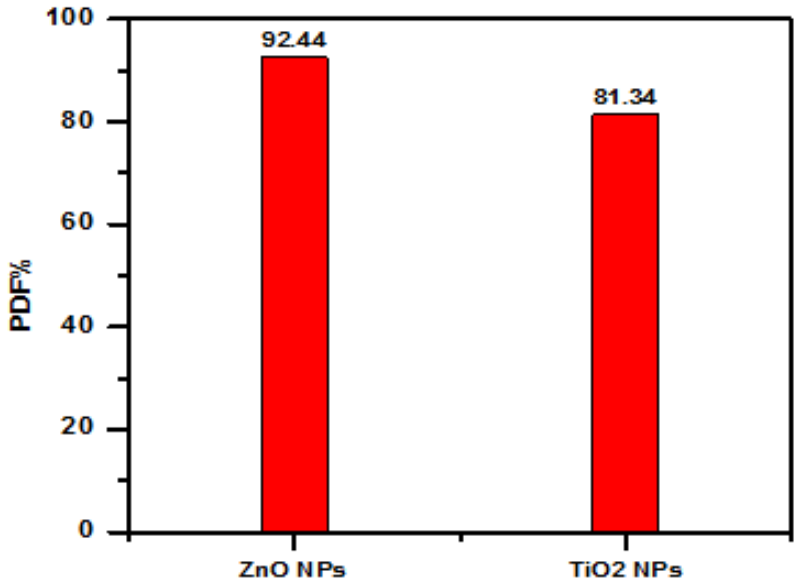


Figure 8. Photocatalytic degradation: A comparison between TiO_2 NPs and ZnO NPs

Conclusion

In this study, the efficiency of photodegradation of zinc oxide and titanium dioxide nanoparticles in decomposing pharmaceuticals in an aqueous solution was determined. Among several types of pharmaceuticals, the tetracycline TC drug was chosen. The decomposition of a TC drug solution was mainly carried out under UV irradiation for a fixed period of time in the presence of a small amount of zinc oxide and titanium dioxide nanoparticles. ZnO NPs decompose TC drugs faster than TiO_2 NPs, so ZnO NPs show better photodegradation efficiency than TiO_2 NPs. Because ZnO NPs have a large surface area and good dispersion in the solution, while TiO_2 NPs have a small specific surface area compared to ZnO NPs, and require a longer time than zinc oxide to decompose TC drug 100%.

Disclosure Statement

No potential conflict of interest was reported by the authors.

Funding

This study was funded by Payame Noor University (PNU) Research Council.

Authors' Contributions

All authors contributed to data analysis, drafting, and revising of the article and agreed to be responsible for all the aspects of this work.

Orcid

Haider Falih Shamikh Al-Saedi
https://orcid.org/0000-0003-0862-9543
Ola Hamad Salah
https://orcid.org/0000-0002-3194-1749
Russul Reidh Abass
https://orcid.org/0000-0002-3697-3926
Manal Morad Karim
https://orcid.org/0000-0003-0061-0389
Salima B. Alsaadi
https://orcid.org/0009-0004-3760-4489
Salam Ahjelf
https://orcid.org/0009-0008-8881-6022
Zainab Jamal Hamoodah
https://orcid.org/0000-0002-5688-441X
Rathab Ali Ahmed
https://orcid.org/0009-0002-7479-3333

References

[1]. Jeong J., Song W., Cooper W.J., Jung J., Greaves J., Degradation of tetracycline antibiotics: mechanisms and kinetic studies for advanced oxidation/reduction processes, *Chemosphere*, 2010, **78**:533 [Crossref], [Google Scholar], [Publisher]

[2]. a) Ahmad F., Zhu D., Sun J. Environmental fate of tetracycline antibiotics: degradation pathway mechanisms, challenges, and perspectives, *Environmental Sciences Europe*, 2021, **33**:64 [Crossref], [Google Scholar], [Publisher]; b) Kumar M R., Misganaw G W., Sagar P S., Jayanthi Juturi L., Shimelash Admasu A., P K., M V., Gaddala B. Investigation of Characterisation for Biosorption of Congo Red from Textile Wastewater Using Spathodea Campanulata Leaves: FTIR, SEM, and XRD Analysis, *Chemical Methodologies*, 2023, **7**:605 [Crossref], [Publisher]; c) Abegunde S., Idowu K., Enhanced Adsorption of Methylene Blue Dye from Water by Alkali Treated Activated Carbon, *Eurasian Journal of Science and Technology*, 2023, **3**:109 [Crossref], [Google Scholar], [Publisher]

[3]. a) Nguyen T.M.P., Van H.T., Yılmaz M., Hoang T.K., Nguyen Q.T., Vi T.M.H., Nga L.T.Q., Removal of Tetracycline from aqueous solution using composite adsorbent of ZnAl layered double hydroxide and bagasse biochar, *Environmental Technology & Innovation*, 2022, **28**:102914 [Crossref], [Google Scholar], [Publisher]; b) Elhadeuf K., Bougdah N., Fouzia B., Chikhi M. Experimental Study and Modeling by the Complete Factorial Plan of the Elimination of Textile Dye by Electrocoagulation Using Aluminum from Recycled Cans, *Chemical Methodologies*, 2023, **7**:183 [Crossref], [Google Scholar], [Publisher]; c) Vaeli N. Laboratory Study of Effective Factors on How to Extract Carvacrol from Oliveria Decumbens Plant with the Help of Supercritical Fluid CO2 and Using Ultrasound Waves, *Eurasian Journal of Science and Technology*, 2022, **2**:34 [Crossref], [Publisher]

[4]. Alhattab Z.D., Aljeboree A.M., Jawad M.A., Sheri F.S., Obaid Aldulaim A.K., Alkaim A.F. Highly adsorption of alginate/bentonite impregnated TiO2 beads for wastewater treatment: Optimization, kinetics, and regeneration studies, *Caspian Journal of Environmental Sciences*, 2023, **21**:657 [Crossref], [Google Scholar], [Publisher]

[5]. Reza K.M., Kurny A., Gulshan F. Parameters affecting the photocatalytic degradation of dyes using TiO 2: a review, *Applied Water Science*, 2017, **7**:1569 [Crossref], [Google Scholar], [Publisher]

[6]. Kamari Y., Ghiaci M. Preparation and characterization of ibuprofen/modified chitosan/TiO2 hybrid composite as a controlled drug-delivery system, *Microporous and Mesoporous Materials*, 2016, **234**:361 [Crossref], [Google Scholar], [Publisher]

[7]. Bakhshi V., Poursadegh H., Amini-Fazl M.S., Salari D., Javanbakht S. Synthesis and characterization of bio-nanocomposite hydrogel beads based on magnetic hydroxyapatite and chitosan: a pH-sensitive drug delivery system for potential implantable anticancer platform, *Polymer Bulletin*, 2023, 1 [Crossref], [Publisher]

[8]. Verdini F., Calcio Gaudino E., Canova E., Colia M.C., Cravotto G. Highly efficient tetracycline degradation under simultaneous hydrodynamic cavitation and electrical discharge plasma in flow, *Industrial & engineering chemistry research*, 2023, **62**:19311 [Crossref], [Google Scholar], [Publisher]

[9]. Ayoub M. Degradation of tetracycline using nanoparticles of zero-valent iron and copper, *Water Practice & Technology*, 2022, **17**:246 [Crossref], [Google Scholar], [Publisher]

[10]. Aljeboree A.M., Alhattab Z.D., Altimari U.S., Aldulaim A.K.O., Mahdi A.K., Alkaim A.F. Enhanced removal of amoxicillin and

chlorophenol as a model of wastewater pollutants using hydrogel nanocomposite: Optimization, thermodynamic, and isotherm studies, *Caspian Journal of Environmental Sciences*, 2023, **21**:411 [Crossref], [Google Scholar], [Publisher]

[11]. Karampour F. Green Synthesis and Characterization of ZnO Nanoparticles from Echinophora Platyloba DC. Extract and HSA Interaction Study. *Asian Journal of Green Chemistry*, 2023, **7**:149 [Crossref], Publisher]

[12]. Ebrahimi A., Samari F., Eftekhar E., Yousefinejad S. Rapid and efficient colorimetric sensing of clindamycin and Fe^{3+} using controllable phyto-synthesized silver/silver chloride nanoparticles by Syzygium cumini fruit extract, *Journal of analytical science and technology*, 2022, **13**:10 [Crossref], [Google Scholar], [Publisher]

[13]. Oluwole A.O., Olatunji O.S. Photocatalytic degradation of tetracycline in aqueous systems under visible light irridiation using needle-like SnO 2 nanoparticles anchored on exfoliated gC 3 N 4, *Environmental Sciences Europe*, 2022, **34**:1 [Crossref], [Google Scholar], [Publisher]

[14]. Mashkour M., Al-Kaim A., Ahmed L., Hussein F. Zinc oxide assisted photocatalytic decolorization of Reactive Red 2 dye, *International Journal of Chemical Sciences*, 2011, **9**:969 [Google Scholar], [Publisher]

[15]. Nithya R., Ragupathy S., Sakthi D., Arun V., Kannadasan N. Photocatalytic efficiency of brilliant green dye on ZnO loaded on cotton stalk activated carbon, *Materials Research Express*, 2020, **7**:075002 [Crossref], [Google Scholar], [Publisher]

[16]. Matinise N., Fuku X., Kaviyarasu K., Mayedwa N., Maaza M. ZnO nanoparticles via Moringa oleifera green synthesis: Physical properties & mechanism of formation, *Applied Surface Science*, 2017, **406**:339 [Crossref], [Google Scholar], [Publisher]

[17]. Dhal J.P., Mishra B.G., Hota G. Hydrothermal synthesis and enhanced photocatalytic activity of ternary Fe 2 O 3/ZnFe 2 O 4/ZnO nanocomposite through cascade electron transfer, *RSC advances*, 2015, **5**:58072 [Crossref], [Google Scholar], [Publisher]

[18]. Ali R., Radhi I.M., Ismail A.A., Abdulrazzak F.H. Modified ZnO for efficient photo-catalysis by silver/graphite oxide nanoparticles, *Journal of Global Pharma Technology*, 2009 [Google Scholar], [Publisher]

[19]. Komala Dewi R., H Kzar H., Suksatan W., Taukhid I., Setia Budi H., F Alkaim A., Aravindhan S. Synthesis and morphological investigation of hydroxyapatite/zinc oxide and evaluation its application in removal of organic pollutants, *Journal of Nanostructures*, 2021, **11**:368 [Crossref], [Google Scholar], [Publisher]

[20]. Laysandra L., Sari M.W.M.K., Soetaredjo F.E., Foe K., Putro J.N., Kurniawan A., Ju Y.H., Ismadji S. Adsorption and photocatalytic performance of bentonite-titanium dioxide composites for methylene blue and rhodamine B decoloration, *Heliyon*, 2017, **3** [Crossref], [Google Scholar], [Publisher]

[21]. Kamari Y., Ghiaci P., Ghiaci M. Study on montmorillonite/insulin/TiO2 hybrid nanocomposite as a new oral drug-delivery system, *Materials Science and Engineering: C*, 2017, **75**:822 [Crossref], [Google Scholar], [Publisher]

[22]. Thakur S., Arotiba O. Synthesis, characterization and adsorption studies of an acrylic acid-grafted sodium alginate-based TiO2 hydrogel nanocomposite, *Adsorption Science & Technology*, 2018, **36**:458 [Crossref], [Google Scholar], [Publisher]

[23]. Wu T.S., Wang K.X., Li G.D., Sun S.Y., Sun J., Chen J.S. Montmorillonite-supported Ag/TiO2 nanoparticles: an efficient visible-light bacteria photodegradation material, *ACS applied materials & interfaces*, 2010, **2**:544 [Crossref], [Google Scholar], [Publisher]

[24]. Abd Alrazzak N., Aowda S.A., Atiyah A.J. Removal Bismarck Brown G dye from aqueous solution over a composite of triazole-polyvinyl chloride polymer and zinc oxide, *Oriental Journal of Chemistry*, 2017, **33**:2476 [Google Scholar], [Publisher]

[25]. Saad S.T., Al-Gubury H.Y., Alrazzak N. Photocatalytic degradation of monoazo dye in ethanol using zinc oxide in ultra-violet radiation, *Asian Journal of Chemistry*, 2018, **30**:2334 [Google Scholar], [Publisher]

[26]. Syah R., Hussein Altajer A., F Abdul-Rasheed O., Amri Tanjung F., M Aljeboree A., Abd Alrazzak N., F Alkaim A. CuMoO4/ZnO nanocomposites: novel synthesis, characterization, and photocatalytic performance, *Journal of Nanostructures*, 2021, **11**:73 [Crossref], [Google Scholar], [Publisher]

[27]. Ma Y., Gao N., Li C. Degradation and pathway of tetracycline hydrochloride in aqueous solution by potassium ferrate, *Environmental Engineering Science*, 2012, **29**:357 [Crossref], [Google Scholar], [Publisher]

[28]. Hamoudi S.A., Hamdi B., Brendlé J. Tetracycline removal from water by adsorption on geomaterial, activated carbon and clay adsorbents, *Ecological Chemistry and Engineering S*, 2021, **28**:303 [Crossref], [Google Scholar], [Publisher]

[29]. Baiee N.A.H., Alkaim A.F. Photocatalytic Degradation of Methylene Blue Dye from Aqueous Solutions in the Presence of Synthesized ZnO Nanoparticles, *NeuroQuantology*, 2021, **19**:53 [Crossref], [Google Scholar], [Publisher]

[30]. García-Uriostegui L., Meléndez-Ortíz H.I., Camacho-Villegas T., Lugo-Fabres P., Toriz G. Synthesis and characterization of mesoporous silica-g-poly (hydroxyethylmethacrylate) nanohybrid particles as a drug delivery system, *Materials Chemistry and Physics*, 2022, **283**:126048 [Crossref], [Google Scholar], [Publisher]

[31]. Aljeboree A.M., Hameed N.M., Saleem H.D., Jasem H., Abood E.S., Abdulrazaq A.G., Aldulaim A.K., Alkaim A.F. Removal of Pollutants from Aqueous Solutions by using Natural Surfaces (Cotton) as a Model for Reusability and Highly Adsorbent Surface, *International Journal of Drug Delivery Technology*, 2022, **12**:583 [Google Scholar], [Publisher]

[32]. Gulen B., Demircivi P. Synthesis and characterization of montmorillonite/ciprofloxacin/TiO2 porous structure for controlled drug release of ciprofloxacin tablet with oral administration, *Applied Clay Science*, 2020, **197**:105768 [Crossref], [Google Scholar], [Publisher]

[33]. Zhang Y., Fu F., Li Y., Zhang D., Chen Y. One-step synthesis of Ag@ TiO2 nanoparticles for enhanced photocatalytic performance, *Nanomaterials*, 2018, **8**:1032 [Crossref], [Google Scholar], [Publisher]

[34]. Zhang J., Cha J.K., Fu G., Cho E.J., Kim H.S., Kim S.H. Aerosol processing of Ag/TiO2 composite nanoparticles for enhanced photocatalytic water treatment under UV and visible light irradiation, *Ceramics International*, 2022, **48**:9434 [Crossref], [Google Scholar], [Publisher]

[35]. Makuła P., Pacia M., Macyk W. How to correctly determine the band gap energy of modified semiconductor photocatalysts based on UV–Vis spectra, ACS Publications, 2018, **9**:6814 [Crossref], [Google Scholar], [Publisher]

[36]. Gao T., Li Q., Wang T. Sonochemical synthesis, optical properties, and electrical properties of core/shell-type ZnO nanorod/CdS nanoparticle composites, *Chemistry of Materials*, 2005, **17**:887 [Crossref], [Google Scholar], [Publisher]

[37]. Sambaza S.S., Maity A., Pillay K. Polyaniline-coated TiO2 nanorods for photocatalytic degradation of bisphenol A in water, *ACS omega*, 2020, **5**:29642 [Crossref], [Google Scholar], [Publisher]

[38]. Sivaranjani K., Sivakumar S., Dharmaraja J. Enhancement photocatalytic activity of Mn doped CdS/ZnO nanocomposites for the degradation of methylene blue under solar light irradiation, *Advances in Materials Science*, 2022, **22**:28 [Crossref], [Google Scholar], [Publisher]
[39]. Mohamed A.J., Ibtesam A., Firas H.A. Compare studies for the photocatalytic degradation of thymol blue with synthesized and commercial MWCNTs, *AIP Conference Proceedings*, 2020 [Crossref], [Google Scholar], [Publisher]

How to cite this manuscript: Haider Falih Shamikh Al-Saedi, Ola Hamad Salah, Russul Reidh Abass, Manal Morad Karim, Salima B. Alsaadi, Salam Ahjel, Zainab Jamal Hamoodah, Rathab Ali Ahmedh. Tetracycline Drug Degradation Using Binary Hybrid Advanced Oxidation Processes of Photocatalytic (UV/ TiO_2 and UV/ZnO) in Aqueous Solutions. *Asian Journal of Green Chemistry*, 8(3) 2024, 336-348. DOI: 10.48309/AJGC.2024.449148.1488