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Analysis of Fe-doped ZnO Thin Films for Degradation of Rhodamine B, Methylene Blue, and Escherichia Coli Under Visible Light

Keywords: thin film, Fe-doped ZnO, photodegradation, rhodamine b, methylene blue, Escherichia coli

Abstract: ZnO is a popular photocatalyst that is often used for the degradation of dyes and bacteria. However, the catalytic performance of ZnO is only optimal under UV light exposure. This study aims to determine the degradation performance of rhodamine b, methylene blue, and Escherichia coli using 0, 5, 10, 15, and 20% Fe-doped ZnO. Deposition of thin film was carried out using the sol-gel method with a spray-coating technique, while the degradation was carried out under halogen light exposure for 3 hours. The optical characterization results show that 20% Fe-doped ZnO has the highest transmittance and the lowest energy band gap of 3.21 eV based on Tauc's plot method. All thin films are hydrophilic with the largest contact angle of 68.54º by 20% Fe-doped ZnO and the lowest contact angle of 52.96º by 5% Fe-doped ZnO. The surface morphology of the thin film resembles a creeping root that is cracked and agglomerated. XRD test results show that the thin film is dominated by ZnO peaks with a wurtzite structure with a hexagonal plane phase and a crystal size of 115.5 Aº. The 20% Fe-doped ZnO thin film had the most efficient degradation performance of 70.79% for rhodamine b, 65.31% for blue, and 67% for E-coli bacteria. Therefore, Fe-doped ZnO is a brilliant photocatalyst material that can degrade various pollutants even under visible light.

1. Introduction

Water pollution by dyes is still a big problem that has not been resolved until now [1-5]. Most of the pollution is caused by the industrial waste of clothing, fabrics, and jeans. Dyestuff waste directly discharged into the environment without being processed first becomes a source of pollution and can cause various hazards, such as toxic effects and reduced light penetration ability in waters [6,7]. Among the various textile dyes, the most widely used are methylene blue and rhodamine b [8,9]. In addition to dyes, water pollution by Escherichia coli bacteria is also a problem [10,11]. Recently, there have been "viral" cases of health problems caused by Escherichia coli contamination such as fever, typhoid, and diarrhoea experienced by residents in several districts in Indonesia, such as Banjarnegara, Gunung Kidul, and Klaten. Based on the investigation results, it was found that the water used by the community in the area contained Escherichia coli bacteria that exceeded the safe threshold as water that humans can consume.

The presence of dye compounds and Escherichia coli bacteria is a serious problem that needs proper handling. Photodegradation is one of the methods to decompose industrial waste dyes. Degradation by utilizing the aid of light offers a relatively inexpensive solution and is easy to implement in Indonesia. In operation, the principle used in this method is to activate photocatalyst materials from semiconductor materials, such as CdS, Fe_2O_3, ZnO, TiO_2, Bi_2O_3, Cu_2O, WO_3 etc. [12-16]. Semiconductor materials that are easily found and most often used are TiO_2 and ZnO [17-19]. ZnO is an excellent oxidizing agent used as a photocatalyst and has a higher efficiency due to the strong absorption of UV from the solar spectrum. Compared to other catalysts, ZnO is suitable for the detoxification process of colour waste in water because it produces H_2O_2 more efficiently [18, 20].

The photocatalytic process occurs when ZnO material in water is exposed to UV light, electrons in the valence band will be excited to the conduction band. This process produces electrons (e-) in the conduction band and holes (h^+) in the valence band. Then electrons react with oxygen molecules from water to form superoxide anion radicals (O_2*), and holes will react with hydroxyl ions from water to form hydroxyl radical compounds (OH*). Superoxide reacts with electrons, and H^+ ions from water form H_2O_2 compounds then react again with electrons to produce hydroxyl radicals [21]. Electrons and holes generate hydroxyl radical compounds then oxidize pollutant molecules (M) to produce degraded compounds (M’). If M is a dye such as a rhodamine b and methylene blue, M’ is dyes with a more straightforward carbon chain and lower concentration. If M is Escherichia coli, M’ is Escherichia coli with less coloni.

In fact, in the actual environment, UV levels in sunlight are microscopic. Therefore, the optical properties of ZnO need to be improved to produce electrons with visible light energy. To improve the properties of ZnO, doping with metal ions is the most effective way to produce structural changes. The optical band gap energy and ferromagnetic properties can be controlled by doping the micro conductor with a transition metal such as Fe. Fe-doped ZnO nanoparticles play an essential role in the photodegradation of organic pollutants [22] and exhibit better photocatalytic activity than undoped ZnO nanoparticles [23]. Several researchers have prepared Fe-doped ZnO nanoparticles, but high doping amounts, control of optical properties to enhance photocatalytic activity, and degradation of multianalyte are rare. In our previous study, the degradation of pollutants by Fe-doped ZnO was still using a UV excitation source [7]. In this study, the degradation was carried out under halogen rays as a representation of actual sunlight. The synthesis results were then characterized and tested for photodegradation of methylene blue, rhodamine b, and Escherichia coli.

2. Materials and Methods

2.1 Materials

The materials used in this study were methylene blue ($C_{16}H_{18}ClN_3S$), rhodamine b ($C_{28}H_{31}N_2O_3Cl$), glass preparation, Iron Nitrate ($Fe(NO_3)_3.9H_2O$, Merck KGaA 99.95%), Zinc Acetate Dehydrate ($Zn(CH_3COO)_2.2H_2O$, Merck KGaA 97%), Acetone (Merck KGaA 99.5%), Isopropanol (Merck KGaA 99.5%), Methanol (Merck KGaA 99.9%), Aquades, Monoethanolamine (Merck KGaA), while the tools used are Mini Compressor (Krisbow, AS 186), VMC digital balance (VB 304), Degradation lamp (20 W), Hotplate Stirrer (Yellow MAG HS 7), and spray (Krisbow HS, 1200333).

2.2 Method

The Fe-doped ZnO sol-gel preparation was prepared by preparing 0.5 M zinc solution from 3.046 g Zn(CH3COO)2.2H2O and 26 ml isopropanol stirred with a magnetic stirrer temperature of 60 °C for 15 minutes until homogeneous [24]. The solution was stirred with a magnetic stirrer at 60 °C for 15 minutes until homogeneous. Monoethanolamine (MEA) was dropped into the solution and stirred on a hotplate at 60 °C for 15 minutes until the solution was colourless or transparent. Fe Nitrate with various concentrations of 0, 5, 10, 15, and 20% was mixed and then stirred for 15 minutes at a temperature of 60 °C. Thin film deposition was carried out using a spray-coating technique, and previously the glass substrate was cleaned using the Radio Corporation of America (RCA) method. The resulting solution is sprayed for 30 minutes at a temperature of 450 °C. The degradation test was carried out by making a solution of 10 ppm dye (rhodamine b and methylene blue). Photodegradation was carried out under visible light for 180 minutes with observations every 30 minutes. The degradation test of Escherichia coli bacteria was carried out with actual samples from the Rasamala river under visible light for 180 minutes.

2.3 Characterization

The morphology and chemical composition of the compounds in the thin film was observed using the Analytical Scanning Electron Microscopy – Energy Dispersive X-ray instrument (SEM – EDX JEOL JSM-6510LA). Analysis of the thin film crystallinity using X-Ray Diffraction instrument (Shimadzu Maxima XRD-700). Determination of the contact angle of the tips film using a digital microscope endoscope camera instrument. Optical characteristics and efficiency photodegradation of dyes and Escherichia coli were analyzed based on the results of the UV-Vis Spectrophotometer (Shimadzu UV-Vis 1240 SA).

3. Results and Discussion

The optical properties of the Fe-doped ZnO thin film obtained from the UV-Vis test are shown by transmittance and absorbance graphs. Tests of the absorbance spectrum of Fe-doped ZnO thin film in the wavelength range of 300-800 nm are shown in Figure 1. The highest transmittance is by 20% Fe-doped ZnO thin film at the same wavelength, while the highest absorbance is undoped ZnO. Substitution of Zn atoms by Fe atoms will form close distances between particles with less light [25]. It indicates that the concentration of Fe is very influential with the transmittance value because it can produce a different number of electron-hole pairs for each sample. The less Fe, the fewer electrons are produced so that the photon energy absorbed by the electrons to be also excited less and the transmittance value is increased.

The energy band gap value is processed using Tauc's plot method. In this study, the shift that occurs is redshift, as shown from the results of 0-20% Fe-doped ZnO, namely 3.32, 3.28, 3.25, 3.21, and 3.21 eV. This result is consistent with the study reported by Ariyakkani et al. (2017) for Fe-doped ZnO thin films, where the Eg value decreased from 3.38 eV for undoped ZnO to 3.07 eV for 20% Fe-doped ZnO [26]. A decrease in the energy bandgap indicates the shift that occurs as the concentration of Fe increases. Thus, the decrease in the energy bandgap occurs because the valence electrons of Fe larger than Zn will cause Fe to become a donor atom near the conduction band so that the electron transition requires less energy to the conduction band [27,28]. The decrease in the energy band gap value can also be explained by the presence of Fe atoms in ZnO can suppress crystal growth. In addition, the inclusion of Fe in ZnO also causes defects in ZnO crystals that cause high light absorption, which is very necessary to increase the photoactivity of visible light from ZnO [29].

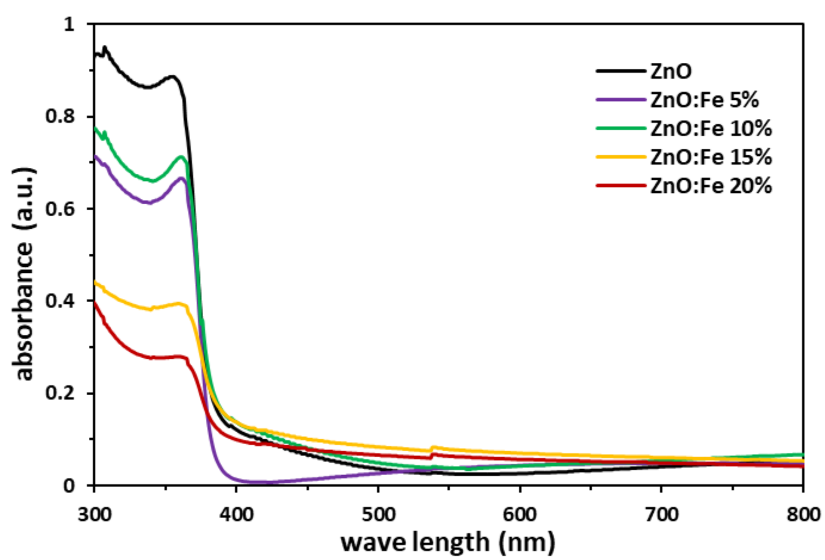


Figure 1. The absorbance spectrum of the Fe-doped ZnO thin film

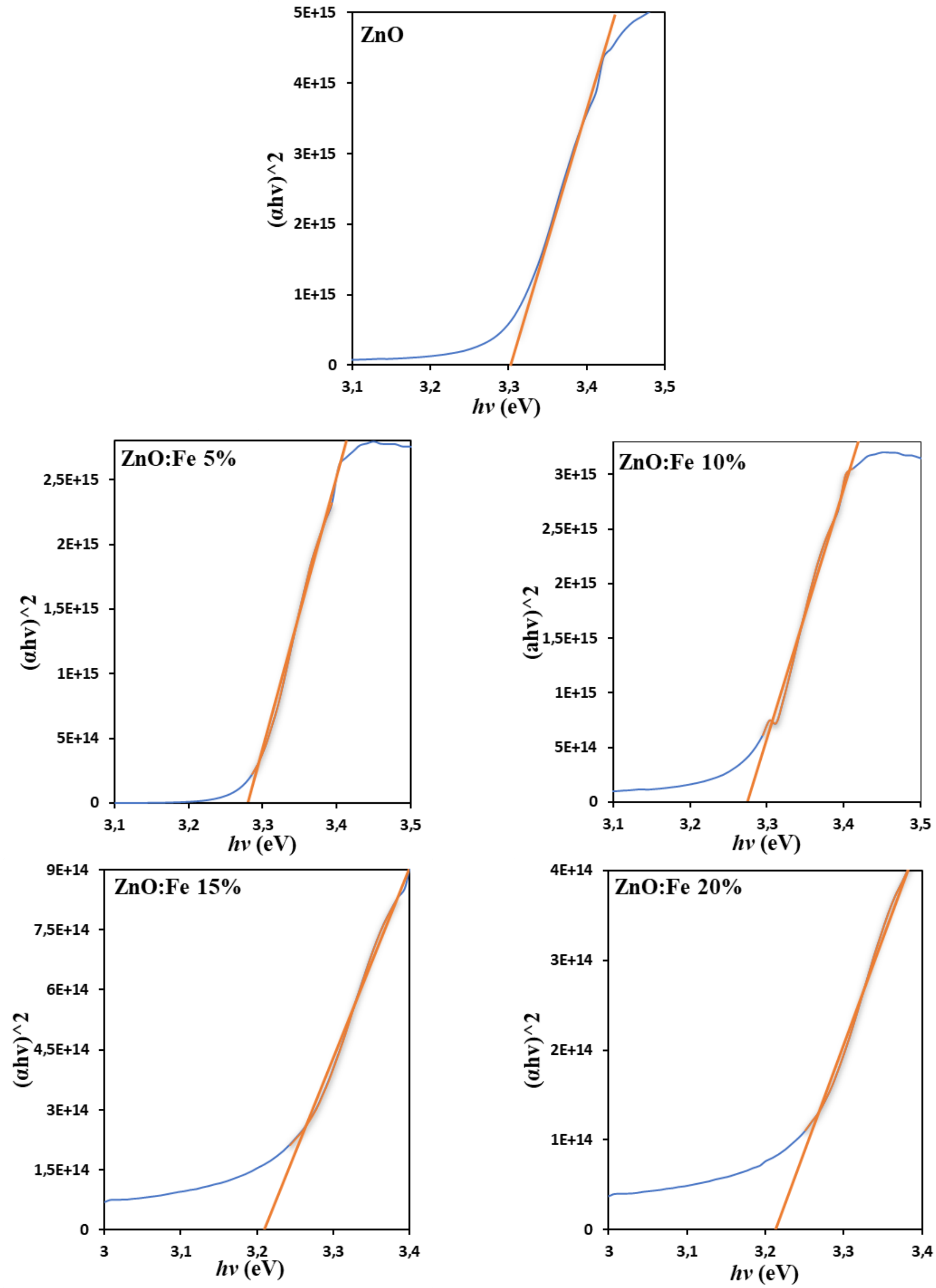


Figure 2. The energy band gap of Fe-doped ZnO thin film using Tauc's plot method

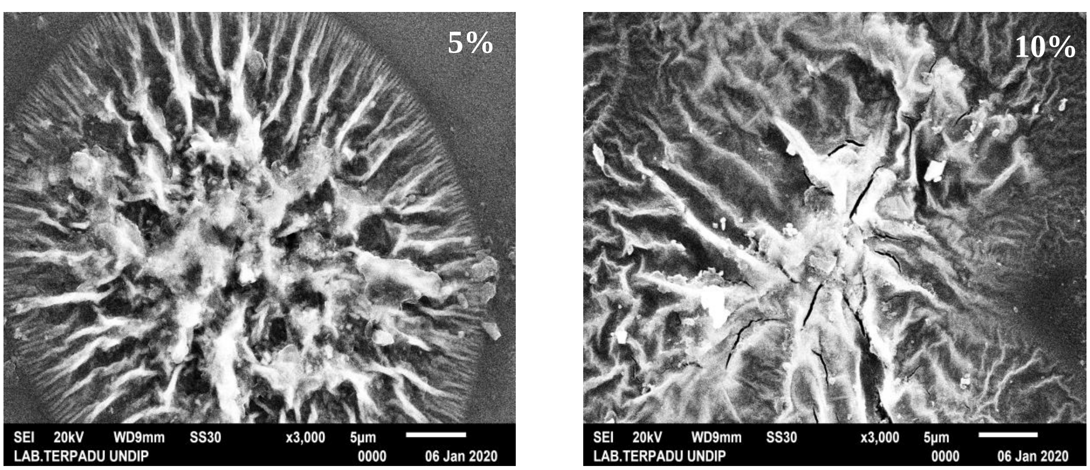
The hydrophilicity test was carried out between the dye solution and the surface of the film that had been coated on a glass plate using a Contact Anglemeter. Table 1 shows all the Fe-doped ZnO films forming a contact angle of less than 90° which means the films can interact well with water. Despite being dropped with two different dyes (rhodamine b and methylene blue), the resulting contact angle increased uniformly as the Fe concentration in ZnO increased, from 62.53° to 66.89° and 63.27° to 68.54°. This result is in contrast to ZnO doped with other materials, such as La, Na, Pb, and Mg, where the addition of a doping concentration of less than 7.5% reduces the contact angle value [30,31], including our previous study where the addition of Fe also lowers the value of the contact angle [7].

Interestingly, these results indicate that the addition of Fe doping less than 10% will increase the hydrophilic properties, while the addition of Fe doping more than 10% will increase the hydrophobic properties. This change in properties is probably due to differences in the microstructure and morphology of each doped film. Changes in properties that lead to an increase in hydrophobic properties will be advantageous for degradation applications because it increases the surface area and causes an increase in analyte adsorption on the catalyst surface [32]. The higher the contact angle formed by the thin film, the higher the degradation efficiency produced under halogen light.

SEM characterization is needed to determine the material's physical properties, including its microstructure and surface morphology. Figure 3 shows the results of Fe-doped ZnO thin film characterization using SEM with 3000x magnification. Fe-doped ZnO thin film appears to have a root-like morphology grouped and evenly distributed over the entire substrate surface. The root-like surface morphology is caused by the bonds between the particles deposited in the presence of high temperatures on the substrate. These bonds result in the grains becoming fused and forming a structure that looks like a root [33]. The diameter of the size formed ranged from 150 nm to 500 nm with an average particle size of 333.75 nm. A high concentration of Fe dopant resulted in the appearance of agglomeration and aggregation in ZnO morphology [34], also causing irregular cracked surfaces.

Table 1. Fe-doped ZnO thin film contact angle against rhodamine b and methylene blue

thin film	Contact angle (°)	
	rhodamin b	methylene blue
Undoped ZnO	62.53	63.27
5% Fe-doped ZnO	52.96	57.62
10% Fe-doped ZnO	55.55	61.81
15% Fe-doped ZnO	65.75	62.83
20% Fe-doped ZnO	66.89	68.54



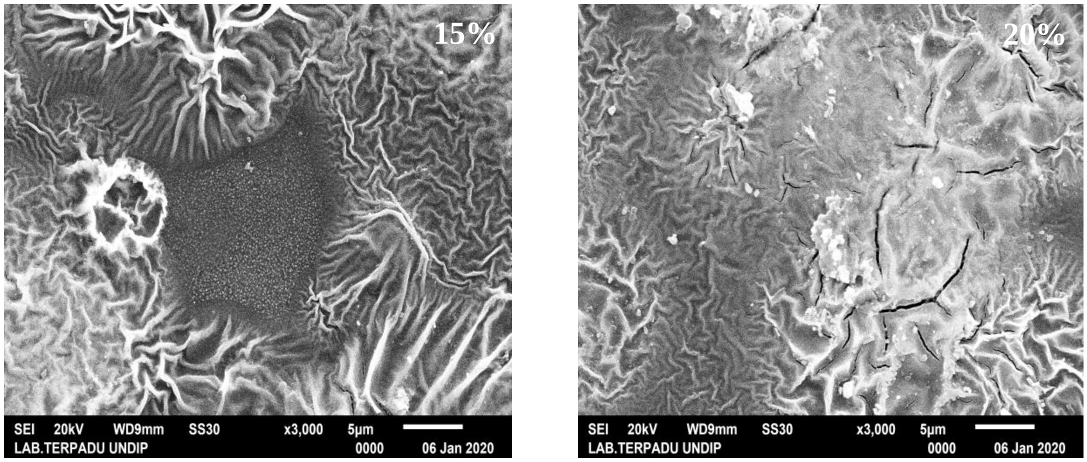


Figure 3. Surface morphology of Fe-doped ZnO thin film 3000x magnification

Characterization using EDX is needed to determine the chemical properties, including the material's elemental composition. Tables 2 show that O, Fe, and Ze atoms were successfully formed on the substrate. The increase in the percentage of mass and Fe atoms and the increase in the number of concentrations. It indicates an increase in the molecules that make up the thin film formed. It causes the resulting energy band gap to become smaller, and the ability of Fe-doped ZnO to photodegrade in the visible light spectrum is getting better. X-ray diffraction test results were carried out to determine the phase contained in the sample. From the results of the XRD test, analysis was carried out by matching the spectrum of the XRD test results with JCPDS data (Joint Committee on Powder Diffraction Standard) which helps know what compounds are contained in the Fe-doped ZnO. JCPDS data used are JCPDS 36–1451 (ZnO), JCPDS 06-0696 (Fe), and JCPDS 24-0072 (Fe_2O_3). Figure 4 shows the XRD test results for 20% Fe-doped ZnO showing several diffraction peaks with the most dominant peak belonging to ZnO with 2θ used between 20-90° and the wavelength used =1.5405Å

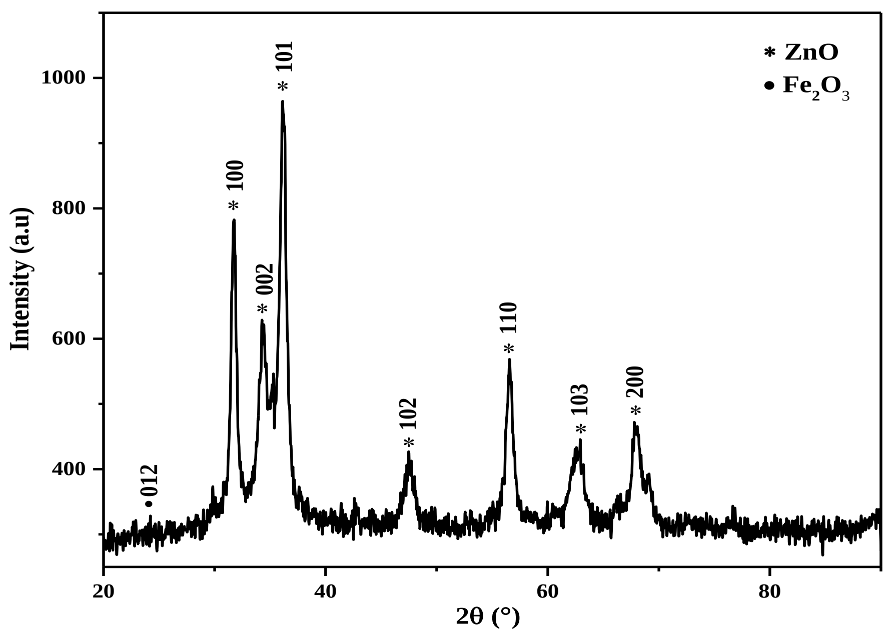


Figure 4. XRD results of 20% Fe-doped ZnO thin film

Table 2. The mass composition of the chemical elements Fe-doped ZnO thin film

Element	Mass of Fe-doped ZnO (%)			
	5%	10%	15%	20%
O	49.03	54.96	58.38	53.31
Fe	2.05	3.41	3.58	5.47
Zn	48.92	41.63	38.04	41.21

The XRD test results showed that the ZnO sample had a wurtzite structure with hexagonal plane phases showed that the diffraction peaks were at 2θ=31.76°, 34.26°, 36.10°, 47.48°, 56.54°, 62.90°, and 67.82° with Miller indices (100), (002), (101), (102), (110), (103), and (200) [35-37]. This result is similar with the study reported by Roguai and Djelloul (2021), where all the peaks of the XRD spectrum (100), (002), (101), (102), (110), (103), and (200) for Fe-doped ZnO (0-10%) are identified as single-phase ZnO wurtzite structure with the space group $P6_3mc$ [38]. During synthesis, the doping addition of ferrite nitrate led to a peak at 24.22°, corresponding to (012) Fe_2O_3 [39]. In the hexagonal structure, lattice constants (a and c), unit cell volume (V), crystal size (D), lattice strain (ε), and atomic packing fraction (APF) are calculated using the following formula [40]:

$$a = \frac{\lambda}{\sqrt{3}\sin\theta_{(100)}} \quad (1)$$

$$c = \frac{\lambda}{\sqrt{3}\sin\theta_{(002)}} \quad (2)$$

$$V = \frac{\sqrt{3}}{2} a^2 c \quad (3)$$

$$APF = \frac{2\pi}{3\sqrt{3}} \frac{a}{c} \quad (4)$$

The results of calculations using the formulas (1) to (5) are shown in Table 3 below:

Table 3. Crystal Parameters of Fe-doped ZnO

Parameter	Value	Unit
a	1.509586	Å
c	1.57997	Å
V	3.118137	$Å^3$
APF	1.154747	%

X-ray diffraction is also used to determine the Crystal Size (D) by using the Debye Sherrer equation approach as follows:

$$D = \frac{K\lambda}{\beta\cos\theta} \quad (5)$$

Where is the wavelength of X-rays (1.54056) and is the value of Full Width at Half Maximum (FWHM), and K is the crystal form factor whose values are between 0.9. The debye Scherrer equation is then modified into equation (6) as follows:

$$\ln\beta = \ln\frac{K\lambda}{D} + \ln\frac{1}{\cos\theta} \quad (6)$$

The calculation of the values (ln 1/cos) and (ln) XRD analysis results in all crystal plane orientations are shown in the following table:

Table 4. Calculation of (ln 1/cos θ) and (ln β)

2θ	Crystal Plane Orientation	$1/\cos\theta$	$\beta\ (rad)$	$\ln\ 1/\cos\theta$	$\ln\beta$
31.76	100	1.1758	0.01193	0.16195	-4.42894425
34.26	002	1.21092	0.01564	0.19138	-4.1582305
36.10	101	1.23844	0.01427	0.21385	-4.24943083
47.48	102	1.4805	0.01908	0.39238	-3.95921498
56.54	110	1.8134	0.01358	0.59521	-4.29944439
62.90	103	2.17099	0.02362	0.77518	-3.74586958
67.82	200	2.66511	0.017	0.98025	-4.07475961

The value of intercept from the graph of the relationship ln (1/cos) as the x-axis and ln as the y axis is equal to ((K)/D), so equation (6) can be modified into the following equation:

$$D = \frac{K\,\lambda}{\exp(y_0)} \qquad (7)$$

Where y_0 is the intercept value of the graph ln (1/cos) versus ln, the intercept value from the graph above is -4.3171, so the crystal size is 115.5 A^o. This result may be the smallest crystal size among other samples not tested, considering that several previous studies increasing the Fe content reduced the lattice parameters and the average crystal size [34,41,42].

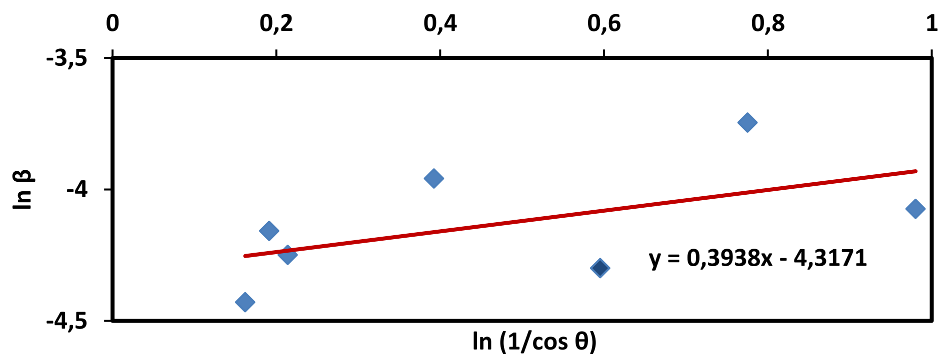


Figure 5. Relationship chart of ln (1/cos θ) versus ln β

Figure 6 and Table 5 show the results of the photodegradation carried out to test the photocatalyst activity of Fe-doped ZnO. Photocatalyst activity was tested to degrade rhodamine b, methylene blue, and Escherichia coli under halogen to represent visible light. Photocatalyst reactions that occur in the reaction of Fe-doped ZnO material with visible light can excite electrons and leave holes [43,44]. The electrons interact with the oxygen present in the water to produce superoxide. Meanwhile, hydroxyl radicals are generated from the interaction between holes and water. Hydroxyl radicals are potent oxidizing agents, so they are capable of oxidizing organic compounds. The radicals formed will break down the dye molecules to produce H_2O and CO_2 [45].

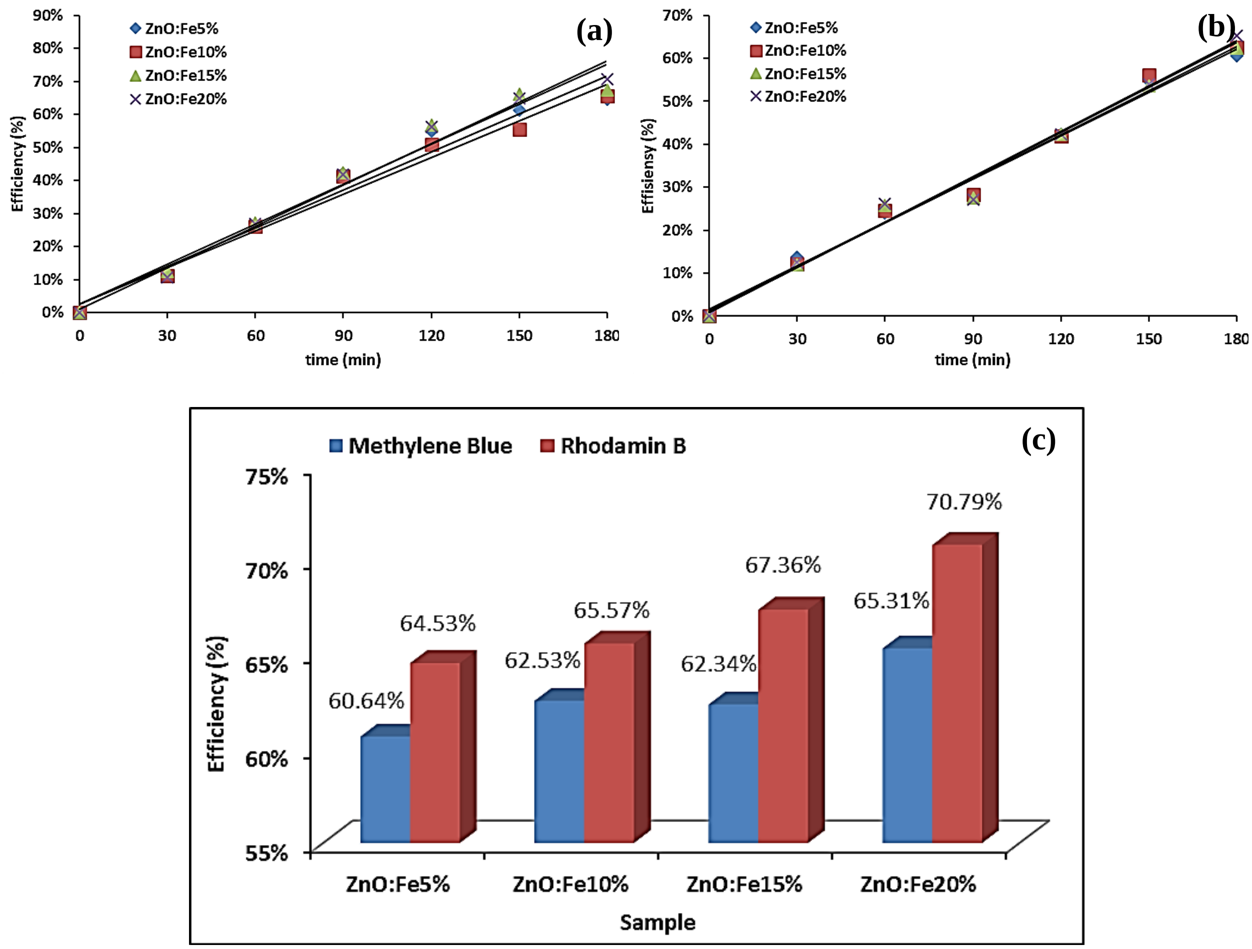


Figure 6. (a) Photodegradation of rhodamine b (b) Photodegradation of methylene blue (c) Photodegradation efficiency diagram of Fe-doped ZnO under visible light

Table 5. Bacterial degradation test results

Thin film	Degradation Efficiency (%)
5% Fe-doped ZnO	25
10% Fe-doped ZnO	42
15% Fe-doped ZnO	50
20% Fe-doped ZnO	67

Photodegradation of the thin film under visible light for rhodamine b and methylene blue degradation based on 5-20% Fe-doped ZnO, are 64.53; 65.57; 67.36; 70.79% and 60.64; 62.53; 62.34; 65.31%. The photodegradation of Escherichia coli was 25, 42, 50, and 67%. The most optimal degradation ability is possessed by a 20% Fe-doped ZnO, both on rhodamine b, methylene blue, and Escherichia coli. The energy band gap value influences the effectiveness of the photocatalyst thin film performance. The small energy band gap indicates that the energy required to excite electrons from the valence band to the conduction band is getting smaller. With the same amount of energy, a thin film with a small energy band gap can produce more electron-hole pairs than a thin film with a large energy bandgap. Therefore, the smaller the energy band gap, the higher the degradation ability under visible light. On the other hand, the larger the energy band gap, the lower the degradation ability under visible light.

The presence of Fe found in EDX measurements is believed to form a trapping state between the conduction band and the valence band, which can inhibit the recombination rate of electron and hole pairs and increase photocatalytic activity. This result was confirmed by Bousslama et al. (2017), who investigated Fe-doped ZnO for the degradation of rhodamine b [28], Saleh and Djaja (2014) for the degradation of methyl orange [46], and Roguai & Djelloul (2021) for the degradation of methylene blue [38]. Fe^{2+} and Fe^{3+} ions obtained are known to form 2 new energy levels in the energy gap. One energy level is above the valence band, which refers to the 3d orbital of Fe^{3+}, and the other is below the conduction band because the electron energy level of the Fe^{2+} ion is lower than the 3d Zn in the conduction band [25,46].

When halogen light is given, electrons can be excited from Fe^{3+} and the valence band to the conduction band. After the electrons are excited, Fe^{3+} will be converted to Fe^{4+}, which can interact with hydroxyl ions and produce OH hydroxyl radicals. Simultaneously, electrons excited to the valence band can react with oxygen and produce superoxide radicals (O_2*). The formed active radical species will initiate a series of redox processes to break down dye compounds into simpler ones and cause Escherichia coli cell membranes to lyse. Therefore, developing a thin film with appropriate optical properties such as 20% Fe-doped ZnO would be advantageous for the degradation of liquid pollutants since electron-hole pairs can be generated only by a visible light excitation source.

Conclusions

The increase in Fe dopant impacts improving optical properties, decreasing the energy bandgap and increasing the photocatalytic properties of ZnO thin films. A Fe-doped ZnO thin film has a root-like surface morphology dominated by ZnO peaks with a wurtzite structure with a hexagonal plane. Fe dopant determines the hydrophilicity of film, and the magnitude of the contact angle is directly proportional to the degradation efficiency. 20% Fe-doped ZnO is the best photocatalyst thin film formula for degrading dyes and Escherichia coli under visible light.

References

[1] Lops, C., Ancona, A., Di Cesare, K., Dumontel, B., Garino, N., Canavese, G., ... & Cauda, V. (2019). Sonophotocatalytic degradation mechanisms of Rhodamine B dye via radicals generation by micro-and nano-particles of ZnO. *Applied Catalysis B: Environmental*, *243*, 629-640.

[2] Nguyen, C. H., Fu, C. C., & Juang, R. S. (2018). Degradation of methylene blue and methyl orange by palladium-doped TiO2 photocatalysis for water reuse: Efficiency and degradation pathways. *Journal of Cleaner Production*, *202*, 413-427

[3] Pang, Y., Ruan, Y., Feng, Y., Diao, Z., Shih, K., Chen, D., & Kong, L. (2019). Ultrasound assisted zero valent iron corrosion for peroxymonosulfate activation for Rhodamine-B degradation. *Chemosphere*, *228*, 412-417.

[4] Pratiwi, N. I., Sari, S. R., Arifan, F., Wulandari, A. T., Alkian, I., Mustasjar, B., & Aji, M. B. F. B. (2020, March). Batik Pemalang Organic Wastewater Composition and Simple Electrocoagulation-Filtration Treatment. In *IOP Conference Series: Earth and Environmental Science* (Vol. 448, No. 1, p. 012037). IOP Publishing.

[5] Larasati, D. A., Alkian, I., Arifan, F., & Sari, S. R. (2020, March). Batik Home Industry Wastewater Treatment Using UVC/Ozon Oxidation Method: Case Study in Cibelok Village, Pemalang, Indonesia. In *IOP Conference Series: Earth and Environmental Science* (Vol. 448, No. 1, p. 012055). IOP Publishing.

[6] Arshad, H. M. U., Anwar, H., Javed, Y., Naz, M. Y., Ghaffar, A., Khan, M. Z., & Farooq, Z. (2019). Investigation of photocatalytic degradation of methylene orange dye using titanium dioxide–zinc oxide nanocomposites. *Materials Research Express*, *6*(12), 125009.

[7] Sutanto, H., Alkian, I., Pramunditya, V. J., Mukholit, M., Hidayanto, E., Duri, I. R., & Priyono. (2020). Effect Of Fe Addition On Zno Thin Films For Photodegradation Under UV And Halogens Light. *International Journal Of Scientific & Technology Research.* 9 (8), 315-318.

[8] Suharyadi, E., Muzakki, A., Nofrianti, A., Istiqomah, N. I., Kato, T., & Iwata, S. (2020). Photocatalytic activity of magnetic core-shell CoFe2O4@ ZnO nanoparticles for purification of methylene blue. *Materials Research Express*, *7*(8), 085013.

[9] Sun, Y., Gao, Y., Zhao, B., Xu, S., Luo, C., & Zhao, Q. (2020). One-step hydrothermal preparation and characterization of ZnO–TiO2 nanocomposites for photocatalytic activity. *Materials Research Express*, *7*(8), 085010.

[10] Wang, W., Zeng, Z., Zeng, G., Zhang, C., Xiao, R., Zhou, C., ... & Zhou, Y. (2019). Sulfur doped carbon quantum dots loaded hollow tubular g-C3N4 as novel photocatalyst for destruction of Escherichia coli and tetracycline degradation under visible light. *Chemical Engineering Journal*, *378*, 122132.

[11] Syahrul, F., Wahyuni, C. U., Notobroto, H. B., Wasito, E. B., Adi, A. C., & Dwirahmadi, F. (2020). Transmission media of foodborne diseases as an index prediction of diarrheagenic Escherichia coli: Study at elementary school, Surabaya, Indonesia. *International Journal of Environmental Research and Public Health*, *17*(21), 8227

[12] Cheng, L., Xiang, Q., Liao, Y., & Zhang, H. (2018). CdS-based photocatalysts. *Energy & Environmental Science*, *11*(6), 1362-1391

[13] Hitam, C. N. C., & Jalil, A. A. (2020). A review on exploration of Fe2O3 photocatalyst towards degradation of dyes and organic contaminants. *Journal of environmental management*, *258*, 110050.

[14] Poorsajadi, F., Sayadi, M. H., Hajiani, M., & Rezaei, M. R. (2020). Synthesis of CuO/Bi2O3 nanocomposite for efficient and recycling photodegradation of methylene blue dye. *International Journal of Environmental Analytical Chemistry*, 1-14.

[15] Bouallouche, R., Dalhatou, S., Kane, A., Nasrallah, N., Hachemi, M., Amrane, A., & Assadi, A. A. (2021). Reconsideration of the contribution of photogenerated ROS in methyl orange degradation on TiO2, Cu2O, WO3, and Bi2O3 under low-intensity simulated solar light: mechanistic understanding of photocatalytic activity. *Euro-Mediterranean Journal for Environmental Integration*, *6*(3), 1-10.

[16] Borade, P. A., Suroshe, J. S., Bogale, K., Garje, S. S., & Jejurikar, S. M. (2020). Photocatalytic performance of ZnO carbon composites for the degradation of methyl orange dye. *Materials Research Express*, *7*(1), 015512.

[17] Sakthivel S, Neppolian B, Shankar M, Arabindoo B, Palanichamy M, Murugesan V. Solar photocatalytic degradation of azo dye: comparison of photocatalytic efficiency of ZnO and TiO2. Solar energy materials and solar cells. 2003;77:65-82.

[18] Bica, B. O., & de Melo, J. V. S. (2020). Concrete blocks nano-modified with zinc oxide (ZnO) for photocatalytic paving: Performance comparison with titanium dioxide (TiO2). *Construction and Building Materials*, *252*, 119120.

[19] Hidayanto E, Sutanto H, Wibowo S, Irwanto M. Morphology and Degradation Kinetics of N-Doped TiO2 Nano Particle Synthesized Using Sonochemical Method. Solid State Phenomena: Trans Tech Publ; 2017. p. 95-100.

[20] Shah, A. A., Chandio, A. D., & Sheikh, A. A. (2021). Boron Doped ZnO Nanostructures for Photo Degradation of Methylene Blue, Methyl Orange and Rhodamine B. *Journal of Nanoscience and Nanotechnology*, *21*(4), 2483-2494.

[21] Sutanto, H., Wibowo, S., Nurhasanah, I., Hidayanto, E., & Hadiyanto, H. (2016). Ag doped ZnO thin films synthesized by spray coating technique for methylene blue photodegradation under UV irradiation. *International Journal of Chemical Engineering*, *2016*.

[22] Ba-Abbad, M. M., Kadhum, A. A. H., Mohamad, A. B., Takriff, M. S., & Sopian, K. (2013). Visible light photocatalytic activity of Fe3+-doped ZnO nanoparticle prepared via sol–gel technique. *Chemosphere*, *91*(11), 1604-1611.

[23] Hui, A., Ma, J., Liu, J., Bao, Y., & Zhang, J. (2017). Morphological evolution of Fe doped sea urchin-shaped ZnO nanoparticles with enhanced photocatalytic activity. *Journal of Alloys and Compounds*, *696*, 639-647.

[24] Sutanto, H., Hidayanto, E., Irwanto, M., & Wibowo, S. (2017). The influence of nitrogen doping concentration on the strain and band gap energy of N-doped zinc oxide prepared using spray

coating technique. In *Solid State Phenomena* (Vol. 266, pp. 141-147). Trans Tech Publications Ltd.

[25] Dong, S., Xu, K., Liu, J., & Cui, H. (2011). Photocatalytic performance of ZnO:Fe array films under sunlight irradiation. Physica B: Condensed Matter. 406 (19), 3609-3612

[26] Ariyakkani, P., Suganya, L., & Sundaresan, B. (2017). Investiagtion of the Structural, Optical and Magnetic Properties of Fe doped ZnO Thin Films Coated on Glass by Sol-Gel Spin COating Method. *Journal of Alloys and Compounds, 695*, 3467-3475.

[27] Cheng, W., & Ma, X. (2009, March). Structural, optical and magnetic properties of Fe-doped ZnO. In *Journal of Physics: Conference Series* (Vol. 152, No. 1, p. 012039). IOP Publishing.

[28] Bousslama, W., Elhouichet, H., & Férid, M. (2017). Enhanced photocatalytic activity of Fe doped ZnO nanocrystals under sunlight irradiation. *Optik*, *134*, 88-98.

[29] Sze-Mun, L., Jin-Chung, S., Honghu, Z., Hua, L., Haixiang, L., Yen-Yi, C., Man-Kit, C., Abdul, R. M., Green synthesis of Fe-ZnO nanoparticles with improved sunlight photocatalytic performance for polyethylene film deterioration and bacterial inactivation, Materials Science in Semiconductor Processing, Volume 123, 1 March 2021, 105574.

[30] Shaban, M., & El Sayed, A. (2016). Effects of Lanthanum and Sodium on the Structural, Optical and Hydrophilic Properties of Sol-Gel Derived ZnO Films: A Comparative Study. *Material Science in Semiconductor Processing, 41*, 323-334.

[31] Shaban, M., & El Sayed, A. M. (2015). Influences of Lead and Magnesium Co-doping on the Nanostructural, Optical Properties and Wettability of Spin Coated Zinc Oxide Films. *Materials Science in Semiconductor Processing, 39*, 136-147.

[32] Najafidoust, A., Asl, E. A., Hakki, H. K., Sarani, M., Bananifard, H., Sillanpaa, M., & Etamadi, M. (2021). Sequential Impregnation and Sol-Gel Synthesis of Fe-ZnO over Hydrophobic Silica Aerogel as a Floating Photocatalyst with Highly Enhanced Photodecomposition of BTX Compounds from Water. *Solar Energy, 225*, 334-356.

[33] Sutanto, H., Durri, S., Wibowo, S., Hadiyanto, H., & Hidayanto, E. (2016). Root-like morphology of Zno: Al thin film deposited on amorphous glass substrate by sol-gel method. *Physics Research International*, *2016*.

[34] Kumar, M. S., & Arunagiri, C. (2021). Efficient photocatalytic degradation of organic dyes using Fe-doped ZnO nanoparticles. *Journal of Materials Science: Materials in Electronics*, 1-11.

[35] Liu, Q., Dai, J., Hu, F., Zhang, Z., Xiong, K., Xu, G. (2021). Core-shell structured Fe/ZnO composite with superior electromagnetic wave absorption performance. *Ceramics International*, *47*(10), 14506-14514.

[36] Ciciliati, M.A., Silva, M.F., Fernandes, D.M., de Melo, M.A.C., Hechenleitner, A.A.W., Pineda, E.A.G. (2015). Fe-doped ZnO nanoparticles: Synthesis by a modified sol–gel method and characterization. *Material Letter*., 159, 84-86.

[37] Basyooni, M. A., Shaban, M., & El Sayed, A. M. (2017). Enhanced gas sensing properties of spin-coated Na-doped ZnO nanostructured films. *Scientific reports*, *7*(1), 1-12

[38] Roguai, D., & Djelloul, P. (2021). Structural, Microstructural and Photocatalytic Degradation of Methylene Blue of Zinc Oxide and Fe-doped ZnO Nanoparticles Prepared by Simple Coprecipitation Method. *Solid State Communications*, *334-335*.

[39] Mohammadigharehbagh, R., Pat, S., Akkurt, N., & Korkmaz, S. (2021). Studies on the Morphological, Structural, Optical and Electrical Properties of Fe-doped ZnO Magnetic Nano-crystal Thin Films. *Physica B: Physics of Condensed Matter*, *609*.

[40] Monshi, A., Mohammad, R. F., Mohammad, R. M., 2012. Modified Scherrer Eqquation to Estimate More Accurately Nano-Crystallite Size Using XRD. World Journal of Nano Science and Engineering, Vol. 2, pp. 154-160.

[41] Pakizeh, Esmaeil (2020). *Optical response and structural properties of Fe-doped Pb(Zr0.52Ti0.48)O3 nanopowders. Journal of Materials Science: Materials in Electronics, (),* –. doi:10.1007/s10854-020-03050-1

[42] Saadi, H.; Rhouma, F.I.H.; Benzarti, Z.; Bougrioua, Z.; Guermazi, S.; Khirouni, K. (2020). Electrical conductivity improvement of Fe doped ZnO nanopowders. Materials Research Bulletin, 129(), 110884–. doi:10.1016/j.materresbull.2020.110884
[43] Srivastava, A., Kumar, N., & Khare, S. (2014). Enhancement in UV emission and band gap by Fe doping in ZnO thin films. *Opto-Electronics Review*, *22*(1), 68-76.
[44] Khalid, N. R., Hammad, A., Tahir, M. B., Rafique, M., Iqbal, T., Nabi, G., & Hussain, M. K. (2019). Enhanced photocatalytic activity of Al and Fe co-doped ZnO nanorods for methylene blue degradation. *Ceramics International*, *45*(17), 21430-21435.
[45] Xu, L., & Li, X. (2010). Influence of Fe-doping on the structural and optical properties of ZnO thin films prepared by sol–gel method. *Journal of Crystal Growth*, *312*(6), 851-855.
[46] Saleh, Rosari; Djaja, Nadia Febiana (2014). UV light photocatalytic degradation of organic dyes with Fe-doped ZnO nanoparticles. Superlattices and Microstructures, 74(), 217–233. doi:10.1016/j.spmi.2014.06.013

RESPONSE TO REFEREE 1

We appreciate the time and effort the reviewers dedicated to providing feedback on our manuscript and are grateful for the insightful comments and valuable improvements to our paper. We have incorporated most of the suggestions made by the reviewers. Those changes are highlighted within the manuscript. A point-by-point response to the reviewers' comments and concerns is coloured blue colour, and the revised parts in the manuscript are highlighted with "*Italic*".

In this manuscript, the author demonstrated the Degradation of Rhodamine B, Methylene Blue, And Escherichia Coli by ZnO: Fe Thin Films under Visible Light. The results are interesting. Therefore, after minor revisions, the paper can be considered by the journal for publication. The detailed comments are listed as follows:

1. In the abstract the overall purposse of the study and the research problem investigated is missing.

   We've added a research problem and improved the abstract thoroughly. The revised abstracts are: *"ZnO is a popular photocatalyst that is often used for the degradation of dyes and bacteria. However, the catalytic performance of ZnO is only optimal under UV light exposure. This study aims to..……………………………………etc."*

   – **(Page 1, No 16)**

2. In 3rd line of the abstract the sentence is not properly written.

   We have corrected the sentences in the abstract. We have discussed and agreed to change some words in order to improve the abstract, the title of the paper, and the entire content of the manuscript.

   - *"ZnO:Fe"* → *"Fe-doped ZnO"*
   - *"tauch's plot"* → *"Tauc's plot"*
   - *"thin layer"* → *"thin film"*

   – (**Almost all pages)**

3. In the introduction section the authors have not explained how ZnO: Fe degrades E.coli (Mechanism). Also, the mechanism of photocatalysis of dyes has not been explained in this manuscript.

   We have improved the introduction and added an explanation of the photocatalytic mechanism against dyes and Escherichia coli. Those improvements are:

"The photocatalytic process occurs when ZnO material in water is exposed to UV light, electrons in the valence band will be excited to the conduction band. This process produces electrons (e-) in the conduction band and holes (h^+) in the valence band……………………………………………………………………………………………..etc".

–(Page 2, No 1)

4. In the materials and methods section the authors have not mentioned the amount of 0.5 M zinc acetate dehydrate and isopropanol solution that is to be mixed. In addition to that, the authors also have not cited any reference for the method used here.

   We have added the precise quantities of $Zn(CH_3COO)_2.2H_2O$ and isopropanol along with the references.

   "The Fe-doped ZnO sol-gel preparation was prepared by preparing 0.5 M zinc solution from 3.046 g $Zn(CH_3COO)_2.2H_2O$ and 26 ml isopropanol stirred with a magnetic stirrer temperature of 60 ^oC for 15 minutes until homogeneous [24]..etc"

   –(Page 2, No 33)

5. The authors have not mentioned the necessary details that are crucial to reproduce the test.

   In this section, we add an explanation:

   *"**2.3 Measurements.** The morphology and chemical composition of the compounds in the thin film was observed using the Analytical Scanning Electron Microscopy – Energy Dispersive X-ray instrument (SEM – EDX JEOL JSM-6510LA). Analysis of the thin film crystallinity using X-Ray Diffraction instrument (Shimadzu Maxima XRD-700)..etc"*

   – **(Page 2, No 46)**

6. In the results and discussion section the manuscript lacks the discussion part and hasn't included a Comparison with prior studies. Which is often considered the most important part of a research paper because it most effectively demonstrates the ability as a researcher to think critically about an issue, to develop creative solutions to problems based on the findings, and to formulate a deeper, more profound understanding of the research problem you are studying.

   We have added an explanation of the critical analysis of the results section and a discussion of the manuscript and comparison with several previous studies.

   "This result is consistent with the study reported by Ariyakkani et al. (2017) for Fe-doped ZnO thin films, where the Eg value decreased from 3.38 eV for undoped ZnO to 3.07 eV for 20% Fe-doped ZnO [26]………………………………………………………………………………..etc"

-(Page 3, No 15)

"This result is in contrast to ZnO doped with other materials, such as La, Na, Pb, and Mg, where the addition of a doping concentration of less than 7.5% reduces the contact angle value [30,31], including our previous study where the addition of Fe also lowers the value of the contact angle [7]. Interestingly, these results indicate that the addition of Fe doping less than 10% will increase the hydrophilic properties, while the addition of Fe doping more than 10% will increase the hydrophobic properties……………………………………………………………………………etc"

-(Page 5, No 3)

"This result is similar with the study reported by Roguai and Djelloul (2021), where all the peaks of the XRD spectrum (100), (002), (101), (102), (110), (103), and (200) for Fe-doped ZnO (0-10%) are identified as single-phase ZnO wurtzite structure with the space group $P6_3mc$ [38].etc"

-(Page 7, No 5)

"The presence of Fe found in EDX measurements is believed to form a trapping state between the conduction band and the valence band, which can inhibit the recombination rate of electron and hole pairs and increase photocatalytic activity. This result was confirmed by Bousslama et al. (2017), who investigated Fe-doped ZnO for the degradation of rhodamine b [28], Saleh and Djaja (2014) for the degradation of methyl orange [45], and Roguai & Djelloul (2021) for the degradation of methylene blue [38]………………………………………………………………………etc"

-(Page 9, No 18)

7. In the conclusion section the authors have only summarized the results that were obtained they should give a summary of what was learned from your research, this summary should be relatively brief since the emphasis in the conclusion is on the implications, evaluations, insights, etc. that you make.

We have improved the conclusions of this study:

"The increase in Fe dopant impacts improving optical properties, decreasing the energy bandgap and increasing the photocatalytic properties of ZnO thin films. A Fe-doped ZnO thin film has a root-like surface morphology dominated by ZnO peaks with a wurtzite structure with a hexagonal plane. Fe dopant determines the hydrophilicity of film, and the magnitude of the contact angle is directly proportional to the degradation efficiency. 20% Fe-doped ZnO is the best photocatalyst thin film formula for degrading dyes and Escherichia coli under visible light"

–**(Page 10, No 17)**

8. The Authors should cite more references it's not adequate.

   We have cited more references, hopefully adequate for this research paper.

RESPONSE TO REFEREE 2

We appreciate the time and effort the reviewers dedicated to providing feedback on our manuscript and are grateful for the insightful comments and valuable improvements to our paper. We have incorporated most of the suggestions made by the reviewers. Those changes are highlighted within the manuscript. A point-by-point response to the reviewers' comments and concerns is colored blue colour and the revised parts in the manuscript are highlighted with *"Italic"*.

This work describes the preparation, by sol-gel and spray deposition, of Fe-doped ZnO films as photo-catalysts to degrade some dyes. This work is interest for waste water treatment and water purification which is essential and vital. The authors gave good introduction and applied several techniques for this study. This work is acceptable for publication in Materials Research Express after some amendments:

1. The first sentence in the abstract: " The deposition of a high concentration ZnO:Fe thin layer was carried out using a sol-gel method with a spray-coating technique.", remove "high concentration". In the second sentence:"... ZnO:Fe 0, 5, 10, 15, and 20% ..... " rewrite as 0, 5, 10, 15, and 20% Fe doped ZnO (and also throughout the manuscript). In the fourth sentence (and throughout the manuscript) rewrite tauch's plot as Tauc's plot. Finally, ad a sentence to mention the application(s) of this study

   We have improved the abstract thoroughly.

   - Eliminate the word "high concentration".
   - Substitute "ZnO:Fe 0, 5, 10, 15, and 20%" → "0, 5, 10, 15, and 20% Fe-doped ZnO" for the abstract, title of the paper, and the entire content of the manuscript.
   - Substitute "tauch's plot" → "Tauc's plot".
   - Substitute "thin layer" → "thin film". **(Page 1, No 16)**

2. In "Materials and Methods" section: Please divide this section into 2.1. Materials (to describe all the used material and their molecular weight). 2.2. Method (To give the details of the sol-gel and spray deposition processes). 2.3. Measurements (or characterization) to describe all the tools/devices and instruments used in this study, including the contact angle measurement)

We have rewritten each sub-chapter 2.1 Materials, 2.2. Methods, and 2.3. Characterization separately. In this section we add the sentence:

"*The morphology and chemical composition of the compounds in the thin film was observed using the Analytical Scanning Electron Microscopy – Energy Dispersive X-ray instrument (SEM – EDX JEOL JSM-6510LA). Analysis of the thin film crystallinity using X-Ray Diffraction instrument (Shimadzu Maxima XRD-700). Determination of the contact angle of the tips film using a digital microscope endoscope camera instrument. Optical characteristics and efficiency photodegradation of dyes and Escherichia coli were analyzed based on the results of the UV-Vis Spectrophotometer (Shimadzu UV-Vis 1240 SA)"* -**(Page 2)**

3. The x-axes in Fig. 2 is the incident photon energy E = hυ, not Eg.

   We have fixed the X-axis of the energy bandgap graph – **(Page 4)**

4. Mention above each peak in Fig. 4, whether the peak belongs to ZnO, Fe or Fe2O3 combined with the Miller's indices.

   We have corrected figure 4 and listed each peak with a Miller index. - **(Page 6)**

5. Correct the typing mistakes in equations 1,2, 5 and 6 concerning with the symbol "θ"

   We have fixed equations 1,2,5, and 6 with the symbol "θ" – **(Page 7)**

6. In Table 3, the parameters A and C should be small a and c and italic.

   We have fixed parameter names in Table 3 – **(Page 7, No. 22)**

7. It is important to compare the obtained results (XRD, optical, SEM, contact angles and Photocatalytic) with the published papers, such as:

   - Enhanced Gas Sensing Properties of Spin-coated Na-doped ZnO Nanostructured Films: Sci. Rep. 7, Article number: 41716 (2017) https://doi.org/10.1038/srep41716

   - Influences of lead and magnesium co-doping on the nanostructural, optical properties and wettability of spin coated zinc oxide films: Mater. Sci Semicond Process., 39, 2015, 136-147 https://doi.org/10.1016/j.mssp.2015.04.008

- Effects of lanthanum and sodium on the structural, optical and hydrophilic properties of sol–gel derived ZnO films: A comparative study: Mater. Sci. Semicond. Process.41, 2016, 323-334. https://doi.org/10.1016/j.mssp.2015.09.002

We have cited more references, hopefully adequate for this research paper. We have added an explanation of the critical analysis of the results section and a discussion of the manuscript and comparison with several previous studies.

"This result is consistent with the study reported by Ariyakkani et al. (2017) for Fe-doped ZnO thin films, where the Eg value decreased from 3.38 eV for undoped ZnO to 3.07 eV for 20% Fe-doped ZnO [26]…………………………………………………………………………......etc"

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"This result is similar with the study reported by Roguai and Djelloul (2021), where all the peaks of the XRD spectrum (100), (002), (101), (102), (110), (103), and (200) for Fe-doped ZnO (0-10%) are identified as single-phase ZnO wurtzite structure with the space group $P6_3mc$ [38]……………………………………………………………………………………......etc"

-(Page 7, No 5)

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Djaja (2014) for the degradation of methyl orange [45], and Roguai & Djelloul (2021) for the degradation of methylene blue [38]………………………………………………………………etc"

-(Page 9, No 18)

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Re: “Analysis of Fe-doped ZnO Thin Films for Degradation of Rhodamine B, Methylene Blue, and Escherichia Coli Under Visible Light"
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We notice the following author has been added to your author list at the revised submission stage:

Marhaendrajaya, Indras
Diponegoro University Faculty of Science and Mathematics - Physics
Jl. Prof. Soedarto, SH Tembalang, Semarang, Indonesia, Semarang 50275
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Dear Dr Sutanto,

Re: "Analysis of Fe-doped ZnO Thin Films for Degradation of Rhodamine B, Methylene Blue, and Escherichia Coli Under Visible Light"
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COMMENTS TO THE AUTHOR(S)
The paper sounds good.

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COMMENTS TO THE AUTHOR(S)

The Authors have made all the necessary changes that were suggested in the previous review and now this article can be considered for publishing in this journal.

Letter reference: ERWSA01

Heri Sutanto <herisutanto@live.undip.ac.id>

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Materials Research Express <onbehalfof@manuscriptcentral.com> Wed, Oct 27, 2021 at 6:14 PM
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Dear Dr Sutanto,

Re: "Analysis of Fe-doped ZnO thin films for degradation of rhodamine b, methylene blue, and Escherichia coli under visible light"
Article reference: MRX-124508.R1

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mrx@ioppublishing.org <mrx@ioppublishing.org> Sat, Oct 30, 2021 at 7:56 PM
To: herisutanto@live.undip.ac.id

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mrx@ioppublishing.org <mrx@ioppublishing.org> Wed, Nov 3, 2021 at 11:48 AM
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