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To cite this article: N A Heshammuddin *et al* 2024 *IOP Conf. Ser.: Earth Environ. Sci.* **1347** 012004

Optimization of eco-waste synthesis in zinc oxide-graphene oxide for cephalexin antibiotics removal

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Abstract. Cephalexin (CFX) is an antibiotic prescribed for a wide variety of conditions, including bacterial infections and urinary tract infections. It has recently been labelled as an emerging pollutant due to the high concentrations of CFX that indicate a potential risk to the environment. This research set out to discover the best conditions for producing a leachate from waste materials utilising the Eco-Zinc Oxide-Graphene Oxide (Eco-ZnO/GO) catalyst for the efficient removal of CFX. The Eco-ZnO/GO nanocomposite that were generated from waste is used for adsorption in an aqueous solution. Field Emission Scanning Electron Microscope (FESEM) was used to characterise Eco-Zno/GO nanocomposite while Response Surface Methodology (RSM) were used to optimise the removal efficiency of CFX across three factors: starting concentration, mixing speed, and contact time. Average nanoparticle sizes for Eco-ZnO and GO were measured to be 23 and 122 nm, respectively. Eco-ZnO/GO has a maximum removal efficiency of 89.55% at the optimal factor of 1000 rpm in 30 minutes when working with a concentration of 100 mg/l of CFX. The results of the study demonstrated that the waste nanocomposite technology is used to remove the material effectively, which can add to our growing body of knowledge in this area.

1. Introduction

Cephalexin (CFX) is among the 18 pharmaceutical compounds that were detected in water samples with concentrations of 75% or higher [1]. Due to its efficacy in combating various bacterial illnesses, such as urinary tract infections and lung infections [2], CFX has become a common treatment option prescribed by doctors. In addition to its effectiveness in treating gonorrhea, bronchitis, pneumonia, scarlet fever, and infections of the lower and upper respiratory tracts, CFX also demonstrates efficacy against beta-lactamase producing staphylococcal infections and streptococcal septicemia [3]. Though it has applications in the medical industry, it is a form of emerging pollutant that has the potential to cause damage to the natural environment. The conventional ways of the water treatment system, lead to the production of secondary by-products, makes the development of new technologies are necessary. The development of nanotechnology, on the other hand, will be one of the solutions to which will finally be provided by modern technological progress.

Nanotechnology is one example of how modern technological progress is facilitating the elimination of previously intractable issues. Due to its superior speed and efficiency, nanotechnology has found widespread use in a variety of fields since its rapid development coincided with the onset of modernisation [4]. As it develops into a more integral aspect of cutting-edge technology, nanotechnology is increasingly vital to modern society. Nanotechnology has a rapidly expanding market in the commercial sector since it is possible in improving the result accuracy [5]. In this research, the removal of CFX antibiotic can be utilised by using nanoparticle-sized Zinc Oxide-Graphene Oxide (Zno/GO) generated from ecological waste.

ZnO nanoparticles are more appealing for wastewater treatment due to their potential as a multifunctional agent with qualities including catalysis, photochemical capabilities, medicinal effects, fungicidal, antibacterial, and UV filtering [6]. On the other hand, graphene oxide's excellent conductivity, selectivity, and sensitivity have piqued the curiosity of numerous nanomaterials researchers [7]. Graphene oxide is a good candidate for use in experimental optimisation studies due to the properties already demonstrated. Hence, Eco-waste from ZnO-GO synthesizing has attracted the attention of many researchers who are interested in its potential for use in the elimination of the CFX antibiotics from water.

Adsorption using ZnO/GO generated from eco-waste was used as the mechanism for CFX elimination in this investigation. The adsorption method outperforms alternative approaches because of its low cost, ease of use, and ability to completely purge pharmaceuticals from contaminated water [8]. The higher removal efficiency of this procedure has made it the method of choice among researchers. Each adsorption agent's nanocomposite has a unique set of capabilities when it comes to removal [9]. The adsorbent capacity of each synthesised nanocomposite is a variable that can affect the efficiency of the elimination [10]. Thus, with the help of the nanocomposite eco-waste generated ZnO/GO, CFX antibiotics was removed from treated effluent and sewage, with the hopes of developing new green technologies.

2. Materials and Methods

The eco waste samples were collected at the landfill in Simpang Renggam. Eco-waste produced ZnO/GO nanocomposite is prepared for synthesis. The characterisation of ZnO/GO nanocomposite that has been synthesized from eco-waste will be utilised by using Field Emission Scanning Electron Microscope (FESEM). The optimization was conducted by using RSM, the relationship between mixing speed and contact time is evaluated in order to determine the optimal nanocomposite for removing CFX.

2.1. Leachate wastewater as eco-waste

Leachate wastewater samples were collected at the Simpang Renggam Landfill. The selection of the location was due to the value of BOD_5/COD ratio greater than 0.1 that originated from long-defunct landfills that were later stabilised. The site is located at 1°57'29.4"N and 102°54'49.2"E, in Johor, Malaysia.

A sample of leachate was collected from the landfill, stored in a 100 ml bottle in a cooling box, and rushed to the lab for microbiological analysis as soon as possible (within 2 hours). The analysis and procedures used in the experiments were conducted in accordance with the guidelines established by the American Public Health Association (APHA, 2008).

2.2. Synthesis of eco-waste derived ZnO/GO

The precipitation process was utilised to synthesise a nanocomposite consisting of ZnO produced from eco-waste and GO. The process of preparing the synthesis of eco-waste ZnO/GO is outlined in Figure 1, which may be found below.

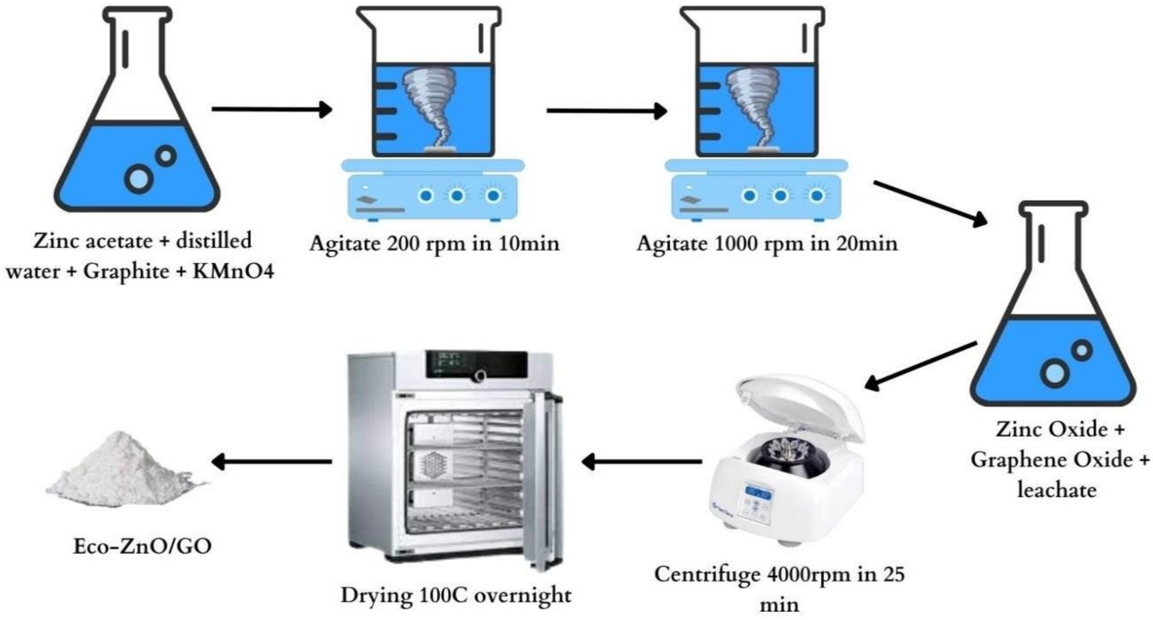

Figure 1. Synthesis of eco-waste derived ZnO/GO

2.3. Optimization of eco-waste derived ZnO/GO

Factors parameters include mixing speed and contact time for CFX removal were subjected to an optimisation analysis and evaluation, respectively, in the RSM. The effectiveness of removing CFX from an aqueous solution was investigated by measuring two factors that have been evaluated by Response Surface Methodology (RSM): the mixing speed and the contact time. The effectiveness of ZnO/GO produced from eco-waste in the elimination of CFX is dependent on the parameters under which it is used. As a promising and accessible technique, adsorption has the potential to purge water of a wide variety of contaminants. The amount of CFX in the sample was used to determine how accurately to compute the adsorption effectiveness of the removal process. It is generally agreed that the removal condition should be at its highest point when possible. The equation describing the effectiveness of the removal was shown as follows:

$$E(\%) = \left(\frac{C_i - C_f}{C_i}\right) \times 100\% \tag{1}$$

Where E is efficiency of adsorption (%), Ci is initial concentration of CFX (mg/L), and Cf is final concentration (mg/L).

3. Results and discussions

The results incorporate both the characterization and optimisation of ZnO/GO produced from eco-waste to maximise the effectiveness of CFX removal.

3.1. Characterization of eco-waste derived ZnO/GO

Figure 2 depicts the nanocomposite synthesis of the Eco-ZnO/GO size grain. Each graphic demonstrates that the voltage of the electron images must be maintained at 5.0 kV so that they can be viewed. As shown in Figure 2, the nanoparticle size of the synthesis Eco-ZnO is 23 nm, whereas the size of the GO particles measured 122 nm. A study using a scanning electron microscope (SEM) found that ZnO NPs are round and that their typical particle size is between 15 and 35 nm [11]. The majority of GO particles ranged in size from 30 to 200 nm, while there was also a sizable population of particles somewhere about 300 nm [12]. Hence, the nanoparticle size for both eco-ZnO and GO is within their average size. Utilising the ImageJ software, the measurements of the synthesized were carried out by averaging their sizes. On Eco-ZnO/GO, all nanoparticles have an irregular form, which can increase the efficiency of the removal process through adsorption.

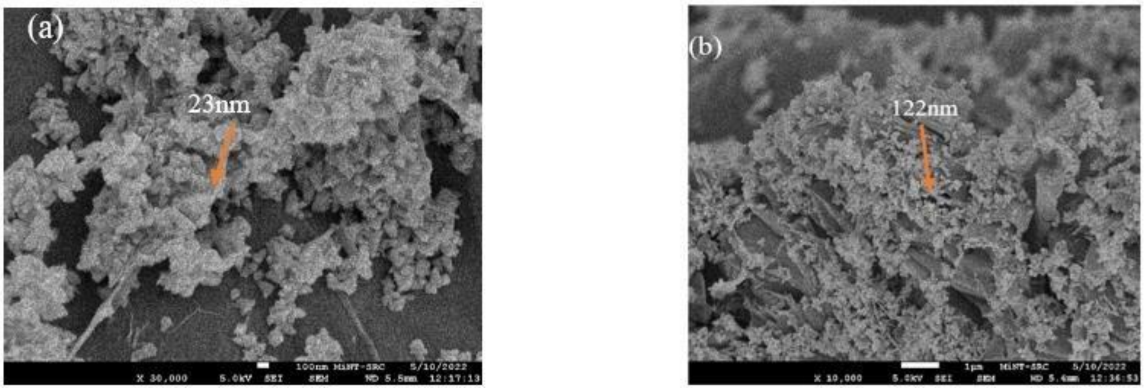

Figure 2. FESEM images synthesized Eco-ZnO/GO NPs (a) Eco-ZnO (b) GO

3.2. Optimization of eco-waste derived ZnO/GO

Table 1 presents the results of the experiments that were conducted using the RSM software and collected data for the parameters mixing speed, contact time and removal efficiency.

Table 1. Data of optimization result based on factor mixing speed and contact time

Std	Run	Factor 1 A: Contact time (min)	Factor 2 B: Mixing speed (Rpm)	Efficiency of Removal CFX (%)
8	1	45	1000	29.89
7	2	45	200	26.44
12	3	45	600	82.46
3	4	30	1000	89.55
5	5	30	600	88.06
10	6	45	600	64.37
4	7	60	1000	78.95
1	8	30	200	71.93
9	9	45	600	5.26
11	10	45	600	83.58
13	11	45	600	56.32
2	12	60	200	85.06
6	13	60	600	87.72

According to Table 1, the maximum mixing speed was 1000rpm, and the maximum contact time was 60 minutes. Contact time of 30 minutes and a mixing speed of 1000rpm, the CFX removal efficiency data showed a maximum of 89.55%. The lowest efficiency was achieved at 600rpm and 45 min, while the average efficiency was 65.35 %. However, the efficiency of mixing at 1000 rpm (the highest possible speed) is consistently above 70%. The 1000rpm mixing speed resulted in effectively reduce the CFX in water. Extremely rapid mixing reveals the equilibrium distribution of adsorbate molecules in the solution and on the adsorbent, demonstrating how CFX may be efficiently removed [13]. This was determined by conducting a contact a study of the optimal mixing speed using five different variations.

Optimization study shows that the 30 minutes had the highest efficiency, ranging between 70% and 90%. This range is indicative of the sweet spot for removing CFX. Although a steep decline in productivity could potentially influence the optimal percentage factor value. In conclusion, this study suggests that leachate can be used as a catalyst to increase CFX removal efficiency to >60%. The results show high efficiency has been achieved for CFX removal.

3.3. Factors that influence the efficiency of eco-waste derived ZnO/GO efficiency in degradation of CFX in aqueous solution

The graph 3D has been plotted for mixing speed, contact time and efficiency removal CFX is shown in the Figure 3.

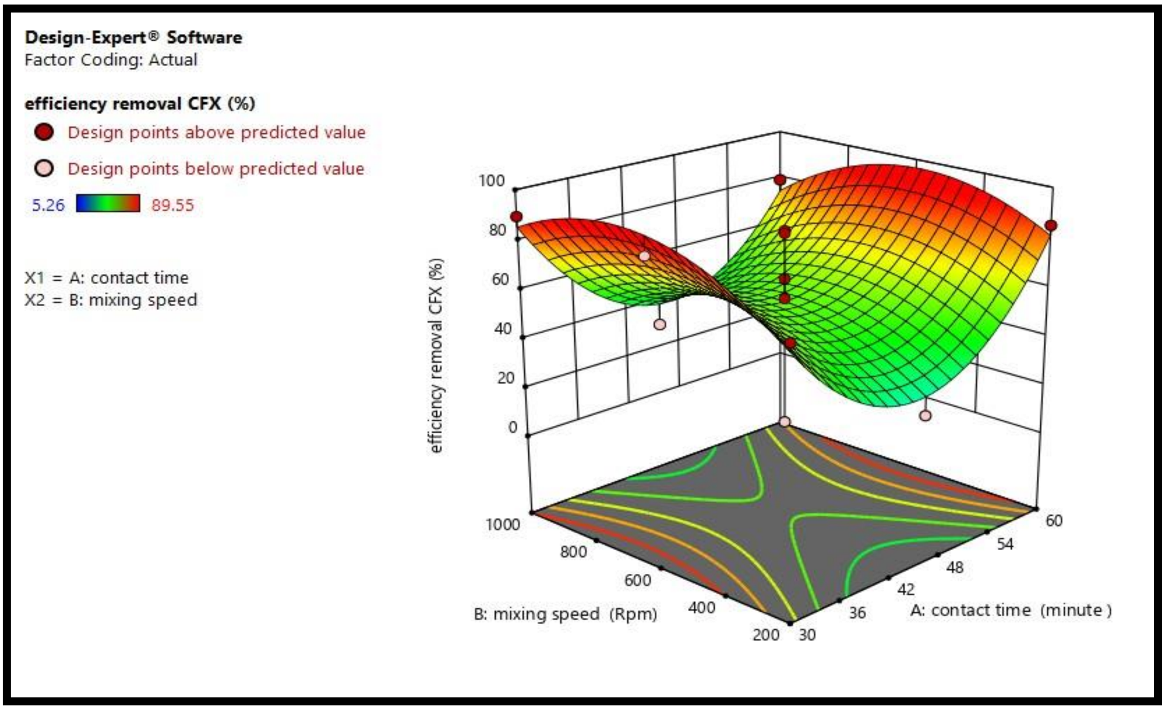

Figure 3. 3D graph model mixing speed (rpm), contact time (min), and efficiency removal CFX (%)

Figure 3 illustrates the optimal removal efficiency. Based on Figure 3, the efficient removal of CFX is in between 30-60 minutes of contact time and 200-1000rpm for mixing speed. Based on the graph, it was concluded that the best removal of CFX can be obtained by doing it in less contact time while raising the mixing speed. The 3D graph shows that there are two possible optimum places. When the mixing speed was set to 200 rpm, the best contact time was found to be between 42 and 48 minutes. It is important to assess how effective removal will be under the two optimal conditions that will be evaluated.

3.4. Statistical analysis

The ANOVA data analysis by using RSM is showed in Table 2.

Table 2. Analysis of variance (ANOVA) table result

Source	Sums of square	df	Mean of square	F-value	P-value
Model	4841.08	5	968.22	1.52	0.2969
A-contact time	0.7994	1	0.7994	0.0013	0.9728
B-mixing speed	37.30	1	37.30	0.0584	0.8159
AB	140.78	1	140.78	0.2206	0.6529
A^2	4629.38	1	4629.38	7.25	0.0309
B^2	974.52	1	974.52	1.53	0.2564
Residual	4467.25	7	638.18		
Lack of fit	390.51	3	130.17	0.1277	0.9388
Pure error	4076.74	4	1019.19		
Cor total	9308.34	12			

As in Table 2, the model has a F-value of 1.52, which indicates that the model is not significantly different from the noise. A F-value of this magnitude might be caused by noise with a probability of 29.69%. P-values that are lower than 0.0500 denote the significance of the model terms. In this instance, A^2 is an important model term which when the model terms are not significant, values that are greater than 0.1000 show this.

The fact that the Lack of Fit F-value is only 0.13 signifies that it is not statistically significant in comparison to the pure error. A Lack of Fit F-value of this magnitude might be caused by noise with a probability of 93.88%, according to the analysis. A lack of fit that is not significant is desirable.

3.5. Fits statistics

The difference between the adjusted R^2 of 0.1773 and the predicted R^2 of 0.0736 is less than 0.2, indicating that there is reasonable agreement between the two values. The signal-to-noise ratio is something that may be measured using Adeq Precision. A ratio of 3.65 shows that there is insufficient signal. The results of the statistical models that were used to fit the data in this investigation are displayed in Table 3.

Table 3. Fit of statistics model of waste derived eco-ZnO/GO

Std dev	25.26	R^2	**0.5201**
Mean	65.35	Adjusted R^2	**0.1773**
C.V.%	38.65	Predicted R^2	**0.0736**
		Adeq precision	**3.6466**

The value of the standard deviation was found to be 25.26, which indicates that the experimental testing may have been subject to some degree of human error. The value of projected R^2 differs significantly from the value of adjusted R^2, which is something that can be produced from the preparation of eco-waste derived ZnO/GO. Since the preparation of the eco-waste could potentially result in an error during testing of the parameter, the inaccuracy cannot be addressed. It is highly encouraged to carry out repeated experiments till an acceptable result is found.

4. Conclusion

Leachate samples extracted from the Simpang Renggam Landfill site was successfully employed in synthesizing waste-derived eco-waste generated ZnO/GO. Characterization based on the electron image indicated that eco-ZnO nanoparticles has an average size of 23 nm, while GO nanoparticles had an average size of 122 nm. The size and stability of the nanoparticles were directly influenced by anions, pH, and concentration. CFX removal efficiency revealed that the highest removal efficiency occurred after a 30-minute contact time at a mixing speed of 1000 rpm could exceeded 60%. However, further research is needed to study the removal mechanism of the CFX by using nanocomposite. Additionally, expanding the research to include more parameters using waste nanocomposites made from various materials is recommended.

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Acknowledgments

The authors would like to thank the Ministry of Higher Education (MOHE) Malaysia through Fundamental Research Grant Scheme (FRGS) (FRGS/1/2021/WAB02/UTHM/02/1) and UTHM through GPPS grant vot Q326. We also thankful to the Micropollutant Research Centre Laboratory Assistant, Universiti Tun Hussein Onn Malaysia for their support.