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Article Information Overview

Manuscript ID	**coatings-1448119**
Status	Website online
DOI	10.3390/coatings11121452
Publication Certificate	Download Publication Certificate (PDF)
Banner	Download Banner (PDF) (/publication/articler/banner/689035)
Website Links	Abstract (https://www.mdpi.com/2079-6412/11/12/1452) HTML version (https://www.mdpi.com/2079-6412/11/12/1452/htm) PDF version (https://www.mdpi.com/2079-6412/11/12/1452/pdf) Manuscript (https://www.mdpi.com/2079-6412/11/12/1452/manuscript)
Article type	Article
Title	Synthesis, Characterization and Photodegradation Studies of Copper Oxide–Graphene Nanocomposites
Journal	*Coatings* (https://www.mdpi.com/journal/coatings)
Volume	11
Issue	12
Section	Corrosion, Wear and Erosion (https://www.mdpi.com/journal/coatings/sections/Corrosion_Wear_Erosion_Protection)
Special Issue	Strengthening, Corrosion and Protection of High Temperature Structural Materials (https://www.mdpi.com/journal/coatings/special_issues/high_temp_struct_mater)
Abstract	In this work, a simple hydrothermal method was employed to prepare a pristine sample of copper oxide (CuO) and three samples of copper oxide–graphene nanocomposites (CuO-xG) with x = 2.5, 5, and 10 mg of graphene. The synthesized samples were characterized using X-ray powder diffractometry (XRD), field emission scanning electron microscopy (FESEM), energy-dispersive X-ray spectroscopy (EDX), Fourier transform infrared spectroscopy (FTIR) and ultraviolet–visible (UV-Vis) spectroscopy. The XRD patterns of CuO-xG nanocomposites exhibited the diffraction peaks related to the crystal planes of monoclinic CuO and hexagonal graphite. The surface morphology of the prepared samples was investigated using FESEM images. EDX analysis was used to investigate the chemical composition of the synthesized samples. FTIR spectroscopy identified the vibrational modes of the covalent bonds present in the samples. The allowed direct optical bandgap energy was calculated for all prepared samples using UV-Vis absorption spectra. The small bandgap of CuO-xG nanocomposites indicates their potential use as an effective photocatalyst in the presence of visible light. Photocatalytic activity of the samples was explored for the degradation of methylene blue (MB) dye contaminant under visible light irradiation. The results showed that the CuO-5G sample has the highest photodegradation efficiency (~56%).
Keywords	CuO–graphene nanocomposite; hydrothermal synthesis; structural properties; FESEM; optical bandgap; photocatalytic activity

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Author Information

Submitting Author	Awais Ahmad
Corresponding Authors	AWAIS AHMAD, Mahyuddin K.M. Nasution, Rafael Luque, Mabkhoot A Alsaiari
Author #1	Indah Raya (https://orcid.org/0000-0001-8575-1994)
Affiliation	1. Department of Chemistry, Faculty Mathematics and Natural Science, Hasanuddin University, Makassar 90245, Indonesia
E-Mail	indahraya@unhas.ac.id **(co-author email has been published)**)
Author #2	Awais Ahmad (https://orcid.org/0000-0003-4774-3111)
Affiliation	2. Departamento de Quimica Organica, Universidad de Cordoba, Edificio Marie Curie (C-3), Ctra Nnal IV-A, Km 396, E14014 Cordoba, Spain
E-Mail	awaisahmed@gcuf.edu.pk **(co-author email has been published)**)
Author #3	Ayad F. Alkaim
Affiliation	3. Department of Chemistry, College of Science for Women, University of Babylon, Hilla 51002, Iraq
E-Mail	alkaimayad@gmail.com **(co-author email has been published)**)
Author #4	Dmitry Bokov
Affiliation	4. Institute of Pharmacy, Sechenov First Moscow State Medical University, 119991 Moscow, Russia
E-Mail	fmmsu@mail.ru **(co-author email has been published)**)
Author #5	Enas R. Alwaily
Affiliation	5. Microbiology Research Group, College of Pharmacy, Al-Ayen University, Nasiriyah 64011, Iraq
E-Mail	enas.kazem@alayen.edu.iq **(co-author email has been published)**)
Author #6	Rafael Luque (https://orcid.org/0000-0003-4190-1916)
Affiliation	2. Departamento de Quimica Organica, Universidad de Cordoba, Edificio Marie Curie (C-3), Ctra Nnal IV-A, Km 396, E14014 Cordoba, Spain
E-Mail	q62alsor@uco.es **(corresponding author email)**

Author #7	Mabkhoot Alsaiari (https://orcid.org/0000-0002-4366-3256)
Affiliation	6. Promising Centre for Sensors and Electronic Devices (PCSED), Advanced Materials and Nano-Research Centre, Najran University, Najran 11001, Saudi Arabia 7. Empty Quarter Research Unit, Department of Chemistry, College of Science and Art in Sharurah, Najran University, Najran 11001, Saudi Arabia
E-Mail	mabkhoot.alsaiari@gmail.com **(corresponding author email)**
Author #8	Mohammed Jalalah
Affiliation	6. Promising Centre for Sensors and Electronic Devices (PCSED), Advanced Materials and Nano-Research Centre, Najran University, Najran 11001, Saudi Arabia 8. Department of Electrical Engineering, College of Engineering, Najran University, Najran 11001, Saudi Arabia
E-Mail	jalalah@gmail.com **(co-author email has been published)**)

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Editor Decision

Decision	Accept in current form
Comments	This manuscript has been revised and replied according to the comments of reviewers. It could be accepted for publication in Coatings.
Decision Date	19 November 2021

Review Report

Reviewer 1	Review Report (Round 1) (/user/manuscripts/review/21776434?report=15279938)
Reviewer 2	Review Report (Round 1) (/user/manuscripts/review/21776709?report=15279925)
Reviewer 3	

Review Report (Round 1) (/user/manuscripts/review/21815199?report=15310964)

Review Report (Round 2) (/user/manuscripts/review/21815199?report=15688041)

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Previously Published Papers

Al-Kahtani, A.A.; Tabassum, S.; Raya, I.; Khlewee, I.H.; Chupradit, S.; Davarpanah, A.; Elveny, M.; Ali, S. Influence of Different Rotations of Organic Formamidinium Molecule on Electronic and Optical Properties of $FAPbBr_3$ Perovskite. *Coatings* **2021**, *11*, 1341. doi: 10.3390/coatings11111341 (https://doi.org/10.3390/coatings11111341)

Related Papers Published in MDPI Journals

Nwanna, E.C.; Imoisili, P.E.; Bitire, S.O.; Jen, T.-C. Biosynthesis and Fabrication of Copper Oxide Thin Films as a P-Type Semiconductor for Solar Cell Applications. *Coatings* **2021**, *11*, 1545. doi: 10.3390/coatings11121545 (https://doi.org/10.3390/coatings11121545)

Witjaksono, G.; Junaid, M.; Khir, M.H.; Ullah, Z.; Tansu, N.; Saheed, M.S.B.M.; Siddiqui, M.A.; Ba-Hashwan, S.S.; Algamili, A.S.; Magsi, S.A.; Aslam, M.Z.; Nawaz, R. Effect of Nitrogen Doping on the Optical Bandgap and Electrical Conductivity of Nitrogen-Doped Reduced Graphene Oxide. *Molecules* **2021**, *26*, 6424. doi: 10.3390/molecules26216424 (https://doi.org/10.3390/molecules26216424)

Sagadevan, S.; Lett, J.A.; Weldegebrieal, G.K.; Garg, S.; Oh, W.-C.; Hamizi, N.A.; Johan, M.R. Enhanced Photocatalytic Activity of rGO-CuO Nanocomposites for the Degradation of Organic Pollutants. *Catalysts* **2021**, *11*, 1008. doi: 10.3390/catal11081008 (https://doi.org/10.3390/catal11081008)

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Journal: Coatings (https://www.mdpi.com/journal/coatings) (ISSN 2079-6412)
Manuscript ID: coatings-1448119
Type: Article

Title: Synthesis, structural, morphological, optical and photocatalytic characterization of CuO-graphene nanocomposites (https://www.mdpi.com/2079-6412/11/12/1452)

Authors: Indah Raya , AWAIS AHMAD * , Afshin Davarpanah , Marischa Elveny , Mahyuddin K.M. Nasution * , Rafael Luque * , Mabkhoot A Alsaiari * , Mohammed Jalalah

Section: Corrosion, Wear and Erosion (https://www.mdpi.com/journal/coatings/sections/Corrosion_Wear_Erosion_Protection)

Special Issue: Strengthening, Corrosion and Protection of High Temperature Structural Materials (https://www.mdpi.com/journal/coatings/special_issues/high_temp_struct_mater)

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Authors' Responses to Reviewer's Comments (Reviewer 1)

Author's Notes: The authors successfully prepared a pristine sample of CuO and three samples of CuO-xG nanocomposites with x= 2.5, 5, and 10 mg of graphene. A lot of works including structural characterizations and mechanism analysis have been done in this paper. The paper was well written and the results are quite interesting. Therefore, I recommend its publication in Coatings after minor revisions. Some specific comments are listed below:

Firstly, we would like to thank you for commenting on our paper. We do believe that your comments improve the quality of our paper. We have addressed your comments below and implemented them in the revised manuscript.

1. A maximum of 6 keywords is considered to be appropriate; the quantity of keywords in this paper should be reduced.

We thank the reviewer for pointing this out. The number of keywords has been reduced to 6.

1. According to Table 4, “The average diameter of grains grown on the graphene sheets is about 28.92, 20.11 and 40.85 nm for CuO-2.5G, CuO-5G and CuO-10G nanocomposites, respectively”, authors should explain why the average diameter of grains for CuO-5G is the minimum value with the amount of graphene increasing.

We appreciate this comment. In fact, when the amount of graphene is low, ultrasonic dispersion can make the graphene disperse well. However, when the amount of addition is large (~ 10 mg), the graphene is not easy to disperse and agglomerates obviously. The explanation of FESEM results has been revised.

1. As shown in Figure 6, authors should illustrate the meaning of Figure 6 and what it can prove, rather than just one sentence.

The description of UV-Visible absorption spectra has been updated in the revised manuscript.

1. Introduction should be supplemented in reference to the graphene and metal oxide composite materials for example, “Vertical carbon skeleton introduced three-dimensional MnO_2 nanostructured composite electrodes for high-performance asymmetric supercapacitors, Journal of Power Sources, Volume 476, 15 November 2020, 228527”, and “Defects engineering of Fe_2O_3@Sn_2O_3 nanosheet arrays for high-performance hybrid supercapacitor, Journal of Energy Storage, Volume 42, October 2021, 103123.”

The references have been modified, according to the reviewer’s comment.

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( ) Moderate English changes required
(x) English language and style are fine/minor spell check required
( ) I am not qualified to assess the quality of English in this paper

	Yes	Can be improved	Must be improved	Not applicable
Does the introduction provide sufficient background and include all relevant references?	( )	(x)	( )	( )
Is the research design appropriate?	(x)	( )	( )	( )
Are the methods adequately described?	(x)	( )	( )	( )
Are the results clearly presented?	(x)	( )	( )	( )
Are the conclusions supported by the results?	(x)	( )	( )	( )

Comments and Suggestions for Authors

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Date of this review	21 Oct 2021 03:23:23

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Journal: Coatings (https://www.mdpi.com/journal/coatings) (ISSN 2079-6412)
Manuscript ID: coatings-1448119
Type: Article

Title: Synthesis, structural, morphological, optical and photocatalytic characterization of CuO-graphene nanocomposites (https://www.mdpi.com/2079-6412/11/12/1452)

Authors: Indah Raya , AWAIS AHMAD * , Afshin Davarpanah , Marischa Elveny , Mahyuddin K.M. Nasution * , Rafael Luque * , Mabkhoot A Alsaiari * , Mohammed Jalalah

Section: Corrosion, Wear and Erosion (https://www.mdpi.com/journal/coatings/sections/Corrosion_Wear_Erosion_Protection)

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Authors' Responses to Reviewer's Comments (Reviewer 2)

Author's Notes: There are some issues which, in my opinion, should be tackled in order to improve the quality of the manuscript:

We sincerely appreciate your valuable comments and suggestions, which helped us to improve the quality of the paper. We have provided a point-by-point response to the reviewer's comments.

1. Add a table with title <list of abbreviations> to the article. Note: The list of abbreviations should be in alphabetical order.

We appreciate the reviewer for these observations. All abbreviations have been checked and defined correctly in the text.

1. Each acronym should be explained the first time it appears in the text, even if it appeared in the abstract. Check all abbreviations in text: each word

should start with capital to explain an abbreviation?

All abbreviations have been checked and defined correctly in the text.

1. An additional English grammar and spelling check should be performed.

The manuscript underwent a rigorous editing process, and all mistakes in English grammar, spelling, subscripts and punctuation have been corrected.

1. Avoid having headlines follow directly - e.g., section “3. Results” and “3.1. Structural and Morphological Analysis. Use the space in between to introduce what comes, why, and how it is structured (add a paragraph).

With respect to the reviewer’s comment, we think the paper is well structured (according to the literature).

1. Conclusions are not sufficient and need to be redesigned.

We thank the reviewer for this comment. The Conclusions section has been revised.

1. Some references are clustered ([1-14] and [15-28] and [29-42] and [43-56]), it would be good to separate them and describe in short, the relevance of each of them.

The references have been checked and modified, according to the reviewer’s comment.

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Quality of English Language

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	Yes	Can be improved	Must be improved	Not applicable
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Are the methods adequately described?	( )	(x)	( )	( )
Are the results clearly presented?	( )	(x)	( )	( )
Are the conclusions supported by the results?	( )	(x)	( )	( )

Comments and Suggestions for Authors

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Submission Date	19 October 2021
Date of this review	19 Oct 2021 14:17:51

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Manuscript ID: coatings-1448119
Type: Article

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Authors: Indah Raya , AWAIS AHMAD * , Afshin Davarpanah , Marischa Elveny , Mahyuddin K.M. Nasution * , Rafael Luque * , Mabkhoot A Alsaiari * , Mohammed Jalalah

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Authors' Responses to Reviewer's Comments (Reviewer 3)

Author's Notes

Dear Authors,

the scientific area of your manuscript entitled Synthesis, structural, morphological, optical, and photocatalytic characterization of CuO-graphene nanocomposites addresses the experimental characterization of carbon-based copper oxide composites is according to the papers published in Coatings journal and this work can be interesting to the readers.

However, your manuscript can be published in Coatings journal after major revision.

You have to improve the presentation of scientific results, i.e., data results are not properly presented, analyzed, and furthermore, the main conclusions are not supported by the data.

Please, the responses to the questions and comments should be added to the text of your corrected manuscript.

We thank the reviewer for the time spent in reviewing our paper.

Materials and Methods

1. In section 2.4. Photocatalytic Activity of the Samples, you have to add to the description information about reactor parameters, temperature for each experiment condition (dark adsorption, and photocatalysis), solution volumes that were taken from the suspensions, and analysed using a UV-Visible spectrophotometer.

According to the literature, we have included all required information on sample preparation, measurements and photocatalytic test conditions.

1. How did you estimate that the error for the average crystallite size (calculated from Debye-Scherrer formula) and average grain diameter (from STEM images) is always 0.01 nm?

We reviewed the previous similar studies and decided to remove the errors of the parameters you mentioned.

1. You have to add more details about the preparation of samples to the measurements using XRD, FESEM, FTIR, and UV-Vis spectroscopy (e.g. resolution, the form of sample, e.g. powder, KBr^{+} sample, or colloid solution) and the procedure of photocatalytic tests (the volume of solution which was collected to the analysis using UV-vis spectroscopy, the separation between catalyst and reaction mixture).

According to the literature, we have included all required information on sample preparation, measurements and photocatalytic test conditions.

Results

1. How you can explain the differences in the average crystallite size determined by X-Ray diffraction patterns and the average grain diameter estimated using SEM images?

Crystallite size is the smallest - most likely single crystal in powder form. The crystallite size commonly determined by XRD (using the Debye–Scherrer equation). Grain is either a single crystalline or polycrystalline material, and is present either in bulk or thin film form. During the processing, smaller crystallites come closer and grow to become larger due to kinetics. Therefore, in the most likely scenario, the grain is larger than a crystallite. And, the grain morphology is commonly determined by FESEM (but not XRD). From the FESEM images, When CuO is incorporated with graphene, we see the formation of graphene sheets on which CuO grains appear to grow. The presence of the sheets becomes clearer with increasing the amount of graphene in the nanocomposite. The average diameter of grains grown on the graphene sheets is about 28.92, 20.11 and 40.85 nm for CuO-2.5G, CuO-5G and CuO-10G nanocomposites, respectively (see Table 4). When the amount of graphene is low, ultrasonic dispersion can make the graphene disperse well. However, when the amount of addition is large (~ 10 mg), the graphene is not easy to disperse and agglomerates obviously.

1. The values of Cu, O, and C contents suggest the differences in the atomic ratio between Cu and O species, e.g. according to the data in Table 5, CuO is represented by CuO_2 (Cu:O = 36.70:63.30), CuO-2.5G by CuO (Cu:O = 22.20:27:58), CuO-5G by CuO4 (Cu:O = 4.78:19.02), and GCuO-10G by CuO7 (Cu:O = 4.28:33.15). How you can explain the differences in the presented results?

These are the direct results of EDX spectroscopy performed on the samples. Obviously, when the amount of graphene in the composition increases, the atomic percentage of copper decreases. Nonlinear variation of the atomic percentage of oxygen can be related to the different covalent bonds present in each sample.

1. The results of EDS analysis (high atomic ratio between oxygen and copper species) and FTIR spectra (the presence of the bands, which can be due to the stretching vibration of C-O, and C=O bonds) suggest that the obtained composites can be a mixture of graphene and graphene oxide with copper oxides.

With respect to the reviewer' opinion, according to the literature, we do believe that the word "graphene" is more appropriate for our study than "graphene oxide"

1. How can you explain the presence of bands in the EDX spectra at around 1.5-2.0 eV?

These band are due to the elements present in the substrate holder. Getting such signal from the sample holder is quite common in EDX analysis.

1. How can you explain the high absorption in the range of 800-1000 nm?

In response to comment 3 by Reviewer 1, we have revised the description of UV-Visible absorption spectra. Please see the section *Evaluation of Optical Bandgap* in the revised manuscript.

1. The quality of Figure 9 must be improved – the colours of the chemical formulas are not visible. You don't have to use the colours like light green, light blue, orange, rose, etc.

We apologize for this mistake. Due to irrelevance, Figure 9 (c) has been removed form the manuscript.

1. Did you perform the recycling test for all catalysts?

No. Unfortunately, the recycling test was not performed

1. Did you check the possibility of any changes in the morphology of studied materials during the catalytic tests?

No. It was not considered.

1. How did you estimate the presence of selected products of MB photocatalytic degradation which are shown in Figure 9?

The photodegradation efficiency was calculated based on the initial absorbance of the MB (A_0) and the absorbance after 1, 2, 3, 4, 5, 6, 7, 8 and 9 h irradiation (A_t), recorded at the maximum absorbance

wavelength for the MB, using a Shimadzu UV-1800 UV-VIS spectrophotometer and applying the following equation:

Photodegradation efficiency (%) = $[(A_0 - A_t) / A_0] *100\% = [(C_0 - C_t) / C_0] *100\%$.

(C_t / C_o) can be calculated simply from the above equation. In order to remove any ambiguity, the equation and its explanation have been changed.

1. The size of particles/grains, which are visible in Figure 2, is not provided with the values of average size presented in Table 4.

We did not understand what the respected reviewer exactly means by this. We estimated the average diameter of grains present in each sample by averaging different diameter sizes observed in FESEM image (e.g. D1, D2, D3 for CuO sample, shown in Figure 2a). The estimated values are reported in Table 4.

1. Did you check the effect of the adsorption of MB dye on the surface of materials? How you can explain the lower activity of CuO-10G than pure CuO?

As mentioned in the paper, the lower photodegradation efficiency of the sample CuO-10G was unexpected, and there is no physical explanation for this phenomenon.

1. Can you expand the discussion regarding electron-hole generation and recombination? Did you have any proof which can confirm the presence of this form of species on the surface of materials, e.g. using EPR spectroscopy?

We think this issue has been sufficiently addressed in the section *Study of Photocatalytic Activity*. Unfortunately, we are not able to perform EPR spectroscopy at this time.

Others

1. You have to correct the description of references because, in the present form, they are not prepared according to the Instruction for authors.

The references have been checked and modified, according to the reviewer's comment.

1. You have to improve all abbreviations. They should be in the same form, e.g. CuO and CuO-Gx (or CuO-xG).

We thank the reviewer for these observations. All abbreviations have been checked and used correctly throughout the manuscript.

Review Report Form

Quality of English Language
( ) English very difficult to understand/incomprehensible
( ) Extensive editing of English language and style required
(x) Moderate English changes required
( ) English language and style are fine/minor spell check required
( ) I am not qualified to assess the quality of English in this paper

	Yes	Can be improved	Must be improved	Not applicable
Does the introduction provide sufficient background and include all relevant references?	(x)	( )	( )	( )
Is the research design appropriate?	( )	( )	(x)	( )
Are the methods adequately described?	( )	( )	(x)	( )
Are the results clearly presented?	( )	( )	(x)	( )
Are the conclusions supported by the results?	( )	( )	(x)	( )

Comments and Suggestions for Authors

Dear Authors,

the scientific area of your manuscript entitled *Synthesis, structural, morphological, optical, and photocatalytic characterization of CuO-graphene nanocomposites* addresses the experimental characterization of carbon-based copper oxide composites is according to the papers published in *Coatings* journal and this work can be interesting to the readers.

However, your manuscript can be published in *Coatings* journal after major revision.

You have to improve the presentation of scientific results, i.e., data results are not properly presented, analyzed, and furthermore, the main conclusions are not supported by the data.

Please, the responses to the questions and comments should be added to the text of your corrected manuscript.

Materials and Methods

1. In section 2.4. Photocatalytic Activity of the Samples, you have to add to the description information about reactor parameters, temperature for each experiment condition (dark adsorption, and photocatalysis), solution volumes that were taken from the suspensions, and analyzed using a UV-Visible spectrophotometer.
2. How did you estimate that the error for the average crystallite size (calculated from Debye-Scherrer formula) and average grain diameter (from STEM images) is always 0.01 nm?
3. You have to add more details about the preparation of samples to the measurements using XRD, FESEM, FTIR, and UV-Vis spectroscopy (e.g. resolution, the form of sample, e.g. powder, KBr+sample, or colloid solution) and the procedure of photocatalytic tests (the volume of solution which was collected to the analysis using UV-vis spectroscopy, the separation between catalyst and reaction mixture).

Results

1. How you can explain the differences in the average crystallite size determined by X-Ray diffraction patterns and the average grain diameter estimated using SEM images?
2. The values of Cu, O, and C contents suggest the differences in the atomic ratio between Cu and O species, e.g. according to the data in Table 5, CuO is represented by CuO_2 (Cu:O = 36.70:63.30), CuO-2.5G by CuO (Cu:O = 22.20:27:58), CuO-5G by CuO4 (Cu:O = 4.78:19.02), and GCuO-10G by

CuO7 (Cu:O = 4.28:33.15). How you can explain the differences in the presented results?

3. The results of EDS analysis (high atomic ratio between oxygen and copper species) and FTIR spectra (the presence of the bands, which can be due to the stretching vibration of C-O, and C=O bonds) suggest that the obtained composites can be a mixture of graphene and graphene oxide with copper oxides.
4. How can you explain the presence of bands in the EDX spectra at around 1.5-2.0 eV?
5. How can you explain the high absorption in the range of 800-1000 nm?
6. The quality of Figure 9 must be improved – the colors of the chemical formulas are not visible. You don't have to use the colors like light green, light blue, orange, rose, etc.
7. Did you perform the recycling test for all catalysts?
8. Did you check the possibility of any changes in the morphology of studied materials during the catalytic tests?
9. How did you estimate the presence of selected products of MB photocatalytic degradation which are shown in Figure 9?
10. The size of particles/grains, which are visible in Figure 2, is not provided with the values of average size presented in Table 4.
11. Did you check the effect of the adsorption of MB dye on the surface of materials? How you can explain the lower activity of CuO-10G than pure CuO?
12. Can you expand the discussion regarding electron-hole generation and recombination? Did you have any proof which can confirm the presence of this form of species on the surface of materials, e.g. using EPR spectroscopy?

Others

1. You have to correct the description of references because, in the present form, they are not prepared according to the Instruction for authors.
2. You have to improve all abbreviations. They should be in the same form, e.g. CuO and CuO-Gx (or CuO-xG).

Submission Date 19 October 2021

Date of this review 25 Oct 2021 16:46:14

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Journal: Coatings (https://www.mdpi.com/journal/coatings) (ISSN 2079-6412)
Manuscript ID: coatings-1448119
Type: Article

Title: Synthesis, structural, morphological, optical and photocatalytic characterization of CuO-graphene nanocomposites (https://www.mdpi.com/2079-6412/11/12/1452)

Authors: Indah Raya , AWAIS AHMAD * , Afshin Davarpanah , Marischa Elveny , Mahyuddin K.M. Nasution * , Rafael Luque * , Mabkhoot A Alsaiari * , Mohammed Jalalah

Section: Corrosion, Wear and Erosion (https://www.mdpi.com/journal/coatings/sections/Corrosion_Wear_Erosion_Protection)

Special Issue: Strengthening, Corrosion and Protection of High Temperature Structural Materials (https://www.mdpi.com/journal/coatings/special_issues/high_temp_struct_mater)

Abstract: In this work, a simple hydrothermal process is utilized to prepare a sample of pure copper oxide nanocomposite (CO) as well as three samples of copper oxide-graphene nanocomposites named GCO 2.5, GCO 5, and GCO 10, which contain 2.5, 5, and 10 mg of graphene, respectively. Using XRD analysis, the structures of the synthesized samples were examined. To study the morphological properties of the grown samples, field emission scanning electron microscopy (FESEM) was car-ried out. Energy-dispersive X-ray spectroscopy (EDX) to identify the elemental composition of the samples. Fourier-transform infrared spectroscopy (FTIR) was used to assess the type of bonds present in the structure of the nanocomposites. The optical and photocatalytic properties of the nanocomposites were studied using UV-Vis spectroscopy. The role of graphene in various prop-erties of the synthesized samples was evaluated.

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Authors' Responses to Reviewer's Comments (Reviewer 3)

Author's Notes

1. In the chapter 2.4. Photocatalytic Activity of the Samples, you have to add to the text of manuscript information about reactor parameters, temperature for each experiment condition (dark adsorption, and photocatalysis), solution volumes that were taken from the suspensions, and analyzed using a UV-Visible spectrophotometer. You have to estimate the error for the average crystallite size (calculated from Debye-Scherrer formula) and average grain diameter (from STEM images).

We are thankful to you for positive suggestions to our manuscript.

Photocatalytic experiments were carried out at room temperature (24 °C) by photodegrading MB dye with a 100 W household light bulb. To do so, 20 mg of each synthesized sample was added separately into 50 mL of 100 ppm MB solution. Before illumination, the suspensions were

continuously stirred at a dark place at room temperature for 30 min to reach an adsorption-desorption equilibrium. Then, the suspensions were exposed to visible light irradiation for 9 hours at room temperature. At regular 1-hour intervals, 5cc (cubic centimeter) samples were taken from the suspensions and analyzed using a Shimadzu UV-1800 UV-VIS spectrophotometer in the wavelength region of 300-1100 nm.

2. You have to add more details about the preparation of samples to the measurements using XRD, FESEM, FTIR, and UV-Vis spectroscopy (e.g. resolution, the form of sample, e.g.: powder, KBr sample, or colloid solution) and the procedure of photocatalytic tests (the volume of solution which was collected to each analysis using UV-vis spectroscopy, the separation between catalyst and the collected reaction mixture).

The comments made by the reviewer are highly accredited and suggestions are addressed as acquired.

To study the structural phases of the synthesized samples, XRD patterns of all synthesized samples were obtained in powdered form at room temperature by a Panalytical PW1730 diffractometer using Cu $K_{\alpha}1$ radiation (λ = 1.540598 Å) with a scanning angle (2θ) varied from 20° to 80° and scanning step width of 0.02. The XRD profiles were analyzed by PANalytical X'Pert HighScore software and compared to standards compiled by the Joint Committee on Powder Diffraction and Standards (JCPDS). Investigation of the surface morphology of the samples and their elemental analysis were performed using a MIRA3-TESCAN FESEM equipped with an EDX system. Suspension of nanomaterials is formed in ethanol and drop casted on glass slides. These samples are sticked on carbon tapes after drying and then subjected to carbon caoting. FTIR spectra of CuO and all synthesized nanocomposites inserted in KBr powder system are investigated in the frequency range of 400–4000 cm^{-1} using a BFRL Rayleigh FTIR WQF-510 spectrometer. The UV-Visible absorption spectra were also recorded at room temperature using a Shimadzu UV-1800 UV-VIS spectrophotometer equipped with an integrating sphere, in which white BaSO4 was served as the reference in the wavelength region of 200-1000 nm.

3. How you can explain the high content of oxygen species in all materials estimated using EDX analysis? The values of Cu, and O contents suggest that the source of oxygen species are not only copper oxide (CuO). The high content of oxygen can be explained by the formation of graphite oxide. The graphite oxide disperses in basic solutions or can be dispersed by sonication in polar solvents to yield monomolecular sheets, known as graphene oxide by analogy to graphene, the single-layer form of graphite. The results of EDX analysis (high atomic ratio between oxygen and copper species) and FTIR spectra (the presence of the bands, which can be due to the stretching vibration of C-O, and C=O bonds) suggest that the obtained composites can be a mixture of graphene and graphene oxide with copper oxides.

Your comments are highly appreciated. Suggestions are made accordingly.

Yes, we have discussed the high content of oxygen species in all materials estimated using EDX analysis

EDX analysis was used to investigate the chemical composition of the synthesized samples. The obtained EDX spectra are shown in Figure 4. The weight and atomic percentages of the elements found in the samples are also provided in Table 5. The EDX spectrum of pristine CuO (see Figure 4a) confirms the presence of Cu and O elements in this sample. Also, the EDX spectra of CuO-xG nanocomposites (x= 2.5, 5 and 10 mg of graphene) indicate the presence of three main elements including Cu, O and C, further demonstrating the successful formation of the samples. The EDX spectra presents the high content of oxygen in the nanocomposites which could be predicted due to formation of graphene oxide [74] which are further confirmed by the C=O and C-O vibrational peaks in FTIR spectra.

4. You have to explain the high absorption in the range of 800-1000 nm. Did you use any reference material for UV-vis measurements?

Thank you for your input to our manuscript. Suggestions are made.

The UV-Visible absorption spectra were also recorded at room temperature using a Shimadzu UV-1800 UV-VIS spectrophotometer equipped with an integrating sphere, in which white $BaSO_4$ was served as the reference material in the wavelength region of 200-1000 nm.

5. You have to perform the recycling test for all catalysts.
6. **We really acknowledge the reviewer suggestion. Next time we will be aware regarding this too but at present time it is not possible because in Pakistan still labs are closed due to pandemic**

7. You have to characterize the morphology of studied materials after catalytic tests.

We really acknowledge the reviewer suggestion. Next time we will be aware regarding this too but at present time it is not possible because in Pakistan still labs are closed due to pandemic

8. How many particles/grains of each material were used to estimate the values of average size for each sample presented in Table 4 (e.g. 10 or 100 particles/grains)?

We are really thankful to reviewer for their kind suggestion. It was 10

9. Can you try to explain the lower catalytic activity of CuO-10G than pure CuO?

Thank you for your input to our manuscript.

The lower catalytic activity of CuO-10G than CuO nanoparticles can be explained by the particles size. It is reported that activity of any material is related to its size [84, 85] . The lesser is the size of nanoparticles, the lesser will be the rate of electron-hole recombination. The order of average grain diameter of synthesize material is CuO-5G (20.11 nm) < CuO-2.5G (28.92 nm) < CuO (31.95 nm) < CuO-10G (40.85 nm). The particle size of CuO is smaller than the CuO-10G as confirmed by the XRD and SEM so it has higher photocatalytic activity than CuO-10G.

10. Have you any proof which can confirm the creation in the studied materials the phenomenon of the electron-hole generation and recombination?

Thank you for your input to our manuscript. The suggestions are made

The photocatalytic activity itself is confirmation of the basic phenomena that how these synthesized reduces the electron-hole pair recombination and how these are generated in samples. Which is further supported by the XRD and EDX data as discussed below.

The graphene has been reported as the highly efficient absorbent for the MB molecules [85, 86]. The graphene/GO sheet induces the strong pi-pi conjugation with the molecules of MB [86, 87]. There is electron transfer from the CuO to MB molecules when the energy matches or exceeds the band gaps of CuO the electrons are excited from the valence to conduction bands, holes are generated. The photoelectrons are transferred to graphene. The conjugated graphene then transfers the electrons towards the MB molecules absorbed on the surface, causes the decomposition reaction. The concentration of MB molecules on surface of nanocomposites (CuO-xG) is greater than the CuO because of greater absorbing capacity of graphene which leads to the greater photocatalytic activity [33]. In addition to this, the presences of surface hydroxyl (eOH) groups causes trapping of holes to produce radicals which are excellent oxidizing agents for organic pollutants in water [88]. On the basis of EDX and XRD data, it is confirmed that CuO and CuO-xG has high amount of CueOH which can provide sources of OH radicals and facilitates the decomposition of MB.

Review Report Form

Quality of English Language

( ) English very difficult to understand/incomprehensible
( ) Extensive editing of English language and style required
(x) Moderate English changes required
( ) English language and style are fine/minor spell check required
( ) I am not qualified to assess the quality of English in this paper

	Yes	Can be improved	Must be improved	Not applicable
Does the introduction provide sufficient background and include all relevant references?	(x)	( )	( )	( )
Is the research design appropriate?	( )	( )	(x)	( )
Are the methods adequately described?	( )	( )	(x)	( )
Are the results clearly presented?	( )	( )	(x)	( )
Are the conclusions supported by the results?	( )	( )	(x)	( )

Comments and Suggestions for Authors

Dear Authors,
in my opinion, your corrected manuscript entitled *Synthesis, structural, morphological, optical and photocatalytic characterization of CuO-graphene nanocomposites* concerning the characterization of carbon-based copper oxide composites is according to the papers published in *Coatings* journal and this work can be interesting to the readers but it can be published after major revision.

You have to improve the presentation of scientific results because data results are not properly presented, analyzed, and furthermore, the main conclusions

are not supported by the data.

In the chapter 2.4. Photocatalytic Activity of the Samples, you have to add to the text of manuscript information about reactor parameters, temperature for each experiment condition (dark adsorption, and photocatalysis), solution volumes that were taken from the suspensions, and analyzed using a UV-Visible spectrophotometer.

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How you can explain the high content of oxygen species in all materials estimated using EDX analysis? The values of Cu, and O contents suggest that the source of oxygen species are not only copper oxide (CuO). The high content of oxygen can be explained by the formation of graphite oxide. The graphite oxide disperses in basic solutions or can be dispersed by sonication in polar solvents to yield monomolecular sheets, known as graphene oxide by analogy to graphene, the single-layer form of graphite. The results of EDX analysis (high atomic ratio between oxygen and copper species) and FTIR spectra (the presence of the bands, which can be due to the stretching vibration of C-O, and C=O bonds) suggest that the obtained composites can be a mixture of graphene and graphene oxide with copper oxides.

You have to explain the high absorption in the range of 800-1000 nm. Did you use any reference material for UV-vis measurements?

You have to perform the recycling test for all catalysts.

You have to characterize the morphology of studied materials after catalytic tests.

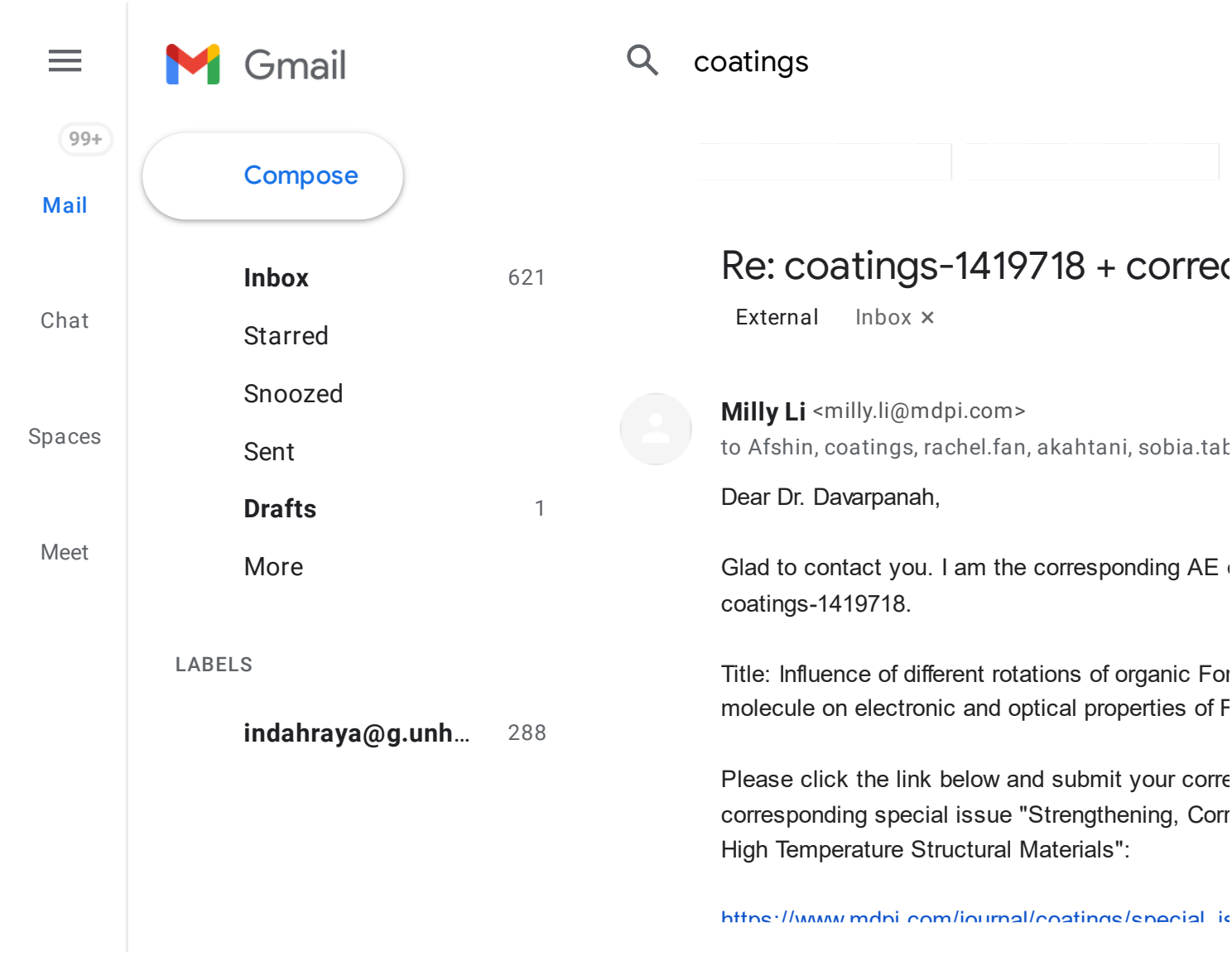
How many particles/grains of each material were used to estimate the values of average size for each sample presented in Table 4 (e.g. 10 or 100 particles/grains)?

Can you try to explain the lower catalytic activity of CuO-10G than pure CuO?

Have you any proof which can confirm the creation in the studied materials the phenomenon of the electron-hole generation and recombination?

Submission Date 19 October 2021

Date of this review 11 Nov 2021 18:03:39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