

Thank you for your submission to Applied Surface Science

From: Applied Surface Science (eesserver@eesmail.elsevier.com)

To: uyi1973@yahoo.com; uyi_sulaeman@yahoo.com

Date: Sunday, May 21, 2017, 10:07 AM GMT+7

Dear Dr. Sulaeman,

Thank you for sending your manuscript Native defects in silver orthophosphate and their effects on photocatalytic activity under visible light Irradiation for consideration to Applied Surface Science. Please accept this message as confirmation of your submission.

When should I expect to receive the Editor's decision?

We publicly share the average editorial times for Applied Surface Science to give you an indication of when you can expect to receive the Editor's decision. These can viewed here: http://journalinsights.elsevier.com/journals/0169-4332/review_speed

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2. Click on [Author Login]. This will take you to the Author Main Menu

3. Click on [Submissions Being Processed]

Many thanks again for your interest in Applied Surface Science.

Kind regards,

Professor Henrik Rudolph

If you require further assistance, you are welcome to contact our Researcher Support team 24/7 by live chat and email or 24/5 by phone: <http://support.elsevier.com>

Your Submission

From: James Whitten (eesserver@eesmail.elsevier.com)

To: uyi1973@yahoo.com; uyi_sulaeman@yahoo.com

Date: Tuesday, July 11, 2017, 07:07 AM GMT+7

Ms. Ref. No.: APSUSC-D-17-05216

Title: Native defects in silver orthophosphate and their effects on photocatalytic activity under visible light Irradiation
Applied Surface Science

Dear Dr. Sulaeman:

Two reviewers have now commented on your paper. You will see that they are advising that you revise your manuscript. If you are prepared to undertake the required work, I would be pleased to consider a revised manuscript. As you will see, some of the reviewer comments may require significant work. It is also possible (especially in the case of reviewer #2), that it might not be possible to perform those experiments. Please do your best to address their concerns in both the cover letter and manuscript and justify why the requested experiments may not be necessary (if this is the case).

If you decide to revise the work, please submit a list of changes or a rebuttal against each point which is being raised when you submit the revised manuscript. Note that in general the best way to address reviewer concerns is to do so within the manuscript and not simply in the cover letter to me.

Please also clearly mark your changes, deletions and additions in the revised manuscript (e.g. by color or underlining).

To submit a revision, please go to <https://ees.elsevier.com/apsusc/> and login as an Author.

Your username is: uyi1973@yahoo.com

If you need to retrieve password details, please go to:

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On your Main Menu page is a folder entitled "Submissions Needing Revision". You will find your submission record there.

Please note that this journal offers a new, free service called AudioSlides: brief, webcast-style presentations that are shown next to published articles on ScienceDirect (see also <http://www.elsevier.com/audioslides>). If your paper is accepted for publication, you will automatically receive an invitation to create an AudioSlides presentation.

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The revised version of your submission is due by Sep 09, 2017.

Yours sincerely,

James E. Whitten, Ph.D.
Editor
Applied Surface Science

Reviewers' comments:

Reviewer #1: This paper deals with the synthesis of Ag₃PO₄ photocatalysts with native defects and their effects on photocatalytic activity under visible light Irradiation. It was found that the activity of Ag₃PO₄ with native defects for Rhodamine B degradation dramatically increased by 5.8 times higher compared to that of the pristine Ag₃PO₄, and the defect might facilitate the enhanced separation of photogenerated electron-hole which enhanced the photocatalytic activity. This paper can be considered to be accepted after addressing the following issues.

(1) In the Introduction Part: Two strategies such as particular morphology and heterostructure materials are carefully introduced. However, the present heterostructure materials are mainly focused on the well-known semiconductor coupling. In fact, the surface modification of Ag₃PO₄ by various electron or hole cocatalysts is also an effective method to improve the photocatalytic performance via promoting the rapid transfer and separation of photogenerated charges. In this case, for a better understood about the Ag₃PO₄ photocatalysts, the surface modification of Ag₃PO₄ should be briefly introduced, such as Phys. Chem, Chem. Phys., 2017, 19, 10309; ACS Catal, 2013, 3, 363; Appl. Catal. B, 2014, 160-161, 658; J. Photochem Photobiol A, 2017, 340, 70.

(2) What is the main native defect in the present Ag₃PO₄ photocatalysts? The author should give some discussion.

(3) During the light irradiation of Ag₃PO₄ photocatalysts, the metallic Ag can be produced. The metallic Ag may has an important effect on the final photocatalytic performance. The author should give some discussion in this paper.

Reviewer #2:

1. The activity of the sythesised catalyst could also be tested with a colorless pollutant in order to avoid self-synthesized degradation

2. In order to have a better understanding of the mechanism involved in these cases, the photoreactions could be carried out in the presence of different electron and hole quenchers

An Itemized List of the Changes in the Revised Manuscript

Journal : Applied Surface Science
Ms. Ref. No. : APSUSC-D-17-05216
Title : Native defects in silver orthophosphate and their effects on photocatalytic activity under visible light irradiation
Authors : Uyi Sulaeman, Dadan Hermawan, Roy Andreas, Ahmad Zuhairi Abdullah, Shu Yin.

The authors greatly appreciate the reviewer's favorable comments. Here are the replies to each point. The response is in brown font, and the changes in the manuscript are in blue font. Please take some time to review this revised manuscript again, thank you very much!

Reviewers' comments:

Reviewer #1: This paper deals with the synthesis of Ag_3PO_4 photocatalysts with native defects and their effects on photocatalytic activity under visible light Irradiation. It was found that the activity of Ag_3PO_4 with native defects for Rhodamine B degradation dramatically increased by 5.8 times higher compared to that of the pristine Ag_3PO_4 , and the defect might facilitate the enhanced separation of photogenerated electron-hole which enhanced the photocatalytic activity. This paper can be considered to be accepted after addressing the following issues.

(1) In the Introduction Part: Two strategies such as particular morphology and heterostructure materials are carefully introduced. However, the present heterostructure materials are mainly focused on the well-known semiconductor coupling. In fact, the surface modification of Ag_3PO_4 by various electron or hole cocatalysts is also an effective method to improve the photocatalytic performance via promoting the rapid transfer and separation of photogenerated charges. In this case, for a better understood about the Ag_3PO_4 photocatalysts, the surface modification of Ag_3PO_4 should be briefly introduced, such as Phys. Chem, Chem. Phys., 2017, 19, 10309; ACS Catal, 2013, 3, 363; Appl. Catal. B, 2014, 160-161, 658; J. Photochem Photobiol A, 2017, 340, 70.

According to the comment, I have added the introduction in paragraph 3.

The third strategy is the surface modification of Ag_3PO_4 by various electron or hole co-catalysts. This method can improve the photocatalytic performance via promoting the rapid transfer and separation of photogenerated charges. The Ag_3PO_4 surface could be modified by Co-Pi as hole cocatalyst that improved the photocatalytic activity [23]. The simultaneous loading of Ag nanoparticles and Fe(III) co-catalyst could improve the photocatalytic activity of Ag_3PO_4 [24]. This Ag nanoparticles can generate the surface plasmon resonance and improves the bandgap visible-light absorption of Ag_3PO_4 , resulting in the generation of more photogenerated charges. The surface of Ag_3PO_4 could also be modified by incorporating graphene to form Ag_3PO_4 /graphene composite [25, 26]. This modification improved the surface area, absorption of organic dyes and separation of photogenerated electron-hole pairs [25]. Incorporating graphene on the Ag_3PO_4 also improved the morphology in which the Ag_3PO_4 dispersed uniformly on the graphene sheets surface [26].

(2) What is the main native defect in the present Ag_3PO_4 photocatalysts? The author should give some discussion.

According to the comment, I have added an explanation in the section of results and discussion at paragraph 8.

The atomic ratio of Ag/P significantly decreases by increasing the ethanol content indicating that the silver vacancy could be easily created (Table 3). Theoretically [27], the main native defects of Ag_3PO_4 could be a silver vacancy (V_{Ag}), oxygen vacancy (V_{O}), silver interstitial (Ag_i), oxygen interstitial (O_i), and interstitial hydrogen (H_i). Among these defects, silver vacancy is easier created in Ag_3PO_4 due to low energy of formation. Previous work, the defects could be designed by non-stoichiometric synthesis of photocatalyst [39]. Here, the different polarity of the mixed solutions might also affect the atomic ratio of Ag_3PO_4 . The decrease of Ag/P atomic ratio, the decrement of the Ag4d FWHM and increment of P2p FWHM of Ag_3PO_4 indicate that the silver vacancies were formed.

(3) During the light irradiation of Ag_3PO_4 photocatalysts, the metallic Ag can be produced. The metallic Ag may has an important effect on the final photocatalytic performance. The author should give some discussion in this paper.

It is very nice suggestion to discuss the formation of metallic Ag during the photoreduction. Usually, the metallic Ag could be produced and decreases the photocatalytic activity. In this paper, we did not investigate the existence of metallic Ag during photoreduction. We have focus on the design of enhanced photocatalytic activity and to explain the reason of increased phototocatalytic activity, especially in the new idea of defect. However, recently, we have also worked on the topic of synergistic effect of defect site and hole co-catalysts to generate both high photocatalytic activity and stability of Ag_3PO_4 for the next paper. It is challenging to produce the catalyst with an excellent photocatalytic activity and high stability.

Reviewer #2:

1. The activity of the synthesized catalyst could also be tested with a colorless pollutant in order to avoid self-synthesized degradation

According to the comment, I have added the data of colorless pollutant (phenol) degradation as shown in. Fig.5, the explanations are in the paragraph 4 of results and discussion.

To ensure the effect of defect sites, the photocatalytic activity was also applied to decompose the phenol compound (the colorless pollutant) under blue light irradiation with the higher power of LED lamp (3 W) (Fig.5). The highest photocatalytic activity of Et-90 was evaluated by phenol decomposition and the result was compared to Et-0. The rate constant of 0.286 min^{-1} could be observed in Et-90 which is significantly higher than that of Et-0 (0.124 min^{-1}) indicating that the photocatalyst is not only active for color pollutant degradation but also for colorless pollutant.

2. In order to have a better understanding of the mechanism involved in these cases, the photoreactions could be carried out in the presence of different electron and hole quenchers

According to the comment, I have added the data of mechanisms as shown in Fig.7 and the explanations are in the last paragraph of results and discussion.

Mechanisms of photocatalytic were investigated by adding the scavengers of radicals and holes to photocatalytic reaction [32]. The benzoquinone (BQ), isopropyl alcohol (IPA) and ammonium oxalate (AO) were added into the reaction solution as scavenger of superoxide ion ($\text{O}_2^{\cdot-}$) radicals, hydroxyl ($\cdot\text{OH}$) radicals and holes, respectively. The effect of these scavengers to photocatalytic reaction can be seen in Fig.7. The BQ addition could

significantly suppress the photocatalytic activity in both Et-0 and Et-50, indicating that their mechanisms involved the superoxide ion radicals. The addition of IPA suppress the photocatalytic activity of Et-50, suggesting that the mechanism involved the hydroxyl radicals which might be generated by the defect sites of Ag_3PO_4 surface, whereas the photocatalytic activity in Et-0 could not be significantly suppressed by adding IPA, indicating that the mechanism might not involve the hydroxyl radical. Generating hydroxyl radical in Et-50 correspond with the XPS analysis that showing the higher ratio of O2/O1 due to higher amount of hydroxyl group adsorption in the surface of Ag_3PO_4 . The PO_4^{3-} on the surface of Ag_3PO_4 might play as the strong bonding ability with H_2O that can promote hydroxyl radical formation. The addition of AO to Et-0 and Et-50 suppressed their photocatalytic activity, indicating both of them involved the holes mechanism. The higher suppression was found in Et-50, suggesting that the holes were highly involved in the mechanism of photocatalytic. The Ag vacancy sites in the band gap of Ag_3PO_4 will act as capture traps for photoexcited holes, which subsequently enhances the separation of electron-hole pairs resulting in improvement in photocatalytic activity. The holes accumulated at the valence band of Ag_3PO_4 might directly oxidize RhB to produce $\text{RhB}^{\bullet+}$ radicals, and the excited electron at the conduction band of Ag_3PO_4 transfer to adsorbed oxygen to form $\cdot\text{O}_2^-$. The mechanism of photocatalytic reaction was proposed in Fig. 8

Your Submission

From: James Whitten (eesserver@eesmail.elsevier.com)

To: uyi1973@yahoo.com; uyi_sulaeman@yahoo.com

Date: Friday, September 22, 2017, 09:57 PM GMT+7

Ms. Ref. No.: APSUSC-D-17-05216R1

Title: Native defects in silver orthophosphate and their effects on photocatalytic activity under visible light irradiation
Applied Surface Science

Dear Dr. Sulaeman:

I am pleased to confirm that your paper "Native defects in silver orthophosphate and their effects on photocatalytic activity under visible light irradiation" has been accepted for publication in Applied Surface Science.

Comments from the Editor and Reviewers can be found below.

When your paper is published on ScienceDirect, you want to make sure it gets the attention it deserves. To help you get your message across, Elsevier has developed a new, free service called AudioSlides: brief, webcast-style presentations that are shown (publicly available) next to your published article. This format gives you the opportunity to explain your research in your own words and attract interest. You will receive an invitation email to create an AudioSlides presentation shortly. For more information and examples, please visit <http://www.elsevier.com/audioslides>.

Your accepted manuscript will now be transferred to our production department and work will begin on creation of the proof. If we need any additional information to create the proof, we will let you know. If not, you will be contacted again in the next few days with a request to approve the proof and to complete a number of online forms that are required for publication.

Thank you for submitting your work to this journal.

With kind regards,

James E. Whitten, Ph.D.
Editor
Applied Surface Science

Comments from the Editors and Reviewers:

Reviewer #1: This paper can be accepted now.

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