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Dear Dr Sutanto,

Re: “Bright Green Fluorescence of Microwave Irradiation-Synthesized Cdots as Sensitive Detector of Iron (III)" by Lewa, Ismira; Sutanto, Heri; Subagio, Agus; Marhaendrajaya, Indras; Sugito, Heri
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Bright Green Fluorescence of Microwave Irradiation-Synthesized Cdots as Sensitive Detector of Iron (III)

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A b s t r a c t

Carbon nanodots (Cdots) are very attractive materials due to their fluorescence and great potential in various fields. One field that still needs to be developed is heavy metal ion detector. In this study, a simple and cost efficient fluorescent detector of Fe^{3+} was built. This material was synthesized by microwave irradiation method using citric acid and urea with purified water as the solvent to get the optimal concentration of urea in order to produce bright green fluorescence. Synthesized Cdots with optimal concentration of urea emit bright green fluorescence under UV light radiation. This bright green fluorescence was then used to detect Fe^{3+}. The fluorescence of this solution was quenched with the addition of Fe^{3+} due to transfer charges and exciton recombination. Optical characterization was carried out using uv-vis spectrophotometer, and fluorescence spectrophotometer. Results showed two absorption peaks on Cdots; at 330 nm and 410 nm. Fluorescence analysis was performed using a beam of 532 nm produced emission at a wavelength of 590 nm. In addition, structural characterization was performed using FTIR and SEM-EDX.

Introduction

Carbon nanodots (Cdots) are zero-dimensional carbon material with a dimension of less than 10 nm. Cdots were first discovered by Xu et al. in 2004 from the purification process of carbon nanotubes [1]. Cdots are spherical [2], non toxic [3] and have high water solubility [4].

Cdots can be used in several application areas such as biomedicine [5], LED [6], energy related applications [7-8], bio imaging [9], and sensors [10]. Cdots are very interesting material with their fluorescence properties [11] and one of the fields that is being developed using this is sensors. Cdots are the right material choice to replace the commonly used probes. Sensor applications typically use quantum dots semiconductor materials such as CdS [12]. However, CdS is toxic and is not easily degraded in the environment. Cdots are an alternative substitute material that are effective because they are not toxic, compatible, and easily soluble in water [12].

Cdots can be synthesized using several methods that are divided into two categories of top down and bottom up. The top down method breaks large molecules into smaller ones. This method includes laser ablation [13], electrochemistry [14] and arc discharge [15]. Meanwhile, the bottom up method, which is also known as the chemical method, is the preparation of a material from small-sized materials. This method includes pyrolysis [16], hydrothermal [17] and microwave [18]. Microwave irradiation is one of the most widely used methods because it can reach certain heat in shortly, efficiently, fast, easily, and inexpensively. Improving the fluorescence properties of this material is an important aspect that should be considered. Various methods can be used such for this purpose, including surface passivation [19]. Passivation process uses organic materials containing an amine group to improve fluorescence. Fluorescence is caused by a surface energy trap that can be modified to get the properties that are suitable for certain applications. Surface modification method is appropriate for sensing applications [20]. Based on research by Sun and coworkers, the surface passivation process of non-luminescent Cdots derived from polyethylene glycol polymers (PEG) shows strong luminescence [20]. By using microwave irradiation, the passivation process can be carried out in one step, hence, microwave irradiation is the right method to synthesize Cdots [21].

Previous studies have proven that Cdots can be used to detect heavy metals with the turn on/turn off fluorescence mechanism. One study by Zhang et al. synthesizes Cdots from melamine and g-C_3N_4 by heating them for 2 hours to detect Pb^{2+} ions [22]. Another study by Kumar et al. synthesizes Cdots using the hydrothermal method for 7 hours to detect Hg^{2+} ions [23]. In this research, Cdots synthesis using microwave irradiation was carried out to obtain a material that has strong luminescence and can be used as a detector of Fe^{3+} heavy metal ions. The precursors used here were citric acid as a source of carbon and urea as passivation agents and distilled water as solvent. Variations of urea concentration were made in order to find the right formula to produce

strong luminescence. Sensitivity tests for Fe^{3+} heavy metal ions were also carried out. Further characterization includes using uv-vis spectrophotometer to observe absorbance spectrum, fluorescent spectrometer to observe emission spectrum of Cdots and FTIR to observe functional groups contained in the synthesized material.

Material and Methods

A synthesis procedure for Cdots can be seen in Figure 1. Cdots were made of citric acid and urea using microwave irradiation method. Citric acid was used as a source of carbon, while urea was used as passivation agent with its amine group content. Purified water was used as solvent in the reaction. Two grams of citric acid were mixed with urea with varied concentration of 1 to 7 grams. The solvent for each solution was 60mL of purified water. Once the solution is homogenized, it was then irradiated using a microwave with power 450W for 30 minutes until a blackish brown crust is formed. The crust was then taken and mashed into powder. Afterwards, the Cdots powder was dissolved in purified water with a concentration 100 ppm. This solution was then characterized and analyzed to determine its properties.

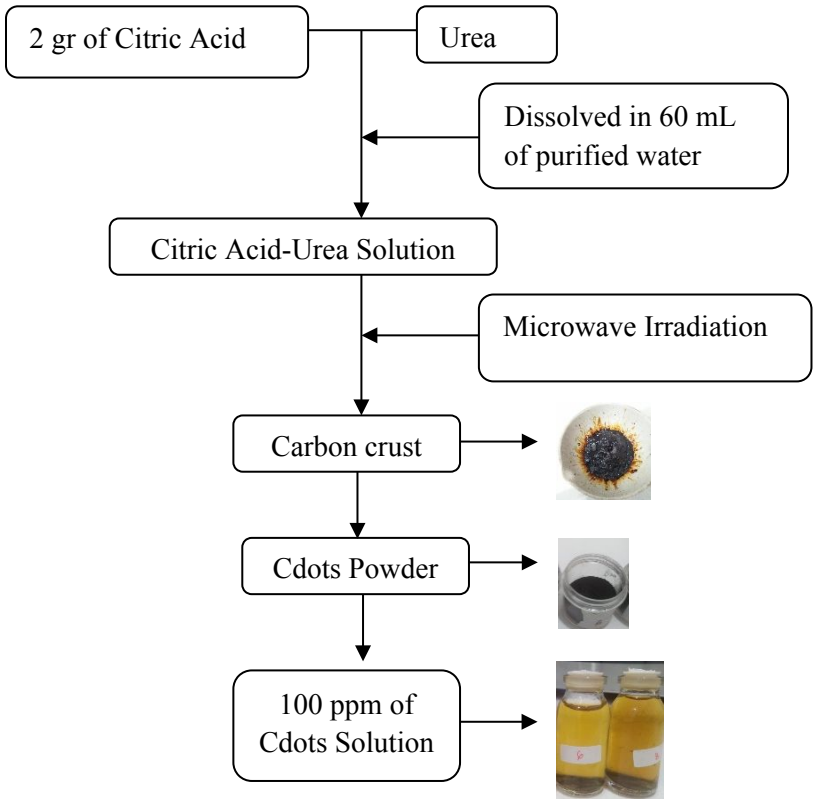


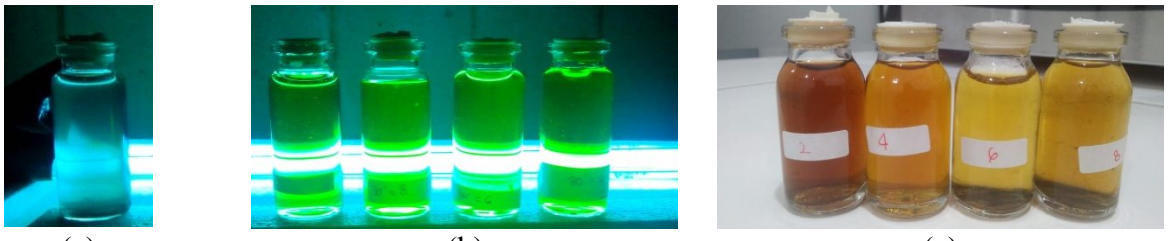
Figure 1. Sample preparation flowchart

Results and Discussion

Cdots Synthesis

The first step is synthesizing Cdots using microwave irradiation. The materials used were citric acid as a source of carbon and urea as a surface passivation agent. Physical processes occurring during synthesis are divides into 4 stages of dehydration, polymerization, carbonization and passivation[20]. Dehydration reaction is the release of water content in the molecules that are reacting. The second step is polymerization. This is where reaction triggers spontaneous nucleation process that is followed by the formation of new longer bonds. The third step is carbonization in which inorganic carbon bonds are derived from citric acid [21]. The final step is surface passivation. This is where coating on the surface of Cdots by compounds of the amine functional groups contained in urea takes place. Cdots surface without coating is not only exposed to contaminants, but also powerless because carbon and oxygen are endogenous to be able to react with organic molecules, hence, it can eliminate opto-electronic properties of Cdots. Therefore, coating is very important because it can maintain the stability of Cdots physical properties and strengthen fluorescence. Surface passivation forms a thin insulating capping layer that can protect Cdots and increase their fluorescence. There are various types of polymers or organic molecules that can be used as passivation agents as long as they do not contain chromophores from visible light to UV and they do not emit the same wavelength, as this may affect the original luminescence of Cdots [24].

Results obtained in this synthesis process are black solids, which were then dissolved in purified water for further observation. Upon observation using UV radiation with a wavelength of 280-300 nm it appeared that the Cdots emit green luminescence, as shown in Figure 2 below.



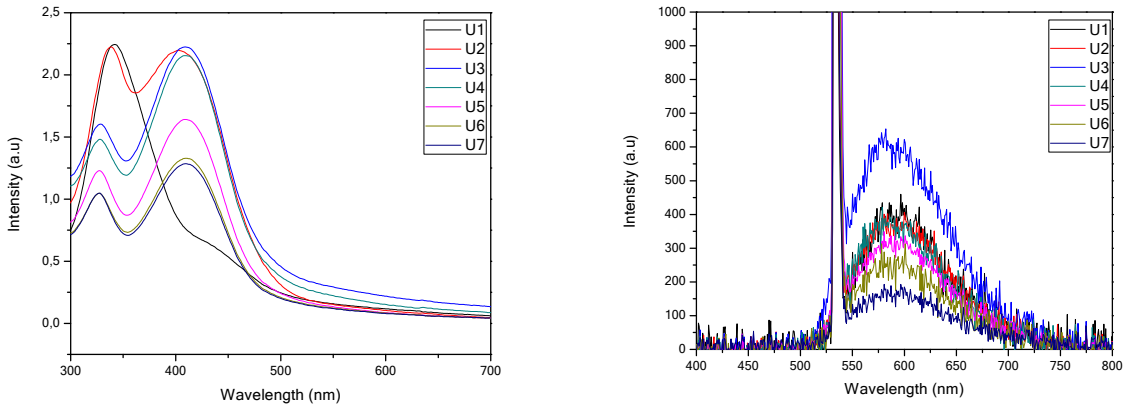
(a) (b) (c)

Figure 2. (a) Activated carbon, and (b) Cdots illuminated by UV radiations, and (c) non-illuminated Cdots

The figure shows that activated carbon (a) has no green luminescence, while Cdots produce green luminescence (b). This indicates change in optical properties when Cdots are changed into a quantum nano materials dimension.

Optical Characterization

Optical characterization of Cdots was performed using UV-visible spectroscopy and fluorescence spectroscopy. Absorption spectra of synthesized Cdots (Fig. 3a) showed two absorption peaks; at 330 nm and 410 nm. These absorption peaks could be associated with $\pi \rightarrow \pi^*$ transition and $n \rightarrow \pi^*$ transition, respectively. These are transitions of carbonyl and oxygen containing compounds. Fig. 3a shows the effect of adding urea that causes a decrease in absorption intensity from the sample. This is possible as the more nitrogen levels contained in urea, the less Cdots absorption is because of lower levels of citric acid and more predominant nitrogen levels. Furthermore, surface functional groups also play an important role in determining the absorption wavelength of Cdots.



(a) (b)

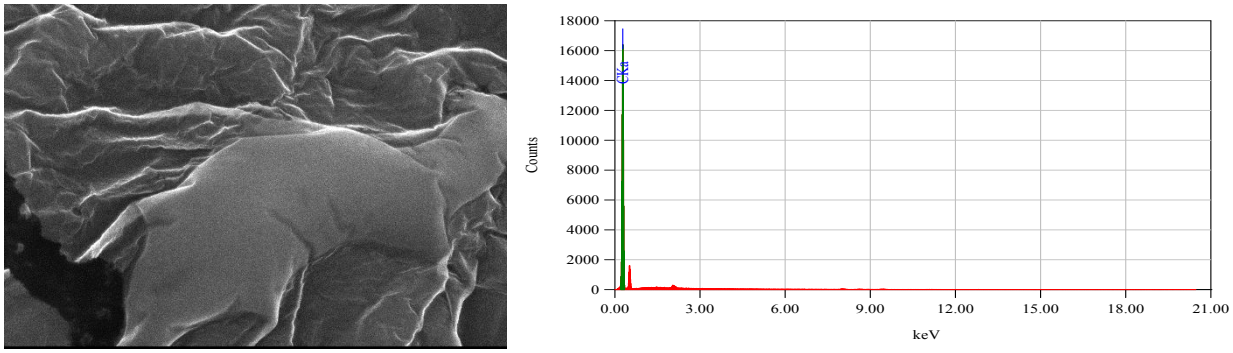
Figure 3. Optical characterization of Cdots (a) absorbance spectra, and (b) fluorescence spectra

The fluorescence spectrum shows a peak at a wavelength of 590 nm. At a concentration of 1 gram, 2 grams and 3 grams of urea, there is an increase in fluorescence intensity. Fluorescence that occurs in Cdots is caused by the presence of surface passivation process by passivation agents. The passivation process results in surface energy trap that enables emission stability and hence, improved fluorescence of Cdots. This is because of the quantum confinement effect of the emission

energy trap on particles, where the ratio between the surface and volume of particles affects passive particles [20]. Fluorescence intensity of Cdots saturated at a variation of 4 grams urea kept on decreasing until the concentration reaches 7 grams of urea. The reason is that at a concentration of 4 grams, the level of urea is more dominant than citric acid as a carbon source. So it is estimated that the number of carbon bonds decreases and this results in reduced fluorescence intensity. The effect of concentration is very important for the fluorescent properties of Cdots. Emissions from strong surface energy levels are caused by changes in concentration [25].

Structural Characterization

In addition to optical characterization, characterization using SEM EDX and FTIR were also carried out. SEM EDX was used to determine the morphological structure of Cdots. Meanwhile, FTIR was used to determine the functional group of synthesized material.



(a) (b)

Figure 4. (a) SEM image from the sample produced at 10,000 magnification, and (b) EDX results showing 100% carbon atoms

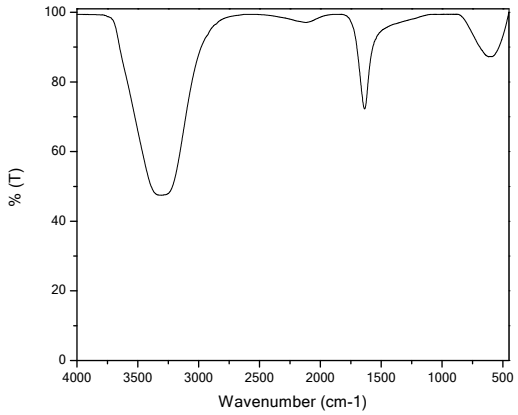
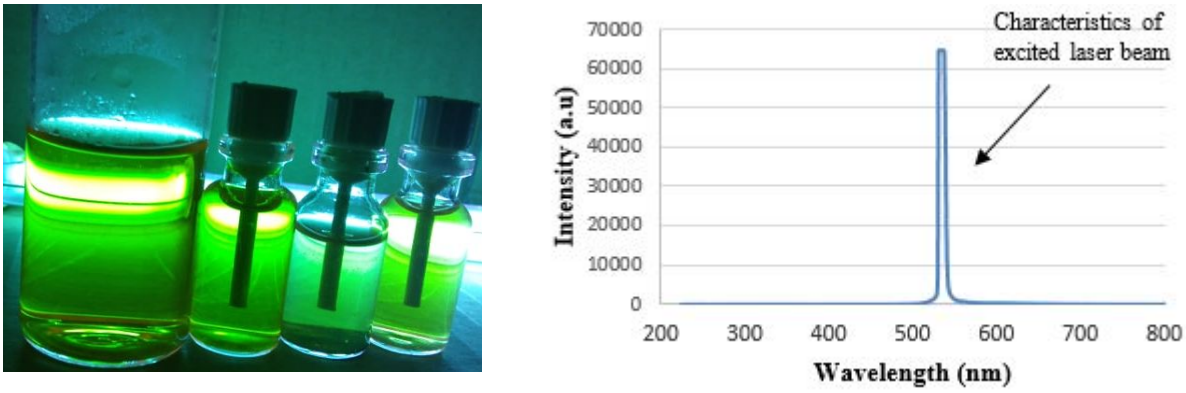


Figure 5. FTIR measurement from citric acid and urea

FTIR analysis can be used to determine the functional groups contained Cdots surface [26]. Liu et al. explain that the functional group of Cdots contain O-H and N-H at wave numbers 3100-3400 cm^{-1} and function group C=O at 1600-1770 cm^{-1} [27]. Results of FTIR measurement show no significant differences with urea variation. Cdots will be more stable and hidrofility increases if there are O-H and N-H groups.[28] Results show OH and NH functional groups at 3307.94 cm^{-1} and C=O groups at 1635.19 cm-1, while the CH_2 groups is found at 1475-1365 cm^{-1} [28].

Fe^{3+} Ions Detection

The Fe^{3+} ions detection test was carried out to find out changes in fluorescence properties of Cdots when some heavy metal solutions were added. Observations show that Cdots fluorescence changes with the addition of heavy metals ions solution, as can be seen in Figure 6.



(a) (b)

Figure 6. Sensitivity test of Cdots against heavy metals (a) fluorescence spectrum of Fe^{3+} Cdots

It can be seen in Figure 6a that when given a standard solution of Cr^{6+} and Pb^{2+} Cdots' fluorescence of cdots remained constant. But it was different when Cdots were given a solution of Fe^{3+} , as the fluorescence began to quench. This can be proven by observing fluorescence spectrum, as shown in Figure 6b. It is easy to see in the spectrumthat there is no peak emission of material, there is only the peak of excited laser beam characteristics. Therefore, we can conclude that Cdots synthesized from citric acids and urea are sensitive to Fe^{3+} ion solutions. Quenched fluorescence of cdots are caused by several functional groups on the their surface. These include hydroxyl and carboxyl groups that are capable to selectively respond to Fe^{3+} ions. The mechanism of thisfluorescence turning offstems from charge transfer and controlled exciton recombination.[29]

Conclusions

A synthesis of Cdots has been successfully carried out using the microwave irradiation method of citric acid and urea via microwave irradiation process. The synthesized Cdots show bright green fluorescence and are sensitive to Fe^{3+} ions. Therefore, it canbe concluded that the addition of urea increases fluorecence. Nonetheless, Cdots saturate at some point and have reduced intensity The proper concentration of urea is 2 : 1 gram to 2 : 3 grams. Cdots fluorescence is quenched upon administration of a standard solution of Fe^{3+} ion. This quenching mechanism is caused by the process of charge transfer charge from the carboxyl and hydroxyl functional groups that reside on the surface of Cdots.

Acknowledgments

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COMMENTS TO THE AUTHOR(S)
This manuscript reports the synthesis of green-emission Cdots by microwave irradiation method, and uses fluorescent detector for Fe3+. They claim the quenching mechanism to be charge transfer and exciton recombination.

Some problems below can be solved to improve the work quality.

1. The absorption peaks at 330 nm and 410 nm in Fig. 3a could be associated with π→π*transition and n→π* transition, respectively. These are transitions of carbonyl and oxygen containing compounds. To our knowledge, the former corresponds C=C bond while the latter attributed to C=O. More information can refer to the paper “Nitrogen doped graphene quantum dots as a fluorescent probe for mercury(II) ions (doi.org/10.1007/s00604-019-3249-4)”, and cite as one reference.
2. “SEM EDX” should be SEM and EDX. Scale bar is more necessary than magnification factor to be specified in SEM. Can the authors give some illustration on the SEM? I cannot get the structural information about the Cdots. Therefore, I consider TEM image necessary to show the structure and size.
3. Space should be given between different words, such as spectrumthat, thatfluorescence, offstems, etc.
4. Corresponding PL spectra of the Cdots in Fig. 6a are quite necessary to show in Fig. 6b.
5. The authors consider the quenching behavior to charge transfer and exciton recombination. Lifetime measurements are necessary to conduct to support this point of view. The study “All–inorganic CsPbBr3 perovskite quantum dots as photoluminescent probe for ultrasensitive Cu2+ detection” can be cited, and give corresponding experiments confirming this mechanism.
6. Why did the author give the title “Bright Green Fluorescence of Microwave Irradiation-Synthesized Cdots as Sensitive Detector of Iron (III)” rather than “Bright Green Fluorescence of Microwave Irradiation-Synthesized Cdots as Sensitive probe of Iron (III)”? Is the latter more suitable for this study?

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Dear Dr Sutanto,

Re: "Bright Green Fluorescence of Microwave Irradiation-Synthesized Cdots as Sensitive Detector of Iron (III)" by Lewa, Ismira; Sutanto, Heri; Subagio, Agus; Marhaendrajaya, Indras; Sugito, Heri
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MRX-115841
Bright Green Fluorescence of Microwave Irradiation Synthesized Cdots as Sensitive Detector of Iron (III)

Referee: 1

COMMENTS TO THE AUTHOR(S)

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AUTHORS GENERAL RESPONSE:

Thank you very much for your valuable comments and suggestions. We have revised the manuscript accordingly.

SPECIFIC RESPONSES:

1. Thank you for the comment and reference of the journal that you provided. We are honored to receive suggestions that are so constructive from you for our manuscripts. We have read the references you gave, revised our manuscripts and cited the references. Please kindly check page 4 line 8.
2. Thank you for your constructive comment. We have revised our manuscript by separating SEM and EDX term. We also gave explanation more about SEM image and added a description of the scale bar in Figure 4a. Please kindly check page 5 Line 14 until 20. From the SEM image we can see that our result has irregularly shaped with a rough

surface. Actually, we are waiting for the TEM results and it takes more time for the results to come out.

3. Thank you for the careful correction you gave. We have revised typographical errors on our manuscript, please kindly check. Hopefully we will be more careful and thorough in writing.
4. We have added a corresponding PL spectra in several conditions with addition of heavy metal ions. Please kindly check at page 6 Figure 6b.
5. Thank you for the constructive comments and excellent references that helped us to answer question about our statement of the mechanism quenching behaviour. We consider this quenching behavior is due to interaction between the carboxyl and hydroxyl groups with heavy metal ions. This statement is in accordance with some of the citation sources that we have studied. The characterization results from FTIR state that our material contains carboxyl and hydroxyl groups which can interact with several solutions of heavy metal ions and it can proven by PL spectrum.Furthermore, we read a several of references including the references that you gave that this interaction phenomenon involved charge transfer and exciton recombination. But we cannot provide lifetime measurement data because of the limited infrastructure we have. In fact, this study aims to provide information about making simple and easy heavy metal ion detectors. We hope you understand our limitations and hope for good results.

6. We agree that recommendation title that you give is more suitable for our manuscript. So we have revised the title of our manuscript. Thankyou very much for the very good suggestion.

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Re: "Bright Green Fluorescence of Microwave Irradiation-Synthesized Cdots as Sensitive Detector of Iron (III)" by Lewa, Ismira; Sutanto, Heri; Subagio, Agus; Marhaendrajaya, Indras; Sugito, Heri
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COMMENTS TO THE AUTHOR(S)
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