Science & Technology Development Journal – Engineering and Technology, 5(3):1577-1585

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Research article

Determination of the efficiency of AgNPs/ZIF-8 coating on different substrates

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ABSTRACT

Depending on different applications or materials that integrate with the substrate as well as the transduction method used in the biosensors, different materials of the substrate are considered. This research focuses on selecting the appropriate substrate in designing biosensors, detecting microorganisms in the environment. In this study, the glass slide (glass), Polyethylen plastic (PE plastic), and the cellulose paper (cellulose acetate (CA) & cellulose nitrate (CN)) are investigated substrates for coating AgNPs/ZIF-8 – material used as an optical signal amplification in further steps, using structural analysis techniques – Scanning Electron Microscopy (SEM) for morphology characterization and the Energy Dispersive Spectrometer (EDS) for optical characterization. A morphology characterization with SEM showed that cellulose paper is a better substrate in comparing with the other substrates (glass, PE), because the sensity of distribution on CN was the highest (85-90%) when it coated with AgNPs/ZIF-8. According to EDS analysis, the ratios of Ag in mixture on CN substrate (15.38±0.39% mass) is slightly higher than CA substrate (13.58±0.39% mass); and about 53 times higher than glass substrate (0.37±0.13% mass) and PE plastic substrate (0.29±0.08%). Therefore, cellulose paper (CA) will be suggested to select for further research as a new material with cost (inexpensive) and effectiveness in coating AgNPs/ZIF-8.

Key words: AgNPs/ZIF8, substrate, biosensor

INTRODUCTION

Over the years, due to the properties of nanoparticles in terms of good biocompatibility, broad structure variety, and notable bioimitative characteristics, nanotechnology has been used in the development of biosensors to improve performance and sensitivity[1]. Various kinds of nanoparticles have been studied in the context of biological detection[1]. Among these nanoparticles, metal nanoparticles (MNPs) such as silver nanoparticles (AgNPs) have been widely used due to their unique physicochemical features, such as the morphological and structural characterization at the nanoscale[2]. As the characteristics of MNPs are closely related to their dispersibility[3], metal-organic frameworks (MOFs) are used to support MNPs[4]. Bagheri et al. have described the use of Ag nanoparticles/Zn-based MOF (AgNPs@Zn-MOF) to enhance the electrochemical activities of MOFs for the selective detection of patulin[5].

Substrates selected for biosensor manufacture must be relatively inert, possess a surface consistency ideal for ligand attachment[6]. There are many different types of substrates, such as glass slide, polymer, paper, etc.[7] Depending on different applications or materials that integrate with the substrate as well as the transduction method used in the biosensors, different materials of the substrate are considered. Silver nanoparticle is obviously one of the excellent candidates for the antibiosis of glass surface, but it is barely absorbed to the surface of the protected glass by a weak force[8]. Sarva.M.P et.al (2016) used cellulose filter paper coated with silver nanoparticles because of it's properties such as inexpensive, abundant, sustainble, and renewable[9]. However, there are no studies comparing coat performance between materials. In this work, AgNPs/ZIF-8 will be synthesized and coated on different materials such as (glass, polyethylene, cellulose paper). Thus, the coating performance of AgNPs/ZIF-8 on these materials is compared.

METHODS

Materials

Zinc nitrate ($Zn(NO_3)_2$, Merck), 2-methylimidazole (2-MeIM - $CH3C_3H_2N_2H$ 99%, Sigma Aldrich), methanol (CH_3OH, Merck), milver nitrate ($AgNO_3$, Merck), modium borohydride ($NaBH_4$, Sigma Aldrich), (3-Aminopropyl) trithoxysilan 98% (APTES - $C_9H_{23}NO_3Si$, A Johnson Matthey company), ethanol (C_2H_5OH, Merck), sulfuric acid 98% (H_2SO_4, Merck), hydrogen peroxide 30% (H_2O_2, Merck), acetic acid 100% (CH_3COOH, Merck) were

Cite this article : Huyen T T B, Phuong L D, Thi P T, Hanh D V B. **Determination of the efficiency of AgNPs/ZIF-8 coating on different substrates** . *Sci. Tech. Dev. J. – Engineering and Technology;* 5(3):1577-1585.

History
• Received: 23-3-2022
• Accepted: 06-9-2022
• Published: 30-9-2022

DOI : 10.32508/stdjet.v5i3.969

used without further purification.

Substrate: Research choose 4 types of substrate include Glass (cover glass made in Germany), PE (Polyethylene) plastic, Cellulose acetate (CA) paper, Cellulose nitrate (CN) paper.

Characterizations: The surface morphology was characterized using Scanning Electron Microscopy (SEM, JEOL JSM-6400) and the Energy Dispersive Spectrometer (EDS, JEOL JSM-IT200).

Experiments:

Figure 1 revealed that know how the experiment of substrate comparison study was conducted.

- First, the ZIF-8 and AgNPs/ZIF-8 were synthesized according to the previous report[4]. In a typical processor, 187.5 ml of 100 mM solution of 2-MeIM in methanol and 125 ml of 100 mM solution of $Zn(NO_3)_2$ in methanol were mixed and stirred for 12h at room temperature. The milky sususpension was centrifuged and washed with methanol for three times and dried at 80 °C overnight. Thereafter, 10 g ZIF-8 was added into 40 mL $AgNO_3$ solution (25 mmol dm^{-3}). Thereafter, the mixture was stirred for 12 h at room temperature. The suspension was successively centrifuged and then dried under vacuum at 80 °C overnight. Afterwards, 0.5 g $NaBH_4$ dissolved in 40 mL water was mixed with the Ag+/ZIF-8 with vigorous stirring for 30 min and the mixture turned dark.
- Substrate preparation: In this study, 4 substrates of glass, PE plastic, cellulose paper (CA, CN), and paper are used to coat AgNPs/ZIF-8.
- Before the experiment the glass slide was treated according to Yaohui Lv, et al. (2008)[8]. The glass slide was immersed in a lightly boiled piranha solution (3:1 v/v 98% H_2SO_4/30% H_2O_2) for 20 min. After being washed with DI water, it was dipped in 1% ethanol solution of APTES for 30 min at room temperature. The pH of this solution was adjusted to 3.5-5.5 by 100% CH_3COOH. Then, washed with ethanol and cured in the oven at 100^oC for at least 2 hr.
- The cellulose papers and PE plastic were rinsed with 95% ethanol before coating[9].
- Coating the AgNPs/ZIF-8 onto the selected substrates.
- Analyze and characterize the substrate modification via SEM and EDS.
- Select the best substrate.

RESULTS

Synthesis of ZIF-8 and AgNPs/ZIF-8

ZIF-8 was synthesized successfully (Figure 2).

Figure 2a showed the milky suspension, which may be to contain ZIF-8 and the residue after being centrifuged (Figure 2b).

The ZIF-8 was in a good crystal morphology with a mean diameter about of 700 nm (Figure 2c), which confirmed the formation of pure ZIF-8. The elements of C and Zn were undoubtedly generated by ZIF-8 (Figure 2d).

Figure 3 showed the characterizations of AgNPs/ZIF-8 composite. The solution containing Ag+/ZIF-8 becomes dark after adding $NaBH_4$ solution, as a result of the formation of AgNPs/ZIF-8 (Figure 3a). As shown in Figure 3b, AgNPs/ZIF-8 with uniform morphology and size were well dispersed. In addition, Figure 3c exhibits the elements of C and Zn are undoubtedly generated by ZIF-8, while the element of Ag comes from AgNPs (9.64 ± 0.32 % mass).

AgNPs/ZIF 8 coating on Glass substrate

The surface morphology of AgNPs/ZIF-8 on immobilized glass substrate was analyzed by SEM, which showed a significant difference between a blank glass (Figure 4a) and glass coated in AgNPs/ZIF 8 (Figure 4b).

Figure 4a & b showed that SEM image of AgNPs/ZIF-8 layer coated on the glass slide. It was obvious that there was no presence of AgNPs/ZIF-8 on the bare glass slide. On the other hand, the EDS data (Figure 4c) showed peaks of Ag came from AgNPs, in which Ag accounted for 0.37±0.13% in mass.

AgNPs/ZIF 8 coating on Polyethylene (PE) plastic substrate

Similar to the glass substrate, the SEM image showed a significant difference between a blank PE plastic (Figure 5a) and PE plastic coated in AgNPs/ZIF-8 (Figure 5b).

Figure 5 a&b showed the amount of AgNPs/ZIF 8 that appeared on the PE plastic. The percentage of Ag on PE plastic was 0.29±0.08% in mass (Figure 5c).

AgNPs/ZIF-8 coating on Cellulose paper substrates

Two kinds of cellulose paper were used in this research, which were CN and CA.

The coated papers images and blank papers are characterized through SEM technique (Figure 6a,b,c; Figure 7a,b).

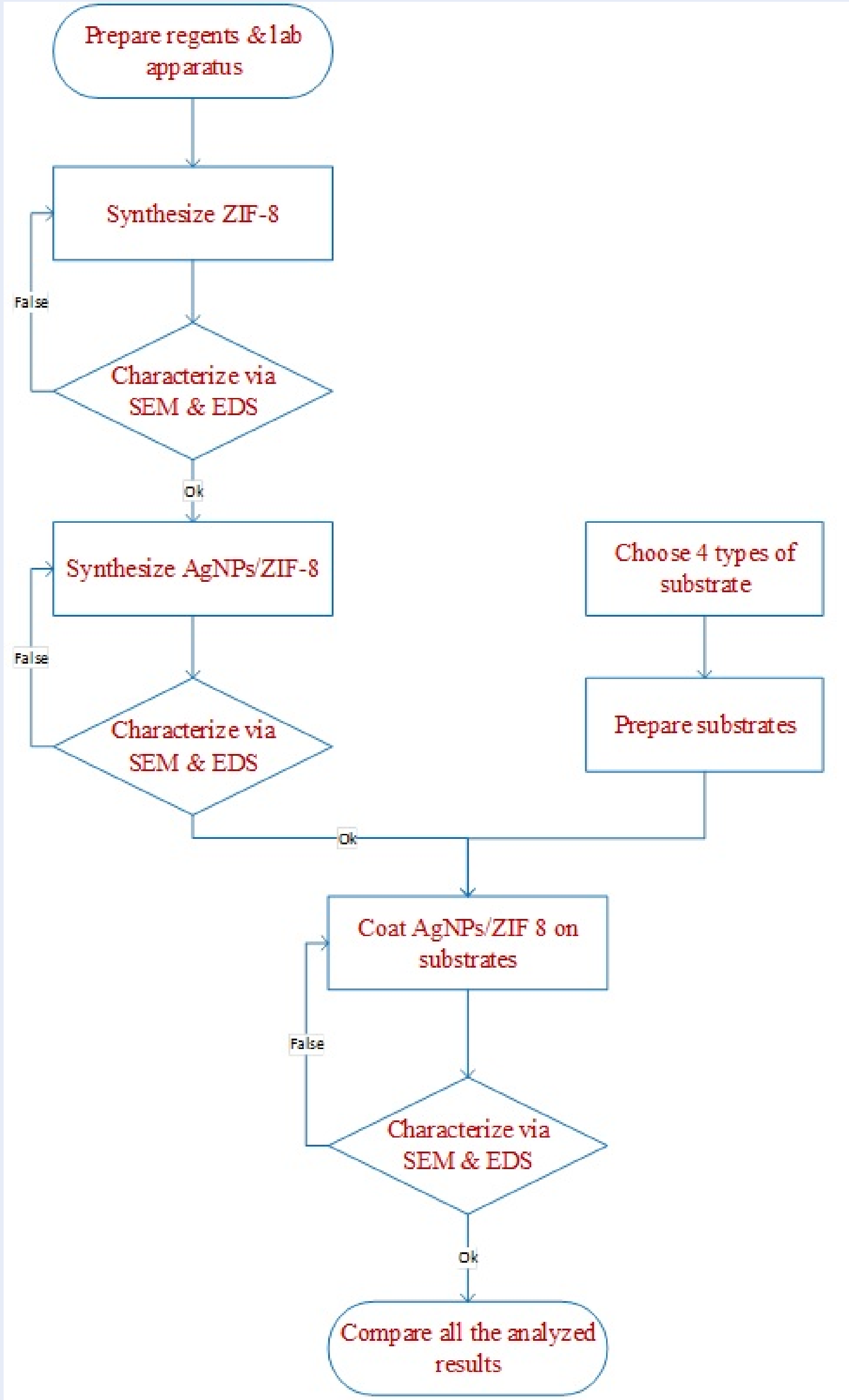

Figure 1: The process of the experiment

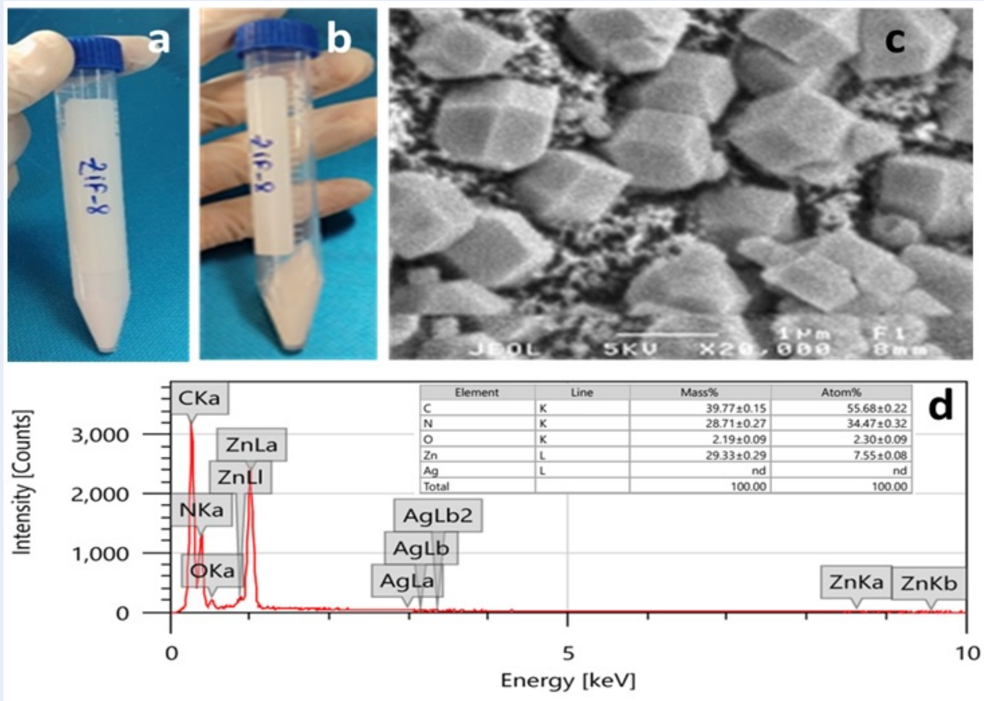

Figure 2: The result of (a)(b) Formation; (c) SEM image; (d) EDS data of ZIF-8

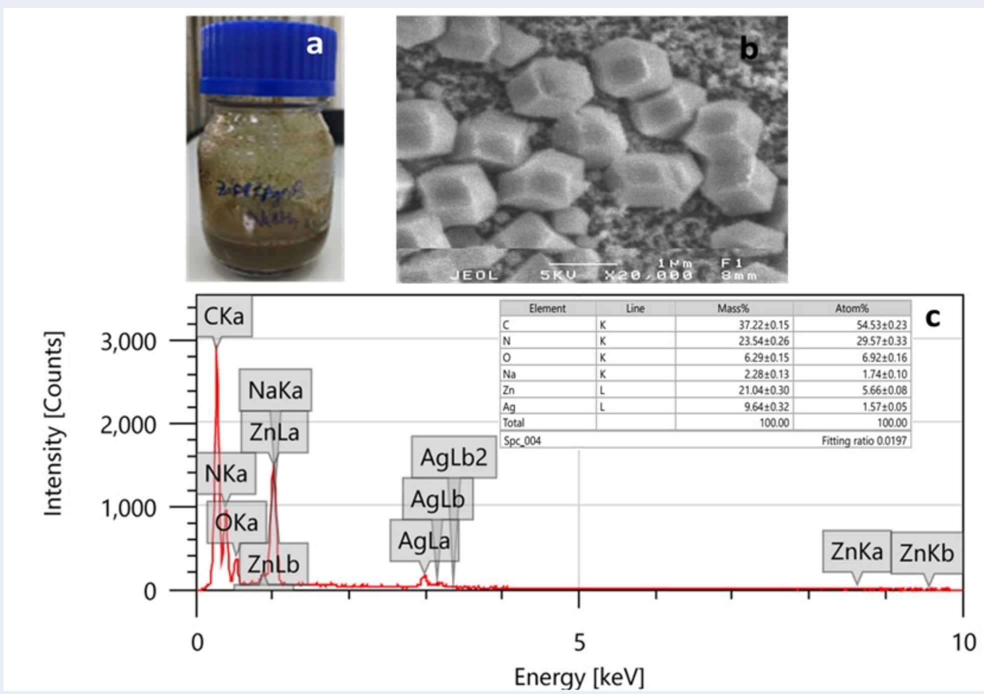

Figure 3: The result of (a) Formation; (b) SEM image; (c) EDS data of AgNPs/ZIF-8

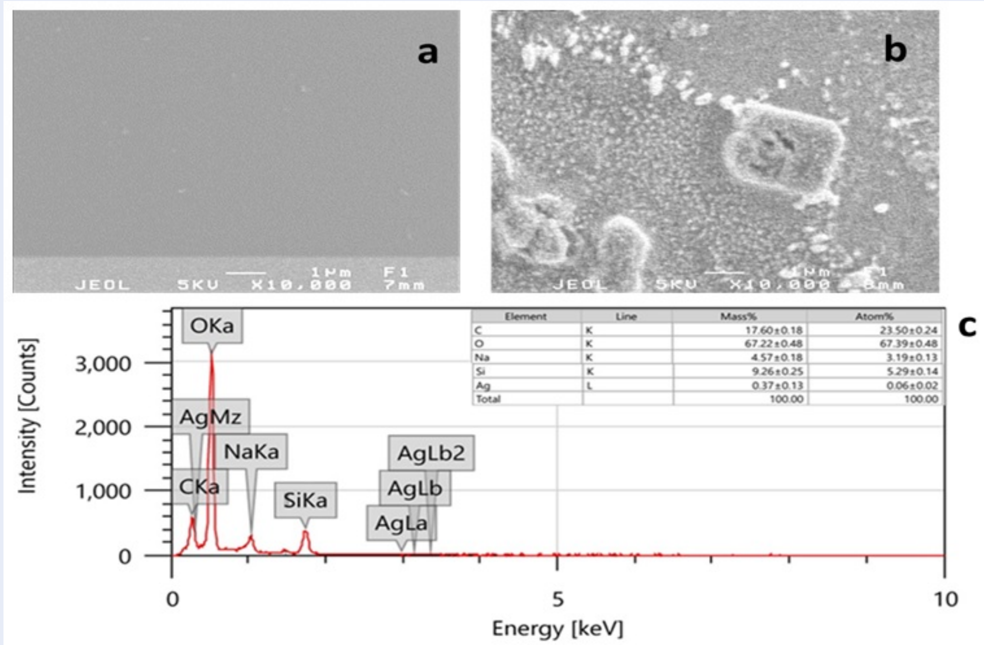

Figure 4: The SEM image glass before (a) and after AgNPs/ZIF-8 coating (b); EDS data of glass after AgNPs/ZIF-8 coating (c)

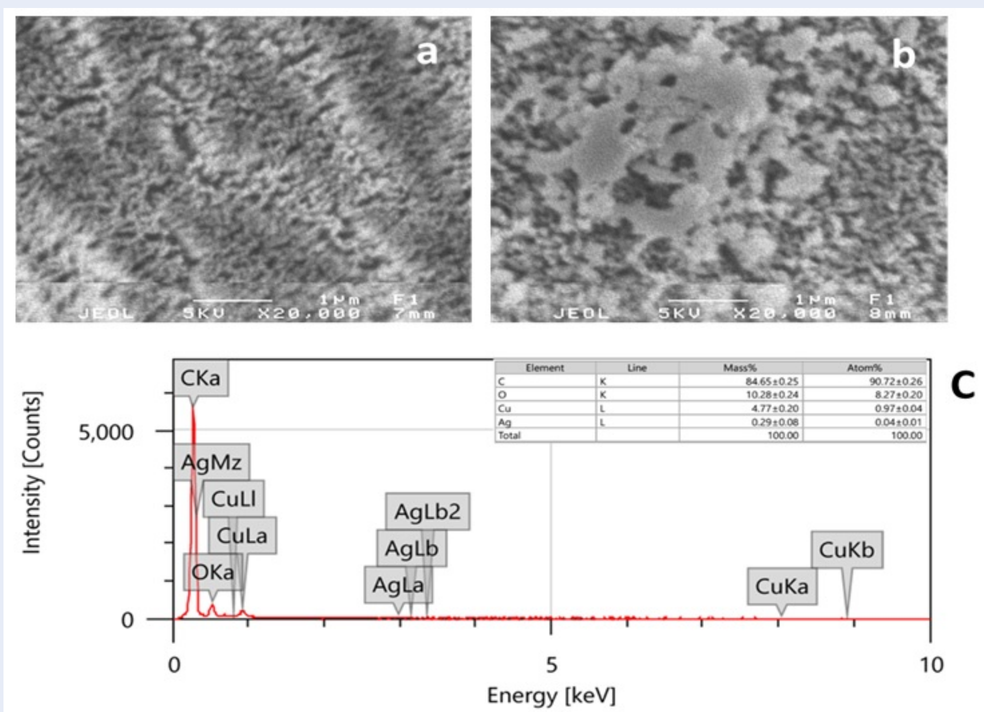

Figure 5: The SEM image of PE plastic before (a) and after AgNPs/ZIF-8 coating (b); EDS data of PE plastic after AgNPs/ZIF-8 coating

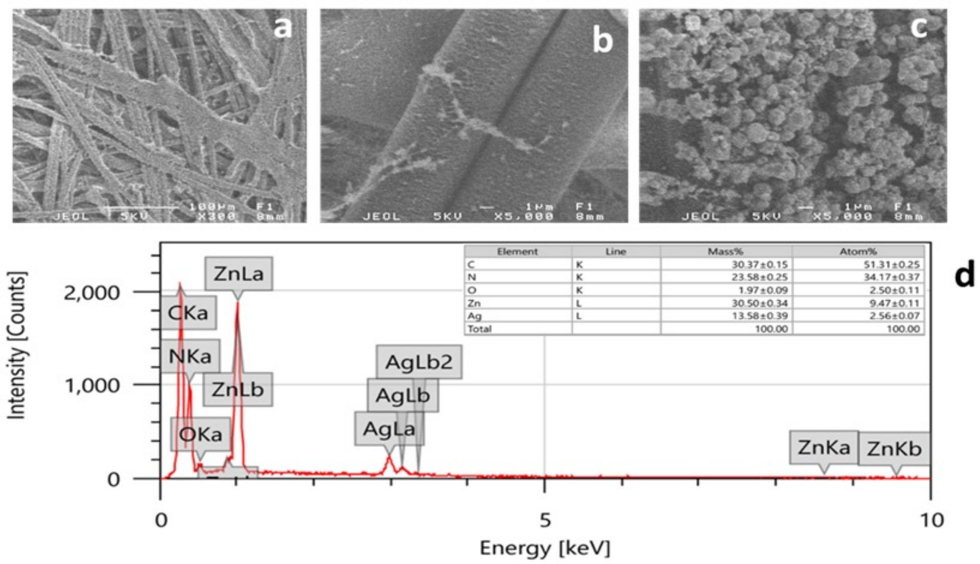

Figure 6: The SEM image of CA before (a)(b) and after AgNPs/ZIF-8 coating (c); EDS data of CA after AgNPs/ZIF-8 coating (d)

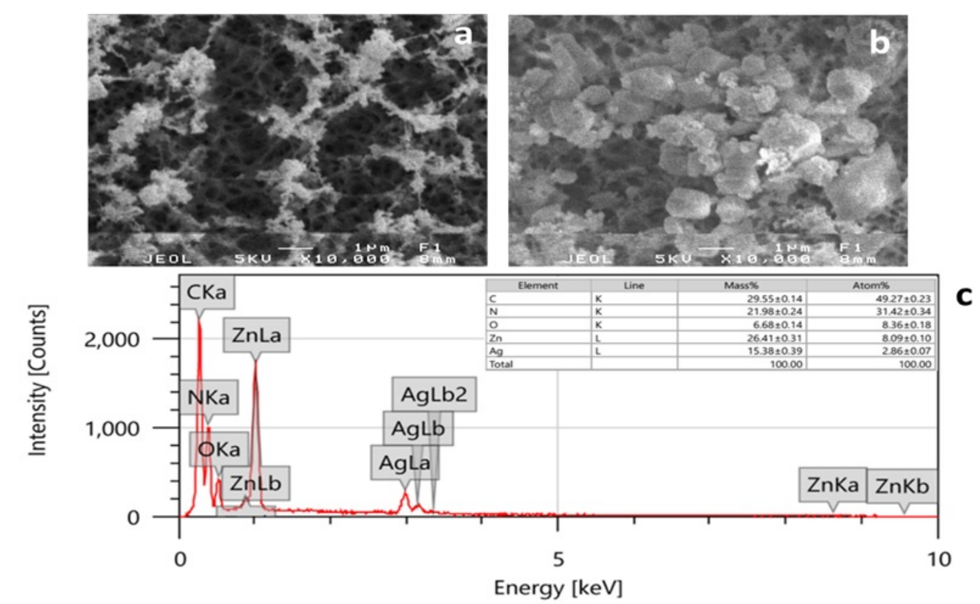

Figure 7: The SEM image of CN before (a) and after AgNPs/ZIF-8 coating (b); EDS data of CN after AgNPs/ZIF-8 coating (c)

In Figure 6a and Figure 7a, it was easy to see that a clear structure of cellulose paper with varies of equal pore size, as the pore size of filter paper is 0.45μm (according to the manufacturer's specifications), in which the cellulose fiber size of CA paper is larger than that of CN paper.

As confirmed by the assessment, the papers have been coated with AgNPs/ZIF-8. Figure 6d and Figure 7c show the peaks of Ag. The mass percentage of Ag on CA and CN paper was 13.58±0.39% and 15.38±0.39%, respectively.

DISCUSSION

The research used SEM and EDS to characterize the morphology of AgNPs/ZIF-8 coating on different substrates. Each substracte was measured about 7 points over total area of 1cm^2 in a zigzag pattern, repeated 3 times. The results showed that the appearance of AgNPs/ZIF-8 at all measurement points, AgNPs/ZIF-8 was evenly coated on the surface of the substractes as well. There ass a significantly different in the efficiency of AgNPs/ZIF-8 coating on substrate types. The amount of AgNPs/ZIF-8 covering Glass substrate and PE substrate was small and uneven on the surface of the substrate (Figure 4b & Figure 5b). Meanwhile, the AgNPs/ZIF-8 has already been well distributed that they got into some typical pores on the surfaces of the cellulose paper (CA, CN substrate) were shown in Figure 6c and Figure 7b.

There were differences in the percentage and density distribution of Ag on substractes, depending on different substrates (Table 1).

The mass percentage of Ag on glass, PE, CA, CN substrate was 0.37±0.13%, 0.29±0.08%, 13.58±0.39%, 15.38±0.39% respectively. For atomic percentages, the % Ag on glass, PE, CA, CN substrate was 0.06±0.02%, 0.04±0.01%, 2.56±0.07%, 2.86±0.07%, respectively.

In terms of both mass and atomic percentage of Ag, the percentage (both mass and atom) of Ag on CN ass the highest. It was higher than glass 41 times (% mass) and 47 times (% atomic); higher than PE 53 times (% mass) and 71 times (% atomic).

The results were similar when considering the average density of distribution of AgNPs/ZIF-8 on the substrate surface at the SEM imaged points. The resulting density of distribution on the substrate surface from high to low was CN, CA, glass and PE, respectively. Sensity of distribution on CN was the highest (85-90%), while on PE the lowest (25-30%).

It means that the coating efficiency of AgNPs/ZIF-8 on glass and PE substrate are very low, while it was high for CA and CN substrates.

Comparing between CA and CN substrate, the percentage of Ag in mixture on CN substrate was slightly higher than CA substrate, but CN substrate was changed its shape after coating (Figure 8), possibly due to the affinity of water for the NO_3^- functional group.

CONCLUSION

In summary, the SEM result and visual observation showed that CA paper is the best substrate in comparation with the other substrate (such as glass, PE, CN). Indeed, the AgNPs/ZIF-8 film was uniformed and fully covered on CA paper. According to EDS analysis, the mass percentate of Ag in mixture on CN substrate (15.38 ± 0.39% mass) was slightly higher than on CA substrate (13.58±0.39% mass); and it was about 53 times higher than that on glass substrate (0.37±0.13% mass) and PE plastic substrate (0.29±0.08%). But CN substrate was changed its shape after coating.

In general, both the morphology characterization and optical properties showed that cellulose paper is a better substrate than suitable for designing a biosensor. Therefore, cellulose paper (CA) will be suggested to select for further research as a new material with cost (inexpensive) and effectiveness in coating AgNPs/ZIF-8.

ACKNOWLEDGMENTS

This research is supported by Environmental analysis Lab, Faculty of Environment and Natural Resources - Ho Chi Minh City University of Technology, and funded by The Murata Science Foundation.

The research received many contributions from Nguyen Huu Viet in the process of completing the research.

LIST OF ABBREVIATIONS

PE: Polyethylen

CA: cellulose acetate

CN: cellulose nitrate

SEM: Scanning Electron Microscopy

EDS: Energy Dispersive Spectrometer

MNPs: metal nanoparticles

AgNPs: silver nanoparticles

MOFs: metal-organic frameworks

ZIF-8: Zeolite imidazolate frameworks-8

JEOL JSM-6400: Model of Scanning Electron Microscopy

JEOL JSM-IT200: Model of Energy Dispersive Spectrometer

DI water: Deionized Water

Table 1: The percentage and the density of distribution of Ag on substractes

Substrate	Mass	Atomic	Density of distribution
Glass	0.37±0.13%	0.06±0.02%	35-40%
PE	0.29±0.08%	0.04±0.01%	25-30%
CA	13.58±0.39%	2.56±0.07%	80-85%
CN	15.38±0.39%	2.86±0.07%	85-90%

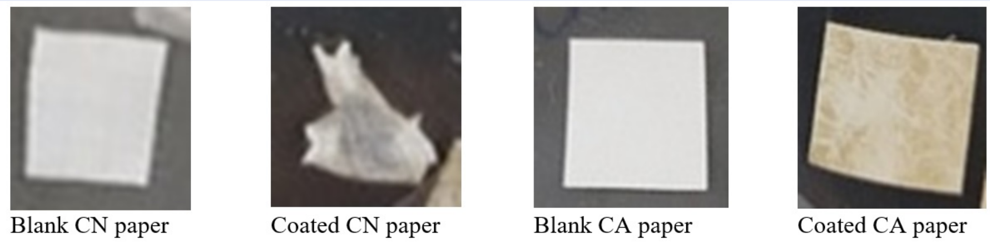

Figure 8: The visual appearance of cellulose papers before and after AgNPs/ZIF-8 coating

CONFLICT OF INTEREST

There is no conflict of interest.

AUTHORS CONTRIBUTION

Huyen TTB contributes in design experiment ideas, conduct experiments, synthesize and analyze data, writing of the manuscript.

Phuong LD contributes in conduct experiments, synthesize and analyze data.

Thi NT contributes in design experiment ideas, references.

Hanh DVB contributes in design experiment ideas, supervision, data checking, proofreading and editing of the manuscript.

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Tạp chí Phát triển Khoa học và Công nghệ – Engineering and Technology, 5(3):1-1
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Bài nghiên cứu

Xác định hiệu quả của việc phủ AgNPs/ZIF-8 trên các đế khác nhau

Trịnh Thị Bích Huyền[1,2,*], Lại Duy Phương[1,2], Phạm Tấn Thi[2,3,4], Đặng Vũ Bích Hạnh[1,2,*]

TÓM TẮT
Tùy thuộc ứng dụng hoặc vật liệu khác nhau của đế cũng như phương pháp chuyển đổi tín hiệu được sử dụng trong cảm biến sinh học mà các vật liệu khác nhau của đế được xem xét. Nghiên cứu này tập trung vào việc lựa chọn đế thích hợp trong việc thiết kế cảm biến sinh học, được sử dụng để phát hiện vi sinh vật trong môi trường. Trong nghiên cứu này, thủy tinh, nhựa PE và giấy cellulose (cellulose acetate và cellulose nitrate), được lựa chọn cho làm đế sẽ được phủ AgNPs/ZIF-8 (vật liệu được sử dụng làm bộ khuếch đại tín hiệu quang học trong các bước tiếp theo). Nghiên cứu sử dụng các kỹ thuật phân tích cấu trúc bao gồm kính hiển vi điện tử quét (SEM) để xác định đặc điểm hình thái và máy quang phổ phân tán năng lượng (EDS) để xác định đặc điểm quang học. Thông qua đặc điểm hình thái học bằng SEM thấy rằng giấy cellulose là vật liệu tốt hơn so với các vật liệu khác (thủy tinh, nhựa PE), bởi khi được phủ AgNPs/ZIF-8, mật độ phân bố trên CN là cao nhất (85-90%). Theo phân tích EDS, tỷ lệ Ag trong hỗn hợp trên đế cellulose nitrate (15,38 ± 0,39% về khối lượng) cao hơn một chút so với Cellulose acetate (13,58 ± 0,39% về khối lượng); và cao hơn khoảng 53 lần so với đế thủy tinh (0,37 ± 0,13% khối lượng) và đế nhựa PE (0,29 ± 0,08%). Nhìn chung, giấy cellulose là đế tốt hơn phù hợp để thiết kế bộ cảm biến sinh học. Do đó, giấy cellulose (CA) sẽ được chọn làm vật liệu dùng làm đế của cảm biến sinh học trong nghiên cứu kế tiếp bởi giá thành (không quá đắt) và hiệu quả phủ AgNPs/ZIF-8.
Từ khoá: AgNPs/ZIF8, đế, cảm biến sinh học

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Trích dẫn bài báo này: Huyền T T B, Phương L D, Thi P T, Hạnh D V B. **Xác định hiệu quả của việc phủ AgNPs/ZIF-8 trên các đế khác nhau.** *Sci. Tech. Dev. J. - Eng. Tech.;* 5(3):1-1.

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