KKU Engineering Journal

https://www.tci-thaijo.org/index.php/easr/index

Published by the Faculty of Engineering, Khon Kaen University, Thailand

Photocatalytic and antibacterial activities of ZnO powders prepared via a sol-gel method

Weerachai Sangchay*[1)] and Kornkanok Ubolchollakhat[2)]

[1)]Faculty of Industrial Technology, Songkhla Rajabhat University, Songkhla 90000, Thailand
[2)]Faculty of Science, Thaksin University, Pattalung 93110, Thailand

Received June 2015
Accepted August 2015

Abstract

We report on the synthesis of ZnO powders via a sol-gel method. The prepared powders were calcined at temperatures of 300, 500 and 700 °C for 1 h with a heating rate of 10 °C/min. The microstructures of the fabricated powders were characterized using SEM and XRD techniques. The results showed that all samples exhibited agglomeration of spherically shaped particles and revealed only a Wurtzite phase. The photocatalytic activities of the powders were tested via the degradation of a methylene blue (MB) solution under UV irradiation. Finally, antibacterial activity was evaluated by observation of the inactivation of *E. coli*. It was seen that samples calcined at higher temperatures gave better photocatalytic and antibacterial activities. At the highest calcination temperature investigated in this experiment (T700 condition), the powders showed photocatalytic and antibacterial activities of 80.06 and 99.00%, respectively.

Keywords: Photocatalytic activity, Antibacterial activity, ZnO powders, *E. coil*, Sol-gel method

1. Introduction

Zinc oxide (ZnO) is a II-VI group semiconductor materials with wide band gap (~3.37 eV) and high excitation binding energy (~60 meV) at room temperature [1]. ZnO is an important inorganic material, which has multiple properties, such as semiconducting properties, photocatalytic activity, antibacterial activity and growth promoter [2]. It is widely applied in the field of optoelectronics [3-4], pharmaceutics [5], cosmetics [6-7], food science [8-9] and agriculture [2]. The antibacterial activity of ZnO has been widely explored [2, 10-12]. It has been documented that concentration, size and healing temperature can affect the antibacterial activity. ZnO as an inorganic antibacterial reagent is more stable than the organic reagents [2].

Up to now, a number of chemical routes have been used to synthesize ZnO powders such as hydrothermal method [13-14], spray pyrolysis [15-16] and sol-gel method [17-18]. Among these methods, sol-gel shows many advantages over other techniques such as its simplicity and low equipment cost. Therefore, in this study we had concentrated the effect of calcined temperature on controlling the structural, photocatalytic activity and antibacterial activity of ZnO powders by sol-gel method.

2. Experimental

2.1 Powders preparation

ZnO powders were prepared via sol-gel method [17]. Firstly, 2.1949 g of zinc acetate dehydrate ($Zn(CH_3COO_2).2H_2O$) was dissolved in 50 ml distilled water and mixture the vigorously stirred at room temperature for 15 min. Finally, 1.6 g NaOH that was dissolved in 50 ml distilled water was slowly added to the zinc precursor solution. The white precipitates were achieved and were then vigorously at room temperature for 45 min before filtering, rising with distilled water, drying at 60 °C for 24 h and calcined at the temperatures at 300, 500 and 700 °C in air for 1 h with a heating rate of 10 °C/min. For this work the ZnO powders calcined at the temperatures at 300, 500 and 700 °C were designated as T300, T500 and T700, respectively.

2.2 Powders characterizations

The morphology and particle size of the fabricated powders were characterized by Scanning Electron Microscope (SEM-Quanta 400). The phase composition was characterized using an X-ray diffractometer (XRD) (Phillips X'pert MPD, Cu-K). The crystallite size was calculated by the Scherrer equation, Eq. 1, [19].

$$D = 0.9\lambda/\beta\cos\theta \qquad (1)$$

Where D is the average crystallite size, λ = 0.15406 nm is the wavelength of the CuK_α radiation, θ is the Bragg angle and β is the full-width at half-maximum (FWHM) in radians.

2.3 Photocatalytic activity test

The photocatalytic activity was evaluated by the degradation of MB under UV irradiation using eleven 50 W

*Corresponding author. Tel.: +66 7433 6933
Email address: weerachai.sang@yahoo.com
doi: 10.14456/kkuenj.2016.4

of black light lamps. A 10 ml of MB with a concentration of 1×10^{-5} M was mixed with 0.0375 g of powders and kept in a dark chamber for 1 h, after that kept in a chamber under UV irradiation for 0, 1, 2, 3, 4, 5 and 6 h [19]. After photo-treatment for a certain time, the concentration of treated solution was measured by UV-vis. The ratio of remained concentration to initial concentration of MB calculated by C/C_0 was plotted against irradiation time in order to observe the photocatalytic degradation and the percentage degradation of the MB molecules (%DMB) was calculated by Eq. 2, [19].

$$\%DMB = 100(C_0-C)/C_0 \quad (2)$$

Where C_0 is the concentration of MB aqueous solution at the beginning (1×10^{-5} M) and C is the concentration of MB aqueous solution after exposure to a light source.

2.4 Antibacterial activity test

The antibacterial activity of powders against the bacteria *Escherichia coli* (*E. coli*) was studied. Aliquots of 10 ml *E. coli* conidial suspension (10^5 CFU/ml) were mixed with 0.05 g of powders. The mixture was then exposed to either UV irradiation (eleven 50 W of black light lamps) for 0, 5, 10, 15 and 20 min. Then, 0.1 ml of mixture suspension was sampled and spread on Macconkey Agar plate and incubated at 37 °C for 24 h. After incubation, the number of viable colonies of *E. coli* on each Macconkey Agar plate was observed and disinfection efficiency of each test was calculated in comparison to that of the initial or control (N/N_0) [19-20]. Percentage bacterial reduction or *E. coli* kill percentage was calculated according to the following equation, Eq. (3) [19-21]. The antibacterial activities were test at 3 samples for 1 condition and averaged value from the results.

$$E = 100(N_0-N)/N_0 \quad (3)$$

Where E is the percentage bacterial reduction or *E. coli* kill percentage, N_0 and N are the average number of live bacterial cells per milliliter in the flask of the initial or control and powders finishing agent or treated fabrics, respectively.

3. Result and discussion

3.1 Powders characterizations

The surface morphology was observed with SEM. Figure 1 shows surface morphologies of ZnO powders. It was seen that for all powders, the agglomeration and spherical shape with increasing calcined temperatures at 300 to 700 °C.

From the XRD study as shown in Figure 2, it was found that ZnO powders calcined at the temperatures at 300, 500 and 700 °C reveal only the ZnO phase. The peaks corresponded to the (100), (002), (101), (102), (110), (103), (200), (112) and 201) plans of ZnO in the wurtzite structure correspondence with JCPDS (card number 36-1451) [16]. It was apparent that calcined temperatures have significantly effect on crystallinity of ZnO phase from XRD, with the crystallinity increases with increases calcined temperatures.

The crystallite size of nanocrystalline ZnO powders which was calculated by Scherrer's formula gave the values of about 28.3, 22.5 and 12.5 nm for calcined at the temperatures at 300, 500 and 700 °C, respectively. It was found that crystallite size decreases with increases calcined temperatures due to the contribution of calcined temperatures effect. The result indicated that ZnO powders calcined at the temperatures at 700 °C exhibits smaller of crystallite size of nanocrystalline ZnO powders.

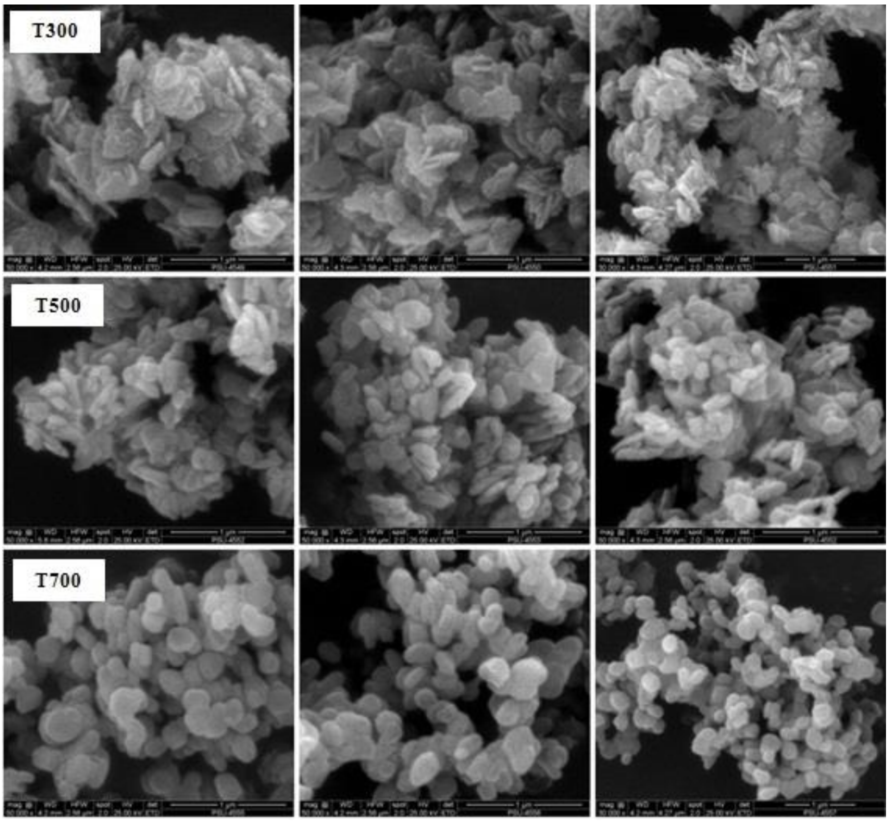

Figure 1 SEM surface morphology images of ZnO powders (magnification 5,000X)

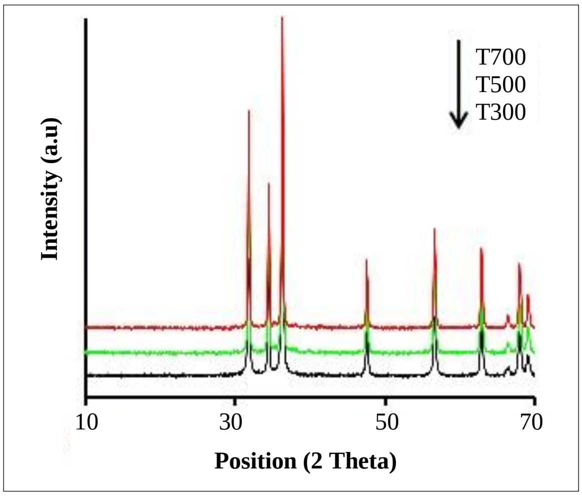

Figure 2 XRD patterns of ZnO powders

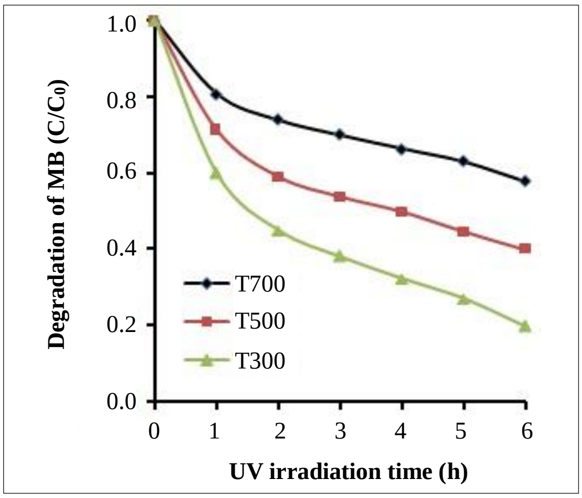

Figure 3 The photocatalytic activity of ZnO powders under UV irradiation

3.2 Photocatalytic activity

The photocatalytic degradation of MB (C/C_0) by using ZnO powders under UV irradiation is show in Figure 3. It was apparent that calcined temperatures have significantly effect on photocatalytic reaction under UV irradiation, with the photocatalytic activity increases with increases calcined temperatures due to effect of calcined temperatures.

The MB degradation percentage of ZnO powders under UV irradiation is shown in Figure 4. It was found that the MB degradation percentage of ZnO powders under UV irradiation for 6 h are 42.10, 60.01 and 80.06% for calcined at the temperatures at 300, 500 and 700 °C, respectively. It was found that ZnO powders calcined at the temperatures at 700 °C show the best photocatalytic activity.

3.3 Antibacterial activity

Figure 5 displays the *E. coli* survival rate (N/N_0) after testing with UV illumination on ZnO powders. The result shows that the *E. coli* survivals decrease with UV irradiation time. It also indicates that the ZnO powders calcined at 700 °C exhibit higher antibacterial activity compared to the ZnO powders calcined at 300 and 500 °C, respectively.

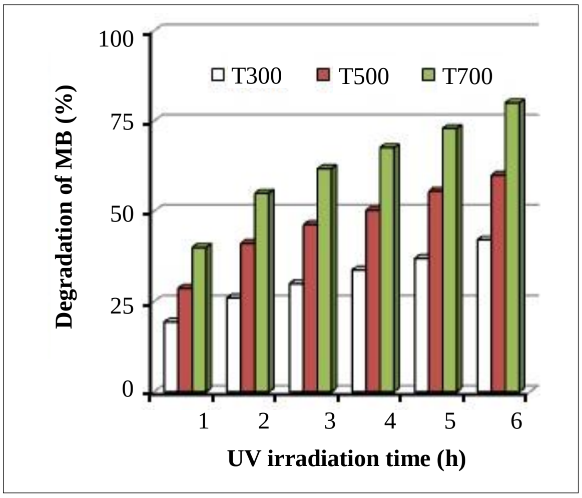

Figure 4 The MB degradation percentage of ZnO powders under UV irradiation

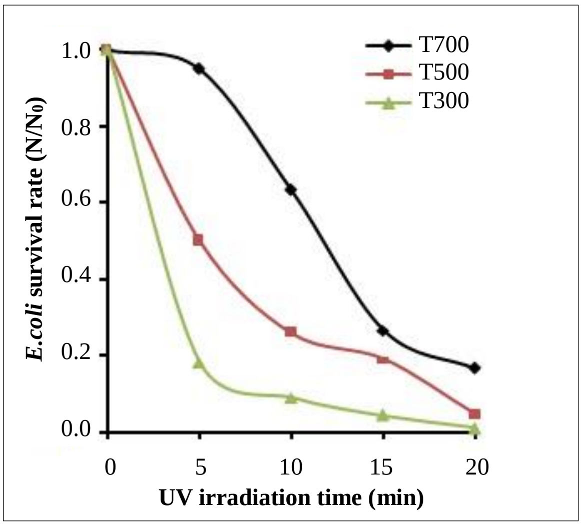

Figure 5 The antibacterial activity of ZnO powders under UV irradiation

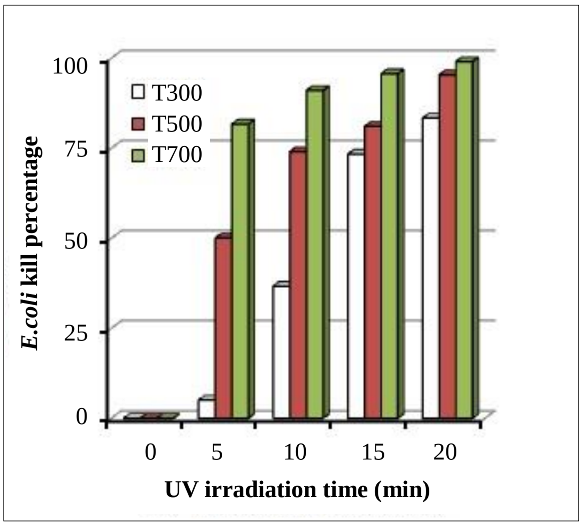

Figure 6 *E. coli* kill percentage of ZnO powders under UV irradiation

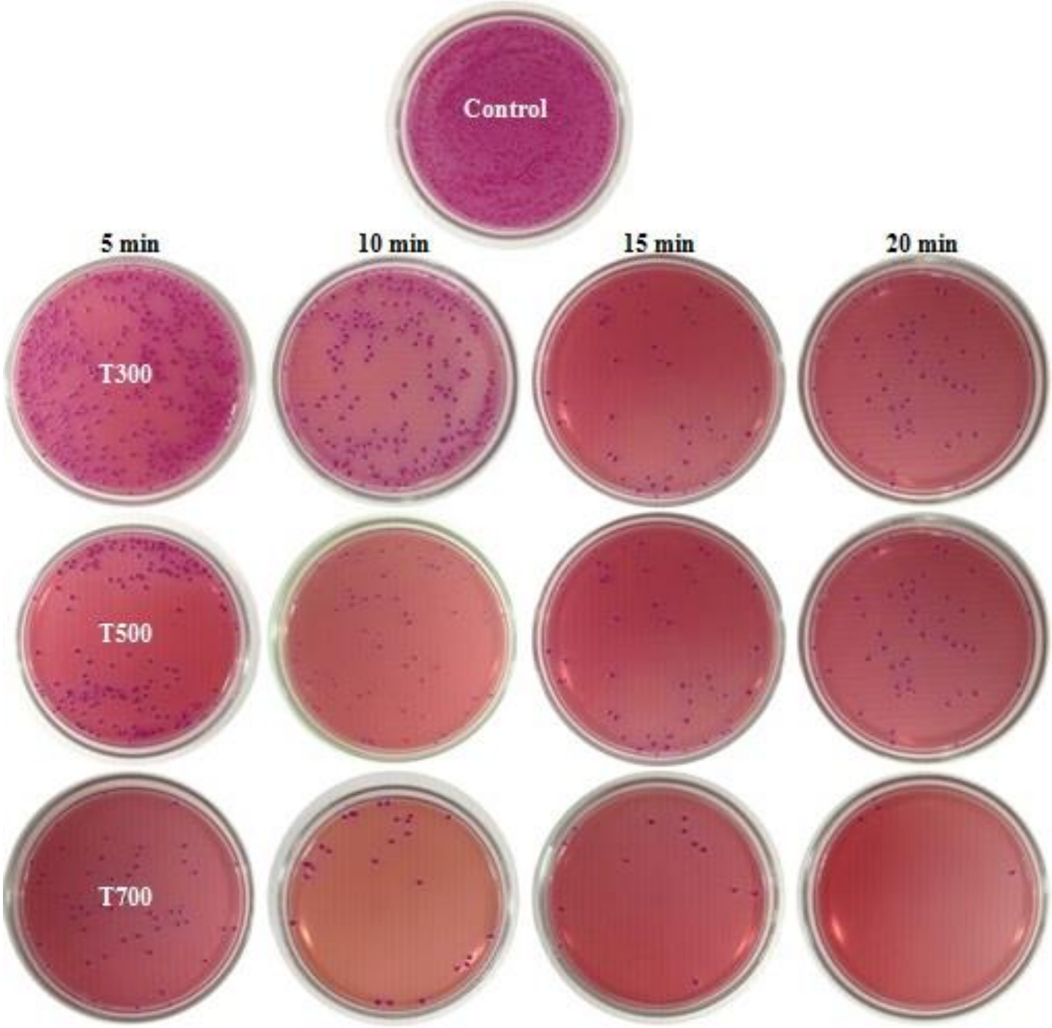

Figure 7 Photo of viable *E. coli* colonies under UV irradiation of ZnO powders compared with control

The *E. coli* kill percentage of ZnO powders under UV irradiation is shown in Figure 6. It is found that the *E. coli* kill percentage of ZnO powders under UV irradiation for 20 min are 83.33, 93.33 and 99.00% for ZnO powders calcined at the temperatures at 300, 500 and 700 °C, respectively. In this research, Researchers have studied the influence of UV disinfection affecting *E. coli* (case no ZnO powders) from testing found the *E. coli* kill percentage infection was very low. The percent mortality was only 5% under UV irradiation for 20 min, so the factors that affect the *E. coli* kill percentage infection for this research came from the influence of powders.

The photo of viable bacterial colonies (red spots) on fabricated ZnO powders and the control treated with UV are illustrated in Figure 7.

4. Conclusions

In this work, ZnO powders were fabricated by sol-gel method. The effect of calcined at the temperatures at 300, 500 and 700 °C on microstructure photocatalytic activity and antibacterial activity were investigated and concluded as followings,

1. ZnO powders reveal only the wurtzite phase and surface morphologies was found that for all powders, the agglomeration and spherical shape was observed and the particle size increases with increasing calcined temperatures.

2. The photocatalytic and antibacterial activities of ZnO powders increases with increasing calcined temperatures. It is note that ZnO powders calcined at the temperatures at 700 °C (T700) exhibits higher photocatalytic and antibacterial activities under UV irradiation with MB degradation percentage of 80.06% for 6 h and *E. coli* kill percentage of 99.00% for 20 min.

5. Acknowledgements

The authors would like to acknowledge Institute of Research & Development, Songkhla Rajabhat University and Faculty of Industrial Technology, Songkhla Rajabhat University, Thailand for financial support of this research.

6. Reference

[1] Sarma H, Sarma KC. X-ray peak broadening analysis of ZnO nanoparticles derived by precipitation method. Int J Sci Res Publ. 2014;4(3):1-7.

[2] Wang C, Liu LL, Zhang AT, Xie P, Lu JJ, Zou XT. Antibacterial effect of zinc oxide nanoparticles on Escherichia coil K88. Afr J Biotechnol. 2012;11(44): 10248-54.

[3] Djurisic AB, Ng AMC, Chen XY. ZnO nanostructures for optoelectronics: Materials properties and device applications. Progr Quant Electron. 2010;34:191-259.

[4] Zhang Z, Bian J, Sun J, Ma X, Wang Y, Cheng C, Luo Y, Liu H. High optical quality ZnO films grown on graphite substrate for transferable optoelectronics devices by ultrasonic spray pyrolysis. Mater Res Bull. 2012;47:2685-8.

[5] Yamamura S, Momose Y. Quantitative analysis of crystalline pharmaceuticals in powders and tablets by a pattern-fitting procedure using x-ray powder diffraction data. Int J Pharm. 2011;212:203-12.

[6] Kuo CL, Wang CL, Ko HH, Hwang WS, Chang KM, Li WL, Huang HH, Chang YH, Wang MC. Synthesis of zince oxide nanocrystalline powders for cosmetic applications. Ceram Int. 2010;36:693-8.

[7] Le TH, Bui AT, Le TK. The effect of Fe doping on the suppression of photocatalytic activity of ZnO nanopowder for the application in sunscreens. Powder Tech. 2014;268:173-6.

[8] Yu N, Zhang M, Islam MN, Lu L, Liu Q, Cheng X. Combined sterilizing effects of nano-ZnO and ultraviolet on convenient vegetable dishes. LWT- Food Sci Tech. 2015;61:638-43.

[9] Shankar S, Teng X, Li G, Rhim JW. Preparation, characterization, and antimicrobial activity of gelatin/ZnO nanocomposites films. Food Hydrocolloids. 2015;45:264-71.

[10] Ravichandran K, Rathi R, Baneto M, Karthika K, Rajkumar PV, Sakthivel B, Damodaran R. Effect of Fe+F doping on the antibacterial activity of ZnO powder. Ceram Int. 2015;41:3390-5.

[11] Talbian N, Amininezhad SM, Doudi M. Controllable synthesis of ZnO nanoparticles and their morphology-dependent antibacterial and optical properties. J Photochem Photobiol B Biol. 2013;120:66-73.

[12] Nair MG, Nirmaka M, Rekha K, Anukaliani A. Structure, optical, photo catalytic and antibacterial of ZnO and co doped ZnO nanoparticles. Mater Lett. 2011;65:1797-800.

[13] Suwanboon S, Klubnuan S, Jantha N, Amornpitoksuk P, Bangrak P. Influence of alkaline solution on morphology of ZnO prepared by hydrothermal method for using as photocatalyst and bactericidal agent. Mater Lett. 2014;115:275-8.

[14] Suwanboon S, Amornpitoksuk P, Bangrak P, Randorn C. Physiscal and chemical properties of multifunctional ZnO nanostructures prepared by precipitation and hydrothermal methods. Ceram Int. 2014;40:975-83.

[15] Chen CY, Weng JC, Chen JH, Ma SH, Chen KH, Horng TL, Tsay CY, Chang CJ, Lin CK, Wu JJ. Photocatalyst ZnO-doped Bi_2O_3 powder prepared by spray pyrolysis. Powder Tech. 2015;272:316-21.

[16] Ozcelik BBK, Ergun C. Synthesis of ZnO nanoparticles by an aerosol process. Ceram Int 2014;40:7107-16.

[17] Suwanboon S. Structural and optical properties of nanocrystalline ZnO powder from sol-gel method. Sci Asia. 2008;34:31-4.

[18] Duan L, Zhao X, Zheng Z, Wang Y, Geng W, Zhang F. Structural, optical and photocatalytic properties of (Mg,Al)-codoped ZnO powders prepared by sol-gel method. J Phys Chem Solid. 2015;76:88-93.

[19] Sangchay W. Photocatalytic and antibacterial activity of Ag-doped TiO_2 nanoparticles. KKU Res J. 2013;8(5):731-8.

[20] Sangchay W. Fe doped TiO_2 thin films coated on glass fiber to inhibit bacterial of *E. coli* prepared by sol-gel method. Dig J Nanomater Bios. 2014;9(4):1593-601.

[21] Sangchay W, Ubonchonlakat K. Photocatalytic disinfection of water containing *E. coli* using Fe^{3+} doped TiO_2 thin films coated on glass fibers. Dig J Nanomater Bios. 2015;10(1):283-90.