Biotemplated synthesis of Ag-ZnO nanoparticles/ bacterial cellulose nanocomposites for photocatalysis application

by Henry Aritonang 3

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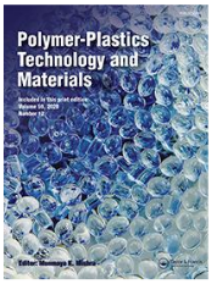

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Biotemplated synthesis of Ag-ZnO nanoparticles/bacterial cellulose nanocomposites for photocatalysis application

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ABSTRACT

Bacterial cellulose (BC) was used as a biotemplate for facile fabrication of silver-zinc oxide nanoparticles/bacteria cellulose (Ag-ZnO/BC) nanocomposite having high potential application in photocatalysis via a one-step method. Scanning electron microscopy images confirmed that BC nanofibers were uniformly coated with Ag-ZnO in aqueous suspension using co-precipitation method. The size of Ag-ZnO nanoparticle in BC and its photodegradability were increased with the increasing concentration of $AgNO_3$ added. The greatest efficiency is demonstrated by the ability of this material to degrade methylene blue (MB) by up to 76% after a 180 min ultraviolet irradiation period, indicated that the Ag-ZnO/BC nanocomposite is a promising candidate as robust ultraviolet responsive photocatalyst.

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Bacterial cellulose; Ag-ZnO nanoparticle; nanocomposite; photocatalyst

1. Introduction

Bacterial cellulose (BC) is a unique material produced by Gram-negative bacterium *Acetobacter xylinum*. During cultivation, the bacteria synthesize fine sub-elementary cellulose fibrils, which are extruded from terminal enzyme complexes into the culture medium. BC is markedly different from cellulose obtained from trees and cotton, obtained free of lignin and hemicellulose, in a 3-D network composed of a bundle of much finer microfibrils of nanometric size.[1,2] In vitro and in vivo studies demonstrated its biocompatibility to produce nanocomposite materials.[3,4] Most of these composites are prepared for long-term durability using non-degradable polymeric resins and high-strength fibers. However, the environmental impact of persistent non-degradable plastic-based wastes is a global concern. Therefore, various biodegradable polymers have been investigated as replacements for the non-degradable plastics.[5,6] Due to its good mechanical properties, water sorption capacity, porosity, stability and conformability, BC has been used in tissue engineering of cartilage,[7] replacement of blood vessels[8] and in the wound healing process.[9]

BC-based nanocomposites can be fabricated statically either by using the synthesized BC gel or modifying the cellulose biosynthesis. For instance, BC nanocomposites are used for environmental applications to purify contaminants from water by utilizing photocatalysts.[10,11] Photocatalysis is an efficient, attractive, and clean technology for pollutant abatement either in aqueous media or in the gas phase.[12–14] ZnO is a photocatalyst[15]; its widely reported band gap matches with that of TiO_2 and its conduction band (CB) and valence band (VB) edges are very close to those of TiO_2.[16] Exposure of semiconductor nanocrystals to light of energy not less than the bandgap leads to the formation of electron/hole pairs, electrons in the CB and holes in the VB.[17,18] Some of these pairs diffuse to the crystal surface and react with the adsorbed substrates, resulting in photocatalysis.[19,20]

Photocatalysis takes place on the surface of the semiconductor and the morphology and surface modification have a strong influence on the photocatalytic activity.[21,22]

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Tuning the performance and activity of nanocrystalline ZnO by doping it with Ag is of current interest.[23,24] ZnO behaves as an n-type semiconductor[25] and Ag doping is found to be effective for the fabrication of *p*-type ZnO.[26,27] In addition, Ag doping modifies the optical and electronic properties, which in turn influence the photocatalytic activity. The present work reveals larger photocatalytic activities of sol–gel synthesized ZnO and Ag-doped ZnO. Preparation of nanoparticulate Ag-ZnO by chemical,[28] photochemical,[29,30] precipitation,[28,31] hydrothermal[32,33] or solvothermal,[34] microwave[35], flame spray pyrolysis[36], and electrospinning[37] methods has been reported. Karunakaran et al.[38] and Wu et al.[34] synthesized Ag-ZnO by sol–gel and solvothermal method, respectively. Some of the methods, however, produce larger size of Ag-ZnO and require reducing agent and synthetic polymer as capping agent.

In this research, for the first time, Ag-ZnO heterostructure nanoparticles were synthesized with BC as a stabilizer in order to prevent the formation of aggregated Ag-ZnO heterostructure nanoparticles and to improve the stability of the nanoparticle dispersion. By this method, the use of synthetic polymer as capping agent is reduced so that it is more economical and reduces environmental risks. These Ag-ZnO/BC materials have been examined for their photocatalytic properties. It was expected that the synthesized Ag-ZnO/BC heterostructure nanoparticles would exhibit excellent photocatalysis, and then, the synthesized Ag-ZnO/BC nanocomposite might be used in the fields of environmental especially in degrading synthetic dyes.

2. Materials and methods

2.1. Materials

$Zn(NO_3)_2.6H_2O$, $AgNO_3$, NaOH, urea, glacial acetic acid, methylene blue, and ethanol were obtained from Sigma-Aldrich (99,9%). Coconut water, sucrose (food-grade white sugar) and *Acetobacter xylinum* were obtained from a local traditional market. All chemical materials and solvents used in the experiments were analytical grade reagents and were used without further purification.

2.2. Synthesis of BC membranes

BC was synthesized according to the procedure previously described. At first, 5 L of coconut water was filtered using gauze and was boiled. 500 g of sucrose, 25 g of urea and 30 mL of glacial acetic acid were then added while stirring. Three hundred milliliters of this solution was poured into a plastic tray and was allowed to cool. After that, 30 mL of *Acetobacter xylinum* was added and then left for 6 days. BC gel produced was rinsed using hot water for 15 min, immersed in NaOH 1% (m/v) for 24 h, and was then immersed in glacial acetic acid 1% (v/v) for 24 h. The BC gel was subsequently rinsed with deionized water (pH 7) and stored at 4°C. BC gel was cut in the size of 4 cm x 4 cm, pressed to reduce the water content and finally air-dried at room temperature for 6 days to produce BC membranes.[39]

2.3. Synthesis of Ag-ZnO/BC nanocomposites

The Ag-modified ZnO inside BC (denoted as Ag-ZnO /BC nanocomposite) was prepared by co-precipitation method. Five grams of $Zn(NO_3)_2.4H_2O$ and $AgNO_3$ (each wt1%, wt3%, wt5%, wt7%, and wt10% of the weight of $Zn(NO_3)_2.4H_2O$) were dissolved in 100 mL of distilled water, hereafter denoted as wt1% Ag/ZnO, wt3% Ag/ZnO, wt5% Ag/ZnO, wt7% Ag/ZnO, and wt9% Ag/ZnO. Then, BC was put into solution under vigorous stirring with sonicator for 1 h. Sonication was continued for 15 min along with dropwise adding an aqueous solution of 1 M NaOH, until pH 13 was reached and precipitate was formed. The above solution was subjected to sonication at the ambient condition with a high-density ultrasonic probe immersed directly in the solution.

Ag-ZnO/BC-gel nanocomposite was removed from the solution and rinsed with distilled water until the composite pH became neutral. Nanocomposite membrane was obtained through a compressive process and air-dried at room temperature. The dried nanocomposite was characterized by SEM-EDS, X-RD, TEM, and was analyzed for their photocatalytic activity.

2.4. Characterization of membrane fiber

2.4.1. X-Ray Diffraction (XDR)

X-ray diffraction spectra were recorded using a PW1710-based diffractometer (Japan) using Cu Kα radiation at 40 kV and 30 mA, step pass of 0.01° and a step time of 3 s, from 5 to 90° (2θ angle).

2.4.2. Scanning Electron Microscopy (SEM)–energy dispersive spectroscopy (EDS)

The BC dried composite samples were coated with gold (MC1000, Hitachi, Tokyo, Japan). Analysis of the structure of the BC and BC composites was performed using a JEOL SEM equipped with an EDS detector with an accelerating voltage of 15 kV. Samples were mounted on Cu tape to avoid any possible artifacts in the resulting EDS spectra from the use of a carbon conductive tape.

2.4.3. Transmission electron microscopy (TEM)

TEM measurements were obtained in a JEOL HT-7700 microscopy, operating at 120 kV. The samples were sonicated to remove Ag and ZnO nanoparticles from BC and the particles were, then, suspended in ethanol. A drop of the suspension was deposited on the copper grid.

2.5. Photocatalytic activities test

The photocatalytic performance of the Ag-ZnO/BC samples was evaluated by using methylene blue (MB). A UV lamp (20 W) served as the UV light source for UV light photocatalysis. Typically, Ag-ZnO/BC nanocomposite with a size of 1 × 1 cm^2 was ultrasonically dispersed into 15 mL aqueous suspensions of MB (5 ppm). Prior to irradiation, the mixture was stirred in the dark for 16 min to establish an adsorption/desorption equilibrium, and it was then irradiated for some periods of time: 60, 120, and 180 min. After a given irradiation time, a certain amount of solutions was collected and centrifuged to remove the photocatalysts. Subsequently, the residual MB concentration was determined by a UV–vis spectrophotometer. The efficiency of the photocatalysts for photocatalytic degradation (D) of MB dye was calculated using the following formula[40]: $D(\%) = (C_o\text{-}C_t)/C_o$ x 100%, where C_o is the concentration of aqueous MB dye before the addition of any photocatalyst and C_t is the concentration of MB in the reaction suspension containing photocatalyst following UV exposure for time t.

3. Results and discussion

3.1. Morphology

A set of selected SEM micrographs of the surface of composite materials was studied. For each material, the BC fibril dispersion and the interfacial adhesion are shown. Obviously, in the case of BC, only the small mat fragments, and not the isolated fibrils, are observed (Figure 1a). Figure 1b–c shows the pressed and dried Ag/BC and ZnO/BC sheets, respectively. SEM micrographs provide evidence of strong interface adhesion between BC and matrix fibers. The surface of BC is covered by round granules of Ag nanoparticles, and needle-shaped particles from ZnO nanoparticles attached to the surface of the BC fibers.

Meanwhile, on Ag-ZnO/BC membranes (Figure 2), the shape of each particle is not obvious because it has joined together, however the particles are seen effectively incorporated into the surface layer of the BC matrix and cover a larger surface area on the surface of the BC matrix because of their 2-D morphology.[41] A drastic change in the material is also observed when the Ag content in the composite is increased. For example, the wt 9% Ag-ZnO covers over BC and is not possible even to distinguish the BC surface. The surface of composites is not smooth at high filler content due to the self-ordering of the high aspect ratio of Ag.[42,43]

The formation of ZnO and Ag nanoparticles in the BC matrix was initiated at the immersion of BC in an aqueous solution of $ZnNO_3$ and $AgNO_3$. Zn^{2+} and Ag^+ ions interacted through electrostatic interactions with oxygen ions from the hydroxyl and ether groups found in BC fibers. Interactions occured in all parts of the fiber because they were diffused into all parts of BC.[39,44,45] When a reduction occurs, the ions were turned into Ag and ZnO nanoparticles. Although interactions occur with all parts of cellulose fiber, particle nucleation processes are limited due to competition between Zn^{2+} and Ag^+ ions.

When Ag and ZnO nanoparticles were removed from the BC matrix by sonication, the cellulose fibers were released into the solution together with the two particles. Based on TEM analysis, it is shown that Ag-ZnO nanoparticles are attached to the surface of cellulose fibers (Figure 3a). This information supports the previous images that particles are attached to the surface of cellulose fibers. In addition, the average size of these particles varies from 11 to 16 nm, and their size increased with the increasing concentration of Ag solution (Figure 3b–c; Table 1).

Table 1 shows that Ag content is increased with the increasing of $AgNO_3$ concentration which in turn increases the nanoparticle size. This is made possible by the increasing number of Ag particles attached to the surface of Ag itself and to the surface of ZnO.

EDS performed on the samples shows the existence of C, O, Zn, and Ag elements. The Ag content is increased based on EDS data and corresponds to the increasing concentration of $AgNO_3$, as presented in Figure 4 and Table 1.

The EDS spectra show that the intensity of the Ag peak is increased because of the increased concentration of the Ag solution added to the BC membrane.

3.2. X-ray diffraction

XRD is the most commonly used in materials science to investigate the crystalline structure, the ratio of crystalline to amorphous regions, crystal size, the distance between planes of crystals and the arrangement patterns of crystals. In this study, XRD analysis were conducted to explore the microstructural changes in the BC matrix caused by the adsorption and penetration of ZnO and Ag particles. The XRD patterns of BC and nanocomposites are shown in Figure 5.

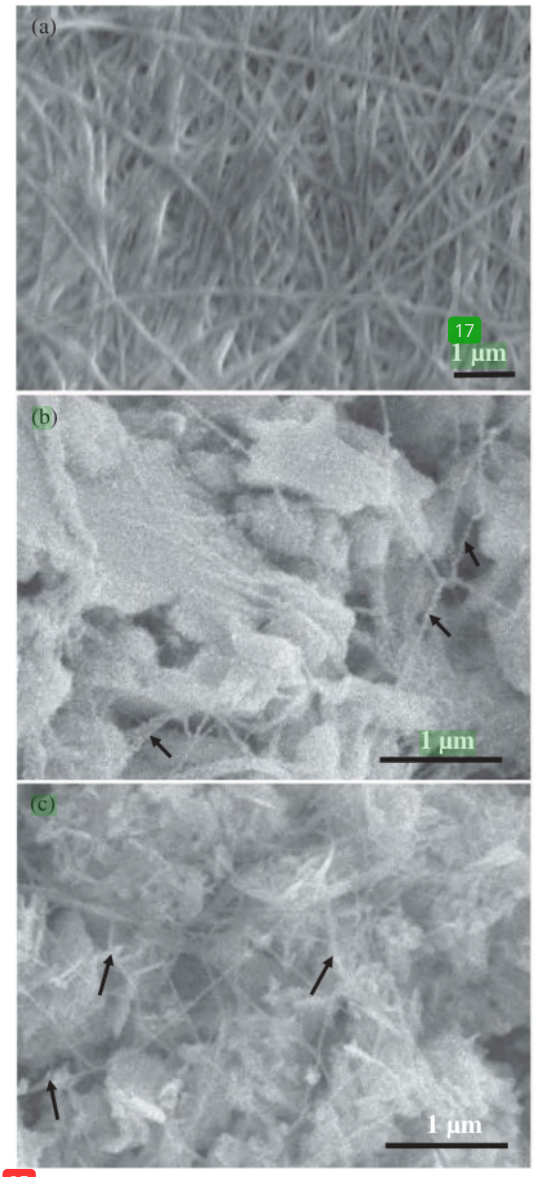

Figure 1. SEM images of (a) pure BC membrane, (b) Ag/BC, and (c) ZnO/BC nanocomposite.

All the diffraction lines are assigned well to the hexagonal phase of the wurtzite structure of ZnO with a reference pattern as mentioned in the ICDD No. 01-079-0206. The XRD diffraction peaks represent the crystalline nature of ZnO nanoparticles. The Ag-doped ZnO composites exhibit diffraction patterns that are well-matched with the reference pattern and the composites reveal some additional diffraction peaks at 2θ 38° and 64° (marked by asterisks) associated with the face-centered cubic (FCC) phase of metallic Ag (ICDD No. 00-004-0783) and the intensity of these peaks increases with the increase of Ag concentration in the composites. clearly indicates the formation of crystalline Ag cluster in the ZnO nanoparticles. The Ag^+ ions in ZnO lattice behave as monovalent dopant, which has the ability to occupy both the lattice and interstitial sites due to the higher ionic radius of Ag^+ ions (1.29 Å) than Zn^{2+} (0.74 Å). The effect of Ag incorporation in the ZnO lattice is studied by monitoring the position and broadening of the diffraction peaks and it is affected severely due to the incorporation of Ag.

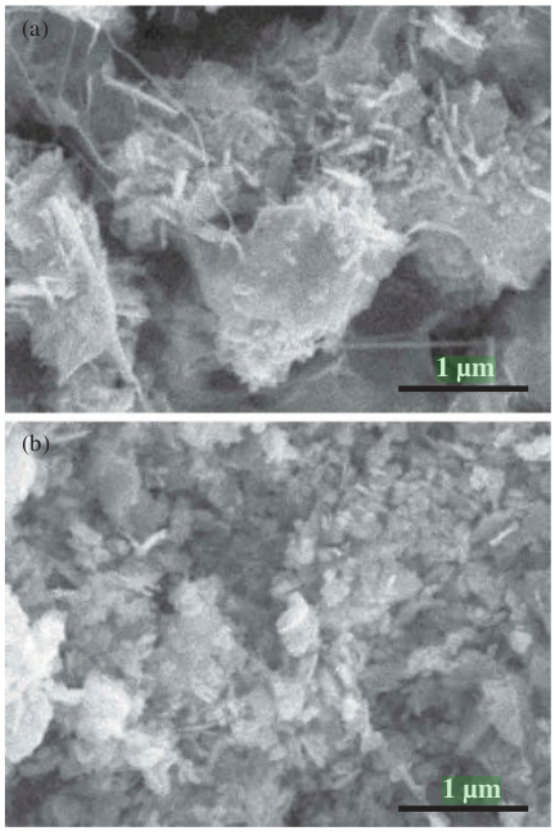

Figure 2. SEM images of (a) wt1% and (b) wt9% Ag-ZnO/BC nanocomposites.

It is shown in the diffractogram that the peak intensity in wt3% is higher than that in wt1%, especially between 2θ 30° and 40°. However, in wt3%, there are also additional peaks which could be the unexpected impurities.

As the concentration of $AgNO_3$ is increased, the surface of BC is increasingly covered. This could be observed in the decreasing peak intensity for BC and this

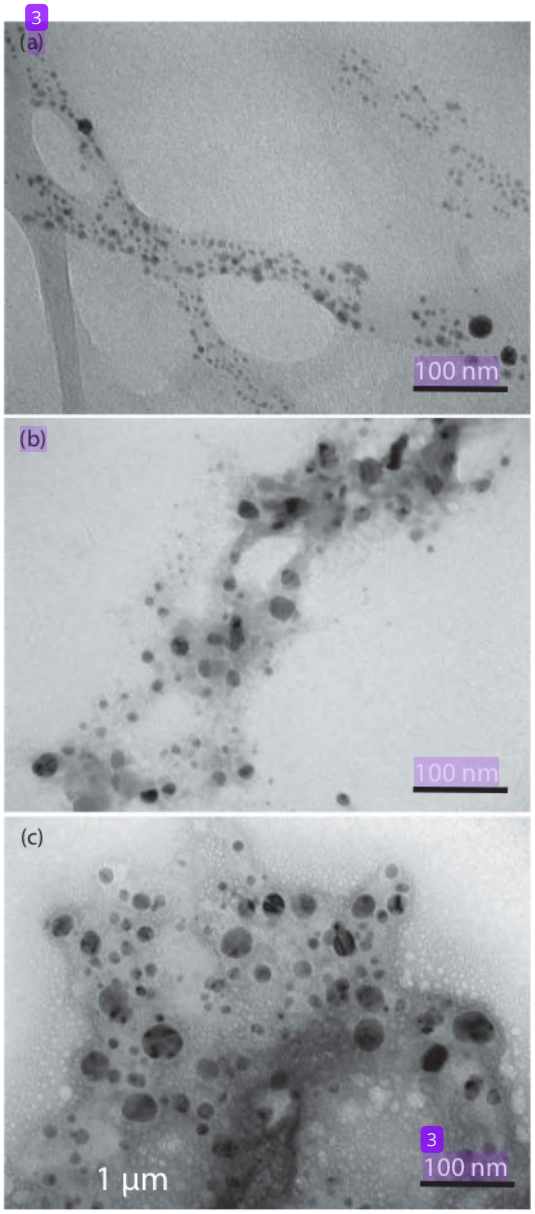

Figure 3. TEM images of (a) Ag-ZnO attached to the surface of cellulose fibers, and (b-c) wt1% and wt9% Ag-ZnO/BC nanocomposites, respectively.

Table 1. The average size and Ag nanoparticles content in each Ag-ZnO/BC nanocomposites based on TEM and EDS data.

Nanocomposites	Particles size (nm)	Ag content (weight %)
wt1%	11.3 ± 5.2	2.27
wt3%	12.0 ± 5.1	4.58
wt5%	15.1 ± 5.1	6.99
wt7%	15.2 ± 7.2	9.40
wt9%	16.1 ± 7.2	10.12

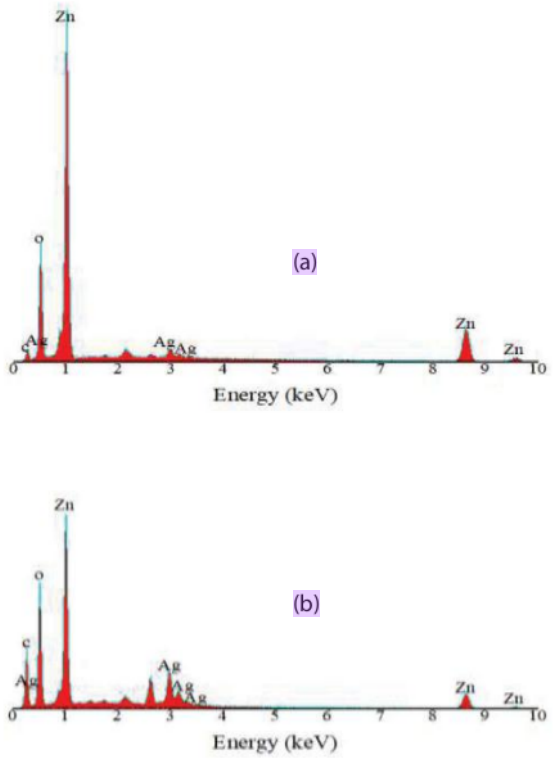

Figure 4. EDX spectra of (a) wt1% and (b) wt9% Ag-ZnO/BC nanocomposites.

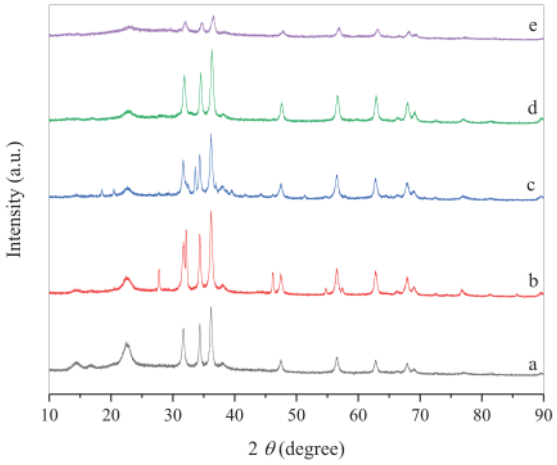

Figure 5. The XRD patterns of (a) wt1%, (b) wt3%, (c) wt5%, (d) wt7%, and (e) wt9% Ag-ZnO/BC nanocomposites.

information was reported in previous studies.[39,46] In addition, the diffractogram shows that the lowest peak intensity exists in wt9%, especially the peak intensity for ZnO. It is suspected that the higher the concentration of

$AgNO_3$, the more Ag^+ and Zn^{2+} ions are overlapped and packed tightly together and, as a result, compete with each other to be reduced into Ag and ZnO. This overlap is occured in the surface of membrane and inner part of BC fiber, as well. The increasing concentration of Ag also drives more Zn^{2+} ions into the interior of the BC membrane. Consequently, most of the trapped metal ions, especially Zn^{2+}, are not reduced.

3.3. Photocatalytic activity

As previously reported[26,29,38], Ag and ZnO nanoparticles are the photocatalytic agents in Ag-ZnO/BC nanocomposite.

Figure 6 shows the degradation efficiencies of MB on the Ag-ZnO/BC nanocomposites with different duration of time using BC as the template. In general, degradation efficiency for all nanocomposites is increased with the increasing irradiation time. When the irradiation time is 60 min, the highest photocatalytic activity is shown by wt9% Ag-ZnO/BC nanocomposite which was 17.55%. This is acceptable because the nanoparticles content found in these nanocomposites is relatively higher than other nanocomposites. Furthermore, the photocatalytic activity is increased sharply from irradiation time of 60 min to 120 min for all nanocomposites, and the highest was still shown by nanocomposites which contain more Ag nanoparticles. However, when the irradiation time is increased to 180 min, the increase in photocatalytic activity is not as large as before and even form a flat line. These results indicate that Ag and ZnO nanoparticles function as photocatalysts despite their presence in the BC membrane which is a support and matrix[39,45] for the two nanoparticles. This is similar to the study reported by Chong et al.[47] on Ni-doped ZnO semiconductors, except that they did not use BC matrix.

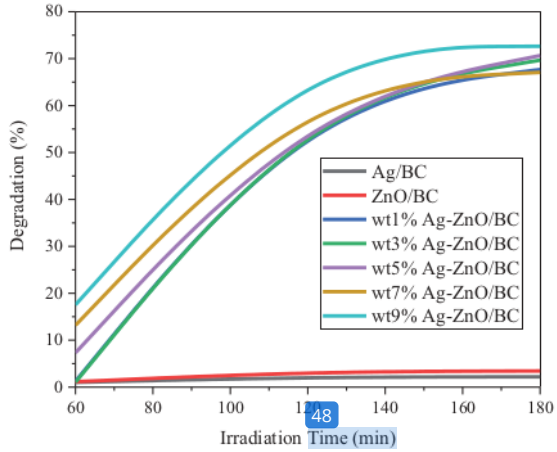

Figure 6. The effect of irradiation time on MB degradation catalyzed by the Ag-ZnO/BC nanocomposite catalysts.

This information shows that particle size and surface area have a major effect on the photocatalytic activities of the material. However, in addition to these factors, other parameters, such as morphology, defect, and impurity contents might affect photocatalytic activity.[48]

4. Conclusions

In this work, Ag-ZnO/BC nanocomposites with good photocatalytic activity were prepared via an easy one-step biotemplated method in aqueous suspension under alkaline conditions using NaOH. Nanocrystals of ZnO with well-defined wurtzite structures and round-shaped Ag were obtained with a size of less than 100 nm. The as-used co-precipitation method is effective to synthesize Ag-ZnO embedded BC. The results of XRD, SEM, and TEM analysis confirmed Ag-ZnO nanoparticles. The photocatalytic ability of the newly developed Ag-ZnO/BC nanocomposite was evaluated by MB degradation. The results indicated that the robust BC-supported Ag-ZnO nanoparticles were effective in photocatalysis of organic dyes like MB. In addition, size-controlled Ag-ZnO on renewable BC nanofibers offered the potentialities as recyclable photocatalyst in the area of the catalytic processes.

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