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Research Article

Effect of Different Mediated Agents on Morphology and Crystallinity of Synthesized Silver Nanowires Prepared by Polyol Process

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Synthesis and characterization of multiple crystalline silver nanowires (NWs) with uniform diameters were carried out by using 1,2-propandiol and ethylene glycol (EG) as comediated solvents and $FeCl_3$ as mediated agent in the presence of poly(vinyl pyrrolidone) (PVP). Experimental data and structural characterizations revealed that AgNWs have evolved from the multiple crystalline seeds initially generated by reduction of $AgNO_3$ with EG and 1,2-propandiol followed by reducing Fe(III) to Fe(II) which in turn reacts with and removes adsorbed atomic oxygen from the surfaces of silver seeds. In addition, uniform silver nanowires were obtained by using $FeCl_2$ and $AlCl_3$ as mediated agents in EG solution. Field emission scanning electron microscopy (FESEM) and transmission electron microscopy (TEM) showed uniform nanowires in both diameter and length. UV-Vis spectra showed adsorption peaks confirming the formation of nanowires. X-ray diffraction (XRD) patterns displayed the final product with high crystallinity and purity. In this study, a growth mechanism for forming AgNWs was proposed and a comparison between different mediated agents was carried out.

1. Introduction

One-dimensional metal nanostructures have received significant attention in a wide range of applications; among them silver nanowires (AgNWs) are of great importance because of having high electrical and thermal conductivity in comparison with other metals [1–5]. Different applications require different silver nanowires with specific properties and among all the parameters affecting them, morphology plays an important role and it is strongly influenced by the production method used for synthesizing the wires. Various methods have been utilized to synthesize silver nanowires, like solvothermal method [5], wet chemical synthesis [6], ambient solution phase [7], soft solution processing [8], polyol method [9], hydrothermal technique [2, 10], ultraviolet irradiation [11], synthesis with DNA, and so forth [12]. Among these methods, polyol process is the most applicable due to its simplicity, low cost, high yield, and less reaction time.

Some researchers synthesized silver nanowires through polyol method and studied different factors affecting the final product.

Lee et al. [13] synthesized ultra-long AgNWs with multi-step growth by polyol process. Other researchers studied the effect of reaction time, temperature, and molecular weight of poly(vinyl pyrrolidone) (PVP) on the morphology of silver nanowires [14]. A few studies have considered the effects of using mediated agents on the growth and the morphology of AgNWs. Jia et al. [15] used EG and glycerol as comediated agents and studied the self-seeding and growth mechanisms

of AgNWs. Tang et al. [16] used air assisted method with NaCl as mediated agent. In other studies different chloride salts such as $FeCl_3$ and $CuCl_2$ have been used to synthesize AgNWs in polyol process [5, 17, 18]. Although these chloride salts play the same role in controlling the growth of AgNWs through oxidative etching and stabilizing the reaction, the solubility and the nature of chloride salts influence the structural uniformity of silver nanowires.

Recently, An et al. [19] have provided a new method to produce AgNWs from AgCl nanocubes, using glycerol mediated solution, in which the viscosity of the solution could control the migration of Ag^+ ions. Moreover, Jia et al. [15] used glycerol and ethylene glycol as comediated agents to control the nucleation in the beginning of the reaction and to control the growth of AgNWs.

In our study, for the first time multiple crystalline AgNWs were prepared by using EG and 1,2-propandiol as comediated agents and $FeCl_3$ as the second mediated agent in the presence of PVP to provide a high stable solution to control the growth of uniform AgNWs with high efficiency and thin diameter in a short reaction time. The results showed that the use of two different mediated agents reduced the reaction time, effectively. It was observed that the color of the solution changed to dark gray, immediately after the injection of the $AgNO_3$ solution, and then changed to light gray after 30 min and it remained unchanged, as an indication of the formation of AgNWs. In addition, $FeCl_2$ and $AlCl_3$ were used in EG solution to study the effect of different chloride ions on the morphology and the crystallinity of AgNWs. Indeed some specific applications such as stretchable and wearable electrochromic devices, elastomer nanocomposites, highly conductive flexible paper, and flexible touch panels can be produced based on AgNWs. Also, AgNWs can be used to produce antibacterial textiles for medical application [1–7].

2. Experimental

2.1. Materials. All the chloride salts ($FeCl_2$ (purity 98.0%), $FeCl_3$ (purity 98.0%), and $AlCl_3$ (purity 99.99%)) and solvents (EG (purity > 99.5%), 1,2-propandiol (purity > 99.5%), and acetone) were purchased from Merck. Silver nitrate ($AgNO_3$ (purity > 99.0%)) was purchased from Fluka and PVP (K-30, Mw 40,000 (purity > 99.0%)) from Dae-Jung (Korea). Deionized water (HPLC grade) was provided using Millipore Equipment, USA, with conductivity of 1000 μs. All chemical materials were used as received without any further purification.

2.2. Synthesis Methods

Method A (Without Any Mediated Agents). 10 mL of 1,2-propandiol was added to a 50 mL round bottom flask and was heated for 2 hours at 170°C in a silicon oil bath, and then 0.5 mL of (0.005 M) $AgNO_3$/1,2-propandiol solution was injected dropwise into the flask under vigorous magnetic stirring (fixed at 300 rpm). Then 3 mL $AgNO_3$/1,2-propandiol (0.1 M) and after that 3 mL PVP/1,2-propandiol (0.45 M) were added dropwise to the flask, at the rate of 36 mL/hr. After 30 minutes the color of the solution changed to pale yellow. The reaction continued for 1 and a half hours, in which the color gradually changed to light gray and after that, the solution was allowed to be cooled to room temperature for 40 minutes. Then, the mixture was washed and centrifuged by acetone and deionized water and the final product was kept in deionized water for later characterization. All solutions used were made from highly pure chemicals, without any moisture. All glassware used in the synthesis process was washed several times by deionized water and acetone and then dried in hot air.

Method B (With $FeCl_2$ as Mediated Agent). 10 mL of EG was added to a round bottom flask and was heated at 155°C for 1 hour in a silicon oil bath. Then 80 μL of (0.04 M) EG/$FeCl_2$ solution was added to the flask and magnetic stirrer was fixed at 500 rpm. After 15 minutes, 3.5 mL of (1.14 M) PVP/EG and 3.5 mL of (0.94 M) $AgNO_3$/EG solutions were injected dropwise into the flask, respectively, in 10 minutes, after which the color of the solution changed to light yellow; 30 minutes later the color of the solution became milky and after 60 minutes it changed to gray. The resulting suspension was allowed to be cooled to room temperature, washed, and centrifuged by acetone and deionized water and the final product was kept in deionized water.

Method C (With $AlCl_3$ as Mediated Agent). In this method the same solution as in the previous methods was prepared but a different mediated agent, $AlCl_3$, was used. 80 μL of (0.04 M) $AlCl_3$/EG solution was used and the magnetic stirring speed was 500 rpm. Reaction temperature was 175°C and the color of the solution changed to muddy yellow after 1 hour and it gradually became gray in 2 hours. The final solution was allowed to be cooled to room temperature, and it was washed and centrifuged similar to methods A and B.

Method D (Comediated Solution and $FeCl_3$ as Mediated Agent). 10 mL of 1,2-propandiol was added to a round bottom flask, heated at 170°C for 1 hour, and then 3 mL of EG was added to the flask and was heated for one more hour. Later 80 μL of (0.04 M) EG/$FeCl_3$ solution was injected into the flask and the magnetic stirrer was set at 500 rpm. After 15 min, 1 mL of (0.005 M) 1,2-propandiol/$AgNO_3$ solution was injected dropwise into the flask. Afterward 4.5 mL of (0.1 M) 1,2-propandiol/$AgNO_3$ and then 4.5 mL of (0.45 M) 1,2-propandiol/PVP solutions were injected dropwise, each in 5 minutes. Five minutes after the injection, the color of the solution changed to dark gray and a lot of bubbles appeared on the surface of the solution. Gradually, after 30 minutes the color became light gray and heating continued for the next 50 minutes. Washing and centrifuging processes were similar to the other methods mentioned before.

2.3. Characterization. Morphology was observed by FESEM (MIRA3 TESCAN, Czech Republic) and TEM (Zeiss, EM10C, 80 KV). UV-visible spectroscopy analyzer used was SHIMADZU UV-1650PC and X-ray diffraction analyzer was V3RD-Philips, Analytica.

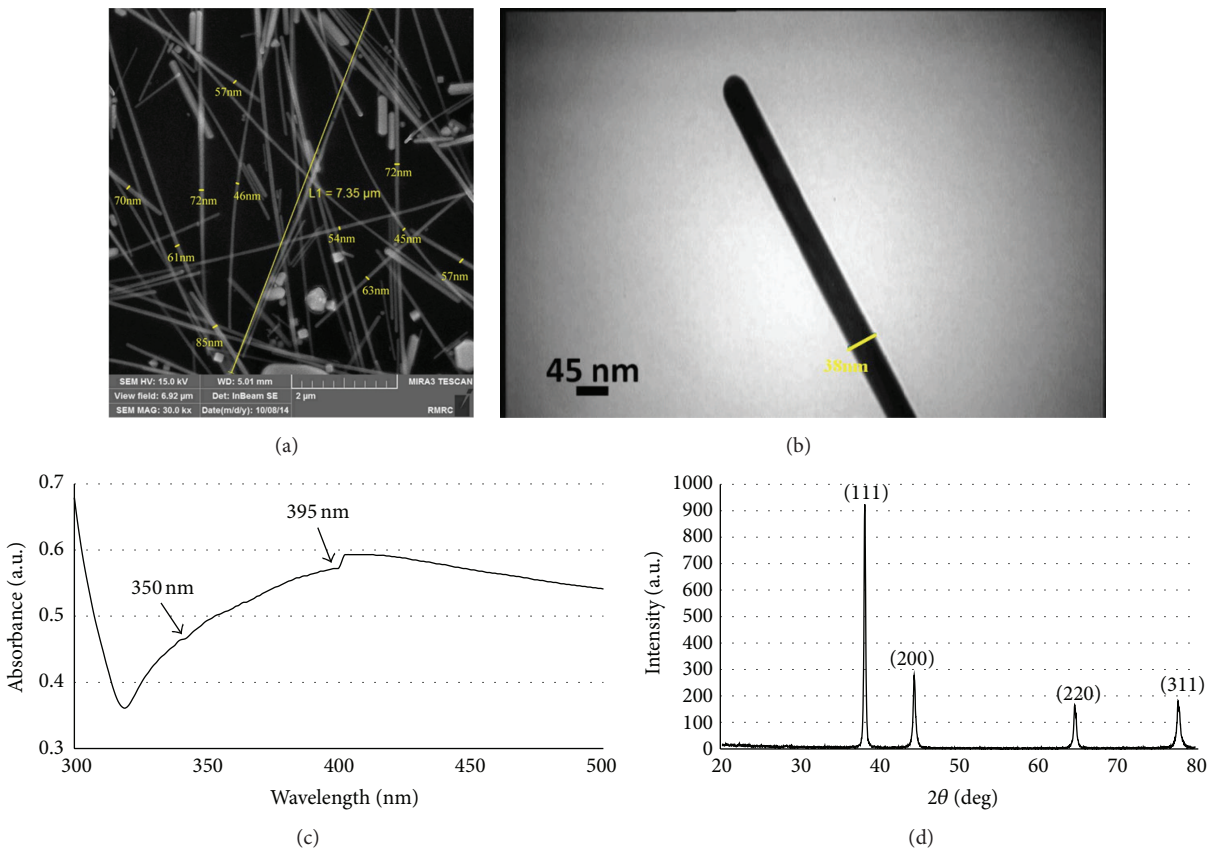

Figure 1: (a) FESEM of AgNWs synthesized without mediated agent. (b) TEM of AgNWs synthesized without mediated agent. (c) UV-Vis absorption spectra of AgNWs synthesized without mediated agent. (d) XRD patterns of AgNWs synthesized without mediated agent.

3. Results and Discussion

3.1. Characterization of the Synthesized Silver Nanowires

Method A. Figures 1(a) and 1(b) show FESEM and TEM images of AgNWs synthesized without any mediated agents. The images show AgNWs with nonuniform length and diameter. From the FESEM micrograph it can be seen that not all of the AgNPs are transformed into nanowires. TEM micrograph shows a single AgNW with average 64 nm in diameter and 12 μm in length (Figure 1(b)).

Figure 1(d) illustrates X-ray diffraction of AgNWs synthesized without any mediated agents. The peaks at 2θ = 38.13°, 44.30°, 64.45°, and 77.41° correspond to the (111), (200), (220), and (311) crystalline planes, respectively, confirming a face centered cubic (FCC) unit cell for silver nanowires. The lattice constant calculated from the diffraction pattern was 0.4086 nm, which is in agreement with the reported value of silver (JCPDS 04-0783). From XRD data shown in Table 2 it can be concluded that Method A does not lead to the formation of highly crystalline structures. In Table 2, the ratio of intensity between (111) and (200) peaks reveals a relatively high value of 3.25 compared to the theoretical ratio value of 2.5. This high value of ratio indicates the enhancement of {111} crystalline planes in the AgNWs.

From UV-Vis spectroscopy, optical properties can be concluded. Surface plasmon resonance (SPR) of Ag nanostructures highly depends on their morphologies. Different surface plasmon resonance peaks located at different frequencies correspond to Ag nanostructures with various shapes and sizes [2, 9].

Figure 1(c) shows UV-Vis absorption spectra of AgNWs synthesized without any mediated agents. The shoulder observed around 395 nm is an indication of low concentration of AgNWs [5, 19]. This test shows that a mixture of AgNPs and AgNWs are the products of this method. The shoulder at 350 nm could be considered as the optical signature of AgNPs.

Method B. Figures 2(a) and 2(b) show FESEM and TEM images of AgNWs synthesized with $FeCl_2$ as mediated agent. FESEM image shows AgNWs with high yield and relatively uniform average diameter equal to 88 nm. TEM image shows AgNWs with 98 nm in diameter and 12 μm in length (Figure 2(b)).

Figure 2(d) shows X-ray diffraction pattern of highly crystalline AgNWs synthesized with $FeCl_2$ as mediated agent. XRD pattern reveals that the synthesis of AgNWs through polyol process with $FeCl_2$ as mediated agent leads to the

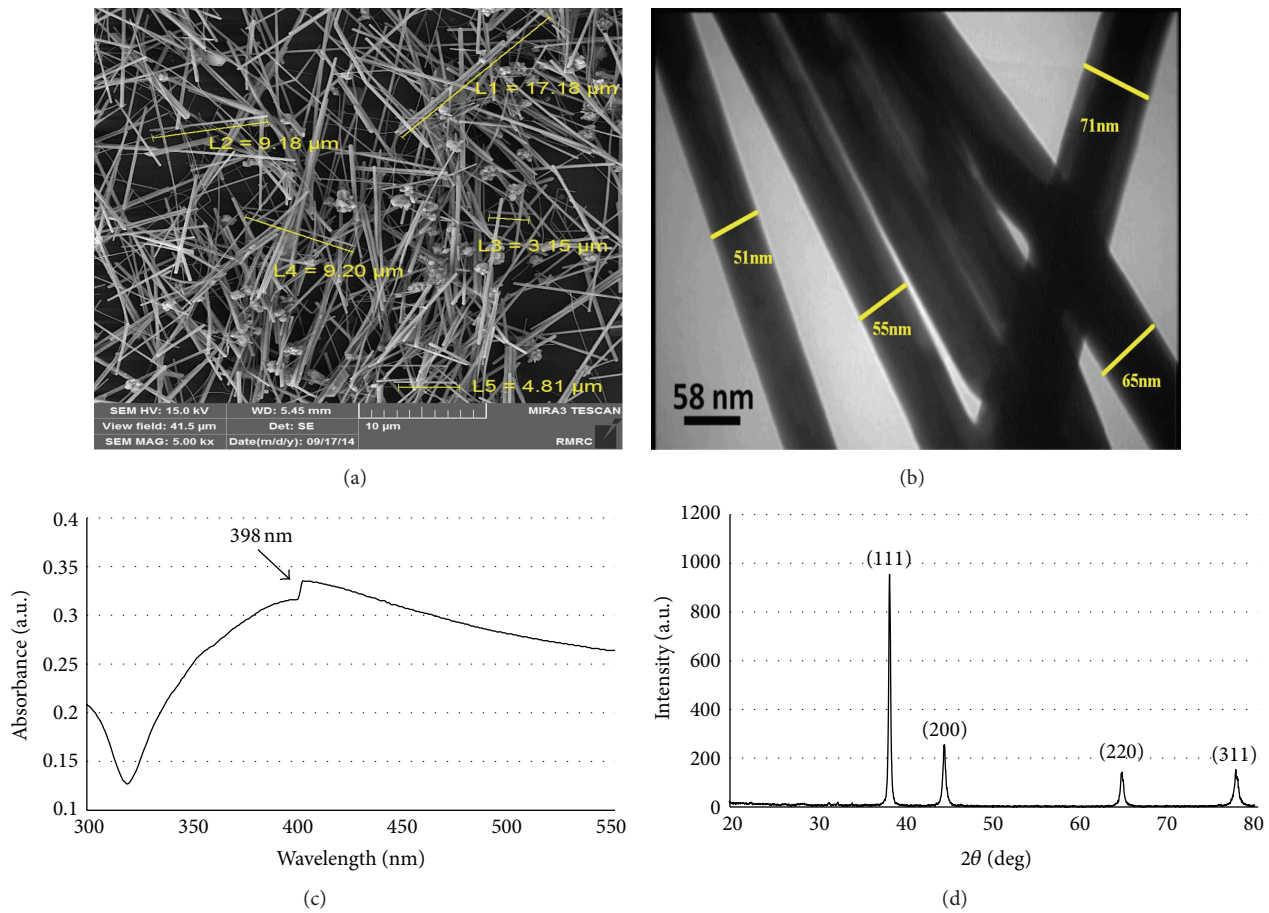

FIGURE 2: (a) FESEM of AgNWs synthesized with $FeCl_2$ mediated agent. (b) TEM of AgNWs synthesis with $FeCl_2$ mediated agent. (c) UV-Vis absorption spectra of AgNWs synthesis with $FeCl_2$ mediated agent. (d) XRD patterns of AgNWs synthesis with $FeCl_2$ mediated agent.

formation of relatively uniform nanowires. From Table 2, the ratio of intensity between (111) and (200) peaks reveals a relatively high value of 3.8. It is obvious that using of $FeCl_2$ as mediated agent affected ratio of intensity between panels and it shows the crystallinity in this method increased.

Figure 2(c) shows the UV-Vis absorption spectra of AgNWs with $FeCl_2$ as mediated agent. The weak peak positioned at 398 nm could be considered as the optical signature of relatively long AgNWs [5, 19]. No peak for AgNPs was observed, indicating the formation of pure AgNWs without any particles.

Method C. Figures 3(a) and 3(b) show FESEM images of AgNWs synthesized at two different concentrations of $AlCl_3$ as mediated agent. From these images it could be observed that the introduction of $AlCl_3$ into the solution greatly helped the formation of silver nanowires. Figure 3(c) shows high yield and uniformity of formed AgNWs. TEM image (Figure 3(c)) shows single AgNWs with average 230 nm in diameter and 20 μm in length. Consequently, it was observed that, after reaching a special length, AgNWs bent and they grew with a different angle. In addition, by increasing the concentration of $AlCl_3$ from 80 to 100 μL, a few square and triangular shaped silver nanoparticles could be observed (Figure 3(b)).

Figure 3(e) illustrates the X-ray diffraction graph of the sample synthesized with $AlCl_3$ as mediated agent. Diffraction peaks were observed and indexed to the (111), (200), (220), and (311) planes of the pure face centered cubic (FCC) with 99.9% crystallinity of AgNWs. The lattice constant was calculated, $a = 4.0862$ Å, JCPDS File 04-0783. Furthermore, it is noteworthy that the intensity ratio of the (111) to (200) peaks is 4 (Table 2), which is higher than other methods, clearly indicating the enrichment of {111} crystalline planes in the silver nanowires [5, 19].

In Figure 3(d), from UV-Vis spectroscopy, a peak around 398 nm appears, indicating the formation of silver nanowires with high purity [19]. In fact the absorbance in the 398 nm position is about 1 (a.u.). This absorption is less than other methods indicating that the diameter of Ag nanowires in Method C is more than other methods.

Method D. Figures 4(a) and 4(b) show FESEM and TEM images of AgNWs synthesized with comediated agents: EG/1,2-propandiol plus $FeCl_3$ mediated agent. It is clearly shown that AgNWs with a length of up to 7 μm and diameter

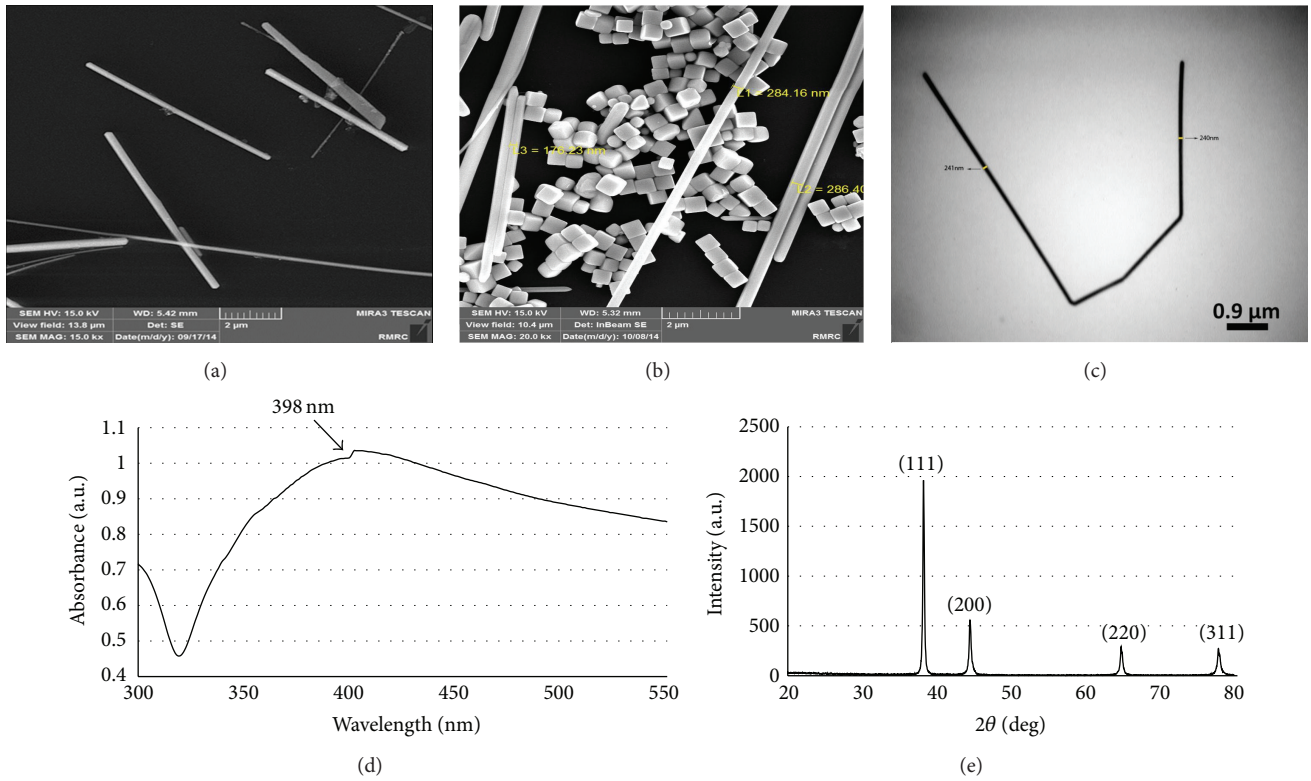

FIGURE 3: (a) FESEM of AgNWs synthesis with 80 μL $AlCl_3$ mediated agent at magnification of 15000. (b) FESEM of AgNWs synthesis with 100 μL $AlCl_3$ mediated agent at magnification of 20000. (c) TEM of AgNWs synthesis with $AlCl_3$ mediated agent. (d) UV-Vis absorption spectra of AgNWs synthesis with $AlCl_3$ mediated agent. (e) XRD patterns of AgNWs synthesis with $AlCl_3$ mediated agent.

around 60 nm have been created with high efficiency. By increasing the volume ratio of EG to 1,2-propandiol compared to increasing the amount of $FeCl_3$, thinner AgNWs with diameters around 52 nm were obtained (Figure 4(c) $V1 : V2 = 5 : 15$). However, some AgNPs and nanowires with surface roughness were observed (Figure 4(c)).

Figure 4(e) shows the typical XRD pattern of AgNWs. The diffraction peaks existing at 38.00°, 44.71°, 64.34°, and 77.32° are indexed as (111), (200), (220), and (311) being consistent well with a face centered cubic (FCC) Ag crystalline structure (lattice constants $a = b = c = 0.4086$ nm, JCPDS 04-0783). No peaks for other crystal types were observed. In general, from XRD pattern it can be concluded that multiple crystalline structures can be obtained through this method [5]. According to our experience, slight differences were observed between lattice parameters of all these four synthesis methods. The ratio of intensity between (111) and (200) in this method was calculated in 2.8. It indicated that crystallinity relative to other methods decreased.

In Figure 4(d), illustrating the UV-Vis spectra of AgNWs, characteristic absorption peaks at 350 nm could be observed; the peak at 395 nm is an indication of AgNWs with high purity. The weak peak around 350 nm can be a sign of formation of few particles during synthesis [5, 19, 20]. Figure 5 shows the UV spectra obtained for different samples prepared by different methods. As can be seen different behaviors are observed due to differences in morphology of AgNWs.

3.2. The Reaction Mechanism Proposed and Discussions. In Method A, at a high temperature 170°C, at first, 1,2-propandiol is converted to propionaldehyde; then homogeneous nucleation takes place by the reduction of Ag^+ ions to Ag^0 [21]. The AgNPs start to grow through a self-seeding mechanism. Then in the presence of PVP which is a polymeric surfactant that could be chemically adsorbed onto the surface of Ag seeds through O–Ag bonding, AgNPs dispersed homogeneously in the solution. As the reaction developed, the small AgNPs were no longer stable in the solution, and they started to dissolve and it contributed to the formation of AgNWs [19, 21].

In Method B, ethylene glycol is converted to glycolaldehyde and AgNPs are formed by the reduction of Ag^+ ions with the help of glycolaldehyde. High Cl^- concentration caused by the decomposition reaction of $FeCl_2$ used as mediated agent reduces the concentration of free Ag^+ ions in the solution through the formation of AgCl and electrostatically stabilizing the initially formed Ag seeds. On the other hand, this phenomenon leads to the slow release of the Ag^+ ions and affects the subsequent reaction and it facilitates the high-yield formation of the thermodynamically more stable multiple twinned Ag seeds that are required for the wire length growth. In addition, Fe^{2+} ions are reduced to Fe^+ ions by EG, which in turn reacts with and scavenges adsorbed atomic oxygen from the surface of AgNPs. Thereupon, Fe^{2+} can remove oxygen from the solvent which prevents twinned seeds dissolved by

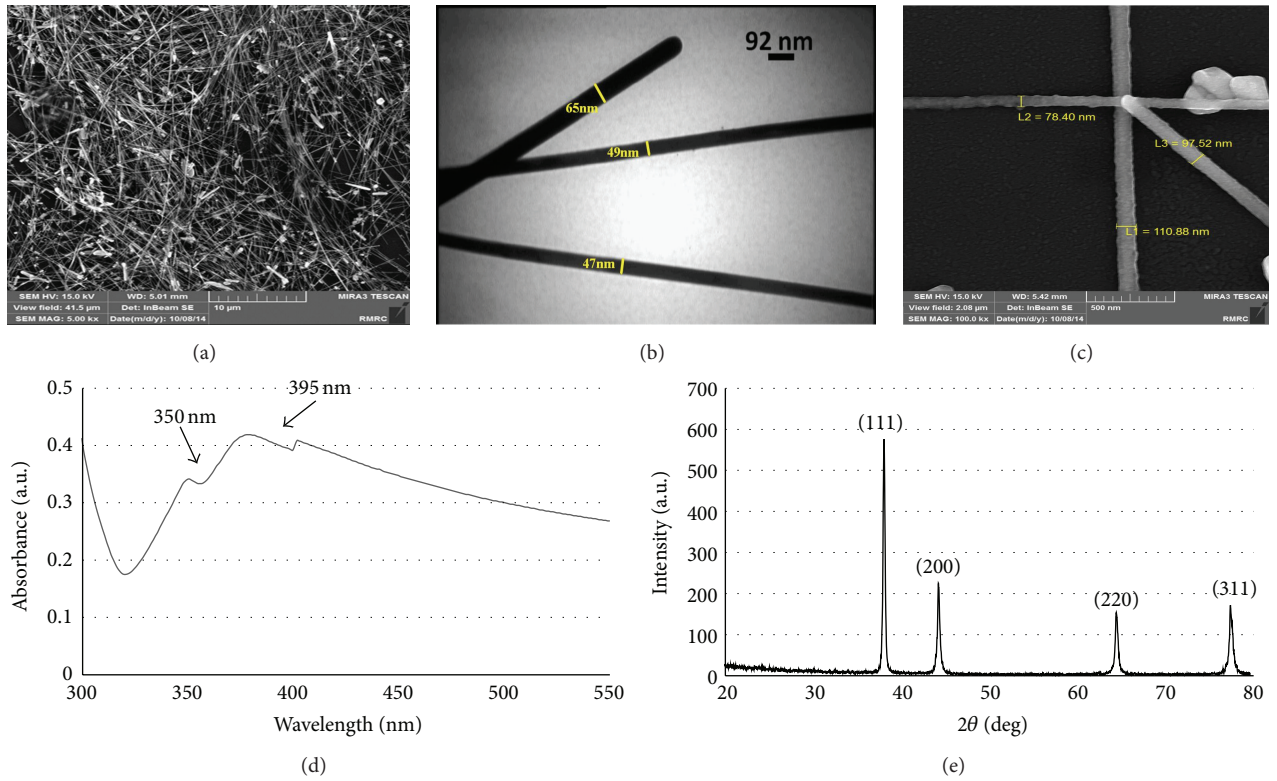

FIGURE 4: (a) FESEM of AgNWs synthesis with EG/1,2-propandiol mediated and $FeCl_3$ as second mediated agent at magnification of 5000. (b) TEM of AgNWs synthesis with EG/1,2-propandiol mediated and $FeCl_3$ as second mediated agent. (c) FESEM of AgNWs synthesis with EG/1,2-propandiol mediated and $FeCl_3$ as second mediated agent at magnification of 100000. (d) UV-Vis absorption spectra of AgNWs synthesis with EG/1,2-propandiol mediated and $FeCl_3$ as second mediated agent. (e) XRD patterns of AgNWs synthesis with EG/1,2-propandiol mediated and $FeCl_3$ as second mediated agent.

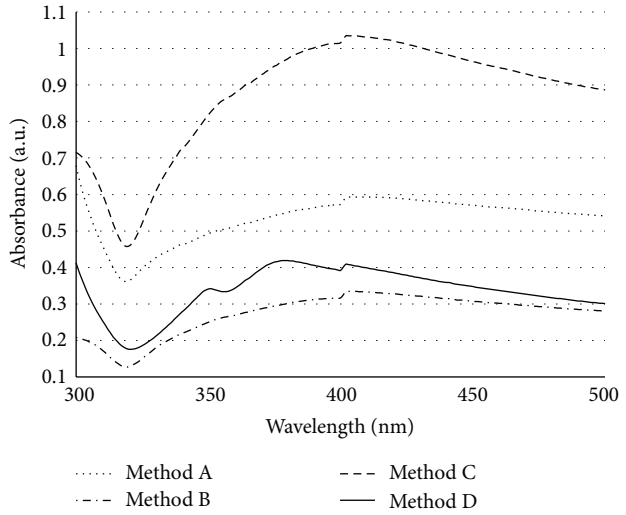

FIGURE 5: UV-Vis spectra of AgNWs prepared by different methods.

oxidative etching and scavenging adsorbed atomic oxygen from the surface of the Ag seeds, facilitating multiple twinned seeds growth to AgNWs [17]. Finally, $AgNO_3$ and PVP were added to the solution, followed by the nucleation and the growth of silver nanowires. When multiple twinned seeds formed the nanoparticles during nucleation, the PVP was bonded by crystalline planes {100} rather than {111} facets, so the growth took place only in the crystalline planes in the [110] direction resulting in the fivefold twinned AgNWs [15].

In Method C, the introduction of $AlCl_3$ into the solution leads to different characteristics of AgNWs in comparison with other methods discussed in this paper. The nucleation and the growth mechanisms are the same as in Method B. The reduction of free Ag^+ ions and the formation of silver seeds are both very slow. However, the slow rate of this reaction may be suitable for the formation of more thermodynamically stable multiple twinned silver seeds required for AgNWs growth [22]. In addition, the number of free Ag^+ ions in the solution decreases as they reduce to Ag^0 by EG. Under these conditions, multiple twinned silver seeds can grow into nanowires with the assistance of PVP during the Ostwald ripening process [23]. The role of PVP is obviously important in the synthesis of AgNWs because of the strong interaction between the oxygen atom of the carboxyl (C=O) group of PVP and the Ag core [24, 25]. Figures 3(a) and 3(c) show uniform AgNWs obtained in the presence of $AlCl_3$. It has been observed that $AlCl_3$ does not tend to release Al_3 ions in low temperatures, but by increasing the temperature (185–190°C), Al_3 can remove all oxygen atoms from the solution

Table 1: Average data of final product of AgNWs in all methods.

Method	Temperature (°C)	Reaction time (mine)	Diameter (nm)	Coefficient of variance (CV)	Diameter (standard division) SD	Length (μm)	Coefficient of variance (CV)	Length (standard division) SD
A	170	90	64	0.16	10.62	12	0.18	1.5
B	155	60	88	0.25	17.97	12	0.24	1.9
C	180	120	180–230	0.19	45.93	20	0.26	5.4
D	170	30	40–60	0.27	17.64	7–10	0.13	1.1

Table 2: The lattice constant was calculated for AgNWs prepared by different methods.

Method	(111) 2θ	(200) 2θ	(220) 2θ	(311) 2θ	Lattice nm	Ratio of intensity between (111) & (200)
A	38.13	44.30	64.45	77.41	0.4077	3.25
B	37.64	43.21	64.41	77.35	0.4085	3.8
C	38.12	44.21	64.11	77.12	0.4074	4
D	38	44.71	64.34	77.32	0.4080	2.8

and help in the growth of AgNWs. In addition, by the use of $AlCl_3$ it was observed that the growth angels of crystalline AgNWs changed during the growth (Figure 3(c)). It is also concluded that there is the possibility of joining two AgNWs while growing as a cause of achieving bent AgNWs. It is also probable that Cl^- ions surrounding the AgNWs led to the formation of bent AgNWs because of the strong ionic forces caused by the presence of $AlCl_3$. The other possibility could be the effect of mechanical stress caused by the stirrer on the crystalline structure during the growth of AgNWs.

Some studies have reported V-shaped and Z-shaped patterns for silver nanorods and nanowires. Jia et al. [15] observed that a single silver nanowire possesses several V-shaped structures with different bending angles. The bending angles were in the range of 100°–150°. Gao and coworkers [25] reported observing Z-shaped wires with bending angles 125° and concluded that they were joined, not bent, wires because of the high surface energy present at this location. Liu et al. [26] reported achieving V-shaped nanorods with bending angles in the range of 110°–150°.

In Method D, EG and 1,2-propandiol have been solution mixed and the volume ratio of EG to 1,2-propandiol is as follows: $V1 : V2 = 3 : 15$. At 170°C the formation of uniform AgNWs may be attributed to the synergetic effect between EG and 1,2-propandiol. Using two kinds of solvents as a mediated agent and $FeCl_3$ as a mediated and nucleation agent is indispensable for the synthesis of high quality AgNWs. The chemical properties of polyol greatly depend on the number of hydroxyl groups and the length of carbon chains. Therefore, 1,2-propandiol with two primary hydroxyl groups has a more powerful reduction ability compared to EG. The viscosity of the EG and 1,2-propandiol solution decreased less than the viscosity of EG solution, by increasing the reaction temperature. Therefore, proper viscosity and the provided electrostatic stability from Cl^- ions lead to a highly stable solution and this controls the growth and morphology of AgNWs by lessening the adding rate of Ag^0 as a result of the slow movement of Ag^+ ions reduced by EG and 1,2-propandiol. Furthermore, sufficient multiple crystalline seeds could be prepared at an initial reaction stage with Cl^-. Fe(III) can be reduced to Fe(II) by mixed solution and can react with and remove the adsorbed atomic oxygen from silver surfaces [25–27]. In addition, Ag seeds are selectively adsorbed by PVP molecules and multiple crystalline seeds grow faster than single crystalline seeds [5]. Consequently, reaction time in this method is less than other methods. The reaction time of each method depends on the mediated agents used in the process and the index was color changed. Finally, in this condition AgNWs with high quality and yield were created by using a mixed solution and $FeCl_3$ as mediated agent. However, by increasing the volume ratio of EG to 1,2-propandiol ($V1 : V2 = 5 : 15$) more AgNPs were formed and surface roughness of AgNWs was increased (Figure 4(c)). Since all chemicals used in this work were 99.9% pure, surface roughness of AgNWs is not caused by the impurity of the chemical substances. Consequently, it is possible that the high concentration of Cl^- ions released from the decomposition of $FeCl_3$ on the surface of Ag particles and wires leads to high electrostatic stability and this electrostatic energy of Cl^- ions could be the cause of the surface roughness of AgNWs. Table 1 shows the results of synthesized AgNWs by different mediated agents.

4. Conclusion

Silver nanowires were synthesized and characterized through polyol process by using different mediated agents. It is discovered that the addition of $FeCl_2$ and $AlCl_3$ greatly facilitates the formation of silver nanowires. For the first time we report that the use of $AlCl_3$ changes the growth angle of the wires during the formation of AgNWs. However, both cations and anions are necessary for the production of AgNWs with high yield. For controlling the growth and nucleation, Cl^- ions in the reaction provide electrostatic stabilization and they combine with Ag^+ ions to form AgCl colloids in the initial stage of formation of silver seeds. Here, multiple crystalline AgNWs were successfully fabricated by mixed solution EG and 1,2-propandiol comediated agent and $FeCl_3$ as second mediated agent in the presence of PVP molecules. The rate of chemical reaction greatly depended on the viscosity and reduction ability of solvents. The novelty of this study is to prepare uniform AgNWs with high efficiency, employing EG and 1,2-propandiol as cosolvent and $FeCl_3$ as mediated

agent, simultaneously. This paper provides an idea of how to grow silver nanowires for electronic device and antibacterial properties. Stretchable and wearable electrochromic devices, elastomer nanocomposites, highly conductive flexible paper, and flexible touch panels are some applications of AgNWs. Also, AgNWs can be used to produce antibacterial textiles for medical application.

Competing Interests

The authors declare that they have no competing interests.

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