Effects of aging conditions on the morphologies of ZnO particles synthesized under hydrothermal conditions from layered zinc hydroxide as a precursor derived from zinc acetate

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ABSTRACT
Zinc oxide is a useful material for various products. ZnO crystals usually grow in the c-axis direction, on the less exposed (001) plane. However, the (001) plane has high chemical reactivity, and plate-like ZnO crystals on large (001) planes have superior UV-lasing and other properties. In this study, layered zinc hydroxide acetate (ZHA), a precursor of ZnO, was aged in deionized water and Zn(OCOCH$_3$)$_2$ or CH$_3$COONa aqueous solutions of various concentrations under hydrothermal conditions at 90 and 120°C. Aging at above 90°C yielded ZnO in all the solutions, and the concentrations of the CH$_3$COONa aqueous solutions did not affect the form of the ZnO particles. On the other hand, when ZHA was aged in Zn(OCOCH$_3$)$_2$ aqueous solution, the crystal growth of ZnO in the c-axis direction was suppressed as the Zn(OCOCH$_3$)$_2$ concentration increased. In the case of aging in 1 molL$^{-1}$ Zn(OCOCH$_3$)$_2$ aqueous solution, plate-like ZnO was obtained, but the yield was about 10% and a large amount of ZHA remained. When the obtained mixture of plate-like ZnO and ZHA was aged again in deionized water at 90 and 120°C after washing, most of the ZHA was transformed into plate-like ZnO after aging for over 72 hours.

1. Introduction
Zinc oxide is a typical metal oxide semiconductor with a wide direct band-gap of about 3.37 eV [1]. ZnO is widely used in varistor elements, gas sensors, etc., due to its unique optical, electrical and piezoelectric properties [2–4]. In addition, ZnO is used in cosmetics for the purpose of protecting against ultraviolet rays, since it has low toxicity to the human body and absorbs ultraviolet light [5–7].

It is well known that ZnO particles have different properties depending on their morphology, e.g. particle size, anisotropy, etc. Thus, controlling the shape of ZnO particles is an effective way to control the properties of ZnO powders [8,9]. Of special note, it is reported that plate-like ZnO with an exposed (001) plane has more effective gas sensitivity and higher photocatalytic activity than ZnO in other shapes, such as rod-like and pyramid shapes [9]. Also, plate-like ZnO with a wide (001) plane has superior UV-lasing properties than rod-like ZnO [10]. For the above reasons, synthesizing plate-like ZnO leads to improvement of the properties of ZnO as a material for use in a variety of industrial fields.

The ZnO crystal structure can be described as a number of alternating planes composed of O$^2-$ and Zn$^{2+}$ ions stacked alternatively along the c-axis. From its crystal structure, hexagonal columnar crystals can be obtained by growing a nonpolar plane separate from the (001) plane with high surface energy [9,11]. Several studies have reported that the growth environment of ZnO affects the growing direction and morphology of the obtained ZnO [8,9,11].

ZnO synthesis methods include the sol-gel method, precipitation method and others [12–14]. These synthesis methods require complex processes or high temperature. By comparison, ZnO can be synthesized easily under hydrothermal conditions with mild reaction processes, and even at below 100°C [15,16].

Layered zinc compounds (formula: $\text{Zn}_3(\text{OH})_{10-x} (A^{m+})_{x/m}n\text{H}_2\text{O}(A:\text{Anion})$) belong to the family of metal hydroxide salts (MHS) with the general formula $M_y(\text{OH})_b(A^{c+})_{(2a-b)k}n\text{H}_2\text{O}(A:\text{Anion})$, where $M = \text{Zn}^{2+}$, $\text{Co}^{2+}$, $\text{Ni}^{2+}$, $\text{Mn}^{2+}$, etc. MHS consists of positively charged brucite-like layers of metal hydroxides that require the presence of interlayer anions to maintain their overall charge neutrality. These interlayer anions can be Cl$^-$, $\text{CO}_3^{2-}$, $\text{CH}_3\text{COO}^-$, or $\text{SO}_4^{2-}$ [17]. Layered zinc hydroxide acetate (ZHA) is synthesized by direct precipitation by adding sodium hydroxide solution to Zn(OCOCH$_3$)$_2$ solution, or by stirring ZnO powder with zinc acetate in solution [18–20], etc. Besides being studied as an ion-exchange application in catalysis [18], layered zinc hydroxide is often studied as a precursor of...
ZnO. Layered zinc hydroxides can provide ZnO powders through dissolution/reprecipitation processes under wet conditions [21].

In this study, conducted to determine a simpler process for obtaining hexagonal plate-like ZnO particles, ZHA was synthesized through a reaction of ZnO powder with Zn(OCOCH$_3$)$_2$ solutions and aged under hydrothermal conditions in various solutions. The effects of the type and concentration of the electrolyte used during aging in aqueous solutions and the aging temperature and time on the c-axis growth of the obtained ZnO were investigated. The obtained ZnO powder was observed by SEM, and the influence of the aqueous solution conditions during aging on the morphology of ZnO was examined.

2. Materials and methods

2.1. Synthesis of ZHA

Zn(OCOCH$_3$)$_2$·2H$_2$O (Guaranteed Reagent, FUJIFILM Wako Pure Chemical Co.) was dissolved in deionized water to prepare a 1 mol L$^{-1}$ raw material solution.

5 g of ZnO (Sakai Chemical Co., Ltd.) was added to 150 mL of the raw material solution and stirred with a magnetic stirrer for 24 hours. After stirring, the product was entered into a centrifuge (H-200 n · Kokusan Co., Ltd.) operating at a speed of 10,000 rpm for 20 minutes to perform solid-liquid separation, after which the precipitates were centrifuged at 10,000 rpm for 20 minutes, decanted, and washed with deionized water. This procedure was repeated three times.

2.2. Hydrothermal aging of ZHA

Precipitations dispersed in 150 mL of deionized water and Zn(OCOCH$_3$)$_2$ and CH$_3$COONa aqueous solutions of specified concentrations (1, 0.1, 0.01 and 0.001 mol L$^{-1}$) were enclosed in Teflon-lined autoclaves and aged at 60°C, 90°C and 120°C for 24 hours. After aging, the autoclaves were allowed to cool at room temperature for 24 hours. Furthermore, precipitates aged in 150 mL of 1 mol L$^{-1}$ Zn(OCOCH$_3$)$_2$ solutions at 90°C and 120°C for 24 hours and washed, were aged again in 150 mL of deionized water at 90°C and 120°C for 24, 72 and 120 hours and cooled. The precipitates were centrifuged at 10,000 rpm for 20 minutes, decanted, and washed with deionized water. This procedure was repeated three times. After freeze-drying, the final products were obtained as powders.

The crystalline phases of the ZHA were identified by powder X-ray diffraction analysis (XRD) (RINT2000, Rigaku Co.) with the following parameters: Cu Kα radiation, tube power 1.6 kW (40 kV at 40 mA), scan speed 4° min$^{-1}$, and scan angle 5°–65°. The morphologies of the ZHA precipitates were investigated with a field emission scanning electron microscope (S-800 Hitachi, LTD) employing an accelerating voltage of 20 kV with a 6–20 μm aperture. The particle size distributions were assessed using a laser diffraction particle size analyzer (Partica LA-950V2, HORIBA). Thermogravimetric-differential analyses were conducted using a Thermoplus Tg8120 (Rigaku Co.) instrument with a heating rate of 10°C min$^{-1}$ in an air atmosphere.

3. Results and discussion

3.1. Synthesis of ZHAs

Figure 1 shows powder XRD patterns of ZnO as starter seed crystals and precipitates obtained from stirring ZnO in 1 mol L$^{-1}$ Zn(OCOCH$_3$)$_2$ solution for 24 hours.

As shown in Figure 1, ZnO seed crystals have only the diffraction peaks of ZnO, and the precipitates have no peaks of ZnO, but only those of ZHA. These results show that most ZnO stirred in 1 mol L$^{-1}$ Zn(OCOCH$_3$)$_2$ solutions have reacted to synthesize ZHA.

The pyrolysis processes of ZHA were examined by the TG and DTA curves shown in Figure 2.

Considering the TG curves, the two weight losses at 70–150°C suggest that the water molecules were physically adsorbed on ZHA and that the intercalating crystal water molecules were desorbed [19–21]. The endothermic process at 60–90°C is associated with about 8% weight loss, and the process at 110–150°C is associated with approximately 10% weight loss. This combined weight loss of 17–18% is close to the theoretical weight loss (17.5%) ascribed to the release of six water molecules from the formula unit, and the two associated reactions can be described by reaction formula (1) and (2) [18,20]:

\[
\begin{align*}
\text{Zn}_5(\text{OH})_8(\text{OCOCH}_3)_2 \cdot 2\text{H}_2\text{O} & \rightarrow \text{Zn}_5(\text{OH})_8(\text{OCOCH}_3)_2 + 2\text{H}_2\text{O} \\
\text{Zn}_5(\text{OH})_8(\text{OCOCH}_3)_2 & \rightarrow 4\text{ZnO} + \text{Zn}(\text{OCOCH}_3)_2 + 4\text{H}_2\text{O}
\end{align*}
\]

The thermal decomposition of Zn(OCOCH$_3$)$_2$ occurs at over 147°C, but the processes involved after reaction (2) are not straightforward. The decomposition of Zn(OCOCH$_3$)$_2$ upon heating is sensitive to the humidity of the surrounding atmosphere [18,20].

In this study, the weight loss observed by TG between 147 and 200°C can be mainly assigned to the processes that form ZnO through reaction (3). In a high-humidity atmosphere, however, Zn(OCOCH$_3$)$_2$
can react with H₂O to form ZnO and acetic acid, as shown by reaction formula (4):

$$Zn(O_{2}COCH_{3})_{2} \rightarrow ZnO + (CH_{3}CO)_{2}O \quad (3)$$

$$Zn(O_{2}COCH_{3})_{2} + H_{2}O \rightarrow ZnO + 2CH_{3}COOH \quad (4)$$

$Zn(O_{2}COCH_{3})_{2}$ can also be converted to $Zn_{4}O_{6}(O_{2}COCH_{3})_{6}$ and acetone at above 250°C in dry atmospheres, and $Zn_{4}O_{6}(O_{2}COCH_{3})_{6}$ may subsequently decompose in ZnO and acetate according to reaction (5):

$$Zn_{4}O_{6}(O_{2}COCH_{3})_{6} \rightarrow 4 ZnO + 3CH_{3}COCH_{3} + 3CO_{2} \quad (5)$$

The theoretical weight loss rate of ZHA is 38% due to the above pyrolysis reaction [20], and that of the obtained precipitates is approximately 38%. The above results suggest that nearly pure ZHA can be obtained by stirring ZnO in a $Zn(O_{2}COCH_{3})_{2}$ solution.

$ZnO$ and ZHA crystals can be observed in Figure 3. Further, Figure 4 shows their particle size distribution curves. The ZnO used as a raw material is in the form of granular crystals ranging from 0.1–10 µm, and ZHA synthesized using the ZnO in $Zn(O_{2}COCH_{3})_{2}$ solutions forms needlelike crystals in the range of 4–300 µm.

**Figure 1.** X-ray diffraction patterns of ZnO (a) as start, seed crystals and precipitates (b) from stirring ZnO in 1 mol/L $Zn(O_{2}COCH_{3})_{2}$ solutions for 24 hours.
In this study, ZHA synthesized by stirring ZnO in Zn(OCOCH$_3$)$_2$ solutions was used as the precursor for ZnO.

### 3.2. Aging ZHA in deionized water

Figure 5 shows powder XRD patterns of ZHA aged at 60, 90 and 120°C for 24 hours in 150 mL of deionized water. As shown in Figure 5, diffraction peaks of both ZHA and ZnO were observed at 60°C. On the other hand, only diffraction peaks of ZnO were observed at 90°C and 120°C. Therefore, it was found that at 90°C or higher, all the ZHA was used to form ZnO. Also, at any aging temperature, the (001) ZnO diffraction peak is weaker in the direction perpendicular to the c-axis compared with the other peaks.

Figure 6 shows SEM images of ZHA aged for 24 hours in 150 mL of deionized water at 60, 90 and 120°C. Hexagonal columnar crystals can be observed in the precipitation obtained at any aging temperature. At above 90°C, the crystals have clear hexagonal shapes, but at 60°C...
C, slightly finer crystals are observed. These hexagonal columnar crystals are probably ZnO, as suggested by contrast with the SEM observations and XRD patterns of precipitates obtained at any aging temperature (Figure 5). Similarly, the fine crystals observed at 60°C seem to be unreacted ZHA. The aspect ratios of ZnO produced by aging at 60, 90 and 120°C were 2.23, 1.71 and 1.39, respectively, and aging at higher temperatures therefore resulted in shorter crystals. This is probably due to the high number of nuclei generated at high temperatures.
Figure 4. PSD frequency curves of ZnO (a) as start, seed crystals and precipitates (b) from stirring ZnO in 1 mol/L Zn(OCOCH$_3$)$_2$ solutions for 24 hours.

Figure 5. X-ray diffraction patterns of ZHAs aged at 60, 90 and 120°C in 150 mL of deionized water for 24 hours.
3.3. Aging of ZHA in various solutions

Figure 7 shows powder XRD patterns of ZHA aged at 120°C for 24 hours in Zn(OCOCH$_3$)$_2$ and CH$_3$COONa aqueous solutions of 1, 0.1, 0.01 and 0.001 molL$^{-1}$. In both solutions, the diffraction peaks of both ZHA and ZnO are observed at 1 mol L$^{-1}$, while only the diffraction peak of ZnO is observed at a concentration lower than 0.1 molL$^{-1}$. The diffraction peak of (001) was highest for ZHA aged in Zn(OCOCH$_3$)$_2$ aqueous solutions at 1 mol L$^{-1}$, and the (001) diffraction peak was lowest at below 0.1 molL$^{-1}$. In CH$_3$COONa aqueous solutions, the (001) diffraction peaks were lower than the (100) diffraction peaks at any concentration. Also, in a 1 molL$^{-1}$ Zn(OCOCH$_3$)$_2$ aqueous solution, the intensities of the ZnO peaks were lower than those in 1 molL$^{-1}$ CH$_3$COONa aqueous solution. ZHA aged in a 1 molL$^{-1}$ Zn(OCOCH$_3$)$_2$ aqueous solution had a peak on the high angle side nearby a ZHA peak. This is probably due to variations in the ZHA interlayer distance caused by an exchange of part of interlayer CH$_3$COO$^-$ and OH$^-$.

Figure 8 shows the TG and DTA curves of ZHA aged in 1, 0.1, 0.01 and 0.001 molL$^{-1}$ Zn(OCOCH$_3$)$_2$ and CH$_3$COONa aqueous solutions at 120°C for 24 hours. Weight loss due to the thermal decomposition of ZHA was observed only at 1 mol L$^{-1}$. The ZHA, on the other hand, including a large quantity of ZnO such as powders obtained by aging at 120°C in Zn(OCOCH$_3$)$_2$ and CH$_3$COONa solutions in concentrations below 0.1 molL$^{-1}$, showed no or only very small endothermic peaks and much lower weight loss than the theoretical weight loss of ZHA (about 38%) [19,20]. This is probably because ZHA tends to agglomerate in high concentration electrolyte solutions, and it is difficult to dissolve ZHA to form ZnO [22]. Also, the weight loss of ZHA aged in CH$_3$COONa solution was much lower than in the case of aging in Zn(OCOCH$_3$)$_2$. The pH of a 1 molL$^{-1}$ Zn(OCOCH$_3$)$_2$ aqueous solution before aging was 6.22 and that of CH$_3$COONa aqueous solution was 8.53. In the CH$_3$COONa aqueous solution, therefore, more ZHA was dissolved and more ZnO generated due to the high pH [18]. Also, in Zn(OCOCH$_3$)$_2$ solutions of higher concentrations containing large amounts of Zn$^{2+}$, ZHA is probably difficult to dissolve and to convert to ZnO.

At 1 molL$^{-1}$ Zn(OCOCH$_3$)$_2$ solution, a weight loss specific to ZHA was observed, and the weight loss was about 33% at 120°C. Thus, a calculation based on the theoretical weight loss of ZHA (38%) shows that the proportion of ZnO in the sample was only about 13% at 120°C.

Figure 9 shows SEM images of ZHA aged at 120°C for 24 hours in 1, 0.1, 0.01 and 0.001 molL$^{-1}$
Figure 7. X-ray diffraction patterns of ZHAs aged in 1(1), 0.1(2), 0.01(3) and 0.001(4) mol\text{L}^{-1} Zn(CH_3COO)_2 (a) or CH_3COONa (b) solutions at 120°C for 24 hours.

Figure 8. TG and DTA curves of ZHAs aged in 1(1), 0.1(2), 0.01(3) and 0.001(4) mol\text{L}^{-1} Zn(CH_3COO)_2 (a) or CH_3COONa (b) solutions at 120°C for 24 hours.
When aging was conducted in 1 mol L\(^{-1}\) Zn(OCOCH\(_3\))\(_2\) aqueous solutions, plate-like crystals of ZnO and ZHA were observed. When the Zn(OCOCH\(_3\))\(_2\) concentration was lower than 0.1 mol L\(^{-1}\), hexagonal columnar crystals were observed, and the lower the concentration, the longer the rod-like crystals observed in the c-axis direction became. In CH\(_3\)COONa solution, rod-like crystals were observed at any concentration, much as in deionized water. In the results in Figure 7, both the plate-like and rod-like crystals are ZnO. From the above results, it was concluded that concentrations of CH\(_3\)COO\(^-\) and Na\(^+\) have little effect on the morphology of ZnO particles. It was considered, moreover, that the growth of ZnO in the c-axis direction is suppressed as the Zn\(^{2+}\) concentration increases.

Here, in order to obtain ZnO in a higher yield, ZHA aged in a 1 mol L\(^{-1}\) Zn(OCOCH\(_3\))\(_2\) aqueous solution was washed and then aged again in deionized water at 90 and 120°C.
Figure 10 shows powder XRD patterns of ZHA aged in deionized water at 90 and 120°C for 24, 72 and 120 hours after aging in 1 molL$^{-1}$ Zn(OCOCH$_3$)$_2$ aqueous solutions for 24 hours and washing. Figure 11 shows SEM images of this aged ZHA, and Figure 12 shows its PSD frequency curves. In either case, only the ZnO diffraction peaks can be observed. Also, among the diffraction peaks of ZnO, the diffraction peak of the (001) plane is the highest. These results show that the ZHA remaining after the first aging reacted to produce ZnO during the second aging. In the SEM images, large hexagonal plate-like and small crystals were observed at 24 hours. On the other hand, after aging for 72 and 120 hours, the small crystals observed at 24 hours had disappeared and many plate-like crystals were observed. These results suggest that large hexagonal plate-like crystals were generated by utilizing the plate-like ZnO obtained after the first aging as seed crystals. As shown in Figure 12, in either case, the PSD curve shifted to the coarse grain side as the aging time increased. Thus, the small crystals disappeared with a longer aging time. The above results suggest that the small crystals observed at 24 hours were probably dissolved, allowing the large plate-like crystals to grow due to the high solubility and surface energy, a process known as Ostwald ripening [23]. Figure 13 shows the changes in the intensity ratio of the (001) diffraction peak to the (100) diffraction peak with aging time. At any aging temperature, the intensity of the diffraction peak of the (001) plane increased relative to that of the (100) plane with increases in aging time. This results support the fact that, as the aging time lengthens, fine particles disappear and the proportion of plate-like ZnO in the sample increases. Therefore, plate-like ZnO was preferentially obtained in high yields through aging for longer than 72 hours.

4. Conclusions

This work describes a novel method of obtaining plate-like ZnO more conveniently from layered zinc hydroxide as a precursor. Layered zinc hydroxide acetate (ZHA) synthesized from Zn(OCOCH$_3$)$_2$ was aged under hydrothermal conditions in deionized water and Zn(OCOCH$_3$)$_2$ and CH$_3$COONa aqueous solutions, and the influence of the aging conditions on the morphology of the ZnO obtained was investigated. As a result, the following findings were obtained:

1. Nearly pure ZHA with needle-shaped particles was obtained by stirring ZnO with a particle size of 0.6 µm in Zn(OCOCH$_3$)$_2$ solutions at room temperature for 24 hours.
2. ZnO crystals with a rod-like shape, the usual form of ZnO crystals, were synthesized from

![Figure 11 SEM images of ZHAs aged in deionized water at 90(1) and 120(2)°C for 24 (a), 72 (b) and 120 (c) hours after aged in a 1 molL$^{-1}$ Zn(OCOCH$_3$)$_2$ aqueous solution for 24 hours and washed.](image)
ZHA by aging in deionized water or 1, 0.1, 0.01 or 0.001 mol L$^{-1}$ CH$_3$COONa solution at above 90°C for 24 hours. These results indicated that Na$^+$ and CH$_3$COO$^-$ have an insignificant effect on the growth direction of ZnO crystals.

(3) When ZHA was aged in a Zn(OCOCH$_3$)$_2$ solution at above 90°C for 24 hours at higher concentrations, shorter hexagonal columnar ZnO crystals were obtained, and a small number of plate-like ZnO crystals was synthesized at 1 mol L$^{-1}$ above 90°C. Therefore, it can be concluded that the Zn$^{2+}$ in the solution suppressed the growth of ZnO in the c-axis direction.

(4) When both plate-like ZnO and unreacted ZHA obtained after the first aging in a 1 mol/L Zn(OCOCH$_3$)$_2$ aqueous solution were aged again in deionized water at above 90°C for over 24 hours, all the ZHA reacted to generate plate-like or many small ZnO crystals.

After aging for over 72 hours, moreover, only plate-like ZnO crystals were obtained in a high yield. This is
probably because unreacted ZHA and small pieces of ZnO crystals dissolved and grew using the plate-like ZnO crystals synthesized in the first aging as seed crystals.

As described above, it is concluded that two-step aging is an effective process for synthesizing hexagonal plate-like ZnO particles.

**Disclosure statement**

No potential conflict of interest was reported by the authors.

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