Co-precipitation synthesis and sintering of yttrium aluminum garnet (YAG) powders: the effect of precipitant

Ji-Guang Li *, Takayasu Ikegami, Jong-Heun Lee, Toshiyuki Mori, Yoshiyuki Yajima

National Institute for Research in Inorganic Materials, Namiki 1-1,Tsukuba-shi, Ibaraki 305-0044, Japan

Abstract

YAG precursors were co-precipitated from a mixed solution of aluminum and yttrium nitrates using ammonia water and ammonium hydrogen carbonate as precipitants, respectively. Phase evolution of the precursors during calcination and sinterability of the resultant YAG powders were compared between the two methods. The use of ammonia water produced a hydroxide precursor with an approximate composition of $Al(OH)_3 \cdot 0.3[Y_2(OH)_5(NO_3) \cdot 3H_2O]$ which transformed to pure YAG at about 1000°C via $YAlO_3$ phase. Severe agglomeration caused poor sinterability of the resultant YAG powders. The use of ammonium hydrogen carbonate produced a carbonate precursor with an approximate composition of $NH_4AlY_{0.6}(CO_3)_{1.9}(OH)_2 \cdot 0.8H_2O$. The precursor directly converted to pure YAG at about 900°C. The precursor was loosely agglomerated and the resultant YAG powders showed good dispersity and excellent sinterability. For the same calcination temperature of 1100°C, YAG powders from the hydroxide precursor and the carbonate precursor densified to ∼81.2 and ∼99.8% of the theoretical, respectively, by vacuum sintering at 1500°C for 2 h. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Phase evolution; Sintering; YAG; Powders-chemical preparation; $Y_3Al_5O_{12}$

1. Introduction

YAG ($Y_3Al_5O_{12}$) is one of the ceramic materials which can be sintered to translucency or transparency.[1] Compared with alumina, YAG shows better optical and high-temperature mechanical properties. Recent work of Parthasarathy et al.[2] revealed that the creep rate of polycrystalline YAG (3 μm grain size) stressed at 75.5 MPa at 1400°C is only one third of that of the polycrystalline Al_2O_3 (3 μm grain size) tested under equivalent conditions. Besides, unlike alumina, YAG is cubic in structure and does not exhibit any birefringence effects at the grain boundaries, showing better optical properties.

YAG powder was traditionally produced by a solid-state reaction[3–5] between the component oxides which requires repeated mechanical mixing and extensive heat treatment at temperatures as high as 1700°C to eliminate YAM ($Y_4Al_2O_9$) and YAP ($YAlO_3$) intermediate phases. It is well recognized that wet-chemical processing of multi-cation oxides provides considerable advantages of good mixing of the starting materials and excellent chemical homogeneity of the final product. Various wet-chemical methods have been developed and successfully used in recent years for low-temperature production of phase-pure YAG powders. These methods include sol–gel processing,[6–8] hydroxide co-precipitation,[9–17] homogeneous precipitation,[18–20] glycothermal treatment,[21] spray pyrolysis,[22] and combustion synthesis.[23–25] Although sol–gel processing and co-precipitation were widely used for powder synthesis, one main disadvantage of these two methods is that ultrafine particles of the gel-like precursors underwent severe agglomeration during drying, causing poor sinterability of the resultant YAG powders. Though special measures were taken during powder processing to alleviate agglomeration, the problem was not well solved and sinterability of the YAG powders was still not so desirable. Vrolijk et al.[12] reported that the YAG powder produced from hydroxide precursor treated carefully with organic liquids to reduce agglomeration densified to >99% of the theoretical density only after vacuum sintering at 1750°C for 4 h. While Steinmann and De With[8] reported that the YAG powder synthesized by sol–gel of $Al(OC_3H_7^i)_3$ and $Y(OC_3H_7^i)_3$ only sintered to a relative density of 95% at

* Corresponding author. Tel.: +81-0298-51-3354, ext. 2247; fax: +81-0298-52-7449.

E-mail address: jgli@nirim.go.jp (Ji-Guang Li).

1700°C though ball-milling was used to break up hard agglomerates. Recently, Manalert and Rahaman[6] used a hypercritical point drying method to prevent severe agglomeration of the sol-gel derived precursors, and the resultant YAG powder was claimed to densify to nearly full density at 1600°C in oxygen. However, the final sintered material contained an aluminum-rich second phase. In fact, the YAG powders produced by sol-gel or hydroxide co-precipitation are difficult to sinter to full density or translucency without external pressure (hot pressing) or a considerable amount of SiO_2/MgO as sintering aids.[1,13]

Chemical composition as well as physical properties of the precursor have dramatic effects on sinterability of the resultant oxide powders. Previous work showed that ammonium hydrogen carbonate (hereafter referred to as AHC for convenience) exceeds ammonia water for the production of less-agglomerated, well-sinterable alpha-alumina[26] and yttria[27] powders via precipitation. In these cases, the carbonate precursors were loosely agglomerated, and the resultant oxide powders densified to transparency at low-temperatures without sintering aids.

In this work, AHC was used to synthesize YAG powders from a mixed solution of aluminum and yttrium nitrates via co-precipitation. For comparison, ammonia water was also used for powder synthesis under equivalent conditions. Phase evolution of the precursors and sinterability of the resultant YAG powders were investigated and compared between the two methods.

2. Experimental

2.1. Materials

The yttrium and aluminum sources for YAG synthesis were 99.99%$Y(NO_3)_3 \cdot 6H_2O$ and >99% $Al(NO_3)_3 \cdot 9H_2O$, respectively. Cation impurity contents of the aluminum nitrate, as provided by the supplier, were <0.02 wt.% of Na, <0.002 wt.% of K, <5 ppm of Cu, <5 ppm of Pb, and <0.002 wt.% of Fe. As precipitants, AHC was ultrahigh purity and ammonia water (25%) was analytical grade. All these chemicals were purchased from Kanto Chemical Co., Inc. Tokyo, Japan, and were used as received without further purification.

The stock solution of mother salts was made by dissolving aluminum and yttrium nitrates in distilled water. Cation contents of the stock solution were assayed by the ICP (Inductively Coupled Plasma) spectrophotometric technique and further adjusted to meet the YAG stoichiometry. Concentration of the stock solution was 0.15 M for Al^{3+}.

The concentration of AHC solution was expected to affect composition of the resultant precipitate. Previous work[28] revealed that Al^{3+} ions may precipitate as pseudo-boehmite (AlOOH) or ammonium dawsonite

$[NH_4Al(OH)_2CO_3]$ mainly depending on the concentration of AHC solution. To avoid the possible formation of gelatinous AlOOH, concentration of the AHC solution was selected as 1.5 M and was made by dissolving AHC into distilled water. For comparison, ammonia water of 1.5 M was also made by diluting the original one with distilled water.

2.2. Powder synthesis

Precipitation processes were performed on a magnetic stirrer at room temperature. Chemical precipitation can be performed by the normal-strike method (adding precipitant solution to the salt solution) or by the reverse-strike technique (adding salt solution to the precipitant solution). The main difference between the two methods is the rate at which pH of the salt solution changes as a function of time. For multi-cation materials, the latter technique has the advantage of higher cation homogeneity in the precursor,[12] and was used in this study.

For the AHC method, precursor precipitate was produced by adding 200 ml of the salt solution at a speed of 3 ml/min into 320 ml of the AHC solution contained in a beaker under mild agitation. The resultant suspension, after aging 1 h, was filtered using a suction filter, washed four times with distilled water, rinsed with ethyl alcohol (except the samples for chemical analysis), and dried at room temperature with flowing nitrogen gas over 24 h. The dried cake was crushed with a zirconia pestle and mortar and calcined at various temperatures for 1 h under flowing oxygen gas.

Powder synthesis using ammonia water (AW method) was the same as the AHC method except that 170 ml of the diluted ammonia water was used to get a final pH value of about 10.[12]

2.3. Powder characterization

Differential thermal analysis and thermal gravimetric analysis (DTA/TG) of the original precursors were made on a TG-DTA analyzer (Model TAS-200, Rigaku, Tokyo, Japan) in flowing air atmosphere (200 ml/min) with a heating rate of 10°C/min. The sample pot was platinum and the reference material was alpha-alumina.

Phase identification was performed by the X-ray diffraction (XRD) method on a Philips PW1700 X-ray diffractometer using nickel filtered CuK_α radiation in the range of $2\theta = 10\sim50°$ with a scanning speed of 1.5° 2θ per min. Crystallite size of the YAG powder was calculated from line-broadening of the (420) peak using the Philips APD 1700 soft ware from the Scherrer's equation.

Chemical analysis was made to determine composition of the precursor. NO_3^- content was analyzed by the spectrophotometric method on a Ubest-35 spectrophotometer

(Japan Spectroscopic Co., Ltd, Tokyo, Japan); NH_4^+ content was determined by the distillation-titrimetric method; Carbon content was assayed on a simultaneous carbon/sulfur determinator (LECO, CS-444LS, USA); Y and Al contents were determined by the chelate-titrimetric method.

Powder morphology and microstructure of the sintered body were observed by scanning electron microscopy (SEM, Model S-5000, Hitachi, Tokyo, Japan). For powders, sample was ultrasonically dispersed into acetone, and the suspension was spread on the surface of silicon plate. For sintered body, surface of the sample was polished to 1 μm finish with diamond paste and thermally etched at 1300°C for 2 h to reveal grain boundaries. All samples were coated with a thin layer of osmium for conductivity before observation.

2.4. Sintering

Two kinds of sintering methods, constant rate of heating (CRH) sintering and vacuum sintering, were used to investigate densification behavior of the YAG powders. Green bodies were obtained by isostatic compaction at 200 MPa pressure. CRH sintering was conducted in air using a TMA unit (Thermal Mechanical Analyzer, TMA 1700, Rigaku, Japan) up to 1500°C at a heating rate of 8°C/min and a cooling rate of 15°C/min. The sintered density, ρ, at any temperature, was determined from the green density ρ_0 and the measured linear shrinkage $\Delta L/L_0$ using the equation:

$$\rho = \rho_0/(1 - \Delta L/L_0)^3 \tag{1}$$

where L_0 is the initial length of the sample and $\Delta L = L_0 - L$, where L is the instantaneous sample length. The green density of powder compact was calculated from its weight and geometric dimensions. The theoretical density of YAG was taken as 4.55 g/cm^3.[6]

Vacuum sintering was performed in a furnace heated by a tungsten-mesh heater (Model M60-3X8-WW-23, Nemus, Tokyo, Japan). Samples were heated at a rate of 10°C/min to 1500°C and cooled down to room temperature at the same speed after holding 2 h.
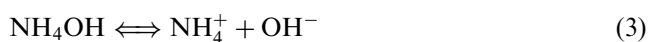
3. Results and discussion

3.1. Precipitation of the YAG precursor

Different ranges of pH variation were observed for the two precipitant solutions during precipitation. For the AW process, pH of the ammonia water decreased from an initial value of 11.78 to 9.92 at the completion of precipitation, and the resultant slurry had a constant pH of 9.92 during the aging period. Compared with ammonia water, the AHC solution showed much weaker alkalinity and had an initial pH value of 7.96. During precipitation, pH decreased only slightly from 7.96 to 7.84 and then increased gradually to the initial value during aging.

The precipitates produced by the two methods differed significantly, primarily due to the presence of different anion species. As expected, the use of ammonia water resulted in a gel-like precipitate which exhibited large volume shrinkage (about 70%) during drying. The dried precursor was strongly agglomerated and was very difficult to pulverize with a pestle and mortar. In principle the precipitate should be hydroxide since the only precipitation participating anion was OH^- generated by the dissociation of NH_4OH. The precursor prepared by the AHC method was apparently not the hydroxide type. It underwent much smaller volume change (about 10%) during drying, and the dried precipitate was softly agglomerated and was very easy to crush with a pestle and mortar, even with fingers. Composition of this precursor will be the result of competition between OH^- and the carbonate species generated by the following chemical reactions during combining with metal cations:

$$NH_4HCO_3 + H_2O \Longleftrightarrow NH_4OH + H_2CO_3 \tag{2}$$

$$NH_4OH \Longleftrightarrow NH_4^+ + OH^- \tag{3}$$

$$H_2CO_3 \Longleftrightarrow H^+ + HCO_3^- \tag{4}$$

$$HCO_3^- \Longleftrightarrow H^+ + CO_3^= \tag{5}$$

As mentioned earlier, Al^{3+} may precipitate as $AlOOH$ or $NH_4Al(OH)_2CO_3$. On the other hand, Y^{3+} may most likely precipitate as normal carbonate of $[Y_2(CO_3)_3 \cdot nH_2O \ (n = 2–3)]$[27] or basic carbonate of $[Y(OH)CO_3]$[29] from the present carbonate anions containing AHC solution.

3.2. X-ray diffraction (XRD) results

XRD spectra of the powders produced by the AW method and the AHC method are shown in Figs. 1 and 2, respectively. The co-precipitated powders by the two methods were found to be amorphous to X-rays until about 850°C. From 900°C, however, remarkable discrepancies concerning phase development were observed for the two precursors. For the powder produced by the AW method (Fig. 1), hexagonal YAP (JCPDS Card No. 16-219) crystallized at 900°C with the presence of small peaks of YAG (JCPDS Card No. 33-40). At 950°C, the YAP phase persisted, but with loss of intensity as more YAG formed. At 1000°C and above, YAG was the only phase detected. These results are consistent with the observations of Kinsman et al.,[11] though direct crystallization of YAG at a lower temperature of 800°C was also reported by other researchers for the co-precipitate from the same system.[10]
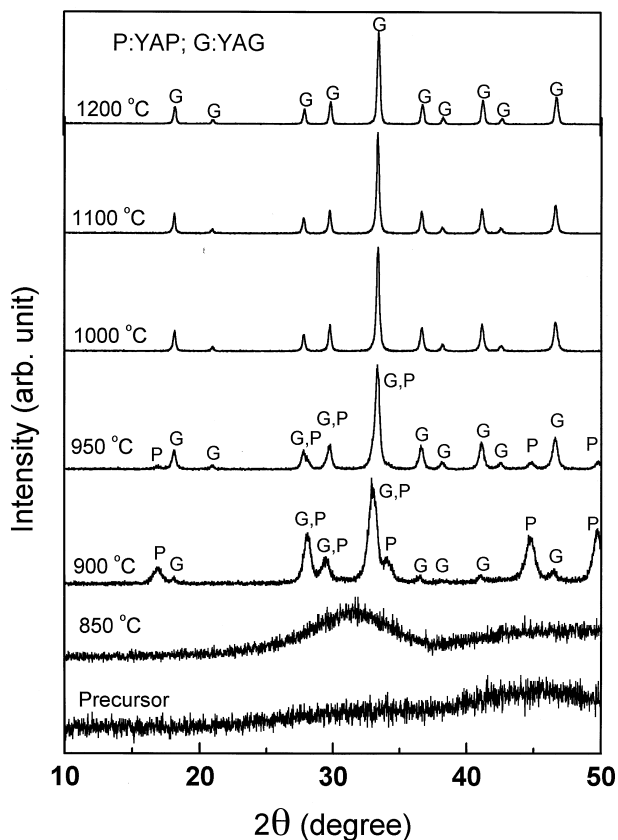
Fig. 1. XRD spectra of the powders synthesized with ammonia water.

Fig. 2. XRD spectra of the powders synthesized with ammonium hydrogen carbonate.

The precursor produced by the AHC method, however, crystallized as pure YAG at 900°C without the formation of any intermediate phases (Fig. 2), indicating higher cation homogeneity of the precursor. Above 900°C, continued refinement of peak shapes and intensities were observed, indicating crystallite growth of the YAG powder as temperature increases.

Fig. 3 exhibits the crystallite size of the YAG powders as a function of calcination temperature. Rapid crystallite growth was observed for both powders from 1100°C. The YAG powder prepared by the AHC method showed higher reactivity and faster crystallite growth rate.

3.3. Thermal analysis and chemical analysis

DTA/TG traces of the precursor produced by the AW method are given in Fig. 4. Three major peaks were identified on the DTA curve. The broad endothermic peak centered at 150°C was assigned to the removal of molecular water. The sharp exotherm at 917°C was caused by the crystallization of YAP, as evidenced by the XRD results in Fig. 1. The diffuse exothermic peak at about 1000°C corresponds to YAP reacting with a polymorph of Al_2O_3 to form YAG by the reaction:[5] $3YAlO_3 + Al_2O_3 \rightarrow Y_3Al_5O_{12}$. Though not detected by XRD, the Al_2O_3 polymorph present at 1000°C is most likely θ-Al_2O_3, which usually shows low crystallinity
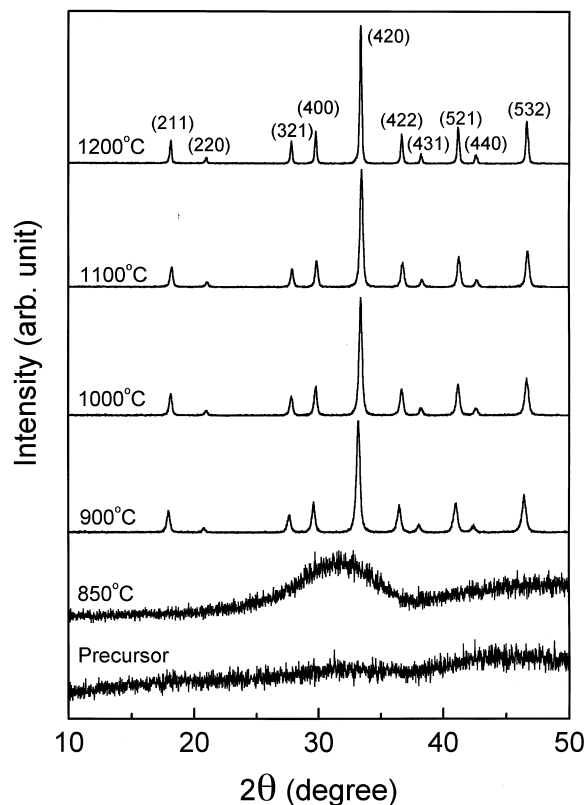
and transforms to highly crystalline α-Al_2O_3 at about 1200°C.[30,31]

The TG curve showed that complete thermal decomposition of the precursor into oxides was achieved at about 900°C with a total weight loss of 37.77% which is higher than the value (26.68%) expected for a precursor of pure hydroxide. Chemical analysis was not performed on this precursor, however, previous work[32] revealed that Y^{3+} usually precipitates as basic salt of approximate formula $Y_2(OH)_5X \cdot nH_2O$ (where X is NO_3^- or Cl^- depending on the type of starting salts, and $n = 1$ to 2) instead of pure hydroxide when ammonia water or sodium hydroxide was used as precipitant. Rasmussen et al.[33] once precipitated $Y_2(OH)_5(NO_3) \cdot 3H_2O$ from yttrium nitrate solution using a 2.5 M ammonia water. In fact, the mass loss of the present precursor is very close to the theoretical value (38.04%) calculated for $Al(OH)_3 \cdot 0.3[Y_2(OH)_5(NO_3) \cdot 3H_2O]$.

Fig. 5 shows DTA/TG curves of the precursor synthesized with AHC. The precursor seemed to undergo several stages of decomposition upon heating. The exothermic peak around 925°C was assigned to the crystallization of YAG, which is evidenced by the XRD results in Fig. 2 where no other phases were found. Major mass loss of the precursor occurred below 400°C, corresponding to about 80% of the total weight loss. The total weight loss (54.2%) is much higher than that of the

hydroxide precursor produced by the AW method, indicating a quite different chemical composition.

Chemical analysis on the precursor yielded 10.3 ± 0.17 wt.% of Al, 20.4 ± 0.29 wt.% of Y, 8.7 ± 0.10 wt.% of C, 6.5 ± 0.22 wt.% of NH_4^+ and 0.04 ± 0.01 wt.% of NO_3^-. These results correspond to a molar ratio of Al:Y:

$C:NH_4^+ = (1.0\pm0.02):(0.6\pm0.01):(1.9\pm0.02):(1.0\pm0.06)$, where NO_3^- was neglected because of its extremely low content. Assuming that C all comes from $CO_3^=$ and considering molecular electrical neutrality, composition of the precursor was approximately expressed as $NH_4AlY_{0.6}(CO_3)_{1.9}(OH)_2 \cdot 0.8H_2O$, where the coefficient
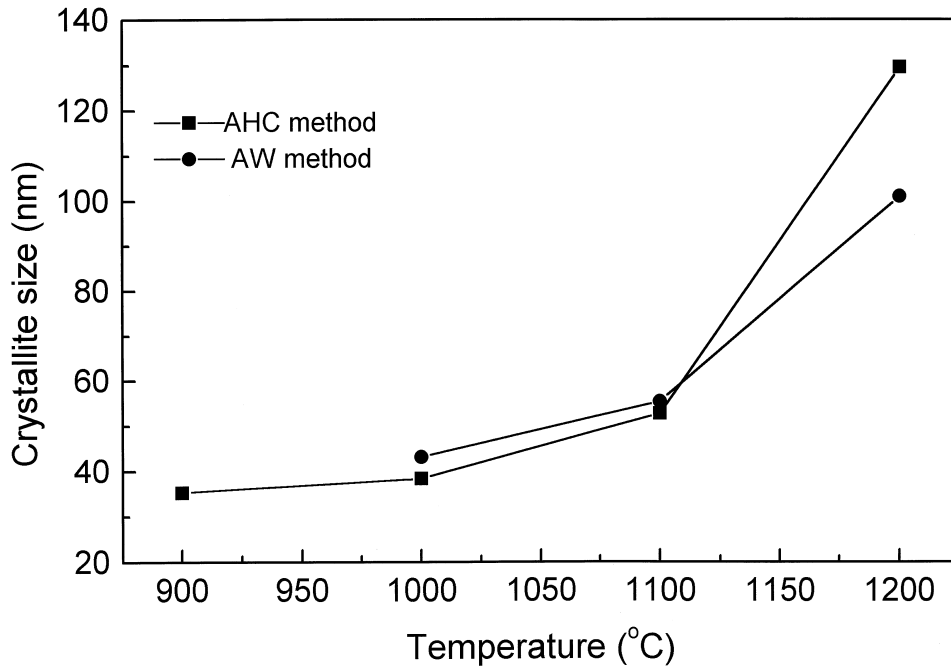

Fig. 3. Crystallite size of the YAG powder as a function of calcination temperature.
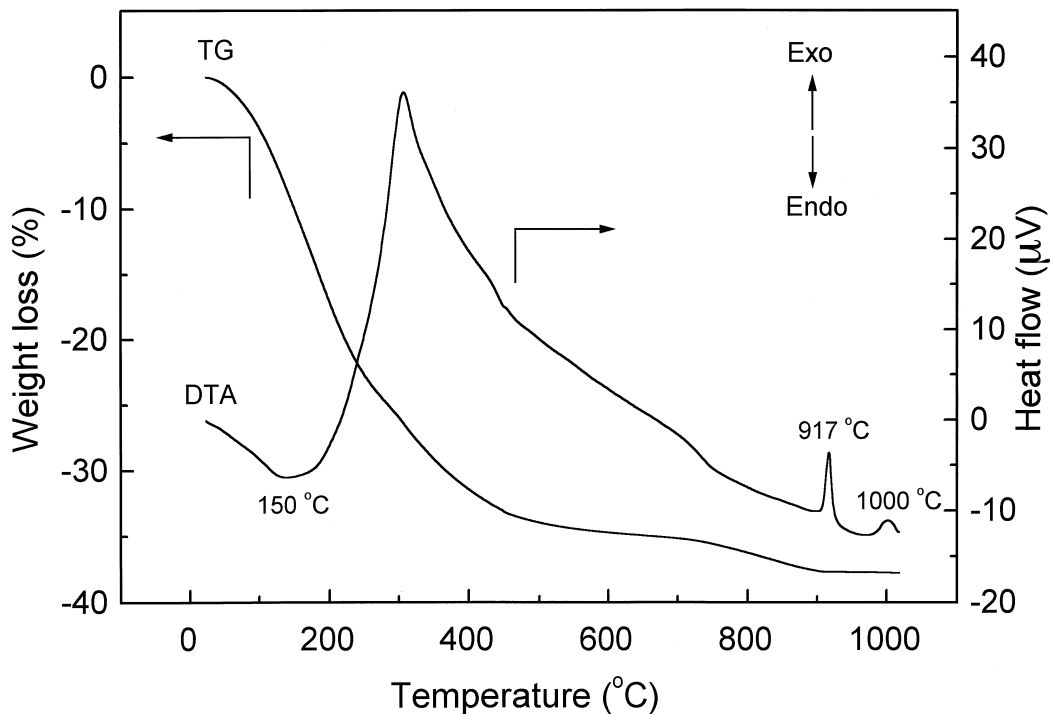

Fig. 4. DTA/TG traces of the precursor produced with ammonia water.

of molecular water was determined from the Y or Al content of the precursor. The theoretical mass loss of this formula (54.46%) shows good consistence with that revealed by the TG curve in Fig. 5. Earlier work produced $[NH_4Al(OH)_2CO_3]^{26,28,30}$ and $[Y_2(CO_3)_3 \cdot nH_2O$ $(n = 2–3)]^{27}$ from Al^{3+} and Y^{3+} containing solutions using AHC as precipitant. So composition of the present precursor may be further expressed as $[NH_4Al(OH)_2CO_3] \cdot 0.3[Y_2(CO_3)_3 \cdot 2.7H_2O]$. The weight loss of the precursor at lower temperatures ($< 400°C$) was mainly ascribed to the release of ammonia and molecular water and the partial decomposition of CO_3 group,[30] while that occurred at higher temperatures ($> 400°C$) was mainly due to the further decomposition of carbonate species.[34]

The direct formation of yttrium normal carbonate rather than basic carbonate was mainly due to the high $CO_3^=$ concentration of the AHC solution. Yttrium basic carbonate of $Y(OH)CO_3$ was classically produced by the so-called homogeneous precipitation process achieved by the forced hydrolysis of urea at elevated temperatures ($> 83°C$).[29] Though precipitation participating anions generated by the decomposition of urea are similar to those contained in the AHC solution, $CO_3^=$ concentration of the homogeneous precipitation system was believed to be very low, partially due to the extremely slow decomposition of urea and partially due to the decreased solubility of H_2CO_3 at elevated temperatures.

3.4. Powder morphology

Fig. 6 shows SEM morphologies of the powders produced by the AW method. The hydroxide precursor (Fig. 6a) mainly contains sub-micrometer sized dense aggregates of nano-sized primary particles. The resultant YAG powders (Fig. 6b–d), at any calcination temperature, were severely agglomerated and showed similar overall morphology to that of the precursor. It is obvious that agglomerate structure of the precursor has retained to the calcined powders.

Fig. 7 shows SEM morphologies of the powders synthesized by the AHC method. The carbonate precursor (Fig. 7a) was composed of extremely fine primary particles which are difficult to distinguish by SEM at the present magnification. Though apparently aggregated, the precursor showed much lower agglomeration strength compared with that of the hydroxide precursor, as mentioned in Section 3.1. The difference concerning agglomeration strength between the two kinds of precursors may be understood by considering their quite different chemical compositions. Severe agglomeration of the hydroxide precursor was mainly due to the bridging of adjacent particles with water by hydrogen bond and the huge capillary force generated during drying.[35] Though washed with alcohol to replace water molecules and decrease the extent of agglomeration, the hydroxide YAG precursor was still strongly agglomerated, as seen in Fig. 6a. For the carbonate precursor, however, the
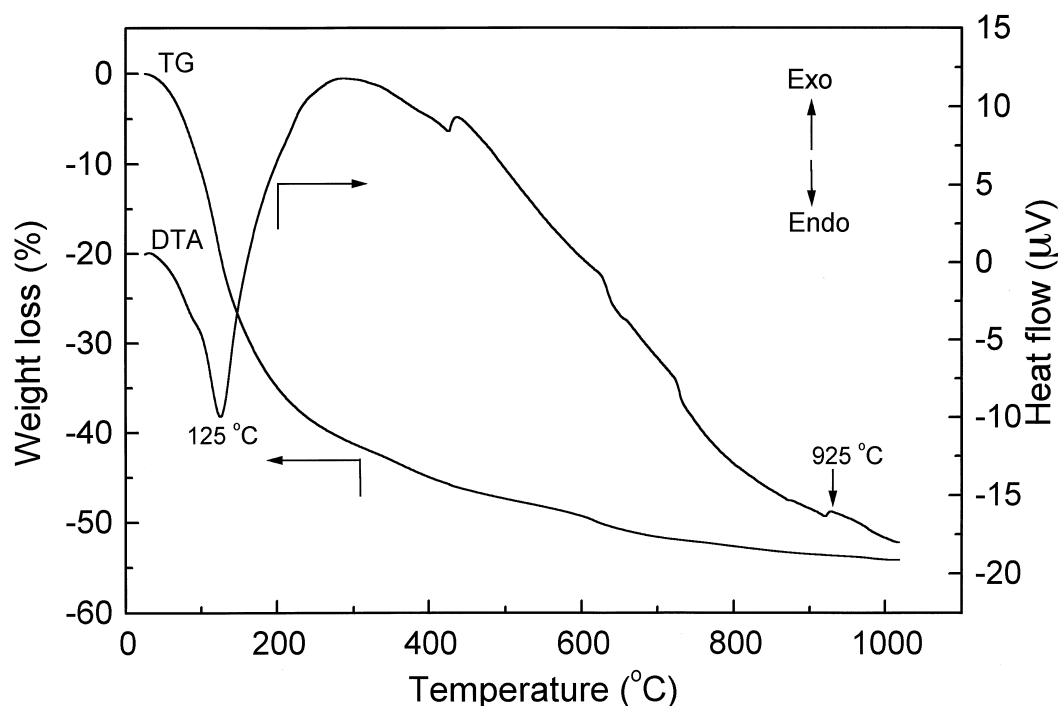

Fig. 5. DTA/TG traces of the precursor produced with ammonium hydrogen carbonate.

possibility of hydrogen bond formation was believed to have been significantly reduced and the water in the precursor was more easily to remove by alcohol washing. In fact, the carbonate precursor for chemical analysis, though not washed with ethanol, showed appreciably lower agglomeration strength than that of the hydroxide precursor washed with alcohol. Further decrease in agglomeration strength was also observed for the carbonate precursor rinsed with ethanol.

The YAG powders from the carbonate precursor (Fig. 7b–e) showed much better dispersity than those from the hydroxide precursor. Though appreciable particle/crystallite growth occurred at higher calcination temperatures, relatively good dispersity persisted.

3.5. Sintering

All samples for CRH sintering were compacted in one batch under 200 MPa pressure. Despite their quite similar crystallite sizes at the same calcination temperature up to 1100°C (Fig. 3), the YAG powder from the AW method, due to the presence of large and dense

agglomerates, exhibited much higher (about 10%) green density than that of the powder from the AHC method.

Fig. 8 shows densification behavior of the YAG powders from the hydroxide precursor. Raising calcination temperature caused higher onset temperature of densification due to crystallite/particle growth and decrease in reactivity. Severe agglomeration caused poor sinterability. After heating to 1500°C at constant rate of 8°C/min, the powders calcined at 1000, 1100 and 1200°C only densified to about 63.4, 62.1 and 61.8% of the theoretical, respectively.

The YAG powders synthesized by the AHC method showed much better sinterability than those by the AW method (Fig. 9). The powders obtained at 900, 1000, 1100 and 1200°C densified to their respective relative densities of about 88.7, 94.0, 93.5 and 83.5% under the same sintering conditions as used for powders by the AW method. Good dispersion was mainly responsible for the excellent sinterability of the YAG powders produced with AHC.

According to Fig. 9, the most favorable calcination temperature for a reactive YAG powder was 1100°C. Higher calcination temperature caused drastic increase
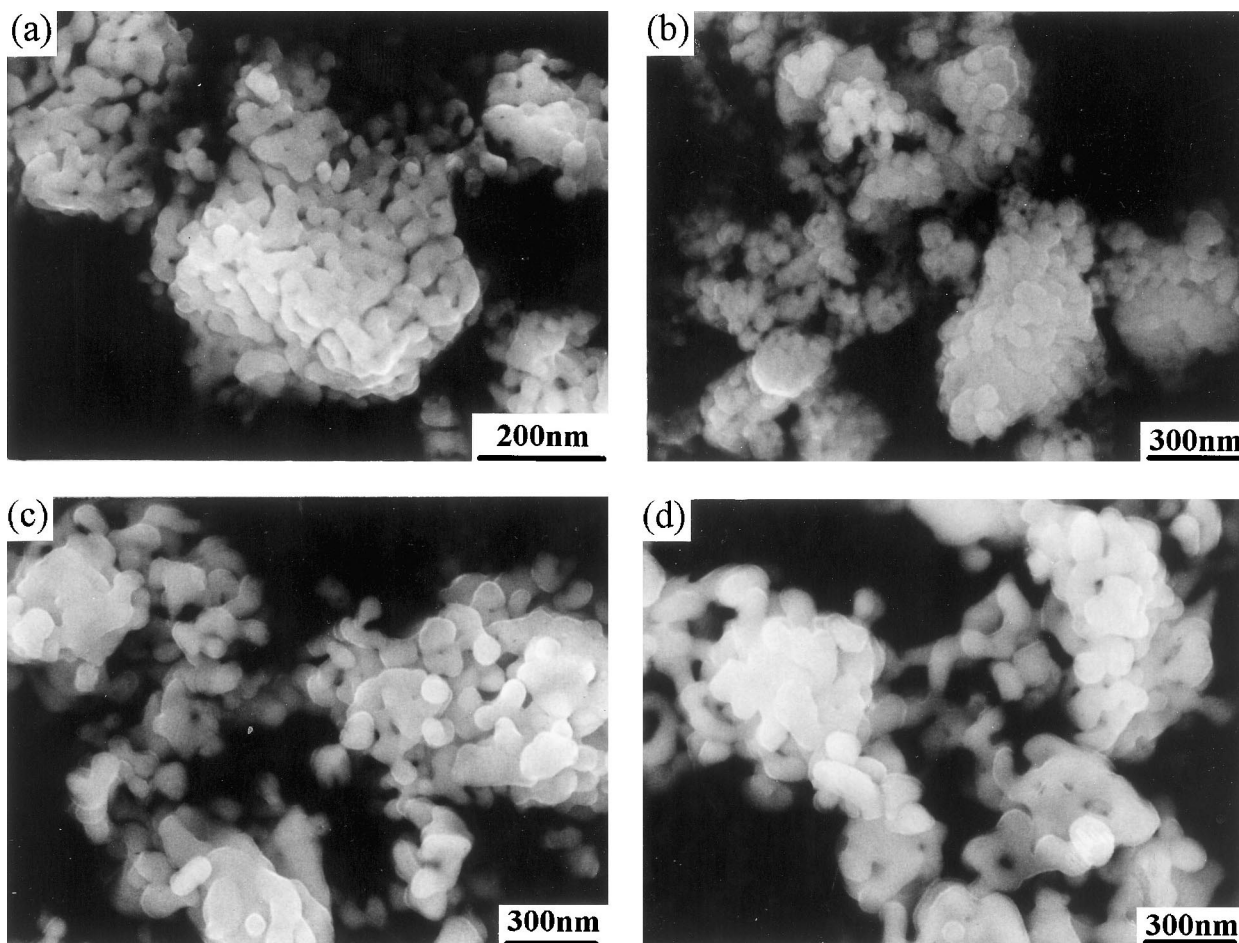

Fig. 6. SEM morphologies of the powders synthesized with ammonia water: (a) the precursor, (b) calcined at 1000°C, (c) 1100°C and (d) 1200°C.

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in crystallite size and decrease in sinterability. While the YAG powders produced at lower temperatures, though have smaller crystallite sizes, exhibited densification rates of slowed down at relatively lower sintered densities, as indicated by the densification curves between 1400 and 1500°C in Fig. 9. This can be explained from the view point of packing uniformity of particles in the green bodies. Smaller particle size, and hence higher friction force during dry compaction, made it more difficult to achieve uniform compaction. While microstructure inhomogeneities limit the powder compact to achieve high final density.[36,37]

The YAG powders calcined at 1100°C were used for vacuum sintering. Crystallite sizes, as determined by the
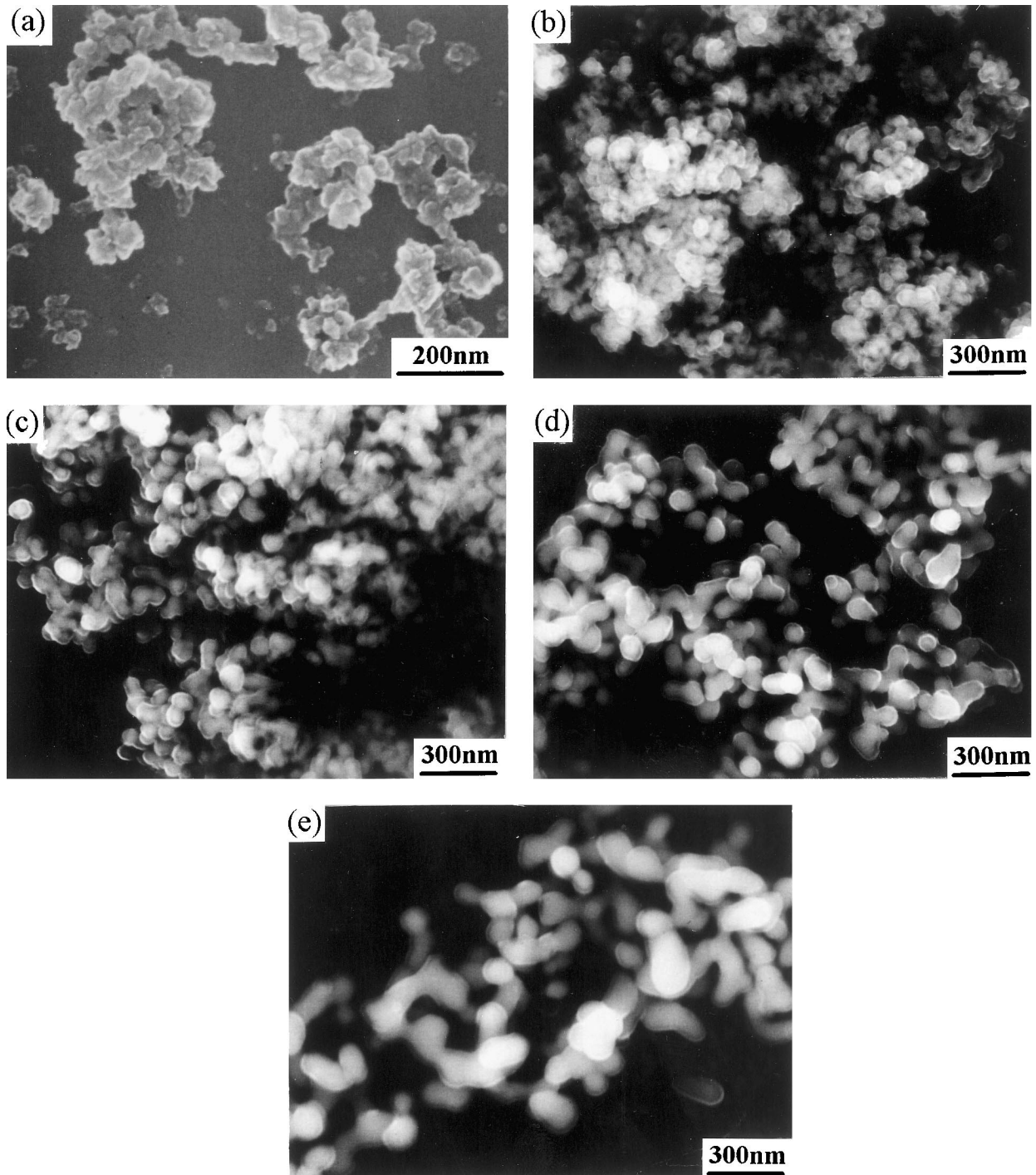

Fig. 7. SEM morphologies of the powders synthesized with ammonium hydrogen carbonate: (a) the precursor, (b) calcined at 900°C, (c) 1000°C, (d) 1100°C and (e) 1200°C.

X-ray line broadening method, were 55.5 and 52.8 nm for the powders by the AW and AHC methods, respectively.

Fig. 10 shows SEM microstructures of the YAG ceramics sintered at 1500°C for 2 h under vacuum. The powder from the AW method only reached a relative density of about 81.2% with a microstructure (Fig. 10a) consisting of porous regions embedded in a much denser background. While the powder synthesized by the AHC method reached nearly full density (~99.8%) at 1500°C. The sintered material has an average grain size of 1.2 μm (Fig. 10b). Translucency, though not so ideal,
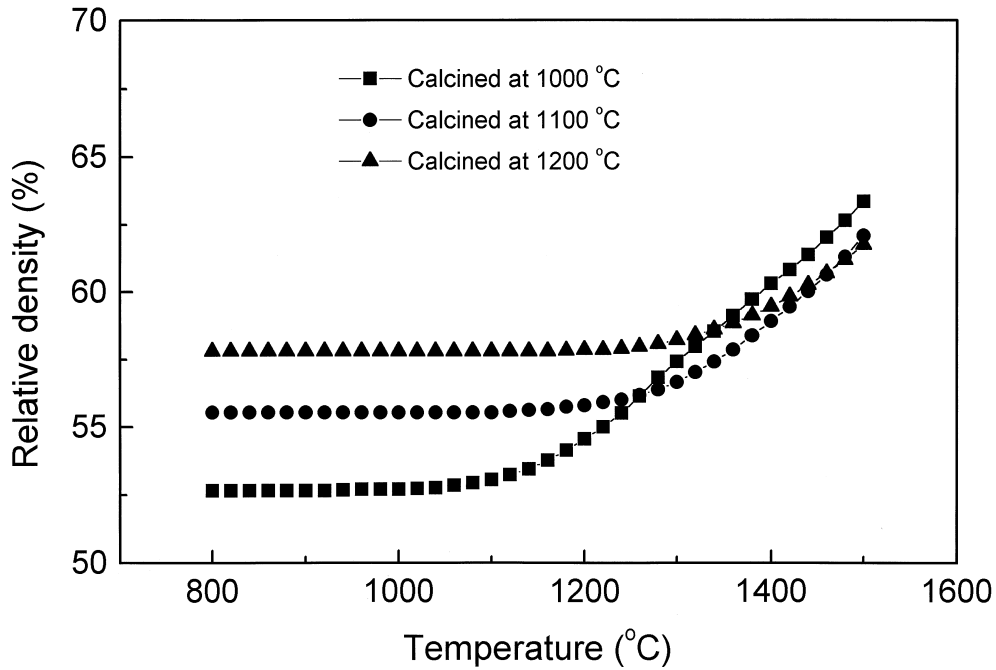

Fig. 8. Relative density versus temperature for YAG powders produced with ammonia water heated at constant rate of 8°C/min in air.
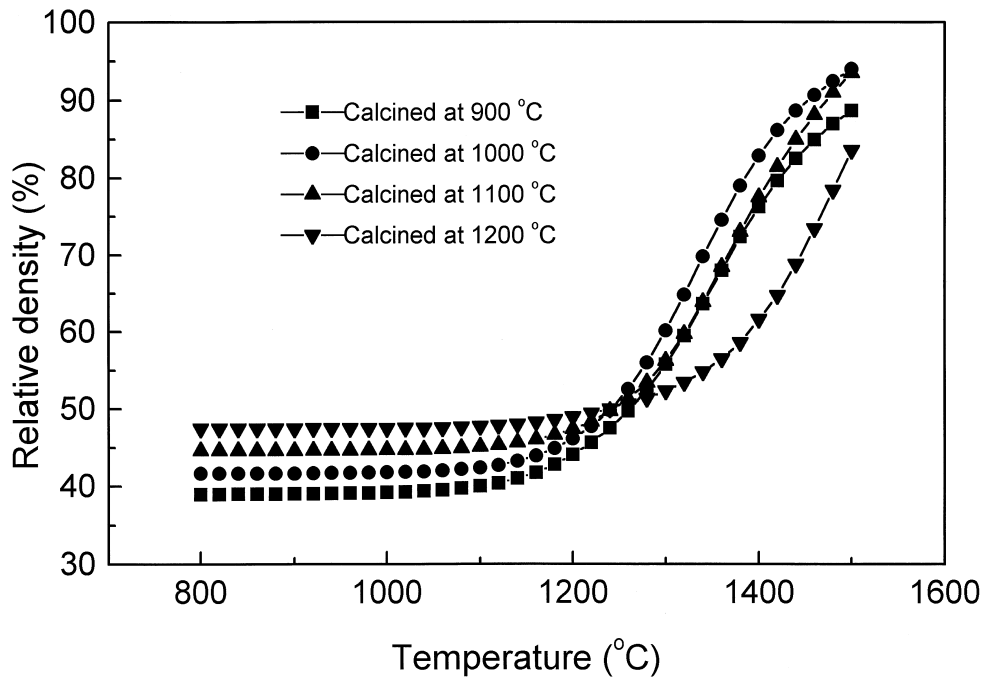

Fig. 9. Relative density versus temperature for YAG powders produced with ammonium hydrogen carbonate heated at constant rate of 8°C/min in air.
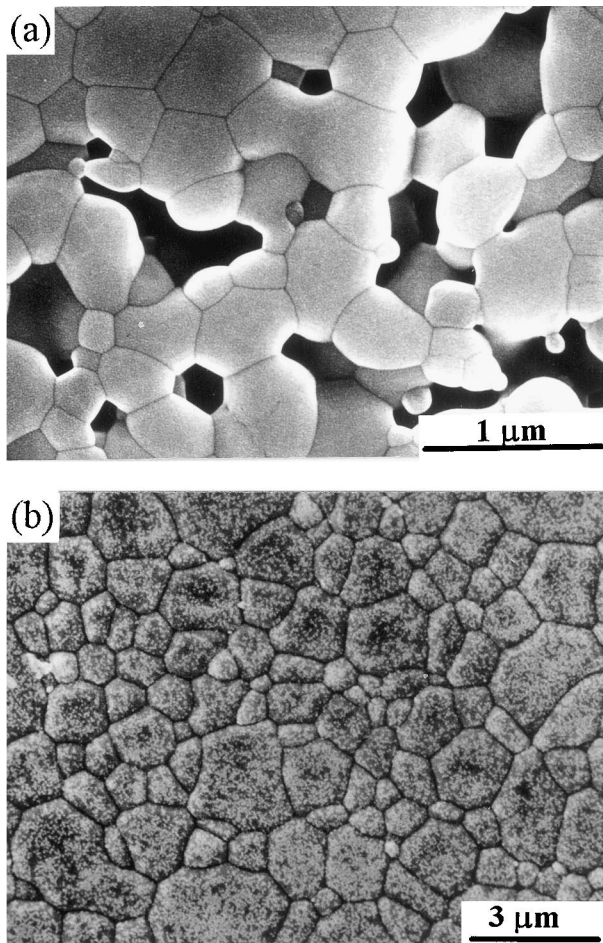
Fig. 10. SEM microstructures of the YAG ceramics sintered under vacuum at 1500°C for 2 h using powders calcined at 1100°C synthesized with: (a) ammonia water and (b) ammonium hydrogen carbonate.

was observed for the sintered body and letters could be read through the pellet of 1 mm thick. Further investigation was being made to improve transparency of the sintered YAG material.

4. Conclusions

YAG powders were produced via co-precipitation from a mixed solution of aluminum and yttrium nitrates using ammonia water and ammonium hydrogen carbonate as precipitants, respectively. Ammonium hydrogen carbonate exceeds ammonia water for the production of well-sinterable YAG powders.

The use of ammonia water produced a gelatinous hydroxide precursor with an approximate composition of $Al(OH)_3 \cdot 0.3[Y_2(OH)_5(NO_3) \cdot 3H_2O]$. The precursor transformed to pure YAG at about 1000°C via YAP intermediate phase. Severe agglomeration caused poor sinterability of the resultant YAG powders.

Carbonate precursor of YAG with an approximate composition of $NH_4AlY_{0.6}(CO_3)_{1.9}(OH)_2 \cdot 0.8H_2O$ were synthesized by using ammonium hydrogen carbonate as precipitant. The precursor converted directly to pure YAG at about 900°C. The precursor was loosely agglomerated after drying and the resultant YAG powders showed good dispersity and sinterability. The most desirable calcination temperature for the carbonate precursor was determined as 1100°C, and the YAG powder produced at this temperature densified to nearly full density by vacuum sintering at 1500°C for 2 h and the sintered body showed translucency.

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