### New Technology and Application Development of High Purity Alumina

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Alumina having a purity of 99.99% (4N) or more is classified as high purity alumina, and has been put into production using several manufacturing methods. Among them, Sumitomo's high purity alumina is manufactured using the hydrolysis method of aluminium alkoxide, and the company has developed various high purity alumina powders to meet customer requirements and expand its business. This report outlines the features of high purity alumina and the development of new grades for future applications.

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### Introduction

Alumina (Al<sub>2</sub>O<sub>3</sub>) is a metal oxide composed of aluminum (Al) and oxygen (O). Of all elements, aluminum has the third highest Clarke number (which indicates the percentage of the element among all elements on the earth's surface), indicating it is relatively abundant in the earth's crust. Alumina is known as one of the most common metal oxides. In addition, alumina has excellent physical and chemical properties such as good heat resistance, insulation performance, wear resistance, and corrosion resistance; thus far, it has been used in a wide range of fields.

Generally, alumina can be obtained by using caustic soda to extract sodium aluminate from bauxite ore, crystalizing a supersaturated solution of the extracted sodium aluminate into aluminum hydroxide, and calcining the obtained aluminum hydroxide. This method is known as the Bayer process; alumina obtained by this method has a purity of 99.6 to 99.9%, and is used in a variety of products such as refractories, sparkplugs, and IC boards.

By contrast, alumina with a purity of at least 99.99% (4N) is called high purity alumina, and is used in boards for LEDs, sapphire glass for watch faces and other single-crystal materials, fine ceramic materials such as for translucent tubes used in high-pressure

sodium lamps and for high-strength ceramic tools, coatings for lithium ion battery separators, and raw materials for high-intensity phosphors. Alumina has fine particles and the particle size distribution is homogeneous. These features enable it to be used in abrasive fillers for magnetic tape as well as precision abrasives for metals, plastics and other materials.

Sumitomo Chemical Co., Ltd. has established a technology for mass-producing high purity alumina by the aluminum alkoxide hydrolysis method. Since entering this market in 1981, Sumitomo Chemical has developed various high purity alumina powders to meet customer requirements and expanded its business.

This report outlines the features of high purity alumina and Sumitomo Chemical's development of new grades for future applications.

# Technologies for manufacturing high purity alumina

Known industrial methods for manufacturing high purity alumina include the aluminum alkoxide hydrolysis method, the ammonium alum pyrolysis method, the ammonium aluminum carbonate hydroxide (AACH) pyrolysis method, the aluminum underwater spark method, and the gas-phase oxidation method. Their respective manufacturing methods are outlined below.

### 1. Aluminum alkoxide hydrolysis method<sup>1)</sup>

Reacting aluminum with alcohol at high temperature produces aluminum alkoxide. Hydrolyzing the aluminum alkoxide as shown in Equation (1) produces aluminum hydroxide. According to Equation (2), calcining the aluminum hydroxide produces intermediate alumina of crystal types  $\gamma$ ,  $\delta$ , and  $\theta$  before finally producing  $\alpha$ -alumina, which is a phase stable at high temperature.

Alcohol produced by such hydrolysis can be separated, collected, and then reused in the alkoxide synthesis process. This method facilitates easy synthesis of high purity aluminum alkoxide, and the produced aluminum alkoxide can be further refined by distillation or other means. Thus, this is the easiest method for manufacturing high purity alumina.

$$AIOR_3 + 3H_2O \rightarrow AI(OH)_3 + 3ROH (R = alkyl group)$$
(1)

$$2AI(OH)_3 \rightarrow AI_2O_3 + 3H_2O \tag{2}$$

#### 2. Ammonium alum pyrolysis method<sup>2)</sup>

This is a typical method for manufacturing high purity alumina to be used as a raw material for artificial gems produced by the Verneuil method, which has been employed since long ago. Ammonium alum is obtained by the reaction of aluminum sulfate and ammonium sulfate; the pyrolysis reaction equation is as shown in Equation (3).

$$2NH_{4}Al(SO_{4})_{2} \cdot 12H_{2}O \rightarrow$$

$$Al_{2}O_{3} + 2NH_{3} + 4SO_{3} + 25H_{2}O \qquad (3)$$

Ammonium alum is refined mainly by recrystallization; its behavior depends on the cations contained in the alum. Generally speaking, sodium, magnesium, or calcium cations are easy to remove by recrystallization. On the other hand, when the alum contains potassium or gallium cations, they are difficult to remove. The removal rate is also influenced by the pH when the alum is precipitated.

When alumina is obtained from ammonium alum, the weight is reduced to about one-ninth by pyrolysis. For this reason, ammonium alum should be refined to a purity that is an order of magnitude higher than the desired purity of alumina. Ammonium alum dissolves into a viscous liquid in its water of crystallization at around 80 °C. Further heating allows it to solidify, pyrolyze while generating NH<sub>3</sub> and SO<sub>3</sub>, and turn into  $\gamma$ -alumina at approximately 900 °C. To completely remove the sulfur content, including SO<sub>3</sub>, the calcination temperature should be at least 1,150 °C. Due to the unique pyrolytic behavior of foaming at the time of solidification, alumina can be obtained that has a low bulk density and a low tendency to form aggregates; however, given the recent increase in awareness of environmental issues, detoxifying NH<sub>3</sub>, SO<sub>3</sub>, and other gases in order to reduce the environmental load involved in production processes has become an issue.

### Ammonium aluminum carbonate hydroxide (AACH) pyrolysis method<sup>3)</sup>

This is a method for obtaining high purity alumina by synthesizing ammonium aluminum carbonate hydroxide (AACH) from ammonium alum and ammonium hydrogen carbonate as shown in Equation (4) and then pyrolyzing the AACH.

$$2NH4Al(SO4)_2 + 8NH4HCO_3 \rightarrow 2NH4AlO(OH)HCO_3$$
$$+ 4(NH4)_2SO_4 + 6CO_2 + 2H_2O \qquad (4)$$

$$2NH_{4}AlO(OH)HCO_{3} \rightarrow$$

$$Al_{2}O_{3} + 2NH_{3} + 2CO_{2} + 3H_{2}O$$
(5)

The conditions under which AACH is synthesized are known to greatly affect alumina sintering and other performance; it is said that under optimum conditions, rice grain-shaped fine AACH particles measuring 1.2 to 2 µm by 0.5 to 0.9 µm are formed, and pyrolyzing these fine AACH particles results in 0.1 to 0.2 µm fine  $\alpha$ -alumina particles with good sintering performance.

With its low affinity for water, AACH is easy to filter, dry, and treat in other ways, and has the advantage of generating less SO<sub>3</sub> gas than the alum pyrolysis method. However, this method is more expensive because alum is used as the raw material to synthesize AACH.

### 4. Aluminum underwater spark method<sup>4)</sup>

When voltage is applied to electrodes (aluminum pellets) that face each other in water in order to cause a spark discharge between them, the aluminum surface is partially melted by the spark discharge and exfoliated. The exfoliated aluminum powder then bonds with OH radicals produced by electrolysis of water to form aluminum hydroxide. By calcining this aluminum hydroxide, high purity alumina can be obtained.

### 5. Gas-phase oxidation method<sup>5)</sup>

By gasifying aluminum chloride and then burning and hydrolyzing the gas in oxyhydrogen flame as shown in Equation (6), high purity alumina can be obtained.

$$2\text{AlCl}_3 + 3\text{H}_2\text{O} \rightarrow \text{Al}_2\text{O}_3 + 6\text{HCl} \tag{6}$$

Generally, when alumina is obtained from a burning reaction in oxyhydrogen flame, its primary particles are small as in the case of synthesis of silica or titania. Next, bulky secondary aggregated particles are formed, and powder that is well dispersible in water and other solvents can be obtained. Alumina obtained by this method is of the crystal type  $\delta$ , and often contains HCl as a by-product impurity.

### **Development of fine alumina**

In order to use alumina in various applications, we have developed various types of fine alumina with particle sizes less than a few  $\mu$ m as shown in **Table 1** and **Fig. 1**. These are used in ceramic sintered bodies, coatings for lithium ion battery separators, abrasives, and many other fields.

Among fine alumina, nanoscale and other fine particles have a larger specific surface area than microsized particles and exhibit other excellent chemical and physical properties; high expectations have been placed in them.<sup>6</sup>

In the field of ceramic materials, which are the major application of fine alumina and have the disadvantage of being generally brittle, it has been reported that by molding and calcining raw alumina processed into nano-sized crystal particles, a tough, less brittle material can be obtained.<sup>7)</sup> A dense sintered body obtained in this way is also advantageous in that it can be calcined at a relatively low temperature. Ceramic materials made of an alumina sintered body are used in the internal parts of semiconductor production equipment exposed to plasma, chemical solutions, and other severe environments due to their excellent heat resistance and corrosion resistance. This field's market is expected to grow as 5G, the next-generation communications standard, is on the cusp of widespread adoption. At the same time, as semiconductor circuits are increasingly miniaturized, production conditions are becoming more severe, and expectations are rising for ceramic parts to have even higher resistance to such conditions. Thus, nano-sized fine alumina particles that

|                           |                      | AKP-20  | AKP-30  | AKP-50  | AKP-3000 | HIT-50 |
|---------------------------|----------------------|---------|---------|---------|----------|--------|
| Crystal structure         |                      | α       | α       | α       | α        | α      |
| Purity                    | (%)                  | >99.99  | >99.99  | >99.99  | >99.995  | >99.95 |
| Particle size             | (µm)                 | 0.4-0.6 | 0.3-0.5 | 0.1-0.3 | 0.4-0.7  | -0.25- |
| Loose bulk density        | (g/cm <sup>3</sup> ) | 0.7–1.1 | 0.7-1.0 | 0.6-1.1 | 0.3-0.6  | -      |
| BET specific surface area | (m <sup>2</sup> /g)  | 4-6     | 5-10    | 9–15    | 4-8      | 6-13   |
| Impurity level            | Si (ppm)             | ≦40     | ≦40     | ≦25     | ≦20      | ≦50    |
|                           | Na (ppm)             | ≦10     | ≦10     | ≦10     | ≦10      | ≦10    |
|                           | Mg (ppm)             | ≦10     | ≦10     | ≦10     | ≦10      | ≦10    |
|                           | Cu (ppm)             | ≦10     | ≦10     | ≦10     | ≦10      | ≦10    |
|                           | Fe (ppm)             | ≦20     | ≦20     | ≦20     | ≦10      | ≦30    |

 Table 1
 Various properties of Sumitomo's high purity alumina

(1) AKP-20









SEM image of Sumitomo's high purity alumina



Fig. 2 TEM image of Sumitomo's ultra fine high purity alumina

can make dense sintered bodies will be in greater demand than ever before.

Since in addition to alumina, other inorganic oxides are becoming increasingly sophisticated due to recent trends toward finer and nano-sized particles, pulverizers, dispensers, classifiers, and other devices essential for developing new grades are being created in rapid succession. By using these devices to optimize production conditions, nano-sized fine alumina powder appropriate for various applications can be developed. Generally, the smaller the primary particles, the higher the tendency for particles to aggregate, and it is also important to develop additives to inhibit such aggregation. Besides being used in ceramic sintered bodies, nano-sized fine alumina is expected to achieve thinner coating layers for lithium ion battery separators, to make highly active catalysts and uniform composite materials, and to exhibit other effects in a wide range of applications.<sup>8)</sup>

As previously mentioned, there are various methods for synthesizing alumina; generally, the transition temperature for an alumina crystal at which intermediate alumina turns into  $\alpha$ -alumina is as high as around 1,200 °C, whereby particles grow and aggregate due to the progress of necking between particles at the same time. This is why it is said to be difficult to produce nano-sized  $\alpha$ -alumina with high dispersibility. There are some laboratory-level methods for synthesizing nano-sized  $\alpha$ -alumina such as the gas phase method, the sol-gel method using alkoxide, the hydrothermal method, the coprecipitation method, and the polymerized complex method. Among these, the sol-gel method is advantageous in that high purity fine particles with a large specific surface area can be obtained.<sup>9)</sup> Technically, it is important to reduce the temperature at which the transition to an  $\alpha$ -type crystal occurs, and processes for realizing the transition to an  $\alpha$ -type crystal at a low temperature (e.g., the addition



ultra fine high purity alumina

Table 2 Ty fit

Typical properties of Sumitomo's ultra fine high purity alumina

|                           |           | Sample A | Sample B |
|---------------------------|-----------|----------|----------|
| Crystal structure         |           | α        | α        |
| Particle size*            | (nm)      | 70       | 110      |
| BET specific surface area | $(m^2/g)$ | 16       | 12       |
| *: Measured by TEM        |           |          |          |

of an alumina precursor or some species) are being studied vigorously.

After earnest examinations based on our know-how accumulated over many years, we were able to develop nano-sized  $\alpha$ -alumina by employing an industrially feasible method. TEM images of two nano-sized  $\alpha$ -alumina samples are shown in **Fig. 2**, their particle size distributions in **Fig. 3**, and their physical properties in **Table 2**. By optimizing this process, particle size can be controlled at the nano-size level, and spherical alumina of crystal type  $\alpha$  is expected to be used in the various applications previously mentioned. In order to expand this technology's availability, we will further combine it with appropriate devices for producing finer particles.

### **Development of large-size alumina**

Recently, equipment has become increasingly motorized and computerized as typified by the automobile industry, and heat-producing parts have rapidly come to be installed in more and more places. Because heat increases the failure rate of electronic equipment and shortens its service life, it is important to introduce thermal design and heat countermeasures to ensure reliability. Generally, as a countermeasure against heat for electronic equipment, a thermal interface material that transfers heat from the heat source to a heatsink, a case, or the like is used. As shown in Fig. 4, a thermal interface material is a composite material composed of a matrix resin (which may be silicone, epoxy resin, or another type of resin) that is filled with a thermal conductive filler, and it combines the thermal conductive property of a filler with the physical properties of a resin. To increase the thermal conductivity of thermal interface materials, it is important to place the thermal conductive filler portions so that they make good contact with each other in order to form a thermal conduction path.



 Table 3
 Typical properties of thermal conductive materials

Two types of thermal conductive fillers are used: electrically conductive fillers made of metal, carbon, or other conductive materials, and insulating fillers made of ceramic or other insulating materials. Electrically conductive fillers made of metal, carbon, or other conductive materials have a high thermal conductivity, but may cause a short circuit or electric shock when used in an IC chip, inverter, or other places where voltage is applied; consequently, they can be used as fillers only in limited areas. On the other hand, insulating fillers withstand voltage, but their thermal conductivity is an order of magnitude less than that of electrically conductive fillers, and their filling rate must be increased to compensate for such inferior thermal conductivity when high thermal conductivity is required.

As the performance of electronic equipment increases, semiconductor devices that have higher processing speeds and that produce more heat have come into increasing use. In addition, as equipment continues to be further downsized, various devices are now mounted more densely, increasing heat density. For these reasons, thermal interface materials now need to have higher thermal conductivity than ever before. In addition, thermal interface materials used in power semiconductors that operate at a high voltage and large current are required to offer more reliable insulation performance than ever before; consequently, it is assumed that the use of ceramic fillers with high insulation performance will increase.

As ceramic fillers, boron nitride, aluminum nitride, and alumina as listed in **Table 3** are mainly used; among ceramic materials, they have a relatively high thermal conductivity. How to improve the thermal dissipation of ceramic materials, which have a thermal conductivity lower than that of electrically conductive materials, is an issue to address. Boron nitride and aluminum nitride have a higher thermal conductivity than

|                      |      | Boron nitride               | Aluminium nitride     | Spherical alumina                     | Advanced Alumina (AA) |
|----------------------|------|-----------------------------|-----------------------|---------------------------------------|-----------------------|
| Thermal conductivity |      | O                           | 0                     | ○<br>(×: Fine size)                   | 0                     |
| Cost                 |      | ×<br>High                   | ×<br>High             | $\bigcirc$ ( $\triangle$ : Fine size) | $\triangle$           |
| Handling ability     |      | ×<br>Crystalline anisotropy | ×<br>Water resistance | O                                     | 0                     |
| Particle size        | (µm) | 0.5-40                      | 1-80                  | <100                                  | 0.2–18                |

 $\bigcirc$ : Excellent  $\bigcirc$ : Good  $\triangle$ : Not bad  $\times$ : Poor



Fig. 5SEM image of Advanced Alumina (AA)

alumina and are expected to increase the thermal conductivity of thermal interface materials, but they are disadvantageous in that they are expensive. In addition, boron nitride shows crystal anisotropy in thermal conductivity, and aluminum nitride is poor in resistance to water as well as difficult to work with. Alumina is relatively inexpensive, provides good electrical insulation, is chemically stable, and is highly reliable when used as a material for thermal dissipation fillers; for these reasons, alumina is currently the most commonly used thermal interface material.

Thus far, we have produced and sold the Advanced Alumina (AA) series, which is high purity alumina having a uniform particle size synthesized by our original synthesis method, the In Situ Chemical Vapor Deposition method.<sup>10)</sup> As shown in Fig. 5, Advanced Alumina consists of polyhedral, sphere-like single-crystal particles. AA is a  $\alpha$ -phase single crystal that has a thermal conductivity that is relatively high for alumina; it takes the form of polyhedral particles, which, when densely packed in a thermal interface material as a filler, make surface contact with other fillers. These features enable its use in highly thermal conductive alumina fillers. Another feature of Advanced Alumina is that it is available in a wide range of central particle sizes, from a submicron size (0.2 µm) to 18 µm. To increase thermal conductivity, it is important to pack thermal conductive fillers as closely as possible. By mixing several types of Advanced Alumina having different central particle sizes, alumina particles can be packed into the closest possible state to achieve high thermal conductivity.

The thermal conductivity of alumina ( $\alpha$  phase) is 30 to 40 W/m K, but even if spherical alumina with excellent fillability is packed closely in a thermal interface material, its thermal conductivity may be as low as 4 to 6 W/m K.<sup>11</sup>) This is probably because as shown in **Fig. 6** (a), resin with a low thermal conductivity inhibits



thermal conduction in the interface between the alumina and the resin. Therefore, to increase thermal conductivity, a thermal conduction path must be formed effectively. Using alumina of a large particle size makes the thermal conduction paths in the alumina particles longer as shown in **Fig. 6** (b), and reduces the interface between the alumina and the resin where thermal dissipation is inhibited. Consequently, the thermal interface material's thermal conductivity can be increased.<sup>12</sup>) Next, large-size alumina's effect on increasing the thermal conductivity of thermal interface materials was evaluated by using our newly developed large-size alumina and common spherical alumina as the main fillers and our Advanced Alumina as the sub-filler.

An electron micrograph of the developed large-size alumina is shown in **Fig. 7**. Its central particle size is 260  $\mu$ m, and it has particles larger than spherical or roundish alumina (10 to 100  $\mu$ m), which are commonly used as main fillers.<sup>11</sup> Sumitomo Chemical's developed large-size alumina has a high  $\alpha$  conversion rate and high true density, and is expected to provide a highly

500 µm

Fig. 7 SEM images of developed large-size alumina

thermal conductive filler that increases the thermal conductivity of thermal interface materials.

The developed large-size alumina was mixed with epoxy resin into a thermal interface material, and its thermal conductivity was evaluated by the laser flash method. The results are shown in **Fig. 8**, which indicates that using the large-size alumina has the effect of increasing the thermal conductivity, and the higher the filler filling rate by volume, the larger the effect. At a filling rate by volume of 80 vol%, the compound containing only alumina as the filler achieved a thermal conductivity of 10.4 W/m K. This value is much higher than the thermal conductivity of 4.2 W/m K when using conventional spherical alumina as the filler, indicating a potential use of alumina in applications that require high thermal conductivity.



Fig. 8 Thermal conductivity of thermal interface material compounded developed large-size alumina

## Application of alumina to porous bodies (inorganic separation membranes)

Porous bodies using alumina are used in separation membranes because of their excellent chemical resistance and heat resistance. Among the various separation technologies, there are high expectations for separation membranes as an energy-saving process. It is said that in the U.S., separation processes are responsible for about half of the industrial sector's energy consumption, and distillation processes account for about half of separation processes. If membrane separation can be used in place of distillation processes, it is thought that energy use can be reduced by approximately 90%.<sup>13)</sup>

Recently, inorganic separation membranes have been actively studied, and it is assumed that inorganic substances can, due to their excellent chemical stability and heat resistance, be used directly in processes involving organic substances or high-temperature processes. For example, inorganic separation membranes are expected to be employed in a variety of processes such as alcohol dehydration, separation of alkane and alkene, separation of the greenhouse gas CO<sub>2</sub>, separation of H<sub>2</sub> (expected to function as a nextgeneration energy source), and membrane reactors that achieve a high conversion rate by combining separation and reaction to shift equilibrium.<sup>13), 14)</sup>

An inorganic separation membrane is roughly composed of a filtration layer and a support as shown in **Fig. 9.** The filtration layer delivers actual separation performance, and may separate particles by different mechanisms (e.g., the molecular sieve mechanism in which smaller particles that can pass through the membrane pores are separated from larger particles that cannot, or the solution-diffusion mechanism which separates particles that are soluble in the membrane from those that are not). The filtration layers



that can be used include zeolite membranes, silica membranes, palladium membranes, carbon membranes, or MOF (Metal-Organic Framework) membranes. To achieve high separation performance, regardless of the material used, it is important to form a dense membrane.<sup>14)</sup>

By contrast, the support provides support for a dense, thin filtration layer, and is mainly made of alumina. The support can be subdivided into a substrate and an intermediate layer. To form a filtration layer, the support's pores must be small in order to increase the smoothness of its outermost surface. At the same time, to increase the separation membrane's permeance performance, the substrate's pore diameter should be large. To meet these two requirements, an intermediate layer is employed. By laying a thin intermediate layer with small pores on a substrate with large pores so as to form the support, the filtration layer's membrane formability can be improved while maintaining the substrate's high permeance performance. We considered developing a small pore support with high permeance performance by producing an intermediate layer using our own alumina powder.

An intermediate layer can be produced by applying slurry, in which particles are dispersed, over a substrate and then drying and calcining the layer. To make the intermediate layer's pores smaller, small particles should be applied over the substrate. However, when small particles are applied on a substrate with large pores, the small particles become embedded in the substrate's pores. Thus, previously it was difficult to lay a thin membrane on a substrate. To resolve this issue, we developed an original method in order to realize a technology for laying alumina powder directly onto a substrate having an average pore size of 15 µm. Fig. 10 shows an SEM image of a cross section of the developed support. This image depicts a 10 to 20 µm thick intermediate layer laid on a porous substrate with large pores, and the surface of the intermediate layer is smooth with little roughness. Generally, a filtration layer is laid on the intermediate layer, and the intermediate layer's uneven surface makes it difficult to produce a dense filtration layer. At present, we are considering producing a filtration layer atop the intermediate layer that we have developed, and we are planning to develop intermediate layers that are compatible with filtration layers.

The gas permeance performance of the developed support was as high as  $1 \times 10^{-5} \text{ m}^3/\text{m}^2 \cdot \text{s} \cdot \text{Pa}$ , while the



100 µm

Fig. 10

SEM image of cross section over support surface region



intermediate layer's pore size was as small as 0.3 to 0.4 µm. This permeance value is about two orders of magnitude higher than commercially available supports as shown in Fig. 11. The high permeance performance is probably because although the substrate's pore size was more than 30 times as large as that of the intermediate layer, the intermediate layer could be laid directly on the substrate, and the thin intermediate layer could be laid on the substrate because the intermediate layer has a unique structure that prevents it from becoming embedded in the substrate. Previous studies have also shown that our alumina powder used herein has high permeance performance due to its unique particle shape,<sup>15)</sup> and it is assumed that high permeance performance supports can be produced by combining the features of alumina particles with layering technology.

### Application to composite oxides

Transparent materials as exemplified by glass, single crystals, and polymers are widely used as window materials, optical materials, and others in both the industrial and consumer product fields. Major transparent materials scatter and absorb less light; they have a single-crystal or amorphous structure. However, in recent years highly transparent, high-strength ceramic has been attracting attention as a transparent material.<sup>16)</sup> Ceramic is already used in practice as an optical material, a high-strength window material, and others and will become more widely used in the industrial and consumer product fields. Though the U.S., Europe, Israel, and other countries are in the lead to develop this material, in Japan, too, a study group on optoceramics has been launched within the Japan Fine Ceramics Association (JFCA), and going forward ceramic will attract more attention in this country as well.

To make polycrystal ceramic transparent, the sintered structure must be made denser and finer; to meet these requirements, the physical properties of the raw powder are very important. Various transparent ceramic materials made of a composite oxide containing aluminum have been put into practical use-for example, AlON, YAG (Y3Al5O12), and magnesia spinel (MgAl<sub>2</sub>O<sub>4</sub>). In particular, magnesia spinel can be made merely by mixing an aluminum source and a magnesium source and then calcining the mixture in the atmosphere; it is a composite oxide that can be synthesized more easily than AlON, which uses aluminum nitride (AlN) as the raw material, or YAG, which has various similar crystal compositions such as YAM (2Y2O3·Al2O3) and YAP (Y2O3·Al2O3). In addition, because magnesia spinel is a cubic crystal, transparent ceramic can easily be obtained from this material by HIP sintering and other methods.<sup>17)</sup> A transparent body of spinel has higher infrared transparency than sapphire, and is expected to be used as a new highstrength transparent material in active sensor windows and other applications.

When spinel is used as a raw material for transparent ceramic, the powder properties (e.g., the particle size distribution and the specific surface area) are important, and it is also necessary to produce high purity spinel powder that does not contain Fe or other unnecessary elements that tint the ceramic by light absorption. This is a simple process, but it is not easy to









prevent contamination caused by the materials of the equipment or containers used for mixing and pulverizing. Based on know-how accumulated in the field of high purity alumina production, we can reduce the concentration of impurity elements in spinel powder to below 10 ppm. In fact, we obtained a highly transparent magnesia spinel single crystal by synthesizing magnesia spinel using our high purity alumina and producing a single crystal therefrom by the Czochralski method. Fig. 12 shows an XRD (X-Ray Diffraction) pattern for magnesia spinel powder, while Fig. 13 shows an image of a spinel single crystal. Fig. 12 indicates that the powder used as the raw material for the single crystal was a single-phase spinel crystal. As a result, a colorless, very highly transparent single crystal was obtained.

Thus far, we have developed technologies for controlling particle shapes to produce high purity aluminabased particles of various shapes such as fine powder, molded bodies, and granular powder. Which properties are required of alumina or magnesia spinel depends on the intended application. This is true for all other types of raw powder as well. For example, in an application in which the powder should be flowable, granular powder is preferred. By using the aforementioned technologies



Fig. 14 Image of spinel granules

for controlling particle shapes with magnesia spinel powder, uniformly sized granules as shown in Fig. 14 can be produced.

Our technologies for increasing purity and controlling particle shapes developed along with the technology for manufacturing high purity alumina can be applied to other composite oxides, and in the future we will develop technologies for producing ultra-fine powder for transparent ceramic.

### Reference

- T. Hashimoto *et al.*, SUMITOMO KAGAKU, Special issue, 1980-II, 59 (1980).
- 2) S. Kazama, Bulletin of the Ceramic Society of Japan, 17 (9), 764 (1982).
- S. Kato, Fain Seramikkusu (Japan), Fine Ceramics, 4, 100 (1983).

- 4) W. Ishibashi *et al.*, Bulletin of the Ceramic Society of Japan, 6 (6), 461 (1971).
- 5) A. Kato *et al.*, Journal of the Society of Materials Science, Japan, 21, 540 (1972).
- 6) P. S. Behera et al., Interceram, 65, 10 (2016).
- H. Muto *et al.*, J. Jpn. Soc. Powder Powder Metallurgy, 54 (12), 839 (2007).
- N. Hezil and M. Fellah, J. Aust. Ceram. Soc., 55, 1167 (2019).
- S. N. S. Mohamad *et al.*, IOP Conf. Series: Materials Science and Engineering, 701, 012034 (2019).
- 10) Y. Uchida, FC Report, 24 (4), 153 (2006).
- 11) M. Seguchi, 2019-Nen netsu seigyo hönetsu buzai shijö no genjö to shin yötö tenkai [2019 Current Statas of Market for Thermal Dissipation Member and Development of New Application], Fuji Keizai Group (2019), p. 223.
- 12) Sumitomo Chemical Co., Ltd., WO 138335 A1 (2020).
- 13) D. S. Sholl and R. P. Lively, Nature, 532, 435 (2016).
- 14) Supervised by Hidetoshi Kita, enerugi·kagaku purosesu ni okeru maku bunri gijutu [Membrane separation technology in energy and chemical processes], S&T Publishing Inc. (2014).
- Sumitomo Chemical Co., Ltd., JP 5362132 B2 (2013).
- 16) K. Morita, FC Report, 37 (4), 139 (2019).
- 17) M. Sokol et al., Adv. Mater., 30, 1706283 (2018).

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