

# A strategy to reduce energy usage in ceramic fabrication

## — Novel binders and related processing technology —

Koji Watari<sup>1\*</sup>, Takaaki Nagaoka<sup>2</sup>, Kimiyasu Sato<sup>2</sup> and Yuji Hotta<sup>2</sup>

[Translation from *Synthesiology*, Vol.2, No.2, p.137-146 (2009)]

Because of serious global environmental problems, the ceramic industry has been concentrating on the reduction of energy usage during manufacturing. In this project, we have investigated low-energy processing techniques for ceramic components. Our research and development approach was carried out with the goal of realizing new ceramics that can be manufactured using conventional manufacturing processes and equipment at low-cost without significant degradation in material properties. After a careful investigation of possible technologies, we concluded that a decrease in the amount of organic binder is the most effective technique to promote low-energy processing, and have successfully developed a novel binder technology. Our technology and knowledge have contributed to greatly reduce the amount of energy required for ceramic fabrication through a collaborative research project with a private company.

**Keywords** : Ceramics, manufacturing, energy saving, binder, process, water

## 1 Introduction

Ceramics are used in the field of industrial machines, and have spread to various other fields including semiconductors and electronic parts, electronic devices, automobiles, processing, environment, energy, and biotechnology. They are recognized as important materials for supporting industry, together with metals and polymers.

Looking at the ceramic manufacturing process from the perspective of environmental load reduction, there are many issues. In Japan, industries including the ceramics industry produce about 40 % of the greenhouse gas generated by energy consumption. Comparing the changes of energy consumption unit index per industrial production index by industries, the index for ceramics related industries is about 1.5 times higher than those of iron and steel, chemical, and paper and pulp industries<sup>[1]</sup>. Particularly, resource saving, energy saving, or environmental load have not been sufficiently considered in the ceramic manufacturing process, and there are many issues in practicing highly efficient manufacturing. Therefore, the development of a process technology to promote energy saving in current ceramic manufacturing as well as the development of an innovative high-efficiency manufacturing process is in immediate demand.

With this social background, our research group has been engaging in R&D to develop a low environmental load manufacturing process including the development of energy-saving process for ceramics. In this paper, we shall present the results of our research and describe how we arrived at the development of the energy-saving process for the existing

ceramic manufacturing.

## 2 State of existing manufacturing process

Although ceramic manufacturing is a high-variety low-production activity compared to other materials such as metals and polymers, it requires many steps including mixing and dispersion, drying, forming, debinding, and sintering. Figure 1 shows the steps for ceramic manufacturing. First step is the preparation of raw material, where the binder and the solvent are added to the starting material such as raw powder and auxiliary agents, and mixing and dispersing the mixture. Auxiliary agent is an additive that promotes densification and expression of function. Binder is an organic additive used to add shape to the green body and to maintain strength. In general, polymer materials are used since they can be broken down and removed by heating. Ceramic products is obtained after following several steps including drying where the solvent is evaporated, forming where the powder is molded into certain shape, debinding where the added binder is removed, and sintering where the body is heated to high temperatures. In addition to these steps, exhaust gas decomposition is added to the manufacturing line, because the gas produced from the binder during debinding contains harmful substances.

Since ceramic manufacturing consists of multiple steps, the development of a manufacturing process involves not just the technological development of a single step, but must involve the steps before and after as well as the preliminary steps. For example, in a case where the formability of the powder is extremely low, it is necessary to investigate the material factors such as raw powder, binder, and solvent, as well as

1. Research and Innovation Promotion Office, AIST Tsukuba Central 2, Umezono1-1-1 Tsukuba 305-8568, Japan \* E-mail : koji-watari@aist.go.jp, 2. Advanced Manufacturing Research Institute, AIST Anagahora 2266-98, Shimo-shidami, Moriyama-ku, Nagoya 463-8560, Japan

Received original manuscript March 2, 2009, Revisions received March 25, 2009, Accepted March 27, 2009

the process control factors of mixing, dispersing, and drying. Only after several experiments, production, and assessment of prototypes, an optimal process condition can be reached. Therefore, ceramic manufacturing requires broad knowledge and experience from material preparation to sintering, in addition to high technological capability in each step. Japanese manufacturing gained high technological capability by repeating the process of extracting issues and then solving them. As a result, the Japanese ceramic products have been leading industrial competitiveness in the world. However, this technological capability may have become a black box of know-how, and no engineering investigations have been conducted. This has prevented technological development of the existing manufacturing process, and increased the distance between the site of production and academic research institutes studying production technology.

### 3 “Valley of death” in energy-saving process technology

It is thought that there are two “valleys of death” that must be overcome in introducing the energy-saving process technology to the site of production. For realization, there exist the “economic valley of death” concerning costs in comparison to existing technology and the “technological valley of death” concerning the introduction of the technology to the existing manufacturing line.

#### 3.1 Economic Valley of Death

The “economic valley of death” is the increased cost in introducing the new technology. The companies are constantly pressured to reduce cost and are wary about any investment in new equipment. Even if the investment is for a technology that is expected to surpass the conventional yield, the companies are reluctant if it departs from the existing process. Therefore, the new technology must be developed under assumptions that it can be incorporated into the existing process and that the existing equipments can be used. Also, since many of the manufacturing lines are in operation 24 hours, results must be obtainable with little change as possible.

#### 3.2 Technological Valley of Death

Various elemental technologies are suggested for energy saving in the manufacturing process. However, many such technologies have issues of complicating the steps, difficulty of incorporation in the successive process, reduction in workability, and of production of waste and hazardous materials. Also, the effort to achieve energy saving may require substantial changes in raw materials and in the existing manufacturing line. As a result, many developed technologies are not put into practical use at the site of ceramic production.

### 4 Scenario for achieving the objective

For the developed technologies to be incorporated effectively at the site of production, it is necessary to overcome the aforementioned two “valleys of death.” To overcome the “valley of death,” it is important to consider the incorporation of developed technology into the existing manufacturing line. In an effort to achieve energy savings of the entire production system, the steps may become complicated and the manufacturing cost tends to rise greatly when several factors are changed. Therefore, we focused on the step that consumes high amount of energy, and clarified the relationship between the process factor and the energy consumed. Based on that result, we investigated elemental technologies that could be incorporated into the existing manufacturing line.

#### 4.1 Consumption energy needed in ceramic manufacturing

Figure 1 shows the steps of the ceramic manufacturing and the flow of the input and output of the materials. Figure 2 shows the percentage of energy consumed in each step. From the result of Fig. 2, it can be seen that the steps that consume large quantities of energy are debinding, exhaust gas decomposition, and sintering. This is because heat energy is necessary for the removal of the organic binder in the green body, the conversion of exhaust gas to vapor or carbon dioxide, and for the ceramic firing. Moreover, energy efficiency is extremely low. Therefore, to reduce the amount of energy consumed by heat energy, it is necessary to: (1)

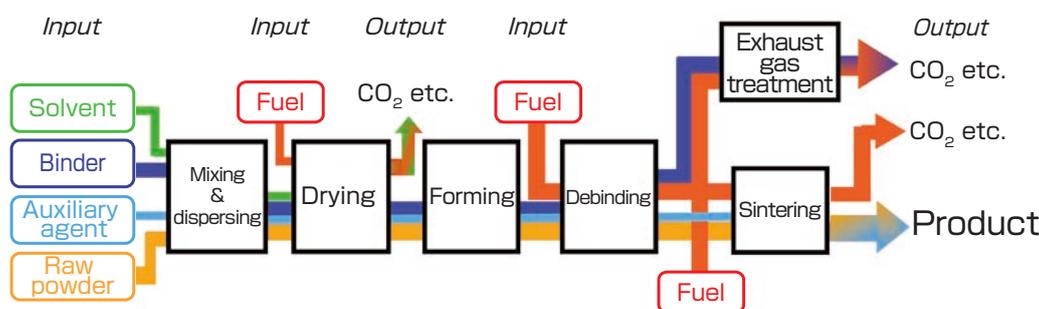


Fig. 1 Steps of ceramic manufacturing and the flow of resource materials.

use highly efficient firing furnace, (2) reduce heat energy by lowering the sintering temperature, and (3) reduce heat energy generated from debinding and exhaust gas decomposition.

#### 4.2 Approach from equipment development

For (1), taking for example the gas firing furnace, the energy needed to sinter the ceramic body at 1300 °C is about 2 % of the total energy consumed. The remainder is about 25 % to heat the furnace wall, about 17 % is heat loss from the furnace wall, and the loss from exhaust gas is over 50 %. Therefore, swift measures such as the development of a firing system for high-efficiency sintering are necessary. Recently, a microwave furnace and a regeneration furnace that recovers high temperature exhaust gas have been developed<sup>[2]</sup>. The development of a high-efficiency sintering furnace is extremely necessary for energy saving in the ceramic manufacturing line. However, this issue was removed from the scenario to achieve the objective, because the main issue here concerns the development of equipment.

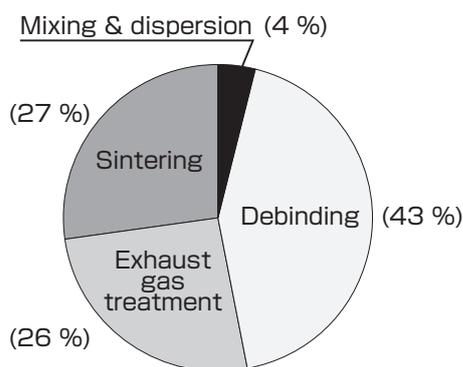
#### 4.3 Approach from sintering technology

For (2), one of the ways is to reduce the total amount of energy needed for heating by using the existing sintering equipment by developing a low-temperature sintering technology. To promote low-temperature sintering of ceramics, it is necessary to mobilize nanoparticle handling technology<sup>[3][4]</sup>, low melting point sintering additive technology<sup>[5]</sup>, dispersion technology<sup>[6]</sup>, surface coating technology (for auxiliary agents that may accelerate the sintering reaction of auxiliaries and particles), high-density forming technology, and others. All technologies work effectively for low-temperature sintering, and enables production of dense sintered body at firing temperature 100–300 °C lower than conventional sintering. However, there are problems such as shrinkage after firing due to addition of

nanoparticles, difficulty in controlling the shrinkage, changes of material property due to addition of low melting point auxiliary agent and pollution of material surface, and reduced workability by using high-density forming technology. Therefore, we withdrew from the approach based on sintering technology.

#### 4.4 Approach from binder technology

The method of (3) is the reduction or elimination of the currently used organic binder. Addition of organic binder enables formation of complex shapes and improves the strength of the green body. However, since an organic binder has low affinity for ceramic raw particle, it causes partial binder aggregation and also weakens the bonding strength between particles. Therefore, for good formability and shape maintenance after forming, a large amount of the organic binder must be added. Although the amount of binders differs according to size, thickness, shape, and processing of the green body, in general, it is 5 wt% or less in the case of dry mold product, 10 wt% or more for sheet mold product, and 20 wt% or more for complex shape product. Since the binder is unneeded after forming, it is removed from the body by heat decomposition or evaporation in the debinding step. The organic material used as a binder normally gasifies when heated at around 600 °C. Since sintering quality decreases when any binder remains on the powder surface as ash or carbon, precise process control is necessary in the debinding step. At the same time, the gas produced may cause structural defects such as pores, flaking, and warping in the green body and sheet, so the temperature must be increased gradually<sup>[7]</sup>. If the temperature at which the binder is completely eliminated is 600 °C, 60 hours of heating is necessary at a heating rate of 10 °C/h to reach the temperature, and 20 hours at 30 °C/h. Moreover, considering the heating and cooling times, the amount of energy required for debinding is extremely high.



**Fig. 2 Percentage of consumption energy in each process (all are laboratory level; energy required for powder production is not included).**

Consumption energy required to sinter 1 kg alumina. Organic binder additive: 10 mass%. Degreasing step: maintain 600 °C 1 hr (12 °C/h). Exhaust gas treatment step: maintain 900 °C. Sintering step: maintain 1400 °C 4 hrs (600 °C/h). For degreasing and sintering steps, 6 KW electric furnace was used. For exhaust gas treatment step, 1.4 KW electric furnace was used.

It is known that the gas generated by heat decomposition of binders contains organic material, and therefore, it is decomposed into harmless substances such as carbon dioxide and water, normally using the afterburner in the exhaust gas decomposition step. The heat decomposition temperature of many organic gases is 600 °C or above, and if the temperature of the afterburner is set above the heat decomposition temperature, the energy required for the treatment of exhaust gas becomes fairly considerable<sup>[8]</sup>. As shown in Fig. 1 and Fig. 2, the energy consumption related to binders is extremely high. If the amount of the organic binder used can be reduced by some sort of technological development, the amounts of heat energy required for debinding and exhaust gas decomposition can be reduced. Therefore, to promote energy savings of the existing ceramic manufacturing process, we decided to make our approach from the binder technology.

If the developed binder technology necessitates major changes in the existing manufacturing line, the initial objective will be

defeated. Therefore, it is important for the developed binder to have almost the same function as the conventional binder. Therefore, we considered a binder technology using a material with high binder function but which can be used at fewer amounts.

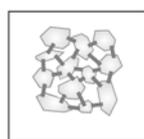
## 5 Development technology and research result

### 5.1 Extraction of key technology

Figure 3 summarizes the functions of the binder. The functions can be roughly categorized into two types. First is to firmly bond the particles and to stably maintain the shape (shape retention). The ceramic green body must have a certain level of strength. One of the important functions of the binder is to maintain the given shape against the weight of the green body itself as well as against the strain of handling at the site of production. The binder with such function is used in the fabrication of ceramic films, sheets, and large products. Second is for the binder to give both fluidity and shape retention to the particles assembly (plasticity). That is, when the raw powder and the binder are mixed, the particles are bonded together weakly through the binder and retain shape (shape retention). Moreover, the shape is changed by simply applying certain amount of force (fluidity), and the shape must be maintained when the force is removed (shape retention). Plasticity is a function required in the production of complexly shaped products such as in extrusion and injection molding.

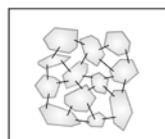
We investigated the materials with high binding functions such as shape retention and plasticity, while aiming to reduce the amount of organic material. Moreover, other than the binder function, we considered properties that are sought in binders in general, such as: (1) low cost, (2) no reaction to raw powder, (3) being soluble in water and solvent, (4) ashes not remaining after decomposition and evaporation, and (4) decomposition gas not being harmful or corrosive.

#### Shape maintenance

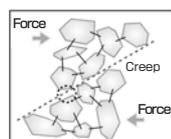


(1) Strong bond

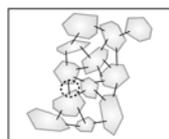
#### Plasticity (fluidity, shape retention)



(1) Weak bond (shape retention)



(2) Bond breaking (fluidity)



(3) Rebonding (shape retention)

Fig. 3 Functions of binder.

### 5.2 Reactive binder technology

We selected the method of reducing the amount of organic binder by adding the same function as the existing organic binder used in the manufacturing line to very small amounts of organic substances. First, we developed a new binder technology where the “shape retention” function (Fig. 3) is expressed with minimum amount of organic material. Highly reactive organic molecules that would be binders were anchored onto the surface of the ceramic raw particle in a form of a film. The whole particle assemblies were formed into desired shapes, the reaction trigger (external stimulus) that excites chemical reaction was applied, and the molecules of the organic film layer were chemically bonded, to create the ceramic green body in which the particles were bonded together firmly. Figure 4 shows the concept of the technology to reduce the amount of organic binders. Unlike in the conventional forming method, the shape of the green body could be maintained efficiently with only a small amount of binders, because of the structure in which the binder molecules linked the ceramic particles with strong chemical bond. Also, partial aggregation of the binder could be prevented since the binder molecules were fixed on the particle surface in film form, and therefore the amount of the organic binder could be reduced.

In this forming method, it was not desirable for the particles to bond before the particle assembly formed the desired shape. Therefore, the chemical bond was introduced at an arbitrary moment by using external stimulus as a reaction trigger. Irradiation of electromagnetic waves (ultraviolet rays<sup>[9]</sup>, microwave<sup>[10]</sup>) and heating at 100 °C<sup>[11]</sup> were used as reaction triggers. When ultraviolet rays were used as reaction triggers, we succeeded in fabricating a solid body, as strong interparticle bond occurred by coating the ceramic particles with amino groups and phenyl azide groups<sup>[9]</sup>. For organic materials that react with microwave, we conducted experiments by referring to past reports, but were unable to reproduce them. Therefore, we looked at water, a high dielectric loss substance that absorbs microwave, and used water-soluble carbodiimide with water as a microwave reactive binder. The oxyethylene (-C<sub>2</sub>H<sub>4</sub>O-) that composes the water-soluble carbodiimide becomes the hydrophilic segment,

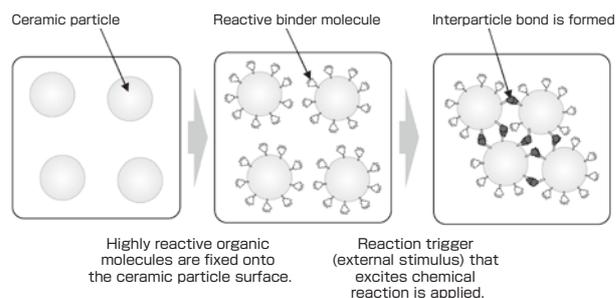


Fig. 4 Fixing of organic molecule and application of reaction trigger to reduce the amount of organic binder.

absorbs microwave, and generates heat. On the other hand, the carbodiimide (-N=C=N-) of the reactive segment firmly bonds the segments present on other particle surfaces<sup>[10]</sup>.

While the UV reactive binder was effective for forming ceramic sheets, ultraviolet rays did not readily reach the interior in large samples since they have short wavelengths. Because microwave possesses longer wavelengths compared to UV, the microwave reactive binder was effective for the fabrication of a large green body. The ceramic green body fabricated using these binders contained only 0.5 wt% of organic substance, and the shape could be maintained with significantly lower amount of organic substance compared to the conventional method. This is an application of surface coating and reactive trigger technologies that we nurtured as elemental technologies to realize the energy-saving process.

### 5.3 Inorganic binder technology

Next, we investigated the inorganic binder technology using inorganic substances, for the total elimination of organic binders that express a “plasticity” function (Fig. 3). First we focused our attention to the fact that clay minerals possess plasticity. Although the expression mechanism of plasticity in clay minerals has not been clarified, it is thought to be related to: (1) effect of water film formed on the particle surface, and (2) slippage caused by the intercalation compound of clay. We focused on (1) and studied the inorganic materials that can retain water. Assuming that the mechanism whereby a new inorganic binder provides shape retention and fluidity is the same as in clay minerals where it is “the effect of water film formed on the particle surface” that does so, we looked for materials in which there was interaction between the particle surface and water, and which could retain sufficient amount of water. Hydrates chemically contain large quantities of water, and take multiple forms in accordance to the chemical bonds of the component elements. Therefore, we thought hydrates could be applied widely to various ceramics. Also, the advantages of hydrates were that most of them were of low cost and had high purity compared to clay minerals. Hence, our research started with the technology for hydration material with fluidity and shape retention properties.

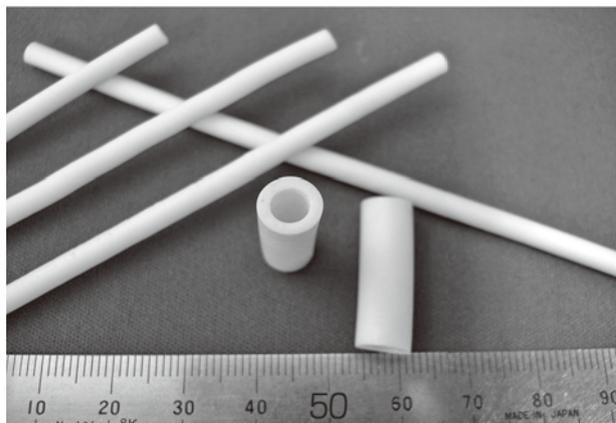
There is a saying that “one who controls binders controls ceramics,” and the importance of binder technology is extremely high in the ceramic manufacturing process. Therefore, the guideline for binder development is almost never publicized. Also, since the binder function involves complex factors, it is difficult to express it in physical quantities. Therefore, at the sites of research and production, for example, in investigation of extrusion technology, it is “ultimately determined by actually extruding the product on the machine.” In the first phase of the research, the definitions of binder functions (shape retention and fluidity) were set originally, and these were evaluated from the behavior of the samples formed by the extruder. Since this method was based

on the observation of the extruded sample and on the relative assessment of shape retention and fluidity, it was far from an absolute assessment of physical quantities. However, it was extremely useful in narrowing down the material and seeking optimal conditions since assessment could be done easily with a small amount of material<sup>[12]</sup>.

For the development of inorganic binder technology, we decided to look for inorganic material that showed the same plasticity behavior as the existing organic binders and clay minerals based on the assessment of binder functions. As a result, for the expression of plasticity of alumina ceramics, we found that the addition of hydraulic alumina ( $\rho$ -alumina) was useful. Hydraulic alumina separated out onto the surface of alumina particle as hydrate particles by hydration. It was found that the particles contained large amounts of water and had high binder functions. As a result, we succeeded in the extrusion of alumina ceramic tubes without using organic binders depending on the added amount, as shown in Fig. 5<sup>[13]</sup>. Moreover, a similar effect was seen in other hydrates, and we confirmed an expression of plasticity in various ceramics.

### 5.4 Assessment of inorganic binder particle surface

The development of the inorganic binder mentioned in the previous section succeeded by hypothesizing that the shape retention and fluidity properties were due to the water film existing on the binder surface. We considered the mechanism of plasticity expression in inorganic binders as follows. Shape is retained since the ceramic particles obtain mild bonding force through the surface tension of water that accompanies the inorganic binder. On the other hand, fluidity is gained because the water film works like a bearing and lubricity occurs between the surfaces of the inorganic binder. While the effect of surface tension could be understood readily, experimental demonstration was necessary for the expression of lubricity. Therefore, our group established the measurement technology for surface-surface interactive force using the atomic force microscope (AFM) to make actual measurements of the interactive force that act between the

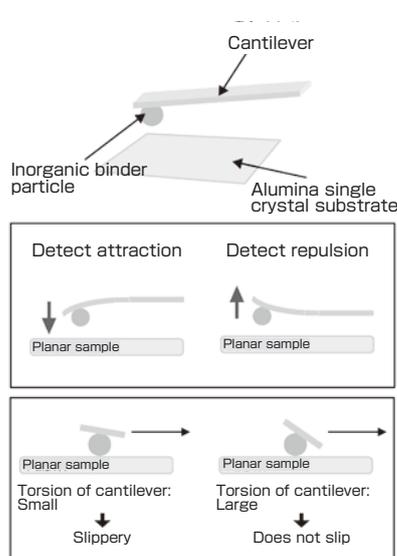


**Fig. 5 Alumina ceramics fabricated using inorganic binder technology.**

surfaces of the ceramic particle and the inorganic binder.

AFM detects the interactive force generated between the sample and the probe as the displacement of cantilever (a plate spring for force detection). Conventionally, it is used as a “microscope” to obtain morphological information of the sample surface, but it can be used to actually measure the interactive force between the particle sample and planar sample by attaching the desired particle onto the tip of the cantilever to be used as a probe (colloid probe method) (Fig. 6). The force in vertical direction (attraction and repulsion) against the planar sample can be estimated from the deflection of the cantilever, and the force in parallel direction (lateral direction) against the planar sample can be estimated from the torsion of the cantilever. We attached the hydraulic alumina particle to the tip of the cantilever, and measured the interactive force with the alumina single crystal substrate set as a model substance of ceramic particle. This was the first attempt to assess the interactive force that the inorganic binder may exert on the ceramic surface.

As a result of measurements, a repulsion that could not be explained by general electrostatic interaction or van der Waals force was detected between the inorganic binder particle and the alumina substrate. It was concluded that this was an interactive force called the hydration repulsion. When a substance with high hydrophilic property is placed in water, a thin layer (hydration layer) where the water molecules are bound and structured by hydrogen bonds is formed on the surface. The repulsion force generated when a foreign surface approaches is hydration repulsion. Also, from the measurement of interactive force in the lateral direction, it was found that a more “slippery” condition occurred when hydration repulsion was observed between the surfaces compared to when there was no hydration repulsion (no hydration layer)<sup>[14]</sup>.



**Fig. 6 Measurement of inter-surface interactive force.**

The existence of the water film that was hypothesized in the development of the inorganic binder was confirmed as a hydration layer that caused hydration repulsion. Also it was indicated that the fluidity was expressed since the hydration layer increased the lubricity between the inorganic binder and the ceramic particle.

## 6 Discussion

The flow of our research is summarized in Fig. 7. Many researchers including us worked on the elemental technologies to realize the energy-saving process, and arrived at various findings. This corresponds to *Type 1 Basic Research*, and some elemental technologies are shown on the left of Fig. 7. However, many of the elemental technologies developed fell into the “valley of death” due to poor compatibility with the existing manufacturing process or increased costs. In this research, we first narrowed down the conditions assuming that the result will be used at the site of production. As a result, the technological and economic issues that may lead to the “valley of death” were clarified. The R&D to solve these issues corresponds to *Type 2 Basic Research*.

The development of low-temperature sintering technology is an attractive area in material process research. There are diverse approaches to low-temperature sintering, and an effective energy-saving process seems to be possible by combining various methods. However, all of the elemental technologies for low-temperature sintering accompany great material transfer for ceramics, and greatly alter the conditions of the existing manufacturing process. On the other hand, the newly developed binder technology has the characteristics of: (1) not greatly changing the property of the raw powder, and (2) not enforcing great material transfer for ceramics. Since we thought we should construct an energy-saving process without requiring great changes to the existing manufacturing process, the research topic became clear. Considering the positioning of this research, it corresponds to the definition of the research theme. However, this definition is “to find simplicity in complexity,” and is based on the knowledge and experience gained over the years.

Since binder function changes significantly due to the type and amount of the raw powder, solvent, and binder, there is an extremely small number of systematic research publication. Also, since several binders are combined to obtain an optimal forming function, the roles of each binder are intricately intertwined. Therefore, although positioned as an extremely important element of ceramics R&D, there was hardly any scientific investigation of binders. Therefore, we decided to aim for reduction or elimination of the organic binder while maintaining the same function as the existing organic binder. We worked on the organic and inorganic binder technologies under these conditions. This is an extraction of technologies to develop a novel binder technology.

Various conditions became clear as the research progressed and the elemental technologies were refined. By conducting R&D for the expression of plasticity in inorganic binders and R&D of the particle surface analysis technology using AFM colloid probe method, the relationship between the particle surface and water was investigated from macro and micro levels. As a result, the importance of water in the plasticity of ceramics, as well as the ways of selection of inorganic binders to increase the water content were demonstrated quantitatively, and we were able to approach the technologies for reduction and elimination of organic binders based on scientific evidence. These researches correspond to the core and basic researches of the *Type 2 Basic Research*.

The flow of inorganic binder research for the reduction and elimination of the organic binder with plasticity shifted from: (1) development of binder assessment technology, (2) process research of inorganic binders, (3) research of water-particle interface, and (4) clarification of process factors that control the inorganic binder technology. As a result, it was shown that inorganic material with high water content could be used as an inorganic binder expressing plasticity. This is the outcome of the *Type 2 Basic Research*.

Discussions within the group of the importance of water in clay minerals and inorganic binders led to the organic binder with hydrophilic properties or those that contain water as candidates of organic binders that reacted to microwave, and this led to the development of a microwave reactive binder. Although the roles of water in inorganic binders

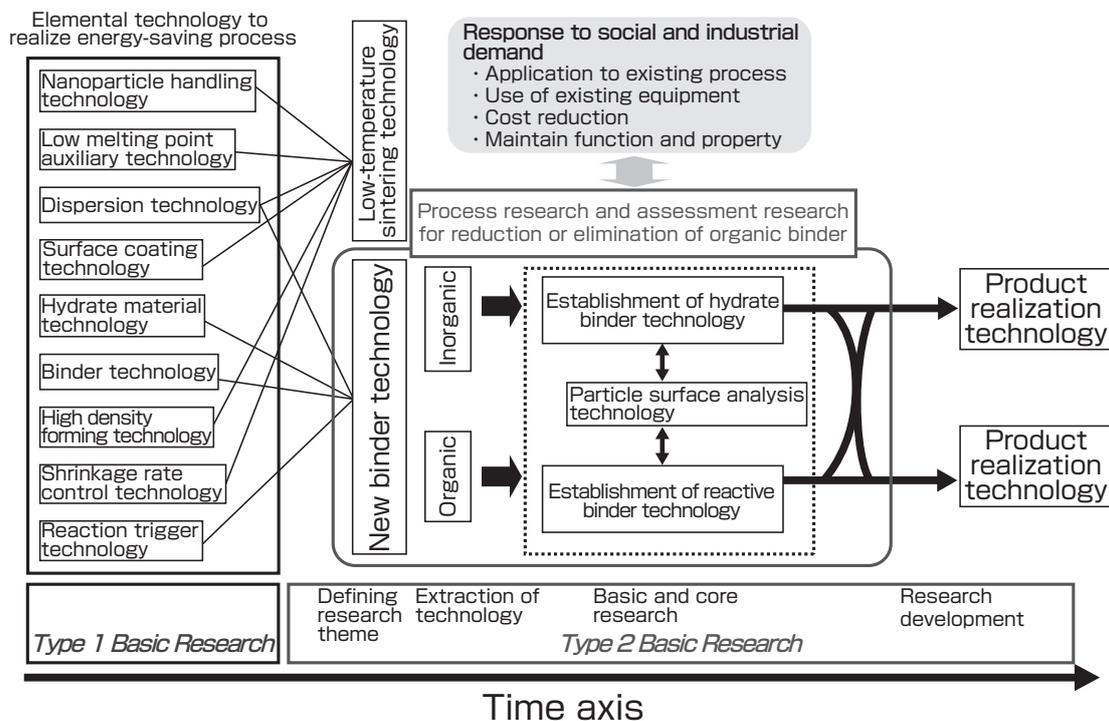
and microwave reactive binders are different, in the greater concept of binders, “water” was an important keyword.

Based on the results and findings obtained, a product was realized by a major material company through joint research. These researches correspond to *Product Realization Research*, and the efficacy of the developed technology was tested in the manufacturing lines.

The advantages of the developed binder include its technological excellence as well as the low cost of incorporating this technology. Since the reactive and inorganic binders can be placed in solvent along with the raw materials and auxiliaries and then mixed and dispersed, no new step is added. Moreover, the binder is used in large amounts for other purposes, and the cost of material is low.

### 7 Evaluation of result and future developments

As a result of engaging in research to fulfill the demand of society and industry such as adaptation to the existing process, use of existing equipment, and low cost, we believe we were able to develop a technology that can be put to actual practice, as the boundary conditions of the technology as well as the elemental technologies became clarified. Extraction of core technology is to find “simplicity in complexity” of the manufacturing process that seems to be complexly intertwined. If the boundary conditions of the research and the core of the technology to be developed are known in the early phase of R&D, resources can be introduced efficiently



**Fig. 7 Building of low-environment load process and mutual relationship in existing ceramic manufacturing steps.**

and the speed of R&D can be accelerated considerably. However, since the manufacturing process is composed of the accumulation of elemental technologies and linkage between the technologies, more time than expected is required for the extraction of core technology. To solve this problem, it is necessary to systematize the elemental technologies and extract the issues and core technology to maintain the succession of the process. This is to create a research scenario for industry. By doing this, we believe various issues in technological realization can be understood, plans to solve the issues can be created, and the speed of research can be further increased.

However, information on the materials and the processes that we can learn from private companies are limited. Therefore, to engage in R&D while writing the scenario for industry, it is important to obtain human resources that have long years of experience and wide-ranging knowledge in the field, and to thoroughly understand the theme and its background.

Also, we believe the public research institute of industrial technology is expected to have a “universal thinking” that does not weather over the years. This is one of the outcomes of the *Type 2 Basic Research* conducted by AIST. In our research, we set the objectives to reduction and elimination of the organic binder, and we obtained a general solution that water on the surface of the raw particle greatly affects the formation of ceramics. When working on research topics that arise from the site of production in joint research, most are technological contributions and one-shot service to a company, but the outcome of *Type 2 Basic Research* may become extremely meaningful by also considering the construction of concepts with scientific universality.

The final target of this research is the complete elimination of organic binders. We succeeded in simple extruding using an inorganic binder. By achieving this technology, ceramic manufacturing without debinding and exhaust gas treatment became possible, and as a result the reduction in CO<sub>2</sub> emission was about 70 % (see Fig. 2). Also, since the amount of the reactive binder was one to two digits less compared to the amount of binders in the ordinary process, the debinding and exhaust gas treatment steps can be eliminated in the future. The estimated reduction in CO<sub>2</sub> will have an impact on the ceramics industry. However, in reality, the demands of industry cannot be met without conventional organic binders in members of certain size or complex shape. Therefore, the current practice is to add small amounts of organic binders to the newly developed inorganic binder.

Since the developed technology can be used in the current ceramic production process, we are working actively to spread the technology. We have engaged in R&D based on materials and processes, and wish to continue the R&D while looking at the development of manufacturing equipment to

improve the energy-saving property based on the findings from this research project. Particularly, we shall fuse the R&D of materials and process with the R&D of manufacturing equipment, bring about a synergy effect, and contribute further to the development of the energy-saving ceramic process.

## Acknowledgement

We are deeply grateful for the cooperation and advice of the researchers and engineers of the companies with whom we engaged in joint research, as well as many people including the researchers of AIST.

## References

- [1] The Institute of Energy Economics, Japan: *Keizai Sangyosho/EDMC Suikei (EDMC Handbook of Energy and Economics Statistics)*, 62-13 (2003) (in Japanese).
- [2] K. Watari: Recent trends in low-energy process technology in ceramics, *Materiaru Integureshon (Material Integration)*, 19, 2-9 (2006) (in Japanese).
- [3] Y. Hotta, C. Duran, K. Sato, T. Nagaoka and K. Watari: Densification and grain growth in BaTiO<sub>3</sub> ceramics fabricated from nanopowders synthesized by ball-milling assisted hydrothermal reaction, *J. Euro. Ceram. Soc.*, 28, 599-604 (2007).
- [4] J. Qiu, Y. Hotta, K. Watari and T. Mitsuishi: Enhancement of densification and thermal conductivity in AlN ceramics by addition of nano-sized particles, *J. Am. Ceram. Soc.*, 89, 377-80 (2006).
- [5] K. Watari, M. C. Valecillos, M. E. Brito, M. Toriyama and S. Kanzaki: Processing and thermal conductivity of aluminum nitride ceramics with concurrent addition of Y<sub>2</sub>O<sub>3</sub>, CaO and Li<sub>2</sub>O, *J. Am. Ceram. Soc.*, 79, 3103-8 (1996).
- [6] T. Isobe, Y. Hotta and K. Watari: Preparation of Al<sub>2</sub>O<sub>3</sub> sheets from nano-sized particles by aqueous tape casting of wet-jet milled slurry, *J. Am. Ceram. Soc.*, 90, 3720-24 (2007).
- [7] Y. Kinemuchi, R. Ito, H. Ishiguro, T. Tsugoshi and K. Watari: Binder burnout from layers of alumina ceramics under centrifugal force, *J. Am. Ceram. Soc.*, 89, 805-809 (2006).
- [8] K. Watari, K. Sato, T. Nagaoka and T. Ozaki: Binder process and energy-saving sintering technology, *Shin Zairyo Series, Kankyo Taio-Gata Ceramics No Gijutsu To Oyo (New Material Series, Technology and Application of Environment-Friendly Ceramics)*, 13-27 (2007) (in Japanese).
- [9] K. Sato, Y. Hotta, T. Nagaoka, K. Watari, M. Asai and S. Kawasaki: Mutual linkage of particles in a ceramic green body through potoreactive organic binders, *J. Ceram. Soc. Japan*, 113, 687-691 (2005).
- [10] K. Sato, M. Kawai, Y. Hotta, T. Nagaoka and K. Watari: Production of ceramic green bodies using a microwave-reactive organic binder, *J. Am. Ceram. Soc.*, 90, 1319-22 (2007).
- [11] C. Duran, K. Sato, Y. Hotta and K. Watari: Covalently connected particles in green bodies fabricated by tape casting, *J. Am. Ceram. Soc.*, 90, 279-282 (2007).
- [12] T. Nagaoka, C. Duran, T. Isobe, Y. Hotta and K. Watari: Hydraulic alumina binder for extrusion of alumina ceramics, *J. Am. Ceram. Soc.*, 90, 3998-4001 (2007).
- [13] T. Nagaoka, K. Sato, Y. Hotta, T. Tsugoshi and K. Watari: Extrusion of alumina ceramics with hydraulic without

organic additives, *J. Ceram. Soc. Japan*, 115, 191-94 (2007).

- [14] M. Polat, K. Sato, T. Nagaoka and K. Watari: Effect of pH and hydration on the normal and lateral interaction forces between alumina surfaces, *J. Colloid Interface Sci.*, 304, 378-387 (2006).

---

## Authors

### Koji Watari

Completed doctorate course at the Graduate School of Engineering, Nagaoka University of Technology in March 1990 (Doctor of Engineering). Joined the Government Industrial Research Institute, Nagoya, Agency of Industrial Science and Technology, Ministry of International Trade and Industry in April 1990. After working at the National Industrial Research Institute of Nagoya, became leader of the Low Environmental Load Sintering Technology Research Group, Advanced Manufacturing Research Institute, AIST in April 2001. Leader of Advanced Sintering Technology Research Group since April 2004. Currently Senior Planning Manager, Research and Innovation Promotion Office, AIST. Also stayed at the Pennsylvania State University (doctorate researcher) during 1998~1999, and worked at the R&D Division, Agency of Industrial Science and Technology, Ministry of International Trade and Industry during 1999~2000. Visiting professor at the Nagaoka University of Technology since February 2004, and Gebze Institute of Technology (Turkey) in March 2009. Advancement Award, The Ceramic Society of Japan (1997); Academic Award, The Nagai Foundation for Science and Technology (2002); Richard M. Fulrath Award, American Ceramic Society (2006); Research Advancement Award, Japan Society of Powder and Powder Metallurgy (2007); Award of Merit, Workshop on Anisotropic Science and Technology of Materials and Devices (Turkey) (2008); Certificate of Merit, The Kazuchika Okura Memorial Foundation (2008); and Best Scientific Photography Award, The Ceramic Society of Japan (2009); Best Paper Award, *Journal of the Ceramic Society of Japan* (2009); and others. Acted as guest editor for the *MRS Bulletin* (June 2001), *Journal of the Ceramics Society of Japan* (February, March 2008), and others. Has worked on the R&D of ceramic process technology, reactive site control process technology, and high heat conductive ceramics. In this research, was in charge of planning the whole proposal, research management and operation, and low-temperature sintering technology.

### Takaaki Nagaoka

Graduated from the Faculty of Science, Tohoku University in 1985. After working at Nihon Cement K.K. (currently Taiheiyo Cement Corporation), joined the Government Industrial Research Institute, Nagoya, Agency of Industrial Science and Technology in 1987. Worked on the next-generation core technology research project. Dispatched to Fine Ceramics Research Association during 1996~1998. Currently working on the R&D of inorganic binder at AIST since 2001. In this research, worked on inorganic binder technology.

### Kimiyasu Sato

Withdrew from the doctorate course at the School of Science, The University of Tokyo in 1997. Became Fellow Researcher

of the National Institute for Research in Inorganic Materials, Science and Technology Agency in 1997. Became CREST researcher of the Japan Science and Technology Agency in 2000. Joined AIST in 2002. Visiting researcher at the Stockholm University during 2008~2009. Currently works in Inorganic-Based Plastics Group, Advanced Manufacturing Research Institute, AIST. Doctor of Science. In this research, worked on reactive organic binder technology and inorganic and organic interface assessment technology.

### Yuji Hotta

Completed doctorate course at the Graduate School of Science, Hokkaido University in 1997. Joined the Government Industrial Research Institute, Nagoya, Agency of Industrial Science and Technology, Ministry of International Trade and Industry (current AIST) in 1997. After being dispatched to the Fine Ceramics Center during 2000~2001, became visiting researcher at the Ytkemiska Institutet (YKI: Institute for Surface Chemistry) of Sweden during 2001~2002. Currently leader of Inorganic-Based Plastics Group, Advanced Manufacturing Research Institute, AIST. Works on nanoparticle handling technology, dispersal technology for ceramic particles, and forming process technology. In this research, was in charge of low-temperature sintering technology using nanoparticles.

---

## Discussion with Reviewers

### 1 Energy saving and reduction of CO<sub>2</sub> emission by the developed binder

**Comment and question (Nobumitsu Murayama, Advanced Manufacturing Research Institute, AIST)**

What were the degrees of reduction in energy and CO<sub>2</sub> emission, or how much is expected by using the inorganic binder or by reducing the organic binder?

**Answer (Koji Watari)**

By using the inorganic binder, so far we have succeeded in simple extrusion forming of alumina and silicon nitride ceramics, and the reduction in CO<sub>2</sub> emission for manufacturing was about 70 % (see Fig. 2). In the actual production line, the amounts of reactive and inorganic binders added are different depending on the forming process, as well as the type, size, and shape complexity of the members. Also, conventional binders must be added in some cases. I have heard that the CO<sub>2</sub> emissions were greatly reduced at the companies to which the technology was transferred, but I shall decline disclosure of specific figures and details of the reduction due to the limitation of joint research.

In the future, we would like to estimate the necessary amount of binders according to the member specs (material, size, shape, etc.) at actual equipment level, and quantitatively assess the CO<sub>2</sub> emission and the relationship of reductions based on the estimate.

### 2 Cost increase or decrease in introducing the developed binder

**Comment and question (Toshimi Shimizu, Research Coordinator, AIST)**

It is described that the cost factor of the newly developed technology is important from the perspective of "economic valley of death" in the energy-saving attempts for manufacturing process. What are the costs of introducing reactive and inorganic binders?

**Answer (Koji Watari)**

One of the advantages of the binder we developed is the low cost of introduction, in addition to its technological excellence. The reactive and inorganic binders are simply mixed

and dispersed in the solvent along with the raw materials and auxiliaries, and no new step is added. Also, the binder we used is abundantly available, and the cost of material can be kept low.

### **3 Relationship between the elimination of organic binders and the introduction of inorganic binders**

#### **Comment and question (Toshimi Shimizu)**

You stated that the objective for the realization of energy-saving process is reduction or elimination of organic binders. Does this mean “elimination = introduction of inorganic binders”?

#### **Answer (Koji Watari)**

It is as you have indicated. The introduction of inorganic binders is the elimination of organic binders, and this leads to significant reduction of CO<sub>2</sub> emission in the ceramic manufacturing process.

### **4 Relationship between material production and assessment technology**

#### **Comment and question (Nobumitsu Murayama)**

In the development of the inorganic binder, first the binder assessment technology was developed, then followed the development of the inorganic binder. This research progression is very interesting. In ordinary material process research, first, the material is developed and then assessed. However, in the research for a new function, it is necessary to establish the assessment technology of that new function. Please comment on the relationship between material production and assessment.

#### **Answer (Takaaki Nagaoka)**

In the R&D of material and process through search of materials and their optimization, it is necessary to conduct tests for assessments. In this research, however, there were two limitations. One was that there was a lack of assessment method for binder functions. As a result, “ultimately it was determined by actually extruding the product on the machine.” Second was that high quantity (several hundred grams) of samples was needed per assessment on an actual machine. Although that amount may be small at the site of production, it was difficult at AIST when considering the time and labor needed. This was more pronounced when several assessments had to be done. Therefore, we set the definition of binder functions (shape retention and fluidity) for extruding, and developed a method for assessing the functions with minimum amount of samples. As a result, we could easily and quickly narrow down the prospect samples using small amounts of samples.

When searching for new material functions, if there is no function assessment technology at the laboratory level, then the researchers themselves must establish the assessment technology. Although this seems to be taking the long way around, by simultaneously developing the assessment technology, the researcher can think deeply about the meaning of the expressed function, and can work on material production from the same perspective as the person doing the assessment. Moreover, by accumulating the assessment technology, the developed technology and material together become a bundle of highly original, advanced knowledge.

### **5 Significance of scientifically investigating the know-how technology**

#### **Comment and question (Nobumitsu Murayama)**

It can be said that this research is a scientific investigation of the know-how technology for binders. While this will certainly promote the advancement of manufacturing process, disclosure of the companies' know-hows, which is the primary practice in the ceramics industry, may lower the competitiveness of these companies. Please comment on the significance of scientific investigation of a technology that was dependent on experience in

the material process field.

#### **Answer (Koji Watari)**

As the reviewer indicated, the basic and core research of binder technology is a scientific consideration of a technology that relied on corporate experience, and in the future, this may lower the competitive edge of the Japanese ceramics companies. Therefore, as a research leader, I considered the contents of patent and paper that will be publicized, explained carefully to the companies engaging in joint research, and disclosed some of the research results with their approval.

Scientific investigation of a technology that relied on experience, i.e. understanding the scientific basis of a technology, systematization of a technology, and extraction of the major factors, are extremely important outcomes of a public research institution like AIST, which is a collection of research professionals. Also, one of the roles of AIST is to find how to overcome the “valley of death” in a scientific perspective, and I think we can provide appropriate elemental and alternative technologies by scientific investigation of a technology that was dependent on experience.

Many of the companies that engaged in joint research with us set the ultimate objective as “development of high value-added product and improvement of production efficiency through basic and core research.” They became aware of the importance of basic and core research and demanded better research results. I think this is an indication that the companies want to make better products and improve production efficiency by scientifically investigating their know-hows that were dependent on experience. On the other hand, technological know-hows are important assets for the companies, and must be handled with care.

### **6 Technological transfer of innovative material process that requires total change in the manufacturing line**

#### **Comment and question (Nobumitsu Murayama)**

To overcome the economic valley of death that you mentioned, I am certain that it is more efficient if “the technology developed is technology that can be incorporated into the existing process, and the existing manufacturing equipment can be used.” However, there is always a possibility of technological transfer of innovative material process that requires total change in the manufacturing line. In case of the latter, one of the ways may be for a public research institute like AIST to have a prototype manufacturing line. Can you comment on this.

#### **Answer (Koji Watari)**

Many private companies are reluctant to invest newly in their manufacturing line. Considering the importance of the technology and its dissemination, it may be greatly significant for a public research institute like AIST to create a prototype manufacturing equipment for trial manufacturing. However, investment is large. Therefore, before making the new investment, it is necessary to set up a business model including the possibility of alternative technology, procurement of user and cost of maintenance, extraction of key application, and understanding of market trend.