Additive manufacturing of ceramic components

—Towards innovation of ceramic industry—

Tatsuki OHJI

Aiming for innovative ceramic manufacturing technologies which enable creative and novel products, a national R&D project "High-Value Added Ceramic Products Manufacturing Technologies (HCMT)" has been initiated since 2014 as part of the Council for Science, Technology and Innovation (CSTI), Cross-ministerial Strategic Innovation Promotion Program (SIP), "Innovative design/manufacturing technologies" program in Japan. The project deals with two key technologies: additive manufacturing (AM) for realizing complex-shaped ceramic products and reducing their lead-times, and hybrid coating on 3D bodies for enhancing their functionality and durability. Following an overview of this project and a brief description on the general status of AM technologies, this article focuses on the R&D strategies and the latest achievements on AM of ceramics in this project. Among a variety of AM approaches, we employ two AM technologies for making ceramic green bodies; powder layer manufacturing (powder bed fusion or indirect selective laser sintering) and slurry layer manufacturing (vat photo-polymerization or stereolithography), because of their dimensional accuracy, shape-flexibility, density-adjustability, *etc.* The former is a dry forming process, and is suitable for large/porous components, while the latter is a wet one, being good for small/dense parts. In addition, intensive research efforts are being devoted to ceramic laser sintering (direct selective laser sintering) which enables concurrent forming and sintering (saving post-sintering-process). This paper describes several 3D prototype models produced for various application targets using the developed AM technologies, which are never attainable with conventional methods. The current issues and future perspective for AM of ceramics will be addressed and discussed as well.

Keywords: Additive manufacturing, ceramics, components, powder, slurry, powder bed fusion, stereolithography, laser sintering, ceramic industry

1 Introduction

Because of their unique and excellent material properties, ceramics are often used as key parts in many advanced products and systems in a variety of fields including manufacturing, energy, environments, IT, electronics, optics, bio-technologies, and transportation. It is also noteworthy that Japanese ceramic industries have maintained the world's highest-level manufacturing technologies, which have brought about almost a half of the global market share of ceramic-related products, thanks to their incessant efforts for technological innovation.

Ceramic manufacturing process has been composed of several miscellaneous steps including powder preparation, mold making, granulation, forming, dewaxing, sintering, machining, finishing, *etc.* (Fig. 1, top). In addition, some of the steps such as granulation, dewaxing and sintering require a great deal of thermal energy, indicating higher ratios of labor and energy expenditures to the total production cost in comparison with those of other materials. As a result, production from countries of lower labor and energy costs has been gradually increasing in recent years, along with their progress of manufacturing technologies. To maintain and consolidate the technological superiority and international competitiveness of Japan's ceramic industry, it is now crucially required to develop innovative manufacturing technologies which enable us to produce creative and novel products of high value. For this purpose, a national R&D project "High-Value Added Ceramic Products Manufacturing Technologies (HCMT)" has been initiated since 2014 in the CSTI, SIP, "Innovative design/manufacturing technologies" program of the government of Japan.[1]-[3] The HCMT project intends to integrate the above-stated miscellaneous steps of manufacturing process into the two key technologies, "additive manufacturing (AM)" and "hybrid coating (HC)" (Fig. 1, middle), which bring about many advantages in terms of production process as well as product performance. AM realizes complex-shaping of ceramic products and reduces their lead-times as will be described later, while HC provides better surface modification of products and enhances their functionality and durability,[4] strengthening the international

Fellow, AIST　2266-98 Anagahora, Shimo-Shidami, Moriyama-ku, Nagoya 463-8560, Japan　E-mail: t-ohji@aist.go.jp

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competitiveness of Japan's ceramic industry. This article will first briefly outline the HCMT project and describe the general status of the current AM technologies; then it will focus on the R&D strategies and the latest achievements on AM of ceramics in this project.

2 Overview of HCMT project

The HCMT project deals with R&D on "product manufacturing (PM)" as well as AM and HC. Figure 2 shows the R&D items in each of AM, HC, and PM. In AM, powder-layer manufacturing (PLM) and slurry-layer manufacturing (SLM) technologies are being developed for actualizing mold-free production of green (or formed) bodies of complex-shaped components, in addition to ceramic laser sintering (CLS) which realizes concurrent forming and sintering. HC focuses on hybrid aerosol deposition (HAD) and fine-particle thermal spraying (FTS) for highly adhesive coating onto 3D shaped substrates (including polymer and metal), enhancing functions and durability of products. Based on these two developed platforms, we try to establish manufacturing technologies for a variety of target products of high value in

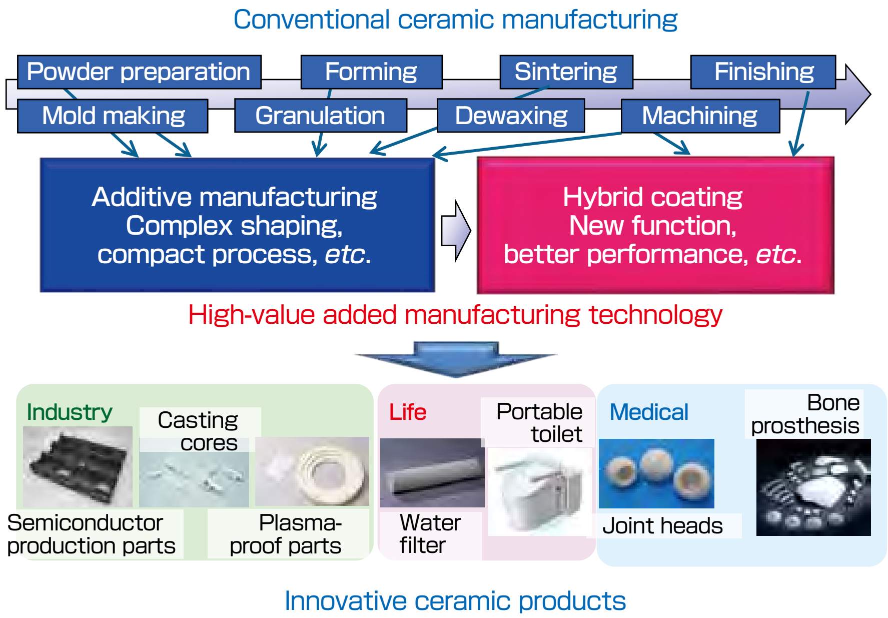


Fig. 1 Conventional ceramic manufacturing process and high-value added manufacturing technology to be developed

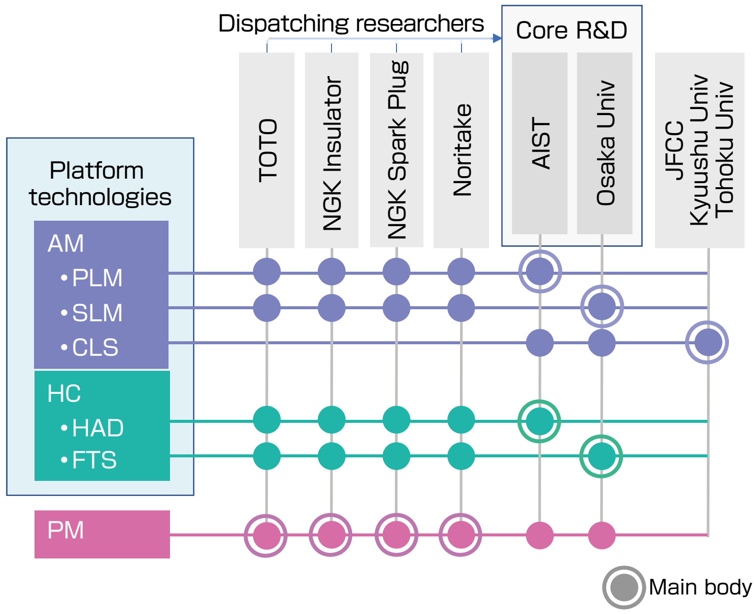


Fig. 2 R&D items and participating organizations of HCMT project
AM: Additive Manufacturing, PLM: Powder-Layer Manufacturing, SLM: Slurry-Layer Manufacturing, CLS: Ceramic Laser Sintering, HC: Hybrid Coating, HAD: Hybrid Aerosol Deposition, FTS: Fine-particle Thermal Spraying, PM: Product Manufacturing

PM. Examples are semiconductor production parts, plasma-resistant parts and ceramic cores of gas-turbine blades in the industrial field; water-purifying filters and portable toilets in areas related to everyday life; bone prostheses and ceramic heads for hip joints in the medical field (Fig. 1, bottom).

Figure 2 also shows the participating organizations of the HCMT project.[1] The core R&D sites are placed at the National Institute of Advanced Industrial Science and Technology (AIST) and Osaka University for intensive R&D using common research facilities and equipment. TOTO Ltd., NGK Insulators, Ltd., NGK Spark Plug Co., Ltd. and Noritake Co., Limited dispatch their researchers to core R&D sites for developing platform technologies in collaboration as well as product manufacturing for their own targets. These four companies are known as the "Morimura" Group which has established the foundation of modern ceramic industries of Japan since the beginning of the 20th century. In addition, Japan Fine Ceramics Center (JFCC), Kyushu University and Tohoku University are in charge of R&D on CLS.

3 R&D strategies for AM of ceramics

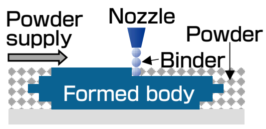
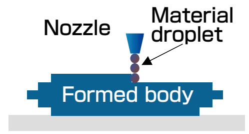
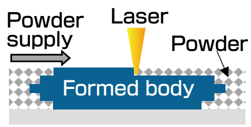
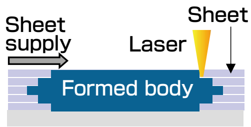
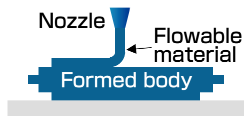
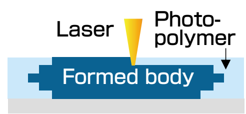
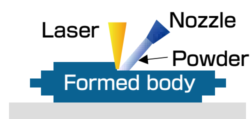
AM (additive manufacturing), also known as 3D printing, is a process by which a three-dimensional body is built through point, line or planar deposition of material typically using a print head, a nozzle, or another appropriate equipment. Objects are produced by not subtracting but adding material, based on computer-aided design (CAD) files or 3D model data, without using machining tools or forming dies and molds. The advantages include the following: (1) Realizing complex-shaped or integral-structured bodies which are never attainable by conventional molding approaches (this enables us to make totally new design of products enhancing their performance and durability), (2) Saving production time and cost due to a moldless process (this is particularly true for large variety-small amount production such as for new product prototypes and artificial bones and teeth), and (3) Saving raw materials since only a necessary amount is consumed while substantial amount of machining loss is generated in subtractive manufacturing, (4) Actualizing unique material structures including compositionally or functionally gradient layer textures. There are a variety of AM methods, which are classified into seven categories according to ASTM F2792-12a, "Standard Terminology for Additive Manufacturing Technologies." Table 1 shows this classification with illustrations.

AM has been well developed in the field of polymers and already has been widely used for fabricating 3D products of this sort of material to such an extent that household 3D printers for resin have been commercially available for some time.[5] Some key metal parts have also been successfully produced by AM;[6]-[8] for example, GE Aviation has introduced the additive-manufactured metal fuel nozzles in combustion systems of aircraft engines that could not be made conventionally.[8] The benefits include "25 % lighter weight than its predecessor part," "the number of parts of the nozzle reduced from 18 to 1," and "5 times higher durability due to more intricate cooling pathways and support ligaments."

Regarding AM of ceramics however, though some complex-shaped 3D bodies have been prepared with relatively high precision using vat photo-polymerization (stereolithography), *etc.*,[9]-[18] their product size has been generally limited, typically to a few centimeters or less, and the status is far from manufacturing technologies to be used in industries. Hence, comprehensive R&D efforts on manufacturing processes including powder preparation, lamination, and post-process suitable for ceramics are crucially required to grow AM of ceramics to the level of industrial application, and this has triggered the HCMT project.[1]-[3]

When applying AM methods, which have been used in the fields of polymers and metal, to ceramics, because of the difficulty in directly obtaining sintered bodies due to their intrinsic nature such as high refractoriness and less-sinterability, it is general to produce green or formed bodies instead, which are to be sintered in a conventional furnace afterwards. For example, in powder bed fusion, laser heat melts polymer binder which is mixed with ceramic powder to form green bodies. The HCMT project employs PLM and SLM for forming green bodies as stated above; the former is categorized into powder bed fusion (also called "indirect selective laser sintering") of the ASTM F2792-12a classification (Table 1), and the latter into vat photo-polymerization thereof. This is because these two approaches are known to be superior to the others in terms of homogeneous microstructure, good properties of

Table 1. Classification of additive manufacturing technologies, according to ASTM F2792-12a

Method	Outline	
Binder jetting (Aka, 3D printing)	Liquid binder through a nozzle is selectively deposited to join powder materials.	
Material jetting	Droplets of build material through a nozzle are selectively deposited.	
Powder bed fusion (Selective laser sintering, PLM)	Heat, typically of laser, selectively fuses area of a powder bed.	
Sheet lamination (Laminated object manufacturing)	Sheets of material are bonded and selectively cut by laser.	
Material extrusion (Fused deposition modeling)	Flowable material is selectively dispensed through a nozzle or orifice.	
Vat photo-polymerization (Stereolithography, SLM)	Liquid photopolymer in a vat is selectively cured by light, typically of laser.	
Directed energy deposition	Focused heat, typically of laser, selectively fuses materials as being deposited.	

the produced materials, wide shape flexibility, and high dimensional accuracy of the obtained products. In addition, material density (or porosity) can be adjusted over a wide range by combining these two approaches. Typical forming procedure of PLM is shown in Fig. 3. It consists of the following: (1) Mixing ceramic powder and polymer binder and putting them in the supply part, (2) Supplying the mixed powder to the forming stage and smoothing them using a squeegee (Fig. 3) or a roller to make a thin layer (typically ~0.1 mm), (3) Melting the binder of the desired part by scanning laser heat and bonding the ceramic powder, (4) Lowering the forming part by the formed layer thickness, (5) Repeating the above process of (2) to (4) for a 3D green body of a desired shape, and (6) Dewaxing and post sintering the obtained green body in a conventional furnace. PLM is a dry forming process itself and does not need a drying process of a wet body which often leads to undesired distortion and deformation of a green body. This, therefore, is advantageous particularly in making large-scaled products. On the other hand, the lack of fluidity of powder results in low density of green and sintered bodies, indicating that PLM is suitable for producing porous bodies. For example, a previous study on similar AM approaches for alumina showed that the green and sintered densities are around 30 % and 40 %, respectively.[19] It has been reported that the sintered density was substantially improved to 80 % or more when additional treatments of warm isostatic press and slurry infiltration on the green bodies were done; however, it is essentially important to increase the densities without such treatments in view of industrial application. For this purpose, in the HCMT project, we adjust and optimize the whole process from powder preparation (binder selection, powder mixture, powder fluidity evaluation, *etc.*) and lamination (powder supply, laser irradiation, *etc.*) to post-process (dewaxing, sintering, *etc.*), resulting in the sintered density of 84 %, without the additional treatments, in a simple-shaped alumina plate (50 x 50 x 5 mm) for specific mixed powder and experiment conditions.[20]

Figure 4 shows SLM's procedure, which includes the following: (1) Mixing fine ceramic powder and liquid photopolymer and putting them into the slurry supply, (2) Supplying the mixed slurry from the slurry supply on the substrate and smoothing them using a knife edge for forming a thin layer (typically several 10 μm), (3) Curing the photopolymer by laser light for the desired part, (4) Lowering the substrate by the formed layer thickness, followed by the same remaining processes as PLM (5, 6). SLM is a wet approach and therefore has characteristics totally opposite to PLM; due to the high fluidity, it is advantageous for producing dense parts of complex-shapes with high precision. It has been reported that careful selection of raw powder in SLM of alumina resulted in a high sintered density of 99 % with a bending strength of ~800 MPa.[14] On the other hand, undesired deformations and distortions often are generated during the drying process, which leads to unsuitability for making large-sized products. Therefore, the HCMT project is aimed at avoiding such deformations and distortions by optimizing the processing conditions and modifying the forming apparatus. It is also critically important to disperse the fine ceramic particles densely into the slurry, for example by using ceramic powder having bi-modal size distribution, with sufficient degassing for reducing sintering-shrinkage as well as obtaining dense bodies.

As stated above, it is extremely difficult to obtain sintered bodies by directly laser-sintering ceramic powder in AM. For example, Qian, *et al.*[21] investigated direct-laser-sintering of alumina, and revealed three detrimental phenomena in the sintered body including glassy parts due to overheating/rapid cooling, unsintered parts due to heat lack, and cracks due to thermal stress. If it is realized successfully, however, it will bring enormous benefits of savings in the post sintering process which needs substantial cost and time. The HCMT project, therefore, also deals with direct laser sintering of oxide and non-oxide ceramics (CLS); the approach includes full-packing ceramic powder in a layer, optimizing laser irradiation conditions for critical temperature control, *etc.*

4 Platform technologies in PLM

For optimizing AM procedures of ceramics, there are a number of technical items that should be carefully examined and properly selected. This chapter discusses what sorts of technical items there are, how they are connected and correlated to each other and what should be considered and

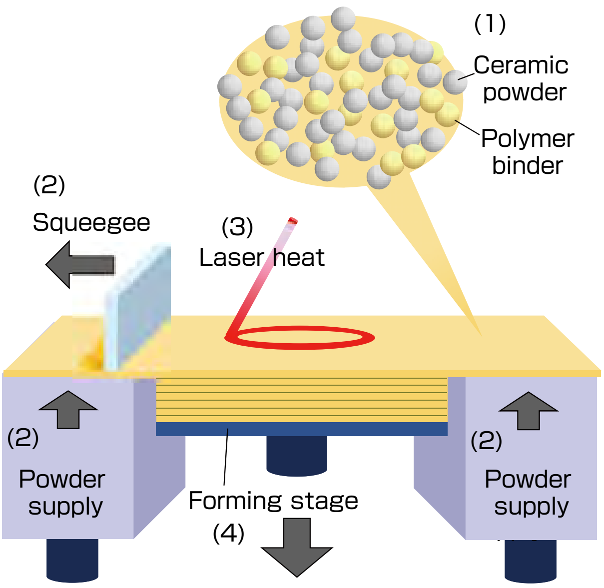


Fig. 3 Typical forming procedure of powder-layer manufacturing (PLM). The number in parenthesis corresponds to that of the description in the text.

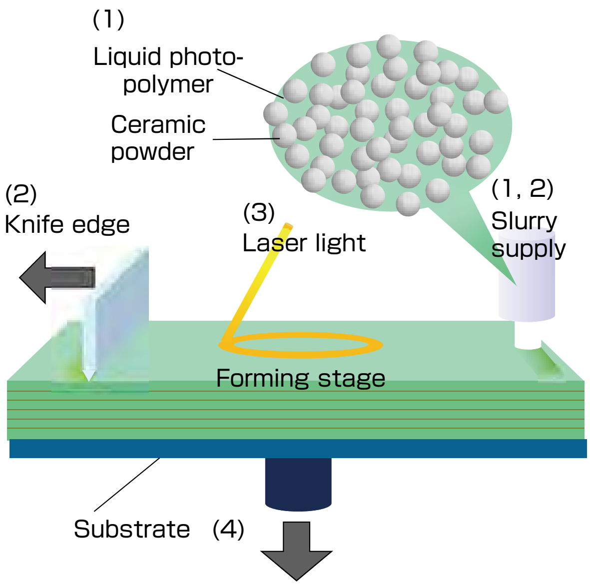


Fig. 4 Typical forming procedure of slurry-layer manufacturing (SLM). The number in parenthesis corresponds to that of the description in the text.

selected in each of the technical items in order to obtain sound products through an AM approach, taking an example of PLM.[22] The total procedure of PLM can be roughly divided into three processes, including powder preparation, lamination (or PLM itself), and post process. Figure 5 shows technical processing items as well as evaluation items in each of these three processes.

In the powder preparation, ceramic powder is mixed with polymer binder to be melted by laser heat as described above by some appropriate method. The mixed powder used in PLM should have sufficient flowability, which is generally obtainable with a spherical shape and size of ~50 μm. Therefore, when the powder size is below 10 μm which is typical for so called fine ceramics, granulation process including spray-drying is frequently used to produce spherical granules of such a size. The mixed powder is evaluated in terms of flowability including angle of repose, compression ratio, Hausner ratio as well as their size and shape, *etc.* Angle of repose is the angle measured in degrees between the horizontal plane and the steepest slope at which loose powder remains in place without sliding. Compression and Hausner ratios are given by $(D_t\text{-}D_b)/D_t$ and D_t/D_b, respectively, where D_t is the tap density and D_b is the static bulk density. When the flowability increases, these three indices all decrease. Suitability of powder to lamination of PLM is also examined in a simple preliminary powder test, where a lump of powder placed in the forming stage is leveled by using a squeegee. If the powder is flowable enough and not so cohesive, the surface becomes smooth without dimples and cracks, which appear with powder cohesive and less flowable, as shown in Fig. 6 (a) and (b),

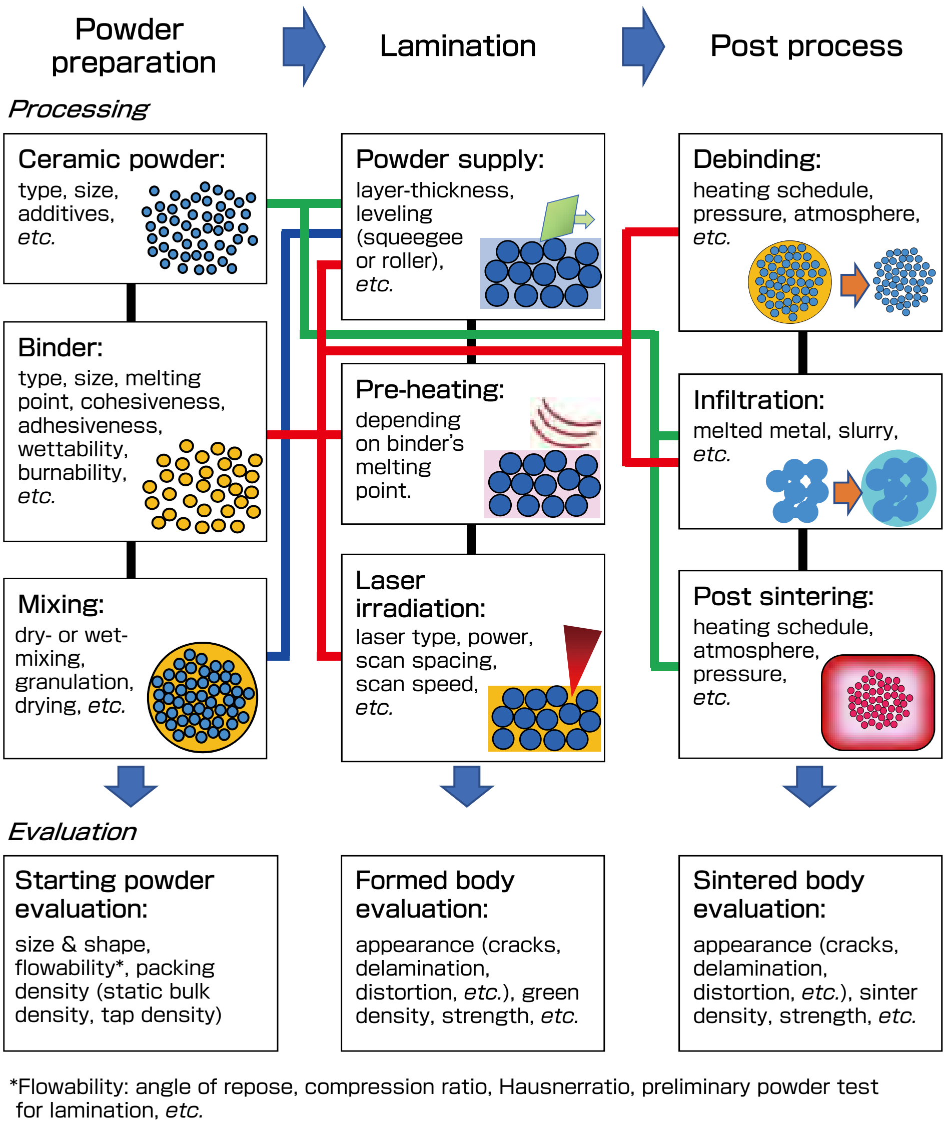


Fig. 5 Technical processing items and evaluation items in starting powder preparation, lamination, and post-process

respectively. The powder flowability is also important for making a sufficiently filled powder layer. Because the layer is formed only by smoothing the powder by a squeegee, *etc.* without pressure, the powder density of the layer is almost equivalent to the static bulk density of the powder. It should be noted that since the mixed powder contains polymer binder as well, the density of ceramic powder itself is further lowered. It is, therefore, essentially required to have a well filled powder layer to get high densities of the resulting green and sintered bodies, which also leads to reduction of undesired deformation/distortion. It has been known that proper combination of coarse and fine powders leads to closer packing; however, powder flowability is generally degraded when containing fine powder.

In the lamination, the first step is formation of the powder layer, whose thickness is to be determined from the mixed powder size; it is preferably 1.5 to 5 times larger than the powder's maximum size. The thicker the layer is, the higher the production rate; however, it results in larger steps of side surfaces. The thickness also should be determined so that laser-heat is sufficiently transferred to the bottom of the layer. Insufficient heat transfer causes a large temperature distribution thickness-wise, frequently resulting in warping and inter-layer delamination of a green body. Next, pre-heating mixed-powder is made before lamination, depending on the binder's melting point. For example, when employing wax-based binder whose melting point is 80–120 ℃, temperature difference is usually small thickness-wise and successful lamination is easily attainable without pre-heating. On the other hand, for nylon-based binder with melting point of 150–200 ℃, higher laser power or slower laser scan is usually required to melt it, which leads to a large temperature difference thickness-wise and frequent appearance of warping as described above. An example of warping which was observed in a green body with nylon-based binder is shown in Fig. 7 (a). Pre-heating the mixed powder closely to the melting point is effective for avoiding such warping. Laser irradiation conditions including laser type, power, scan spacing, and scan speed should be carefully chosen depending on the types of the polymer binder and ceramic powder, *etc.* An issue often occurring during the lamination is sliding of a green body embedded in powder and formation of a gap, as shown in Fig. 7 (b). This sliding is

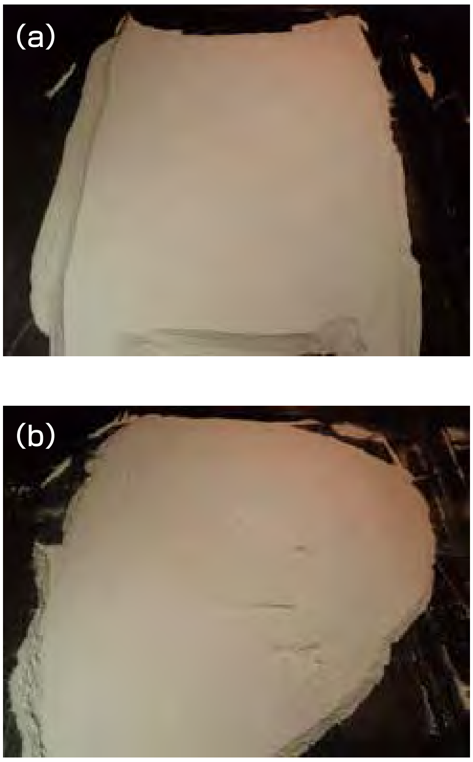


Fig. 6 Preliminary powder test for lamination, (a) Smooth surface with flowable powder, (b) Dimples and cracks with less flowable powder

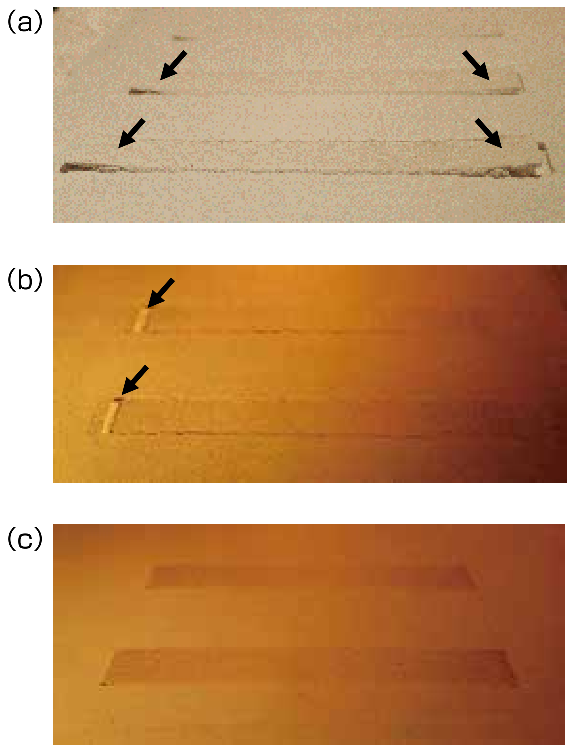


Fig. 7 (a) Warping observed in a green body with nylon-based binder, (b) Sliding of a green body and formation of a gap, (c) Successful formation of a green body embedded in powder. The body is rectangular-shaped with 7 mm width and 50 mm length.

made when the body is trailed by powder being re-coated on it and is more likely to occur typically in the following cases: (1) the binder is still heated and adhesive, (2) the squeegee moves too fast, or (3) the mixed powder is too flowable. Thus, it can be resolved by cooling the binder sufficiently, lowering the squeegee movement speed, or lowering the powder flowability. Figure 7 (c) shows an example of successful formation of a green bogy without the above issues (embedded in powder).

In the post process, first the binder is removed from the obtained green body by burning it out. The heating schedule, pressure, atmosphere, *etc.* for this process should be carefully selected so that undesired deformation and distortion would be minimized while the binder is melted and burnt out. Infiltration is often employed for densification. A typical example is siliconized silicon carbides (SiSiC), where melted Si is infiltrated through porous SiC-C green bodies produced by PLM, followed by reaction between Si and C for formation of secondary SiC and densification.[23] Free carbon produced during burning the polymer binder can be used for this reaction. Slurry infiltration into green bodies also can increase green and sinter densities as already stated.[19] In post-sintering, selection of conditions including heating schedule, pressure, atmosphere, *etc.* is crucial for obtaining sound sintered bodies, and knowledge and experiences so far on sintering of conventional green bodies are of great use for it. The green and sintered bodies are evaluated in terms of appearance (cracks, delamination, distortion, *etc.*), green/sinter densities, strength, and others.

As seen so far, many of the technical processing items are closely connected and correlated to each other; such close relations are expressed by solid lines in Fig. 5. It can be said that particularly powder preparation substantially affects many of the subsequent processes of lamination and post process. For example, the properties of the binder are critically important for the powder supply, pre-heating and laser irradiation of the lamination (melting point, cohesiveness, adhesiveness, wettability, *etc.*), while they are also crucial to the debinding and infiltration (where burnt binder is often used for reaction with infiltrated ones) of the post process (melting point, burnability, *etc.*). Thus, powder preparation is the most essential process in PLM, similarly to the cases of conventional ceramic processing. Sound products can be obtained only after all the technical items are properly selected and performed. It should be noted that the approach for examining and integrating the technical items into optimal AM technology described in this chapter is employed similarly in SLM and CLS.

Table 2. Bulk density, Young’s modulus, specific stiffness (Young’s modulus/bulk density) and flexural strength of PLM-produced SiSiC and conventional one (molding approach).

	PLM	Conventional
Bulk density	3.0	3.0
Young’s modulus (GPa)	340	340
Specific stiffness	113	113
Flexural strength (MPa)	290	320

5 Prototype models produced by AM

Taking advantage of the developed AM technologies, the HCMT project has manufactured several types of unique prototypes aimed at various target applications, some of which are described in this chapter. The first are stage models produced by PLM, which are anticipated as basic structures for ceramic exposure stages used in future semiconductor industries; some examples are shown in Fig. 8, in comparison with a conventional structure.[1][24]-[26] A light and stiff exposure stage of large scale and complex shape is critically needed for next generation IC chip production where more accurate positioning and higher throughput will be strongly required. While the conventional rib structure produced by molding consists of simple walls (a), AM can make that having windows in the walls (b), and furthermore truss structures of light weight/high stiffness (c-e), which were not obtainable until now. The models of (b-e) are siliconized silicon carbides (SiSiC) which are obtained by Si infiltration into SiC-C green bodies followed by reaction-sintering, as described above. Their feature is high specific stiffness (Young’s modulus/bulk density) and very little sintering-shrinkage, both of which are advantageous for application to large-sized exposure stage products. In order to fully recognize AM as industrial manufacturing technologies, it is crucially important for products produced by AM to have properties equivalent to those of conventional ones. Table 2 compares bulk density, Young’s modulus, specific stiffness, and flexural strength of PLM-produced SiSiC with those of the conventionally manufactured high-rigidity SiSiC.[26] It should be noted that specific stiffness, which is the most important property for stage application, equals to that of the conventional ones. R&D should proceed for improving flexural strength to 320 MPa or higher.

One of the advantages of AM is its capability of producing complex-shaped parts directly from computer aided design files or data. Using computer simulation based on structural topology optimization techniques, it is now possible to optimize rib structures of stage models. Figure 9 shows an optimal rib structure obtained thereby in comparison with a conventional one, and a SiSiC exposure stage model (green body) produced by PLM based on that structure.[22] The weight is reduced to half or less while maintaining the same vertical stiffness (the simulation neglects horizontal stiffness). The thinnest part of the model is approximately 3 mm in thickness.

Because of their chemical resistance, durability, and other properties, ceramic filters currently are used for various applications, one of which is in a water-purification device. The water paths of the filters are unidirectional straight channels, simply because they are produced by extrusion molding. Employing the AM techniques, however, enables us to make the channels more complicated, such as, for example, spiral channels as shown in Fig. 10 (a).[1][26] It will bring several potential benefits including increase of contact area between water flow and the channel, local control of water flow (flow velocity, laminar flow vs. turbulence, *etc.*) and others, which may lead to improving the performance and miniaturizing the device. Figure 10 (b) shows an alumina filter model produced by PLM, containing spiral channels of 3 mm diameter, which can be identified from the traces of the cut model.[1][22] Similar filter models have been also manufactured by SLM, and the joining technologies for making a long-sized filter base are also under development.[26]

Besides the above-stated ones, the HCMT project has manufactured several other types of prototype models, including artificial alumina knee joints whose internal surface has salient parts, which were difficult to make conventionally, to improve fixation into bones,[1][27] bone prosthesis with uniform pore size and no closed pores which leads to sufficient infiltration of bone cells and dense bones,[1][27] and ceramic cores for cooling systems of gas-turbine blades with remarkably shortened production time and wide flexibility of the structure design.[1][28]

Notable progresses have been made in direct laser sintering of ceramics as well.[1][29][30] A thin formed layer of alumina with high green density of 83 % has been successfully obtained by dewaxing and drying slurry layer containing

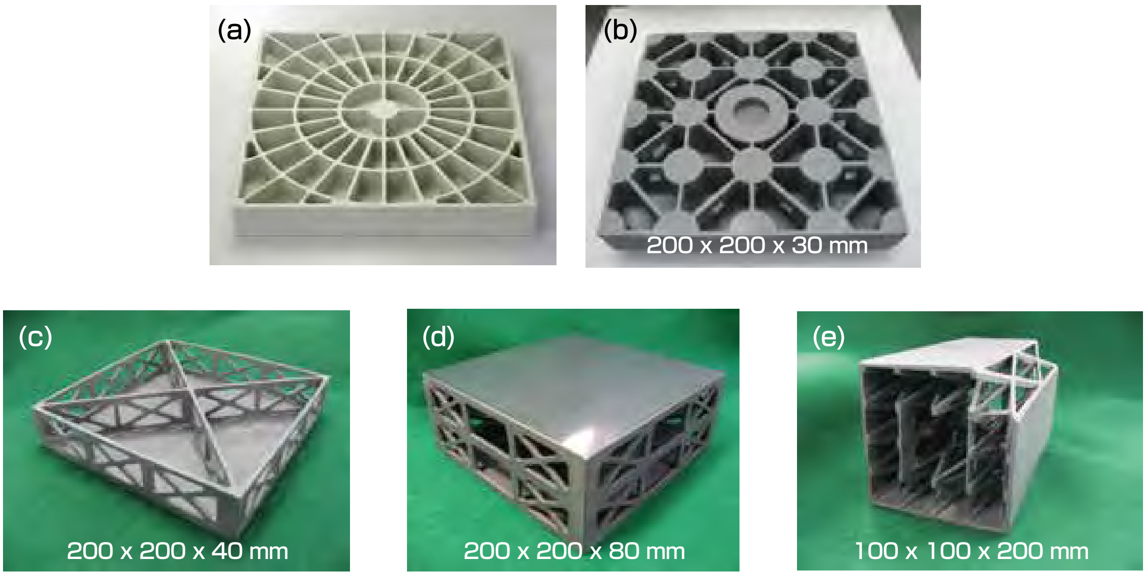


Fig. 8 SiSiC exposure stage models for IC chip production
(a) Conventional rib structure produced by molding approach, (b) PLM-produced rib structure having windows in the walls, and (c), (d) & (e) PLM-Produced truss structure of light weight/high stiffness

optimal mixture of different sorts of alumina powder. Laser-irradiating this highly-packed green alumina layer has led to full densification without such glassy parts, unsintered ones, and cracks as reported previously.[21] The laser absorption coefficient was adjusted by using a proper amount of material having a different coefficient from alumina.

6 Summary and future perspectives

This article describes the R&D strategies and the current achievements on AM of ceramics in the HCMT project, which has been initiated since 2014. The two AM technologies, PLM and SLM, which are advantageous in terms of dimensional accuracy, shape-flexibility, density-controllability, *etc*., are being developed for producing ceramic green bodies. A variety of 3D prototype models for varied target products have been manufactured so far by using the developed AM technologies. Furthermore, intensive research efforts are being devoted to ceramic laser sintering. In order to ensure the developed technologies, 7 and 26 patents have been applied for on PLM and on the HCMT project as a whole, respectively, as of March, 2018.

While AM of ceramics has numerous advantages as already stated, there are also several issues which should be taken into consideration when using it for industrial applications: (1) Profitable almost only for large variety-small amount products, (2) Facilities sometimes can be very expensive, requiring substantial initial costs, (3) Difficult to apply to ceramics, for which melting and solidification are not available in principle, (4) Currently usable only for producing green bodies, which are to be post-sintered in a conventional furnace, (5) Whether AM products give us the same properties as those of conventional ones is not certain, (6) Some restrictions apply to powder used, *e.g.*, the grains should be free flowing, requiring a preferably spherical shape and size of ~50 μm when supplying them to the forming stage in some methods including binder jetting and powder bed fusion. In order to overcome these issues and establish AM as manufacturing technologies in ceramic industry, further R&D efforts are critically required in the future.

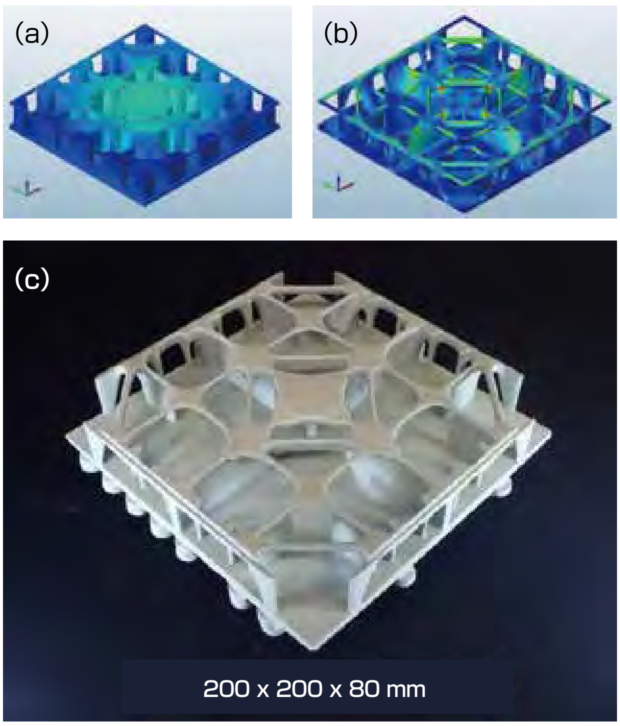


Fig. 9 (a) Conventional rib structure, (b) Optimal rib structure obtained by computer simulation based on structural topology analysis, and (c) an SiSiC exposure stage model (green body) produced by PLM based on the structure (b)

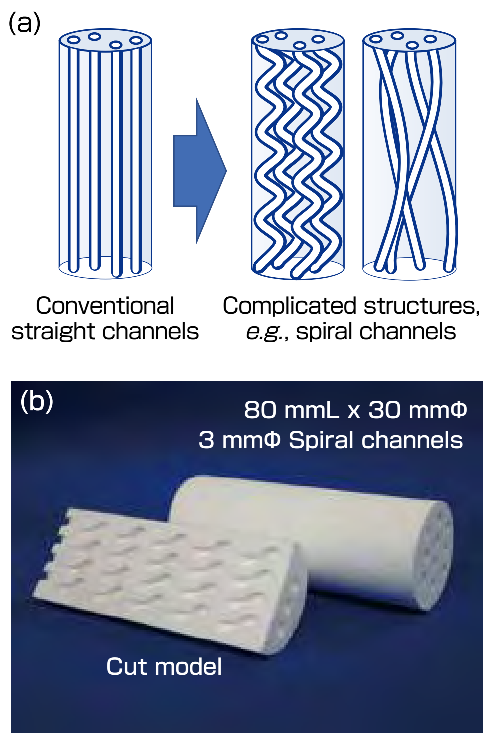


Fig.10 (a) Conventional water-purification ceramic filter has unidirectional straight channels, while the AM techniques enable more complicated channels, *e.g.*, spiral ones. (b) an alumina filter model produced by PLM, containing spiral channels of 3 mm diameter whose traces are observed in the cut model

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Author

Tatsuki Ohji

Joined AIST in 1983, and currently is Fellow thereof since 2017. BS and MS in mechanical engineering from Nagoya Institute of Technology and Ph. D. in inorganic materials engineering from Tokyo Institute of Technology. Has authored more than 350 peer-reviewed papers and 20 book chapters, and has edited more than 40 books and conference volumes. Research interests include mechanical/physical properties, microstructural control, and novel processing of ceramics and related materials. Fellow of American Association for the Advancement of Science (AAAS), American Ceramic Society and ASM International, and Academician of World Academy of Ceramics. Currently serves as Governor of Acta Materialia Inc., Vice President of the Ceramic Society of Japan, and Editor in Chief of *Ceramic International* and *Journal of the American Ceramic Society.*

Discussions with Reviewers

1 Overall

Comment (Toshimi Shimizu, AIST)

This “report” describes research centered on additive manufacturing (AM), of which the author serves as the leader, of the R&D project on high-value added ceramic products manufacturing technologies (HCMT) of the Cross-ministerial Strategic Innovation Promotion Program (SIP). It is a focused presentation of the technological platform of powder-layer manufacturing (PLM), an AM technology for making ceramic products that have uneven thickness or complex shapes that are difficult to process, and of prototypes made from the technology. It is a very effective presentation in understanding the processes that form PLM and the interconnection of the related technological elements. As a report that presents an example of development of technology that has practical value that is expected to lead to innovation, it is suitable for *Synthesiology.*

Comment (Akira Kageyama, AIST)

Of ceramic technology and industry in which Japan has excelled, this report summarizes the process by which the HCMT project was started as a national strategical project, and how it succeeded, against the decline of Japan’s world market share along with the improvement of technological level of developing countries. It presents the selection of elemental technologies and integration of each technology, focusing on AM as technology that forms ceramic products of complex shapes that were difficult to make with the conventional technology. It is also worth reading as a report on management of R&D in which collaboration and role assignment was necessary among the participating universities, AIST, and companies. Of the necessary technologies covering a wide range, PLM is selected, and it describes in detail, within the allowed range of disclosure, each of the elemental technologies such as the optimization of raw material powder, which is bound by confidentiality. R&D discussion is presented in an easy to understand manner, and it is well worth being printed in this journal.

2 Content, structure, research scenario (draft report)

Comment (Toshimi Shimizu)

The first draft that was submitted as a research paper and the second draft that was submitted as a report did not satisfy the qualities that are required of *Synthesiology.* For a writing to be accepted, it needs to cover seven topics: 1. research objective, 2. research target and relationship to society, 3. scenario, 4. selection of elements, 5. relationship among elements and their integration, 6. evaluation of results and future perspectives, 7. originality. The first and second draft did not cover these. Specifically, 1) for 3. scenario, the scenario of hypothesis was not rationally described; 2) for 5. relationship among elements and their integration, the relationship among elements and integration were not rationally described using scientific words; 3) for 7. originality, much information that was already publicly known was presented and it lacked originality. Therefore, its content as it was unfortunately did not apply to a research paper or a report of *Synthesiology*, which need to focus on the research scenario and specific research processes. The draft covered the whole R&D of AM, and lacked depth in its description of the scenario and hypothesis, and the relationship and integration among elements.

The reviewer suggests focusing on PLM technology actively being promoted at AIST as a revision of the content of the report. How about using a diagram which would help the reader to understand and describing in detail each elemental technology related to PLM, how they are corelated, and the process by which they are integrated and optimized?

Answer (Tatsuki Ohji)

According to your advice, I revised the text, and in Chapter 4 “Platform technologies in PLM,” I have categorized and described in detail each elemental technology of the three processes of powder preparation, lamination, and post process, have presented the relation to other elemental technologies using a diagram (Fig. 5). Especially with strongly related elemental technologies, they were emphasized in the diagram, and the process by which they were integrated and optimized was described. Evaluation items for each process are also considered. Furthermore, in Chapter 5 “Prototype models produced by AM,” the subject was narrowed down to a ceramic stage model and a ceramic filter model attained by PLM, and other prototype models were simply described along with references.

Comment (Akira Kageyama)

The first draft was submitted as a research paper, and after a review, additions were made, and the manuscript was resubmitted as a report. In the second draft, the significance of the technologies and their positions are easier to understand as more detailed descriptions were added of the R&D that was bound by confidentiality. However, as a report, it needs to strongly state how technology is introduced to society and how it is to be useful, but this point is not expressed clearly. I suggest a revision on this line. If you rearrange and reorganize the structure, the way of expression, and figures and tables, the point of the report will become clear. For example, a report according to the editorial policy needs to present 1) the aim, 2) the process of development (the course to the goal), and 3) the outcome. Keeping this in mind, the report can be arranged to include 1) the global state of ceramic industry and the situation that Japan faces (situation analysis), 2) the issues that need to be solved in order to overcome the situation, and why the issues were chosen (problem analysis), 3) what was decided technically and in management in order to clear or solve the issues (decision analysis), and 4) the results of R&D and management executed in this project with the above-mentioned 1) to 3) in mind (output and/or outcome). The manuscript already includes these aspects, so why not revise the whole structure and try to make it simpler and clearer?

The manuscript is also rather long. In the second draft, as, taking PLM as an example, the relationship and integration of elemental technologies are described, couldn’t the section concerning other R&D related to AM be reduced or deleted by stating that “similar integration of elemental technologies is attempted as PLM”? It is also useful to use references. I also suggest narrowing down examples presented in the Chapter 5 “Prototype models produced by AM.”

Answer (Tatsuki Ohji)

As you pointed out, I have revised the manuscript as follows: The global state of ceramic industry and the situation of Japan, and the significance of the HCMT project in order to break through the situation was described in Chapter 1 “Introduction”; R&D under this project and the managerial system was written in Chapter 2 “Overview of HCMT project”; the technical strategy of AM was summarized in Chapter 3 “R&D strategies for AM of ceramics” and Chapter 4 “Platform technologies in PLM”; and the outcomes up to now were presented in Chapter 5 “Prototype models produced by AM.”

As I have responded to the previous comment, I have focused on the prototype model of PLM in Chapter 5 “Prototype models produced by AM,” and have greatly reduced the descriptions of technologies other than PLM in Chapter 3 “R&D strategies for AM of ceramics,” and have added a sentence that you suggested to the end of Chapter 4 “Platform technologies in PLM.”

3 Technical terms and drawings (figures)

Comment (Toshimi Shimizu)

The readers of *Synthesiology* are not specialists of ceramic technology, but general engineers, researchers, and readers. For the engineers not related to the ceramic field, it would be difficult to understand the various and numerous technical terms that are used throughout this report. Since the report is written assuming that the technical terms are understood, understanding of the content by the general reader would not be deepened with such difficulty. Lack of unity of terms can also be seen.

The shortest course to explain technical terms is to use drawings. Extreme examples are the processes of shaping the raw material, sintering, and the post process, which are nothing special for engineers of the field, need to be explained from the basics by using drawings to the general reader and engineers from other fields. Regarding classification of AM, and technological explanation of PLM and slurry layer manufacturing (SLM), how about organizing technologies with overviews and characteristics by using drawings?

Answer (Tatsuki Ohji)

As you pointed out, I have used drawings in Table 1 “Classification of AM,” and diagrams (Figs. 3, 4) of PLM and SLM of the revised (final) draft. In the diagrams on PLM and SLM, I have used numbers that correspond to explanations in the text. In the related diagram of elemental technologies (Fig. 5), I have also used simple illustrations. Concerning technical terms, they have been unified to the most general terms that are used in the materials field.

4 Intellectual properties (draft)

Comment (Akira Kageyama)

I imagine that there are many intellectual properties generated through this project. Couldn’t you write about this in the last chapter, Chapter 6” Summary and future perspectives”? For example, it can be a phrase like “so and so number of patents on PLM, and so and so number of patents on the HCMT project as a whole have been filed.” Intellectual property is one of the important outcomes of R&D, and is a yardstick to measure international competitiveness of technology.

Answer (Tatsuki Ohji)

As you indicated, I have added the following sentence to Chapter 6 “Summary and future perspectives”: “In order to ensure the developed technologies, 7 and 26 patents have been applied for on PLM and on the HCMT project as a whole, respectively, as of March, 2018.”