

# Development of high power and high capacity lithium secondary battery based on the advanced nanotechnology

## — The convergence innovation strategy employing the inter-disciplinary research and inter-organization straightforward technology transfer —

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The development of high power and high capacity lithium secondary battery has been executed in a new strategic innovation scheme. This paper reports the significance of adopting the idea of interdisciplinary research to straightforward technology transfer from university, through AIST and battery company down to automobile company. A joint project of these organizations covering the upper stream science (at a university) and the down stream battery products (at a company) has been found a good scheme for rapid R&D when the university or AIST has innovative seeds. AIST has superiority in producing new technological seeds because of the interdisciplinary research between energy and nanotechnology as well as a wide spectrum of research fields covered by the institute. In this paper, an application of nanocrystalline electrode materials to high power battery is reported where the technology was successfully transferred to the commercial development of superior battery products.

**Keywords :** Lithium secondary battery, nanocrystal, high capacity and high power electrode, plug-in hybrid car, hybrid electric vehicle (HEV)

### 1 Background of research

Japan and other advanced nations are faced with two major energy crises today. One is structural energy limitation due to rapid increase of energy demand from BRIC countries and fierce international competition over procurement of resource for third world economic growth. Second is decrease in food production due to dramatic changes in atmospheric environment caused by global warming, as well as increased risk to sustainable biosphere due to reduction of space suitable for human survival, caused by increased abnormal climate. Japan, which is a resource-poor country, suffers from uncertainty factor of energy supply that is the foundation of nation's economic development, and innovations in energy technology is the most important issue for ensuring national energy security.

High-power lithium secondary battery is highly effective against global warming, and it is expected to accelerate the market entry of plug-in hybrid electric vehicles (HEV) and electric vehicles (EV) that may raise industrial competitiveness. Fierce competition is taking place throughout the world for its development. The desired high output performance of battery cannot be achieved with conventional material technology, and innovations in battery technology have been awaited. With the background of recent advances in nanoscience and nanotechnology, there is great interest in development of high-power high-capacity electrode material based on fine nanostructure control of electrochemical active materials.

The Author became interested in innovative active materials based on state-of-art nanotechnology, and worked on the development of high-speed charge-discharge electrode. Since

charge transfer rate increases dramatically in nanocrystal where the diffusion length of ions becomes extremely short, the possibility of designing high-speed charge-discharge battery has been theorized. The Author's research team applied advanced nanotechnology process and fabricated electrodes with nanocrystal and nanoporous structures that could not be made with conventional material science, and showed that high output property that far surpassed conventional performance could be achieved under laboratory conditions. Therefore, we decided to investigate whether these new material design concepts could be used as power source of next-generation HEV in industry-academia-government collaboration project, with involvement of battery and automobile manufacturers. In this paper, the efficacy of nanotechnology for innovations in the field of energy storage will be discussed, and the efficacy of vertical collaboration project of industry-academia-government for swift commercialization of innovation will be addressed, as well as introducing the actual results of the technological development.

### 2 Development of innovative battery technology through fusion of nanotechnology and energy technology

Figure 1 is the Ragone plot of performances of lithium secondary batteries that have been realized dramatically in recent years, and examples of innovative energy devices and products. Currently they are put to practical use in cell phones and laptop computers. These devices use small low-capacity lithium battery and do not require high output density. Recently, middle-sizing and higher power for lithium battery became possible, and the batteries are used as power source for electric bicycles and power tools. The frontline

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of lithium battery technology is moving from small to large devices, and higher power, higher capacity, and lower cost are important issues assuming application as power source for automobiles. As shown in Fig. 1, the frontline of battery technology application is shifting from lower right to upper left. The middle- and large-size, highly reliable lithium battery is expected to become central energy technology for robots, HEVs, and recyclable energy in the future. To accomplish this, it is necessary to develop high-power cell of several kW/kg level as mentioned above, and innovative technological development using nanotechnology is drawing great expectation and investments.

In fact, assuming use as auxiliary power for clean vehicles represented by hybrid and fuel cell cars that are key technologies to counter global warming, power source with both sufficient energy and output densities is necessary. Figure 2 is a Ragone plot of target performance values of auxiliary power source for HEV of Japanese automobile manufacturers and U.S. Department of Energy (DOE). When targeting energy regeneration for general-use automobile, about 30 Wh/kg energy density and 3 kW/kg output density are required for battery cell, and such performance is intermediate level for lithium secondary battery and electric double-layer capacitor (EDLC). This performance (30 Wh/kg, 3 kW/kg) is equivalent to charge-discharge rate of 100 times per hour (completely charged in only 36 sec), and it is impossible to construct a storage device with such energy and output densities using bulk size intercalation electrode materials currently available. If ion intercalation mechanism was used for secondary battery active material, charge-transfer rate must be improved 100 times. That is, if storage mechanism that is midway between secondary battery and EDLC is used to obtain both energy and output densities, dramatic acceleration of ion dispersal within electrode and electronic conductivity must be achieved simultaneously.

This paper is a report of the result of the industry-academia-government vertical collaboration project where the goal was development of nanoporous electrode composed of nanospace

and nanocrystal active material, to create power source that enables high-speed input/output of electric energy, and to create high-power battery that can charge and discharge 100 times faster compared to ordinary lithium secondary battery. Particularly, I shall explain the original concept of the National Institute of Advanced Industrial Science and Technology (AIST) that conducted application of nanocrystal electrode material to high-power battery, and describe how nanotechnology can greatly contribute to the innovation of electric power storage. In the vertical collaboration R&D, automobile manufacturer who was end user participated from the planning stage of the project. For electrode material development that enabled high-output power source performance (strategic goal value in Fig. 2 about 3 kW/kg) required for plug-in HEV, we had the automaker indicate the direction of basic research at university and AIST, to maintain sufficient market competitiveness in terms of safety and low cost. To demonstrate the efficacy of vertical collaboration R&D that lead to product realization in shortest distance without major changes in specification due to result of core technology development, we conducted industry-academia-government collaboration product by four organizations with support from New Energy and Industrial Technology Development Organization (NEDO).

First, I shall explain the physicochemical basis of how high-power battery that is an innovation of storage technology using nanotechnology can be realized. The storage mechanism originates from electrochemical reaction accompanied by charge transfer by ion diffusion and electronic conduction. Here, diffusion length  $L$  in certain time can be expressed by the following equation, when diffusion coefficient of lithium ion in solid is  $D_{Li}$  and diffusion time is  $\tau$ .

$$L = (D_{Li} \tau)^{1/2} \tag{1}$$

If the diffusion coefficient of lithium ion in active material is estimated to be about  $10^{-13} \text{ cm}^2/\text{s}$ , time  $\tau$  required for ion

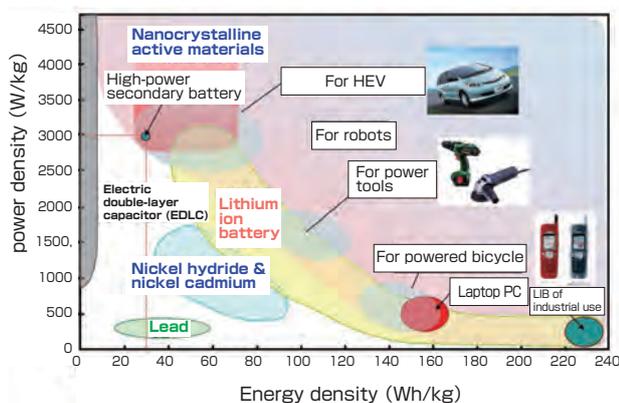


Fig. 1 Industrial products realized by innovations in battery technology.

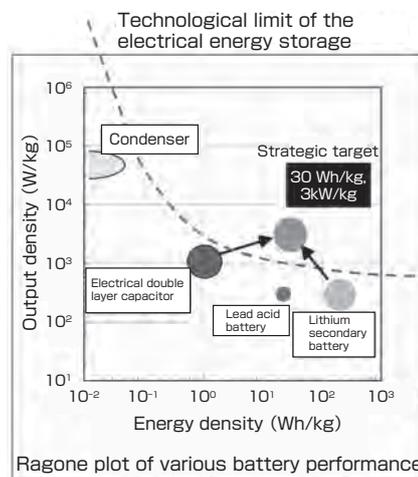


Fig. 2 Frontier in battery performance.

to diffuse inside the active material of width 5 nm is 1 sec at most. When the diffusion coefficient of ion in electrolyte within the pore is estimated to be about  $10^{-6}$  cm<sup>2</sup>/s, the time required for diffusion inside pore with length 10 μm can be estimated as 1 sec at most. Therefore, as shown in Fig. 3, if fast ion diffusion ( $k_1$ ) in electrolyte within nanopore and ion diffusion rate ( $k_3$ ) in solid at nanometer level within active material are used, charge and discharge can be realized in second order even if the particle size of nanoporous structure electrode is larger than micrometer. However, this is theoretical consideration under condition that electronic conduction ( $k_4$ ) of active material framework and surface reaction of active material ( $k_2$ ) are sufficiently fast in the rate determining process shown in Fig. 3. On the other hand, since specific surface area is large in nanoporous electrode, there is interest in energy property unique to nano material such as lithium storage caused by electrochemical reaction on the surface. If the surface is utilized, there may be possibility of storing lithium above stoichiometric composition.

Various nano-size electrode active materials were made based on migration kinetics at AIST to check the high capacity electrode property of nanoporous crystal material, and high-speed charge-discharge property was evaluated. In principle, if nanoporous electrode could be fabricated, charge-discharge in 36 sec, which was required as power source of HEV, would be possible. To accomplish this, it was necessary to integrate the elemental technologies of the frontier of nanotechnology fields such as solution process, molecular template synthesis technology, self-assembling process, and mass synthesis process for nanocrystals, and then to check the efficacy of innovative electrode active materials at battery cell level jointly with manufacturers. The competition for development is becoming fierce around the world since such high-power battery is long awaited in the industry and the market scale is great.

### 3 Execution and result of R&D

I shall describe the outline of development of high-power lithium secondary battery through interdisciplinary fusion

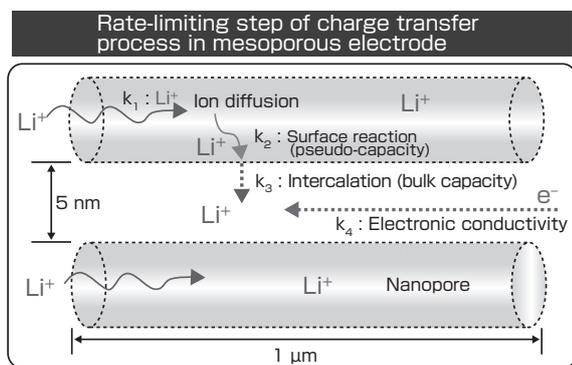


Fig. 3 Charge transfer process and rate-controlling step within electrode.

and industry-academia-government vertical collaboration conducted as NEDO project, and discuss whether the strategy for “shortening distance” to innovation was effective. This R&D was a vertical collaboration project by four organizations, Nagasaki University, AIST, Hitachi Maxell, Ltd., and Fuji Heavy Industries Ltd., as “Research and Development of High Capacity Secondary Battery by Low Resistance, High Ion Diffusion Nanoporous electrode” under the R&D for Practical Utilization of Nanotechnology and Advanced Materials conducted in FY 2005~2007. This project was conducted to develop high-power lithium secondary battery for HEV using advanced nanotechnology, and it was original because it used nanotech in energy technology effectively. It was characteristic that the end user automobile manufacturer was included in the project from commencement, and it was also distinct that the use of electrode technology of university and AIST as power source of HEV in short period was placed as central topic of R&D.

Nagasaki University, which was located upstream, investigated the inorganic chemical synthesis process of nanoporous electrode material from which high output property could be expected, from standpoint of basic chemistry, and developed new synthesis method that could be applied to practical electrode. Mesoporous material using molecular template such as surfactants could be applied to amorphous structure such as silica but were difficult to apply to crystalline active material such as LiCoO<sub>2</sub> and LiFePO<sub>4</sub> that were positive electrode materials for lithium secondary battery. Therefore, a process to fabricate inverse-opal form electrode framework using template structure with colloid polystyrene (PS) was developed, as shown in Fig. 4.

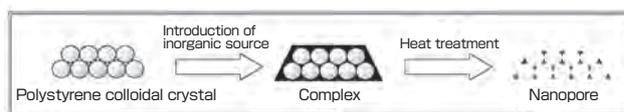


Fig. 4 Potential of university: nanoporous electrode synthesis process.

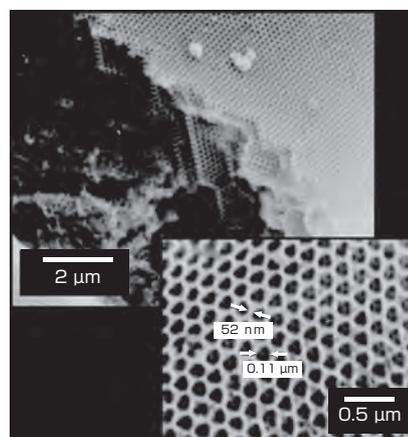


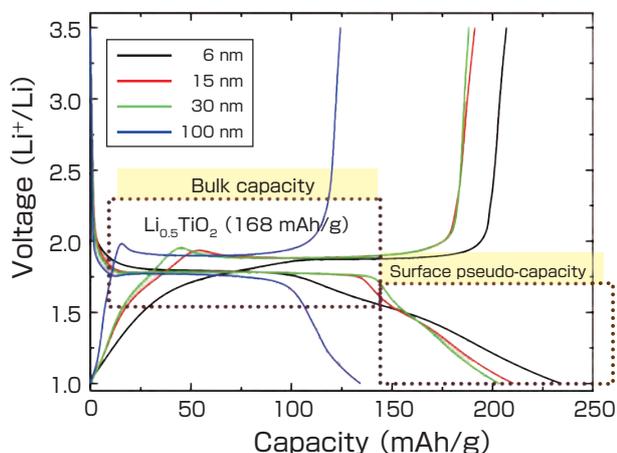
Fig. 5 Titania nanoporous electrode.

The example of synthesis of titania nanoporous electrode is shown as specific example. Ethanol solution of titanium alkoxide was added to PS colloid crystal, and the solution was mixed with titania nanoporous body consisting of anatase crystal by firing at 450 °C, and followed by niobium alkoxide, to obtain  $Ti_xNb_{1-x}O_2$  nanoporous electrode (Fig. 5). Because these electrodes had sequential structure of three-dimensional framework and had regular arrangement of nanopores and active materials, it was possible to obtain high ion conductivity, electronic conductivity, and lithium storage capacity.

Next, AIST conducted synthesis and structural evaluation of high-power battery electrode material using these nanocrystals followed by electrochemical property evaluation, and investigated the physical chemical mechanism of high-speed charge transfer property for how such innovative energy property could be obtained from this nanomaterial technology<sup>[1]</sup>.

In actual material development, the electrochemically active nanocrystal titania was set as model material for high-power electrode, and anatase and rutile structures were prepared in size ranges 6 nm ~ 100 nm, to conduct systematic evaluation of size dependency of lithium battery electrode property<sup>[2][3]</sup>.

In nanocrystal titania, specific surface area was large since the particle diameter was in nanosize range, and surface energy storage property was readily available, and surface and interior of solid were thought to be two phases in coexistence as region that reacted electrochemically with lithium. That is, there were two different lithium storage mechanisms, where former was surface lithium adsorbed accompanied by charge transfer on surface of nanocrystal, while latter was lithium stored by intercalation inside the nanocrystal. The faradaic capacity by surface adsorption manifested as pseudo-capacity, and stored lithium inside the solid manifested as intercalation capacity. These two different lithium storage mechanisms were shown clearly in



**Fig. 6 Charge-discharge curve of nanocrystalline titania.**

charge-discharge curve, as shown in Fig. 6. In nanocrystal titania, charge-discharge curve differed greatly from that of bulk material. The intercalation capacity in ordinary titania solid was normally 168 mAh/g for equilibrium composition  $Li_{0.5}TiO_2$ , whereas this increased to about 230 mAh/g in the research result. This capacity increased with smaller crystal size. It was thought that increase in capacity occurred due to lithium stored on the nanocrystal surface as pseudo-capacity on the surface in addition to intercalation capacity.

In fact, as shown by the charge-discharge curve, lithium was stored at certain site potential up to  $Li_{0.5}TiO_2$ , which is theoretical capacity of lithium composition, so discharge curve showed constant voltage of about 1.75 V. When this lithium composition was surpassed, pseudo-capacity discharge curve appeared with gradual decline in voltage along with increased capacity. Large pseudo-capacity that appeared in region below 1.75 V was apparently oxidative-reductive capacity of titania, and might represent the capacity of lithium that was electrochemically adsorbed to the surface of active material. These did not have constant site potential, and the voltage was thought to decrease, as the surface turned metallic as lithium concentration on titania surface increased.

On the other hand, the most important issue when designing high-power active material was question of most appropriate nanosize. From the result of size dependency of Fig. 6, when crystal size of titania was 100 nm and 30 nm, the electrode property increased, and the effect of nanosizing became apparent for active material of 30 nm size. Moreover, nanocrystal property became dominant in 6 nm size active material, different from the bulk material. From these results, there was great possibility in using active material with size less than several 10 nm when utilizing innovative nanocrystal property.

For charge-discharge mechanisms using pseudo-capacity on titania surface, high-speed charge transfer became possible since they did not involve slow diffusion process inside the solid. As shown in Fig. 7, intercalation inside the nanocrystal showed good electrochemical property for battery material since plateau voltage occurred in two-phase coexistence. However, this storage mechanism was not suitable for high-speed charge-discharge since it accompanied slow charge transfer process due to diffusion within solid. On the other hand, surface pseudo-capacity mechanism by adsorption and desorption of ion on the surface may enable charge transfer and lithium storage in second order since they did not accompany diffusion process. In fact, the change of discharge capacity when charge-discharge current density of titania crystal with anatase structure of 6 nm and 30 nm were changed is shown in the figure. In both cases, intercalation capacity tended to decrease as the current density was raised, and while capacity decreases severely in titania of 30 nm

crystal size, titania with 6 nm crystal size showed little decrease under condition of large current density of 10 A/g, and we found that high-speed intercalation was taking place within the crystal.

In charge-discharge curve, capacity decreased severely in the flat part of voltage in 30 nm crystal, and it was found that the intercalation of lithium ion inside the crystal was inhibited under condition of large current density. On the other hand, when pseudo-capacity mechanism of nanocrystal surface was used, the pseudo-capacity at surface did not decrease much even when the current density was raised. This showed that charge transfer at surface reaction occurred at extremely high speed. In 6 nm nanocrystal, the pseudo-capacity content of the surface did not decrease even at high-speed charge-discharge to 40 A/g, and this implied that lithium ion possessed reversible high-speed storage property. That is, in nanocrystal active material, pseudo-capacity appeared due to large specific surface area, and lithium storage at over stoichiometric composition became possible. Moreover, new electrode property where high-speed lithium storage mechanism appeared at surface without intercalation into the solid was discovered. Using this energy storage property unique to nanocrystal, it would be possible to realize innovative high-capacity high-power lithium battery electrode material.

In industry-academia-government vertical collaboration development, it was necessary to synthesize nanocrystals using active materials, for which the battery manufacturer was considering product realization, and to investigate their high output property. If nanocrystals were synthesized using active materials used in the products and their high-capacity, high-power, and high-cycle properties were demonstrated,

it should lead directly to practical use. To utilize the concept of nanocrystal active material in product development of batteries for power tools by Hitachi Maxell, one of the participant of this project, we developed the fabrication process of nanoporous structure and nanocrystal synthesis of lithium titanate  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  from which high output property could be expected among titanium oxide materials<sup>[4][5]</sup>.  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  active material is electrode active material with negligible level of crystal structure change in insertion and extraction of lithium ions, and therefore has drawn attention as electrode material with excellent charge-discharge cycle property. Therefore, AIST added polymer as dispersing agent for introducing mesoporous structure when synthesizing the active material, fabricated nanoporous structure electrode composed of framework of sequential mesopore and nanocrystal active material, and conducted evaluation of high output property.

To introduce mesopore with high ion diffusability to nanocrystal electrode body, template polymer P123 (Pluronic; EO20PO70EO20) was added during sol-gel synthesis of  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  electrode to promote dispersal of nanocrystal  $\text{Li}_4\text{Ti}_5\text{O}_{12}$ . After adding polymer to primer, and firing in air for 6 h at 400 °C and then for 2 h at 750 °C, we fabricated nanoporous structure electrode where  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  particles with size about 60 nm were linked in highly dispersed manner. When the electrode properties were evaluated, as can be projected from the concept of low-resistance, high-ion diffusion electrode, we obtained sufficient electrode capacity at high charge-discharge current density for  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  electrode with nanoporous structure, and the cycle property was good.

According to AIST research, it was found that the output

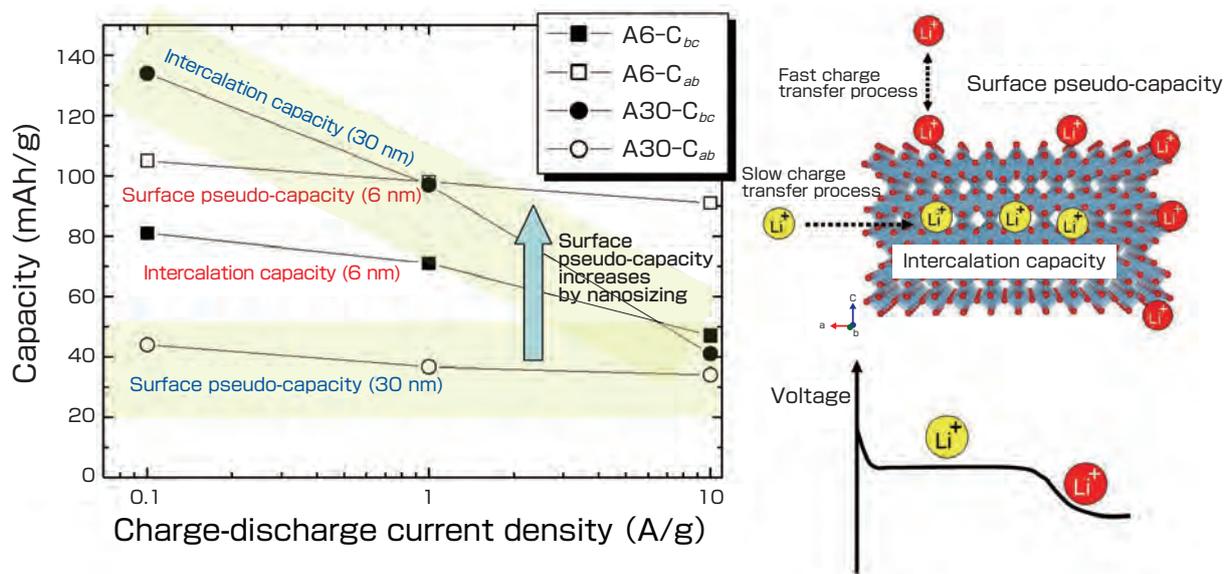


Fig. 7 Pseudo-capacity mechanism of nanocrystalline titania surface and its high-rate charge-discharge property.

property improved by adding nanoporous property to active material size of several 10 nm in  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  with high possibility for high-power negative electrode. In titania ( $\text{TiO}_2$ ), which is similar titanium oxide, we studied the electrode property to 6 nm size, and clarified the energy storage property characteristic of nanosize material such as high-speed charge-discharge property and pseudo-capacity from the perspective of basic chemistry. On the other hand, existing battery products used bulk ( $\mu\text{m}$  level) size electrode, and there was no product realization using such small active material. Therefore, there was no systematic study on active material size for optimizing the battery power property in either industry or academia. Therefore, it is necessary to continue industry-academia-government collaboration research to investigate which nanosize shall be set as goal for nanotech electrode in product realization phase, or to constantly keep in mind what is the optimal active material size for high output property.

This was an important technological item for linear connection of R&D by university, AIST, and battery manufacturer. As shown in Fig. 8, there was unexplored area or “missing region” of size between the size regions explored for active materials in basic research by university and AIST and the size regions sought for the products by battery manufacturer. It was imagined that the optimal value for output property lies in this missing region. In vertical collaboration, it was important to study this size region from both basic and practical sides to quickly clarify optimal size, and to investigate the optimal solution in battery cell. In fact, region of active materials of several 10 nm to several 100 nm was not systematically surveyed until now, and R&D in the missing region was an important milestone in this vertical collaboration. In fact, as explained in the following example of Hitachi Maxell, battery prototypes were created using active materials of different sizes from 55 nm ~ 200 nm using

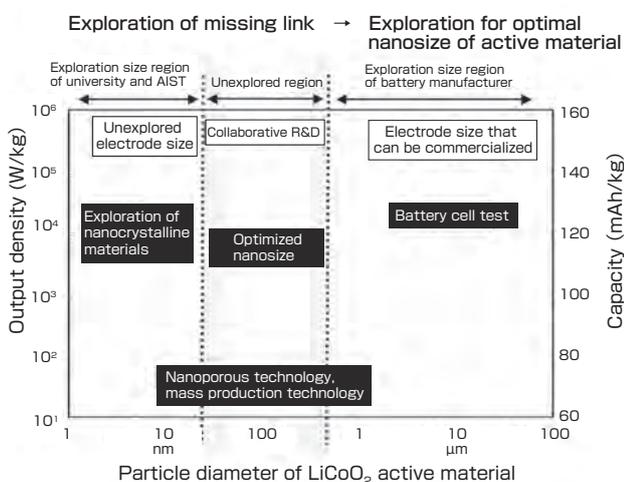


Fig. 8 Exploring active material size in the missing region to optimize the electrode property.

spinel structure manganese  $\text{LiMn}_2\text{O}_4$  for practical electrode, and the output characteristic was evaluated. Active material size was shifted from bulk range to nanosize range in steps, and actual exploration was done on which size would be optimal for capacity and output within the missing region for intercalation electrode. This R&D project was probably the first systematic investigation of size dependency of electrode property for optimization of active material size for product realization, rather than just synthesis and property clarification of nanosize active material.

Next, in vertical collaboration, investigation of quick innovation potential of university and AIST was necessary. To investigate the potential for application to product using the excellent electrode properties of nanocrystal material that were learned from basic research, prototype creation and performance assessment of battery cells were conducted. Prototype fabrication was done by Hitachi Maxell that fabricated and evaluated standard spec laminated cell.

First, I shall describe the fabrication of the prototype battery. The electrode body used in the experiment was fabricated by mixing electrode active material, conductivity aid, and PVDF dispersion fluid binder, that were each weighed to target composition, in planetary ball mill, and paint-form electrode sol was fabricated after adjusting the viscosity. This paint-form electrode sol was applied using an applicator on of 15  $\mu\text{m}$  thick aluminum foil so the dried weight would be 5.0~7.0  $\text{mg}/\text{cm}^2$  for both positive and negative electrodes, and the electrode body was finished by pressing after drying. The structure and photograph of laminated battery cell are shown in Fig. 9.

Considering the result of high output property of nanocrystal electrode at AIST, prototype cell with nanosize electrodes for both negative and positive electrode was fabricated. Negative electrode was made of 100 nm  $\text{Li}_4\text{Ti}_5\text{O}_{12}$ , while positive electrode was made of  $\text{LiMn}_2\text{O}_4$  nanocrystal active material of different sizes from 55 nm to 200 nm, for purpose of finding optimal active material size in the missing region.

Although there were some variations in  $\text{LiMn}_2\text{O}_4$  for active material size 150 nm or less, no significant decrease of discharge capacity occurred to 2 A/g rate. From active material

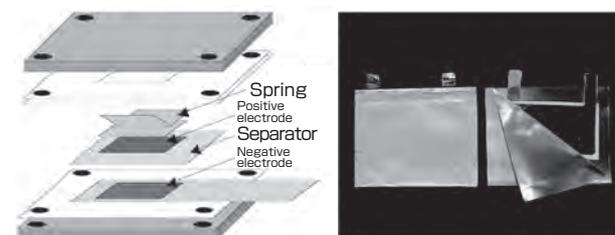


Fig. 9 Structure and photograph of laminated battery cell prototype.

size dependency of capacity retention, the property increased under high-speed charge-discharge condition of 5 A/g or over when the particle size decreased, and active material with 55 nm size showed best output property (Figure 10). According to AIST's calculation based on diffusion theory, for particle diameter of 100 nm or less, lithium ion diffusion within the particle would be sufficient for 100 charge-discharge per hour. Looking at the actual prototype cell data, when electrode using active material size of 150 nm or less was used, charge-discharge was possible even at high current density, and it was found that output property improved as active material size decreased. As result of optimizing the active material size in the missing region, best output property was obtain in battery cell using negative electrode (100 nm) of nanocrystal active material  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  with spinel structure and positive electrode (55 nm) of  $\text{LiMn}_2\text{O}_4$ . Its high output property was demonstrated at battery cell level using the innovative energy storage mechanism of nanocrystal electrodes. Finding the optimal size for nanocrystal active material to be used in high output battery was the most important issue since

the commencement of the project. While the development of high-power battery using nanosize active material is becoming fierce around the world, to find out were the optimal solution lies in the 1 nm ~ 100 nm size range will solidify the foundation of innovation in storage technology.

In this R&D, AIST clarified by experiment that nanosize active material was effective for realizing high-capacity and high-power properties through its characteristic lithium storage mechanism. Moreover, by engaging in systematic exploration of missing region through vertical collaborative development for quick practical application, and by evaluating the output and cycle properties of the prototype battery, it was clarified by experiment that optimal solution was active material of size around 50 nm. Currently, while R&D of high-power lithium secondary battery using nanosize active material is accelerating around the world, this was the first R&D that specifically pursued which active material size was optimal for battery products. In the vertical collaboration project lead by the Author, optimal active

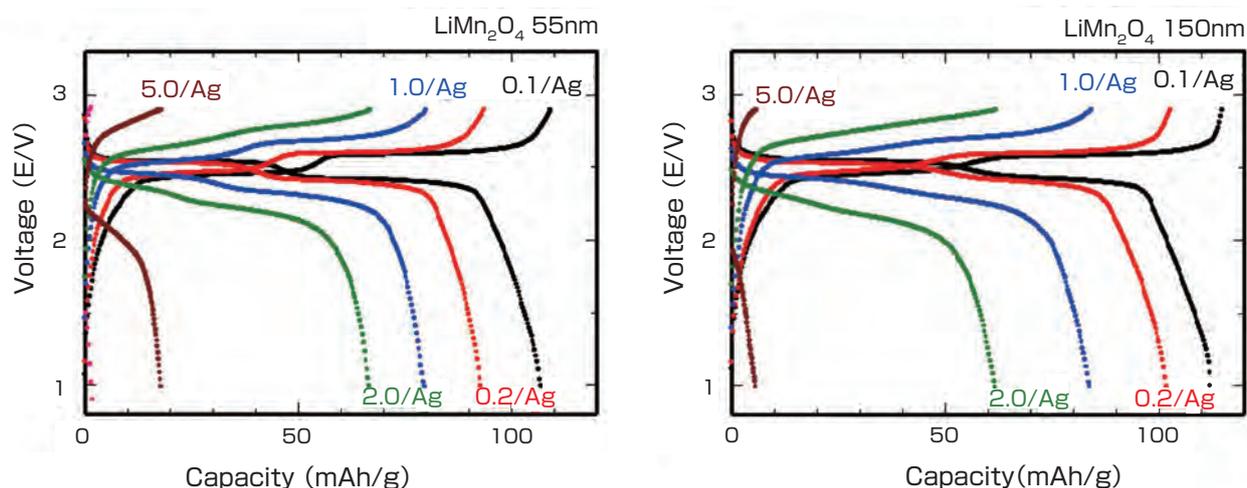


Fig. 10 Output property of lithium battery using nanocrystal electrode.

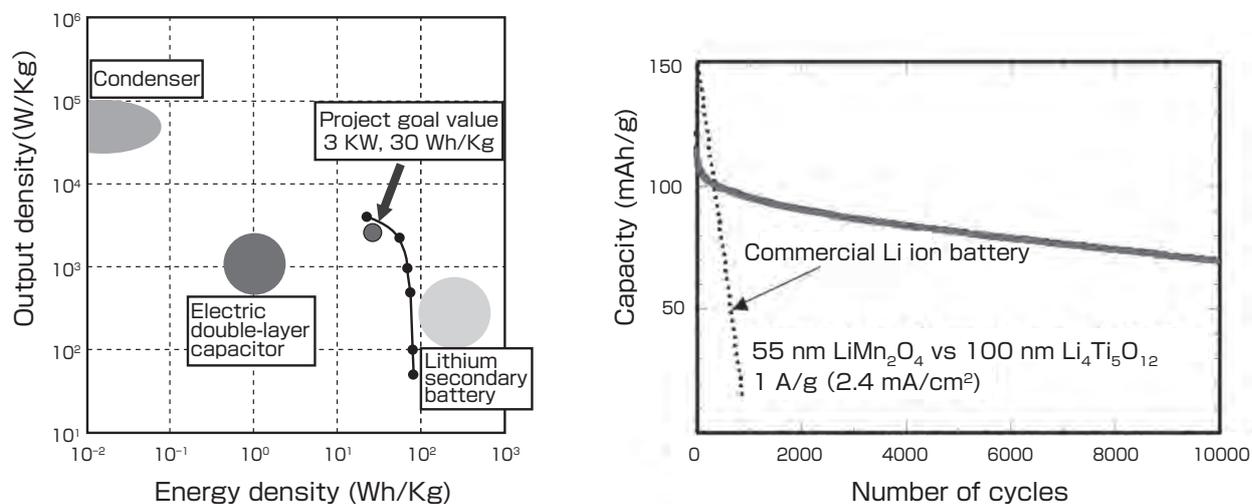


Fig. 11 Ragone plot of battery performance of prototype and charge-discharge cycle property (higher performances compared to existing batteries were observed for both items).

material size was found for product specification as result of systematic exploration of missing range of active material size in order to create innovative battery product quickly using excellent lithium storage property of nanosize active material that was found by university and AIST. Moreover, we fabricated the prototype battery cell was fabricated, and high output and cycle properties were confirmed to be applicable as product.

Figure 11 is a Ragone plot of the battery performance of prototype fabricated for the project. It surpassed the performance of existing lithium secondary battery products, and achieved high capacity and output properties that were set as project goal values. In addition, charge-discharge cycle property, which is the most important specification in product realization, was investigated to 10,000 times under high output condition, and capacity retention of 60 % was obtained. Hence, its superiority compared to existing battery products was shown, and it was found that it had reliability that could withstand product realization. Currently, product development of high-power battery utilizing the innovative energy property of these nanocrystal active materials is in progress. AIST generated the concept of nanocrystal electrode as innovative energy storage material, realized high output property at basic research phase, academically clarified the innovative energy storage property that arose from surface effect and size effect, and then demonstrated high-power lithium battery utilizing the excellent electrode properties of nanocrystal active material through collaboration with battery manufacturer.

#### 4 “Shortening distance” to innovation through interdisciplinary fusion and industry-academia-government vertical collaboration

As strategy for “shortening distance” to innovation as described in the paper, Full Research was accelerated by interdisciplinary fusion and vertical collaboration of industry-academia-government, as shown in Fig. 12. I shall emphasize the point that characteristic of AIST is that innovative new technology is often generated through interdisciplinary fusion. Since it has high number and density of researchers, and has wide research spectrum that covers almost the entire range of industrial technology from standard, geology, bio, to information, fusion of diverse disciplines and integration of elemental technologies can be done readily. This means that it is a research organization with high potential for innovation appropriate for creating new technologies and concepts never seen before. Utilizing this organization structure, it is capable of generating innovative sprout technology in the interface of the disciplines. That is, bud of new technology can be created in diverse interface regions such as bio and energy, standard and nanotech, or electronics and environment, and high innovation potential that will serve as platform of innovative technology can be created.

Next point to be considered is the methodology of how to investigate the efficacy of such innovation potential in short time, or synthesesiology of innovation. Taking as example the R&D for high power lithium battery, which the Author lead in the NEDO project, I shall discuss the R&D process that can investigate the efficacy of innovation potential in short time, for example in few years, through the vertical collaboration scheme of industry-academia-government. As shown in Fig. 12, vertical collaboration is organized by gathering the university (located upstream) that engages in basic research, AIST that engages in Full Research, and battery manufacturer and automobile manufacturer who is the end user (downstream) into one project that is vertically

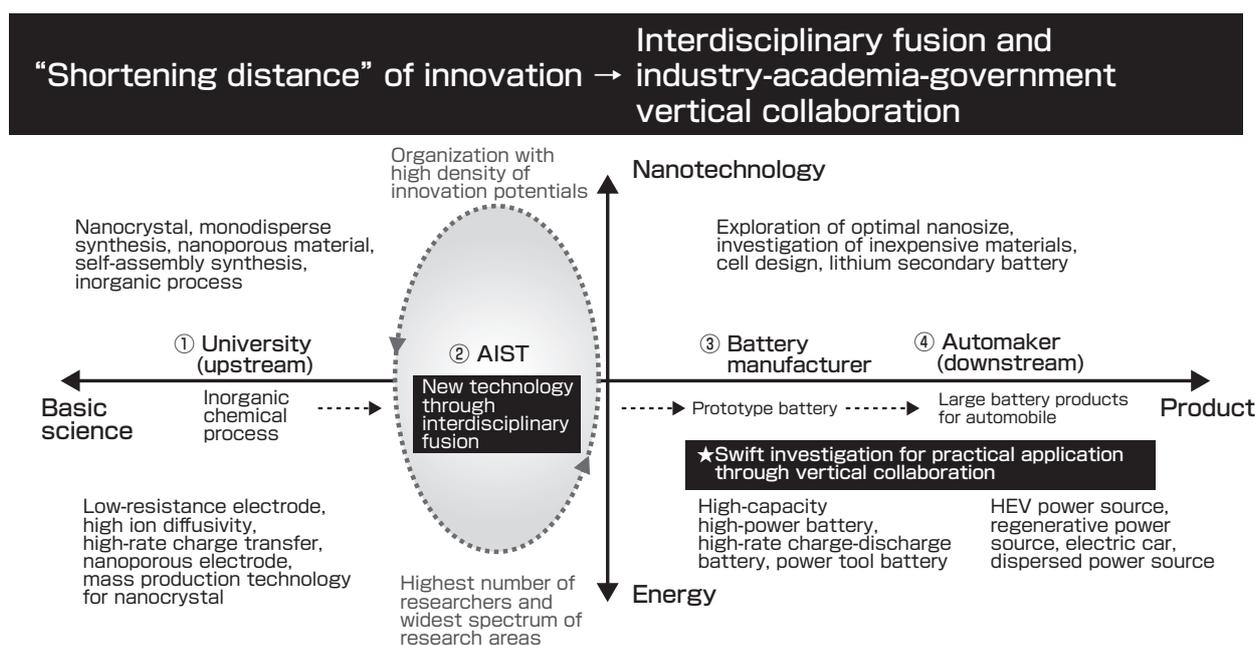


Fig. 12 Synthesiological method for innovation by industry-academia-government vertical collaboration.

linked, to push the innovation potential quickly to realization. In this collaboration scheme, the new technology of university and AIST is transferred quickly to the automaker, which enables quick investigation of efficacy and reliability. Technological request of the automaker can be back cast (fed back) to AIST and university through the battery maker, and this may provide direction for the basic research. By sharing this bi-directionality among the participating organizations, close information exchange and basic research that matches the product specification will be possible, and may lead to effective R&D process that enables practical result in short period of few years.

Next, what is the role of AIST in such new R&D process? In short, AIST can function as central innovation hub in such vertical collaboration. It is capable of creating innovation potential effectively and efficiently through interdisciplinary fusion because of the high number and density of researchers and wide spectrum of research fields. To find subjects that can be applied to product amongst the new technologies generated by interdisciplinary fusion, the industry-academia-government vertical collaboration project is effective as explained in this paper. As shown in Fig. 12, the process of quick technological transfer of innovative technology created by interdisciplinary fusion to product that has been set as clear goal using the industry-academia-government vertical collaboration scheme is an effective convergent R&D process that enables “shortening distance” to innovation. AIST functions as central innovation hub in this process.

In this paper, it was demonstrated that convergent R&D

process was successful in innovation of high-capacity high-power lithium secondary battery, which is key technology for HEV power source. In synesthesiology of innovation where collaboration is assumed, there are two methods: (1) vertical collaboration development as convergence process where clear product goal is set and various elemental technologies are integrated, and (2) horizontal collaborative development as co-creation process where diverse new technology is created without setting clear product goal, as shown in Fig. 13. In the NEDO project for which Author was the R&D leader, there was clear goal of developing a specific product i.e. high-power high-capacity lithium secondary battery for HEV, in shortest time possible. Therefore, the former convergence process was employed, R&D was conducted under industry-academia-government vertical collaboration scheme that involved university and company, and quick realization of innovation potential was attempted through fusion of nanotech and energy fields. The convergence mechanism that effectively integrates elemental technology is most appropriate to achieve clear product goal in “shortest distance,” and it was actually possible to develop innovative high-power battery in short period.

In conventional collaborative process, the idea of basic research from university is transferred to AIST, technological development is conducted right up to turn-over to industry such as measurement of performance and mass production process, and then technology is transferred from battery manufacturer to automakers. However, in such successive collaborative development, when the direction of R&D shifts according to the interest of the researchers at each

### Synesthesiological method for innovation in R&D process

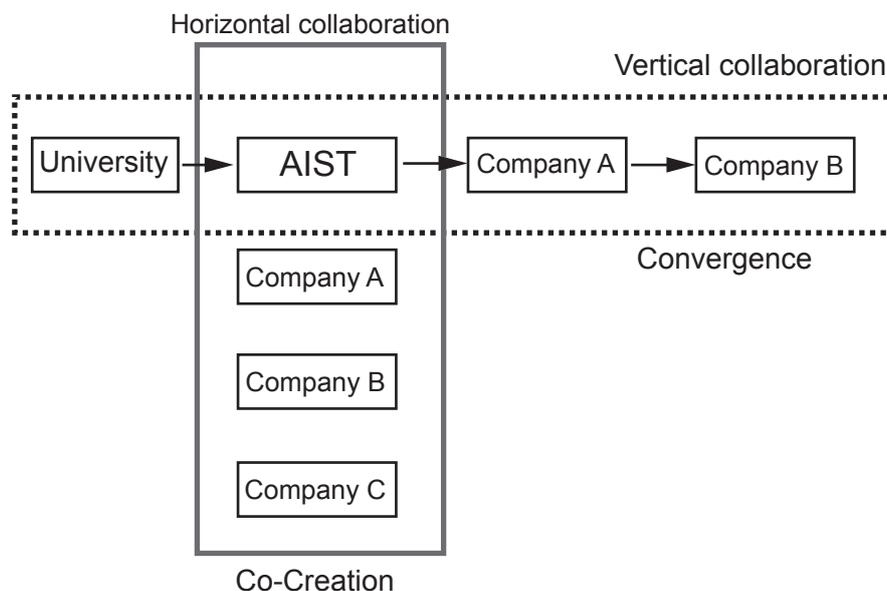


Fig. 13 Two-dimensional *synesthesiological* method for innovation.

organization, the end user downstream may receive some technology quite different from one they were expecting. In this industry-academia-government vertical collaboration, the direction of basic research was set by participation of all organizations at commencement of the project and the technological development demanded by the automaker was fed back to university and AIST, to speed up downstream technological transfer that usually takes time and to ensure accurate technological transfer needed for the product. Distance between upstream and downstream was “shortened” by maintaining straight passage of technology flow. We succeeded in creating extremely hopeful innovative material, nanocrystal electrode, in three-year development period, and were fortunate to be able to develop innovative technology and built bridge between nanotechnology and energy technology that are generally considered difficult to join. Although the project achieved sufficient performance as power source for HEV, the product development of small cell is still in progress, using nanocrystal electrode for power tool that can be commercialized in few years.

Figure 13 shows the synthesiological method of innovation that are categorized as R&D processes, and convergence mechanism with vertical collaboration as explained in this paper is effective for product for which final goal is clear. For realization in short time period, it is most efficient to converge technology into the final product by integrating various elemental technology, and under vertical collaborative system, the technology transfer can be accomplished in shortest time period. On the other hand, if the goal is to create innovative sprout technology that does not exist or to seek diverse and highly generalized technological standard, co-creation mechanism with horizontal collaboration is better. This can generate diverse innovation potential and contributes to wide-ranging industry in horizontal manner. Although this paper described the example of convergence mechanism, both mechanisms are important as *Synthesiological* method of innovation at AIST. It is necessary to continue refining the methodology.

## 5 Future issues

Figure 1 shows the overview of the battery industry, and for application to automobile power source, upsizing process, safety, and cost performance are required at high level in addition to capacity and cycle properties. Since the development period of this research was only three years, we did not reach realization of automobile power source. The vertical collaboration scheme is mechanism that enables swift and sufficient technological transfer to all collaborating organizations. On the other hand, there is still very high hurdle in directing research through feedback from automaker to university and AIST and straight technological transfer from basic research to automaker. For upsizing battery, there will be no technological leap forward from

small size level of cell phones and laptops to large battery for cars, but the technology will develop through several intermediate phases. In that sense, to create innovations, it is necessary to conduct industry-academia-government vertical collaboration that takes in consideration realistic conditions such as limited time and budget, as well as market strategy and technological potential of participating companies.

## 6 Summary

As diversity and speed are demanded in innovations and various collaborative R&D are sought, this paper discussed the convergent *Synthesiological* method of innovation by interdisciplinary fusion and industry-academia-government vertical collaboration as effective R&D process, using the example of high-power battery development. Based on the chemical synthesis process created at the university, AIST succeeded in developing nanocrystal electrode, which is high-performance active material that realizes high capacity and high output properties. As result of collaborative development with battery manufacturer to apply the active material concept to products, it succeeded in fabricating prototype of high-performance lithium battery with 30 Wh/kg and 3 kW/kg, which are performances demanded for regeneration power source for HEV. Moreover, superior cycle property needed for product realization was obtained compared to current product. Currently, R&D of product that will be commercialized in few years as power tool battery is in progress to utilize the result of this vertical collaboration development at participating battery manufacturer. Nanocrystal electrode is innovation of storage technology that was born from the fusion of nanotechnology and energy technology, and industry-academia-government vertical collaboration was a scheme appropriate for achieving strategic goal in short time through swift investigation of efficacy of innovation potential.

As discussed in this paper, creation of innovation potential that cannot be done by university or industry alone can be done easily at AIST, which is a consolidated research center that is capable of becoming core research institute in collaborative research. The reasons are because there are many researchers and wide research spectrum exists within one organization, and AIST can create diverse innovation potential highly efficiently through fusion of various disciplines. It was also demonstrated that R&D in vertical collaboration with participation by end user company was effective to swiftly check the possibility of application to target product. The new *Synthesiological* method of innovation (interdisciplinary fusion plus vertical collaboration) is an extremely effective scenario in speeding up R&D. In the R&D for high-power lithium secondary battery using nanotechnology, it was possible to develop the nanocrystal electrode, which is innovative material technology, to product realization phase in short time of three

years. This scenario is also effective R&D process in other industries such as bio, information, nanotech, manufacturing, environment, and energy, particularly in short range project where the final product goal is clear.

## 7 Acknowledgements

I thank all people who cooperated in this industry-academia-government vertical collaboration development for high-power lithium secondary battery. Particularly, Zhou Haoshen, Group Leader of Energy Interface Technology Group, AIST contributed greatly for the property data when titanium oxide nanocrystal material was used as electrode material. Tetsuichi Kudo, Professor Emeritus of The University of Tokyo gave us essential idea for the concept of high-speed charge transfer that takes place within active material. I am grateful to the people of Hitachi Maxell, Ltd. for fabricating the prototype laminated battery and testing the battery properties. We received advice from Professor Moriguchi of Nagasaki University on the synthesis of porous electrode structure. I am also thankful to people of Nanotech Division, NEDO and Collaboration Promotion Department, AIST who gave us various advices in conducting this project.

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Institute, AIST. Doctor of Engineering. While at university, worked widely on new material for amorphous silicon solar cell and functional material process. After joining Electrotechnical Laboratory, worked on R&D for innovative power source devices based on nanotechnology such as polymer electrolyte fuel cell and super capacitor. Currently works on material development for high-capacity high-power lithium ion secondary battery. Worked as leader of R&D in the vertical collaboration project by four organizations, Nagasaki University, AIST, Hitachi Maxell, Ltd., and Fuji Heavy Industries Ltd. as “Research and Development of High Capacity & High Power Density Secondary Battery by Low Resistance, High Ion Diffusion Nanoporous Electrode” under R&D for Practical Utilization of Nanotechnology and Advanced Materials, NEDO conducted from 2005 to 2007.

## Discussion with Reviewers

### 1 The flow of vertical collaboration

#### Question and Comment (Koichi Mizuno)

Joint research in vertical collaboration style is method that has been addressed frequently. Can you indicate if there was any reverse transfer of R&D from the company to AIST, since collaborative research is not mere one-way flow from AIST to the company (battery manufacturer)? Of course, I see that the main topics of development were suggested by the company, but I think it will help explain vertical collaboration if you explain any bi-directional exchange of technological contents.

#### Answer (Itaru Honma)

AIST selected titania TiO<sub>2</sub>, which was appropriate material for basic research since it was available in various sizes, to academically clarify the nanosize effect of electrode material, and systematically studied the electrode property of this oxide. In the basic research phase, we studied the appearance of surface capacity accompanying nanosizing, clarified that the nanocrystal active materials were suitable for high-speed charge-discharge, and explored optimal nanosize. Joint development was done by communicating to the companies in this industry-academia-government project the fact that excellent high capacity and high output properties of nanosize active material could be used in actual active material.

In fact, Hitachi Maxell was using Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub>, which is similar titanium oxide to titania on which AIST was working, for negative electrode material, but to downsize this material to nano level and optimize capacity and output properties, AIST and companies pointed out the necessity for clarifying which Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub> size was optimal for use in commercial battery by exploring the missing range of active material size, as shown in Fig. 8.

Optimal nanosizing of practical electrode material listed in the commercialization plan was conducted through bi-directional information exchange and development plan of technological transfer of sizing effect of titania (fore-cast from AIST to Hitachi Maxell) and practical exploration of nanosize active material (back-cast from Hitachi Maxell to AIST). The 55 nm LiMn<sub>2</sub>O<sub>4</sub> and 100 nm Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub> used in prototype cell data were results for optimal nanosize found in this collaborative development, and as described in the paper, output property that far surpassed the conventional battery performance was obtained.

### 2 Surface pseudo-capacity

#### Question and Comment (Koichi Mizuno)

The “pseudo-capacity” on surface is the key point in increasing charge-discharge capacity rather than intercalation

inside the electrode solid, and I think it is a good example of application of nanosizing of active material. Although it is described that in surface pseudo-capacity, “the surface becomes metallic as lithium concentration increases and voltage decreases,” it may be helpful if the author explain the reaction mechanism and specific image of the surface related to the pseudo-capacity.

**Answer (Itaru Honma)**

To provide simple explanation, as shown in Fig. 7, lithium ion causes intercalation (insertion of ion) inside the solid in bulk reaction, and at the same time, oxidation reduction of the compositional metal occurs to store electric energy. It arises from the essential (perhaps standard?) electrochemical reaction of the material. Therefore, no matter who conducts experiment anywhere in the world, as long as material with same stoichiometric composition is used, same amount of lithium ion is stored in equilibrium voltage. However, the storage mechanism at surface is different from the essential (bulk) property unique to the material, and it is non-unique property that manifests because a surface exists. Therefore, difference occurs in capacity property due to change in specific surface area or plane direction (crystal morphology) (that is, data will differ according to experiment group).

In Figure 7, it is described as surface reaction where electron enters the titania surface layer and causes single-electron reduction of  $Ti^{4+}/Ti^{3+}$  at same time as adsorption of lithium ion to the surface in the fast charge transfer process. That is, regardless of crystal structure, it is thought that pseudo-capacity originates from surface reaction. Moreover, the two-electron reduction of  $Ti^{4+}/Ti^{2+}$  that does not occur in bulk becomes possible in nanocrystal surface with special chemical bond status for lithium ion, and that produces large capacity. The lithium ion shown in red in Fig. 7 is presenting special surface reaction, and shows that the lithium is stored at higher concentration than bulk. If two-electron reduction takes place,  $Ti^{2+}$  will appear, so voltage decreases and the surface becomes more metallic as shown in bottom figure.

In addition, grid expansion occurs in nanocrystal and crystal structure (phase) different from bulk becomes stable, and the surface is not necessarily same as surface of bulk. These are extremely interesting research subject. In other words, there is possibility that new storage mechanism may appear, and we intend to pursue this basic research further.

### 3 Collaboration of AIST with battery manufacturer and automobile manufacturer

**Question and Comment (Akira Ono)**

I think this research demonstrates advantages of the vertical collaboration of industry-academia-government. Then, I ask the Author, who was the project leader, about the collaboration. In this project, what specific suggestions did AIST receive from the battery manufacturer, and how were they reflected in the project? Also, what specific suggestions did AIST or the battery manufacturer receive from the automobile manufacturer, and how were they reflected in the project? Please answer from the standpoint of project leader.

**Answer (Itaru Honma)**

It has been found that high output can be obtained using

nanosize active material and basic research is accelerating in this area, but I was surprised that there was absolutely no research on which nanosize materials were optimal for  $LiMn_2O_4$  and  $Li_4Ti_5O_{12}$  that are practical electrode materials. As shown in Fig. 8, there is missing range in active material size, and the battery manufacturer suggested that we should seek physicochemical knowledge of nanosizing effect by exploring this unexplored range, and search for sizes that generate highest capacity and output properties, as most important items of development. It is important from perspective of basic research to systematically investigate the size effect of active materials in nano range, and the surface effect that becomes clear in the process and size effect of charge transfer process of ion and electron in that process become very important guides for material design in developing high-power electrode. AIST set direction of basic research to respond to the expectation of the battery manufacturer, and studied the nanosizing effect of metal oxide materials such as titania, as described in the paper. The automobile manufacturer indicated that low cost and productivity are important issues in actual commercialization, and we worked simultaneously on mass production process of nanosize active material based on this suggestion. Although not included in this paper, we developed new active material synthesis process where nanocrystal active material could be mass-produced in kilogram level using molten salt method.

### 4 Collaboration of AIST and university

**Question (Akira Ono)**

What specifically was the collaboration between Nagasaki University and AIST? Rather than transferring just the technological potential of Nagasaki University to AIST, can you talk specifically about the interaction and coordination between the two organizations from the standpoint of project leader?

**Answer (Itaru Honma)**

Nagasaki University (NU) made major contribution in developing the basic chemical process. AIST has been studying the nanosizing effect of active material, and NU selected the reaction process appropriate for synthesis of the material. Specifically, they conducted reaction kinetic investigation for the hydrothermal synthesis and molten salt synthesis methods used in this project, and we received basic but very important advices on starting material and solvent types. Moreover, they conducted basic research on low cost synthesis that was requested by the automobile manufacturer. Particularly NU investigated the practical process where synthesis could be accomplished as a one-step firing process for carbon high-capacity electrode material. The result is being utilized in battery development by the automobile manufacturer.

In the collaboration of AIST and NU, to maximize each other's research potential in extremely short period of three years, the development of nanocrystal active material of metal oxides was done by AIST, while the development of nanoporous and high-power carbon materials that are essential as conductivity aid in actual battery electrode was done by NU. By doing so, we were able to design innovative high-capacity high-power electrode through combining the two results during the course of project.