1 Introduction

With the goal of promoting energy conservation and exploring forms of energy alternative to petrified sources, as well as of reducing environmental burden, much effort has been spent on developing new breeds of vehicles. These include Hybrid vehicles (HV) that run on a combined system of a fossil fuel engine and an electric motor, Plug-in HVs (PHVs) that allow recharging of the in-vehicle battery using a commercial electric source, and Electric Vehicles (EVs). As the electric source to drive these vehicles, the development and introduction of a high-power, large capacity secondary battery is a challenge of urgent concern, and the lithium-ion battery has become the focus of attention.

The lithium-ion battery, with its advantage of high energy density, has scored impressive advances in the field of information communication and the home appliance sector, especially as a portable power source. Against the backdrop of widespread proliferation of mobile phones and notebook PCs, global shipments of lithium-ion batteries amounted to ¥390 billion in FY 2008.[1]

The lithium-ion battery has a host of advantages including: that next to no side reaction occurs when a lithium ion intercalates into or desorbs from the positive/negative electrode material, the possibility that energy efficiency may be further enhanced by the lowering of the battery’s inner resistance, a small self-discharge, and no memory effect. Research and development, up to the present, has focused on upgrading the energy density. Japan put the lithium-ion into practical use ahead of all other countries, and has maintained the global lead in terms of both technological edge and production.

For highly efficient use of energy for automobiles in the future, full use of the secondary batteries should be an integral part of automobile development. The battery provides electric power for driving, while enabling instantaneous recovery and storage of braking energy in quick response to the running conditions. Among in-vehicle secondary batteries, the lithium-ion battery is the most promising because of its superior characteristics in terms of power and energy density per unit weight or volume. The majority of the currently used secondary batteries are nickel-hydrogen type, and the shipment value as of FY 2009 is expected to reach ¥220 billion. This value is based on the market size forecast for HVs in 2009 (approximately 1.1 million vehicles[3]) and assuming a battery price of ¥200,000 per unit.[2]

According to a Nomura Research Institute forecast, the HVs market size will grow to about 2.2 million units in 2012.[3] Given that the lithium-ion battery will be used in all these vehicles in place of the nickel-hydrogen battery and that EVs will enter widespread use, the shipping value of lithium-ion batteries for vehicle use will far exceed ¥440 billion. Which means that the shipping value of lithium-ion batteries for vehicles has the potential of catching up and even exceeding the shipping value of lithium-ion batteries for information communication and home appliance devices. Mitsubishi Motors Corporation and Fuji Heavy Industries Ltd. have started mass production of EVs equipped with a lithium-ion battery, and Toyota Motor Corporation has launched its PHVs that utilize lithium-ion batteries into a limited market area starting in mid-December of 2009, aiming at full-scale commercial availability in the market starting in 2011. Nissan Motor Co., Ltd. also announced in 2010 its plan to mass-produce lithium-ion batteries for EVs in Japan, U.S., and Europe. It is very likely that the adoption of lithium-ion batteries for HVs will continue to expand, and according to a Fuji-Keizai forecast, the
market size of the lithium-ion battery for vehicles is expected to step across the ¥2 trillion mark around FY 2014.\(^5\)

In sectors other than the transport equipment sector, the adoption of a small- to large-scale lithium-ion battery system is desired for storage and for the stable supply of electric power. For example, renewable energy sources such as solar and wind power generation require power storage because of their weather dependent nature, and large-scale power generation requires electric-load leveling to compensate for the time lag between production and consumption.\(^6\) Therefore, in the long term, the demand for lithium-ion batteries has the potential for becoming larger than the estimated value based on the assumptions described above.

This article describes, among other subjects in the general R&D trends of in-vehicle lithium-ion batteries, the status and challenges of the electrode and electrolyte materials implemented in the battery, which may hold the key to improving performance. It also deals with the desirable future scheme of proceeding with R&D of electrode and electrolyte materials for efficient realization of high output power and large capacity, as well as achieving superior safety.

\section{High-power, Large-capacity, Secondary Batteries Pave the Way to a Low-carbon Society: Options Available for Research}

In the sectors of renewable energy systems and transport equipments, a substantial reduction of environmental burden can be achieved by efficient use of high-power, large-capacity, secondary batteries. In the transport equipment sector especially, domestic CO\(_2\) emissions have turned into a general downward trend thanks to the progressive use of environmentally friendly cars, typically HVs, which began around 2000. Promoting the adoption of PHVs and EVs in the coming years will enable further reduction of CO\(_2\) emission. Wide spread use of EVs, which typically emit 1/4 of the amount of CO\(_2\) (approximately 50g/km) that conventional gasoline engine vehicles emit, can facilitate a substantial reduction of vehicle-originated CO\(_2\) emissions.\(^6,7\) For example, if 50\% of vehicles owned in Japan (75.7 million vehicles)\(^8\) were replaced by EVs, the reduction achievable is estimated to be approximately 40\% of the current total CO\(_2\) emissions from vehicles (approximately 100 million tons out of 257 million tons, as of 2007).

Currently available secondary batteries (nickel-hydrogen batteries) have a typical capacity of around 1.5kWh, and are capable of providing mileage of only about 10 km.\(^6,10\) Upgrading the battery capacity to 50kWh would extend the continuous running mileage to approximately 330 km. A significant enlargement of the secondary battery’s capacity is essential for HVs and EVs to run longer. EVs and HVs require different power and energy characteristics from batteries. HVs have limited space for mounting battery cells because they have a fossil fuel engine as well, and the weight of the battery cells is also restricted for the same reason. Thus, a higher power density is required from the battery cells to secure enhanced driving performance. EVs have the advantage of having a simplified vehicle driving system and no fuel tank, which results in fewer components. This enables a significant trimming of vehicle weight as well as a larger space for mounting a larger number of battery cells. The priority of the secondary battery for EVs is enhanced energy density, rather than power density, in order to extend travel distance. The secondary battery for a PHV has a use environment between that of a HV and an EV.\(^11,12\)

Figure 1 shows the relationship between power density and energy density of currently available major secondary batteries (lithium-ion, nickel-hydrogen, electric double layer capacitor, and others),\(^13\) and the directions of the development program aiming at vehicle applications. An ideal secondary battery should have a high power-per-unit volume/weight, large capacity, long charge-discharge cycle life, and a long chargeable life, in addition to being safe and affordable.

From the data shown in Figure 1, it can be seen that the methods for realizing a high-power, large-capacity secondary battery are broadly divided into three options, which are: (1) further efforts to enhance the power and enlarge the capacity of the currently available lines of lithium-ion batteries, (2) enlargement of the electric double layer capacitor, and (3) the development of a lithium-ion capacitor. Although the electric double layer capacitor, (2), with its inherent high power density and superior endurance for repetitive charge-discharge cycles is counted on for vehicle applications, it is still in the basic research
phase and is currently viewed to have limited applications because of its low energy density. Some research efforts are underway to develop a lithium-ion capacitor, e.g., a combination of the lithium-ion battery and the electric double layer capacitor. In the immediate future, however, focus should be placed on R&D aiming at boosting the power and enhancing the capacity of the lithium-ion battery (1). The emphasis throughout the following chapters is on the R&D trends related to lithium-ion batteries (1).

3 Configuration of a Lithium-ion Battery and Charge/Discharge Mechanism

Among the metallic elements, lithium has the smallest electrochemical equivalent (e.g., the quantity of electric charge required to be transferred to trigger an oxidation-reduction reaction). The lithium-ion battery harnesses a set of reversible oxidation/reduction reactions, in which a lithium-ion dissolved by a discharge reaction returns to metallic lithium by a charging reaction. The negative electrode, which must be capable of directly incorporating a lithium-ion in its matrix, normally has a layered structure made of a carbon-based material. The lithium ion penetrates into (intercalation), and desorbs from the gaps between the layers of this carbon-based material (e.g., graphite).

At present, a type of lithium-ion battery (average output voltage: 3.6V) with a combination of graphite (negative) and lithium cobaltate (LiCoO$_2$) (positive) electrodes has become commercially practical.

Figure 2 schematically illustrates the lithium-ion battery’s charge/discharge mechanisms and the electrochemical reactions taking place in this negative and positive electrode construction. An electron and an ion are generated by an oxidation reaction (a reductant loses an electron) and a reduction reaction (an oxidant accepts an electron), and the energy released from this electron and ion provides the source of electric energy. The electromotive force (EMF) of the battery is determined by the difference of electrical potential between the positive and negative electrode materials. The material that generates EMF is called an active material. A battery requires, in addition to the electrodes, an electrolyte that allows ionic transport between the positive and negative electrodes, and a separator that prevents direct contact of the electrodes. Each reductant and oxidant has an oxidation-reduction potential, which is determined by the chemical species that constitutes these chemical agents. This electric potential is called the electrode potential. A material with low electric potential has a strong reducing power, and a material with high electric potential

![Figure 1: Power density vs. energy density of currently available major batteries: directions for their vehicles applications](image_url)
has a strong oxidizing power; thus a combination of these two electrodes can form a battery. While a battery is being charged, chemical reactions take place in the direction reverse to discharging, during which a reduction reaction takes place on the negative electrode and an oxidation reaction takes place on the positive electrode.\(^{[14-16]}\)

The power density designates the capacity of a battery to release energy in a short period of time, and is represented by the product of current and voltage (each per unit weight or unit volume of the battery). As the voltage is determined by the average voltage inherent to the electrode material, a reduction of the battery’s inner resistance is essential for obtaining a higher current, or power density. Electric charge retrievable from the battery \((Q)^*\) and the battery voltage \((V)\) determines the energy obtainable from a battery, and is represented by their product (i.e., \(U = Q \times V\)). The EMF of a battery is affected by such factors as the state of charge and activities of substances involved in reactions, and it tends to undergo change during the discharging process. This instability is mainly due to the inner resistance of the battery, which suppresses the voltage downward as the current increases. In the circumstance where the battery voltage fluctuates, both the current and voltage must be integrated to estimate an exact value. For simplicity, however, it is a normal practice to use an average voltage to calculate energy. Energy density per unit volume/weight is calculated by taking only the active materials on the positive and negative electrodes into consideration, and the obtained value is interpreted as the theoretical power density. More specifically, the energy density of a battery is obtained by calculating the product of the average voltage difference between the positive and negative electrodes and the electric capacitance density.\(^{[14]}\)

**Figure 2**: Charge/discharge mechanism of lithium-ion battery: examples of electrochemical reactions taking place on the electrode

**Glossary**

* Quantity of electricity: A value obtained by integrating current with time. In the case of constant current discharge, this value coincides with the product of discharge time and discharge current. The capacity of a battery is represented by the quantity of electricity it can discharge until the battery voltage falls off to a predefined value (final voltage).
4 | Current Status of Lithium-ion Battery R&D

4-1 Superiority of Japan’s Lithium-ion Battery Technology

Japanese companies were among the first to commercialize the lithium-ion battery and their share of global production was 100% in FY 1998. Since then they have maintained the global lead in R&D. However, as shown in Table 1, the global share of the Japanese companies is shrinking rapidly, both in terms of production volume and shipping value.\(^{(6,17,18)}\) Japanese companies occupied all the top six positions in terms of global share in FY 2000, but the global share of Japanese companies dwindled down to approximately 60% in FY 2005. As of FY 2008, Japanese companies scarcely maintained their top position with a global share of 50%. Foreign companies have been proactively boosting their efforts for R&D of lithium-ion batteries and the increase of their global share well reflects their efforts.

Korean and Chinese companies have made especially significant R&D efforts to catch up with Japan since 2005. Samsung SDI Co. Ltd. and LG Chemical Ltd. (both Korean), and BYD Co. Ltd. (China) have received attention in their R&D of the secondary batteries for use in HVs and fuel-cell HVs (FCHVs). Among other notable companies, SAFT (France) is pushing forward with secondary battery development for use in EVs and HVs. A123 Systems Inc. (U.S.) has also started production and marketing of cost-effective, safety-oriented lithium batteries, aiming at in-vehicle applications for HVs.\(^{(19)}\)

Let us review the activities of two international conferences, both of which rank as the most representative for understanding the current trends in lithium-ion battery research, whereby the focus is placed on R&D of materials. These conferences are the “International Meeting on Lithium Batteries (IMLB 2006),” held in France in 2006, and the “Pacific Rim Meeting on Electrochemical and Solid-State Science (PriME 2008),” held in Hawaii in 2008 and co-hosted by the American Electrochemical Society and the Electrochemical Society of Japan. Figure 3 summarizes the country-by-country number of the papers presented in these conferences related to

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Note: Panasonic’s TOB (takeover bid) against Sanyo succeeded on December 11, 2009. The global share of lithium-ion batteries of these two companies combined amounted to 29% in fiscal 2008.

Prepared by STFC based on Reference\(^{(1,2,6,17)}\)
lithium-ion batteries. In IMLB 2006, the number of papers presented by Japanese researchers ranked either first or second in the areas of positive/negative electrodes and electrolytes. In PRiME 2008, the combined number of presentations from the U.S. and Japan on positive electrodes occupied approximately 65% of all presentations. Among other countries, Korea and China gave more presentations than European countries did.

4-2 R&D Projects on the Initiative of the Ministry of Economy, Trade and Industry of Japan

In the third period of the “Basic Program for Science and Technology”, the subject “High-performance electric power storage technology that overcomes differences in sources and uses” has been selected as one of the technological areas of strategic importance, and the technological development of the next-generation in-vehicle battery was placed among the priority areas by the Economic Growth Initiative laid down in 2006. Following these policies, the New Energy and Industrial Technology Development Organization (NEDO, an independent corporation under the jurisdiction of METI) started in FY 2007 a project (Li-ion and Advanced Battery Development) to develop a high-performance, low-cost secondary battery with a view to accelerating commercialization of new vehicles such as HVs, EVs, and FCHVs. The project is currently underway for R&D of a 3kWh-grade lithium-ion battery that has the following target specifications and is slated for completion by FY 2015: energy density of 100Wh/kg, power density of 2kW/kg, and a cost of $30,000/kWh. The project also includes long-term basic research to develop new types of batteries that step far ahead of the performance available from the lithium-ion battery.

In addition, a separate R&D project (Research & Development Initiative for Scientific Innovation of New Generation Batteries) started in FY 2009, and over its projected span of seven years it is to establish fundamental technology in view of realizing EV-oriented secondary batteries capable of providing driving performance comparable to that of vehicles powered by gasoline engines, whereby the elucidation of a fundamental reaction mechanism inside the battery is of importance. The fruits of the R&D project are expected to pave the way for the realization of innovative lithium-ion batteries with breakthrough enhancements in performance, safety, and reliability, which will enable a significant upgrade of driving performance for EVs and PHVs.

4-3 R&D projects on the Initiative of U.S. Government

Figure 5 shows the framework and content of the R&D of secondary batteries for vehicle use guided by the Department of Energy (DOE).

DOE has been pushing forward the development of secondary batteries for EVs by granting 50% of development overheads to the United States Advanced Battery Consortium (USABC) and encouraging battery manufacturers (A123 Systems Inc., Compact
Power Inc., EnerDel Inc. et al.) to participate in the project. (USABC is an organization established in 1991 by automobile manufacturers, centered on the “Big Three.”) In addition to the support program for USABC, DOE is undertaking R&D activities rigorously to develop lithium-ion batteries for vehicle uses (HV, PHV, and EV) through such programs as ATD (Applied Technology Development), ABR (Applied Battery Research), and BATT (Batteries for Advanced Transportation Technologies). The important elements of technical development for these projects (high-energy-density batteries for PHV and high-power batteries for HV) include the cost, characteristics, safety, and useful lifespan.

In the USABC program, the participants are undertaking R&D using individual high-potential materials for enhanced energy density. Compact Power Inc. uses a composite material consisting of layered oxide and manganese spinline oxide (positive electrode), A123 Systems Inc. uses a phosphoric-acid-based material (positive electrode), and EnerDel Inc. uses spinel structure titanium acid manufactured by dispersing nano-scale particles (negative electrode) and manganese-nickel acid lithium (LiMn1.5Ni0.5O4) (positive electrode).[26,27]

The R&D of battery cells being carried out in the ATD program and ABR program places emphasis on the development of a lithium-ion battery for PHV uses. The objectives of this R&D include the realization of energy density that allows a PHV to run 40 miles, and of a useful lifespan that allows up to 5,000 charge/discharge cycles. The Argonne National Laboratory plays a central role in carrying out these programs, while Brookhaven, Idaho, and Sandia National Laboratories participate. The objectives of these programs include the R&D of battery cell materials, calendar life (the period during which a battery can maintain a charged state continuously), charge/discharge cycle life, and resistance to overloading. The BATT program, on the other hand, places emphasis on fundamental research for the development of positive and negative electrode materials and electrolytes to be used in HV, PHV, and EV. The program is being carried out under the initiative of Lawrence Berkeley Laboratory.[28-30]

4-4 R&D projects in Europe, Korea, and China

In Europe, the R&D of lithium-ion batteries started in 1993, under the initiative of EU’s JOULE (Joint Opportunities for Unconventional or Long Team Energy Supply) project, whose main R&D subject has been the secondary batteries used in EVs. New projects have also been launched, under the financial support of the EU executive body and the European Commission, to develop a new lithium secondary battery. An example of this effort is the ALISTORE (Advanced Lithium Energy Storage Systems Based on the Use of Nano-powders and Nano-composite Electrodes/Electrolytes) project, which is now underway with the participation of 16 battery-related
research groups within the EU, and is slated to continue for 5 years starting in FY 2004.

In Korea, a large 5-year national project was launched in 2004, and this government-led project has been working on the development of an extra-large capacity lithium-ion battery and electric double-layer capacitor.

In China, a technology development project (called the “863 project”) has been underway since 1986, and R&D of a secondary battery for use in EVs and HVs was carried out from 2001 to 2005. From 2006, the development of a lithium-ion battery has been carried out, whereby FCHVs were added as a research objective in addition to HVs and EVs. The presence of Korea and China in sales shares of lithium-ion batteries has been growing as shown in Figure 3, and the proactive stance of these countries towards lithium-ion batteries seems to be a major driving factor for this growth.

5 Current R&D Status of Electrode and Electrolyte Materials and Future Direction

5-1 Electric Potential and Discharge Capacity Density of Electrode/electrolyte materials

The power and energy density of a lithium-ion battery largely depends on the electrode materials and electrolyte materials. Figure 6 summarizes the relationship between electric potential and charge-capacity density of electrode materials that have been studied.\[16,31-33\]

In small-capacity batteries, LiCoO\textsubscript{2} is mainly used as a positive electrode material, while for applications that require an especially high level of safety, lithium manganate (LiMn\textsubscript{2}O\textsubscript{4}) is selected, which has a spinel type crystal structure (an octahedral crystal structure consisting of metal elements A, B, and oxygen; composition formula: AB\textsubscript{2}O\textsubscript{4}). The majority of the new materials currently under R&D with a view to providing a superior positive electrode are oxides of transition metals (generic name designating the group of elements that occupy positions between the third...
and eleventh family in the periodic table). Generally, these positive electrode materials tend to provide lower discharge potential as the electric-capacity density increases.

At present, carbon-based materials are mainly used as negative electrode material. The advantages of carbon-based negative electrode materials include: long charge/discharge cycle life (even in the 100% depth of charge/discharge), the capability of attaining a high lithium-filling fraction, and a configurability of the electrode potential in the vicinity of a metallic lithium electrode. A variety of carbon-based negative electrode materials are under scrutiny in terms of electric capacity and electrode potential. A hard carbon (non-graphitizable carbon) material is a favored choice if energy density is the main objective, and a graphite-based material (graphitizable carbon) is more suited if high-power characteristics are the target. Although a variety of materials with capacity superior to carbon-based materials have been proposed, many of them need further improvement in respect to electrode potential and charge/discharge cycle life concerns.

Several options are available for the electrolyte: nonaqueous (organic and ionic liquids), gelled electrolyte, organic solid electrolyte, and inorganic solid electrolyte materials. Among these, mainstream solutions are those based on electrolytes. Major electrolyte systems currently used are prepared using mixtures of several compounds, such as the mixture of dimethyl carbonate and a cyclic carbonate (e.g., propylene carbonate, ethylene carbonate), and the mixture of lithium-4-phosphorous fluoride (LiPF$_4$) and a chain carbonate (e.g., diethyl carbonate or methyl carbonate).

5-2 Electrode and Electrolyte Materials for Higher Power and Larger Capacity

The power source to drive a vehicle requires a secondary battery with a breakthrough upgrade of power and capacity that is contained in a lightweight and compact package, which is essential in conjunction with the advancement of power electronics technology (electric motor and inverter). Currently available lithium-ion batteries already have such advantages as relatively high energy density and non-susceptibility to memory effect (a phenomenon whereby the repetition of charge/discharge cycles while the battery is not completely depleted has downward effects on the discharge voltage). Still, the currently available power level is not sufficient to drive an HV, and the production cost is still too high, mainly because of the electrode and electrolyte materials and the expenses involved in their production processes.

In order to yield a higher power density, enhancement of current magnitude is of importance, in conjunction with a boost of output voltage. For this, a reduction of inner resistance is needed. Figure 7 summarized the main factors that give rise to the lithium-ion battery’s inner resistance during a discharge process.$^{[15]}$ The inner resistance during discharge is broadly divided into two factors: resistance that accompanies lithium-ion transport, and the electron conduction resistance that occurs both inside the electrode and the electrode-collector interface region. The resistance due to lithium-ion transport is further divided into several factors: the resistance a lithium ion suffers while conducting in the electrolyte, the resistance while migrating within the positive and negative electrode, and the resistance while a lithium ion is diffusing in the active materials inside the positive and negative electrode. Contributions from these factors depend on the nature of the materials used in the positive/negative electrode.

The energy density of the currently used positive electrode materials (e.g., LiCoO$_2$) is, as a matter of course, lower than that of negative electrode materials. For energy density enhancement of positive electrode materials, a new material with a higher average electric potential than LiCoO$_2$ and a larger electric capacity must be explored.

5-3 Safety and Reliability of Electrode/Electrolyte Materials

The lithium-ion batteries currently available still have safety and reliability problems, such as the danger of excess heat generation and reduced lifespan, both typically caused by the reaction products and impurities that comes into existence during the oxidation reaction inside the battery. These problems can be ascribed to the generation of metallic lithium. In cases where LiCoO$_2$ is used as a positive electrode, metallic lithium is deposited on the surface of the negative electrode due to the abnormal charge/discharge process, and the very reactive metallic element can impair the safety of the system. When the battery falls into a deviated state, for example a state of over-charge, LiCoO$_2$ used in the positive
electrode can trigger an internal short circuit that generates excess heat and, if an organic-solvent-based electrolyte is used, a fire will eventually be caused. In cases where the system uses a graphite negative electrode, as its electric potential lies in the vicinity of the generation potential of metallic lithium, a rise in internal resistance for some reason can promote the formation of metallic lithium.

On the surface of the particles (electrode’s active material), the creation and annihilation of such a compound as Li$_2$CO$_3$ can take place through the repetition of the charge/discharge processes. On the positive/negative electrode surfaces, simple contact with the electrolyte can trigger an undesired or
unintentional oxidation/reduction reaction in the interface between them. For example, a trace amount of impurity (e.g., water) contained in the electrolyte can generate an undesired reaction product, and the thin, nano-scale layer thus produced can cover the active material on the surface of both electrodes, exerting an adverse effect of causing charge/discharge characteristics such as the rise of resistance in the interface, heat generation, lowering of power and capacity, and shortening of useful lifespan. In the case of a nickel oxide positive electrode, a thin high-resistance layer (SEI: Solid-Electrolyte Interface layer) is generated on the surface of the active material by the action of discharge potential and the layer has the adverse effect of reducing system safety. A thin layer of aluminum (Al) can form on the surface of the positive electrode’s collector. In many cases, the battery can be operated with the layer in place, but, as it may induce a corrosive reaction that produces an oxide layer (Al2O3, Al2(CO3)3, etc.) with a thickness around several tens of nanometers, finally reducing battery performance.

To avoid these undesirable effects, some systems use thermally stable LiMn2O4 as the positive electrode and lithium titanate (LTO: Li4Ti4O9) material as the negative electrode. The electric potential of LTO-based materials is high enough (1.6V) to hinder the formation of metallic lithium. The use of graphite can invite such problems as the loss of active materials and the elimination of the electron path through the long-term repetition of charge-discharge cycles due to the volume variation that accompanies the intercalation/desorption process of lithium ions. In contrast, LTO-based negative electrode materials do not undergo volume variation even when the intercalation/desorption takes place in charge/discharge processes, thus giving the battery a longer useful lifespan. Although the theoretical capacity of LTO-based materials is not very high (approximately 170Ah/kg), its merit from safety aspects is good enough to make it a candidate for the negative electrode of a secondary battery for vehicle use.

### 6 Challenges and Approaches toward Better Electrode/Electrolyte Materials

#### 6-1 Priority Electrode Materials for Future R&D

(1) Positive Electrode Materials

The use of a material with high electric potential and large discharge capacity can increase the power and energy density of the battery. Figure 8 shows the positive electrode materials on which the focus of future R&D should be placed, and their potential vs. discharge capacity-density relationships. The capacity density of the positive electrode materials, now in practical use, is around 200Ah/kg at the most, and this poses a major problem that stymies the enlargement of the lithium-ion battery’s capacity. Several materials have been gathering R&D momentum, including spinels, layered oxides, olivines, Li2MO3-based solid solutions (M represents a metallic element), and silicate based materials. With the exception of silicate-based materials, the discharge capacity density of these materials lies at or below 300Ah/kg.

A spinel material (composition formula: LiM2O4) is characterized by its stable crystal structure, providing superior safety. Although the power density of spinel materials can be enhanced by increasing electric potential, it has a growth ceiling mainly due to inherently low-capacity density (approximately 150Ah/kg). Variants of layered oxide materials (LiMO2) obtained by substituting a portion of cobalt (Co) in LiCoO2 structure with such metals as nickel (Ni) and manganese (Mn) have been extensively investigated. These include NiCo-, NiCoMn-, and NiMn-systems and several of them have come into practical use. The capacity density of these materials, however, is around a maximum of 270Ah/kg. A complete substitution of cobalt with nickel can provide a capacity density approximately 30% higher, but higher nickel ratios may raise safety concerns. Substitution with manganese increases output potential but lowers the capacity density.

Some of the olivine materials, generically represented by the composition formula LiMPO4, show a relatively high electric potential, but their capacity density lies at or below 200Ah/kg. These materials include lithium manganese phosphate (LiMnPPO4), lithium iron phosphate (LiFePO4), and...
lithium cobalt phosphate (LiCoPO₄). Research has been made to utilize the inexpensive and chemically stable LiFePO₄; according to some reports, the system enables acceleration of charging and discharging while maintaining the power density comparable to conventional lithium-ion batteries. The increased charge/discharge speed was obtained by using miniaturized LiFePO₄ particles (several to several tens of nanometers) as the positive electrode material, thus enhancing the speed of lithium ion diffusion and electron conduction towards the electrode surface and inside the matrix. Because of a superior level of safety, in spite of their less favorable capacity density, these olivine materials have been considered as suitable for large-scale secondary batteries for use in vehicles. However, olivine materials generally have a problem in that the capacity density falls at elevated temperatures. In comparison with the nickel oxide materials, the olivine materials generate a smaller amount of heat when they thermally decompose, making them more favorable in terms of safety. The fluoride olivine materials feature high capacity density, and hold promise in realizing a high energy density electrode, but they also face challenges such as a susceptibility to corrosion.

With a view towards attaining higher power, the positive electrode materials that feature high electric potential have been explored. Among these, olivine materials and oxide materials typically containing cobalt, manganese, and vanadium (V) are the development objects with immediate priority. These materials have discharge capacities around 150–160 Ah/kg; thus they are unsuited for realizing high energy density. The batteries that use these positive electrode materials, taking advantage of their high power density, aim at reducing the number of serially connected cells. Layer-structured vanadium pentoxide (V₂O₅) has been investigated as one of the materials with the potential of high energy density, but it has the drawback of losing reversibility because of a phase transition that takes place during the charging/discharging process.

With a view toward attaining higher capacity, on the other hand, silicate-based materials and sulfur (S)-based materials have been gaining attention. Lithium iron silicate (Li₂FeSiO₄), one of the silicate-based materials, features superior stability at elevated temperatures when used as an electrode material and, with two lithium atoms in a molecule, is expected to produce a substantial upgrade of discharge capacity density. Even the crystal structure of Li₂FeSiO₄, which had been unknown, was elucidated recently using the full power of analysis techniques including high-resolution X-ray powder diffraction. With the information on basic crystal structure in hand, expectations are high that the new research developments for the use of this material as a positive electrode will proceed. The sulfur-based materials

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**Figure 8**: Priority positive electrode materials for future research: electric potential vs. charge-capacity density

Reconstructed by SFTC based on Reference[37,38]
deserve attention for the fact that they have 300Ah/kg or more of capacity, but their output potential is generally low (2.7V) and R&D to substantially upgrade this value is needed.

In order to seek a breakthrough enhancement in power and capacity density, a significant increase in discharge density must be achieved through the R&D on positive electrodes. The R&D, therefore, should focus attention on materials that have large capacity density (the layered-oxide materials, fluoride-olivine materials, silicate-based materials, and sulfur-based materials), whereby emphasis should be placed on resolving the challenges each material presents. The following could offer useful clues for carrying out these tasks: doping of trace components, coating of electrode surface with a conducting material (enhancement of reactivity), and application of inorganic chemical synthesis processes that enable the control of material structure at the nano-scale level.

(2) Negative Electrode Materials

Large discharge capacity is the necessary criteria, just as is the case with the positive electrode, for a material to be used as the negative electrode. In addition, the use of a material with lower electric potential can increase the power and energy density of the battery. Figure 9 illustrates the relationships between electric potential and discharge-capacity density of the materials on which future R&D should place emphasis.\[37,38\]

Use of lithium-intercalated graphite-based materials for a negative electrode is a common practice at present, and they provide energy density in the range between 200 and 800Ah/kg. Graphite negative electrodes have an electric potential of approx. 0.1V. As the majority of currently available electrolyte solvents cannot stably exist with such a low potential as 0.1V, the solvent undergoes decomposition continuously during the course of lithium intercalation. A usual method to prevent this is to coat the graphite surface with a layer (SEI layer) that enables stable intercalation/desorption of lithium ions, but the layer also creates the problem of a resistance increase during the intercalation/desorption process. For graphite-based negative-electrode materials, much research has already been carried out on the enhancement of power and energy density, and now the capacity density of these materials seems to have reached their theoretical limits.\[14,16\]

The LTO-based negative electrode has a higher electric potential, allowing many electrolytes to stably exist. This enables intercalation/desorption of lithium ions without the help of a SEI layer, thus reducing the internal resistance. However, the average electric potential of lithium-ion batteries that use LTO-based materials in their negative electrode is relatively low (2.4V).\[42\]

From the point of view on enhancing capacity

![Figure 9: Priority negative electrode materials for future research: electric potential vs. charge-capacity density](image)
density, such a line of materials as sulfide-based, alloy-based, lithium-metal-based materials indicate another direction of research. Silicon-based materials have more than five-times the theoretical discharge capacity of graphite-based materials, and silicon-lithium intermetallic compounds have also gathered attention to be exploited in future. Silicon-based compounds (intermetallics with copper, tin, zinc, and titanium) have relatively high conductivity and their capacity density can be augmented up to 800 Ah/kg. Their charge/discharge characteristics are in the process of being improved.\[41\] Although lithium metal and its alloys potentially have very high energy density, they have never been used. In addition to safety concerns, lithium reactions that take place in the electrode-electrolyte interface during charge/discharge processes are beyond control, resulting in quite inadequate charge/discharge cycle characteristics. There are still many challenges to be overcome related to silicon-based materials, lithium-silicon intermetallics, and lithium-metal-based materials; thus the focus of R&D should continue to be placed on them. These problems include a degradation of charge/discharge cycle characteristics and a lowering of capacity due to volume variation during the charge/discharge process, an increase of resistance in the electrode-electrolyte interface, and the need for longer life and better safety.

6-2 Highly Important Electrolyte Materials for Future Research

The main objective expected in the R&D of electrolyte materials is to enhance safety. Insufficient safety has been one of the factors that have caused delay in the implementation of the lithium-ion batteries on board HVs and EVs. Use of a flammable solvent as an electrolyte ingredient is always accompanied by the danger of ignition in such cases as: intrusion of impurities in the production processes, occurrence of a short circuit in the electrode, and overcharging. If a light, solid hydride featuring rapid lithium-ion conduction becomes usable in practice, a lighter and safer lithium-ion battery than those that use conventional organic-solvent-based electrolytes can be realized. It also eliminates the need for anti-overcharging measures. The major challenge in doing so is to enhance the lithium-ion conduction in the electrolyte to the level possible in an electrolytic solution.

Figure 10 summarizes ion conduction characteristics of the electrolytes currently under R&D, including: organic/ionic nonaqueous electrolytes, gel electrolytes, and organic/inorganic solid electrolytes.\[37,38\] Up to the present, a carbonate-based organic solvent, mixed with the lithium ion salt of 6-phosphorous fluoride (LiPF6), has been the mainstream electrolyte, due to its high ionic conductivity over a wide temperature range (~30 to 80°C) and its stable chemical properties. Another requirement for an electrolyte is that it will not impair other members of the battery because of the voltage imposed during the charge/discharge process.

In order to make an electrolyte flame resistant, fire retardant compounds such as phosphorous (P) and boron (B) are often added to the electrolyte solution. These compounds have the effect of shielding oxygen when a fire breaks out, but they cannot guarantee sufficient safety.

In order to upgrade safety substantially, research has been carried out to replace liquid materials with solid counterparts. However, a solid electrolyte generally slows down the lithium-ion conduction, resulting in reduced battery performance. Other than liquid-based electrolytes, gel polymer electrolytes and organic/inorganic solid electrolytes have been the objectives of research. The gel polymer electrolyte consists of a polymer carrier with an electrolyte liquid included/re retained in it, and its polymer network has the effect of suppressing the dendritic crystal growth of lithium that often occurs on the surface of negative electrodes under the condition of high current density. The organic solid electrolytes have, with great difficulty, attained an ionic conduction level of 10^-5 S/cm at normal temperatures, but they still need a 1000-times enhancement to catch up to the level of organic-solvent-based electrolytes.

Inorganic materials, such as lithium nitride (Li3N), sulfur-containing glass, and sulfur-based amorphous electrolytes (Li2S-P2S5) have been investigated for use as a solid electrolyte. They generally show a 10^-3 S/cm range of ionic conduction at room temperature, and are reported to remain stable in combination with LiCoO2-based materials (positive electrode) and metallic-lithium-based materials (negative electrode).\[39,42\] A case of an all-solid lithium-ion battery development has been reported, in which the use of a conventional flammable solvent is eliminated by such method as mixing solid electrolyte materials and positive electrode materials. This all-solid battery is reported to have a power level of conventional
batteries, while eliminating such problems as ignition and liquid leaks. This example uses LiCoO₂ materials as the positive electrode and lithium-sulfide (Li₂S)-based materials as the electrolyte, and a grafting material (nano-scale LiCoO₂ particles are mixed with Li₂S) is applied in the electrode-electrolyte interface to enhance lithium ion migration between them.[44, 45]

Lithium boro-hydride is another example of a solid electrolyte material that has been a target of R&D. It is a solid hydride that consists of lithium (Li), boron (B) and hydrogen (H) and allows rapid lithium ion conduction at room temperature. Partial introduction of halogenated lithium, such as lithium iodide (LiI), into a solid hydride was found to have the effect of stabilizing the crystal structures that enable lithium ion conduction at room temperature.[46, 47]

When exploring a new electrolyte material, due attention should be paid to the findings on the ionic conduction mechanisms of dissimilar ionic electrolytes (e.g., H⁺, O²⁻) (they are used, for example, in solid-oxide fuel cells).[13] Nano-scale level control of the electrode-electrolyte interface structure will become necessary in the electrolyte research for all-solid batteries.

6-3 Technology for Basic Analysis and Evaluation

With the objective of attaining breakthrough high power and large capacity, as well as enhanced safety, it is essential to pursue a fundamental mechanism that helps develop battery characteristics, whereby a variety of analyses, evaluations, and simulation techniques are exploited. Among others, the following mechanisms need elucidation from the viewpoint of nano-scale structure: the charge-storage mechanism of electrochemical reactions that involves charge transfer through the ion diffusion and electron conduction inside the active materials of the electrode, and the mechanism that causes impairment and degradation of electrode/electrolyte materials.

Figure 11 schematically illustrates the methods used in basic analysis and evaluation of electrode/electrolyte materials. If the charge/discharge mechanism is governed only by the adsorption/desorption processes on the surface of a nano-crystal, fast charge transfer becomes feasible because of the exclusion of slow intra-solid charge diffusion processes. It is known that active nanocrystalline materials are capable of storing a large amount of lithium, in excess of stoichiometric prediction, owing to their huge specific surface, and enable rapid lithium storage only by the surface. Application of this lithium storage mechanism taking place inside a nano-crystal to the electrode material may raise the possibility of achieving a lithium-ion battery with innovative high power and large capacity.

The electric potential of a battery is related to the electron transfers during oxidation/reduction reactions that take place between different electronic structures and states of valency of the electrode material, and the output characteristics are mainly related to ion diffusion. Therefore, R&D based on microscopic information is becoming increasingly important. This includes a material exploration based on the elucidation of such factors as the changes of the electrode material’s crystal structure and electron conformation that takes place during charging/discharging, and electrochemical reactions that occur in the electrode-electrolyte interface. Many approaches have been tried along this line including: a lithium cluster formation reaction taking advantage of the nano-size void inside the electrode structure, and a method to utilize the wide oxidation/reduction range of transition metal elements through the use of nitrides. However, details of these factors and electrochemical reactions in electrode-electrolyte interfaces are not yet fully understood. In order to develop a battery that can retain its innovative characteristics, the fundamental mechanism must be explored more actively than ever before. Many measurement methods are currently employed for evaluation from an electrochemical standpoint. These include Alternating Chemical Impedance Spectrometry (ACIS), Soft X-ray Absorption Spectroscopy (XAS), X-ray Photoemission Spectroscopy (XPS), and Fourier-transform Infrared Spectrometry (FT-IR). These techniques will continue to provide useful tools for identifying the chemical species that are formed in the interface between an electrode and electrolyte.[41]

6-4 New Electrode-forming Technology

The secondary battery at present is manufactured by winding three sheet-formed planer elements (positive and negative electrodes, and a separator in between) into a roll, and containing the roll in a cylindrical or square shaped vessel made of laminated material, whereby the space between the sheets is filled with electrolyte. These electrodes are manufactured by forming a mixture of paste and powder into a sheet. New attempts have been made to transform the
High speed Li-ion conduction inside the electrolyte
High speed Li-ion conduction in the electrolyte-electrode interface
High speed Li-ion diffusion into the electrode matrix

High power, large capacity next generation secondary battery
Lithium-ion battery
New battery that operates on novel principles

Advanced measurements/analyses methods
ACIS, XAS, XPS, FT-IR, Light emission, etc.

Advanced observation methods
High resolution electron microscopy etc.

Advanced simulations
Molecular dynamics, Molecular orbital, First-principle methods etc.

Technology for basic analyses and evaluation
High electron conduction mechanism
High ionic conduction mechanism
Behavior of electrochemical reaction

Figure 10: Ionic conduction characteristics of electrolyte materials
Reconstructed by SFTC based on Reference[37,38]

Figure 11: Approaches for basic analyses and evaluation of electrodes/electrolytes
Prepared by the STFC

Nonaqueous electrolyte liquid (organic, ionic liquid)
(LiPF₆/EC-PC)+(PVDF-HFP)

Inorganic solid electrolytes
Li₂S
Li₂S-P₂S₅
3LiBH₄·LiI

Organic solid electrolyte
LiN(CF₃SO₂)₂(CH₂CH₂O)₈

Gel electrolyte
EC: Ethylene carbonate
PC: Propylene carbonate
DMC: Dimethyl carbonate
PVDF: Polyvinylidene fluoride
HFP: Hexafluoropropylene
EMI: Ethyl-methyl imidazolium

Nonaqueous electrolyte liquid
(LiPF₆/EC-PC)+(PVDF-HFP)

LiBF₄/EMIBF₄
LiPF₆/EC-DMC

LiBF₄/EMIBF₄
LiPF₆/EC-DMC

10⁻⁵
10⁻⁴
10⁻³
10⁻²
10⁻¹

-40 -20 0 20 40 60
Temperature (°C)

Ionic conductivity (S/cm)

-40 -20 0 20 40 60
Temperature (°C)
two-dimensional battery into a three-dimensional structure. The new approach forms, through the use of a semiconductor microfabrication technique, a countless number of minute protrusions (height: several tens of micrometers) on a silicon substrate, and three battery elements (positive electrode, negative electrode, and solid electrolyte) are formed sterically upon the surface of each protrusion. This structure is effective for enhancing energy density because it helps increase discharge capacity and the specific surface of electrode materials per unit area. Another feature of this structure is that it reduces inter-electrode distance between the positive and negative electrodes. Narrow inter-electrode distance is expected to help enhance ionic diffusion, leading to a higher power density. An example along this line is a joint research project conducted by Physical Sciences Inc. and MIT, which made a prototype of a lithium-ion battery that has numerous tiny cylindrical electrodes. According to the project report, performance reached twice that of the conventional parallel-structure lithium-ion battery (energy density ≥ 100Wh/kg, power density 1kW/kg) [48,49]. At present, tiny chip-type batteries (typical dimension: several cm) are under development for use as a source for small devices. Application of the 3D structure to medium- and large-size batteries would become possible upon the development of a high ionic-conduction solid electrolyte and a successful scale-up of the production process.

8 Proposal for Advancement of Generic/Fundamental Research

In the present situation, where a rapid transition into a low-carbon society is of importance, an acceleration of approaches toward the realization of a next-generation automobile society is urgently needed. The high-power, large-capacity lithium-ion battery is one of the technologies that are gathering special attention. R&D of materials used in electrodes and electrolytes concerns generic technology that will help pave the way to the batteries with breakthrough performance. Government-affiliated organizations should indicate approaches toward future technology from the standpoint of a long-term strategy. In particular, there is much expected of the technology development of the materials used in all-solid lithium-ion batteries, but high risks and challenges are also involved. The research efforts in this area should be funded by public investment.

Publicly-funded research projects should be run based on a grand design that is formulated by taking the overall aspects of development processes into consideration, from the incubation of generic/fundamental technology to practical realization and wide dissemination, wherein the special focus should be placed on assisting generic/fundamental R&D of the electrode and electrolyte materials used in all-solid lithium-ion batteries, from which the emergence of breakthrough properties is strongly expected. To facilitate such R&D oriented toward generic/fundamental technology, projects need a well-organized, interdisciplinary framework.

7 Research and Development of an All-solid Lithium-ion Battery

For an EV to run a distance of 300 km on single charge, assuming a mileage of 6 km per 1kWh, the battery must have an energy capacity of 50kWh. The batteries with more than 500Wh/kg of energy density generally weigh as much as 100 kg. Thus, the use of lithium-ion battery technology is much desired to realize a large energy density (≥ 500Wh/kg) battery because of weight considerations. However, it is generally accepted that the currently used structure of lithium-ion batteries, even with some sophistication in the future, has an energy density ceiling of about 250 Wh/kg [37]. Several types of innovative secondary batteries have been proposed that can theoretically have an energy density larger than 500Wh/kg. These include: a metal-air battery, an all-solid lithium-ion battery, and a multivalent cation battery. Among all these types, the lithium-sulfur-based all-solid battery is, from feasibility considerations, the most promising candidate for the next-generation secondary battery. However, several challenges remain to be solved by future R&D such as: the low electron conductivity of sulfur and its tendency to dissolve into the electrolyte. According to the currently accepted view, an inorganic all-solid lithium-battery holds the possibility of achieving a breakthrough upgrade of energy density through the use of sulfur-based material as the positive electrode and metallic lithium as the negative electrode. This configuration is also expected to eliminate short circuits caused by a leaking electrolyte solution; thus it has the potential to drastically enhance safety.
that involves industry-government-academia collaboration. An interdisciplinary organization consisting of dissimilar fields of activities has the advantage of producing innovative technology in its boundary areas in a relatively short time, and allows a quick verification of its validity as well. Close, bi-directional communication between universities and public research organizations (generic/fundamental research information) and automobile and battery manufacturers (technical needs) will accelerate the process of transforming research results into practical use.

Research of a very high level on electrode and electrolyte materials, from the nano scale to the bulk level, are being pushed forward in each of the advanced countries, but Japan should maintain its leading role, as it has been up to now, among these countries. Success in the development of technology for a high-power, large-capacity all-solid lithium-ion battery with superior cost-effectiveness, ahead of all other countries, will have widespread repercussions not only in the automobile industry, but also in the wide spectrum of industries that aim at energy-saving and lowering environmental burden. Up to the present, Japan has had the global lead in the lithium-ion technology. However, the government agencies and private enterprises in the U.S., European countries, Korea, and China, among others, have been boosting their research efforts in recent years, aiming at greater sophistication of the in-vehicle secondary batteries, which poses a threat to Japan’s retaining of its advantages in the future. Lithium-ion technology for vehicles is the research area in which Japan should maintain a technical edge over other countries into the future and continue to secure its global leadership position.

9 Conclusion

Although not discussed in this article in depth, important aspects of battery research include the environmental friendliness of the materials used and concerns about stable resource supply. Full-fledged introduction of EVs into the market is expected to start around 2012, and widespread use of them requires many preparations, such as the implementation of infrastructure for charging/replacing batteries, further enhancement of performance and safety of the secondary batteries. In addition to these preparations, concern is deepening around the stable supply of battery materials, especially the metallic lithium, because the lithium-ion battery is expected to be the mainstream driving source for these EVs. Lithium deposits that can be mined efficiently and in an economically feasible manner are concentrated in certain regions. At present, Chile, Australia, China, Russia, and Argentina are the major lithium-producing countries. Although about 50% of the world’s total lithium deposits are estimated to lie in Bolivia, they are largely undeveloped. As lithium is a recyclable material, the fear of resource depletion is not serious if its usage in confined to portable electronics devices. However, the dissemination of EVs will trigger a precipitous increase in lithium demand, far surpassing the present trend of gradual production increase. According to a tentative estimation, producing EVs to replace 10% of vehicles owned in advanced countries (about 60 million units) will require about 0.42 million tons of lithium posing the fear of fierce battles for resources. Thus, the building of production infrastructure and a market for lithium is needed, and they must be planned from a long-term standpoint.

Other aspects from which a breakthrough is urgently desired for the widespread use in the future include the drastic cost reduction in every step of battery production, from synthesis of materials to the final product. The price tag for an in-vehicle lithium battery is more than twice as high as that of a nickel-hydrogen secondary battery, amounting to approximately ¥200,000 per one kWh output. Cost reduction is a must in addition to further efforts toward higher power and larger capacity. Currently, LiCoO₂ (a positive electrode material), used in lithium-ion batteries, has an especially high in price. The cost largely depends on the material and structure of the primary members of the battery, such as the positive and negative electrode and the separator. In order to attain cost reduction, efforts by material manufactures are also essential; they must keep an eye on the demand trends of in-vehicle lithium-ion batteries and plan investment in mass production facilities accordingly.
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Profile

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