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Trends in Research and Development on Plastics of Plant Origin — From the Perspective of Nanocomposite Polylactic Acid for Automobile Use —

Hiroshi Kawamoto
Nanotechnology and Materials Unit

1 Introduction

The age of mass production and mass consumption is ending and the conservation, reuse, and recycling of resources have become an important issue for the creation of a regenerating and recycling-oriented society. To solve the problems regarding energy consumption and global warming, the recycling of resources, and environmental burdens, it is necessary for business enterprises to take specific measures by setting up targets in various fields of business activities including product design and development, procurement and production, sales, and recycling of resources. In the automobile industry, since further global motorization is expected to proceed, making proactive efforts to reduce environmental burdens in the total life cycle of automobiles including the development, production, use, disposal, and recycling is strongly required. It is an absolute must to promote the reduction of the global carbon dioxide emission and establish a sustainable society based on earth-friendly technologies aiming at a regenerating and recycling-oriented society[1].

In the "Third Science and Technology Basic Plan," the development of innovative materials and components, which are responsible for the next-generation contributing to the maintenance and enhancement of industrial competitiveness, is listed as a theme in the nanobiotechnology field aiming at solving issues on scarce and deficit resources, measures for handling harmful substances, and improvement and conservation of the environment. The purpose of these themes is to realize a sustainable recycling-oriented society maintaining a balance between environment and economy[2]. The "Biomass-Nippon Strategy" aims at promoting the effective utilization of plastics of plant origin as a technology for converting biomass to products such as plastics[3], and recommends the utilization of biomass as a substitute for products of fossil origin as one of the measures for reducing the emission of carbon dioxide, which is a greenhouse effect gas, among those that have been investigated.

In this report, the trends in the research and development of polylactic acid, which is one of the plastics of plant origin presently attracting widespread attention as a carbon neutral material*1, are described particularly from the perspective of nanocomposite material for automobile use. Properties of polylactic acid as an industrial material are explained through the present status of its applications to interior materials for automobiles. Furthermore, the effect on the worldwide sugar yield from biomass when all the plastics for the automobile application are replaced with polylactic acid is considered.

2 Plastics of plant origin as carbon neutral materials

Plastics of biomass origin are produced using biomass as the raw material, and broadly classified into three categories: those of microbe origin that are polymerized by microorganisms in their body, those of natural product origin for which biomass itself is used as polymers, and those that are chemically synthesized using

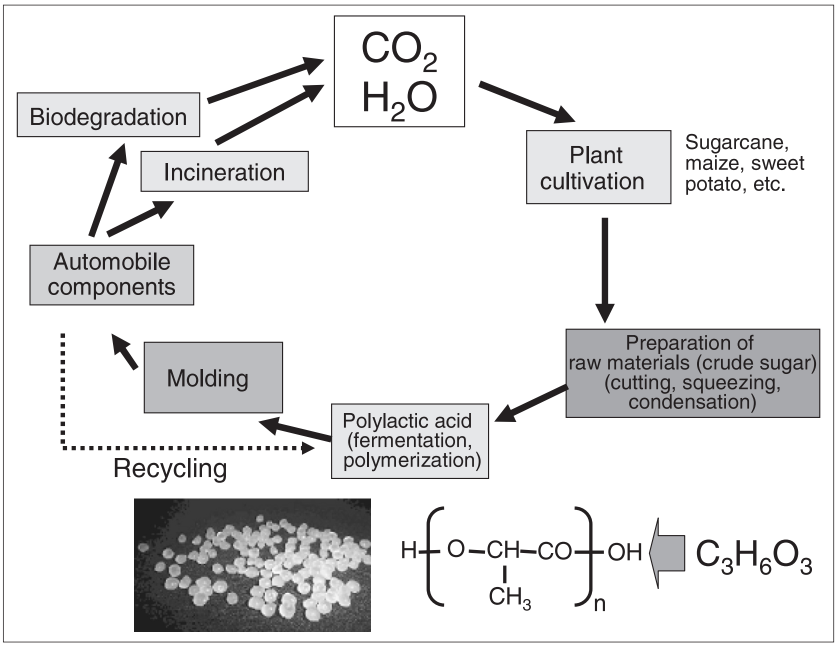


Figure 1 : Carbon cycle in the use of plastics of plant origin (polylactic acid)
Prepared by the STFC based on References[7, 8] with partial modification and addition

monomers of biomass[4-6]. Among these plastics, the plastics of plant origin refer to those that use plants as the raw materials, and account for the major part of plastics of biomass origin excluding those that use fossil resources as the raw materials. The carbon dioxide generated by the decomposition of plastics of plant origin originally existed in the atmosphere, and does not contribute to the increase of carbon dioxide in the total life cycle excluding the energy consumption during the production stage. Therefore, the plastic of plant origin can be defined as a carbon neutral and renewable material.

Polylactic plastic is a material of plant origin that has recently been attracting attention as a promising material for buildings, enclosures for home electric appliances, and interior and exterior materials for automobiles. Figure 1 shows the carbon cycle of a plastic of plant origin taking polylactic acid as an example[7, 8]. Polylactic acid is produced mainly from the sugar obtained from maize, sugarcane, sweet potato, etc., by polymerizing lactic acid which has a structure consisting of three carbon atoms. These carbons are originally derived from the carbon dioxide in the atmosphere so that the absolute amount of carbon in the atmosphere is not affected whether it is biodegraded or incinerated; therefore, polylactic acid can be said to be a carbon neutral material. Furthermore, if the components made of polylactic acid are efficiently recovered from used cars at low cost, it becomes possible to regenerate high-purity lactic acid monomers from these recovered components. The recycling of polylactic acid products enables not only the effective utilization of limited plant resources, but also the reduction of carbon dioxide generated in the incineration of components.

3 Present status of research and development on plastics of plant origin

3-1 Reasons why polylactic acid attracts attention

Figure 2 schematically shows the advantages of polylactic acid among other plastics of plant origin from the perspectives of material supply and industrial merits. Lactic acid is produced by fermenting sugar, which is generated by decomposing starch obtained from maize, sugarcane, sweet potato, etc., using enzymes. Then the lactic acid is polymerized to produce polylactic acid, and various types of plastic products are manufactured through the reforming and fabrication processes. These plastics are biodegradable since they are decomposed by bacteria in several months and return to the soil. Polylactic acid can be

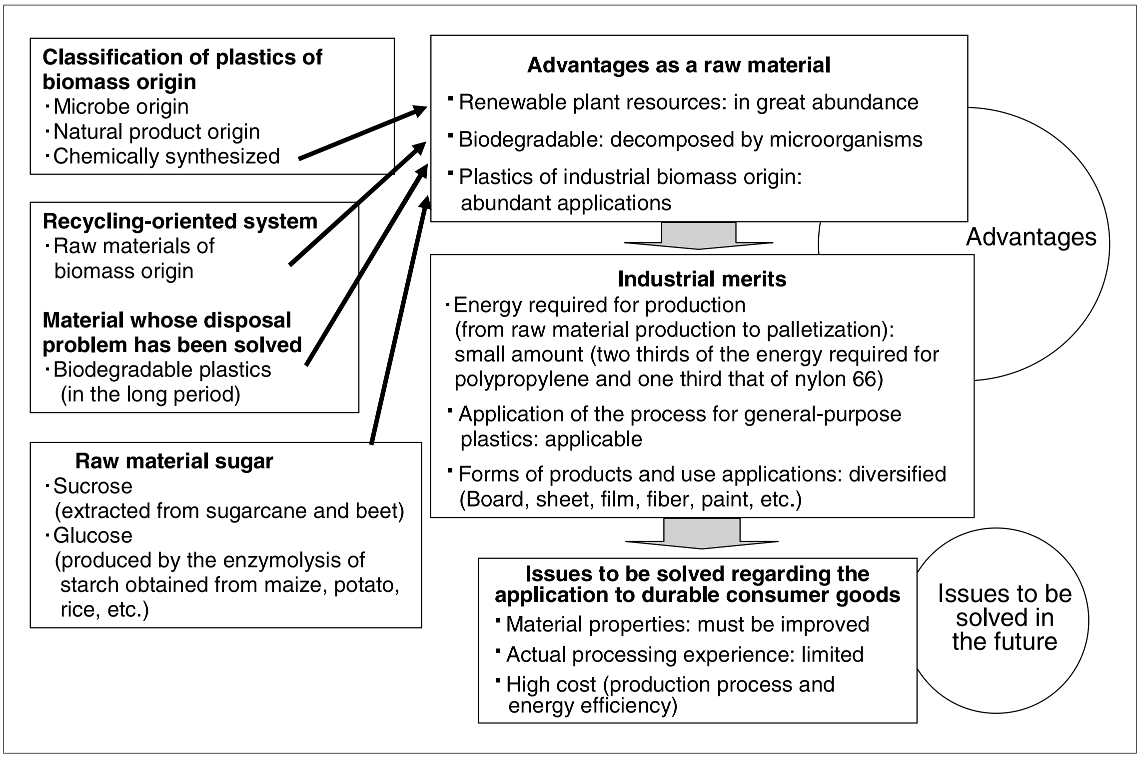


Figure 2 : Advantages of polylactic acid as an industrial material and issues to be solved

Prepared by the STFC

synthesized not only from starch, but also from abundant renewable biomass resources such as disposed paper, leftover food, residual lumber, lumber from thinning, straw, and chaff, and it is one of the most popular plastics of plant origin used as industrial materials. Polylactic acid has many advantages as follows: the energy required for the production of material and palletizing is low compared with that required for the plastics of petroleum origin; the production process for the plastics of petroleum origin can be applied; and forms of products and use applications are diversified. However, in order to apply polylactic acid to durable consumer goods, further improvement of the material properties including heat resistance and mechanical strength is indispensable. Other issues to be solved in the future include insufficient experience in the production process and high cost compared with the plastics of petroleum origin[6, 9, 10].

3-2 Application of polylactic acid to interior components for automobiles

The advantage to the recycling-oriented society brought about by the application of components made of polylactic acid is considered to be enormous. In particular, since the automobile industry is going to apply such components on a full scale, components developed in the automobile industry are expected to broadly expand into other industries. Figure 3 shows a spare tire cover made of kenaf-fiber-reinforced polylactic acid and a floor mat made of polylactic acid fiber, both of which have already been used for Japanese commercial automobiles[7, 11]. Kenaf is a fiber-rich annual grass that grows rapidly in the temperate and tropical zones. It absorbs a lot of carbon dioxide and abundantly contains useful cellulose and long, strong fibers are obtained from the stems. The spare tire cover shown in the chart is made of kenaf-fiber-reinforced polylactic acid. The combination of polylactic acid and kenaf fiber prevents the deterioration in elasticity modulus at high ambient temperature and improves impact resistance. Polylactic acid is hydrolysable and it has been found that the degree of hydrolysis is affected by the remaining lactide, which is a dimmer of lactic acid monomers, in the polylactic acid. By drastically reducing the content of the remaining lactide, the resistance of the spare tire covers to humidity, which is one of the important environmental service conditions for automobiles, is secured for a long period[7].

Spare tire cover made of kenaf-fiber-reinforced polylactic acid

Floor carpet made of polylactic acid fiber

Figure 3 : Components made of polylactic acid used for Japanese commercial automobiles

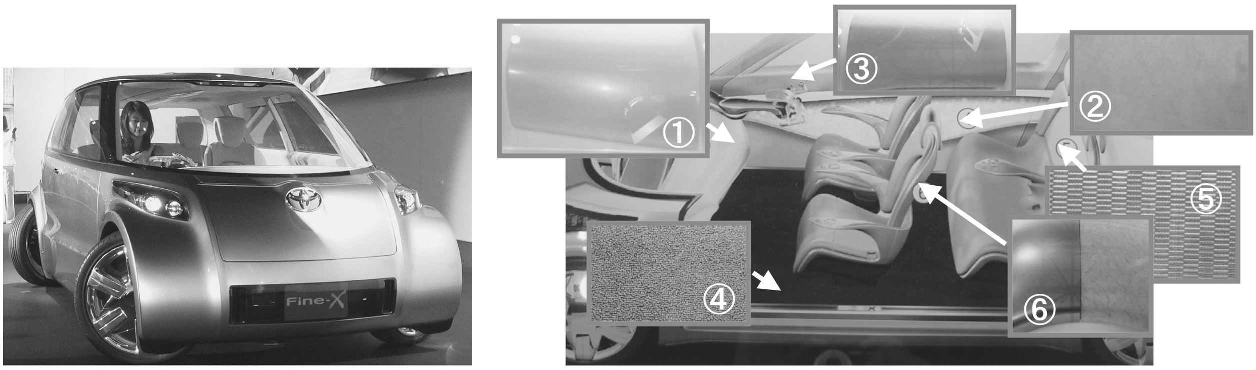
Prepared by the STFC based on References[7, 11] with modification

The life cycle assessment of automobile components made of polylactic acid shows that the emission of carbon dioxide is reduced by about as much as 85% compared with the components made of polypropylene which is a plastic of petroleum origin[11, 12]. Therefore, the adoption of components made of polylactic acid not only significantly reduces the energy consumption in the production process, but also reduces the emission of carbon dioxide because carbon, a component of carbon dioxide, circulates in the life cycle of the products, which means that polylactic acid is an earth-friendly material.

The components made of polylactic acid shown in Figure 3 may cover only a small portion of the total components used for automobiles, but the application is expected to be expanded into other components and vehicle types so that a recycling-oriented society is realized in the future. Material properties will be improved in the course of research and development searching for the mechanisms of the development on the properties of polylactic acid. For example, research and development to prepare all the interior components for automobiles using only materials of plant origin is under progress. Figure 4 shows the interior components of plant origin for a concept car[13]. These components are made mainly of polylactic acid using only plants as the raw materials (plant content: 100%), and kenaf-fiber-reinforced polylactic acid is used for some of the components as in the case of Figure 3.

3-3 Improvement of material properties of polylactic acid by kenaf-fiber reinforcement

To use polylactic acid as a material for the interior components of automobiles, high heat resistance must be achieved and impact resistance, which is the most important mechanical property, must be comparable to or higher than those of polypropylene. However, the impact resistance of simple polylactic acid is as low as 50% less than that of polypropylene. Figure 5 shows an example of a component with improved shock resistance that has been developed aiming at significantly high impact resistance and 100% plant content[12]. The material used was prepared by mixing kenaf fiber and polylactic acid fiber in a weight ratio of 7:3 and heating the mixture so that the polylactic acid fiber was melted. After that, the mixture was pressed at room temperature to form the component. The shock resistance of the composite polylactic acid reinforced with kenaf fiber was three times higher than that of polypropylene. Figure 5 shows the surface condition and cross-sectional structure of kenaf-fiber-reinforced polylactic acid. It seems possible to improve the mechanical properties other than impact resistance by densifying the matrix resin of polylactic acid, which bonds the kenaf fibers, with an improved filling method.



Component	Material	Material constitution and characteristics
1. Upper part of instrument panel, package tray	Polylactic acid	Transparent material
2. Inner lining (ceiling, pillar, trim), seat	Extra-fine polylactic acid fiber	Suede-touch skin material
3. Door trim ornament	Hemp	Japanese-paper-like skin material
4. Floor	Polylactic acid fiber	Carpet material
5. Seat back	Polylactic acid fiber	Meshed seat back material
6. Seat back board	Kenaf-fiber-reinforced polylactic acid	Board material

Figure 4 : Future interior components made of plant-origin materials equipped on a concept car

Prepared by the STFC based on Reference[13] with modification

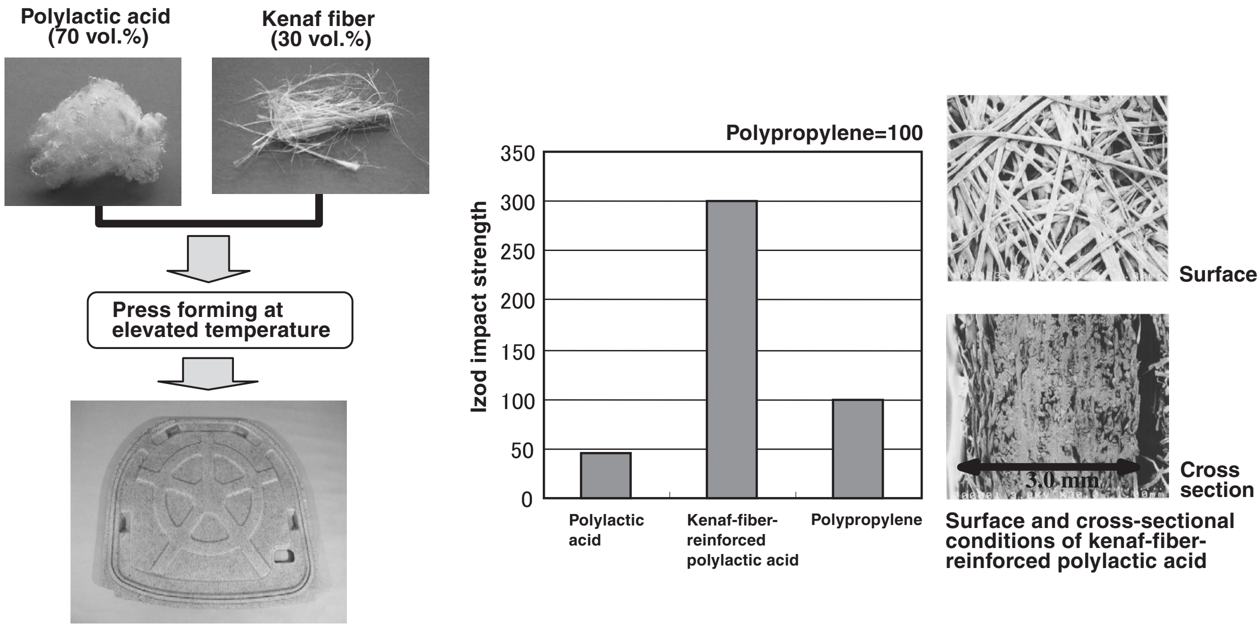


Figure 5 : Production process for a component (spare tire cover) made of kenaf-fiber-reinforced polylactic acid and Izod impact strength of the material

Izod impact strength: a rectangular specimen with a notch at the center is fixed and a hammer is used on the notched side to break the specimen. Izod impact strength is calculated from the energy required for the fracture.

Prepared by the STFC based on Reference[12] with modification

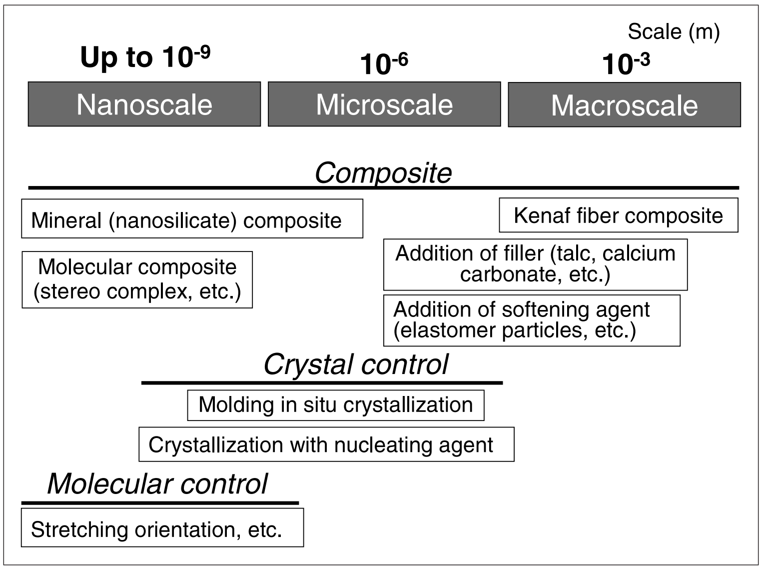


Figure 6 : Major methods to improve mechanical properties of polylactic acid

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4 Themes of research and development for the improvement of material properties of polylactic acid

4-1 Improving material properties by a nanoscale composite process

Efforts are being made to improve the mechanical properties of polylactic acid from the perspectives of composite process, crystal control, and molecular control of the material structure on a nanoscale, microscale, and macroscale. Figure 6 shows the specific methods on each of these scales[12]. The following is an example of research and development for material improvement using a composite process on a nanoscale. As the crystallinity increases, thermal distortion temperature rises so that crystallization proceeds during the injection process, which results in improved heat resistance[14]. In the past, the crystallization speed of polylactic acid was so low that it was necessary to obtain easily crystallizing polylactic acid in order to improve the heat resistance. To improve the heat resistance from the perspective of the nanoscale composite process, it is important to study methods to form composite with nano constituents of inorganic clay so that crystallization is promoted.

It is also extensively being studied to uniformly disperse nanoscale inorganic additives as filler among the organic molecules. In the past nanocomposite synthesis process, inorganic materials (such as montmorillonite, which is a type of clay, and synthesized mica) that have a microstructure of laminated plate crystals have been used as the filler. These inorganic materials are added to the organic monomers after being modified by the ion exchange process using organic cation compounds, and then the monomers are polymerized with the filler being uniformly dispersed[15]. Figure 7 shows the crystal structure of sodium montmorillonite, which is natural mineral clay and a type of silicate having a nanoscale laminar silicate structure[15, 16].

Figure 8 schematically shows an example of the production process for preparing nanocomposite of polylactic acid using sodium montmorillonite[16]. In this process, nanoscale sized clay is dispersed in the polymer so that the interaction between the constituents is increased and the softening of polylactic acid under a high temperature environment is suppressed thereby improving the heat resistance of the composite. Sodium montmorillonite is used as the clay and organic ammonium salts are used for the organizing process. Clay is added before polylactic acid is polymerized from the lactide. The layer interfaces of the clay being added are organized to increase the affinity with the monomer. As a result, the lactide penetrates between the clay layers causing ring-opening polymerization, and the laminated structure of

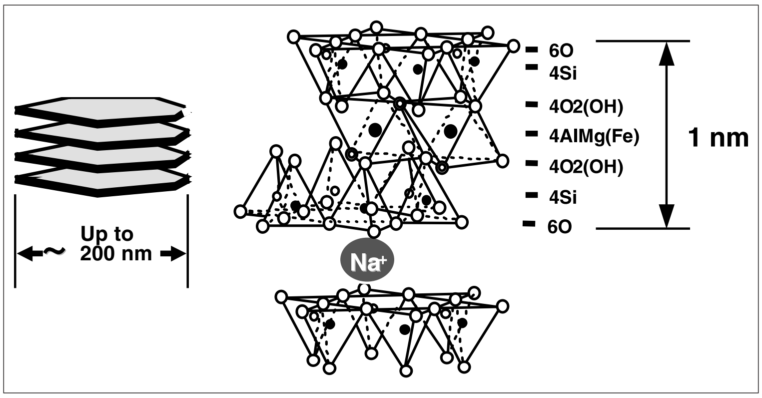

Figure 7 : Crystal structure of sodium montmorillonite

Prepared by the STFC based on References[15, 16] with modification

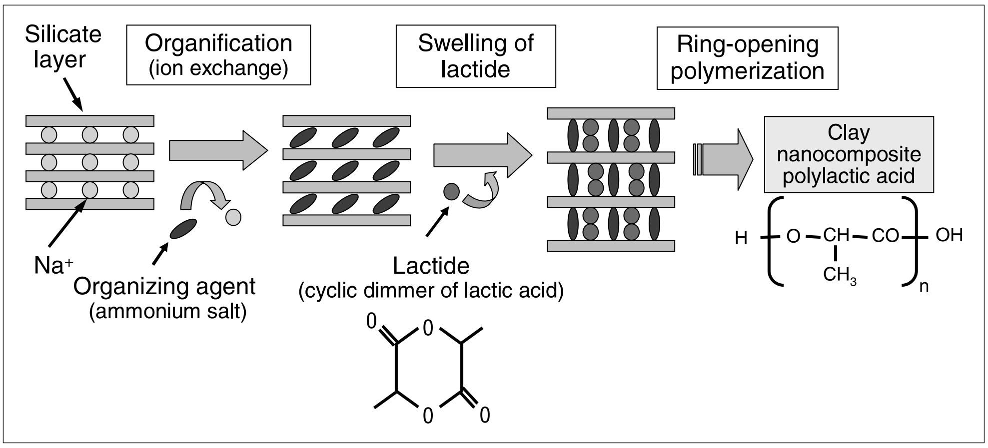

Figure 8 : Schematic diagram for synthesis process of clay nanocomposite polylactic acid

Prepared by the STFC based on Reference[16] with modification

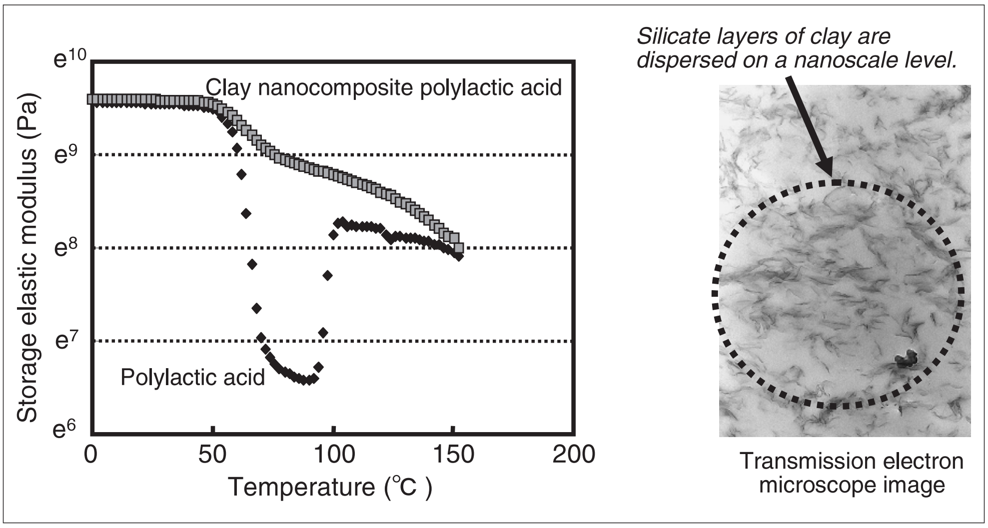

Figure 9 : Viscoelasticity and transmission electron microscope image of clay nanocomposite polylactic acid

Storage elastic modulus: refers to the part of the mechanical properties of a polymer that has elasticity combined with viscosity corresponding to elasticity. The temperature dependency of this property is obtained by applying dynamic load to the material and measuring the transmitted force.

Prepared by the STFC based on References[8, 14, 16] with modification

the clay is separated layer by layer, which results in nanoscale dispersion in the polylactic acid[14, 17].

Figure 9 shows the viscoelasticity characteristics and a transmission electron microscope image of clay nanocomposite polylactic acid formed by high-temperature injection molding[8, 14, 16]. To prepare the sample, molding conditions such as the mold temperature and cooling time were optimized to cause crystallization in the mold, and the elasticity modulus of polylactic acid at high temperature was drastically increased with the additional effect of reinforcement by the nanodispersed silicate layers. The clay nanocomposite polylactic acid enables crystallization at practical temperatures so that the heat resistance is significantly improved. The transmission electron microscope image of the clay nanocomposite polylactic acid indicates that the silicate layers of clay are dispersed on a nanoscale in the polylactic acid. This shows that the silicate layers are separated layer by layer and uniformly dispersed on a nanoscale, but not that the ring-opening polymerization occurs predominantly outside the layers while the layer structure is maintained[14, 17].

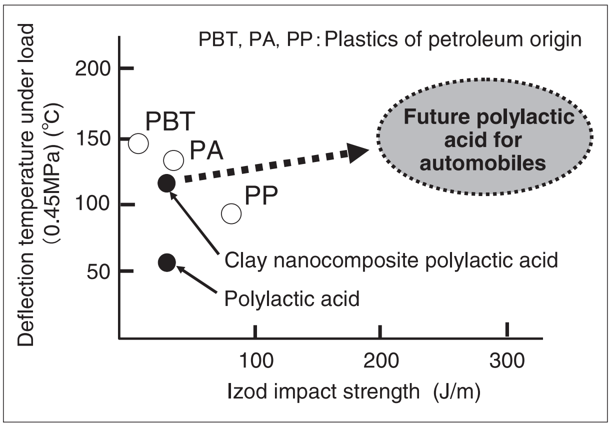

Figure 10 : Relationship between the deflection temperature under load and shock strength of clay nanocomposite polylactic acid

Deflection temperature under load: the temperature at which deflection reaches the specified value (0.32 mm) when the temperature of the specimen immersed in a liquid heating medium is raised under a bending stress (0.45MPa).

Prepared by the STFC

4-2 Nanocomposite technologies to further improve mechanical properties

Figure 10 shows the relationship between the deflection temperature under load and the impact strength of clay nanocomposite polylactic acid by high-temperature injection molding in comparison with that of the plastics of petroleum origin. At present, simple clay nanocomposite polylactic acid can improve the deflection temperature under load but it cannot improve shock strength, and composite processing with plant fiber such as kenaf is indispensable when applying to structural materials.

Figure 11 shows the future orientation for material design and strength assessment of polylactic acid toward significant improvement in mechanical properties. To realize the full-scale use of kenaf-fiber-reinforced polylactic acid as a structural material, it is necessary to obtain the optimum composite effect by statistical treatment considering the variation of the strength of kenaf fiber[18]. It is also necessary to understand the mechanical properties under various conditions of the service environment in which automobiles are operated. It has been shown that the shock strength of kenaf-fiber-reinforced polylactic acid is three times higher than that of polypropylene, and further expansion into the applications for structural materials is expected when the material structures on the microscale and

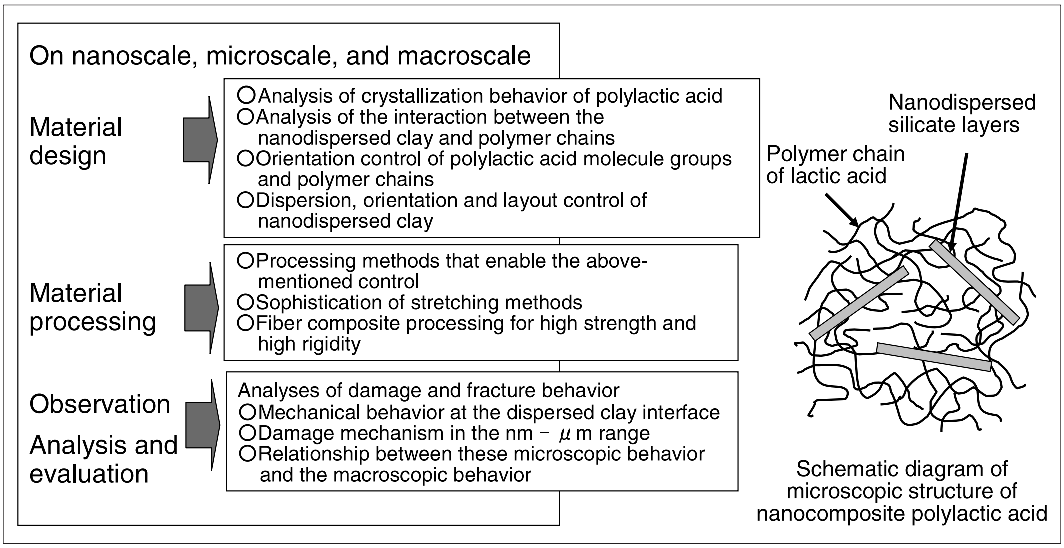

Figure 11 : Orientation for material design and strength evaluation of polylactic acid toward significant improvement in mechanical properties

Prepared by the STFC

macroscale are further controlled. Kenaf bast contains much cellulose and long, and strong fibers are obtained from the bast. However, to use kenaf as an industrial material, it is necessary to secure the quality and a stable supply[19].

For polylactic acid to be used as an injection molding material, it is further required to improve the crystallization speed, moldability, heat resistance, and shock resistance. Themes for material design and material processing from the nanoscale through microscale to macroscale include: analysis of crystallization behavior of polylactic acid, analysis of the interaction between the nanodispersed silicate layers and polylactic acid molecular chains, orientation control of polylactic acid molecular chains, dispersion, orientation, and layout control of nanodispersed silicate layers. Regarding the observation and analysis for strength design and evaluation, it is indispensable to analyze the damage and fracture process by understanding the mechanical behavior at the interface between the nanodispersed silicate layer and base material, damage evolution in the nanoscale through macroscale range, and the relationship with macroscopic mechanical behavior[20, 21].

In the composite technology on a nanoscale for inorganic nanomaterials and organic polymers, the viscosity of organic polymers drastically increases as the filler material is increased in the organizing treatment, and there is a limit to the improvement of physical properties. Therefore, in order to drastically improve the material properties, research and development of synthesis through material design at the atomic and molecular level is necessary. One of such synthetic methods is a process in which metal alcoxide is used as the raw material for the inorganic component. To uniformly disperse the filler finely in the organic polymer on a nanoscale, an electroviscous fluid is used as the solvent in the metal alcoxide method. Here, an electroviscous fluid refers to a fluid whose rheological characteristics vary in an electric field. Research and development related to the nanocomposite technology that applies an electroviscous fluid for polyamide plastics has been conducted[22-24]. It may be possible to further improve the material properties of polylactic acid by applying such conventional technologies for the plastics of petroleum origin.

5 Future themes other than material properties

5-1 Development of low-cost production process

One of the major causes that disturbs the application and diffusion of biomass plastics is its high cost. When polylactic acid is used as the raw material for plastics, the cost is two to five times higher than that of plastics of petroleum origin[25]. While the present price of plastics of petroleum origin is about 80 to 100 yen/kg, the "Biomass-Nippon Strategy" sets the target price of plastics of biomass origin to about 200 yen/kg in 2010, which is about twice as high as that of plastics of petroleum origin[3, 6, 28].

Figure 12 shows the production process of polylactic acid and the approximate cost ratio

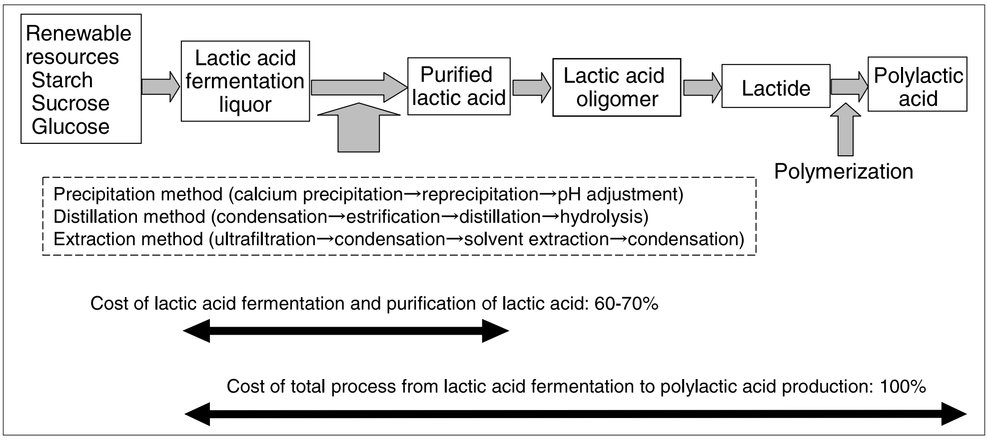

Figure 12 : Production process of polylactic acid and approximate cost ratio of purified lactic acid

Prepared by summarizing the contents of References[4, 6, 10, 26, 27]. References[6, 10] calculate the cost on a production scale of 37 thousand tons of polylactic acid using old rice as the raw material.

of purified lactic acid in the total cost[4, 6, 10, 26, 27]. In the production of polylactic acid, the cost required for purified lactic acid is the largest, which accounts for about 70% of the total cost. Therefore, it is indispensable to reduce the production cost of plastics made from polylactic acid to a level comparable to that of plastics of petroleum origin (one half to one fifth of the present level), by significantly reducing the cost of the purified lactic acid used for polymerization. To obtain high purity lactic acid from fermented liquor, precipitation, distillation, or the solvent extraction method is used, and all of these processes consume much energy. Therefore, it is essential to develop a technology that uses energy more efficiently[27].

In order to reduce the cost of lactic acid, technologies utilizing enzymes or lactic acid producing bacteria that reduce or eliminate processes causing high cost such as neutralization, distillation, and condensation in the purification process must be developed. Innovative processing that applies microorganisms or biotechnology is expected to contribute to the enhancement of price competitiveness for plastics of plant origin. It is also possible to reduce the cost by recycling the lactic acid recovered from used polylactic acid so that the costly process of producing lactic acid is eliminated. On the other hand, since the price of plastics of petroleum origin that use naphtha as the raw material is expected to significantly increase due to the plateaued level of petroleum production, the relative cost-effectiveness of polylactic acid will improve in the future as a result of the increased cost of petroleum.

The “Strategic Technology Roadmap for the Green-bio Field” of the “Environment and Energy Sector of Biotechnology Strategy Guidelines” lists the production of useful materials such as high-quality, high-value-added products and the efficient production of materials as technical themes, and defines a road map by classifying the necessary technologies into such categories as those used for “material production making use of microorganisms” and “material production making use of plants.”[29] The road map lists the development of microorganisms and enzymes that are resistant to organic solvents and the development of material production using bio-refineries and closed plant factories as important technologies to be developed. Japan is considered to be dominant in these technologies. According to the road map in the green-bio field, for example, the development of a technology that enables bioprocess making use of microorganisms under special conditions in an organic solvent is expected to realize a significant reduction of production costs by 2025. Among these chemical products that make use of biofunctions, plastics of plant origin are considered to be the core materials and polylactic acid will dominate among them. Therefore, further improvement of the mechanical properties of polylactic acid and the development of low cost production processes are dual issues for the diffusion of this material.

5-2 Effect on the international demand and supply of food

Destabilizing factors for the demand and supply of food, such as population growth, increase in food demand in Asia due to economic development, and the progress of global warming, are increasing worldwide[30, 31]. The expansion of food production and stable supply are limited by factors such as the decrease in cultivated acreage caused by the desertification and depletion of water resources, and the balance between population growth and food supply is being eroded. Therefore, taking the global trends in the demand and supply of food into consideration, the effect of the use of a large amount of polylactic acid for automobile components is estimated as follows.

Figure 13 shows the results of the estimation of the worldwide yield of biomass sugar[32] and the demand for sugar necessary to produce polylactic acid in the automobile industry. The chart shows the results of estimation assuming that all the plastics used for automobiles in the world are replaced with plastics of plant origin (polylactic acid). The estimated total consumption of plastics for automobiles worldwide is about 5.7 million tons at present. To replace all these plastics used for automobiles with polylactic acid, 8.4 million

Biomass	Worldwide yield (100 million tons/year)	Yield ratio of sugar (%)	Yield of sugar (100 million tons/year)
Wheat	5.6	60	3.36
Maize	5.3	60	3.18
Rice	5.3	60	3.18
Potato	2.7	15	0.14
Cassava	1.5	20	0.30
Sweet potato	1.3	25	0.33
Sugarcane	11.0	15	1.56
Beat	2.8	15	0.42
Total	35.5		12.8

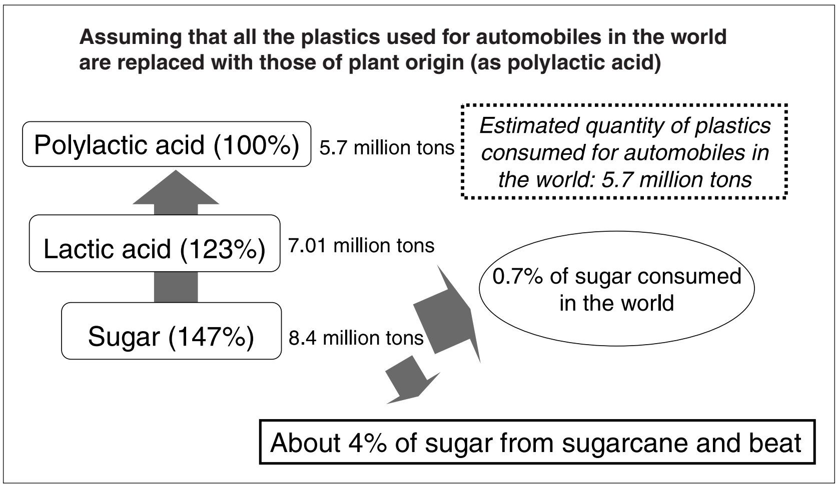

Figure 13 : Estimation of worldwide yield of biomass sugar and demand of sugar for polylactic acid used for automobiles

The yield of biomass sugar is calculated based on Reference[32]. Prepared by the STFC

tons of sugar is required taking the yield ratio into consideration. Since the total yield of sugar in the world is about 1.3 billion tons, about 0.7% of the total sugar produced in the world is consumed by automobiles. If all the sugar used for automobiles is taken from sugarcane and beat only, about 4% is consumed for automobiles. Although it is unlikely that the amount of sugar used for automobiles will immediately cause a food problem, it is desirable to obtain as much of this sugar as possible from surplus sugar of food or surplus biomass resources. Since biomass resources are unevenly distributed among countries and regions, and types and quantities are diversified, comprehensive utilization of biomass on a global level must be promoted.

Although the number of automobiles in the world is expected to exceed a billion in 2010[33], it is not a dream to provide all the interior components of automobiles only with plastics of plant origin, mainly based on polylactic acid, if the mechanical properties of polylactic acid are improved and the cost reduction is achieved as expected in the future. Furthermore, if the technologies developed for automobile components are expanded into other consumer durable goods, it will become necessary to reconsider the effect of the sugar demand for polylactic acid on the demand and supply balance of food.

6 Conclusion

Since the number of automobiles in the world is certain to increase, future technologies that reduce the environmental impact in the total life cycle of automobiles must be developed. In this sense, the role of environmentally-friendly material technology that does not waste natural resources is becoming more important than ever.

For the broad utilization of plastics of plant origin in the future, it is important to grow stable plant materials and develop components that make use of the advantages of natural raw materials. The material structures from microscale to macroscale must be matched to the use environment of automobiles. From the viewpoint of synthesizing technology, research and development for the sophistication of the production process regarding crystallization and molding properties, which are unique to the plastics of plant origin, and significant improvement of material properties such as heat resistance and shock resistance must be conducted. In such research and development, conventional technologies for the plastics of petroleum origin should be applied or taken as a model. To realize the full-scale application of plastics of plant origin to structural materials in the future, it is essential to analyze and evaluate the mechanical properties of materials under the environment where automobiles are used by clarifying the underlying mechanism.

To solve the present cost problem of plastics of plant origin, innovative processes using microorganisms and biotechnology are expected to be developed in the future. In addition, research and development to utilize the advantageous functions of plastics of plant origin, such as transparency and high water absorption, must be promoted to create added values. The research and development of effective production and collecting systems for biomass is also required to improve the economic efficiency by utilizing various types of biomass so that plastics of plant origin further penetrate into various social systems.

Glossary

*1 Carbon neutral material
a material that does not affect the amount of carbon dioxide in the atmosphere through its life cycle.

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Hiroshi KAWAMOTO, Dr. Eng.
Head, Nanotechnology and Materials Unit

Doctor of engineering, the Japan Society of Mechanical Engineers Fellow. He was in charge of strength design and assessment in the development of automobile components at Toyota Motor Corporation, and engaged in projects sponsored by the Ministry of Economy, Trade and Industry (such as research and development of fine ceramics) at Japan Fine Ceramics Center. Specialist in strength design and reliability evaluation technology for structural components.

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