Contribution to the achievement of SDGs
by Furukawa Electric Group

Development of Environmentally-Friendly Cellulose Fiber Reinforced Resin CELRe With Excellent Molding Processability

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ABSTRACT CELRe is a cellulose fiber-reinforced resin that utilizes environmentally friendly, high-strength, plant-derived cellulose fibers. These cellulose fibers are highly dispersed within the resin through advanced kneading technology using a twin-screw extruder. CELRe can be injection molded in the same way as conventional thermoplastic resins, and by adjusting the molding conditions, it exhibits an excellent fluidity and a moldability. Additionally, appropriate moisture content control and drying treatments enable the production of high-quality molded products. Furthermore, it is highly recyclable and has little deterioration in its physical properties, making it easy to reuse. This will promote the reduction of CO_2 emissions and the effective use of resources, contributing to the realization of a sustainable society. In the future, CELRe is expected to be applied to many industries as an important material in today's world, where environmental consideration and technological innovation are required.

1. INTRODUCTION

Cellulose fibers are derived from plants and have excellent properties such as a low environmental impact, a light weight and a high strength. Therefore, it is desirable to use them in automobile parts and structural members by combining them with resins such as polypropylene (PP). However, cellulose fibers are hydrophilic and have low affinity with general-purpose hydrophobic resins such as PP. Therefore, when trying to put cellulose fiber reinforced resins into practical use, there was the issue that the cellulose fibers tended not to disperse but to aggregate in the hydrophobic resins[1].

Taking advantage of the resin mixing technology using extruders that Furukawa Electric has developed in the manufacture of optical fiber cables and power cables, we have established a technique to mix pulp and thermoplastic resin in a twin-screw extruder and to highly disperse cellulose fibers in the resin in a single step.

CELRe requires less energy and can be produced at low cost compared to fiber reinforced resins using nano-cellulose fibers, owing to a unique process that directly disperses cellulose fibers in thermoplastic resin without going through the process of nano-sizing or hydrophobizing the cellulose fibers (Figure 1).

The cellulose fibers of CELRe are defibrated during mixing with the resin in a twin-screw extruder and dispersed in the resin, with a size of the cellulose fibers of approximately 50 to 700 μm. In other words, the CELRe process is a technique for dispersing micro-sized cellulose fibers in resin (Figure 2).

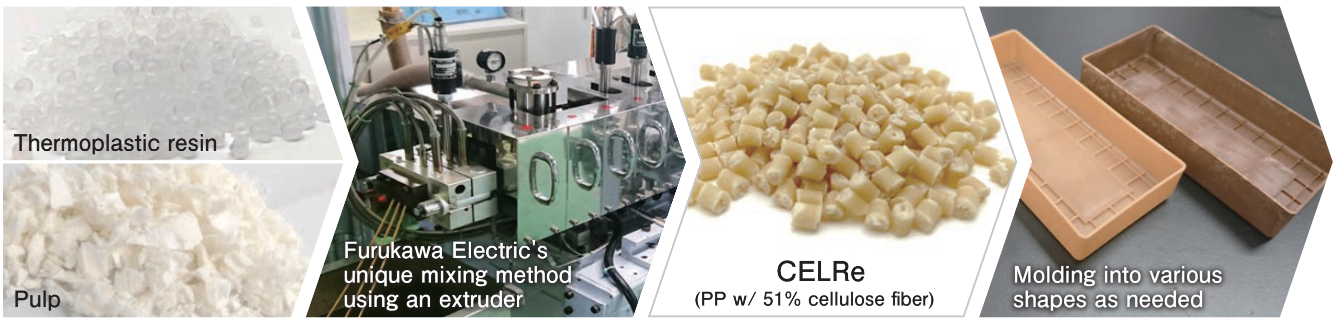


Figure 1 Manufacturing process of CELRe.

[*1] Material Laboratory, Research and Development Division

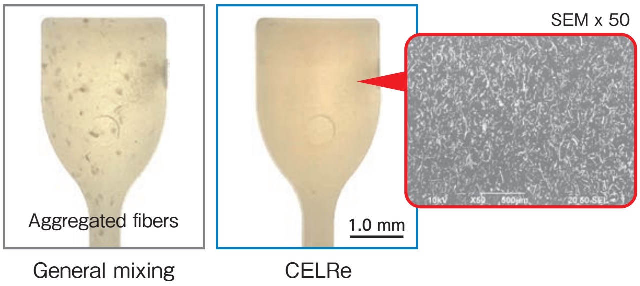


Figure 2 High Dispersion of cellulose fibers in the resin at micro size using our unique technology.

2. MOLDABILITY

Cellulose fiber reinforced resins primarily use thermoplastic resins. The most common method for molding these thermoplastic resins is injection molding. Compared to other molding methods, injection molding is suitable for producing products with complex shapes in a short period of time[2], and is widely used in various fields such as automobiles, home appliances, and office equipment.

This cellulose fiber reinforced resin not only contributes to carbon neutrality through the use of biomass, but also has an excellent mechanical strength. Furthermore, despite its cellulose fiber concentration of 51% and more, CELRe also has an excellent moldability, allowing it to be molded using an injection molding machine and a mold in the same way as conventional thermoplastic resins.

Figure 3 shows examples of molded products using CELRe. In the next section, we will introduce the injection moldability of CELRe in more details.

2.1 Influence of Fluidity

By mixing cellulose fibers into thermoplastic resin, the fluidity is reduced when it is heated and melted. Melt Flow Rate (MFR) is an index often used to evaluate the injection moldability of thermoplastic resins. Since this is measured under static conditions, the shear stress applied to the resin during measurement is much smaller than that during injection molding, which does not necessarily match the behavior during actual molding. Therefore, the spiral flow test is used to evaluate the resin fluidity and the moldability during actual injection molding[3]. In this test, injection molding is performed under certain conditions using a mold with spiral grooves, and the flow length of the resin is measured. This enables to confirm the dependence of the resin flow length on the molding temperature, the injection speed, and the injection pressure.

Figures 4 and 5 show the results of the spiral flow test of CELRe. Under molding conditions similar to those for PP, which is the base resin of CELRe, the spiral flow length tended to be shorter. However, it was found that by adjusting the injection pressure and the molding temperature, the spiral flow length equivalent to that of PP alone could be reached. As a result, it was confirmed that by setting appropriate molding conditions, cellulose fiber reinforced resin can be injection molded in the same way as PP, a general-purpose resin.

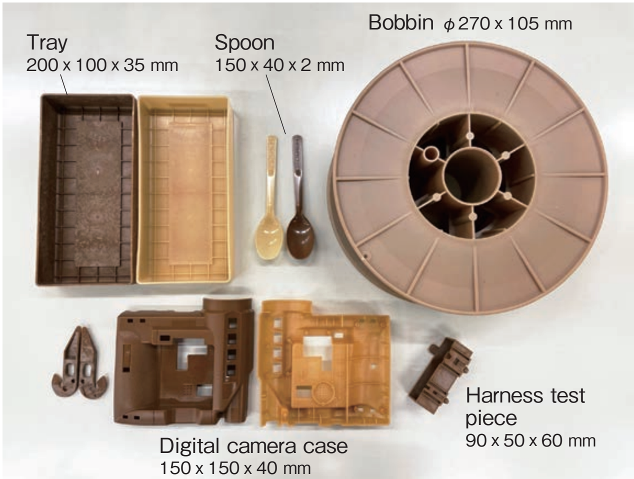
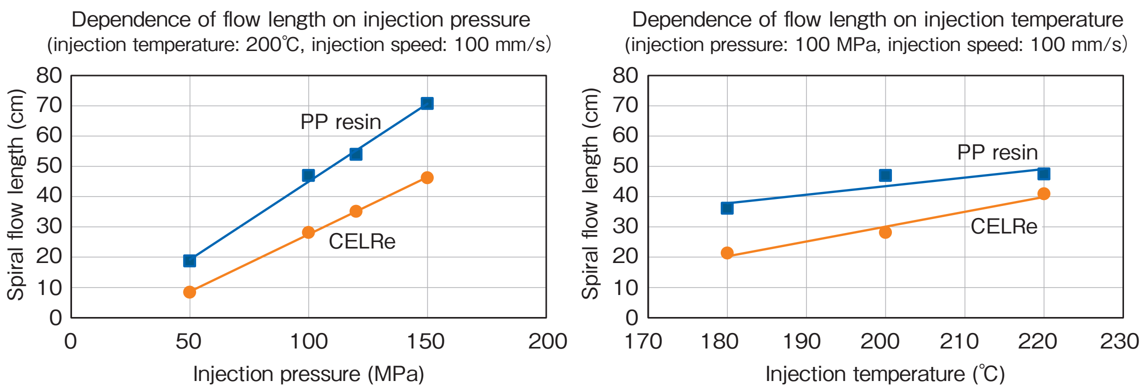


Figure 3 Examples of Various Molded Products Using CELRe.

Figure 4 Examples of the spiral flow test of CELRe (from left: injection pressure 50 MPa, 100 MPa, 150 MPa).



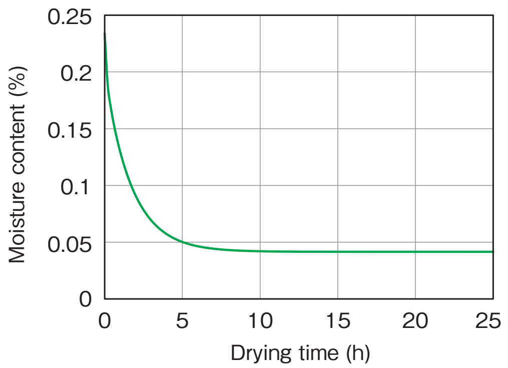
*Spiral shape: Archimedes type spiral flow mold (width: 5 mm, thickness: 3 mm) *Injection machine for testing: 18 tons

Figure 5 Dependence of spiral flow length on injection pressure and injection temperature.

2.2 Effect of the Moisture Content

The moisture content of a resin directly affects its injection moldability and determines the quality of the final molded product. If injection molding is performed when the material contains excess moisture, silver streaks and voids may appear on the surface of the molded product, or decomposition gas may cause short shots and burn marks[4]. In particular, cellulose fiber reinforced resins have the characteristic of easily absorbing moisture from the atmosphere because cellulose fibers are hygroscopic[5]. Therefore, it is more important to control the moisture content than with thermoplastic resins that do not contain cellulose fibers, and proper pre-drying before injection molding is essential.

For example, as Figure 6 shows, when exposed to the atmosphere, CELRe absorbs moisture until it reaches a moisture content of approximately 0.2%. By drying this at 80°C for five hours or more, the moisture content can be reduced to 0.05% and the material can be kept in a stable state. It has been confirmed that when injection molding is performed at this moisture content, the molded product does not show any appearance defects such as foaming or burn marks.



Pre-drying conditions

Drying machine type	Box type hot air dryer
Drying temperature	80℃
Drying time	Five hours or more
Notes	The amount of pellets loaded into a bat should be less than 3 cm.

Figure 6 Moisture content of CELRe and drying conditions before injection molding.

As shown in Figure 6, proper drying treatment is essential when handling cellulose fiber reinforced resin, which results in excellent moldability and high-quality molded products.

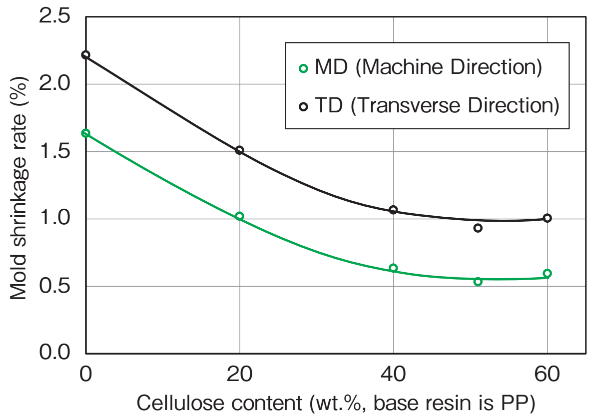
2.3 Mold Shrinkage

After injection molding, the dimensions of resin products decrease as they cool. This phenomenon is known as mold shrinkage. Therefore, molds are designed to compensate for this shrinkage, ensuring that the final product dimensions match the design specifications. In particular, crystalline resins such as PP have the characteristic of having a high mold shrinkage rate because they form a crystalline structure during the cooling process after injection molding[7].

However, adding cellulose fibers to PP can reduce this mold shrinkage rate. As Figure 7 shows, it was confirmed that the mold shrinkage rate decreases as the cellulose fiber content increases, and that when the cellulose fiber content reaches 40% or more, the mold shrinkage rate remains stably low.

PP resin

CELRe molded product (cellulose fiber 51%)



Dependence of cellulose fiber concentration on mold shrinkage rate

Figure 7 Mold shrinkage rate of CELRe molded products.

3. CONTRIBUTING TO A SUSTAINABLE SOCIETY

In order to realize a sustainable society, active efforts are being made around the world to reuse resources and to reduce the use of fossil fuels. Among these efforts, cellulose fiber reinforced resin is attracting attention. This material contributes to conserving petroleum resources and reducing CO_2 emissions by using biomass cellulose fibers instead of conventional naphtha-based resin materials. Furthermore, this cellulose fiber reinforced resin has an excellent recyclability as a molded product, and is expected to contribute further to environmental protection.

3.1 Reducing CO_2 Emissions

Figure 8 shows the carbon footprint of the CELRe manufacturing process. Here, about 41% of cellulose fiber is composed of carbon, and it can be considered that 1.0 kg of cellulose fiber stores 1.5 kg of CO_2. Taking this characteristic into account, as 1.1 kg of CO_2 is emitted during the production of cellulose fibers, its storage capacity is subtracted to calculate the final carbon footprint[8)-10)]. This calculation shows that adding 51% cellulose fibers to petroleum-based PP can reduce CO_2 emissions by about 60% compared to the CO_2 emissions of PP.

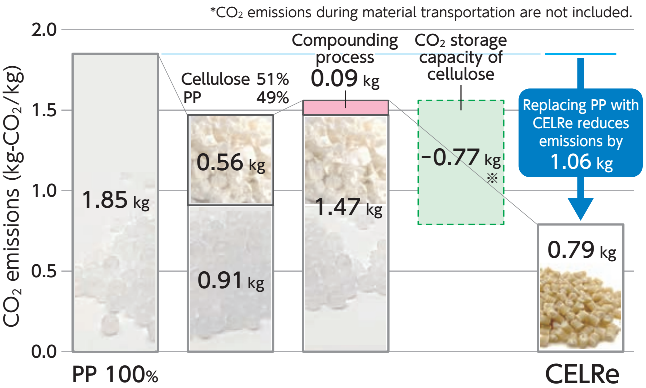


Figure 8 **The carbon footprint during the manufacturing of CELRe (Cradle-to-Gate).**

3.2 Material Recyclability

Cellulose fiber reinforced resin also has an excellent recyclability. It has been confirmed that this material does not significantly lose strength because the cellulose fibers are not easily cut when the CELRe molded products are pulverized. For example, Figure 9 shows the change in strength when injection-molded products made with CELRe are repeatedly pulverized and re-molded. As a result, CELRe retained 90% or more of its tensile strength even after the four recycling cycles. In other words, by reusing cellulose fiber reinforced resin, it is possible to significantly reduce the resource input and consumption of products.

3.3 Closed-loop Recycling

In order to realize a circular society, there is a system called closed-loop recycling, where used products are collected, recycled and then used as raw materials to create new products. This system requires the development of an efficient collection system and advanced recycling technology that can maintain quality. For example, as shown in 3.2, CELRe is a material that can be recycled multiple times.

CELRe, shown in Figure 10, is a typical example of it. With respect to this material, even if scraps (such as spool runners) generated during the manufacturing process are pulverized and reused in injection molding, there is almost no loss in strength. This property will enable a closed-loop recycling system, which will result in a significant reduction in CO_2 emissions and in waste. Such efforts are a significant step towards a sustainable future, paving the way to produce high-quality products while minimizing environmental impact.

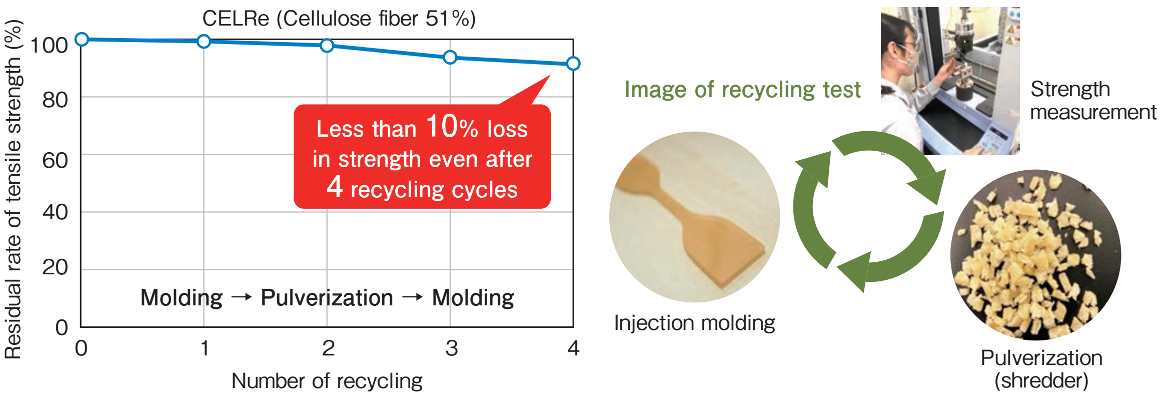


Figure 9 **CELRe's residual rate of tensile strength depending on the number of recycling.**

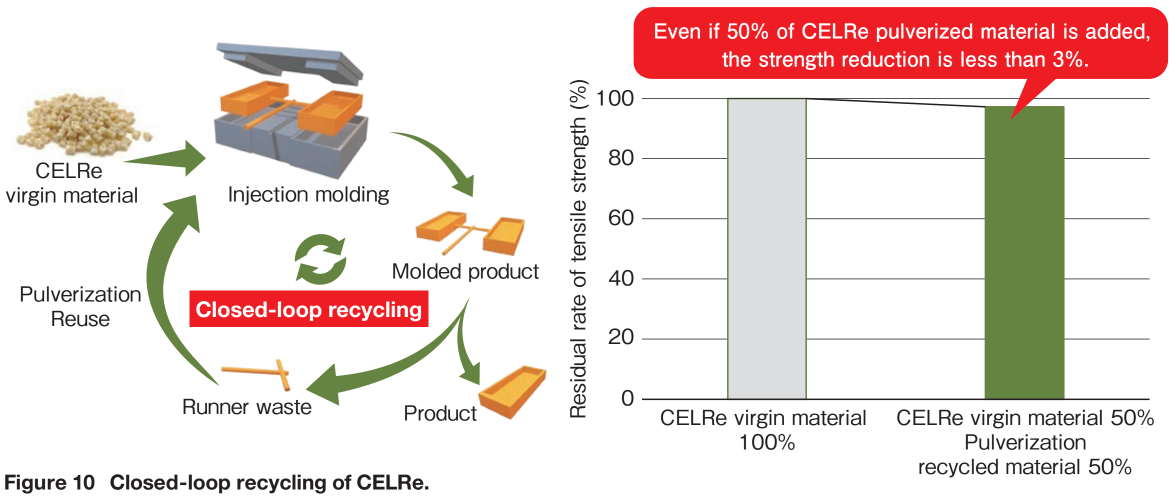


Figure 10 **Closed-loop recycling of CELRe.**

3.4 Biomass Mark

Recently, resin composites containing a large amount of cellulose fibers have been attracting attention as materials that can significantly reduce plastic usage and lower environmental impact.

So far, cellulose fiber reinforced resins, in which a small amount of cellulose fibers is dispersed, have been mainly studied. However, from the perspective of reducing the environmental impact (carbon neutrality), efforts are being made to replace plastic with wood materials as much as possible. In other words, it is expected that new resin composites containing a large amount of cellulose fibers will be created, and research is being conducted to realize this.

As part of this, CELRe's KCP511 grade has already acquired the "Biomass Mark 50" certification from a third-party organization (Japan Organic Resources Association) (Figure 11). By replacing conventional resin materials derived from naphtha with biomass materials such as cellulose fiber reinforced resin, we contribute to conserving petroleum resources and reducing CO_2 emissions.

Figure 11 Biomass Mark of CELRe issued by Japan Organics Recycling Association.

4. CONCLUSION

We focused on the properties of the cellulose fiber reinforced resin CELRe and introduced its advantages. Cellulose fiber reinforced resins are generally thought to be unsuitable for injection molding because they have low fluidity and high water absorption. However, by setting appropriate molding conditions, CELRe is actually a material that can exhibit a high injection molding performance similar to that of conventional general-purpose resins.

Due to the recent growing interest in environmental issues such as global warming and the depletion of petroleum resources, the demand for cellulose fiber reinforced resins as a biomass material, is expected to increase further in the future. Starting from fiscal year 2023, CELRe has established a production capacity of 10 tons a month and has commenced providing samples. Through CELRe, we will continue to contribute to reducing CO_2 emissions and to promoting the recycling of the products, aiming to realize a sustainable society.

REFERENCES

1) Yutaka Yoshida: "Development of Composite Resins with Homogeneously Dispersed CNF, Seikei-Kakou", 35 (2023), 108. (in Japanese)
2) Michihiro Tatsuno: "Injection Molding, Seikei-Kakou", 31 (2019), 254. (in Japanese)
3) Takeshi Sakamoto, Masayoshi Ando: "A Measure of Moldability of Polymers in Injection Molding", Koubunshi-Kagaku, 19 (1962), 351. (in Japanese)
4) Yoshiyuki Nakao, Hiroyuki Narahara, Michio Kojima, Hiroshi Suzuki: "Study on Metal Laser Sintering Mold having gas vent structure to avoid molding defects", Proceedings of the 2006 Spring Meeting of the Japan Society for Precision Engineering, 791. (in Japanese)
5) Takeshi Semba: "Cellulose Nano-fiber and Plastic", Seikei-Kakou, 30 (2018), 361. (in Japanese)
6) Takao Kameda, Tatsuhiro Takahashi, Kiyohito Koyama: "Effect of Packing Pressure on Molding Shrinkage Distribution of Injection Molded Parts of Acrylonitrile-Styrene (AS) resin", Seikei-Kakou, 17 (2005), 571. (in Japanese)
7) Naoichi Takashima, Kunio Mizutani: "Injection Molding of polypropylene, Koubunshi", 12 (1963), 504. (in Japanese)
8) S.V. Joshi, L.T Drzal, A.K Mohanty, S. Arora: "Are natural fiber composites environmentally superior to glass fiber reinforced composites?", Composites Part A: Applied Science and Manufacturing, 35 (2004), 371.
9) C.S. Boland, R.D. Kleine, G.A. Keoleian, E.C. Lee, H.C. Kim, T.J. Wallington: "Life cycle impacts of natural fiber composites for automotive applications: Effects of renewable energy content and lightweighting", Journal of Industrial Ecology, 20 (2016), 179.
10) Shinji Nakayama, Tokiya Yaguchi: "The LCI Calculation Method for LCA of Pulp and Paper Products" JAPAN TAPPI JOURNAL, 56 (2002), 1035. (in Japanese)