Research on Environmental and Scientific Issues in Carbon Fiber-Reinforced Plastics (CFRP) Recycling

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(炭素繊維強化プラスチック(CFRP)のリサイクルにおける環境科学的課題に関する研究)

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1. OVERVIEW

Carbon fiber-reinforced plastics (CFRP) are used for structural materials in the aircraft and automotive industries due to their lightweight and high strength characteristics.

CFRP is a very hard and chemically stable material and therefore the disposal of waste CFRP has been a big issue over the last few years.

In the U.S., it is widely known that retired aircrafts made of CFRP have been left on the desert due to a lack of proper disposal method. At present, there are still no suitable disposal methods in terms of price and environmental impact (energy consumption, LCA etc) and as a result, landfill disposal, which is relatively inexpensive, is the preferred method.

As for the world movement, in Japan, the "Circular Economy Vision 2020" (issued by the Ministry of Economy, Trade and Industry, Japan on 22 May 2020) designates CFRP as one of the five areas (Plastics, Textiles, CFRP, Batteries, and Solar panels) that are in urgent need of consideration for recycling systems.

In Germany, landfilling of untreated CFRP waste is prohibited since 2005.

For such a situation, the establishment of recycle/reuse system is being urged around the world.

In this study, firstly I organized the issues for disseminating recycling technologies of CFRP, especially in light of Japan. As a result, it was clarified that CFRP recycling process inevitably generates carbon fiber (CF) fine particles (dust/milled) as a byproduct, and while there is an urgent need to establish recycling technology, only the technical aspects of the recycling process have been paying attention and little has been known about how CFs including not only the recycled CFs but also the secondary dust affect the ecosystem.

There also exist reports that CF is nanoscaling and finally become NOAA (Nano-objects, and their aggregates and agglomerates greater than 100 nm). With regard to NOAA, there is concern about its health effects.

In this background, secondly I delved into the effects of milled CFs or CF dust generated during a CFRP recycling process on the environment and living organisms when CFs are released into the water environment as a by-product unintentionally.

Specifically, the toxicity evaluation of CFs was conducted by using Japanese medaka fish (*O. latipes*, orange-red variety or himedaka).

This study will help us to predict how CF fine particles generated from the recycling process and unintentionally contained in industrial wastewater will affect aquatic organisms in rivers, lakes and marshes etc.

It is also helpful to formulate countermeasures in the future when CFRP recycling is prevailed and large amounts of recycled CFs are used in our daily lives.

Finally, an occupational health risk assessment study by using Control Banding approach for CF dust which includes NOAA was conducted on a pilot basis for the aim of future implementation on SMEs (small and medium-sized enterprises) work site.

2. REVIEW AND EXPERIMENT

2-1 Organizing current issues in CFRP recycling

2-1-1 Organizational issues for disseminating recycling technologies of CFRP in the Japanese industrial landscape

2-1-1-1 Introduction

Carbon-fiber-reinforced plastics (CFRP) are used for structural materials in the aircraft and automotive industries due to their lightweight and high strength characteristics. Global CFRP market demand was 92,900 tons in 2015 and is expected to reach 269,500 tons by 2024 [1]. As an alternative to metals, they are expected to expand their application into many fields. However, the recycling businesses/technology of waste CFRP is still virtually unestablished especially in Japan and waste CFRP has been mostly landfilled across the globe. The main reason for landfill disposal of CFRP is that it is incombustible and thus requires a large amount of fuel to burn [2]. In addition, in aircraft applications, in-plant waste such as trimming offcuts are often regarded as “strategic supplies” because carbon fiber lamination and knitting are areas of corporate technology expertise. Therefore, small- and medium-sized companies that have been commissioned by aircraft manufacturers run the risk of being penalized by the outsourcing companies if the waste is mishandled and consequently leaks outside. To prevent such risks, landfill disposal has been prevalent so far. Consequently, landfill is still preferred in many countries as the most cost-effective disposal option in monetary terms.

For reference information, in the UK, the landfill cost of waste CFRP is reported to be approximately £82.60 per tonne [3] and there is also an article stating that in 2015 some 18,000 tonnes of CF waste was produced from manufacturing operations globally, of which only around 1,600 tonnes (8.8%) was recycled [4]. In Germany, landfilling of untreated waste is prohibited since 2005. Under such circumstances, to further spread and expand the use of CFRP in the future, it is highly desirable to reduce the environmental load of the entire value chain from production to recycling through the development of recycling and reuse technology. As a remarkable movement within the EU, there is a report that the European Union is aiming for zero landfill disposal by 2025 [5]. There is also a report that the market size of CFRP recycling will reach approximately 100 billion yen in 2030 [6]. In anticipation of the growth of waste that is derived from the growth of demand for CFRP in the future, it is necessary to create a waste material treatment system. Therefore, the establishment of CFRP recycling technology and the development of applications for recycled products are an urgent requirement. However, up until now, there have been almost no cases where reclaimed CFs have been used for mass production, only prototypes, such as aircraft seat arm rests, have been produced, except for the case of BMW. Ahead of the rest of the world, BMW started to use recycled CF in the roof and rear seat shells of their electric i3 and the hybrid i8 vehicles [7, 8]. More recently, especially in Japan, it has become desirable to expand applications by utilizing functional characteristics such as low thermal expansion rate, thermal conductivity, corrosion resistance, and rustlessness, besides the more conventional mechanical properties such as lightness, strength, and hardness. New businesses have been developed by utilizing these additional benefits of CF other than lightness and strength. Concretely, a number of Japanese companies are developing its use in new fields, such as medical field, by utilizing virgin/reclaimed CF as electromagnetic shielding materials [9, 10]. To conclude this section, the flow of collecting and recycling CFs from waste CFRP consists of material recycling and thermal recycling as shown in Fig.1. CFRP waste comes in many forms, such as dry fiber, prepreg (intermediate feedstock that is impregnated with resin in lined CF) scraps, cured parts and currently, regarding CF waste from the aircraft manufacturing process, prepreg, preform (pre-molded CF that fits the product shape), and molded waste are mainly discharged. However, in the future, a large amount of end of life CFRP waste materials are expected to be discharged.

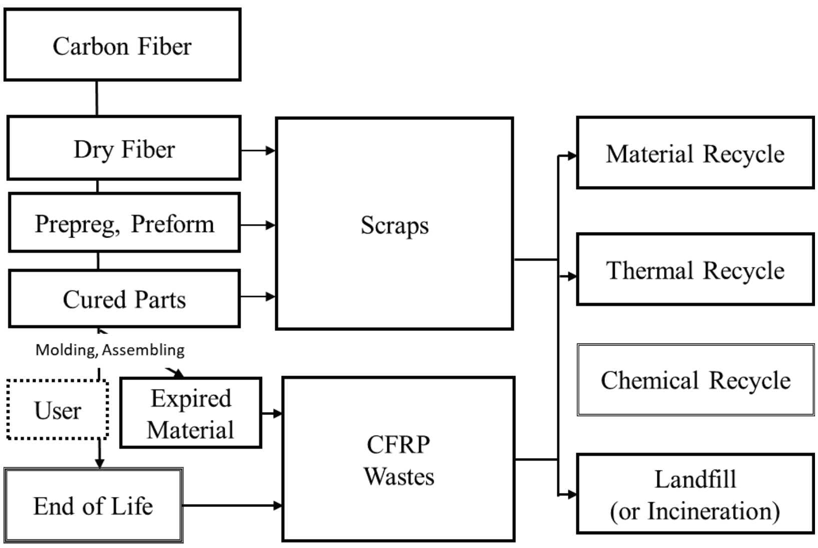


Fig.1 Flow chart of collecting and recycling CFs from waste CFRP

2-1-1-2 CFRP recycling technology

Categorizing crudely, there are two types of recycling technologies [8].
As shown in Fig.2, the first one is a direct reuse (mechanical recycling) technology that can use waste CFRP as it is for the purpose of filler, reinforcement, and so on. Uhlmann and Meier [11] have studied the application of CFRP milling dust as a filler material. They regard the approach to consider milling dust as an additive for reinforcement purposes as one of many possibilities to close the material cycle of used CFs.
The second one is a full recycling process technology. It can be divided into fiber reclamation (recovery of the fiber from the matrix) and remanufacturing into a valuable material (the production of a new feedstock or component).
In terms of fiber reclamation, techniques can be categorized in two intersecting groups: the thermal processes (pyrolysis, fluidized bed pyrolysis, microwave-assisted pyrolysis) and the chemical processes (solvolysis, super-critical fluid solvolysis). In terms of remanufacturing, it is possible to use the recycled CFs (rCFs) as reinforcement in direct molding techniques (injection molding, bulk compound molding, extrusion molding) or to create non-woven or chop strand mats for injection molding [12, 13].
In a full recycling process technology, technique that can recycle both resin and fiber is ideal and research into processes such as the effective separation of resin and fiber is under way by AIST, Japan etc [14]. With regard to recycling technology aimed at recovering CF threads, many companies and research institutions in Japan and overseas are involved, as shown in Tables. 1, 2, and 3. Wet methods such as solvolysis [15, 16] have a better separation accuracy than dry methods from an academic perspective. However, it is expensive and necessary to conduct cumbersome waste liquid disposal and therefore many companies, especially overseas recyclers, adopt pyrolysis (thermic treatment) technology [17]. Generically, the pyrolysis method is energy intensive and can leave char residue on the surface of CFs which is hampering further processing though it depends on process conditions.

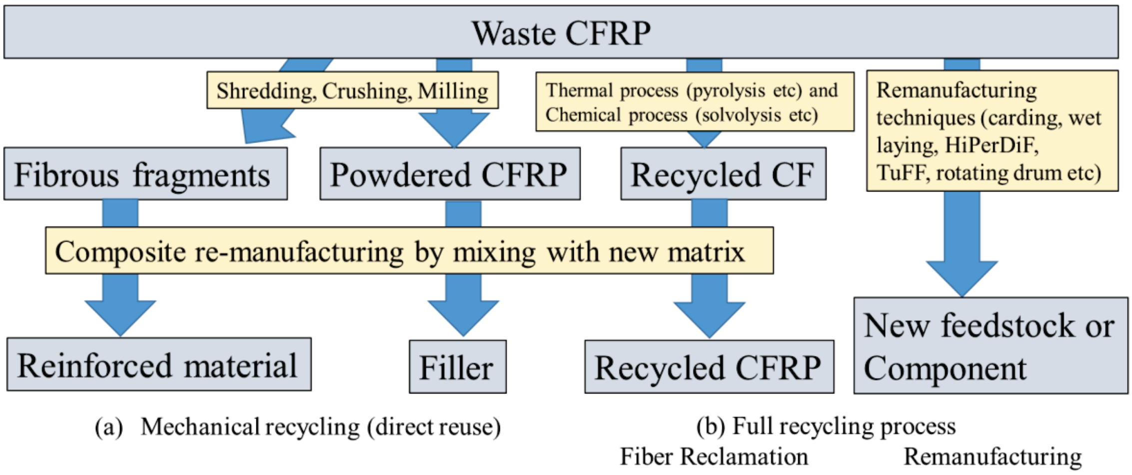


Fig.2 Two concepts for CFRP recycling

Table 1. Example of major recycling companies in the world classified by major recycling method

Solvent Method	CA TACKH (South Korea), Vartega (USA), Ai-Carbon Co., Ltd. (Japan) (Electrolysis), ACA Co., Ltd. (Japan) (Technology to strike CFRP with an air flow close to sound speed and finalize up to 1 µm in diameter) (1), SANWAYUKA INDUSTRY CORPORATION (Japan) (Solvolysis - wet and low temperature (100-150 °C) CFRP recycling technology by using sulfuric acid), Earth Recycle Co., Ltd. (Japan)(Wet depolymerization method)
Pyrolysis	Karborek (Italy), ELG Carbon Fibre (UK)(2), CarboNXT GmbH - CFK Valley Stade Recycling GmbH & Co. KG (Germany), Composites United (formerly CFK Valley) (3) (Germany), Carbon conversion (United States), GEM Co.,Ltd (China) (details are unknown)(4), Shinryo Corporation (Japan), KUREHA CORPORATION (Japan), Fuji design co.,ltd - Re-Tem Corporation (Japan), Teijin Limited (Japan)

(1) ACA CO.,LTD. website http://www.ss-aca.jp/english/reuse/ Accessed 21 August 2020

(2) Holmes M (2018) Recycled carbon fiber composites become a reality. Reinforced Plastics, Volume 62, Number 3, May/June 2018. doi: https://doi.org/10.1016/j.repl.2017.11.012

(3) JEC GROUP NEWS (2019) CFK Valley and Carbon Composites to merge into Composites United. http://www.jeccomposites.com/knowledge/international-composites-news/cfk-valley-and-carbon-composites-merge-composites-united Accessed 21 August 2020

(4) GEM (2014) Mitsui & Co. to expand its car recycling business in China, establishing a joint venture with GEM Co.,Ltd. (2014.05.26) (in Chinese) http://www.gemchina.com/gongsidongtai/2014/05-26/599.html Accessed 21 August 2020

Table 2. Comparison of Japanese recycling technology

Technology classification	Two-step pyrolysis	Pyrolysis	Thermal activation of semiconductors (TASC)	Depolymerization of Epoxy Resin under Ordinary Pressure[6]	Supercritical fluid method[6]	Subcritical fluid method [6]
Research Institute	Carbon Fiber Recycle Industry co.ltd. (CFRI)	The Japan Carbon Fiber Manufacturers Association (JCMA),	JinTech Corporation [5]	Hitachi Chemical Company, Ltd (withdrawal from the business)	Shizuoka University	Kumamoto University
Type of resin	All resins that do not generate toxic gas during pyrolysis	All resins that do not generate toxic gas during pyrolysis	Unknown	Ester	Ester	Ester
Type of solvent					Methanol	Benzyl alcohol
Recoverable fiber type	Long CF, Resin pyrolysis gas	Milled CF	Long CF	Long CF, Resin degradation product	CF, Resin oligomer (Dissolved in alcohol)	CF, Resin degradation product
Temperature	500℃(1st step) 440℃(2nd step)	500-700℃	400℃	200℃	250-350℃	300-400℃
Pressure	ordinary pressure	ordinary pressure	ordinary pressure	ordinary pressure	5-10MPa	1-4MPa
Time(minute)	30	120	10	90-180	Unknown	Unknown
Preliminary process	unnecessary	smash	unnecessary	unnecessary	smash	unnecessary
Scale(ton/year)	2,000	1,000	Unknown	12	Laboratory level	Laboratory level

(5) JinTech (2013) Outline of the TASC technology in English http://jintech.org/?page_id=342 Accessed 21 August 2020

(6) Tanaka K, Yoshimura T, Ijiri M, Nakagawa D (2019) Approach from cavitation processing for new recycling technology development of carbon fiber reinforced plastics. The Japan Society of Mechanical Engineers Vol.85, No.874.
doi: 10.1299/transjsme.18-00342https://www.jstage.jst.go.jp/article/transjsme/85/874/85_18-00342/_pdf/-char/ja Accessed 21 August 2020

Table 3. Comparison of recycling technology other than Japan

Country	the United Kingdom			Italy		Germany			Swiss	France	Spain	the United States		China	
Research Institute	ELG Carbon Fibre Ltd [2]	Imperial College London	University of Nottingham	Karborek Recycling Carbon Fibres	RYMYC	Fraunhofer IGCV	CarboNXT GmbH, CFK Valley Stade Recycling GmbH & Co. KG	SGL Automotive Carbon Fibers	V-CARBON	I2M Laboratory (engineering and mechanics institute, University of Bordeaux)	Formoso Technologies Group [7]	Carbon Conversions, Inc.	Adherent Technologies, Inc [8]	UHT Unitech ,Trans. Com	Fuyuan new material technology Co. LTD (FUY) [9]

Technology type	Pyrolysis	Unknown	Microwave pyrolysis flow, Moving layer, thermic fluid treatment	Unknown	Non-chemical process, likely Superheated Steam	Pyrolysis (Ar, N_2, O_2, (oxidation possible)), max. temp: 800°C	Pyrolysis	Pyrolysis	Pyrolysis and chemical reaction	Solvolysis [10]	Thermal decomposition (recovery of oil from the resins and fibres)	Pyrolysis	Wet chemical degradation of the polymer matrix	Microwave pyrolysis	Pyrolysis
Scale (ton/year)	2,000	Laboratory level	Laboratory level	1,000	300	Under joint research with companies	3,500	Unknown	Unknown	Laboratory level	Pilot plant level	2,000	Pilot plant level	88	1,500

(7) Oliveux Géraldine, Luke O Dandy, Gary A Leeke (2015) Current status of recycling of fibre reinforced polymers: Review of technologies, reuse and resulting properties. Progress in Materials Science 72, 61–99. doi: http://dx.doi.org/10.1016/j.pmatsci.2015.01.004

(8) Adherent Technologies (2020) Recycling Technologies. https://www.adherent-tech.com/recycling_technologies/carbon_fiber_reclamation_faqs Accessed 21 August 2020

(9) Fuyuan new material technology Co. LTD (FUY) website http://www.rcffy.com/, http://www.rcffy.com/news/info.aspx?itemid=74. Accessed 21 August 2020 Recently, three association standards associated with CF recycling were drafted under their lead and had been released.

(10) Matthieu Ayfre, Olivier Mantaux (2019) Innovative semi-products made of recycled carbon fibres. JEC COMPOSITES MAGAZINE 2019, N°131, November-December 2019.

2-1-1-3 Issues in commercializing CFRP recycling technology

The high important issues of commercializing CFRP recycling technology are as follows:

(a) Quality assurance and standards for the fiber properties of reclaimed CFs do not exist.

Catalog values for reclaimed fibers do not exist and very few recyclers such as ELG Carbon Fibre specify their properties on their homepage. Therefore, users cannot choose reclaimed fibers with confidence.

As a background for the above, with regard to CFRP recycling, the quality and physical properties of in-plant waste and other waste materials used as raw (input) materials are not uniform. It is therefore difficult to maintain the quality of recycled products within a certain range. As Longana et al. [13] state, it is necessary to develop new quality control and characterization techniques able to provide reliable mechanical properties in a production-compatible timescale to promote the industrial application of recycled composite materials. Furthermore, the need for ad hoc characterization and quality control techniques for recycled composite materials is further justified by the fact that the waste, and consequently the rCFs, can come from multiple, and potentially unknown or scarcely documented, sources. Regarding the issues related to remanufacturing, needless to say, the quality assurance and standardization will be also applied to the remanufactured feedstock. While there are efficient industrial scale setups for fiber reclamation, remanufacturing methods capable of producing high performance recycled composites are still rare. The most immediate way to remanufacturer CF is to use indirect molding techniques, such as injection molding, bulk molding compound (BMC), and Sheet Molding Compound (SMC) compression [12].

(b) Lack of a reliable supply chain and established market outlet.

For the input, CFRP scrap materials are not continuously available. Furthermore, for the output, the application development of reclaimed CFs has not yet been established and therefore customers and applications of recycled products have not been secured. If the number of lots on the input side cannot be secured, the recycling business itself will not become established.

(c) With regard to the effects on humans and ecosystems caused by CF (dust/milled), enough studies on their bio-toxicity and environmental toxicity have not been conducted.

The impact of microplastics on ecosystems has been noted in recent years and selected reports are as follows. Though CFRP is a variation of the same kind of plastic and attracting attention as an environmentally friendly product, there are few reports on its environmental impact, especially regarding the effects on living organisms, compared to microplastics [18, 25].

・Intake of harmful substances, such as adsorbed microplastics, carries a risk of causing health damage to the ecology [19, 20].

・Low-density polyethylene plastic (LDPE) causes liver dysfunction in Oryzias latipes [21]. Note that human and oryzias latipes share 70% of the same genes.

・The accumulation of 14 plastic particles in the intestines of sea turtles leads to 50% mortality [22].

・The rate of infection to coral diseases under the influence of plastics increases from 4 to 89% [23].

Considering and summarizing the above, Fig.3 shows the whole issue related to each recycling process.

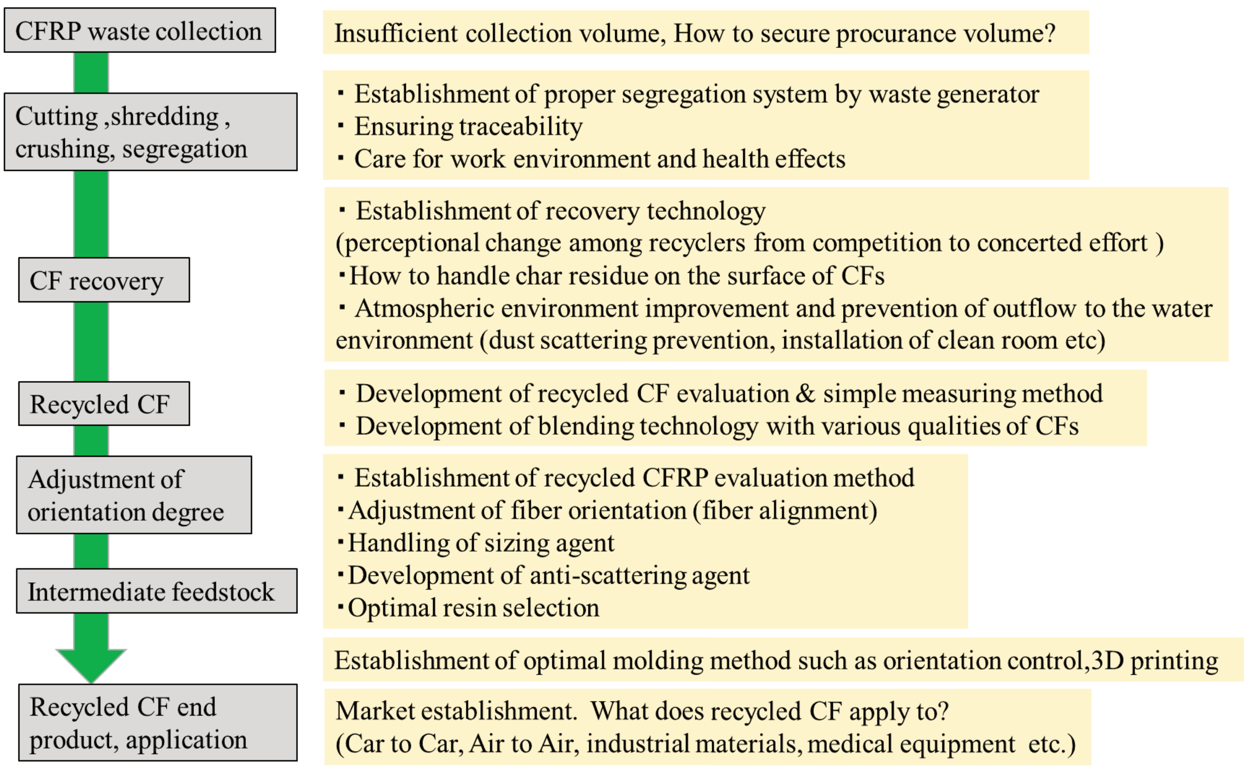


Fig.3 Issues in each step of CF recycling process

In addition to the above (a) to (c), there are other aspects regarding the impact of CFRP recycling both in terms of process and in terms of LCA impact categories. Needless to say, the aspect of economic and environmental assessment is important. [18, 24-26]
The choice of technology route of CFRP recycling and cost/profit consideration are also high important issues since there are many technology routs for CFRP recycling.

Moreover, the choice of technology itself is a key factor for the commercial operation since it is highly related to the cost and discharge type of CFRP recycling process. [7, 27]

2-1-1-4 Solutions and prospects for CF recycling business

The solutions and prospects for CF recycling business are as follows:

(1) Quality assurance and standardization

For virgin CF, the test methods and standards have been established to some extent in Japan etc [28]. Conversely, with regard to reclaimed CF, there are still no established test methods and standards and therefore each batch of reclaimed CF products is shipped in a specified quality range according to user demand. In addition, it is expected that CFRP waste from unknown sources will be found in the future. The quality assurance and standardization will be applied to both the reclaimed fibers and the remanufactured feedstock. Longana et al. [13] describe issue of collection and sorting of the waste and reclamation-induced alterations to fiber geometry, architecture, mechanical and physical properties. Moreover, they describe a study of quality control and property assurance methodology based on the HiPerDiF (High Performance Discontinuous Fibre) method aimed at blending of different fiber lengths. The HiPerDiF method has been used to remanufacture rCFs to obtain high mechanical properties in intermingled hybrids with virgin fibers and interlaminated hybrids with continuous glass fibers and to demonstrate the possibility to maintain high performance after multiple closed loop recycling processes. In the future, it will be necessary for Japan to standardize resin type, formulation ratio, shape, fiber length and so on, and propose a standard measurement method or at the very least a guideline in cooperation with the European norm etc so that sellers and buyers can trade with confidence. Currently, in Japan, a study on the reclaimed CF testing and evaluation method (method to quantitatively evaluate the strength [29], length, orientation (fiber alignment), residue and so on for reclaimed CFs) has been conducted mainly at the AIST and Kyoto University. Such standardizations are also required overseas (such as Airbus). We would therefore like to suggest the launch of a global standard from Japan. It is also expected to proceed with a collaborative study for the establishment of a test and evaluation method in cooperation with Fraunhofer IGCV and the Brightlands Materials Center in the Netherlands, among others.

(2) Lack of a reliable supply chain and established market outlet

There is an urgent need for establishing a waste supply chain. As mentioned so far, there are still no predominant applications and this is the most challenging issue especially for Japan. Reclaimed CF is a cut fiber and it is therefore difficult to apply to structural materials requiring strength. However, reclaimed CF is adequate if users seek a weight reduction equivalent to those of general reinforced plastics. Through further improvement of the existing remanufacturing techniques (carding [30, 31], wet laying [32], HiPerDiF [12, 13, 33, 34], TuFF (the Tailorable Feedstock for Forming) [35, 36], rotating drum [37] etc), new remanufacturing methods capable of producing high performance recycled composites are expected to be developed. This conceivably contributes significantly to the expansion of application. In the future, it is expected to expand its application into automotive parts, civil engineering and construction materials, medical components, and daily necessities. For civil engineering and construction materials, for example, by replacing steel (iron) in bridges with lightweight CF, the burden of repair work can be reduced and maintenance-free is potentially possible. This will have a significant impact on the aging society and participation of women in society. This effort has already been made by a number of Japanese companies by combining virgin CF with glass fiber and the use of CFRP as a reinforcement for the pillars of houses is also under consideration [38]. Conversely, in automotive component applications, if automatic operation progresses in the future, it is expected that the material strength for personnel protection will no longer be required and robust composites such as CFRP will become over-specification with general types of plastic being good enough. As a solution to application development especially in Japan, we would like to suggest listening to the opinions of downstream users (end product manufacturers) as to which type of intermediate feedstock are better among drylaid nonwoven and wetlaid nonwoven. Specifically, the exchange of opinions involving all stakeholders across upstream (recyclers), midstream (intermediate feedstock manufacturers), and downstream (end-product manufacturers) is considered effective as shown in Fig.4. Moreover, it is believed that collaborative market development will be effective through mutual cooperation among intermediate feedstock manufacturers, resin manufacturers, and processing manufacturers beyond the watershed area.

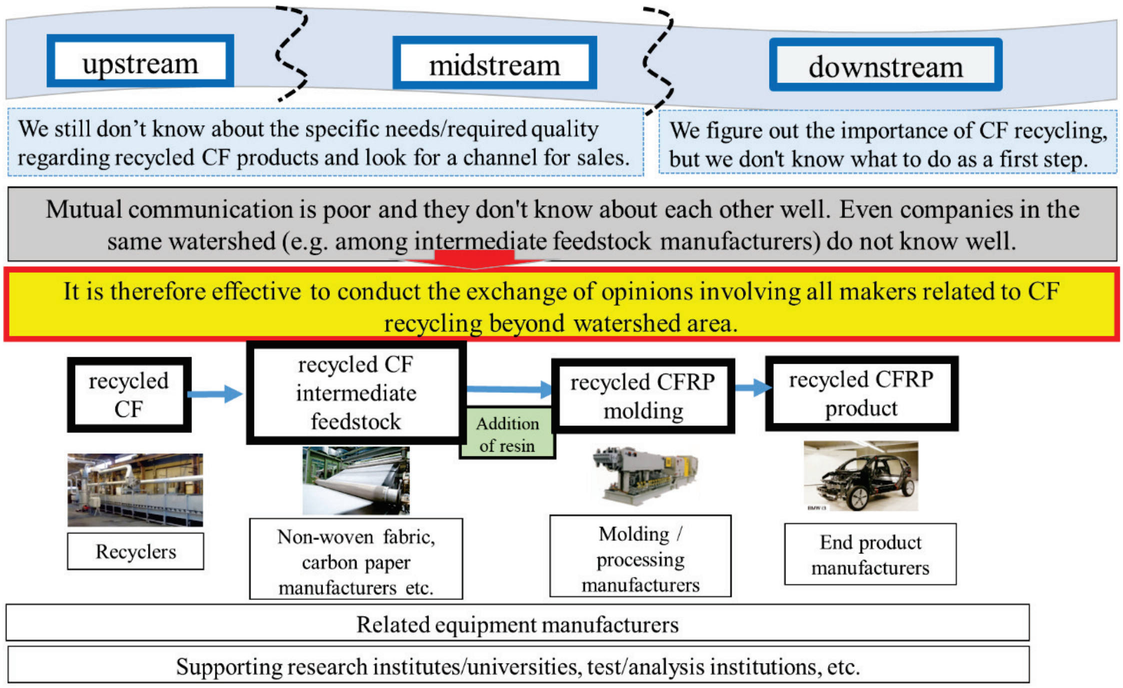


Fig.4 Example of application development solution– exchange of opinions involving all manufacturers involved in CF recycling

(3) Effect of CF on humans and ecosystems

Generally, carbonized CF (after partial firing treatment) recovered from the pyrolysis method contains many impurities such as tar and char [39]. Therefore, its users are more likely to be anxious about its impact on workers and ecosystems than with virgin CF. In particular, reclaimed CF recovered from CFRP retains more tar, char etc than that recovered from CFRTP due to the difference in matrix resin decomposition properties. In addition, unintended impurities such as glass fiber, metal foil, and dust can be containing on the surface of reclaimed CF [40]. When wet treatment method is used as the production of intermediate feedstock or water jet cutting is used, the transfer of CF dust to the water system through discharged water is a significant concern. Even when scrubber systems are used, a risk remains that, if mishandled, the CF dust-tainted water recovered from the atmosphere transfers to external water bodies. If robotic automation systems [41] are not installed for mass production, comfortable working environments will need to be established. For not only in the intermediate feedstock manufacturing process but also in subsequent processes, the safety of workers continues to be a concern because CF dust will always be generated when the CF is cut, smashed, and so on.

As for the impact on humans and ecosystems, the following are clarified:

First, the impact on human health;

-Carbon is considered to be biocompatible and CF and its composite materials are also used in artificial biomaterials [42].

-The diameter of original CF on the market is 5 μm or more, and its particle size differs from "black carbon (2.5 μm or less in diameter)" contained in PM2.5 and "carbon nanotubes (CNTs) (0.4–50 nm in diameter)," which have associated concerns with regard to health effects on the respiratory system. Particularly, it is elucidated that CNTs pose serious health risks similar to those of asbestos and carry a high risk of inducing lung cancer [43-48].

-Unlike asbestos, dust aspiration does not affect red blood cells, alveolar macrophages, and so on, or is considered to have a small effect. [49]

-Waritz RS et al. conducted histopathological evaluations by exposing rats to CFs in the atmosphere. CFs used in their study were made from polyacrylonitrile fiber and 3.5 μm in diameter and 72% were 10–60 μm long. Rats were exposed for 6 h /day, 5 days /week, for 16 weeks to an atmosphere of 20 mg /m3 of CFs (25 × 106 fibers /m3).

As a result, no effects due to the exposure were seen, as judged by clinical signs, body weight, organ weight, organ-body weight ratios, organ-brain weight ratios etc. There was also no indication of fibrosis. [50]

-Judging from the particle size of original CF, it is unlikely to cause serious damage to human health through inhalation. However, fine particles such as those of milled short fiber (fiber diameter less than 3 μm, fiber length less than 80 μm) treated by industrial processes have a likelihood of reaching the human alveoli by inhalation. Therefore, Nakano et al. of Keio University have recommended that a reassessment of their toxicity should be conducted using those small/fine fibers. In addition, they have recommended that a re-evaluation of special CFs, such as those treated with a chemical surface coating, should be conducted. [51]

-Moriyama and his team, of Gifu University, have studied and analyzed the toxicity and health effects on workers of CF dust, which is generated during a pyrolysis recycling process at the molecular level, by using live mice. They suggested the inherent impurities, such as dioxins, attached to the CF rather than the physical state of the fiber (such as fiber length) may impart toxicity to CF dust. They also found that the toxicity and potential hazard to human health changes depending on the degree of firing treatment, has no severe impact on human health as a consequence. [52]

Secondly, the effect on aquatic organisms;

Although CF has characteristics that do not affect the ecological environment of the organism as a biocompatible material [53], there are very few formal reports, such as papers and articles, with scientific evidence [54]. Regarding other types of plastics, like microplastics as mentioned in Subsection (c) of Section 3, the ecological effects have been analyzed to some extent. However, the effects of CF migrated to water sources, such as rivers and lakes, remain unclear and those effects have yet to be investigated. In addition, there is concern over the transfer of CF to groundwater and surrounding sea areas from the final disposal area. Regarding CF, the yarn itself is treated as a stable final disposal site, however, in the form of CFRP, it is treated as a managed final disposal site in Japan. [55] In the event that the capacity of the landfill site is full, although it is unknown whether CF will separate from CF containing products in seawater over the years, it is feared that waste CF products will be dumped into the ocean illegally. Since the announcement of Professor Kojima of National Institute of Technology, Gunma College, Japan in 1994 [56], it has been confirmed that CF can purify water by attaching to microorganisms and sludge in water. In 2012, Professor Hori of Nagoya University, Japan along with TEIJIN LIMITED succeeded in scientifically elucidating the mechanism regarding the ease of adherence between microorganisms and CFs from an energy perspective as follows [57]:

1. The repulsion between negative charges acting between microorganisms and fibers is small when using CFs compared to other fibers.

2. The attractive force, “intermolecular force,” between the fibers and microorganisms is strong when using CFs compared to other fibers.

As mentioned above, mainly for pitch-derived CFs, the study on CF use in water purification materials has already been conducted in Japan [53, 58] and the product has been sold [59]. It is believed that from this good image of “biocompatible materials, water purification materials,” the environmental and ecological toxicity of CF may have been overlooked so far. As for technical solutions to prevent dust from scattering, wrapping with paper, surface coating, and slivers (cotton strips) are considered to be effective. In addition, we would like to propose the development of a dedicated capture and dust collection technology for CF dust/milled and the introduction of downdraft equipment that drains water to the ground. It is also effective to develop an excellent filter system that can recover floating CF dust/milled from water.

2-2 Assessment of the impact and toxicity on aquatic organisms caused by carbon fibers generated during CFRP recycling

2-2-1 Assessment of biological effects and harm to Japanese medaka due to carbonized CFs generated by a pyrolysis carbon fiber recycling process

2-2-1-1 Introduction

Carbon-fiber-reinforced plastics (CFRPs) are used as structural materials in the aircraft and automotive industries owing to their light weight and high strength. The global demand for CFRPs tripled between 2010 and 2020 and is expected to exceed 190 kt by 2050 [60]. Their application as an alternative to metals is expected to expand into many fields. Carbon fiber (CF) has unique functionalities, such as electromagnetic shielding [9, 10], depending on the composite resin that is combined. Thus, CF products, such as CFRPs, have attracted substantial attention and are used worldwide. However, it was recently reported that there is little scientific information on the safety of CFs [61].

In recent years, the environmental impact of microplastics has become a major issue worldwide. Specifically, the effects of the accumulation of plastics in living bodies and elution of chemical substances attached to the plastics are thought to be issues of concern. Therefore, the impact of microplastics on ecosystems has attracted attention in recent years [62], and the following risks have been reported. The intake of harmful substances adsorbed by microplastics may cause ecological damage [19, 20].

Low-density polyethylene plastics cause liver dysfunction in Japanese medaka (*Oryzias latipes*) [21]. The accumulation of 14 plastic particles in the intestines of sea turtles results in 50% mortality [22]. The rate of infectious diseases in coral species increased from 4% to 89% in the presence of plastics [23].

In contrast, there are few reports on the toxicity of CFs to ecosystems compared to that of microplastics; however, two remarkable reports should be noted by those planning for a circular economy, which is an industrial system in which products are reused, remanufactured, and recycled at the end-of-life stage.

According to Nakano et al. [51], the inhalation of CFs with diameters of 5 μm or more is unlikely to seriously affect human health. However, fine particles such as milled short fibers with diameters of less than 3 μm and lengths of less than 80 μm after treatment by industrial processes are likely to reach the human alveoli after inhalation.

According to Moriyama [52], when mice were fed CFs, the CFs were not acutely toxic, in contrast to carbon nanotubes and asbestos. However, the results of a gene expression study suggested that carbonized CFs with residual carbons, etc., on the surface may be mutagenic in the long term; this possibility was not confirmed in mice injected with recycled CFs having a clean surface and few impurities. Another report indicates that the transfer of milled CFs or CF dust to external water sources during manufacturing, processing, and recycling of CF-related products may be of concern [61].

The selection of sample organisms is an important first step in the observation of the effects of chemical substances on aquatic organisms. As a standard strain for the assessment of ecological toxicity, the Japanese medaka is an important species, especially in Japan [63]. Medaka (*O. latipes*, orange-red variety or himedaka) is a freshwater teleost fish that is widely distributed in the Japanese freshwater environment. Its habitat is typically near the water surface, where they are commonly seen during spring in Japan. Medaka is registered as an index organism for the assessment of the ecological toxicity of newly synthesized chemicals of which more than 10 ton/year are produced in Japan. A recently published medaka draft genome showed that humans and medaka share 70% of their genes.

Therefore, it is highly probable milled CFs or CF dust will ultimately result in the same problems as microplastics if they are released into the aquatic environment. Therefore, referring to studies on microplastics, we tested the ecotoxicity of CFs using Japanese medaka.

2-2-1-2 Materials and methods

2-2-1-2-1 Medaka culture conditions

Medaka were purchased by mail order over the internet from FOCUS (Kumamoto, Japan) and CHARM (Gunma, Japan) as a standard strain for the assessment of ecological toxicity. They were bred in tap water dechlorinated by a water conditioner. The fish were fed Tetra KilliMin from Spectrum Brands Japan, Inc. (Yokohama).

The purchased medaka were held for one week in a place other than the test aquarium (fish tank) for acclimatization. Medaka with deformities (such as backbone deformities) were eliminated, and finally 10 medaka per beaker were selected for the ecological toxicity test.

2-2-1-2-2 Ecological toxicity experiments

Ecological toxicity experiments were performed under two conditions. The semistatic experiment was performed according to the OECD TG 204 guideline: Fish, Prolonged Toxicity Test: 14-day Study [64] under CF inputs of 0 mg/L (the control), 5 mg/L, and 50 mg/L. Table 4 shows the detailed test conditions.

Table 4 Conditions for semistatic ecotoxicity experiment

Item	Experimental methodology and condition
Organism species	Japanese medaka, *Oryzias latipes* (orange-red variety or "Himedaka")
Test medium	De-chlorinated (chlorine removed) tap water
Test period	4 weeks (28 days)
Number of organisms	10
Test method	Semi-static test in accordance with OECD TG 204: Fish, Prolonged Toxicity Test: 14-day Study [64] (water is renewed continuously in the test chambers)
Test temperature	21–25 °C
Feeding frequency	Once a day
Number of tests	3
Measurement item	Number of deaths, abnormal shape, behavioral inhibition, food intake, water temperature and pH

For the static experiment, we set our own test conditions, referring to the OECD TG204 guideline, and used the same CF inputs as in the semistatic experiment. Table 5 shows the detailed test conditions.

Table 5 Conditions for static ecotoxicity experiment

Item	Experimental methodology and condition
Organism species	Japanese medaka, *Oryzias latipes* (orange-red variety or "Himedaka")
Test medium	De-chlorinated (chlorine removed) tap water
Test period	4 weeks (28 days)
Number of organisms	10
Test method	Static test (water is not renewed in the test chambers)
Test temperature	21–25 °C
Feeding frequency	Once a day
Number of tests	4
Measurement item	Number of deaths, abnormal shape, behavioral inhibition, food intake, water temperature and pH

The CFs placed in the aquarium consisted of primary heated product samples (carbonized CFs after partial firing treatment, where the fiber length was 0.2–3.0 mm) were obtained from Carbon Fiber Recycle Industry Co. Ltd. (Gifu, Japan).

2-2-1-2-3 Microscopic observation of medaka

After the death of the medaka, the epidermis and internal organs were observed with a phase-contrast microscope to check for the presence of CFs. Further, the internal organs of the medaka were excised, crushed, and examined for the presence of CFs.

2-2-1-2-4 Medaka transparency experiment

To observe the effects of the CFs more clearly, pigments were removed from the epidermis of the medaka using trypsin and alkaline procedures according to Yoshioka et al. [65]. Note that to stop the action of the trypsin, the trypsin was removed from the sample bottle, and the medaka were washed with water several times. Then the medaka were soaked in a liquid containing 0.5% KOH and glycerin.

2-2-1-2-5 Determining the number of colonies in aquarium water on standard agar medium

The water in an aquarium with surviving medaka was diluted 100-fold with phosphate-buffered saline (PBS) and applied to a standard agar medium. The medium was prepared by growing Saccharomyces cerevisiae for two days at 30 ℃ in a yeast extract-peptone dextrose medium (15.0 g of agar, 2.5 g of yeast extract, 5.0 g of peptone, and 1.0 g of glucose) [66].

2-2-1-2-6 DNA extraction

The DNA of microorganisms in the water was extracted using an Extrap Soil DNA Kit Plus, Ver. 2 (Nippon Steel Eco-Tech Corporation, Kisarazu, Japan) according to the manual [67, 68]. Water (1,000 mL) was collected from a beaker in which medaka were living and filtered through a membrane filter (pore size, 0.1 μm) manufactured by Advantec Toyo Kaisha, Ltd. (Tokyo, Japan).

2-2-1-2-7 Gene amplification

Gene amplification was performed by the polymerase chain reaction (PCR) using universal primers. The designed sequence of primers is shown in Table 6. The reagents were prepared using a KAPA kit (Kapa Biosystems, Inc., Wilmington, DE, US) according to the manufacturer's instructions [69]. The 16S rRNA gene V3-V4 region was amplified by PCR using 2X KAPA HiFi HotStart ReadyMix.

Table 6 Entire primer sequence

Area	Species	Sequence of the entire primer (Illumina adapter | index (for forward primer only) | Region-specific sequence) [70]	
		Forward primer	Reverse primer
16S	Bacteria	TCGTCGGCAGCGTCAGATGTGTATAAGAGACAG | index | CCTACGGGNGGCWGCAG	GTCTCGTGGGCTCGGAGATGTGTATAAGAGACAG| GACTACHVGGGTATCTAATCC

As a primer, an adapter manufactured by Illumina, Inc. (San Diego, CA, US) was added to 341fF805R (S-D-Bact-0341-b-S-17/S-D-Bact-0785-a-A-21) [70]. As the forward primer, a six-base index sequence [71] for sample separation was added between the adapter manufactured by Illumina, Inc. and each specific sequence.

2-2-1-2-8 Next-generation sequencer data analysis

The PCR amplification products were submitted to the next-generation sequencer service at Gifu University, which employs Illumina MiSeq (San Diego, CA, US).

First, using the obtained FASTQ sequence files, each sample was separated on the basis of the index sequence, and the primer and index sequences were removed. Next, for sequencing after separation, the forward and reverse sequences were combined, and quality filtering was performed using a Q value of 30 as the cutoff. Then, operational taxonomic units (OTUs) were formed by clustering based on 97% sequence similarity. Finally, chimeric sequences and low-frequency OTUs (less than four sequences in total) were removed.

The above series of operations was performed using the software package Claident v0.2.2017.05.22 (https://www.claident.org/). Representative OTU sequences were classified using RDP Naive Bayesian rRNA Classifier, version 2.11 [72].
During the classification, RDP 16S rRNA training set 16 was used as a reference database, and the 80% confidence threshold was used.

2-2-1-3 Results

2-2-1-3-1 Medaka survival rate

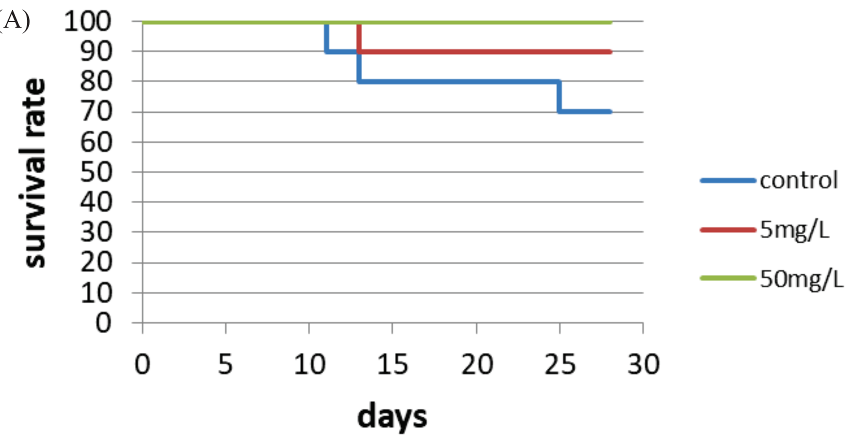
2-2-1-3-1-1 Survival rate of medaka under semistatic conditions

To examine the effect of CFs on ecosystems, we determined the effect of CFs in water on medaka. Therefore, on the basis of the standard ecotoxicity test OECD TG 204: Fish, Prolonged Toxicity Test: 14-day Study, we conducted an ecotoxicity experiment in which medaka were exposed to CFs for 4 weeks (28 days) under semistatic conditions. Under these test conditions, we continuously renewed the water in the aquarium.

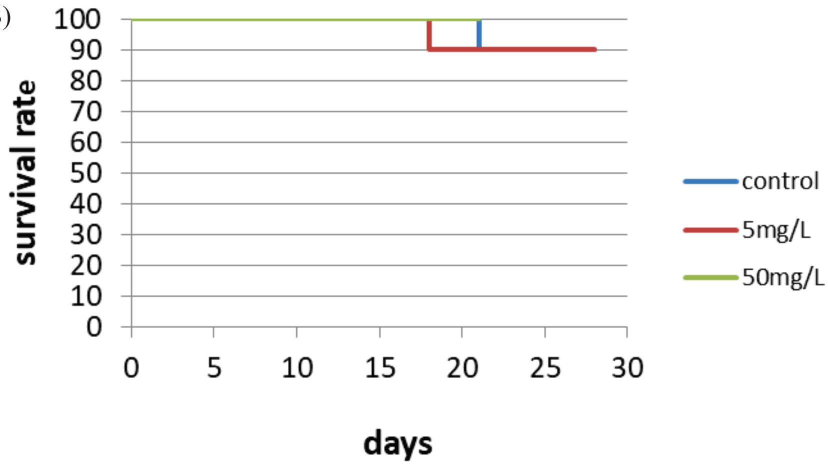
The graph in Fig. 5 shows the survival rate of the medaka for 28 days and that in Fig. 6 shows the results of the Log-rank test [73, 74] for all samples under semistatic conditions.
The p-values of the Log-rank test were all greater than 0.05, and the test results showed that there was no significant difference between the survival curve of medaka at CF inputs of 5 and 50 mg/L and the control samples.

Although there is some dispersion, the three experimental results under semistatic conditions do not differ significantly. Therefore, exposure to CFs is unlikely to affect medaka under such conditions.

(A)



(B)



(C)

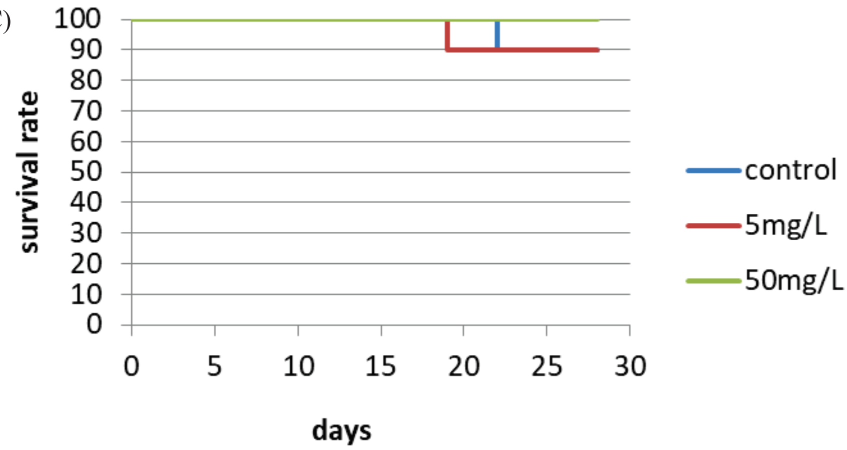


Fig. 5 Survival rate of medaka under semistatic conditions

(A) First experiment: after approximately two weeks, the number of surviving individuals decreased. Ultimately, there was no significant difference in the number of surviving individuals under both CF inputs.

(B) Second experiment: the number of surviving individuals began to change on day 17, and the number of surviving individuals was ultimately the same under all conditions.

(C) Third experiment: there were no significant differences in the number of surviving individuals.

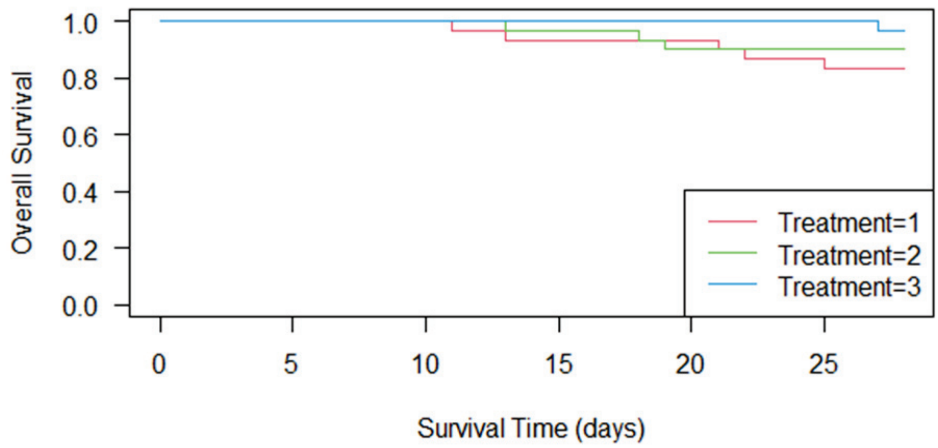


Fig. 6 Results of the Log-rank test of all samples under semistatic conditions

Treatment 1: control, Treatment 2: CF input of 5 mg/L, Treatment 3: CF input of 50 mg/L

Log-rank p-values for comparison between: CF inputs of 5 and 50 mg/L and control samples = 0.2,

CF input of 5 mg/L and control samples = 0.5,
CF input of 50 mg/L and control samples = 0.08, and
CF input of 5 mg/L and CF input of 50 mg/L = 0.3.
Note: In general, p = 0.05 or less indicates a significant difference.

2-2-1-3-1-2 Survival rate of medaka under static conditions

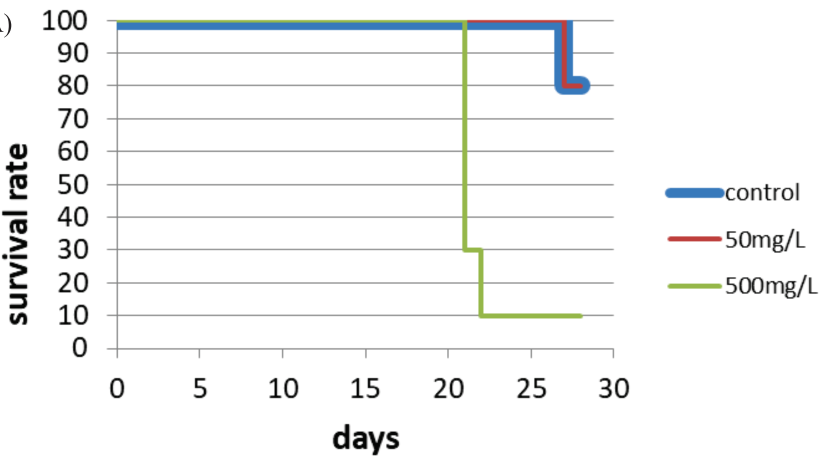
CFs are reportedly able to purify water [53, 56], which might be relevant for the experimental results obtained under semistatic conditions. If CFs have a purifying effect, it is thought that raising medaka under static conditions for a long period (4 weeks or more) may result in improved water quality and thus extend the life of the medaka. Thus, we conducted an additional experiment under static conditions.

Under static conditions, the survival rates of medaka at CF inputs of 50 and 500 mg/L were significantly lower than that of the control sample, as shown in Fig. 7. The survival rate of medaka at a CF input of 50 mg/L was lower than that under semistatic conditions, except for the first experiment.

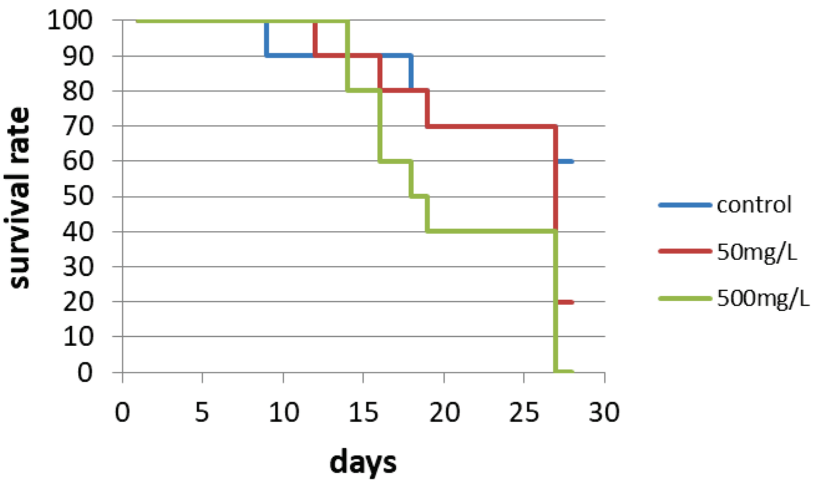
The Log-rank test p-values were less than 0.05, except for the comparison between CF inputs of 50 and 500 mg/L, as shown in Fig. 8.

The results of the survival time analysis (Log-rank test) of all samples showed that there was a significant difference between the survival curve of medaka at CF inputs of 50 and 500 mg/L and that of the control samples.

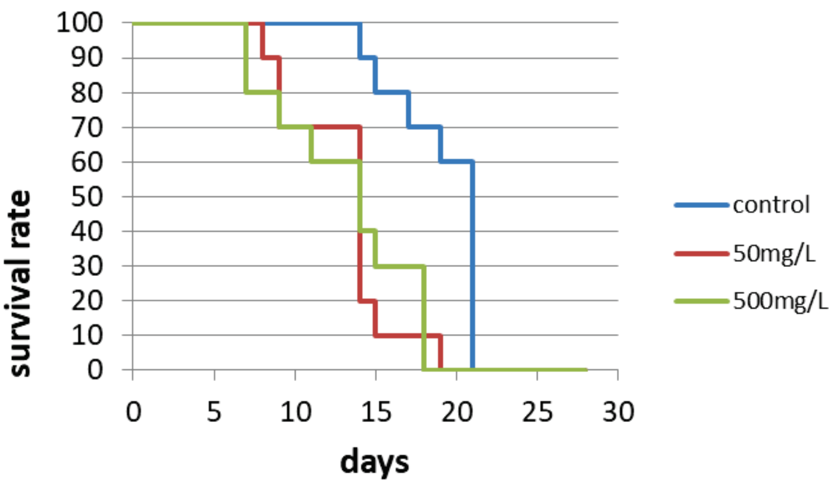
(A)



(B)



(C)



(D)

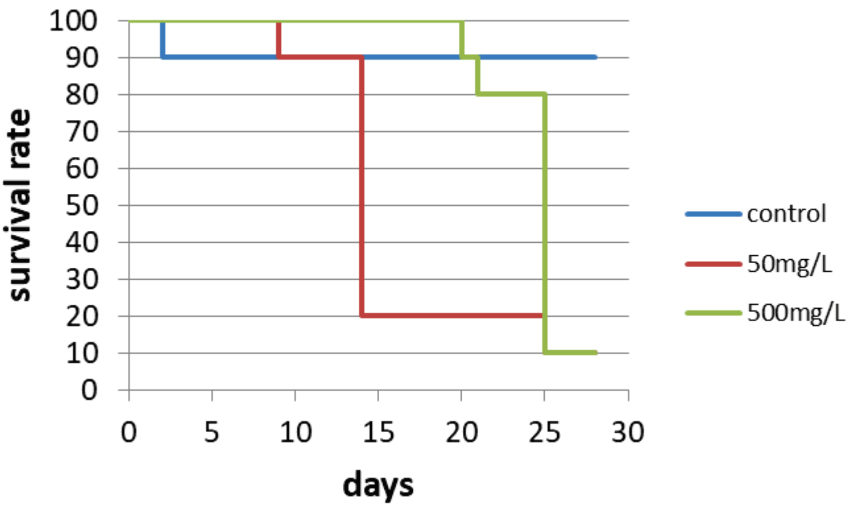


Fig. 7 Survival rate of medaka under static conditions

(A) First experiment: after approximately three weeks, the number of surviving individuals changed; ultimately, the number of surviving individuals was lowest at a CF input of 500 mg/L.

(B) Second experiment: after approximately two weeks, the number of surviving individuals at a CF input of 500 mg/L decreased sharply.

(C) Third experiment: after approximately one week, the number of surviving individuals decreased sharply at CF inputs of 50 and 500 mg/L.

(D) Fourth experiment: after approximately 10 days, the number of surviving individuals decreased sharply at CF inputs of 50 and 500 mg/L.

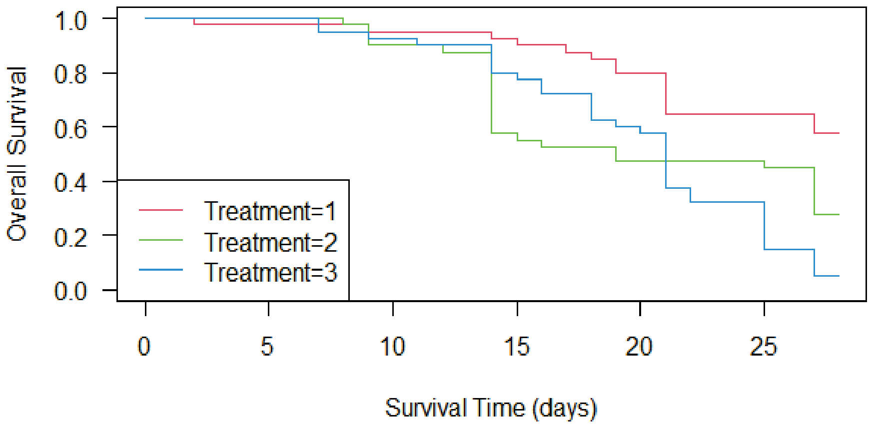


Fig. 8 Results of the Log-rank test of all samples under static conditions

Treatment 1: control, Treatment 2: CF input of 50 mg/L, Treatment 3: CF input of 500 mg/L

Log-rank test p-values of comparison between: CF inputs of 50 and 500 mg/L and control samples

= 2e-05,

CF input of 50 mg/L and control samples = 0.004,

CF input of 500 mg/L and control samples = 2e-06, and

CF input of 50 mg/L and CF input of 500 mg/L = 0.06.

Note: In general, $p = 0.05$ or less indicates a significant difference

This result suggests that CF exposure is lethal to medaka under static conditions. Moreover, for the control samples, the survival rate under static conditions was evidently lower than that under semistatic conditions. This possibly occurred due to the contamination of the growing environment (i.e., changes in the microbiota) under static conditions and in the CF input conditions the degree of the contamination was not lower than that of CF free conditions. Therefore, the medaka mortality under static conditions is attributed in part to the CFs.

2-2-1-3-2 Presence of CFs in medaka epidermis, viscera, and feces

CF exposure is lethal to medaka under static conditions. Thus, the CFs must come into contact with the medaka. We observed the CFs in or on medaka. To observe how CFs penetrate the medaka, CFs were placed in a medaka aquarium. The results demonstrated that needle-like CFs had adhered to the entire skin of the medaka, as shown in Fig. 9. The attached CFs may have direct physical effects or systemic effects on the medaka.

CFs were observed in the viscera of the medakas at CF inputs of 50 and 500 mg/L. This result indicates that the medaka ingested CFs with food or mistakenly ate CFs as food; it is thought that some of the ingested CFs remained in the body, and the rest were excreted as feces. As expected, CFs were not present in either the viscera or the feces of the control samples, as shown in Fig. 9.

Oshima et al. [75] investigated the pharmacokinetics of medaka using virgin microplastic (polystyrene, 2–200 μm) and reported that the microplastic that entered the body was excreted in approximately 1 day. However, our experiment demonstrated that CFs are less likely to be discharged from the body.

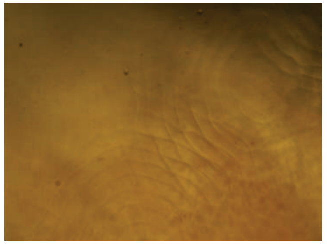
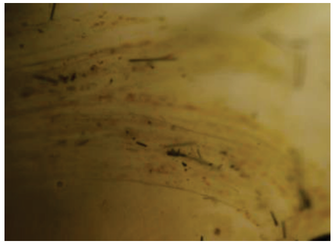
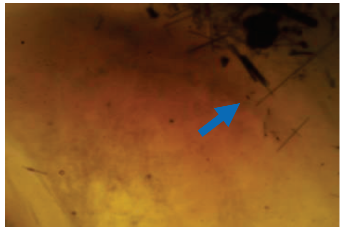
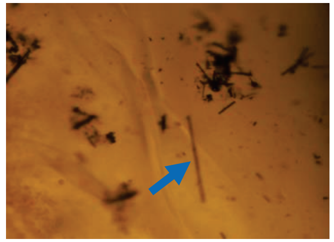
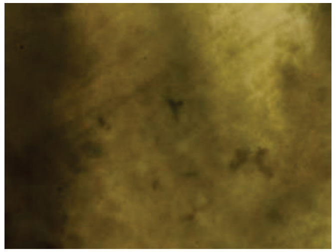
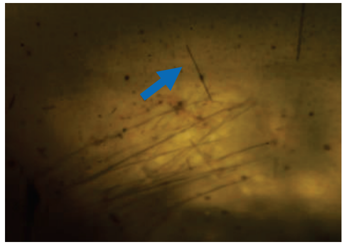
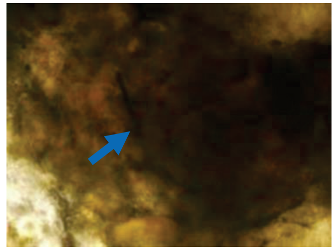
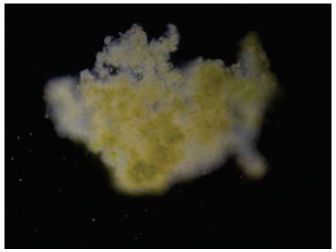
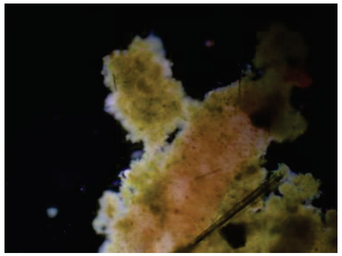
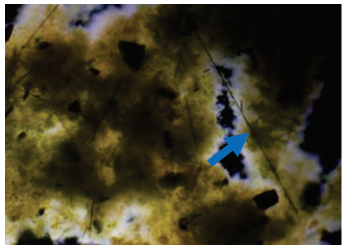
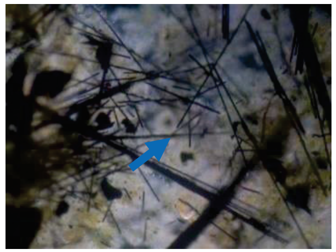
	Semi-static conditions in accordance with OECD TG 204		Static conditions	
	Control	CF 5mg/L input	CF 50mg/L input	CF 500mg/L input
epidermis				
viscera				
feces				

Fig. 9 Observations of CFs　Arrows indicate examples of CFs adhering to or penetrating the medaka.

2-2-1-3-3 Medaka transparency experiment

To observe the CFs on the medaka epidermis more clearly, a medaka from the experiment with a CF input of 50 mg/L was transparentized according to a general preparation procedure [65]. Consequently, the appearance of CFs adhering to the skin of the medaka became clearer, as shown in Fig. 10.

The results in sections 3.2 and 3.3 indicate that CFs adhere to the body surface of the medaka and that the medaka take CFs into their bodies. We assume that over the long term, injuries would appear, followed by infections at the wound site, and finally the medaka would die.

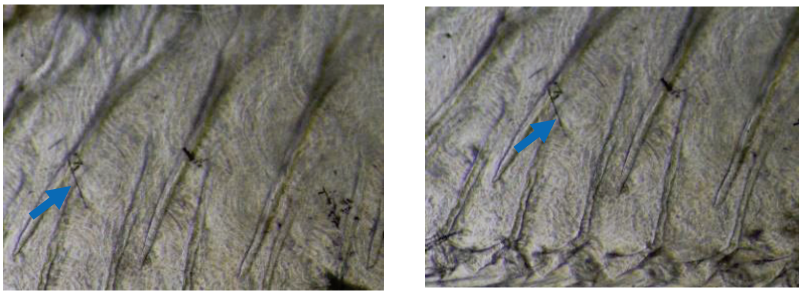


Fig. 10 Observations of medaka epidermis (caudal fin) after transparency processing (50 mg/L CF input under static conditions)
Arrows indicate examples of CFs adhering to or penetrating the epidermis.

2-2-1-3-4 Contamination of medaka aquarium by bacteria

An apparent difference between the semistatic and static conditions is the water quality. The static water appeared to be contaminated by microbes. To examine the contamination of the medaka aquarium, we measured the number of colony-forming unit (CFUs) on a standard agar medium.

Water (100 µL) from the medaka aquarium was applied directly to a standard agar medium. Except on day 0, the samples were diluted 100-fold with PBS, and the CFUs were counted after two days of incubation at 30 °C.

Fig. 11 shows the number of viable bacteria under static conditions. Compared to that on day 0, the number of viable bacteria increased for 7 days and then remained constant; all three samples showed similar behavior over time.

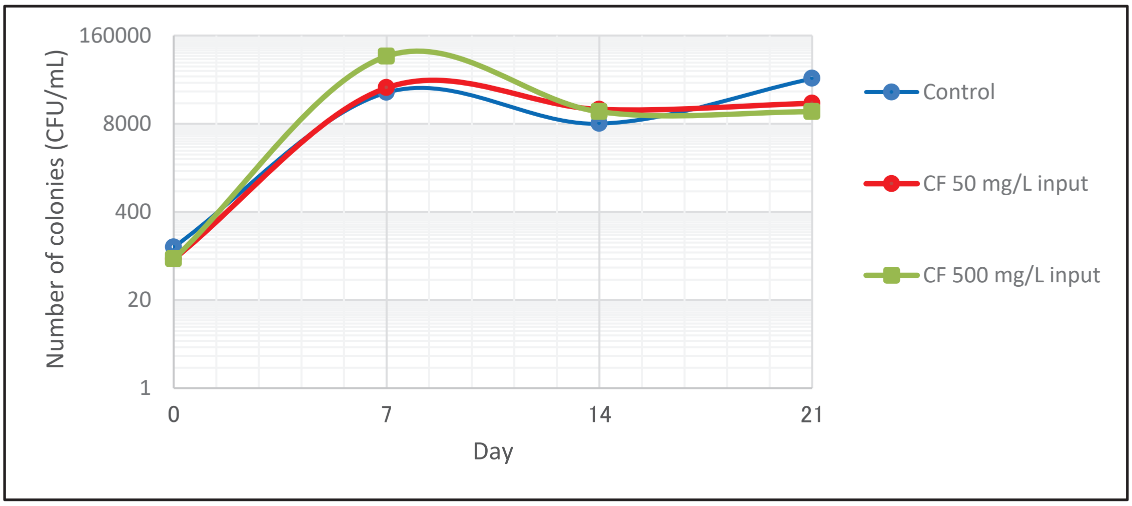


Fig. 11 Number of viable bacteria under static conditions.

2-2-1-3-5 16S rRNA gene analysis

The medaka aquarium under static conditions is clearly contaminated with bacteria. We analyzed the bacterial flora. Fig. 12 shows the structures of the bacterial community at the genus level for each experiment. The genus names of each bacterium are given on the right side of the figure. Colors indicate different genera of bacteria.

To elucidate the effect of CFs attached to the body surface and inside the body of the medaka, water was sampled from each medaka aquarium with CF inputs of 0, 50, and 500 mg/L under static conditions, and the 16S rRNA gene was analyzed.

The results revealed changes in the microbiota of all the samples, and the flora were found to become more complex over time. That is, the water was initially a single ecosystem, but over time, the numbers of various microorganisms increased. This result indicates that there was a risk of infection.

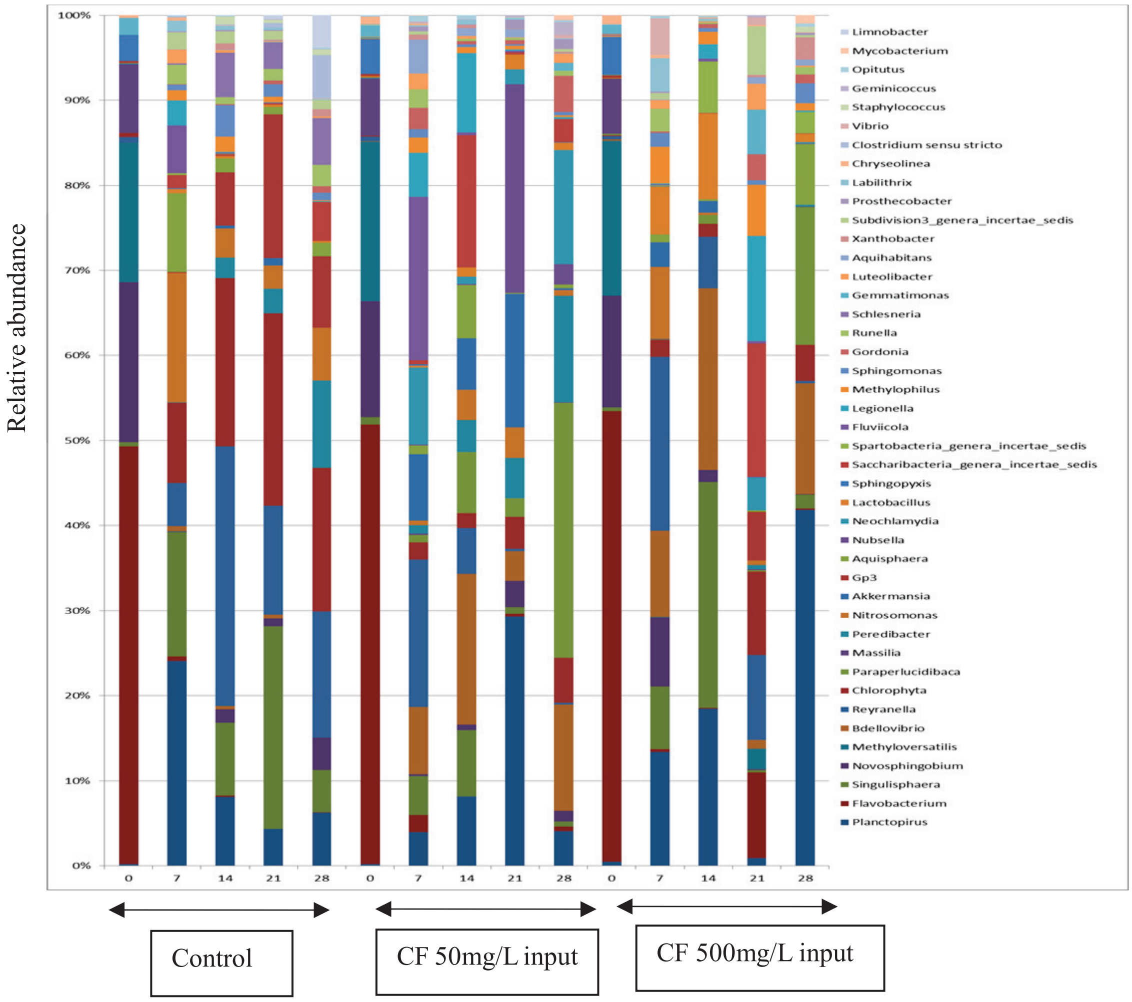


Fig. 12 Microbiota analysis results under static conditions.
Bar plots of the taxonomic profiles of each sample at the genus level.
Colors show the relative abundance of each genus.

2-2-1-4 Discussion

The carbonized CFs used in this experiment were 0.2–3.0 mm in length; thus, they were longer than those used by Nakano et al. [51], which were less than 80 μm in length. However, adverse effects on some medaka were observed.

Under semistatic conditions, none of the obtained data indicated that the CFs affected the survival of medaka. We may conclude that the mortality was low under these conditions.

By contrast, under static conditions, the number of surviving medaka decreased remarkably. The number of surviving medaka at a CF input of 50 mg/L was much lower than that under semistatic conditions. This result shows that the suspension of CFs in the water is not always environmentally safe. As the CFs had little effect under semistatic conditions, the water quality of the samples may affect the medaka health.

We observed dead medaka and confirmed that CFs were attached not only to the epidermis and viscera but also to the feces of the medaka, and the CFs were needle-like and easily adsorbed. This result indicates that the medaka ingested CFs along with food or mistakenly ate CFs as food; it is thought that some of the ingested CFs remained in the body, and the rest were excreted as feces. The CFs in the bodies and gastrointestinal organs were similar in shape to the CFs before exposure, and they could penetrate the medaka body and organs.

Bacterial infections can occur where CFs enter the body, and these may be fatal. The microbial number increased for 7 days under static conditions, and the microbiota became diverse. These results support an increase in bacterial infections. Therefore, they suggest that in environments where the water is stagnant, CFs can be considered to be actively toxic to medaka.

It is not uncommon for fish to be injured by objects in the water in a real environment. Therefore, depending on the growth environment, wounds are assumed to be fatal. Further studies are needed to determine whether and to what extent CFs that are released into the environment will increase the number of wounds to fish.

Herein, we apply our experimental results to natural environments. It can be assumed that semistatic conditions occur in rivers, whereas static conditions occur in areas of enclosed water, such as enclosed gulfs, lakes, and reservoir. Rivers are typically in flux and the water is changing; however, enclosed water shows little change, and the water is stable.

In rivers, it is possible that CFs accumulate near the banks and in deep pools where the water is calm; however, most CFs are expected to flow easily. By contrast, in lakes and reservoirs, because there is almost no flow, CFs are likely to accumulate.

Furthermore, in terms of hygienic analysis, the relationship between the survival rate of medaka and the CF input was analyzed using the difference in the microbiota of the water in the medaka aquarium.

Flavobacterium is a pathogen of freshwater fish, and Flavobacterium columnare causes infectious diseases such as tail diseases (for example, frayed and ragged fins), which is known as columnaris disease in warm-water fish such as medaka. In this experiment, the proportion of Flavobacterium was low except on day 0. This result suggests that tail disease is not likely to be the main cause of medaka fatality.

Mycobacterium, which causes swimming disease, was also found at CF inputs of 50 and 500 mg/L, but not in the control samples. During the observation of the medaka in the aquarium, swimming disease was confirmed several times. It is therefore highly likely that Mycobacterium is one of the causes of medaka mortality.

In summary, it is highly probable that the reduction in the number of living medaka under static conditions resulted from the presence of CFs, which increased the occurrence of infections. However, we do not expect a CF concentration of 50 mg/L in our actual environment under normal circumstances, and in practical situations, injured fish account for a small proportion of all medaka.

In addition, as medaka mortality can be affected by various factors including bacteria, parasites, and physical effects, we conclude that it is difficult to clarify a specific cause of death in terms of infectious disease.

2-3. Measurement of airborne carbon fibers at SME's site where CFRPs are handled

2-3-1 Methods of air sampling

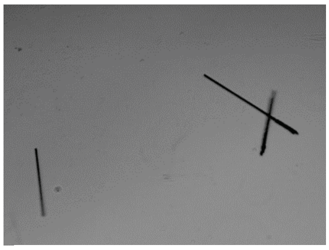


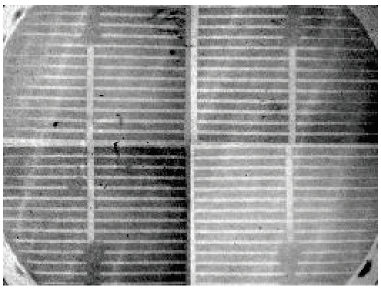
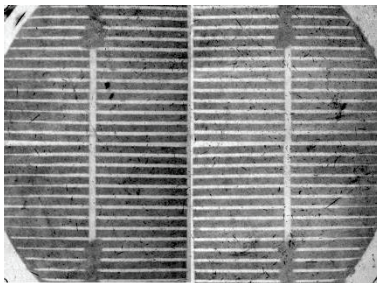
Photo of airborne CFs

・Location: Laboratory, Moritomi Environmental Engineering Laboratory at Gifu prefecture, Japan
・Measurement date: 2021.3.29, 30
・Equipment: Particle counter (Lafil400)
・Flow rate: 23 L/min (60 Hz)
・Measurement hours:12 H
・Carbon fiber was collected on a membrane filter (4.5 cm in diameter, 1 μm in mesh)
・Work in the laboratory: Cutting, rubbing, bending, and twisting of recycled carbon fibers to make twisted yarn.
・Room environment: Windows basically closed, four commercial air purifiers running all day.
・Equipment location: on the floor

Lafil400

2-3-2 Results of data analysis
Analysis conducted by AIST Chubu Center

Table 7. Results of the measurement of the number of airborne carbon fibers in the laboratory atmosphere

	Measurement date: afternoon of 2021/03/29 Carbon fiber related work was <u>not</u> conducted in the morning of the day.	Measurement date: afternoon of 2021/03/30 Carbon fiber related work was conducted in the morning of the day.
Photo of membrane filter after image correction		
0.3μm	1504792	1599565
0.5μm	239544	265169
1.0μm	13245	15296
2.0μm	1084	2109
5.0μm	644	2054
10.0μm	215	893

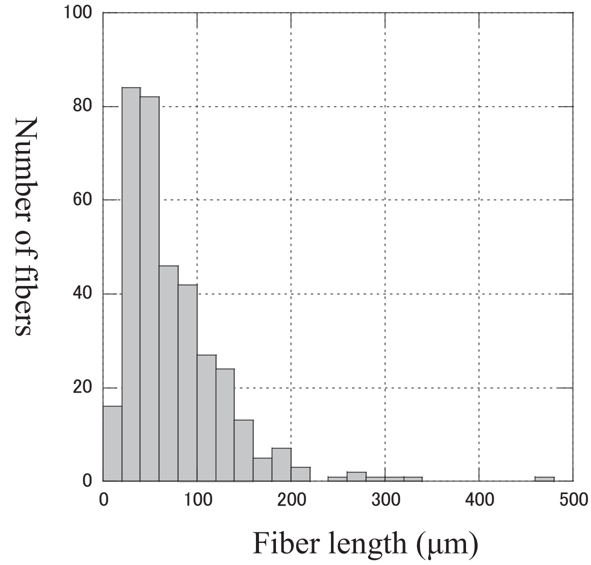


Fig.13 Measurement result of the number of carbon fibers on the membrane filter used on 30 March 2021 (automatically analyzed by software)

The membrane filters measured on 30 March 2021, just after the carbon fiber related work was done, were observed by a stereo microscope, and after image correction, the fiber length was identified.

As a result, carbon fibers (long and thin needles) were recognized.
Most of the carbon fibers were 20 to 60 μm in length, and the maximum length was about 0.5 mm (500 μm) as shown in Fig. 13.
However, there were also many dust particles other than carbon fiber, such as dust and dirt.

2-4. Study on risk management approach for recycled CFRP manufacturing/handling sites using control banding method

2-4-1 Introduction

CFRP is mainly used for structural materials in the aircraft and automotive industries due to its light weight, high strength, and other properties. As an alternative to metal and glass fiber composite materials, CFRP is expected to be used in many future applications such as wind power generation [76] and flying cars, and the amount of CFRP used is expected to increase year by year, with demand exceeding 190,000 tons/year by 2050. [2]
However, the recycling of CFRP is currently not progressing well, especially in Japan, and almost all of the waste CFRP is disposed of in landfills, which is considered to be a problem.

In addition, in response to the recent trend toward decarbonization, some shareholders and others are questioning the existence itself of the CFRP manufacturing industry, which emits a large amount of CO_2 during production. [77]

Given these circumstances, in order to further expand the use of CFRP in the future, it is strongly desired to reduce the environmental impact of the entire value chain from production to recycling by converting CFRP into recycled products. [78]

Carbon fiber (CF) (primary heated product) extracted from the pyrolysis process, which is currently the leading technology for CFRP recycling, contains a large amount of impurities such as tar, and there are concerns about its impact on workers and the ecosystem compared to virgin products. The possibility of unintentional containment of impurities such as glass fiber, metal foil, and dust has also been pointed out in research reports by AIST [40], [79].

Ueda et al.[61] summarized the issues to be addressed to increase social acceptability and promote CFRP recycling and stated that it is necessary to resolve concerns about the biological and ecological effects of CF dust released from recycling-related operations at workplaces and other sites in the future.

Moreover, Moriyama et al.[52] had conducted cell tests using recycled CF artificially finely ground in a ball mill and reported that CF can be finely ground down to several 10 nm by prolonged grinding in a ball mill.

Through measuring by dynamic light scattering (DLS), they reported that with proper fractionation and centrifugation, it is quite possible to refine CFs to 100-300 nm in diameter, some of which are several 10 nm in diameter.

Given the fact that CF can be made into nanoscale materials under artificial conditions, it can be said that if recycling technology is further advanced in the future [81] and CF is used and utilized over and over again, it may eventually be micronized to the nano-level.

Therefore, in recycled CFRP, it is desirable to conduct control measures at the nanoscale level, rather than at the microscale level normally found in virgin products.

In recent years, studies in Europe and the United States have reported the formation of NOAAs (nano-objects, and their aggregates and agglomerates greater than 100 nm).

The composition of NOAA is shown in Figure 14 [82].

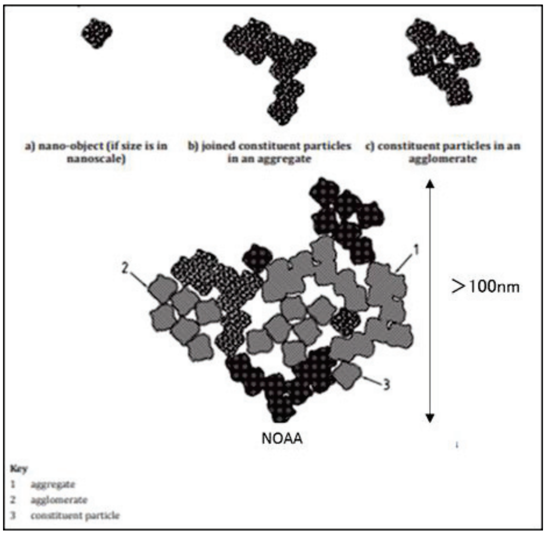


Fig. 14 Composition of NOAA[82]

For example, with regard to occupational exposure to NOAA, studies have been conducted on the occupational exposure risk of airborne NOAA [83]-[87] and measurements of NOAA suspended in actual factories [88]-[92]. The results of actual sampling show that NOAA was present in more than 70% of the samples [89]. It has also been reported that NOAA is formed not only in air but also in liquids such as cell culture medium [93]. Moreover, with regard to industrial products, the effects of TiO2 nano bjects and NOAA on microorganisms under UV irradiation [94], the dermal exposure of NOAA transferred to humans via sweat from antimicrobial textiles containing silver nanoparticles and UV absorbing textiles containing TiO2 nanoparticles [95], and the dermal exposure of NOAA transferred from TiO2 nanoparticles. A wide range of studies have been conducted, including an evaluation of the release potential of NOAA from paints containing NOAA [96] and a study of the release potential of NOAA from wood products treated with coatings containing nano-objects [97].

However, these studies do not include studies on the conversion of CF, including CFRP, to NOAA.

As a noteworthy Japanese move, the Labor Standards Bureau of the Ministry of Health, Labor and Welfare issued a notice "Preventive Measures to Prevent Exposure to Nanomatearials" in 2009 [98].

In this notice, it is clearly stated that exposure control measures should be taken for dust generated during operations involving the handling of nanomaterials.

CF is inherently a brittle material, susceptible to breakage and frying (dust).

These characteristics are more represented in recycled products, which are inferior in strength to virgin products.

Since agents such as sizing agents that modify the surface have been removed, it has the negative characteristics of fiber breakage and frequent flyings during processing.

Therefore, dust control measures in workplaces related to recycled CFRP should be more stringent than the current measures for virgin products because of concerns about the impact aspects on people and the environment.

2-4-2 Current worker safety management measures

Figure 15 [99] shows a diagram that systematically organizes the manufacture, use, and disposal of recycled CFRP from the perspective of the entire life cycle.

In the life cycle of recycled CFRP, some relevant studies have been conducted on the effects of particulate CF on humans (workers) and the environment, including gene expression analysis [52] using animals. However, there are still many unknowns and concerns [100].

Although different from CF, there have been relatively few studies in the evaluation of toxicity related to nano carbon materials.

For example, Coyle et al. [101] evaluated the cytotoxicity of products incorporating carbon nanotubes as fillers or additives into thermoplastics for potential pathological effects due to particulates released upon recycling or disposal, especially incineration.

The results show endocytosis, but no acute cytotoxicity, and that further investigation is needed.

At CFRP handling sites, dust dispersion is a major bottleneck with respect to the impact on people (workers).

For reference, Figure 16 shows a photograph of dust control measures at a public research institute that conducts trial production and press working tests of recycled CF nonwoven fabrics.

In particular, in the trial manufacture of non-woven fabrics using recycled CF, a large amount of CF dust is generated by carding (combing) and other processes, so the workers always wear personal protective equipment (PPE) that covers their entire body.

In a past METI-Chubu project, survey about recycling CF-related work sites of domestic companies was conducted. As a result, it was found that countermeasure methods against CF dust vary depending on the size of the company and other factors. [102]

In many cases, large companies, such as materials manufacturers and aircraft manufacturers, have completely isolated workers from direct contact with CF dust.

On the other hand, in small and medium-sized companies, including venture companies, the measures taken varied greatly from company to company, and in many cases, measures such as natural ventilation with open windows and doors were found to be passive measures with minimal capital investment.

In particular, during the summer, there were many sites where CF-related processing work, such as grinding and cutting, was performed with windows and doors fully opened.

Considering the above situation, it is important to develop product technology such as dust prevention technology by surface coating treatment of materials in consideration of the expected increase in recycled CFRP-related businesses.

However, it would be desirable to have guidance that can be used as a reference when small, medium, and micro enterprises implement dust control measures onsite.

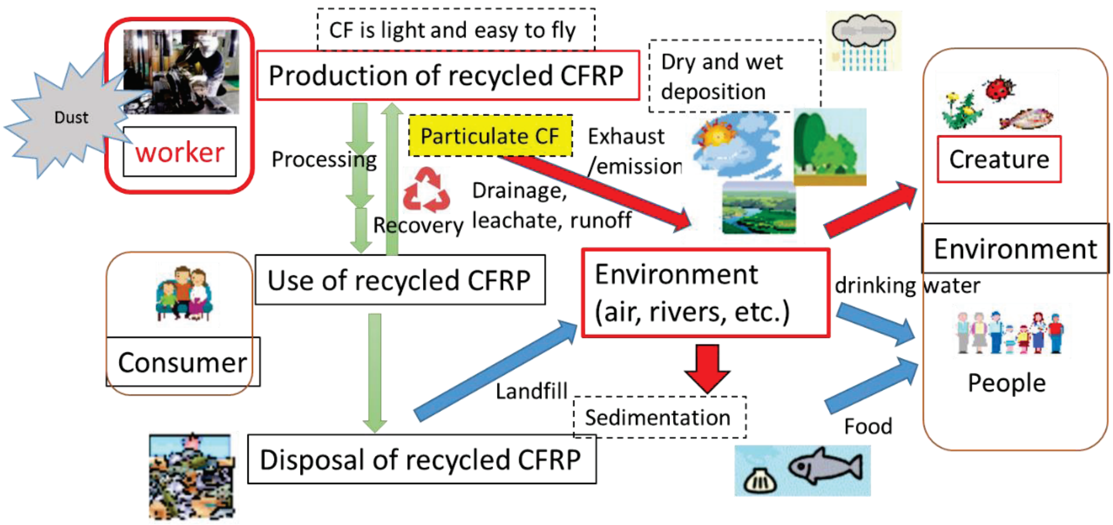


Fig. 15 Production, use, and disposal of recycled CFRP from the perspective of the entire life cycle (red arrows indicate areas of concern) [99]

Fig. 16 Photograph of dust control measures at a public research institute

2-4-3 Proposal for a worker safety management method using control banding

In a nutshell, control banding is a risk management (=control) method based on classification (=banding).

Originally, COHSS Essentials was developed by the Health and Safety Executive (HSE) [103] in the UK in 1998 for SMEs, and later the International Labor Organization (ILO) arranged it as a toolkit for SMEs in developing countries. The main advantage is that it enables a simple risk assessment of the health hazards of chemicals without specialized chemical knowledge.

This method has been used previously for aerosols containing ultrafine particles (welding fumes, carbon black, viruses, etc.).

The target is occupational exposure to unknown hazardous substances whose toxicity is unknown or uncertain and for which quantitative exposure estimates are not available.

The main feature of this method is that highly skilled measurement of exposure concentrations of chemical substances is not required.

However, it should be noted that this is only a guideline-level risk management method and is not legally binding, that it only covers inhalation exposure of workers, and that it assumes normal on-site work such as maintenance and cleaning, and does not consider accidental incidents and accidents.

Controlled banding of nanomaterials (NOAA) in industrial working environments is specified internationally in ISO/TS 12901-2 (2014) [104].

It covers NOAAs that can be unintentionally released throughout their life cycle.

This control banding approach for NOAA, like the case for common chemicals, uses Hazard Bands (HB) and Exposure Bands (EB) to identify the level of risk to workers at the nanomaterials handling site and presents appropriate control measures.

With regard to research trends in control banding, Kuijpers et al. [105] and Bekker [106] have conducted studies for further development of control banding tools, including validation of existing NOAA exposure assessment models, which tend to be conservative.

Bekker [107] discusses advanced modeling approaches for quantitative exposure estimation, the second step after applying control banding.They also evaluate the effectiveness and applicability of the "Advanced REACH Tool-ART" software.

The banding procedure for NOAA is shown in Figure 17.

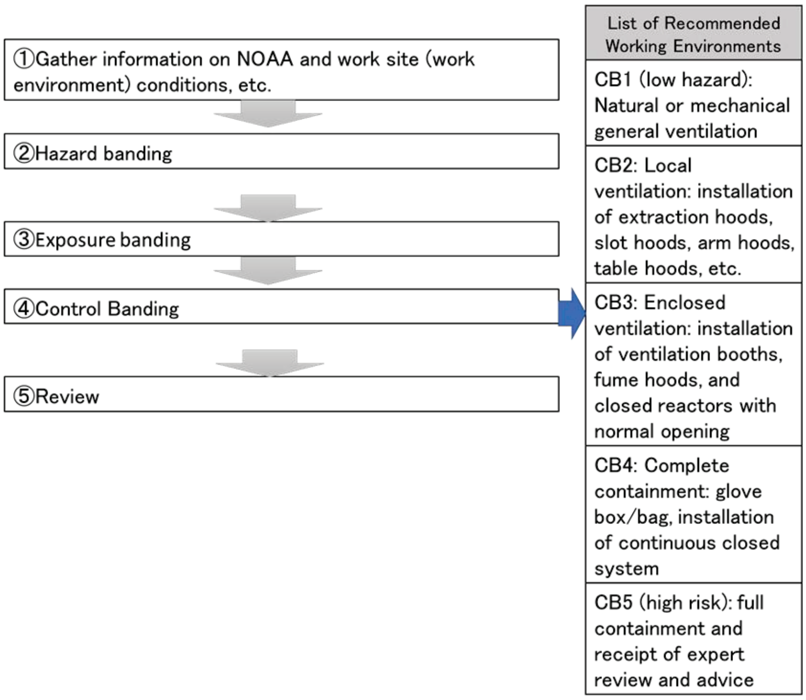


Fig. 17 Banding Procedure

Note that in the hazard banding in (2), various factors such as in vivo persistence, ease of particles reaching the airways, ease of deposition in the airways, and biological response, as well as toxicity, are taken into account in assigning hazard levels for the NOAA in question. The bands will be classified based on the toxicity of basic, similar known chemicals.

In the exposure banding of (3), the exposure potential to humans shall be assigned, taking into account the ease of airborne release of the target NOAA.

Based on recent research results on dust generated in various recycled CFRP-related operations, such as the firing process, paper making process, cutting, polishing, and machining of recycled CF, it is undeniable that the dust may change from micro-scale to nano-scale.

The possibility of formation of NOAA by nano scaling cannot be denied at the same time.

NOAA is currently being discussed in international forums.

For example, ISO/TC229 (Nanotechnology)/WG3 (Environment and Safety Subcommittee) is engaged in activities related to standardization and standardization of methods for measuring airborne NOAA amounts, etc. In this activity, it is also stated that NOAA will become a target for control in the workplace and environment [108].

In spite of the above situation, at present, no consideration has been given to the nanoscaling of CF dust in the process of recycling and other industrial processes, and to the conversion of CF dust to NOAA.

Therefore, the application of NOAA's controlled banding approach in an industrial work environment to recycled CFRP-related work sites is useful in terms of improving worker safety management aspects.

2-4-4 Examples and results of control banding applications in the field

In this study, we attempted to apply NOAA's control banding to the CFRP recycling operations of Company A, a SME located in Gifu Prefecture.

Company A is mainly engaged in removing CFRP from waste CFRP products using the calcination method, and has 10 primary pyrolysis furnaces and one secondary carbonization furnace in its plant. In addition, the removed CF is bagged into flexible container bags by hand.

The banding procedure is shown in Figures 18 [104],19[104] and Table 8.

Table 9 shows the results of the comparison with the current management measures.

For reference, Figure 20 shows the measurement results of particle size and particle size distribution of CF dust in the laboratory of a SME that actually conducts processing operations such as fiber opening using recycled CF manufactured by Company A.

In this case, a particle counter Lafil 400-LF30 (Rocker Scientific) was used to measure for 12 hours.

Figure 20 shows that although the data is not from the actual Company A work site, there are some particulates below 0-20 μm that could be categorized as NOAA. In the future, it would be desirable to further improve the sampling accuracy to cover the nanoscale and further investigate the presence or absence of NOAA.

As a result, there is a difference between the content of the control measures recommended from the application of control banding (CB-5) and the content of the control measures of the current Company A (CB-2), which has already obtained ISO 14001 (environmental management system) and ISO 9001 (quality management system) in 2020.

Currently, Company A is conducting measures ranging from general ventilation to the installation of local exhaust ventilation, however the control measures recommended by the banding this time present complete containment (total isolation) so that workers do not come into contact with any CF dust. This result was consistent with the real situation of the work environment in which workers are exposed to CF dust every time when they remove recycled CF from the furnace and pack it in bags.

Therefore, the results of the application of NOAA's control banding to a CFRP recycling-related work site indicate that further studies are needed for other sites, and that it does not present an ideal control method when considered in total, including cost-effectiveness, etc. However, considering its advantages, such as the fact that measures can be easily studied without measurement, control banding is a useful risk management method.

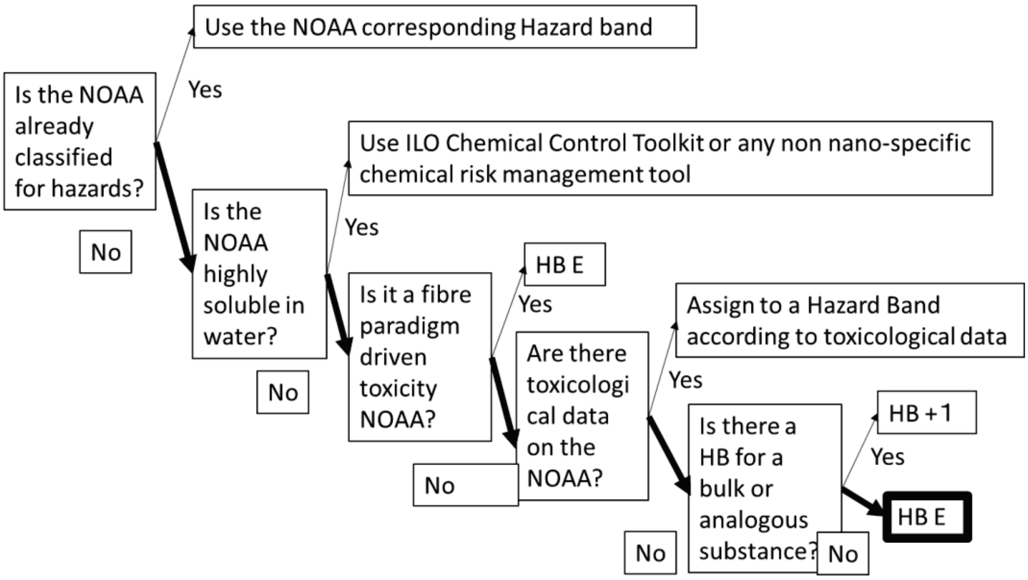


Fig. 18　Hazard Band (HB) assignment [104]

The term "fiber paradigm" refers to the physiopathological mechanisms that cause asbestos-like mesothelioma.CF is not an asbestos-like WHO fiber (inhalant fiber (WHO fiber definition): "a fiber that is at least 5 μm long, less than 3 μm in diameter, and with an aspect ratio (length to diameter ratio) of at least 3:1"), so the answer is "No"

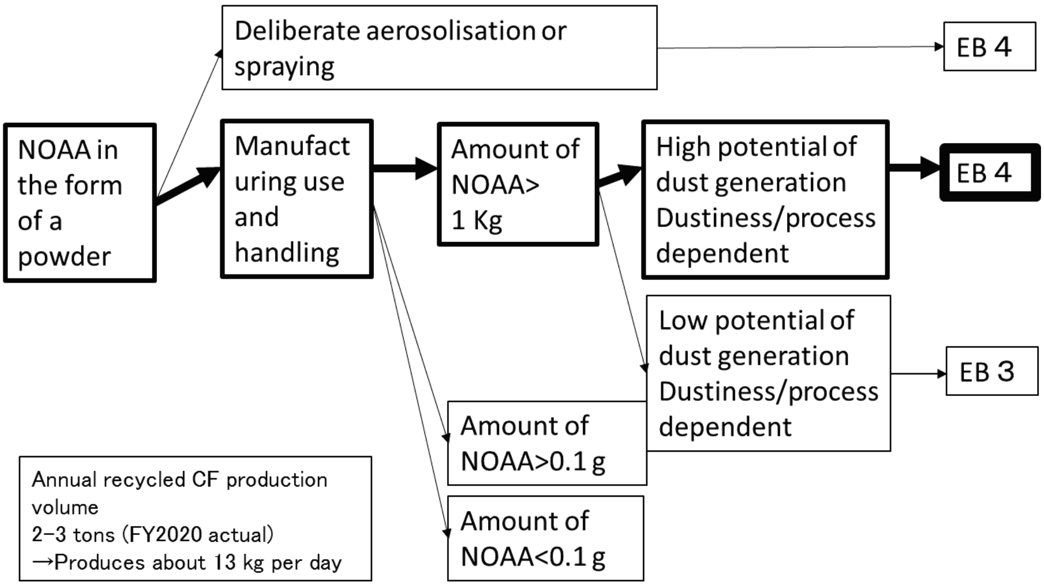


Fig. 19　Exposure band (EB) assignment [104]

Table 8.　Control band (CB) fitting

Hazard Band	Exposure Band			
	EB-1	EB-2	EB-3	EB-4
HB-A	CB-1	CB-1	CB-1	CB-2
HB-B	CB-1	CB-1	CB-2	CB-3
HB-C	CB-2	CB-3	CB-3	CB-4
HB-D	CB-3	CB-4	CB-4	CB-5
HB-E	CB-4	CB-5	CB-5	CB-5

Table 9.　Results of applying NOAA-covered control banding to Company A
(Comparison with current management measures)

Company A's current management measures (ISO 14001 and 9001 certification in 2020)	Control measures recommended as a result of the application of control banding
・The plant as a whole has 11 ventilation fans (total ventilation system) installed in the ceiling. ・Local exhaust ventilation has already been installed in the secondary firing furnace. ・The CF is constantly exposed to CF dust at the removal of CF from the secondary firing furnace (bagging into flexible container bags). Currently, CB−2 level is conducted	Complete containment, and receive professional review and advice. The recommended measure is the CB−5 level.

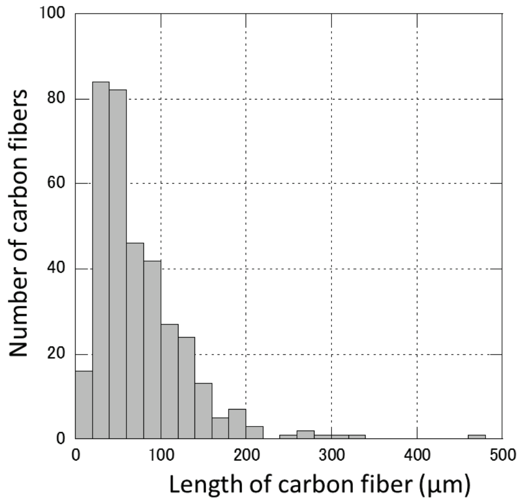


Fig. 20　Results of air sampling in the laboratory where processing work is performed using recycled CF manufactured by Company A.

2-4-5 Discussion and Conclusion

In response to the recent trend toward decarbonization, it is important to recycle CFRP, which emits a large amount of CO_2 during manufacturing process in virgin products, in order to exist as a material.

The current recycling technology [109] is basically a down-cycle, and the fiber length of CF gradually shortens with each repeated recycling.

And eventually, it may be micronized to nanoscale particles to form NOAA, based on a discussion of recent research results.

However, toxicity derived from the nanosize of some nanomaterials has been reported in recent years, raising concerns about their impact on human health and the environment, and methods for evaluating their safety remain unestablished [110].

Through studies by the Health and Safety Executive (HSE) [103] in the UK and the National Centre for Occupational and Environmental Research [111] in Denmark, it has become clear in recent years that nanoscale particles do not exist on their own but polymerize to form NOAA, and their properties, including toxicity, are still poorly understood.

In such a worrisome situation, proactive implementation approach before exposure is more important than retroactive implementation approach.

Currently, not enough research has been done on CF, including CFRP, to evaluate toxicity, including human and animal studies.

In this controlled banding, the lack of toxicity data led to a risk assessment on the safe side and the most conservative control measures, which resulted in the recommendation of complete isolation. The full isolation measure is difficult to implement in practice for SMEs due to the high capital investment, and as a result, they tend to wear PPE that covers the entire body.

As everybody can easily imagine, PPE that covers the entire body is cumbersome to wear and makes work difficult, especially in the summer.

Therefore, further research on toxicity assessment of CF, including CFRP, will be needed to improve the accuracy of recommended control measures when applying control banding.

The application of control banding to the work site where recycled CFRP is handled, as described in this report, has several advantages, including the fact that sampling (measurement) of exposure concentrations, which requires advanced technology, is not necessary, and the study of countermeasures is easy to perform.

The simplicity and accuracy of the method make it useful for SMEs to consider control measures, and we recommend its application to SMEs.

Finally, it is believed that the application of this control banding to SMEs will contribute to the achievement of the UN Sustainable Development Goals (SDGs) "8: Decent Work and Economic Growth" and "12: Responsible Consumption and Production".

3. FUTURE PROSPECTS IN CFRP RECYCLING

3-5-1 Movement of further reducing of environmental impact

Table 10 summarizes the products in which recycled carbon fibers have actually been adopted. All of these cases are from Europe and the United States, not Japan. In particular, in Japan, although the recycling technology of carbon fiber materials has been established in some areas, it is difficult to obtain added values due to its material characteristics, and the use of recycled materials has not progressed smoothly. The development of technology to recycle carbon fiber-based materials with high added values is still underway.

Table 10. List of major applications of recycled carbon fibers

Product area	Examples of recycled carbon fiber applications
Electronic products	Laptop body In the U.S., recycled carbon fiber products have been adopted mainly by DELL for notebook PC cases, and some other brands have also started to adopt them. DELL has teamed up with Carbon Conversions Inc. of the U.S., a supplier of recycled carbon fiber, and is working with aircraft manufacturers to collect off-spec, surplus, and scrap CFs and cut them into small pieces and pelletize them, and then compound them with resins for use in notebook PC covers. For example, in the Latitude 7300, launched in 2019, by using rCF (18.8% content) for the lid of the notebook, the weight has been reduced by 24g compared to the previous model.
	IC chip tray
Energy field	Wind power generation-related products (wind turbine nacelle covers, etc.) In Europe, where wind power generation has been developed as an energy source, recycled carbon fibers have been adopted for wind turbine nacelle covers. Compared to glass fibers, the use of carbon fibers can reduce the deflection of wind turbine blades.

Automotive parts	Roof cover Brake rotors Hood Trunk partitions A-pillar C pillar For automotive applications, the amount of use is increasing in Europe and US for OEMs. In addition to C-pillars, which attracted attention in BMW's 7 series, recycled carbon fibers are also used in roof covers and other applications.
Aircraft field	Side panel Recycled carbon fibers have also been used in airplane side panels and access doors. In December 2014, Boeing issued a press release stating that the access doors on the underside of the wings of the *Boeing 787 Dreamliner* are manufactured by using recycled CFRPs made from scrap materials from the Boeing 787 manufacturing process. This is the first time that carbon fibers which were recycled from aerospace components had been used as a component in commercial aircrafts. Boeing is working with Rolls Royce, a global engine manufacturer, and other companies to promote carbon fiber recycling business. 
Space field	AC power supply components Recycled carbon fibers have also been used for AC power components in spacecraft. However, the hurdles to entry in the aerospace industry are high due to their high specifications for material properties and performance.
Other products Sports playground equipment, Molding materials related to 3D printing materials, Medical equipment (X-ray diagnostic beds, X-ray equipment for medical use, etc.)	

It has been shown that a weight reduction of 100 kg in an automobile vehicle is worth a reduction of 20 g/km in CO2 emissions, and the wide use of recycled CFRPs in vehicles is expected [112]. The National Composites Center (NCC) at Nagoya University has successfully fabricated a 100% thermoplastic CFRP automobile chassis by using the

Long Fiber Thermoplastics Direct (LFT-D) method, which mixes thermoplastic resin and virgin carbon fiber. Under these circumstances, in recent years, there has been a shift in thinking in terms of reducing environmental impact, aiming for "plastic-free" composites with paper (pulp) (so to speak, "*Carbon fiber-reinforced paper*"), rather than conventional composites with petroleum-based plastics. As an advanced example, SANKEN CORPORATION is mixing carbon fiber with paper to make carbon fiber sheets (paper), which they are aiming to use as heating elements (components that dissolve ink in inkjet printers). Although traditionally, keeping affinity with matrix resins has been a challenge in composite materials, there has been a changing of the way of thinking by focusing on the concept of ”reuse”, in which recycled CFs (rCFs) are not returned to the composite, but are used as a nonwoven fabric as they are, such as sputter sheets (sheets for catching welding sparks). When considering the reuse of CFRPs, it is also a good point that they can be manufactured directly by recyclers themselves without going through a compounder, a company that processes materials into pellets etc and sells them to molding and processing manufacturers.

Moreover, the introduction of less environmentally damaging recycling methods is also highly required, and there is a movement to devise ways to make the products easier to recycle from the production design stage. It is known that separating the resin matrix from waste CFRP at the end-of-life stage is very difficult. In order to dissolve current thermoset resins, aggressive chemical or high-temperature reaction conditions are required, which can cause damage to the recovered CFs. Therefore, it is a good idea to use biodegradable resins in the manufacturing stage of new products [60]. Lastly, I regard it is effective to aim for alternative applications of “activated carbon” (utilizing the performance of carbon itself), in addition to the reuse of non-woven fabrics such as sputter sheet. For example, it is a way to look for applications in environmental fields such as exhaust gas treatment, water quality, and soil improvement.

3-5-2 Advantages of recycled compared to virgin CFs and its utilization

Regarding energy consumption (CO_2 emissions), the following has been elucidated [60].

- vCFs: 198–595MJ/Kg (about 10 times higher energy consumption than glass fibers)
- rCFs: 38.4MJ/Kg (when using fluidized bed method)

- Regarding cost, for automotive applications, if the plant's production capacity is 500–6,000 tons per year, it is expected to be less than $5 per kilogram ($5/Kg), which is about 15% of vCFs. However, at the production capacity of 100 tons per year, the cost will be about $15/Kg.

In summary, the advantages of rCF products over virgin products are mainly environmental and price aspect. In recent years, when evaluating upstream recycling technologies, downstream automakers have been asking how much their recycling technologies can contribute to achieving the SDGs and decarbonization. Not only if they have the ability to recover clean rCF from waste CFRP, but also the ability to recover them with a low LCA values is becoming an important factor.

Here, we further pursued the superiority of rCFs and reorganized the other advantages.

It is often said that vCFs are strong, but have weak elongation and cannot be bent well, whereas rCFs have less strength than virgin ones, however rCFs are easier to form when creating a three-dimensional structures. In terms of compensating for the shortcomings of vCFs as mentioned above, twisted yarn is a very viable option. The reason is that twisting can change the elongation and shrinkage, and also can change the formability. The low performance of ordinary rCF short fibers makes it difficult to apply them to structural components such as automobiles, however, by making them into continuous yarns like twisted yarns, the performance can be improved. By establishing the twisted yarn technology, it is highly likely to meet the needs of all users. By mixing rCF milled fibers during 3D printing and using rCF powder as a reinforcement material in the polymer, it is expected to make the created objects less distorted. Therefore, if 3D printers are used widely in the future, rCFs can be expected to be used more [113]. It is generally believed that milled carbon fiber with a length of less than 200 μm (0.2 mm) does not have sufficient strength. This has been an issue in the development of applications for recycled carbon fibers. In these circumstances, Chubu Center of AIST has developed a new technology to improve oxidation resistance (heat resistance) and add insulating properties of milled carbon fibers with a length of about 200 μm (0.2 mm) by coating its surface with silicon carbide or silicon nitride (ceramic coating) [114]. This technology is so-called "upcycling", which means a recycle that improves physical properties and increases added value and makes it possible to improve the decomposition temperature to 600–700°C, which is higher than that of ordinary carbon fibers (around 500°C). rCFs have different surface properties compared to vCFs. The interfacial adhesion between CFs and matrix resin when using rCFs is higher and at the same time the number of functional groups on the surface is lower than when using vCFs [115]. CFRP products using rCFs have different surface treatment characteristics from those using vCFs. It is therefore hoped that applications will be developed that can make good use of this difference in characteristics. Research on the superheated steam (SHS) method [116], in which high temperature steam, nitrogen, carbon dioxide, etc. are used at the firing stage in order to increase the surface activity, is also being conducted under the support of the Japan Fine Ceramics Center (JFCC). By using this technology, it can be possible to modify the fiber surface (improvement of the adhesion to resin). Technically, SHS method is expected to improve the adhesion between fibers and resin in addition to fiber recovery. As a result, post-treatment processes such as sizing treatment for recovered fibers can be deleted, and the manufacturing cost of CFRP can be reduced. Moreover, surface pre-treatment technology of CFRP to activate and clean the surface of carbon fiber prior to resin bonding by using a dry process atmospheric pressure plasma has already been developed. The research on the evaluation of the interface properties of rCFs and the quantitative measurement of carbon residue and analysis of impurity elements on the surface of rCFs which are both simple/efficient and accurate has been conducted by AIST as part of the "Innovative Structural Materials R&D" project from FY2013 -2022. They have been working on the development of a simple analytical method for impurity elements using microwave acid digestion (pretreatment) and flow injection ICP-MS

3-5-3 Organizing the unique functionalities of CFRP (irrespective of virgin or recycled)

At The Japan Carbon Fiber Manufacturers Association's 32nd Composite Material Seminar in February 2019, Airbus executive expert said "one of the key areas where more effort is being made is the development of next generation multifunctional CFRP which includes anti-contamination, self-cleaning." The key will be how much we can promote the use of rCF and gain citizenship by taking advantage of its unique functionalities (vibration damping, electromagnetic shielding, etc.) other than its light weight and high strength. The functionalities of CFRP are listed below Table 11.

Although not related to functionality, there are also some products (motorcycles, racing cars, etc.) that use CFRP because of the attractive appearance of its crease pattern.

Table 11. Functionalities of CFRP

Functionalities	Major factor that produces functionality
Lightweight, high strength, high elasticity (high rigidity)	CF
Excellent radio wave transmissivity	CF
Excellent x-ray permeability	CF
Excellent vibrational damping	Matrix resin
Excellent dimensional stability (very low coefficient of thermal expansion)	CF
High thermal conductivity	CF
Electromagnetic shielding	CF
High radiation resistance	CF [117]
Microwave absorption	CF [114]
High sliding properties (high wear resistance, self-lubrication)	CF
Anti-static performance	CF
Sound absorption properties (soundproofing, sound absorption, sound modulation)	CF and Matrix resin
Impact energy absorption properties	CF and Matrix resin
Chemical resistance (acid and alkali resistance), corrosion resistance (rust and salt damage resistance), heat resistance (insulation), fire resistance	CF, Adhesive (binder) and Matrix resin
Non-magnetic properties	CF
Water purification characteristics	CF

Please note that above properties vary depending on the type and grade of used CFs, the matrix resin, and the laminate structure etc. The properties and rigidity of CFRP vary greatly depending on the material used and the molding method.

3-5-4 Future prospects in CFRP recycling

Companies on the user side, such as automobile manufacturers, have been hoping to further reduce the price of rCFs. However, there is a limit to the approach to lowering the price of rCFs. In particular, rCFs come from discarded CFRPs are finally recovered after a series of work processes including collection, dismantling, separation, and treatment of used products. Hence, it is very time-consuming. Though of course, manufacturers who sell recycled carbon fibers must continue to make efforts to reduce their costs, it is factual that there is very little room for recycled carbon fiber manufacturers to reduce the costs. At the same time, however, it is also necessary to have an attitude to appeal the merits of using rCFs to users.

For example, the use of recycled materials can contribute to the SDGs and the circular economy, and therefore efforts to promote a branding for recycled materials, so-called "eco-branding" are effective. In the future, it will be necessary to create a social system that can generate a demand even in such a high price range. Circular economy (CE) aims at using renewable energy, eliminating toxic chemicals, and eliminating waste by better design of materials, products, systems and business models. As the concept of CE is accelerated in Japan as well, it is safe to say that it will become more and more important to evaluate the ecological impact of milled CFs or CF dust generated during the recycling process [118].

Understandably, with repeated recycling, the length of the fiber shortens and eventually becomes milled. Therefore, the issue of the health effects of milled CFs or CF dust on humans and other living things, as discussed in this study, is the last issue that must be addressed when promoting a resource circulation, including a recycling. Therefore, we consider the issue of the health effects of milled CFs or CF dust on humans and other living things is an important theme that cannot be avoided and where we end up eventually.

4. CONCLUSION

The use of recycled CF is expected to expand in the future and it is also expected that not only in-plant waste and scrap materials but also waste materials associated with end of life machines and vehicles will be discharged in the future.

Most of the CFRP matrix resins used worldwide are thermosetting resins such as epoxy resin. The main issue in recycling CFRP is that it is difficult to reuse by remolding because of the nature of the thermosetting resins. That means thermosetting resins have high mechanical properties and account for a large part of the market but are not easy to recycle.

Epoxy resins have a three-dimensional cross-linked structure, thus the complete thermal decomposition in CFRP requires treatment in an oxidizing environment, making a certain amount of damage to CF inevitable [119].

In this study, we organized the issues in the Japanese industrial landscape as to why CFRP recycling businesses and technology have not yet been established.

In consequence, Environmental, health, and safety (EHS) issues still exist in promoting CFRP recycling and among the presence of various issues, such as quality assurance and standardization of the evaluation methods of reclaimed CF, we noticed that, conspicuously, information with scientific evidence on the toxicity of CF itself is poor and CF is seemingly treated as a non-toxic substance without adequate verification especially in Japan.

Despite that situation, two journal articles [51, 52] indicate that fine particles such as milled short CF treated by industrial processes have a likelihood of reaching the human alveoli by inhalation and CF may possibly exhibit toxicity depending on the method of treatment and management such as firing treatment during the pyrolysis recycling process because the inherent impurities such as dioxins attached to CF may impart toxicity to CF dust.

The conclusions that emerged from organizing the issues are we should regard the improvement of CFRP recycling and processing methods as an important issue, because they have a correlation with biological and health effects as well as environmental and energy issues.

We therefore suggest that further analysis focused on elucidating the anxiety of the effects of CF (dust/milled) itself on people and ecosystems be conducted in order to gain a true understanding of the impact on citizens at the time of using CFRP recycled products on an industrial scale.

Through distributing appropriate information, confirmed on a scientific basis, that will not disrupt society, it will be possible to obtain safety and relief from anxiety for ordinary citizens. By doing so, it will also be possible to develop a socially receptive recycling business implementation system to make it easy for new recyclers to start a business.

In order to elucidate above issue, I delved into the effects of milled CFs or CF dust generated during a CFRP recycling process on the environment and living organisms when CFs are released into the water environment as a by-product unintentionally.

Specifically, the effect of CFs on Japanese medaka fish (*O. latipes*, orange-red variety or himedaka) in the coexistence of rCFs was confirmed by experiments in a water tank.

As a result, no remarkable toxicity was observed in the CFs. However, the CFs clearly injured the medaka. The wounds would be expected to heal in a prepared environment such as an artificial experimental model. However, in an actual environment, especially in one with high biochemical oxygen demand or chemical oxygen demand, the rate of infection of wounds by microorganisms is expected to increase.

Although only small amounts of milled CFs or CF dust are entering the aquatic environment, the amount is expected to increase in the future as disposal and recycling increase. If large amounts of milled CFs or CF dust are discharged in the future, the effects on aquatic organisms such as fish will be a major concern.

We recommend formulating a solution such as preventing the transfer of milled CFs and CF dust generated during various industrial processes to natural aquatic areas before recycled CFs are used to replace virgin CFs for the purpose of resource circulation.

Actual air sampling in the laboratory of a SME showed that CF remained in the indoor air for some time after the CF-related work was completed. In addition, particulates in the 0-20 μm range were also found to be present. This is a fact that should be of concern.

Obviously, with each repeated recycling, the fiber length of CF gradually shortens. Eventually, there is concern that they will become nanoscale fine particles (NOAA). Therefore, as part of the evaluation of occupational exposure to recycled CF minute dust, an occupational risk assessment using a controlled banding (CB) approach was also piloted.

The results showed that the CB method is useful for small and micro enterprises to consider control measures as a first step.

Finally, we hope that our efforts will contribute to the achievement of sustainable development goals such as Goal 14 of the United Nations, which covers life below water and Goal 8: Decent Work and Economic Growth and 12: Responsible Consumption and Production.

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