
Advanced Composite Materials
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/tacm20

# Recycling of resin matrix composite materials VII: future perspective of FRP recycling

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Version of record first published: 02 Apr 2012.

*Adv. Composite Mater.*, Vol. 6, No. 3, pp. 215–225 (1997)


# Recycling of resin matrix composite materials VII: future perspective of FRP recycling

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Received 16 December 1995; accepted 16 October 1996

**Abstract**—Resin matrix composites reinforced by glass fibers and carbon fibers (FRPs) are used extensively for many industrial applications, such as construction materials, automobiles, ships and sporting goods, because of their high strength, ease of molding, light weight, durability, and resistance to corrosion, shock and abrasion. However, despite its many superior properties, it is difficult to resource and recycle FRP after its disposition. In this report, the recycling technology of FRP that has been developed is introduced as well as an outline of new recycling technology of FRP developed by the authors.

FRP fine powders are made from waste FTP moldings using a fine powder crusher. The average grain size of these fine powders is 15–20 μm. FRP fine powder was examined as a possible additive to paints and it was found that it increased the tensile strength of the paints. FRP can also be recycled as filler for FRP resin. FRPs were also examined for possible civil and construction materials and they were found to be useful as aggregates for concrete secondary products. After examining these manufacturing conditions in detail, by fabricating light and high-strength mortars and investigating strength prediction and water solubility after extended time, it was found to be a practical material. Burned ashes from FRP showed superior properties as mortar concrete expansive admixtures. It was concluded that FRPs, which used to be hard-to-destroy and hard-to-recycle materials, can be recycled for many industrial materials after crushing into fine powders.

*Keywords*: FRP; material recycling; recycle; aggregates; construction materials.



## 1. INTRODUCTION

Environmental problems concerning materials such as advanced composites of FRP include recycling, environmental problems that arise during their service periods and usage and their disposal as waste. The severity of these problems depends on the quantity and quality of the materials to be disposed. A yearly production quantity exceeding 1 million tons may pose a social problem in Japan. For example, polyethylene (2 million tons produced yearly) and vinyl chloride (1.4 million tons produced yearly) pose problems in terms of quantities while other materials whose production is in the order of 100 000 tons per year create problems only when they are difficult to dispose of.

Since the yearly production quantity of FRP is only 45 000 tons, there is no social problem in terms of quantities. However, FRP's superior characteristics [1] such as easy molding, high strength, light weight, durability, corrosion resistance, damping characteristics, impact resistance and abrasion resistance work against developing an efficient recycling method [2–5]. Due to the weight reduction of transportation equipment and environmental problems that accompany recycling and disposal, recycling of FRP has finally begun.

The recent attempts at recycling FRP by the Shikoku Industrial Technology Center of MITI, the Synthetic Resin Industry Association of Japan and the FRP Association of Japan is significant and many achievements have been reported [6–10]. The recycling business of FRP is also aggressively pursued in the US and Europe [6, 7].

The characteristics of FRP are determined by its birth and growth environments. FRP wastes are classified into those wastes generated from various factories such as resin manufacturing plants, molding businesses and users and those wastes generated from the consumers of FRP products after their lifespans. The former include trimming gas discharged from factories at the time of manufacturing, polymer residues and failed moldings. Although it is possible to trace back the history of these FRP products before their disposal, once they are land-filled by industrial waste disposal contractors, it is impossible to know their history. Also, if consumers dispose of FRP products by themselves, it is almost impossible to trace the history of FRPs. For these reasons, it is necessary to label the birth and growth environments of FRP at the time of manufacturing.

It is important for FRP recycling to clearly identify which stage of FRP wastes is to be recycled. Recycling of FRP is very challenging even with the latest processing technology from the economic standpoint of view. However, as the birth and growth records of industrial FRP wastes from factories are usually well maintained, recycling of these materials is easier than scrapped FRP ships or general FRP wastes. Therefore, if a recycling technology for industrial FRP is established first, such a technology can be applicable to other types of FRP wastes as well.

The speciality of one of the authors (Kojima) is carbon composite materials and the other's speciality (Furukawa) is concrete composite materials. The two have been working together to conduct research on manufacturing and high functionalization of carbon fiber reinforced concretes and the emphasis is focused on developing an FRP recycling technology that also involves a social problem. This report is concerned with recycling of glass fiber reinforced plastics as well as advanced composite materials reinforced by carbon fibers which have better properties than glass fiber reinforced composites even though the usage is limited. The future perspective is also discussed.

## 2. CURRENT STATUS OF FRP RECYCLING

Although recycling of FRP is being studied by the FRP Association of Japan, the Shikoku Industrial Technology Center and various concerned industries (resin, molding, automobile, sail boat, yacht, poly-bath, sports goods),no outstanding method has been established.

FRP is often used in combination with metals and wood materials in addition to being used on its own. For this reason, it is necessary to separate FRP wastes into each raw material that constitutes the FRP product for recycling [2–5]. To separate FRP into each raw material, it is necessary to perform preprocessing such as cutting and crushing. For both methods, the reader may refer to an excellent review by Miyairi *et al.* Each method has its own advantages and disadvantages and the following is a rough classification of FRP recycling methods applied after separation.

### *2.1. Direct utilization technology*

This technology involves crushing FRP wastes and mixing them with organic or inorganic matrices for recycling. The recycled FRP is then used:

(1) As resin material. For example, FRP products of Class A that are not inferior to conventional products in terms of strength and surface smoothness can be made by crushing SMC mold wastes into particles and using them as SMC fillers for automobile parts. In EC countries (Germany in particular), a plan to crush SMC of abandoned automobile parts into powders by a shredder for reuse is underway.

(2) As aggregate for concretes (for example, manhole caps using BMC waste materials are being made).

(3) As aggregate for gypsum boards.

### *2.2. Indirect utilization technology (thermal decomposition technology)*

This technology comprises separation of FRP wastes into organic and inorganic components by thermal decomposition for reuse. The technology is divided into thermal recycling [2–5], in which FRP is incinerated and the heat generated is utilized, and chemical recycling, in which generated oil-like materials are reused as fuels.

(1) A recycling method to extract various oil components by thermal decomposition for reuse. General Motors of the US is producing oil components equivalent to C heavy oils by drying SMC made external panels of scrapped automobiles at 380–450°C. They are decomposed at 760°C to generate methane gas and low viscosity fuel oils. However, no commercialization has been achieved so far.

(2) A recycling method to extract aromatic compounds in resin and use them as chemical compounds [8, 9].

(3) A recycling method to singly-separate glass fibers by thermal decomposition. Glass fibers from thermally decomposed residues are used as fillers for SMC resin, concrete products and aggregates for roof tiles.

(4) A recycling method to use generated thermal energy at thermal decomposition as fuels and use ashes as a mixing material for mortars.

(5) A recycling method to control thermal decomposition conditions to produce silicon carbide whiskers [8–10].

(6) A recycling method to produce fuels for cement manufacturing plants.

Attempts with item (6) have recently begun and it is a promising method as mass processing is possible. Cement manufacturing plants burn 300 000 tons of scrapped tires yearly which amounts to one third of scrapped rubber tires. FRP can be used as fuels by throwing them into cement kilns. Since glass fibers or other fillers in FRP are similar to the elements that compose cement, no problem arises when they are thrown in kilns. However, this method depends on the structure of kilns in cement manufacturing plants and it does not work for certain types of kilns. A problem with this disposal method lies not only in technological issues but also in economical issues of whether the FRPs are valued items or who pays for shipping of FRP.

The types of furnaces for FRP include the rotation method, the mechanical method and the mobile bed method. As FRP does not burn by itself, it is necessary to add tires, scrapped plastics and city garbage for incineration. There are many unresolved issues such as control of combustion temperatures, deposition of glass fibers on the furnace walls and disposal of generated ashes and the technology has not yet been perfected. Air contamination, odor and workability must also be properly handled for incineration of FRP.

## 3. ULTRA FINE POWDERIZATION OF FRP

To recycle FRP waste materials, it is necessary to change the shape of the FRP object to one which is easy to recycle, such as one-dimensional or two-dimensional shapes. One-dimensional shapes refer to pointwise shapes which transform FRP waste materials into pointwise materials (fine powder) by cutting or crushing. Two-dimensional shapes refer to fiber-like shapes that use glass fibers in elongated state without cutting.

In the current crushing technology, it is possible to make FRP into particles or fiber bundles with an order of a few millimetres by the shredder, crusher or ballmill method. However, it is extremely difficult to make FRP into fine powders with an order of a few micrometres. Our research group has developed a new grinding method using a grinder that embeds polished diamond particles and we have successfully achieved finer powderization of FRP. For the composition and performance of the FRP crusher, refer to [11, 12, 15–18].

Fine powderized FRP powders were separated using four types of sieves. The weight of the FRP fine powders on each sieve was measured to obtain particle degree distribution. Most of the particle degree distribution was less than 44 μm. The shape of FRP fine powders on each sieve, observed by a scanning electron microscope, showed instant cut with very sharp edges.

As the separation method using a sieve cannot obtain the particle degree distribution of consolidated FRP fine powders, measurement was conducted by using a laser refraction type particle degree distribution tester using water as a dispersion agent. The particle size of FRP fine powders was within a range between 0.4 μm and 100 μm and the number of powders with the size of 16 μm was the largest with the average particle size of 13.5 μm.

There are some issues concerning fine powderization of FRP.

(1) How to maintain the quality of fine powders without which recycling is not possible.

(2) Abrasion of diamond edges at the cutter. It is confirmed that the abrasion is not as severe as expected even though many years have passed since the first manufacturing.

(3) Size of crushable samples, since FRP must be cut and crushed to match the size to the entrance of the crusher.

(4) Separation from other raw materials that are part of the composite. Although a crusher can fine-powderize both metals and ceramics, their mixing must be avoided because (a) separation and removal operations after crushing are still needed, (b) the quality as a new material may be degraded and (c) the life of a diamond cutter may be shortened. Consequently, raw materials such as metals except for FRP must be separated before the crushing which requires advanced separation technologies that have been developed.



## 4. RECYCLING AS FILLERS FOR PLASTICS

Recycling of FRP is not practical unless it pays economically, even though it is a technologically superior material. What is important in recycling is to develop a method which does not require moving (transporting) scrapped FRP products. For this reason, recycling of FRP is most practicable when all the processes are carried out in one manufacturing plant where FRP is actually made. Then, recycled FRP ultra fine powders would have great utility as fillers for plastics.

FRP uses fine powders such as calcium carbonate and aluminum hydroxide in addition to resin and reinforcing glass fibers. When FRP fine powders were used as a replacement of these fillers to fabricate resin boards, their surfaces were sufficiently smooth and were no worse than the boards when calcium carbonate was added. The mechanical strength (specific strength) of recycling samples filled with FRP fine powders was 1.3 times higher than those filled with calcium carbonate [11]. Resin boards were also made by filling FRP fine particles in unsaturated polyester resin and phenol resin (resol type) and the workability during molding and the mechanical properties of the molding were not degraded. Automobile spoilers using FRP fine powders produced by the SMC method are already in production. In this case, it is possible to recycle within the same company using fine powders whose history is well known.

## 5. RECYCLING AS PAINT FILLERS

FRP fine powders with superior properties were examined as paint fillers (body extender pigments). Proper types of pigments must be added to paints and in many cases

**Table 1.**
Influence of FRP fine powder on paint characteristics

| Particle degree distribution μm | Tensile strength (MPa) | Young's modulus (MPa) | Extension (%) | Brightness | Pencil hardness |
|---|---|---|---|---|---|
| FRP < 300 | 3.2 | 35 | 14 | 4 | 4B |
| FRP < 75 | 9.2 | 106 | 17 | 8 | 2B |
| 75 < FRP < 150 | 6.2 | 68 | 15 | 8 | 2B |
| 150 < FRP < 300 | 3.4 | 45 | 12 | 5 | 2B |

FRP (hand-lay)/oil paint (phthalic acid resin) coating.
FRP added amount: 20%.



adding body extender pigments along with coloring pigments can control viscosity, dispersity and brightness.

Therefore, an investigation was performed on whether FRP fine powders can be used as paint fillers. FRP fine powders were added into a compound resin paint and were coated on a glass board with a constant thickness after sufficient mixing to make paint films. Pencil hardness and brightness of the paint films (a thickness of about 200 μm) were measured and the tensile strength of the paint film was also measured by peeling it from the glass board. The properties of the paint film depended more on the type of paint (water acrylic paint, oil alkyd paint, phthalic acid resin paint, etc.) than on the type of FRP. The tensile strength and brightness of the paint film were degraded as a result of adding FRP fine powders that could be used as frosting and a tendency of hardening was found.

The strength of paint film itself is thought to depend on the particle size range of added FRP fine powders. For this reason, FRP fine powders were sieved into three types. They are (1) a group whose size is less than 75 μm, (2) a group whose size is 75–150 μm and (3) a group whose size is 150–300 μm each of which was added to paints separately to make films and their tensile strength was obtained (see Table 1). The tensile strength of the paint films made of those paints that did not disperse FRP fine powders was 3.2 MPa and the tensile strength of the paint films where FRP fine powders of less than 75 μm were added at 20% concentration was 9.2 MPa, which was a threefold increase in strength. The Young's modulus of the paint film also increased threefold from 35 MPa to 106 MPa.

In order to examine if the tensile strength of paint films can be increased by adding fine materials, calcium carbonate powders with an average particle size of 10 μm were added to the paints and measurements were conducted similar to those with FRP fine powders. The tensile strength of the paint film was reduced when calcium carbonate was added and the larger the amount, the lower the strength. It was found from this observation that scrapped FRP whose resin is hard to dispose of can be utilized as paint fillers by fine-crushing.

## 6. RECYCLING AS AGGREGATES FOR MORTARS

The raw materials of mortar concrete products include fine aggregates, coarse aggregate and cement and the scarcity of the resources for aggregates is becoming a serious problem. For example, river sands are desirable for the sands used as fine aggregates but they are difficult to obtain so that a replacement for them needs to be sought. In particular, pumice stones and sands are utilized as the aggregates of blocks used for residential walls and fences. However, such pumice stones are becoming difficult to obtain from the standpoint of environmental destruction problems and new aggregates need to be found. In order to examine whether FRP fine powders can be used as aggregates for construction materials, various concrete products have been fabricated for testing.

Plane blocks (30 cm × 30 cm, thickness 6 cm), artificial wood of concrete and interlocking blocks were manufactured by pressing FRP fine powders, sands and cement mixtures under high pressure vibration by a company in Gunma Prefecture. Mortars that contain FRP fine powders have strong freeze resistance and solubility, and possess surface smoothness so that they can be used as the mortars (60 cm × 60 cm, thickness 5 cm) of the retaining wall or as revetment overlaying the block surface [13, 14, 18].

The relation between the manufacturing conditions of FRP fine powder/mortar composites and the mechanical strength was studied and it was found that mortars with reasonable practical strength can be manufactured and the weight can be reduced even if they contain FRP fine particles. Consequently, concrete mortars that contain FRP fine powders are suitable for reduction of transportation cost and for construction of high-rise buildings [16, 17].



## 7. LONG TERM STRENGTH PREDICTION OF MORTARS THAT CONTAIN FRP FINE POWDERS

As construction materials are expected to be used for a long time, maintenance of long term strength is essential. However, it is pointed out that the alkali resistance of glass fibers in FRP is poor and strength reduction may result if they are used for a long time. It is also feared that unsaturated polyester resin which is the matrix material for FRP may be decomposed by the alkali components of cements.

Long-term strength was examined by a hot water soaking test commonly used by the construction material industries. As soaking in hot water at 80°C for one day is believed to be equivalent to 2.5 years of outdoor exposure, the samples were heated for a maximum of 30 days (equivalent to 75 years of outdoor exposure) so that the mortars containing FRP fine powders were forced to be degraded. Mechanical properties of the samples such as unit volume weight, flexural strength and compressive strength were measured. The failure surface of the samples after the flexural strength test was observed by an electron scanning microscope and erosion of the glass fibers was also examined.

Table 2 shows the strength before and after the hot water soaking test of each sample. Since the mechanical strength of mortars containing FRP fine powders was not degraded by the hot water soaking test, it was concluded that they have long-term stability. The presence of many glass fibers was observed on the sample failure surfaces before the soaking test and this did not change much even after six and ten

**Table 2.**
Influence of FRP fine powder contained mortars on mechanical properties by hot water soaking test

| Soaking duration (days) | Outdoor exposure (year) | Weight of unit volume (ton/m$^3$) | | Flexural strength (MPa) | | Compressive strength (MPa) | |
|---|---|---|---|---|---|---|---|
| | | hand | SMC | hand | SMC | hand | SMC |
| 0 | 0.0 | 0.98 | 1.08 | 2.0 | 2.0 | 9.5 | 10.9 |
| 1 | 2.5 | 0.96 | – | 2.4 | – | 9.2 | – |
| 3 | 7.5 | 0.99 | 1.11 | 2.5 | 2.5 | 9.2 | 13.3 |
| 6 | 15.0 | 1.03 | 1.10 | 2.6 | 2.5 | 10.0 | 12.7 |
| 10 | 25.0 | 1.02 | 1.09 | 2.3 | 2.4 | 9.7 | 11.8 |
| 30 | 75.0 | 1.07 | 1.14 | 2.3 | 2.9 | 8.7 | 11.4 |

Water/cement W/C: 0.71.
Aggregate (sand + FRP)/cement: 0.67.
FRP/(sand + FRP): 0.43.
Water curing: 28 days.
Hand: FRP fine powders molded by hand lay-up.
SMC: FRP fine powders molded by SMC method.

days of soaking. Many glass fibers in the surface state that were not damaged were observed on the failure surface of the samples after 30 days. It is concluded that the solubility of glass fibers and unsaturated polyester resin is small even after 30 days of soaking in hot water (equivalent to 75 years of outdoor exposure) [19].

## 8. RECYCLING OF BURNED ASHES

Recycling of FRP residues after thermal decomposition of FRP (FRP ashes) was studied. Attempts to use FRP ashes as aggregates for mortars and concretes have been explored, but no other applications. It was found after various studies that they can be recycled as expansive admixtures. Expansive admixtures can be used to suppress cracking of mortars and concretes due to their dry contraction and to improve flexural strength by chemical prestress. The usage quantity of expansive admixtures as construction materials is about 50 000 tons yearly.

FRP ashes used in our experiment were produced when FRP made by the SMC method was burned at a temperature ranging from 800–900°C in a furnace. The chemical components of FRP ashes were found to be calcium oxide, calcium carbonate and calcium silicate from an X-ray refraction analysis. FRP ashes composed of these compounds possess water affinity and self-solidification that are necessary for expansive admixtures. For this reason, the mortar characteristics that use FRP ashes as part of cement were investigated. For comparison purposes, commercial expansive admixtures (CSA, ettringite series made by Denki-Kagaku KK) were also used. Table 3 shows the mechanical properties (strength after 28 days) of the mortars. The flexural strength and compressive strength were about the same as those of commercial expansive admixtures if the replacement by FRP ashes was about 10% by weight. The expansion rate of the sample in the longitudinal direction was about the

**Table 3.**
Mechanical properties of mortars containing FRP combusted ashes

| Additive | Cement reprecement rate (%) | Flow value | Flexural strength (MPa) | Compressive strength (MPa) | Expansion rate ($\times 10^{-6}$) |
|---|---|---|---|---|---|
| None | 0 | 240 | 6.3 | 47 | None |
| FRP ashes | 10 | 190 | 5.0 | 33 | 550 |
| | 20 | 160 | 4.6 | 21 | 1450 |
| | 30 | 140 | 2.8 | 11 | – |
| Commercial expansive admixture material | | | | | |
| | 10 | 230 | 6.0 | 38 | 600 |
| | 20 | 210 | expansive admixture destroy | 9 | 1500 |

Water/(cement + FRP ashes) = 65%.
Sand/(cement + FRP ashes) = 2.0.
Cement replacement rate = FRP ashes/(cement + FRP ashes).
Water/cement = 65%.

same as that of the commercial expansive admixture. The time change of expansion was also almost the same for both the samples and commercial expansive admixture. Consequently, FRP ashes that possess these characteristics can be used as a cement hardener (hardener of weak foundation and organic soil, and industrial waste disposal) if used with cements.

## 9. DISPOSAL AND RECYCLING OF CARBON FIBER REINFORCED PLASTICS

Fibers used as a reinforcement material of FRP may be made of carbon, glass, Aramid, or polyethylene. The last two types are organic fibers that can be combusted and pose no environmental hazards. On the other hand, carbon fiber reinforced plastics (CFRP) pose various problems. Many new technologies developed using glass fiber FRP cannot be automatically applied to carbon fiber FRP. The current annual production rate of CFRP is about 10 000 tons and this figure has not posed a major social problem yet. However, if CFRP is adopted by a large scale industry such as the automobile industry, the magnitude of production will be increased significantly. As the mechanical properties of CFRP are superior to those of glass fiber FRP, and CFRP is electrically conductive, environmental problems are bound to emerge. Consequently, the future of CFRP may not be that bright.

The easiest method for recycling CFRP is combustion similar to glass fiber FRP. Although carbon fibers are said to be combustion-resistant, they are oxidized in the air and can be burned. When this happens, the combustion temperature rises so that the combustion method and its condition need to be examined. Other problems still remain such as how to collect burned ashes and to disperse unburned ashes and, in particular, the dispersion of carbon fibers may cause damage to surrounding electric equipment.

The second method of disposal of CFRP is to crush CFRP and use the resulting material as expansive admixtures. This method is close to the one applied to FRP by the authors except for the fundamental difference in that CFRP is electrically conductive. As glass fiber FRP is insulative, there is no problem when it is placed in concretes or mortars but embedding CFRP makes concrete conductive. Although it is still possible to take advantage of this as a concrete for static removal, the problems of grounding and electric leakage remain unsolved, and these may be very critical.

The third method of disposal is to recycle CFRP as solid fuels or other carbon materials after carbonization by heat.

The fourth method is to bio-degrade CFRP. Bacteria are known that eat carbon fibers [20–22]. If FRP is made of carbon fibers and bio-degradable resin, both the reinforcement material and the matrix are decomposed and no environmental problems due to disposal would result.

It is necessary to conduct basic study no matter which method will be adopted in the future. The technology and experience accumulated through the study of glass fiber FRP will be fully utilized.



## 10. FUTURE PERSPECTIVE

The greatest strength of FRP is its superior characteristics and its weakness is the difficulty of recycling, unlike steels or aluminum. However, a recycling technology for FRP, hard-to-crush resin, was finally developed and the best method seems to depend on the place where FRP was originally manufactured and processed. The recycling technology in general is governed by the principle of economy. Recycling is possible only when the supply and demand match. For this to be realized, it is important that wastes are not moved at all. If any transportation process is involved, a whole new system must be reconstructed as the balance is no longer held. For FRP recycling, R&D&M is important as well.

### *Acknowledgment*

Thanks are due to Professor Hiroshi Maki of Takushoku University for valuable guidance.

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