

## Recycling of Polymeric Matrix Composites

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**Abstract.** This work brings up the issue of the recycling of residues made of polyester reinforced glass fibres (PRGF), as well as the involved recycling technology. The future PRGF growth projections have led to an increased attention of the post-consumed products. For PRGF, it is observed that three recycling routes are available: mechanical, chemical or energy recovery. The mechanical recycling is attained by means of grinding a material, which has characteristics that allow its reuse. This paper deals with powder technology processing of residues, by reviewing the available technologies and processes aiming the PRGF recycling.

### Introduction

A composite material in its most basic form is composed of at least two combined elements to produce a material with more favourable properties. In practice, most composites consist of a bulk material (the matrix), and a reinforcement of some kind, added primarily to increase the strength and stiffness of the matrix. This reinforcement can be in a fibre form. Nowadays, the most common fabricated composites can be divided into three main groups: polymer matrix composites (PMC's), metal matrix composites (MMC's) and ceramic matrix composites (CMC's)[1]. Lightness and mechanical strength allied to the low relative production cost, are the qualities of the polymer matrix composites that make them competitive, allowing the substitution of conventional materials, such as wood and metal.

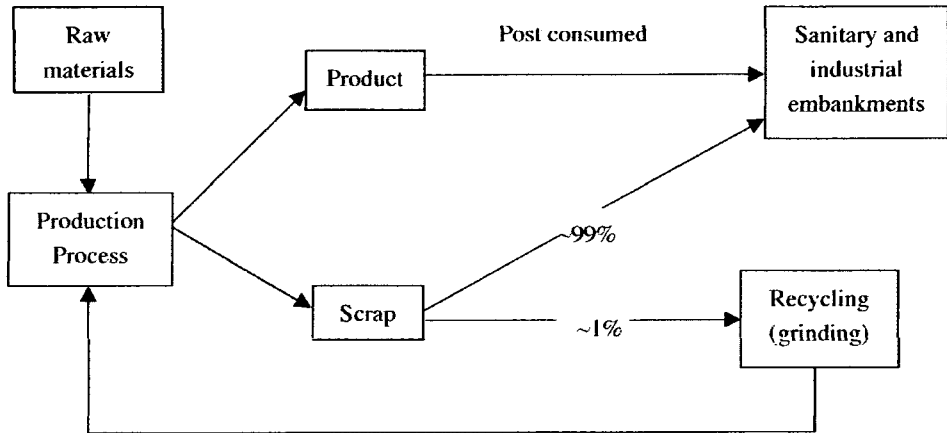
The PRGF composite is an engineering material that consists of the thermoset resin and the fibre. The liquid resin is combined with the fibre in the moulding process, and cures into a solid laminate. There are many types of composite resins and reinforcements and each of these imparts specific properties to the PRGF product. The resin system selection is based on the functional and cost requirements of the product. There are a number of reinforcement fibres used in composites; however, the PRGF contribution to the total amount of fabricated polymer matrix composites is over 90%.

PRGF composites present exceptional versatility adapting to countless application. From roof tiles to car parts, it has guaranteed use. From the ecological point of view, however, it poses a problem. It is considered that the thermoset matrix composites industry in Brazil generates approximately 10,000 tons of solid residues per year, which are usually disposed in industrial embankments [2]. The recycled amount is less than one percent of the total generated, excluding the post-consumed products, see Fig. 1.

The solid residues generated during processing of PRGF products must be appropriately managed

(collection, packaging, transport, treatment and/or final disposition), as well as the post-consumed products. This is intended to minimise the environment damage, and to preserve the health and the welfare of the population.

While the use of thermoplastics for packing produces hundreds of tons per year of solid residues, the thermoset polymer composite material generates only a small percentage of this one. However, it has an aggravating factor, which is the non-melting resin used as matrix. It must also be taken into consideration the growth projection of use of this material in the Brazilian market. This is corroborated by the arrival of foreign industries that produces resins.



Closed mold process

Fig. 1. Scheme of the productive cycle of polyester reinforced glass fibres (PRGF) in Brazil.

There are many management options for PRGF waste recycling, see Fig. 2. Among the options, the chemical recycling, energy recovery and mechanical recycling are the usually available routes to recycle PRGF.

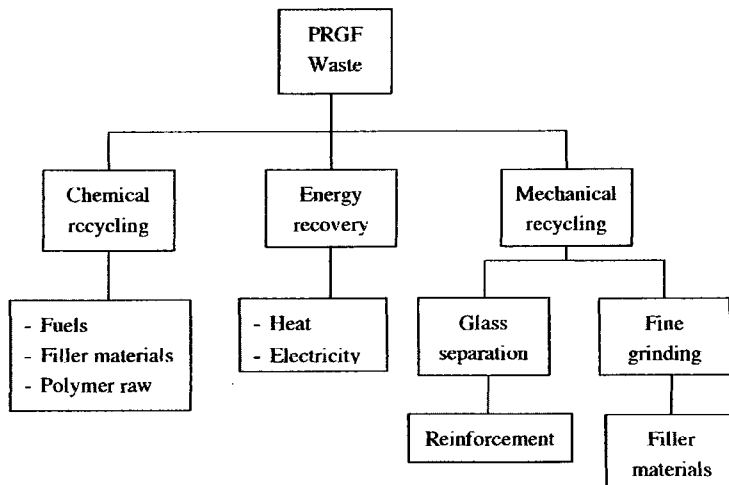


Fig. 2. Scheme of PRGF waste management options.

**Energy recovery.** The energy generation by incineration of plastics waste is in principle a viable

use for recovered waste polymers since hydrocarbon polymers replace fossil fuels, and thus, reduce the CO<sub>2</sub> burden on the environment. Table 1 shows some plastics calorific values compared with that of conventional fuels [3]. However, in fibre reinforced thermoset composites, the fibres are more valuable than the energy content of the polymers.

Table 1. Calorific values of plastics compared with conventional fuels [3].

Fuel	Calorific value (MJ/kg)
Methane	53
Gasoline	46
Fuel oil	43
Coal	30
Polyethylene	~43
Mixed plastics	30-40
Municipal waste	~10

**Chemical recycling.** The chemical PRGF recycling route can be divided into three main categories, pyrolysis, hydrogenation and cracking.

- *Pyrolysis.* This process consists of the thermal degradation of organic materials between 400 °C and 800 °C. The material is burnt in the complete absence of oxygen, or at very reduced level of oxygen, producing gases (hydrocarbons, ammonia and chloride of hydrogen) up to 50 %, plus 25-40% of oil (syncrude), and solid residues.
- *Hydrogenation.* The material is processed in a hydrogen atmosphere at 300 °C to 500 °C and pressures of 100 bar to 400 bar, to yield 65-90% oil (syncrude), 10-20% gases, and up to 20% of solid residues.
- *Cracking.* The material is heated at 400 °C to 600°C, and at pressures slightly above atmospheric, to produce oligomeric waxy liquids suitable for further catalytic cracking. The gases formed are used to fuel the process [4,5].

**Mechanical recycling.** The Powder Processing Centre - CPP, has particular interest in the recycling of PRGF composites by mechanical grinding. The mechanical recycling route may be divided into glass separation and fine grinding.

- *Glass separation* [6]. The composite scrap must first be reduced to a size and shape that allows transportation and handling with some degree of efficiency. Then, the shopped material is processed. When composite materials are processed in a fluidised bed operating at high temperatures, the polymer breaks down and burns. This causes the release of fibres and any filler that are carried out of the fluidised bed in the gas stream. The fibres and fillers can be removed from this gas stream by, e.g. a cyclone. The recovered fibre has some recycling value, but less than virgin fibres for several reasons: the fibre length is no longer uniform or of the same size; the glass will no longer wet out as fast as virgin fibre glass; the glass has filler and resin attached that reduces the weight of fibre per weight used; the recovered fibres have about half the strength of virgin glass fibres.
- *Fine grinding.* This consists in the reduction of the PRGF scraps and process chips to particles

size of approximately 200  $\mu\text{m}$ , see Fig. 3. This is achieved through grinding and the fine material obtained is used as filler. The stages of the grinding process are size reduction of parts measuring about 50 x 200 mm; grinding in an attrition mill; classification of particles (particle size distribution); treatment of the particulate material. However, this route poses some difficulties. Grinding of the Fibreglas contained in the system is difficult [7]. The chemical reactivity of the particulate material (resin and particulate) has to be reduced to avoid low curing times (hardening).

The alternatives for use of the PRGF scrap are, fillers, concrete reinforcement, asphalt reinforcement, energy source and oil. The filler is mainly used in the composition of products manufactured by processes of closed mould. Its function is to reduce the cost, substituting the more expensive resin, besides improving characteristics such as, increase of the stiffness and reduction of thermal expansion. It is worth to stand out that the filler is broadly used in closed mould process, differently from what happens with the open mould process. In concrete or asphalt, the residue of PRGF is used as reinforcement. However, this use is not yet diffused due to lack of regulations. As energy source it is possible to obtain, from incineration, energy equivalent to that of fuel oils. From the oil obtained in chemical recycling processes (pyrolysis, hydrogenation and cracking), it is possible to produce, for instance, xylem, benzene and toluene. Those processes are highly cost demanding [2,8].

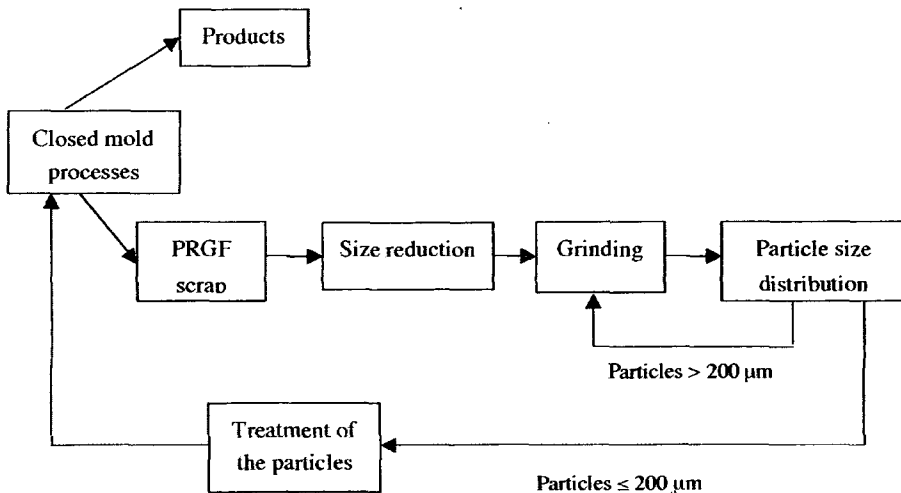


Fig. 3. Scheme for PRGF fine grinding.

### Concluding remarks

Due to the growth of PRGF use, the amount of their residues tends to increase. This implies in the need of effective disposable means for the residues. The search for alternative ways for the use of the PRGF-powder is necessary and should justify the cost of the process.

The most common use for the PRGF-powder nowadays is as filler in thermoset and thermoplastic materials for the plastic industry. There is also a possibility to use PRGF powder as a component for ceramic materials. In general, the use of PRGF-powder is limited. For instance, in the cosmetic and veterinary industries, the presence of pollutants is the prohibitive factor for the use of PRGF-powder.

It is likely that chemical recycling becomes a future tendency in the recycling of thermoplastic

and thermoset polymers. Studies are underway to turn the plastics recycling a profitable activity, or at least, solemnity-maintainable.

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