UPDATED REVIEW ON RECYCLING AND REUSE OF FIBRE REINFORCED POLYMER WASTES INTO CONCRETE BASED MATERIALS

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Abstract

The need for cost-effective end-use applications for fibre reinforced polymer (FRP) wastes has led to a relative great amount of research work on recycling techniques and related potential added value applications. Mechanical recycling, with size reduction to fibrous and/or powdered products, has been considered the most cost-effective recycling technique, at least for relative low cost and clean thermoset FRP materials, but more feasible markets outlets for the recyclates should be identified. The most extensive research work in this field has been carried out on Portland cement and polymer concretes in which grinded FRP scrap has been incorporated either as reinforcement, aggregate or filler replacement. As function of specific concrete mix design formulation and sorting techniques of recyclates, reported added values comprise sometimes slight increase on mechanical properties, decrease of permeability, a less drying shrinkage and wear loss, and a global cost reduction of raw materials. Potential applications of FRP recyclates in concrete include precast paving slabs, roof tiles, railroad sleepers, wall panels, paving blocks and architectural cladding materials. However, these products have not been yet successfully reflected in the industry market.

This paper is aimed at enclosing and summarizing an updated review regarding all these issues with special emphasis on glass FRP (GFRP) wastes: recycling techniques, mix design formulations of GFRP recyclates modified concrete materials, end-use applications and market outlook.

Keywords: FRP Wastes; Recycling; GFRP recyclates modified concretes; Cost-effectiveness; Sustainability

1. Introduction

Worldwide volume production and consumption of fibre reinforced polymers (FRP) have increased in the last decades in several fields, mostly in the construction, automobile, aeronautic and wind energy sectors [1-4]. FRP composite materials are generally made of

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glass (GFRP), carbon (CFRP) or aramid (AFRP) reinforcing fibres dispersed in an organic matrix, usually polyester, epoxy or vinyl-ester thermoset resins. GFRP are by far the largest group of materials in the composites industry, representing over than 95% over the total of FRP composites [5]. According to Lucintel market report, a leading global management consulting and market research firm, the global glass fibre market is expected to grow at a Compound Annual Growth Rate of 5.4% over the five years-period 2015–2020 [6]. Although some contraction in specific market sectors (e.g. sheet and bulk moulding compounds), and in some European countries (e.g., Scandinavian countries and France), the last AVC market report of the European Composites Industry Association (EuCIA) also confirms the steady global growth of GFRP composites industry over the last three years and estimates the European-GFRP production by volume during 2014 in 1043 million tonnes [5].

Despite all the advantages of GFRP based products over traditional materials, the increasing production and consumption also drives to an increasing amount of GFRP wastes, either end-of-life (EoL) products or manufacturing rejects. Since FRP based products present, in general, a long life-span (20-25 years), end-of life disposable was not a major concern until few years ago. However, waste amount resulting from EoL GFRP products will increase strongly within the next few years, and this issue has become particularly worrying. Just the wind energy sector is expected to cover 15.7% of the total EU electricity demand by 2020 and 50% by 2050, and the resulting EoL wind turbine blade material, mainly constituted of GFRP based components, is estimated to reach 100,000 tons per year in Europe [4, 7]. Additionally, the total amount of production wastes per year of GFRP composite industry (e.g., non-conform products and manufacturing rejects) is also following the raising production. According to Fons Harbers, Chairmen of the European Composite Recycling Services Company (ECRC), the total combined volume of EoL and production waste generated by the GFRP composite market in Europe is expected to reach 304,000 tonnes by this year (2015) [8].

Taking into account the above figures, FRP waste management has become an important and concerning issue. Whereas thermoplastic based FRP materials can be easily recycled by remelting and remoulding, recyclability of thermosetting FRP with fibre recovering is a more difficult task due to inherent cross-linked nature of resin matrix [9, 10].

Until now, landfilling and incineration have been the most common end-routes for EoL thermoset FRP products and scrap material. However, considering the actual and impending EU framework legislation on waste management, as well as the increasing price of landfill taxes, these end-routes will be progressively unavailable. Waste management legislation focuses on dealing waste through waste hierarchy and will therefore put more pressure on solving FRP waste management through recycling and reuse [11]. In particular, Waste Framework Directive 2008/98/EC stipulates that "*Member States shall take the necessary measures designed to achieve that by 2020 a minimum of 70% (by weight) of non-hazardous construction and demolition waste…shall be prepared for re-use, recycled or undergo other material recovery*" [12]. Therefore, in the near future, due to these more restrictive EU directives, FRP suppliers could lose their market share to metals and other industries if they cannot ensure that their FRP components can be reused or recycled at the end of their service life cycle [11]. Thus, at the present time, the perceived lack of economical recyclability of thermoset FRP composites is more and more important and seen as a crucial barrier to the development or even continued use of these materials in some markets.

This increase awareness of environmental matters and the seeking for further sustainable materials have driven that several recycling techniques have been analysed and proposed for FRP composites, mainly for GFRP and CFRP waste materials. Although research on recycling methods is underway, related research on end-use applications for the recyclates is still at a very elementary stage; though, in order to be cost-effectives, recycling approaches should always embrace both interdependent issues.

The aim of this work is to enclose and summarize an updated review regarding all these features with special emphasis on GFRP wastes: available recycling techniques, end-use applications for the recyclates, especially, into concrete based materials, and market outlook.

2. Recycling Processes for thermoset FRP Wastes

At present, there are basically three main recycling processes that can be used to get some value from FRP thermoset materials: (a) incineration, with partial energy recovery from heat generated during combustion of the organic part; (b) thermal and/or chemical recycling, such as solvolysis, pyrolysis and similar decomposition processes, with partial recovering of energy and reinforcing fibres; and (c) mechanical recycling, involving the composite break-down by shredding, milling, comminution or other similar mechanical processes, resulting in size reduction to fibrous and/or powdered products. A detailed description of these methods can be found on Pickering [9, 13] and Asmatulu *et al*. [14]. The main key points are summarized in the following items.

2.1 Incineration and Co-incineration

Incineration of FRP scrap with energy recovery is listed as recycling method in some literature, but this feature is still up for debate. Incineration does recover part of the energy of the scrap materials whereas landfilling does not; however, air pollution resulting from incineration is a drawback of this method. On the other hand, the fibre and filler content of the materials still end up as landfilled waste, potentially becoming hazardous waste depending on chemical analysis of the ash [15]. According to the current legislation, limits are settled concerning levels of emissions to air, water and soil, and the residues from the incineration process should be minimized in their amount and harmfulness. The benefit of energy recovering is also discussable: calorific value will depend on the organic fraction and for typical GFRP/CFRP composites that accounts only for 30%-40% in weight. On the other hand, incinerator operators actually charge more for accepting FRP waste in order not to overload the system. Burning plastic wastes limits the amount of household waste that can be processed, which means that large volumes of domestic waste (of which there is an unlimited supply) must be sent to landfill [11]. At present, incineration, with partial energy recovering, as the first alternative to landfilling, is less and less considered as a cost-effective end-route for composite wastes.

Co-incineration in cement kilns constitutes a recent alternative end-route for GFRP wastes and is thought as a slightly better and cost-effective option as this offers combined material and energy recovering. GFRP typically contains E-glass, which is alumina-borosilicate, along with the organic resin and often calcium carbonate filler. When fed into a cement kiln the organic resin burns providing energy and the mineral constituents provide feedstock for the cement clinker, namely Si, Ca and Al. This means that no residue is left at the end. However, there is still a significant gate fee for this process. Also, the total amount of fuel replacement in cement kilns by GFRP waste is limited due to the presence of boron commonly found in the E-glass fibre reinforcement. More than 0,2% of boron oxide in the cement increases the setting time and reduces the early strength. In practical terms, this means that no more than about 10% of the fuel input to a cement kiln could be replaced by GFRP waste material if no significant effect on the performance of the cement is required [13]. Other drawbacks of co-incineration in cement kilns rely on the requirements that GFRP wastes must comply: fragments of composite waste should be smaller than a designated size (20 mm x 20mm), contain low concentrations of toxic materials and heavy metals, contain no foreign material (such as metal inserts or fasteners), have a specific calorific value (higher than 5000 kcal/kg), and must not generate dust such as pulverized glass fibres [16]. At present, co-incineration in cement kilns is commercially active in Germany through Compocycle and is supported by EuCIA [17].

2.2 Thermal/Chemical Recycling

For fibre and partial energy recovering, thermo-chemical decomposition processes have been applied, mostly for CFRP composite wastes due to the inherent economic value of carbon fibres. Although both the energy and material recovering, these recycling processes are only cost-effective in the areas where paybacks are the highest (high economic value of the fibres) and where the volume of material to be processed is large enough to justify the capital cost of the technical plant.

The most common thermal process is pyrolysis which consists on heating the scrap material in an inert atmosphere in order to recover the polymer material as oil. This kind of atmosphere prevents combustion, and as result the air pollution effects are less harmful in this process than in incineration. Another advantage is that the recovered oil can be used either as fuel or be refined to regenerate resin feedstock chemicals. As limitation of this technique, the surface fragilities induced by the thermal stress on the recovered fibres, reducing thus its original strength have been reported [10]. Oxidation in fluidised bed is another thermal process for FRP recycling and it consists in combusting the polymer matrix in a hot and oxygen-rich flow. Recovered fibres by this process are clean and show very little surface contamination by char deposition; though, strength and fibre length degradation also occur [9, 13]. Some recent researches have shown that specific etching processes can significantly recover the original strength of reinforcing fibres that has been previously damaged by a thermal treatment [18, 19], but this approach is still far away from an industrial realization.

The chemical methods of recycling involve dissolution of the resin by means of chemical products and are based on a reactive medium (e.g., catalytic solutions and supercritical fluids) under low temperature [20]. Being a thermal stress-free process allows the fibres to retain most of their original strength. Though, this method involves the use of hazardous solvents and, additionally, it requires the previous granulation of scrap material in order to improve the specific surface, which causes length reduction of recovered fibres. Reduced adhesion to polymer matrix in posterior applications is another common drawback of chemical recycling methods [10].

2.3 Mechanical Recycling

Among the recycling technologies available for thermoset FRP composite materials, the most mature technique is mechanical recycling, with size reduction by shredding, crushing or milling processes. The resultant recyclates, a mix of powdered and fibrous material, can be incorporated as filler or reinforcement replacement into new composite materials or as a closed-loop recycling process. This technique usually involves three steps: (a) Initial size reduction of scrap material in some primary crushing process to pieces in the order of 50-100 mm in size; (b) Final size reduction in jaw crushers, hammer or knife mills where the waste material is ground into a finer product ranging to 10 mm in size down to particles less than 50 µm; and (c) Sorting and classifying operations to grade the resultant recyclates into fractions of different size (through cyclones or air zig-zag separators combined with sieving techniques) [21]. Typically, the finer graded fractions are mostly of powdered nature with high proportion of filler and resin particles, whereas the coarser fractions tend to be of a fibrous nature where the particles have a high aspect ratio and fibre content [9]. Although mechanical recycling has been considered mostly for GFRP composites, in which reinforcing fibres have a relatively low economic value, it can also be applied to process CFRP wastes with environmental benefits as demonstrated by Howarth *et al*. [22].

Mechanical recycling shows significant environmental and economic advantages when compared to the previous recycling routes. In fact, mechanical size reduction: (a) does not produce atmospheric pollution by gas emission or water pollution by chemical solvents effluents; (b) does not require sophisticated, and expectably expensive, equipment like the ones that are required in the other processes; and (c) allows the processing of larger amounts of waste at higher throughputs. As drawbacks, two less attractive features have been pointed out: (a) safety issues due to risk of ignition during shredding process; and (b) the lower value of the final product hardly competitive with homologous virgin raw materials such as calcium carbonate or shopped glass fibres. Nevertheless, ensuring that economically viable end-use applications for the recyclates exist, mechanical recycling at industrial scale processing is so far considered the most suitable recovery technique, at least for relatively low cost and promoter-free FRP materials.

3. End-Use Applications for GFRP Recyclates

Over the last 25 years several end-use applications were investigated for mechanically recycled thermoset GFRP wastes or recovered glass fibres, either as raw material for new composites or into a closed-loop recycling process for the same source-material. In the envisioned applications, GFRP recyclates were applied as filler, reinforcement or core material replacement as follows: (a) filler material for artificial wood (Demura et al., 1995), high density polyethylene plastic lumber (George and Dillman, 2000), rubber pavement blocks (Itoh and Kaneko, 2002), dense bitumen macadam (Woodside et al., 2003), and bulk or sheet (BMC/SMC) moulding compounds (DeRosa et al., 2005); (b) reinforcement for wood particleboard (Reynolds et al., 2004) and soils (Ahmad et al., 2012; Mujah et al., 2013); and (c) core material for textile sandwich structures (Adolphs and Branca, 2001) [4]. Most of the foreseen applications have not succeeded for one or both of the following reasons: (a) tendency of the recyclate addition to negatively affect the mechanical properties of final composite; and (b) negative cost balance, where mechanical recycling and sorting operational costs outweighed the market value of the virgin product.

Among the several potential applications of mechanically recycled FRP wastes in new composite materials, a significant amount of research work have been carried out on Portland cement concrete in which the effect of GFRP recyclates, and more rarely CFRP

[4] All the references of above cited works can be found in Ribeiro *et al*. [23], and Meira Castro *et al.* [24].

recyclates, has been analysed and assessed either as reinforcement, aggregate or filler replacement [25-39]. In the analysed studies, a wide-range of replacement amounts was assessed: between 1% up to 20% in weight of total aggregates (after conversion of volume content to weight content). The applied FRP waste size fraction also differ widely, to relatively large pieces of GFRP or CFRP waste (5-30 mm square by 0,02-10 mm depth) [25, 26] down to very fine grade fractions with particle average diameter less than few microns [27-31, 39]. However, in the most part of research studies, GFRP waste addition consisted of fluffy mixtures of powdered and fibrous particulate material with different length of glass fibres [32-38]. Also, recovered glass fibres through pyrolysis recycling process were investigated for short reinforcement in cement mortars [40].

Besides the environmental benefits, and as function of specific mix design formulation, reported added values due to FRP recyclates incorporation in cement based materials include slight to strong decreases of permeability with subsequent improved durability [27, 29-31, 33], less drying shrinkage [27, 33], better workability [25, 28-30], reduced risk of cracking induced by restrained shrinkage [29, 33], improved fracture and tensile behaviour [26, 29, 38], higher thermal insulation [30, 39], and a global cost reduction of raw materials. In some particular cases, for lower sand replacement ratios, slender increases on compressive [32-37], splitting tensile [25, 33], and/or flexural strengths [26, 30, 32, 35-37], were observed. However, most of the times undesirable features were noticed such as significant losses in the mechanical properties (in most of the cases due to high water-cement ratio required to achieve the desirable workability) [25-29, 31, 32, 34-37, 39, 40], higher wear loss [31], higher setting times [28, 29], potential incompatibility problems derived from alkalis-silica reaction (depending upon glass fibre nature and content) [27-37], higher susceptibility to chloride ions penetration [40], and weak adhesion at recyclate-binder interface [25]. This last issue, commonly found in the design process of composite materials modified with recycled plastic, was also addressed in some research works through the combined incorporation of GFRP recyclates and chemical coupling agents [30].

The global outputs of part of the above research works was addressed by Yazdanbakhsh and Bank (as 2013) in their revision study [41]. The resultant main highlights, benefits and drawbacks of FRP waste incorporation into Portland cement concrete materials, are still valid up-today even considering the most recent advances in that field. As it states in their conclusions, in general, 'the partial replacement of aggregates in Portland cement concretes and mortars by mechanically recycled FRP wastes seem do not considerably affect the durability of the final cementitious materials, but significantly reduce their mechanical properties'. Also, the partial replacement of mineral aggregates by GFRP recyclates generally leads to minor decays in mechanical properties of final composite if fibrous GFRP waste fractions are applied instead of finely powdered GFRP recyclates.

Recently, some experiments were also carried out undertaken the incorporation of GFRP recyclates into polymer concretes (PC) and polymer mortars (PM) [23, 24, 42-45]. The effect of different replacement ratios of sand aggregates by both fine and coarse GFRP waste fractions on final mechanical properties of polyester based PM was assessed, as well as the effect of the incorporation of silane coupling agents. Obtained results showed that the partial replacement of sand aggregates by either of both GFRP waste fractions (up to 15% in weight of total aggregates) improves the compressive and flexural behaviours of resultant PC/PM materials. Lately, the *Global Fiberglass Solutions*^{TM} group (GFSI) also started to investigate this recycling route [46]. Comparing to the end-use applications in cementitious based concrete materials, already reported, the proposed solution overcomes some of the problems found, namely: (a) the possible incompatibilities problems due to alkalis-silica reaction; (b) the decrease in the mechanical properties, and (c) the poor bond GFRP recyclates and matrix binder. Taking into account obtained results, this last approach seems to be a very promising alternative end-route for mechanically recycled GFRP wastes in concrete materials.

4. Market Outlook and Future Perspectives

The intended perspectives for final applications of concrete and/or composite materials modified with GFRP recyclates include, among others, moulding compounds, precast slabs, paving blocks, railroad sleepers, wall panels, manhole covers, valve chambers, cement floor screeds, valley gutters, roofing sheet and flat sheets for signage; however, few of these products came out of the investigation field and had an industrialized expression. One of the few successful applications (manhole covers, utility boxes and urban furniture made of thermoset and glass fibre flakes wastes aggregated in a resin under high pressure cold moulding), was developed by *Reprocover*, in Belgium, and it has been commercialized since 2011 [46]. Nevertheless, apart from some in-house recycling (such as the above example), attempts to commercialize these products as a recycling route for GFRP wastes have failed. Regarding GFRP recyclates into PC materials, recently, this investigation line that was started by Ribeiro and co-workers in 2010 [23, 24, 42-45], also called the attention of *Global Fiberglass Solutions*^{TM} group. Over the last two years, this company has invested significant efforts on research and product development, and expect to commercialize final precast products for rail and roadways infrastructures under the trademark of '*Ecopolycrete*' [46]. Even so, and although all the efforts that had been done on developing cost-effective recycling routes, GFRP wastes still remain mired by the scarceness of reliable outlet markets for the recyclates and clearly developed recycling paths (logistics, infrastructures and recycling facilities) between waste producers and potential consumers for the recyclates. However, it is foreseen that this scenario will change in the next few years as strong investments are being made in this field.

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