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Current Technologies for Recycling Fiber-Reinforced Composites

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High strength, high toughness, and low weight make fiber-reinforced composite materials important as an alternative to traditional materials. Due to their application in different fields, such as construction, aviation, marine, automotive technologies and biomedicine, their production has increased leading to the increasement of composite wastes. New technologies for managing fiber-reinforced composite wastes have been developed to solve the issue of end-of-life of these materials. The aim of this paper is to emphasize recycling technologies used for fiber reinforced composites, and their potential reusage

Key words: **fiber reinforced composites; composite waste; recycling; "end-of-life"**

Introduction

THANKS to their highly desirable material properties, including high strength - to - weight and stiffness - to - weight ratios, and corrosion resistance, advanced fiber-reinforced polymer (FRP) composite materials have a significant increase in engineering applications: aerospace and defense, automobile, building, marine, and oil and gas industry [1, 2]. The global market size for composites was expected to rise from USD 74.0 billion in 2020 to USD 112.8 billion by 2025 [3]. Although there is a lack of standardization in manufacturing technologies of composites, there is a growing demand for FRP from the automobile and transport industry that are expected to have the highest rate in the composites market, especially due to the expected cost reduction of carbon fiber reinforced composites (CFRP). However, there are issues related to the recycling of composites that have to be solved [4]. The obvious lack of recyclability of composites is important since it is a significant barrier to the growth and sustained commercial use of fibers and other composite materials. As a short-term solution, landfilling could be an option; however, in this situation, the lack of recoverable materials must be substituted with novel or supporting materials. With less landfill allocation, recycled composites would also reduce resource consumption, meaning that fiber composites could continue in service for a longer period of time in the same or different applications [5].

The aim of this paper is to bring closer and explain the importance of recycling process of fiber reinforced composites (FRC), current technologies of recycling of FRC, possible applications of recycled materials, and its environmental impact.

FRC Recycling System

In order to conduct the recycling process of any FRC adequately, it is important to follow a chain of operations that depend on one another. First, the composite scrap, originating from the post-consumer products or manufacturing process, is collected and transported. The next step represents the main phase of operations of the recycling system. However, the latest recycling technologies used for FRC have trouble fulfilling the criteria of product quality, environmental legislation, and processing economics. In order to satisfy all the above requirements, there is a great need for more effective separation technologies. Nevertheless, the demand of recyclates with high quality and low price compared to the virgin FRP is one of the most important factors influencing the entire recycling system.

FRC recycling technology

The demand for FRC has grown over the past decades and is expected to increase. Because of wider applications of glass fiber reinforced plastics (GFRP) [6–8] and carbon fiber reinforced composites (CFRC) [9, 10], novel recycling technologies have been developed. These types of recycling are known as mechanical, chemical , and thermal [11, 12] (Fig.1). Nevertheless, a novel method such as the use of high-voltage fragmentation (HVF) help to implement carbon recycling as an environmentally safe, efficient and safe manner [13, 14].

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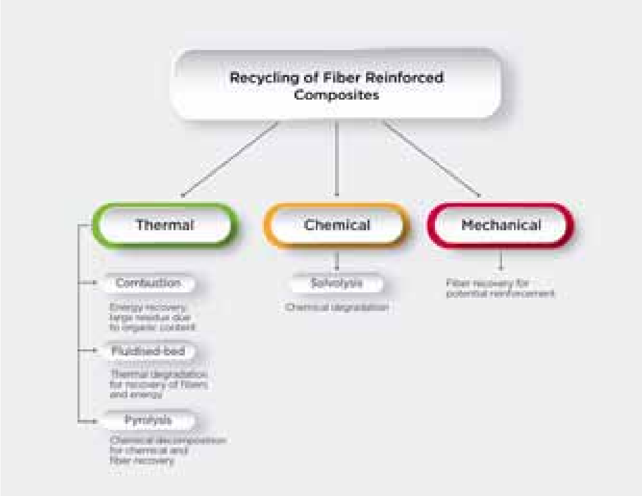

Figure 1. Recycling methods of fiber reinforced composites

Mechanical recycling

Multiple techniques of mechanical recycling of FRC have been investigated [15, 16]. Mechanical recycling is generally used in order to reduce the size of the composites. The process starts with cutting and grinding of the composites into smaller parts and sizing the different fragments to enhance the separation among the matrix and the fiber reinforcement [5]. Scrap composites are cut using slow cutting or crushing mills to aid in the removal of metal inserts and transportation to a size of 50-100 mm, while in the second stage high-speed mills or hammer mills are used to reduce the composite size to 10 – 50 mm [17, 18]. Ultimately, cyclones and sieves are used to grade recyclate volumes into various scale fractions [19]: fine powders [20], coarse [21] or fibrous [22]. There are many reports on possible uses of the obtained recyclates. Patel et al. (2019) reported about the lab-scale separation of glass fibers and polymer resin from the fiber-reinforced polymer acrylic waste. Low filler ratios were added to concrete mixes and the obtained results showed up to 20% enhanced compressive, split tensile, and flexural strength [23]. Caggiano et al. (2017) recovered steel fibers from cementitious composites and reused as substitution and/or addition of the industrial steel fibers. The obtained materials were tested in compression and bending and the results confirmed the possibility to reused as fillers without a significant decay in mechanical properties [24]. Meira Castro et al. (2014) used computational intelligence for the process improvement overcoming the limitations, obtaining a cost-effective process of incorporating GFRC recyclates as fillers in concrete-polymer composites with enhanced compressive and flexural strength [25]. Kočevar and Kržar (2018) used a hammer mill to separate 70% of the GF using 30% of waste as a filler in thermoplastics [26]. Yazdanbakhsh and Bank (2014) reviewed possible applications of FRP waste in concrete and mortar reporting several reduces of strength od cementitious materials and little effects on durability, and gave recommendations for producing stronger mortars and concretes by incorporating FRP wastes [27].

Latest findings have concentrated on alternative methods using high voltages to minimize bulk. Electrodynamic fragmentation (EDF) was one such technique in which a high voltage pulse between 50 and 200 kV was applied to ionized water to break down the CFRC waste into smaller parts [28, 29]. Mativenga et al. (2016) used high voltage fragmentation (HVF) with a high voltage pulse of 160 kV to breakdown the GFRC waste producing long and clean fibers [30].

Thermal recycling

Depending on the nature of the process, thermal recycling can be classified into three main groups, including combustion, fluidized bed, and pyrolysis [16, 26, 31].

Combustion

Combustion process of composite waste are performed in the presence of oxygen. Calorific values of fiber reinforced composites depend on the type, components, and matrix and reinforcement ratio. Thermosetting polymers are often recycled using combustion process due to its high calorific value and potential application as energy source [32]. The key reason why this process is considered unacceptable is the release of CO_2 and harmful emissions and several attempts have been made to explore alternative methods [33].

The obtained incombustible materials are usually disposed in landfills [34, 35] or used as fillers in cement [21–23]. Fraisse et al. (2016) have investigated the impact of using thermally recycled glass fibers in re-manufactured composite materials that exhibited lower fiber volume fraction, unchanged values of modulus of elasticity, and a significant reduction in the composites' maximum stress [31].

Fluidized bed fiber recovery

For the processing of thermoset composites in the form of process scrap or end-of-life products, a fluidized bed combustion process was created. Generally, it is used for the recovery glass and carbon fibers from thermoset composites [36]. The process includes reducing the size of the scrap to be fed into a fluidizing bed of silica sand in the presence of hot air with a specific velocity and temperature. A schematic preview of the fluidized bed combustion process is shown in Fig.2.

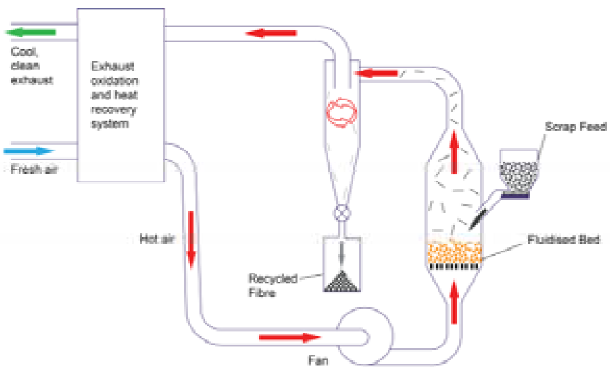

Figure 2. Fluidized bed fiber recovery

Pickering et al. (2000) used contaminated scrap, by process of fluidized bed decomposed the feeds at a bed temperature of 450°C and a fluidizing velocity of 1.3m/s. The obtained fibers were 5 mm long and at purity up to 80% [37]. However, the strength of the obtained fibers is usually reduced due to high temperatures during the fluidized bed process. Yip et al. (2002) used fluidized bed technology to recover carbon fibers from carbon fiber composite scrap. The obtained fibers were 10 mm long, retaining ~75% of their tensile strength and unchanged values of modulus of elasticity. It was concluded that the sand particle abrasion and high process temperature led to degradation of tensile strength of carbon fibers [38].

Pyrolysis

A two-step technique, pyrolysis, emerged as the most promising means of recycling not only precious materials, but also processing fuel and chemicals [39]. Pyrolysis makes it possible to extract the fibers via thermal decomposition of the polymeric resin, but the structural nature of the resins has not yet been restored [40]. However, it is important to conduct the pyrolysis process at an adequate temperature that would not deteriorate the structure of the recycled fibers affecting the strength of the fibers. The pyrolysis process of FRC is used to break down the resin network into lower molecular chains of organic compounds, liquid, gas, and solid carbon char products. Due to the possibility to reuse all waste components, it is believed that pyrolysis holds a great potential. Several studies were conducted on the pyrolysis of GFRC and carbon reinforced composites in the presence of nitrogen showing different results and output (gasses, solid, and oil) [40–43].

Torres et al. (2009) have investigated the pyrolysis process as an alternative method for recycling a thermoset composite of polyester and fiberglass using a laboratory stainless steel autoclave in the range of 300 to 700°C for 30 minutes confirming the theory that the obtained fiberglass could be used to replace virgin filler [44]. Kim et al. (2017) performed a pyrolysis treatment on carbon reinforced composite waste with a super-heated steam at 550°C in a fixed bed reactor for varying reaction temperatures concluding the strong dependance of the recycling efficiency on the pyrolysis temperature, reaction time, and super-heated steam rate [45].

Several studies [46–48] report the application of the microwaves for heating the waste in order to decompose the matrix into gasses and oil. The results showed higher calorific values than the amount of energy used for the recycling process. However, microwave assisted pyrolysis represents a new method with potential applications that need to be developed.

Chemical recycling

One of the major problems in composites recycling is the maintenance of the high-values mechanical properties. Chemical recycling of composite waste is the most effective and preferred choice for ensuring minimal damage to fiber properties. The process includes degradation of the resin using solvents: water, alcohols [49, 50], alkaline catalysts [51, 52], acidic catalysts [53].

Yuyan et al. (2009) used water as a solvent to decompose carbon fiber reinforced thermosetting epoxy composite. The results showed that the fibers were clean with no defects or cracks with tensile strength value 98.2% comparing to virgin fibers [54]. Zhao et al. (2020) performed an efficient recycling of fibers from carbon fiber reinforced polymer composites under a binary alkali solvent system of monoethanolamine containing 10 wt% potassium amine hydroxide at 160°C in 90 minutes. The results of the obtained carbon fibers suggested a small decrease in tensile strength of 6.5% [55]. Si et al. (2020) used thiols to degrade epoxy vitrimer successfully recycling carbon fibers that were used as reinforcement in composite material exhibiting great mechanical properties [56]. Liu et al. (2017) performed chemical recycling treatment of the carbon fiber reinforced composites using $ZnCl_2$/ethanol catalyst system at mild degradation temperature (<200 ºC) [57]. The decomposed polymer matrix was reused as an addition to new epoxy materials, the obtained results showed high strength and modulus. However, the process temperature influenced a mild damage to the recovered fibers. Yang et al. (2015) identified the main problems in degradation of glass fiber properties after chemical recycling treatment of composite wastes and developed two individual chemical treatments: chemical etching and post-salinization that lead to significant regeneration of fiber properties [58].

Conclusions

The key challenge for FRC recycling operations, with modern advances in recycling technologies followed by remanufacturing technologies, is the development of a stronger recycling chain that supports an effective selling of the recycled materials. In order to evaluate the proposed recycling methods for FRC, a great number of research studies has been published, and few of the key techniques in the part of the recycling process have been mentioned in this paper. The focus of this paper were several methods of FRP treatment, significance and application of different methods. It was concluded that chemical recycling offers the highest strength of fibers among the three main methods of FRP technologies. However, this method has been used in large part to recycle useful resin materials for re-use in new resins due to low price of virgin fibers. Mechanical recycling, led by thermal innovations, is the next supplier of high-strength fibers. Pyrolysis is currently commercially exploited. This work has shown that whether they are recycled or reused as composite structures rather than composite materials, composite products tend to have greater performance. Nevertheless, recycled fibers, particularly those made from long fibers, are unlikely to be incorporated in highly structural applications, i.e. new fibers would still be required. Although the recycling of FRC is on the right path, in order to actually make it a commercial reality, obstacles need to be removed and replaced.

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Trenutne tehnologije u reciklaži kompozita ojačanih vlaknima

Velika čvrstoća, velika žilavost i mala težina čine kompozitne materijale ojačane vlaknima važnim kao alternativu tradicionalnim materijalima. Zahvaljujući njihovoj primeni u različitim oblastima, kao što su građevinarstvo, vazduhoplovstvo, pomorstvo, automobilska industrija i biomedicina, njihova proizvodnja je povećana što je dovelo do povećanja kompozitnog otpada. Nove tehnologije za upravljanje kompozitnim otpadom ojačanim vlaknima razvijene su kako bi se rešilo pitanje kraja životnog ciklusa. Cilj ovog rada je da naglasi tehnologije reciklaže koje se koriste za kompozite ojačane vlaknima i njihova potencijalna ponovna upotreba.

Ključne reči: **vlaknima ojačani kompoziti; kompozitni otpad; reciklaža; „end-of-life“.**