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Cam Takviyeli Termoset Kompozit Atıkların Geri Dönüşüm Yöntemlerinin İncelenmesi

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Öz

Termoset reçine matrisli cam fiber takviyeli kompozitler istenilen mekanik özellikleri karşılamaları, hafiflikleri, korozyona karşı dirençli olmaları, kolay üretilebilirlikleri, esnek tasarım özellikleri ve çevresel etkilere karşı dayanıklı olmaları gibi avantajları nedeniyle tüm dünyada gün geçtikçe artan alan ve miktarlarda kullanılmaktadır. Termoset reçinelerin içerdikleri yoğun çapraz bağlar nedeniyle ısı ile yeniden şekillendirilmesi mümkün değildir. Bu özellikleri nedeniyle termoset reçine matrisli kompozit malzemeler kullanım ömürleri sonunda ekolojik ve çevresel açıdan sorun oluşturmaktadır. Bu çalışmada endüstride yaygın olarak kullanılan TsGFRP (Cam Elyaf Takviyeli Termoset Polimer) kompozit malzemelerin servis ömürlerini tamamladıklarında oluşan atıkların geri dönüşüm özellikleri incelenmiştir. Günümüzde farklı ihtiyaçların karşılanmasına yönelik çok farklı özelliklere sahip malzemeler tasarlanmakta ve büyük miktarlarda üretilmektedir. Bununla birlikte artan atık yükünün bertarafı konusunda sürdürülebilir çözümler üretilmesi vazgeçilmez bir gereklilik olarak karşımıza çıkmaktadır. TsGFRP kompozit malzemelerin geridönüşümü konusunda çeşitli çalışmalar yapılmıştır. Ancak bugüne kadar yapılan çalışmalar maliyet ve kazanım açısından değerlendirildiğinde etkin ve sürdürülebilir bir yöntem oluşturulabilmiş ve yaygın bir kullanım alanı sağlanabilmiş değildir. Bu bağlamda yeni araştırmalar yapılarak servis ömürlerini tamamlamış TsGFRP kompozit malzemelerin yeniden kullanımları ve geridönüşümlerine yönelik sürdürülebilir çözüm yöntemlerinin araştırılmasının öncelikli bir gereklilik olduğu anlaşılmıştır.

Anahtar Kelimeler
Ömrünü Tamamlamış Cam Elyaf Takviyeli Kompozit; Cam Elyaf Takviyeli Termoset Polimer; Geri Dönüşüm; Atık Bertarafı

An Investigation on Recycling of the Glass Fiber Reinforced Thermoset Composite Wastes

Abstract

Thermosetting resin matrix glass fiber reinforced composites are used in increasing areas and quantities all over the world due to their advantages such as meeting the desired mechanical properties, lightweight, resistant to corrosion, easy production, flexible design features, and resistant to environmental effects. However, it is not possible to reshape thermoset resins with heat due to the cross-linked networks. Due to these properties, composite materials with thermo-setting resin matrix cause ecological and environmental problems at the end of their life of usage. In this study, the recycling properties of end-of-life TsGFRP (Glass Fiber Reinforced Thermoset Polymer) composite waste materials, which are widely used in the industry were investigated. Today, materials with different properties are designed and produced in large quantities to meet advanced needs. However, it is an indispensable necessity to produce sustainable solutions for the disposal of the increasing waste load. Various studies have been carried out on the recycling of TsG-FRP composite materials. Unfortunately, the studies carried out so far, evaluated in terms of cost and gain, an effective and sustainable method has not been established and a widespread usage area has not been achieved. In this context, it has been understood that researching sustainable solution methods for the reuse and recycling of end-of-life TsGFRP comosite waste materials is a priority requirement in new studies

Keywords
End-of-life GFRP Composite; Glass Fiber Reinforced Thermoset Polymer; Recycling; Waste Treatment.

1. Introduction

GFRP1 Composite materials are design materials produced for achieving different properties and functionality. While fiber is used as a reinforcing element in these materials, thermosetting resins are used as matrix material in larger quantities than the reinforcing element in volume. Various fillers and dyestuffs can be added to the matrix according to the properties desired to be achieved in the designed material. The matrix also contains substances that require a chemical reaction. Glass fibers in the composite can exist in either a continuous or discontinuous phase (Demirel2008).

While glass fiber is used for increasing the mechanical strength in composite materials in general, matrix materials play an important role in increasing the corrosion resistance, distributing the incoming load and the integrity of the structure, while also playing an important role in determining the physical, chemical, thermal properties, and strength of the structure (Ianasi2020, Kaya2016)

GFRP composites can be produced with various production techniques that made them preferred for different purposes. Thanks to the wide advantages they provide, they are used in a very wide area and scale all over the world. The first use of glass-reinforced composites was seen in airplanes and boat hulls. Fiber Reinforced Composite materials first started with the use of aircraft and boat hulls in the field of transportation, and then with the development of different production techniques over time, the usage area has increased and continues to expand (IntKyn.1).

The size of the global composite market has reached 88.4 billion dollars as of 2019. Even though the composite market shrank to 74 billion dollars in 2020 due to the global economic crisis caused by the COVID-19 epidemic, it is estimated that the demand will revive and reach 112.8 billion dollars by 2025 with the recovery in the supply chains. Today, the Turkish composite material market has reached 1.5 billion Euros and a volume of 280,000 tons. The sector is growing in the range of 8-12% in Turkey, above the growth rate of Europe and the World (IntKyn.2).

Fiber Reinforced Composite materials are used all over the world in the fields of aviation, wind energy, consumer goods, marine, sports and entertainment equipment, transportation, defense industry, electrical electronics, pipes, tanks, and infrastructure. Fiber Reinforced Composite materials are used for many different purposes in the field of transportation due to their lightweight, superior resistance to water, corrosion, different weather conditions, and self-colouring abilities.

Construction and sub-sectors related to construction are the areas where fiber reinforced composite materials are used the most, due to their advantages such as strength/weight ratio, design flexibility, high resistance to weather conditions, and non-flammability. In the construction industry; It finds use in applications such as infrastructure (piping systems), structural parts, cladding panels, cladding panels (e.g. for prefabricated buildings), roofing tiles, pipes as well as bathroom furniture (e.g. bath and shower trays). Many decorative details such as columns and balustrades are carved from rigid foam, then coated with glass fiber and polyester resin, strengthened, and made resistant to weather conditions (IntKyn.1).

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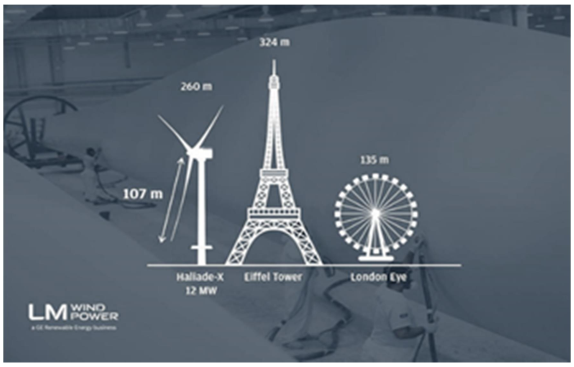

Figure 1. GE Haliade-X Wind Turbine (IntKyn.2).

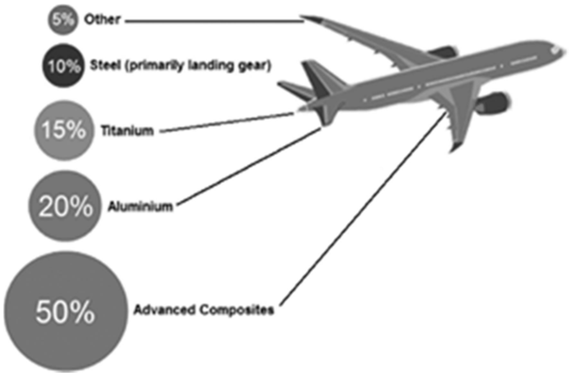

Figure 2. Materials of an airplane (IntKyn.2)

The benefits of GFRPs are listed below;

- Strength/weight ratio advantage and hardness,
- Easy and numerous production techniques, at low cost.
- Few production possibilities at low cost, (hand layup method) and unlimited moulding sizes
- Self colouring possibility and design flexibility and opportunity to harmonize with other materials
- Resistance to water, many chemicals, weather conditions, UV rays
- Good electrical and thermal properties
- Possibility of producing materials in different layers and in different combinations to obtain different mechanical properties
- In the volumetric phase, less energy is needed for the production of GRP than metals (Demirel2008).

Despite all advantages of GFRP composites, which are increasingly used in very large areas, end-of-life composite wastes pose a problem. With the developing technology, the increasing production amount of GFRP composite materials that meet different needs in the industry also causes a great increase in the waste load.

The ecological problems caused by the depletion of natural resources and the increasing waste load increase the importance of developing sustainable waste management solutions all over the World Commission on Environment and Development, published "Our Common Future Report", in April 1987. Sustainable development is defined as "development that meets the needs of the present without compromising the ability of future generations to meet their own needs" on that report. Therefore, environmental protection requirements should be considered manufacture and design of materials and taking into account future generations. Adopting sustainable development to industrial production has become one of the greatest challenges of the century. For this reason, there has been an increasing global trend in waste disposal. Designing and production plans should be done by considering waste disposal and environmental effects (Ming et al. 2021, Ulewics 2021).

Various studies have been carried out for years on the recycling properties of thermoset composites. The descriptions related to the technologies regarding the potential recycling and/or reusing methods for end-of-life composites including, thermal, material, a chemical recycling are given in the literature (Cunliffe and Williams 2003, Oliveux et al.).

Nouigues et al. evaluated the thermo-mechanical behaviours of composites to understand their recycling properties. Various static and dynamic tests at different temperatures (ambient-150°C) were applied over glass transition temperatures (Tg). Higher resistance results were obtained in static flexion and traction from woven glass fibers reinforced unsaturated polyester composites than mixed chopped and woven glass fibers. Results revealed that in the case of the end-of-life wastes total degree of deformation (hydrolyzed composites) is higher than production wastes (virgin composites), recycling end-of-life wastes would be easier than production wastes (Nouigues et al. 2020).

Re-using potential of Glass Reinforced Polyester (GRP) pipe dust, which is a waste that occurs during the manufacturing process, in GRP pipe production has been investigated by Memioğlu et al. GRP pipe dust, was used as 2.5%, 5%, 7.5% and 10% of the total amount of silica sand that is used for pipe producing. It has been determined that the manufacturing of GRP pipe, which has better mechanical properties, fulfil the required values defined in standards and can be designed for different kind of projects can be done by using waste pipe dust. Also, it has been presented that the integration of waste dust in production is a useful approach in terms of waste management (Memioğlu and Dağlı 2019).

A study of using Grp pipe production wet cutting stage wastes in situ $CaCO_3$ which is used as filler in PVC pipe production proposed by Özüyağlı et al. Wet cutting stage wastes consist of silica, unsaturated polyester resin, and glass fiber. It has been presented that using GFRP waste powder in PVC production as a filler result in high flexure and tensile strength and hardness while thus lowering the density of composite (Özüyağlı et al. 2016).

Beycioğlu et al. evaluated using the GFRP pipe production wastes powders as a filler in asphalt mixes by a lab test. Positive results were obtained from applied tests from 4.5% binder content, 3.75% GRP-WP, and 1.25% limestone filler content samples. Utilizing GFRP production waste powders in hot mix asphalt production is a good solution for GFRP waste powder recycling (Beycioğlu et al. 2020).

In another study, Cunliffe et al., evaluated mass balance, pyrolysis gas, and condensable products properties through pyrolyzing GRP/styrene copolymer waste in a fixed-bed reactor. Re-using recovered glass fibers from pyrolyzing was also considered in this study. It is presented that it is easy to obtain glass fibers from the mixture. It was found that glass fibers can be used in test plaques and sample pieces as DMC in polyester composites (Cunliffe and Williams 2003).

Sabau et al. studied determining new composites that consist of recovered waste glass fibers. The polyester was used as a matrix and waste glass fibers were used as reinforcement and sand as filler in this new composite. It is mentioned that the satisfying mechanical properties have been gathered from new composite materials (Sabau et al. 2012).

Machinability of GFRP Khan et al. evaluated the composite material using alumina cutting tools. The performance of the SiC whisker reinforced alumina cutting tool is better than that of the Ti[C,N] mixed alumina cutting tool on machining GFRP composite (Khan and Kumar 2010).

Yıldız et al. applied various lab tests on composites to evaluate the utilizing properties of GRP wastes. Mechanical size reduction was applied to GRP wastes and the obtained three different grain sizes were used as filler in the production of new composites. The hand lay-up moulding technique was recommended as a production method since it allows adding wastes during the production and thus it is easy to apply gel-coat. Lower linear shrinkage and mechanically derived from the applied tests. It is also observed that using higher grain sizes rises glass transition temperature. It is recommended as an environmental solution that utilizes the reduced wastes particles in composites' structure (Yıldız et al. 2020).

In another study implemented by Ribeiro et al., utilizing GFRP pultrusion production end-of-life wastes in new composite production possibilities was investigated. GFRP grinded wastes are utilized in the polyester matrix as filler and reinforcement at various size distributions and loads. It is presented that using GFRP wastes as filler material rises the flexural and compressive behaviour of the composites (Ribiero et al. 2014).

2. TsGFRP Composite Waste Management

Polymers and composites are divided into two different groups as thermoset and thermoplastic. Thermoplastics soften and melt with the effect of heat, they can be reshaped. They are easily soluble in suitable solvents. These features allow the development of various solutions for the recovery of thermoplastic composites. Thermoset resins, on the other hand, cannot be melted or reshaped by heat due to the intense cross-linked network. At high temperatures, they decompose as a result of chain and bond breaks. Thus covalent cross-linked networks limited TsGFRP composites' recyclability.

The European Parliament and the Council Waste Management Hierarchy;

1 - Prevention should be preferred as the first application. This part consists of the precautions that can be taken before the product is manufactured. Just as re-designing product content. Thus maybe using environmental friendly, sustainable raw materials aiming to produce easily recyclable and re-usable products.

2 -Reusing the waste should be the second preference. Reusing the tolerable damaged GFRP pipes in irrigation and drainage lines can be given as an example.

3 - Recycling is the third preference of the waste management hierarchy. The most preferred method of recycling is grinding the wastes in different sizes and utilizing these particles in other material production. It is difficult to find effective usage ara for that kind of production and the energy demand of grinding should be considered.

4 - Since the incineration process has its own wastes needed to be managed on account of environmental concerns "incineration" is considered as the last preference.

5 - The disposal method is used when it is not possible to use other options and it should be mentioned that landfilling is not legal in some countries. Unfortunately, limited recycling structure of GFRP composites typically comes out with landfilling (Bernates et al. 2021, Post et al. 2020).

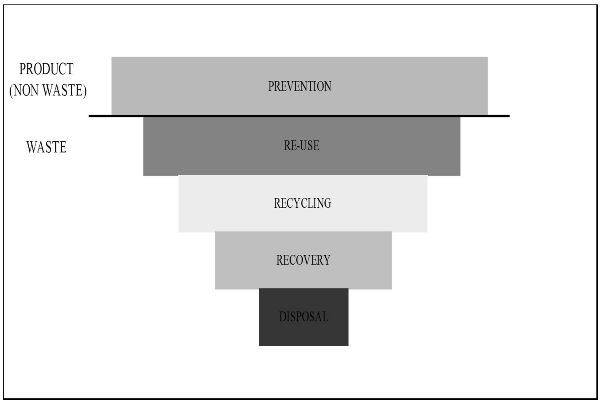

Figure 3. Waste management hierarchy (Bernates et al.)

3. TsGFRP Composite Waste Management

Mostly used TsGFRP composite waste recycling methods are chemical, thermal, and mechanical.

3.1. Chemical Methods

Chemical recycling can be defined as a dissolving matrix of composite waste materials in solvent or with heated water (<400°C) to recover valuable fillers and fibers or resin used as a matrix or monomers. This method is especially suitable for long fibers (Oliveux et al. 2012, Shuaib and Mativenga 2016).

3.2. Thermal Methods

Composite materials exposed with temperature treatment for leading recovery of intended reinforcement materials or energy sources.

3.2.1. Conventional Pyrolysis

Pyrolysing method depends on thermally degrading thermoset polymer with oven heating in an inert atmosphere. Recovered oil and gas from the pyrolyze process can be used as an energy source. Additionally, fiber sizing should be needed for use in other composites (Oliveux et al. 2012, Shuaib and Mativenga 2016).

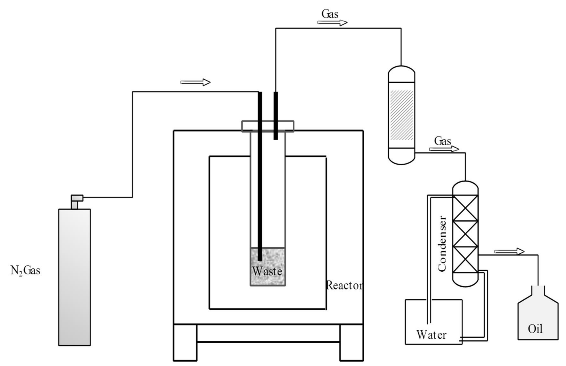

Figure 4. A diagram of the bench-scale pyrolysis reactor (Shuaib and Mativenga 2016).

3.2.2. Microwave Pyrolysis:

Microwave pyrolyzing method depends on degrading thermoset polymer with microwave heating in an inert atmosphere. Fastly heating process and low energy demand considered as a benefit but being is applicable in laboratory scale is a handicap of this method. Obtaining cost-effective products from the pyrolysis process (Shuaib and Mativenga 2016, Akesson et al. 2013).

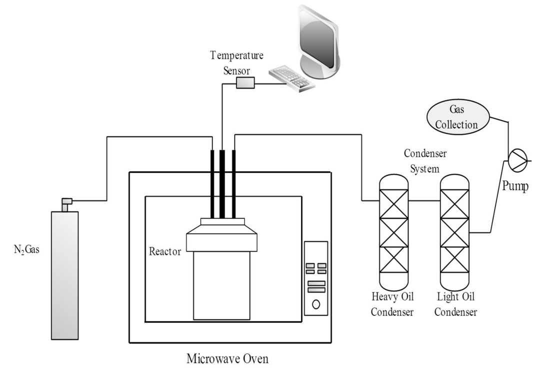

Figure 5. Schematic outline of the experimental set-up for micro-wave pyrolysis (Shuaib and Mativenga 2016).

3.2.3. Fluidized Bed:

The process depends on heating (450-650 °C) composites on a silica bed with an airstream for dissolving the matrix of composite materials for gaining reinforcement or fillers (Shuaib and Mativenga 2016).

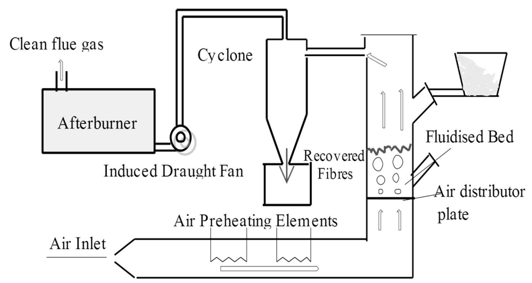

Figure 6. Schematic diagram of a fluidized bed thermal process

3.2.4. Incineration

TsGFRP composite wastes are valuable fuel sources such as coal because of their heating value. It is possible to reduce the waste volume by about 90–95% after incineration. Nonetheless, this process has residues such as fly ash and bottom ash which require treatment or disposal. This process occurs another wastes problem and this made it a noneffective method for recycling TsGFRP composite wastes (Kutluata 2009).

3.3. Mechanical Methods

The Mechanical recycling method depends on reducing the size of composite wastes to utilize recycles in another composite production as partial reinforcement or fillers.

Material sizes that are less than 100 mm in size are considered powder-rich and sizes around 5-10 mm in length are considered fiber-rich. It is applicable on an industrial scale. Limited secondary market values are a disadvantage of this method (Özüyağlı et al 2016, Shuaib and Mativenga 2016).

4. Conclusion and Recommendations

Composite materials are high-performance products produced with the latest technologies. They consist of many different fibers and fillers used in matrices with different properties for gaining the desired properties. That matter complicates the recycling properties of TsGFRP composite wastes. In addition to providing superior performance to the materials, the recycling properties of end-of-life TsGFRP composite wastes are very limited due to their intense covalent crosslinked bonds. Recycling methods of TsGFRP composite waste disposal methods have advantages and disadvantages, but the most important issue of these methods is being low value-added and typically requiring a high energy input and requiring additional costs to the manufacturers.

Since recycling and recovery infrastructures for TsGFRP composite end-of-life wastes have not been established yet, it is widely used as a landfilling and incineration disposal method. These methods, which are used because they are easily applicable and low-cost solutions, are not considered sustainable practices due to economic, social, and environmental concerns.

In the studies carried out to find solutions in this regard, it is aimed to use TsGFRP composite wastes as reinforcement and/or filling material in other composite structures by reducing the size of the wastes after mechanical processes, such as crushing, shredding, and grinding. In the literature, there are many studies on the use of TSGFRP composite wastes, which are reduced in size, as reinforcement or filling material. Among the recycling applications applied to TsGFRP composite waste, mechanical processes are accepted as the most effective and efficient technique with commercial potential. It also provides an advantage in terms of not having a direct environmental impact. Despite all its advantages and studies in the literature, its industrial potential is limited due to the limited availability of easy-to-apply and cost-effective end-uses and the lack of a secondary market for recycled materials.

Despite all the advantages of TsGFRP composite materials, the recycling of end-of-life TsGFRP composite wastes become challenging for manufacturers. For this reason, designing the composites to be produced as the raw material of another product at the end of their service life, and prioritizing recycling technologies with advanced separation and cleaning methods will be the best solution to the problem.

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