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## Recycling of polymer-matrix composites used in the aerospace industry- A comprehensive review

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### ABSTRACT

Polymer matrix composites are in high demand in many industries, including wind turbine blade manufacturing, pressure vessel manufacturing, and aerospace. In order to reduce unsustainable deconstruction that harms the environment and the potential litigation risk associated with the reintroduction of uncertified salvaged components to the aviation market, flight vehicle manufacturers have started collaborating with the recycling sector. Polymer matrix composites have strong mechanical qualities that make them appealing for a wide range of applications, but because of their heterogeneous nature, recovering composite waste is challenging. This paper compares various polymer-matrix composites (PMC) waste recycling techniques utilizing a multi-criteria decision-making approach and a literature study (MCDA). PMC waste recycling is becoming more popular, much work needs to be done to build a widespread, efficient system that is both financially sustainable and produces the highest-quality recovered materials.

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### 1. Introduction

In its most basic form, when two or more components are combined to create a single material with better qualities is known as composites. In reality, most composites are made up of a bulk material (the matrix) and some sort of reinforcement, which is generally used to boost the matrix's toughness and rigidity. This reinforcement could take the shape of fibers. Polymer matrix composites (PMCs), metal matrix composites (MMCs), and ceramic matrix composites (CMCs) make up the three primary categories of man-made composites used today [1]. The properties of polymer matrix composites that make them competitive and enable the replacement of traditional materials, such as wood and metal, are their lightness and mechanical strength in combination with their cheap relative production costs.

Materials that use organic polymers as the matrix and fibers as the reinforcement are called as polymer matrix composites (PMC) or fiber reinforced plastics (FRP). There are many PMC variants based on the type of polymer used, including amorphous and

semi-crystalline polymers, thermoset and thermoplastic polymers, epoxies, and non-epoxy polymers [2].

Low-viscosity resin is used to create thermoplastic polymers, which may alter their physical state in response to temperature. Composites made of thermoset materials cannot be melted, even after curing [1]. This makes it more difficult to recycle in exchange for a wider range of uses at higher temperatures. Epoxies, vinyl esters, and unsaturated polyesters are some of the commonly used types of thermosets. Matrix, which typically takes about 40–50% of the composite, keeps reinforcing fibers together. The composite material's reinforcement fibers, which can make up to 70% of it, are tougher and more powerful than the matrix [3]. Glass or aramid fibers are often employed in FRP carbon [1]. Due to their high specific mechanical properties, carbon-fiber reinforced polymers in particular are frequently utilised in a variety of industries (such as aerospace, aviation, autos, infrastructures, etc [4]). In the last ten years, the demand for carbon-fiber reinforced polymers nearly tripled, rising from 60 to 170 kilotons. According to Figure, the growth rate of the anticipated demand for these materials is expected to slow down over the next 20 years, although there is still a chance that the demand for carbon-fiber reinforced polymers will increase globally. Investigating individual fibre and polymer

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demand on the world market is equally fascinating. Only 500 million metric tonnes of the 8300 million metric tonnes of polymers produced between 1950 and 2015 were recycled. The enormous amount of waste produced by fiber-reinforced polymer composites made it imperative to develop effective recycling strategies. [4].

In-depth research and development have been put into polymer matrix composites (PMCs), which are employed in a range of technological fields, including aerospace. In aerospace industry the demand of composites is increasing due to its light weight, which uses less fuel and which in-turn saves money for the fuel, advanced lightweight substitutes for heavier materials are made possible by the ability of polymers as the matrix element and the high load-bearing properties of the reinforcing phase. Large passenger airplanes are increasingly using fiber-reinforced polymers (FRPs). FRP composites have developed into a different range of materials which can offer considerable benefits in terms of density and fatigue characteristics over traditional metallic materials. Because thermoset composites are lighter than similar metal constructions, the aeronautical industry has noticed a growth in their usage of them in aircraft, particularly in airliners. FRP composites now make up the majority of the structural mass of a number of commercial aircraft, including the Boeing 787 and Airbus A350 XWB. In the space industry, composites play a crucial role in the development and production of space habitats like the International Space Station as well as rockets, satellites, and other spacecraft. Despite the fact that PMCs, and in particular FRPs, are the most widely used composites in the aerospace industry and have exceptional strength-to-weight ratio potential, their use is unquestionably constrained by their inability to deform plastically. As a result, they can only absorb energy through damage generation, which leads to failure mechanisms like delamination, fiber-matrix debonding, and fibre fracture. Inadequate out-of-plane (impact) loading performances are also quite important.

Epoxyes are the most often utilized polymers as matrix in FRPs in the aerospace industry. Compared to other thermosetting polymers (such as polyesters), epoxyes are particularly suited to the aerospace industry due to their excellent mechanical properties, adhesion to substrates and fibers, resistance to moisture absorption and resistance to corrosive conditions. They also operate well at high temperatures because of their high glass transition temperatures.

This study compares different recycling techniques for FRP trash in order to assess their effectiveness in light of the selected sustainability criteria. To do this, a literature analysis was conducted. This sustainability performance evaluation is carried out to examine the applicability of MCDA for this type of study and to better understand the issues related to FRP waste recycling. In order to determine the best-performing approach based on the selected sustainability factors, this study compares several FRP waste recycling systems using multicriteria analysis.

Recycling offers a source of inexpensive, high-quality material, it can help to lower the high cost of raw materials for aerospace components. Recycling more complicated materials, such as polymer-matrix composites, is becoming increasingly feasible as recycling technology advances. This indicates that the aerospace industry's potential uses for recycled materials are growing. Overall, the future potential of recycling polymer-matrix composites utilised in the aerospace sector is positive. Recycling's economic and environmental advantages will become more evident as technology advances, prompting increasing investment in this field. Furthermore, the regulatory climate is projected to become more favourable to recycling, increasing the industry's expansion.

### 1.1. Trash production and recycling potential

An aircraft's manufacturer determines how long it will last. The number of pressurization cycles is used to determine an aircraft's life cycle. The duration of a flight during which an aircraft is kept under pressure is known as a "pressurization cycle." An airplane can be between 5 and 70 years old. A typical aircraft has a 20-year lifetime with a certain amount of flight cycles and hours. Most airplanes are transferred to the aircraft boneyard after their useful lives. Retired airplanes are kept at Aircraft Boneyard as storage.

By 2010, the most of materials employed by the air transport industry were composites, partly due to a dramatic increase in the use of composites as raw goods for production that started back in the 1970 s. Assessing that an aircraft has a lifespan of around 20 to 25 years, reclaiming or reworking the composite trash that has collected in the aviation sector over the years would be critical in three to five decades [5].

Composite materials account for about 18.7% of the resources used by aerospace companies to construct aircraft structures, but this figure is expected to rise in the coming years. A significant user of composites is the aviation sector [6]. Composites are widely used in the marine, pipelines and tanks, wind energy, construction, and other sectors, which increases the quantity of waste produced and the requirement for recycling. Because thermoset resin is the most often used composite (62%), and it is largely constituted of epoxy resin, the recycling and reuse of these kind of epoxy-based thermoset composites is critical [7 5].

### 1.2. Recycling waste composite materials

Recycling garbage is only ranked fourth among the best ways to manage waste by the European Union, with avoidance, reduction, and reuse being preferred over recycling. Mechanical, thermal, and chemical waste treatment techniques for FRP may generally be split into these three categories. Although the technological technique for recycling this form of garbage is the easiest, the final product is also the least valued. Chemical recycling techniques can be used to acquire the highest-grade material [8]. Chemical techniques are still mostly under research and are not employed in the industry [7].

#### 1.2.1. Mechanical recycling

The first step in recycling mechanical composite materials is to cut them into smaller pieces that have a maximum size of 50 to 100 mm. The pieces are then reduced in size to 10–50 mm using a hammer mill or comparable high-speed grinding equipment. Composite material is sorted into fiber-rich and matrix-rich components using cyclones and sieves [9]. Both sorting and the initial cut need less energy. The crushing operation consumes the majority of the energy [7 10].

This is the most popular technique of recycling FRP waste due to the technological ease of the process, however grinding significantly lowers the value of the materials. According to projections, the cost of carbon fiber utilized in the aerospace sector per kilogram must be 81.90 USD on average in 2018. However, this priceless raw material can only be used so much after crushing [7 10 11]. When carbon fibre and glass fibre are employed as reinforcing fibres, mechanical polymer matrix composites are formed.

Since the fibre are broken by crushing, thus larger particles can be used as reinforcing material for other FRP. Because the fibre particles are not completely detached from the polymer matrix after mechanical processing, they are tough to use as reinforcement in another FRP because they are not as tightly connected to the polymer matrix [7 10 11].

To alleviate adverse environmental effects of rising energy utilization for fibre creation, more recycled fibres must be used, as

seen by the disparity in energy-intensive recycling and production procedures [12]. Cement and concrete products, sculpting compounds, roofing materials, drainage, and different boxes made of mechanically processed polymer matrix composites are all examples of FRP applications.

When compared to alternative methods of recycling composite materials, the mechanical process has the benefit of being technologically simple. Both the processing of glass fibre reinforced polymer composites and carbon fibre reinforced polymer composites are acceptable and practicable. In terms of energy consumption, mechanical recycling is less hazardous to the environment than other recycling procedures [12]. Chemical processing procedures need between 63 and 91 MJ/kg, pyrolysis requires between 3 and 30 MJ/kg. Due to the significantly poor fibre quality, reuse is no more financially feasible after two recycling cycles [10].

### 1.2.2. Thermal recycling

Garbage is cooked at temperatures ranging from 300 to 700 °C without the use of oxygen during the pyrolysis process. According to one study, the lowest heating temperature for pyrolysis of wind turbine waste is 500 °C. Heating produces char and synthetic gas or oil as byproducts [13]. Although recovered fibres can be reused, the options are limited, and they may be badly damaged depending on the heating temperature.

Studies have shown that glass fiber-reinforced polymer matrix composites may be recovered with high-quality fibers at a temperature of 450–500 °C [14]. Carbon fiber is a more economically viable option for FRP reinforcement. Moreover, composites containing this fibre may be treated at temperatures ranging from 450 to 600 °C [10]. Still the information available shows that the glass fiber is mainly deteriorates during the pyrolysis process, lowering mechanical characteristics by 50% when compared to virgin filaments [15].

The oil generated during the process or synthetic gas (typically CH<sub>4</sub>, H<sub>2</sub>, CO, and CO<sub>2</sub>) is used for energy recovery, such as the pyrolysis process [13]. In theory, it is also possible to use oil to recover the chemical components needed to produce polymer resins; for example, PMMA's monomer may be recovered by pyrolysis. Different pyrolysis oils provide 15–20 MJ/kg of combustion energy compared to PMMA's 25 MJ/kg. Depending on the heating temperature, various oils, synthetic gases, and solids contribute differently to pyrolysis. The proportion of solids obtained is greater when heating at lower temperatures, whereas the percentage of oil and gas obtained is greater when heating at higher temperatures [5].

The pyrolysis process of recycling fibers can also be used to find a use for them in the automobile sector. Despite the numerous research on pyrolysis, there are no large-scale enterprises that make things from recycled glass or carbon fibre. Moreover, no quality comparisons exist between the same product when recycled fibres are used in place of brand-new fibers [15].

The main difference between microwave and conventional pyrolysis is that microwaves are utilised to heat FRP waste. Microwaves heat up rapidly, and the heating takes place within the recyclable material itself. Additionally, garbage is heated in an oxygen-free atmosphere in this type of pyrolysis. By heating the area around the material as it is heated from the inside, the recycling material lowers heat loss that happens during regular pyrolysis. This saves energy. [16] Microwave pyrolysis in an argon environment produced the highest quality result, and recycled fibre had almost the same tensile strength as new fibre. When carbon fibres are recovered using microwave pyrolysis, the surface is uniform and flat, with strength properties of 72% and a module of 90% of the new fibre. [16].

For the recovery of treated carbon and glass fibres for FRP recycling, a fluidized bed gasifier is ideal. The original polymer matrix

composite's waste is broken up into bits. They are then introduced into a tank of liquid silicon sand. Sand is liquefied at temperatures ranging from 450 to 550 °C using a hot air or nitrogen stream. The polymer matrix evaporates in a heated sand mass, releasing fibres and other components. Heated gases are used to transport solid particles to a "cyclone," where they are separated from the gas mass in a separate area. The afterburner receives polymer resin gases, which totally oxidize there at a temperature of 1000 °C and are then used for heat recovery [17]. The temperature at which the procedure is carried out has an impact on the quality of the treated fibers, much like with thermal processing techniques. The tests found that tensile strength declines less at lower temperatures than it does at higher temperatures.

### 1.2.3. Chemical recycling

The disadvantage of chemical processing is the requirement to apply it to recycled material, depending on the chemical structure. For example, the bulk of studies on epoxy resin matrix composites have been completed, and reinforcing fibres require diverse recycling fluids, time, environment, and temperature to get the optimum outcomes. Chemical recycling is also riskier than mechanical and thermal recycling since it involves potentially hazardous chemicals that might harm the environment and takes place at potentially dangerously high temperatures and pressure. Owing to these considerations, it is difficult to create an industrial-scale chemical FRP waste recycling facility since it is both costly and technically difficult [18–19].

Low-temperature chemical recycling, also known as solvolysis, occurs at temperatures less than 200 °C at normal atmospheric pressure. Throughout the method, acid or other solvents are used to break down the chemical linkages that make up the polymer matrix. In acid solvolysis, pre-treatment is required to hasten the polymer's chemical chain breakdown, particularly when processing FRP since it is made up of several laminas and is therefore simpler to split [13]. The chemical makeup of the recycled glass fibers used in this investigation caused variations in the quality of the materials. The amount of soluble chemicals recycled fibers have in the material used for solvolysis affects their quality. Al<sub>2</sub>O<sub>3</sub> and CaO, two more soluble chemicals, cause fibers to lose up to 60% of their original weight over time [20].

Water is heated to a temperature exceeding 374 °C and crushed to a pressure of 22.1 MPa. Other solvents, such as methanol, ethanol, propanol, and acetone, can be employed with glycols with catalysts and co-formulants to reduce the required temperature and pressure. This technology recovers high-quality recycled carbon and glass fibres from FRP.

In general, temperatures ranging from 230 °C to 500 °C were employed to investigate the chemical FRP waste recycling processes utilising supercritical water mixed to various liquids. Some studies have succeeded in obtaining recycled fibres that have lost only 0.08% of their weight while retaining their original tensile strength, while others have obtained non-recyclable materials that are not suitable for fulfilling new material functions, being used as a matrix, or being used as reinforcement [21]. However, research is conducted in the lab utilizing tiny FRP samples and a broad range of chemical combinations, experiment setups, and methodologies.

For supercritical solvolysis, other liquids such as methanol, ethanol, propanol, acetone, or glycol are used, as well as catalysts in the case of supercritical water solvolysis. Experiments that used these fluids as the principal element in solvolysis had great results in terms of recycled fibre quality, including glass and carbon fibre [22]. Many studies found that the mechanical properties of recycled fibre and fibre used to enhance fresh polymer matrix composites were equal. Nevertheless, the polymer matrices used in composite waste recycling are far from being commercially viable for supercritical solvolysis. The experimental design of the study serves as

proof of this. Small samples are investigated, the ideal solvolysis conditions are still being sought, different liquids, temperatures, and pressures are examined, as well as the machinery that is employed to make the recycling of chemical composite waste as effective as feasible [23].

## 2. Methodology and techniques

The polymer matrix composites are only minimally recycled. At the local or international level, no effective trash recycling system has been built. Despite its numerous potential uses, its widespread adoption is impeded by a lack of market for recovered polymer matrix composite waste. Another difficulty is the expensive and complicated technological aspects of recycling FRP. Not only should facilities be built so that FRP trash may be recycled, but the optimal recycling process should also be chosen. Due to the multiplicity of variables influencing the most effective option in terms of sustainability, making this decision is challenging. Economic, ecological, and social variables are often the three key aspects taken into account in this approach [5]. Despite the fact that almost all studies related to sustainability evaluation contain such a classification, the most recent research on the sustainability assessment analysed in this study has a bigger categorization of criteria that includes also technology and performance management [24].

The two primary categories of sustainability evaluation criteria quantitative and qualitative are separated into the aforementioned basic groupings. Each has advantages and disadvantages. Quantitative criteria can be measured and computed using standard measurement and calculation methods and procedures. Energy use (kWh/kg trash) and the amount of GHG emissions produced (tCO<sub>2</sub>/twaste) are two regularly used quantitative sustainability measures [25]. The qualitative aspects that must be quantified during the review process are based on surveys or the stakeholders' experience and general knowledge. Innovativeness and public perception are two examples of qualitative criteria. Qualitative criteria allow for studies with a broader variety of conclusions—conclusions for which there are inadequate numerical measurements or for which mathematical or physical units cannot be utilised to quantify them. Yet, applying qualitative criteria makes the results more arbitrary. However, as shown by the scholarly publications examined in this work, they are often utilized in sustainability assessment techniques that permit the inclusion of such criteria, such as multi-criteria analysis [26].

A range of sustainability evaluation approaches are used in the scientific literature on the recycling of FRP debris. While identifying the most appropriate kind of analysis for this study, it should be noted that multi-criteria analysis is commonly used in research related to the sustainability evaluation of various trash recycling technologies. This method has a wide range of applications since it allows for the selection of numerous indicator types and their application to the important assessment of a given process, as well as the precise determination of each criterion's significance through the assignment of a specific significance [27]. To evaluate the relevance of the criteria, both the subjective analytical hierarchy method and the objective entropy technique can be applied. The AHP is based on the paired comparisons concept, which compares each of the two indications and ranks them from 1 to 9 according to importance. One indicates that the criteria are equally important, whereas 9 indicates that the particular criterion is more important than the criterion it is being compared to. The AHP technique may also be used to normalise and assess one's own sustainability, however because it does not prioritise alternatives, it is simply utilised to rank the criteria in this study [5]. The assessment and normalization of sustainability performance are done using

TOPSIS (Technique for Order of Preference by Similarity to Ideal Solution) [28]. When doing MCDA, these two multicriteria analysis techniques are combined. These may, for example, be used to assess and compare the sustainability performance of various electronic waste recycling systems. Despite the fact that the specific piece of literature uses fuzzy numbers AHP and TOPSIS, the basic concept is the same as when utilising traditional MCDA methodologies.

A concrete and simple-to-understand result is achieved through a clear and sequential set of steps in a multi-criteria analysis. Since that the MCDA results may be arbitrary and influenced by the researchers' opinions, a sensitivity analysis is also performed to check the validity of the results.

## 3. Results and discussion

A detailed analysis was carried out for this work in order to assess and compare various polymer matrix composite material waste recycling systems. The literature research and analysis of the information that is currently accessible on FRP waste recycling alternatives are done in the introductory chapter. Based on this information, four recycling systems are chosen for further evaluation using specified sustainability criteria and multi-criteria decision-making analyses, particularly the analytical hierarchy process and TOPSIS. [5].

Eight sustainability criteria are chosen for MCDA based on information gained from literature research on FRP waste recycling systems and methodology guidelines for sustainability assessment framework. The four groupings of criteria—economic, ecological, technological, and social impact—represent the four fundamental SA aspects. Because certain FRP recycling systems have received more investigation than others and because MCDA requires comparable data, the criteria selection process is constrained by the knowledge that is already available [29]. Three criteria make up the majority of the technological criteria group, and workplace safety makes up the sole social criterion.

The Fig. 1 above provides the mechanical recycling technique has a score of 0.63, making it the closest to the ideal solution  $P_i$  of the four FRP waste recycling methods according to the results of the MCDA evaluation. Microwave pyrolysis comes in second place with a score of 0.59, followed by conventional pyrolysis with a score of 0.45, and supercritical water solvolysis with a score of 0.38. The greatest choice is just 0.13 units higher than the halfway point to the perfect answer, making all four options far from the optimum solution. Additionally, the resulting scores for the top and second-best performances are close. However, mechanical recycling outperforms supercritical water solvolysis by 40%.

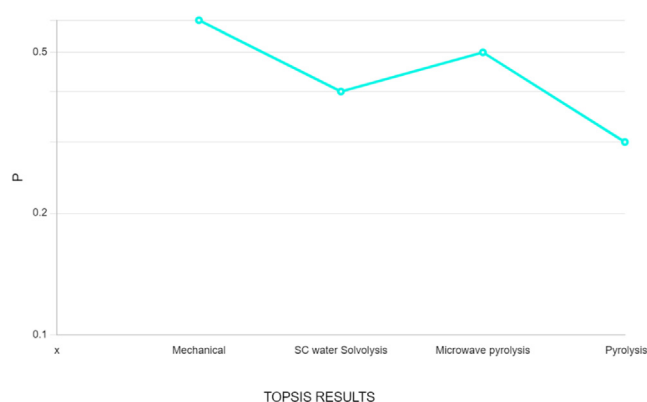


Fig. 1. TOPSIS Results [5].



For all of the examined FRP waste recycling processes, it is far from the best option, which can be understood by examining the patterns of criterion values. Some of the solutions that have been compared perform much better in some areas while having the poorest results in others. For instance, mechanical recycling performs well across six of the eight criteria, but poorly across the other two. Mechanical recycling, out of the four options considered in this study, is predicted to perform the best, but its performance is hampered by the fact that it is the least effective approach in other areas. In conclusion, although getting the highest assessment score, it has several shortcomings that are evident in how far it is from the perfect answer. [5].

Microwave pyrolysis is the second-best alternative, according to the outcomes of a multi-criteria decision-making study used to assess and compare the sustainability performance of four technologies for recycling fiber-reinforced plastic waste. The recycled fiber tensile strength performance of this FRP recycling process is the poorest, but it does not outperform any of the other options in any of the analyzed categories.

According to these findings and a review of the literature, the most sustainable approach for recycling FRP trash is being employed the most. One may argue that this outcome is indicative of the current state of FRP recycling in general. The most archaic technique that has relatively minimal environmental and economical costs, is mechanical recycling. Also, it does not provide high-quality recovered fibre or matrix. Other polymer matrix composite waste recycling techniques have greater ecological effects, but it should be noted that this study does not account for indirect effects, such as reduced global warming potential, which could potentially result in recovering higher-quality materials that could be used in high-technology composites applications, reducing the need for new FRP manufacturing, and saving both material and energy used for virgin fibre and polymer resin.

### 3.1. Conclusion

While FRP has several advantages over homogeneous materials, recycling FRP is challenging due to its composite nature. Industrial-scale application of chemical recycling, which yields the highest grade recovered materials, is not common [21]. Despite the fact that manufacturing polymer matrix composites is a costly and energy-intensive process, the vast majority of FRP plastic waste is landfilled. Recycling this sort of rubbish might reduce the amount of garbage discarded in landfills as well as the requirement for new FRP manufacture. As recycled fibre quality, chemical components recovered from polymer resin matrix, and perhaps energy recovered amount all improve, so does the complexity of technology. According to research, FRP may be chemically recycled to recover high-quality fibres that can be utilised in the same applications as virgin fibre. Another obstacle to creating a worldwide, industrial-scale FRP recycling system is the lack of demand for recovered reinforcing fiber. Based on the literature review, multi-criteria decision-making analysis, and sensitivity analysis performed in this paper to compare FRP waste recycling methods, it is determined that the mechanical recycling method is the most sustainable when the specific sustainability criteria used in this paper are applied. Further study on polymer matrix composite waste recycling methods is needed because there is a scarcity of knowledge on other possible sustainability criteria. If there was a greater range of correct data, MCDA results would be less reliant on a small number of criteria for which information is accessible.

### CRedit authorship contribution statement

**Nandini Ramawat:** Supervision, Resources, Investigation, Data curation, Conceptualization, Writing - original draft, Writing - review & editing. **Madhavchary A T Sanidhi:** Data curation.

### Data availability

Data will be made available on request.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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