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Evaluation of Polymer Matrix Composite Waste Recycling Methods

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***Abstract* – Polymer-based matrix composite materials are in high demand in many different fields: aeronautics, pressure vessel manufacturing, wind turbine blade manufacturing, and others. Due to the great mechanical properties of fiber reinforced plastics, it is a desirable material for various applications, but at the same time its heterogenic structure makes the composite waste hard to recycle. This paper focuses on different fiber reinforced plastics (FRP) waste recycling methods and their comparison by carrying out literature review and using multi-criteria decision making analysis (MCDA). Four polymer matrix composite waste recycling methods are compared to calculate which one has the best sustainability performance based on the chosen criteria. Analytical Hierarchy Process and TOPSIS are applied for criteria weighing and method comparison. Sensitivity analysis is used to evaluate the obtained results. It is concluded that more studies concerning different FRP waste recycling method sustainability performance need to be done, to derive more data, that would make MCDA more reliable and also other FRP waste recycling methods could be compared. Another conclusion is that different methods have different strengths which makes it hard to compare them. While FRP waste recycling is getting more broadly used, there still is a lot of work to establish wide spread effective system of FRP waste recycling that is both economically viable and gives the best results concerning recycled material quality.**

***Keywords* – AHP (analytical hierarchy process); FRP (fiber reinforced plastics); MCDA (multi-criteria decision making analysis); mechanical, sensitivity analysis; solvolysis, thermal, TOPSIS (The Technique for Order of Preference by Similarity to Ideal Solution)**

1. INTRODUCTION

Different types of polymer matrix composite materials are widely used in many different industries. Industrial manufacturing of FRP started in the beginning of the 20th century, but wider commercial consumption began in the 1980s in aeronautics [1]. Polymer matrix composite materials have been made for about a century. There is, however, no effective, globally working recycling system for this kind of waste. There are several FRP waste recycling methods, but not all are established enough to fully function on an industrial scale. This paper focuses on FRP waste recycling method comparison by carrying out literature

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review and using MCDA for comparing different recycling methods to evaluate their performance based on the chosen sustainability criteria. This sustainability performance assessment is done to help better understand the problems associated with FRP waste recycling and to assess MCDA applicability for this kind of analysis [2]. This study aims to compare various FRP waste recycling methods by using multicriteria analysis to find the best performing method based on the chosen sustainability indicators.

Composite material can be defined as a combination of at least two different materials that are combined to obtain a material with mechanical properties that are superior to its components. As opposed to alloys, the homogenic parts of composite materials do not mix with each other; they keep original chemical and physical properties [3]. The main parts of composites are reinforcement fiber and matrix. Polymer matrix composites can be thermoset or thermoplastic. Thermoplastic polymer is made from low viscosity resin that can change its physical state depending on temperature. Even when it is cured, thermoset composites cannot be melted. That gives more application possibilities in higher temperatures, but makes them harder to recycle. Some of the commonly used thermoset resin types are epoxy, vinyl-ester and unsaturated polyester. Matrix holds reinforcement fibers together and usually composes 40–50 % of the composite. Reinforcement fiber is harder and stronger than matrix and can constitute up to 70% of the composite material. For FRP carbon, glass or aramid fibers are mainly used. Reinforcement can be continuous or chopped. Continuous fibers that have smaller diameter are used for higher technology applications, for example in aerospace industry, due to their great mechanical performance that is determined by length-diameter ratio [3].

Composites of continuous fibers are mostly laminates in which each individual layer is positioned as to improve the properties of the laminate in the directions of primary load, thereby preventing the orthotropy. Laminates are extremely durable and rigid when the load is only received at an angle of 0° to the fiber direction, but when loaded, at 90° to the fibers, they are fragile because the load is on the polymer matrix. This is due to the fact that the high strength fiber can reach 3500 Mpa or more, but the usual strength of the polymer matrix is 35–75 MPa, so the matrix is much more fragile [3].

1.1. Waste

In the aircraft manufacturing industry 30–50 % of the FRP becomes manufacturing scrap [4]. No information is gathered on how much FRP manufacturing scrap or waste in general is generated. However, the amount of waste that can be recycled in the future can be deduced from the volume produced. This information is available for the most common polymer matrix composite materials CFRP and GFRP. In 2010 alone, 87 000 tons of GFRP and 2500 tons of CFRP were produced. Manufacturing scrap has not been included in this amount [4]. It can be concluded that incomplete information makes it difficult to implement a successful methodology for recycling the polymer composites and composite materials, since the introduction of recycling technologies requires the amount of waste to be recycled and the material to be recycled [4]. It should also be considered that polymer composite waste, both created in the production process and after usage, consists not only of the main parts – reinforcement and matrix, but also various substances used during the manufacturing process, for processing and preparation of FRP. Furthermore, in composite materials, in addition to their main components, various fillings may also be used, which partially perform the functions of the matrix [5].

In the information sources analysed during the study, the material flow analysis method was applied to determine the amount of waste generated for both the manufacturing scrap and usage waste. That is a systematic estimation of material flows and stocks within a given location and time. MPA can also be used to evaluate the efficiency of waste recycling [6], [7].

For example, in a study where four different FRP production units entered the data in the same system about the flow of composite material manufacturing scrap at the company, the data varied significantly in all four cases both in percentage of total manufacturing scrap (27 %, 15.5 %, 46 %, 7 %), and the percentage produced at certain stages of the production process, such as the amount of cured composite waste (5.5 %, 15 %, 20 %, 0 %) [4]. The most valuable manufacturing scrap is unprocessed reinforcement fibers, as they can be used in any other manufacturing process without any additional processing or processing operations with only minimal processing operations compared to any material, energy and work investments that are needed to recover the fiber particles from the already made composite material [7].

Residue from composite material in the prepreg is also easy to use for producing another composite material, however, there is no demand from consumers [8]. Prepreg waste must be pre-treated before delivering it to a recycling facility to prevent unwanted, potentially hazardous chemicals from entering the environment from the unprocessed material. Thus, without recycling or re-processing prepreg, it becomes a more complex material to recycle with lower potential for waste recycling [8].

FRP waste that is not counted as manufacturing scrap originates mainly for three reasons: depreciation, damage and due to substitution by some other product or component. Manufacturing scrap with good technical properties also ends up as waste. However, this waste could theoretically be upcycled or reused, transformed into other parts or products, thus reducing the cost needed for extending the life cycle of the material. Similarly, to FRP of manufacturing scrap flow, also polymer matrix composites end of life waste lacks comprehensive and reliable information. The data is only collected for individual industry-generated FRP waste. There is no separate indication of how much of it is manufacturing scrap. As well as the information studied during the research, only described the composite materials, where carbon fiber is used as reinforcement. The reason for this is that the carbon fiber is more expensive and a higher quality material with wider reusability options [1], [6], [9].

Already there are aircrafts being manufactured with a significant amount of carbon-fiber-reinforced composites, such as the Embraer ERJ, 20 % of which are approaching the end of their life cycle. (assuming that the life cycle of a commercial aircraft is around 25 years) [6]. Forecasts predict that by 2030 alone, six to eight thousand aircrafts in the United States of America and Europe will become waste due to wear and tear. By 2050, the amount of carbon fiber composite waste will reach 162 000 tons in the United States of America and 145 000 tonnes in Europe, and most of this waste will come directly from air transport. Air transport is the largest sector of carbon fiber consumption [8].

Different data sources provide different data on the percentage of the carbon fiber reinforced polymer composites market which is represented by each industry. According to the data analysed in this study, the aeronautics industry along with the wind turbine manufacturing industry will constitute no more than 56 % of the total carbon fiber market in 2022 or amount to 68 760 tons. Together with other carbon fiber consumer industries, the projected volume is 122 780 tons, only carbon fibers that need to be accompanied by an unknown amount of polymer material and fiberglass, as well as aramid fibers and other less common reinforcement fibers such as boron, SiC, Al_2O_3 and steel [9].

1.2. Recycling Waste Composite Materials

According to the European Union guidelines, waste recycling is only the fourth most desirable method of waste management, with options of prevention, minimization and reuse being more favourable. In general, the waste treatment methods of FRP can be divided into three groups: mechanical, thermal, chemical. Mechanical recycling methods for this kind of waste are the simplest in terms of the technological process, but the final product is also least valuable. The best quality material can be obtained using chemical recycling methods. Chemical methods are still mostly under development and are not commercially used [7].

1.3. Mechanical Recycling

The recycling process of mechanical composite materials begins with the splitting of the composite into smaller pieces with no dimension exceeding 50 to 100 mm. Then the size of pieces is reduced to 10–50 mm using a hammer mill or other high-speed grinding machine. Then, with the help of cyclones and sieves, pieces of composite material are sorted into two separate parts – fiber-rich and matrix-rich. Sorting and first cutting processes are less energy intensive. Most energy is consumed in the crushing process [10], [11].

Due to the technological simplicity of the process, this is the most used FRP waste recycling method, but grinding results in a significant reduction in the value of the materials. Forecasts show that in 2018 the average price per kilo of carbon fiber used in the aeronautics industry must be 81.90 USD [1]. However, after crushing, the use of this valuable raw material is limited. The processing of the mechanical polymer matrix composites is performed both when carbon fiber and glass fiber have been used as reinforcement fiber [11], [12].

Larger particles can be used as reinforcement material for other FRP, because the fibers are short, of a random size and the surface of the fibers are damaged by crushing. It should be noted that after mechanical processing, the fiber particles are not completely purified from the polymer matrix, which also makes it difficult to use them as reinforcement in another FRP because the fibers will not be as well connected to the polymer matrix as if the fiber surface was smooth.

The big difference in energy intensive recycling and manufacturing processes points to the need to use more of the recycled fibers in order to reduce the negative environmental impact caused by the high energy consumption required for fiber production [11], [12]. Examples of the use of FRP for real mechanically processed polymer matrix composites include: cement and concrete products, molding compounds, roofing materials, drainage, various boxes [11].

The advantage of the mechanical process is its technological simplicity when compared to other types of composite material recycling. It is equally suitable and feasible for the processing of both glass fiber and carbon fiber reinforcement polymer composites. In terms of energy consumption, mechanical recycling also has less environmental impact than other recycling methods. Pyrolysis consumes 3–30 MJ/kg, chemical processing methods 63–91 MJ/kg and high volatility fragmentation 4 MJ/kg [12]. After two recycling cycles, reuse is no longer economically feasible due to the very low fiber quality [13].

1.4. Thermal Recycling

Pyrolysis is a thermal waste recycling method in which waste is heated at 300–700 °C, without the presence of oxygen. In a study on pyrolysis of waste generated by the wind turbine industry, the lowest heating temperature is indicated at 500 °C [10]. After the heating process, char and synthetic gas or oil remain. Recovered fibers can be reused but, depending on the heating temperature, they can be very damaged and with little options for their use. Studies show that the temperature of 450–500 °C is suitable for recovering quality fibers from glass fiber reinforcement polymer matrix composites. It is more economically justifiable if carbon fiber is used as FRP reinforcement. Composites containing this fiber can also be processed at a higher temperature range of 450–600 °C. Available information suggests that glass fiber is highly degraded during the pyrolysis process, reducing mechanical properties by up to 50 % compared to virgin fibers, if the temperatures do not fit into the desired range and the prepreg is processed, where the polymer resin is not completely incorporated into the fiber [13], [14].

The char can be used in agriculture as a fertilizer, but the oil obtained during the process (mostly containing toluene, benzene and ethylbenzene) or synthetic gas (mostly containing – CH_4, H_2, CO and CO_2) is used for energy recovery, for example, for the pyrolysis process. It is also theoretically possible to use oil to recover the chemical elements needed to produce polymer resins, for example, PMMA can be pyrolyzed so that its monomer is recovered. PMMA combustion energy output is 25 MJ/kg, while other pyrolysis oils have lower combustion energy output of 15–20 MJ/kg. The ratio of oils, synthetic gases and solids to pyrolysis changes depending on the heating temperature. By heating at lower temperatures, the percentage of solids acquired is higher but by heating at higher temperatures – the oil and gas percentage is higher. This is valid for both CFRP and GFRP by recycling with pyrolysis [13].

In the automotive industry, it is also possible to find purpose for fibers recycled with the pyrolysis method. Despite the large volume of studies on pyrolysis, there are no industrial-scale factories that produce products using recycled glass or carbon fiber. There are also no comparisons of quality between the same product, where recycled fibers would be used instead of new fibers [14].

Microwave pyrolysis is very similar to pyrolysis; only microwaves are used for FRP waste heating. Microwaves heat up very quickly and the heating process takes place inside the recyclable material. Also, in this form of pyrolysis, waste is heated in an oxygen-free environment. As the material for recycling is heated from the inside, it reduces heat loss, which occurs during normal pyrolysis by heating the space around the material, thus energy is saved [15]. The highest quality product of microwave pyrolysis has been produced in argon environment, and the tensile strength of the recycled fiber is almost the same as that of the virgin fiber. However, it should be considered that the study was performed only in laboratory conditions on small material samples. By using microwave pyrolysis to recover carbon fibers, it has been determined that after processing, the surface of the fibers is homogeneous and smooth but has a tensile strength of 72 % and a module of 90 % of the new fiber [16].

Fluidized bed gasifier is suitable for FRP recycling to recover processed carbon and glass fibers. The waste of the initial polymer matrix composite is crushed into pieces. Then they are fed into a liquefied silicon sand tank. The sand is liquefied with a stream of hot air or nitrogen and can reach a temperature of 450 °C to 550 °C. In a hot sand mass, the polymer matrix is evaporated, thereby releasing fibers and other fillers. Solid particles with a flow of hot gases are brought to 'cyclone' and separated from the gas mass in a separate compartment. There they can be further sorted. Polymer resin gases come in the afterburner, where at 1000 °C they completely oxidize and are used for heat recovery [4]. As with the thermal processing methods, the quality of the processed fibers is also influenced by the temperature at which the process takes place. The studies conducted concluded that at lower temperatures tensile strength decreases less than at higher temperatures. For example, at 450 °C carbon fiber tensile strength is reduced by 50–52 %, but by 90 % at 650 °C. Similarly, to the pyrolysis process, the percentage of gases increases with increasing temperature and the rate of gas flow used for heating. In the fluidized bed gasifier, the carbon fiber retains a similar Jiang

Module, as it is for new fibers, as well as a similar surface texture, and recycling process does not reduce the fiber's ability to form links with resins [15], [17].

1.5. Chemical Recycling

Chemical FRP recycling process has various advantages but the drawback of chemical processing is a necessity to apply it to recycled material, depending on the chemical structure, i.e. for different types of polymers, for example, most of the studies have been done on epoxy resin matrix composites, and reinforcement fibers require different recycling fluids, time, environment and temperature to get the best possible result [18]. Chemical methods are also more dangerous compared to mechanical and thermal recycling, as potentially hazardous substances are being used that could harm the environment, as well as the process take places under extremely hazardous conditions, high temperatures and possibly high pressure. These reasons make it difficult to create an industrial-level chemical FRP waste recycling site because it is an expensive and technologically complex process.

Low temperature chemical recycling or solvolysis takes place at less than 200 °C under normal atmospheric pressure. The process uses acid or other solvents to break down the chemical bonds that make up the polymer matrix. In acid solvolysis, it is necessary to use pre-treatment to accelerate the process of chemical chain breakdown of the polymer, especially in the processing of FRP, which consists of several laminas, then it is easier to split. For example, eight-layer FRP waste can be recycled within an hour if pre-treatment has been applied, without it 15 hours are required for recycling [18].

The quality of the recycled glass fibers processed within this study differed from one another according to their chemical composition. The quality of recycled fibers is influenced by how much soluble compounds they contain in the substance used for solvolysis. Fibers containing more soluble compounds Al_2O_3 and CaO lose even 60 % of their initial weight over a long period of time [19].

Supercritical solvolysis – water is heated above 374 °C and compressed at 22.1 MPa. Other solvents, such as methanol, ethanol, propanol and acetone, or glycols with catalysts and co-formulants, can be used together to reduce the required temperature and pressure. This way, high quality recycled carbon and glass fibers from FRP can be recovered [15].

In general, the chemical FRP waste recycling methods with supercritical water, added to various liquids have been studied using a wide range of temperatures from 230 °C to 500 °C. The results vary widely, ranging from acquiring non-recyclable materials that are not suitable for fulfilling new material functions, being used as a matrix or reinforcement, as well as there are studies which succeeded to gain recycled fibers that have lost only 0.08 % of weight and retained their original tensile strength. However, studies are done at the laboratory using small FRP samples and widely varying substance combinations, experiment conditions and techniques [15], [18], [20].

Other liquids, such as methanol, ethanol, propanol, acetone or glycol, are used as catalysts in case of supercritical water solvolysis, are also used for supercritical solvolysis as main substances. In studies where these fluids have been used as the main substance for solvolysis, very good results have been obtained in terms of the quality of recycled fibers, both carbon and glass fiber. In some studies, the recycled fiber in terms of mechanical properties did not differ from the fiber used for the reinforcement of the new polymer matrix composites. However, for the supercritical solvolysis, the polymer matrices in composite waste recycling are far from being used in an economically viable way. This is evidenced by the experimental nature of the study. Small samples are studied, the best conditions for performing solvolysis are still being sought, not only the various liquids, temperatures and pressures are tested, but also the devices that are used to make the chemical composite waste recycling as efficient as possible [15].

2. METHODS AND METHODOLOGY

Only a small amount of the polymer matrix composites is recycled. No efficient waste recycling system has been established at local or global level. Its implementation is burdened by the lack of demand for recycled polymer matrix composite waste, despite having a wide range of potential usage. The technological complexity and costly recycling of FRP is also a challenge. By promoting a sustainability policy, especially in the EU with legislation such as the EU End of Life Vehicle Directive (2000/53/EC) and Directive (1999/31/EC) on Landfill of Waste, which limits composite disposal to landfill and creates more favourable conditions for the introduction of such recycling system [21], it is important not only to create facilities where the recycling of the FRP waste can happen, but also choose the best recycling method. Such choice is difficult because of the many factors that affect what is the most successful solution in the context of sustainability. In general, there are three main factors that are considered in such an analysis – economical, ecological and social [22]. Despite the fact that almost all studies related to sustainability assessment have such a categorization, the latest research on the sustainability assessment analysed during this study, has a broader categorization of criteria including also technological and performance management [23], [24].

The sustainability assessment criteria, which are divided into the above-mentioned main groups, can be of two types: quantitative and qualitative. Each of them has advantages and disadvantages. Quantitative criteria can be measured and calculated using standard measurement and calculation techniques and formulas. For example, in the context of sustainability, widely used quantitative criteria are energy consumption (kWh/kg_{waste}) and the amount of GHG emissions generated (tCO_2/t_{waste}). The qualitative criteria that require quantification during the assessment process are based on surveys or the stakeholders experience and common knowledge. Qualitative criteria include, for example, innovativeness, public attitude. Qualitative criteria make it possible to carry out a wider range of studies including conclusions for which there are not enough numerical measurements, or they cannot be measured by mathematical or physical units. However, qualitative criteria make the results more subjective [22]. Nonetheless, they are widely used in sustainability assessment methods that allow the use of such criteria, for example in multi-criteria analysis, as evidenced by the scientific articles analysed in this paper [25].

Analysing the available information on the polymer matrix waste recycling methods, various product analysis methods are more common, such as material flow analysis to find raw material flowing in, to calculate the amount of manufacturing scrap produced [6], [9]. Life-cycle assessment studies focusing on the evaluation of high-performance FRP products, wind turbines and aircraft are also available [11], [26]. There is also a study using life-cycle analysis concerning potential viability of using recycled carbon fiber [27]. However, in this context, the differences between waste recycling methods and the suitability of particular methods for recycling a certain product are not analysed using MCDA. Recently a study was conducted using multiple MCDA methods to rank renewable energy sources [28].

Various sustainability assessment tools are used in the scientific literature related to FRP waste recycling. However, when choosing the most appropriate type of analysis for this study, it should be considered that multi-criteria analysis is widely used in research related to sustainability assessment of different waste recycling processes [25], [29], [30]. This method has a wide range of applications, because it is possible to choose different types of indicators by applying them to the necessary evaluation of the particular process, as well as precisely defining the importance of each criterion by assigning a certain importance [31], [32]. The importance of the criteria can be determined by using subjective methods such as the analytical hierarchy process and the objective, such as the entropy method. The AHP is based on the principle of pairing comparisons, comparing each of the two indicators by determining the most important one, on a scale of 1 to 9. One means the criteria of equal importance, but 9 that the particular criterion is of absolute importance more than the one against which it is compared. The AHP method can also be used for normalization and self-assessment of sustainability, but it does not consider different alternatives at one level of importance, so it is used in this study only to determine the importance of criteria. TOPSIS is used for normalization and assessment of sustainability performance. These two methods of multi-criteria analysis are used together to perform MCDA, for example to use them to assess the sustainability performance of different electronic waste recycling methods and to compare them. The specific source of literature, however, uses fuzzy numbers AHP and TOPSIS, but the basic principle is the same as using standard MCDA methods.

Multi-criteria analysis has a clear and sequential set of actions that lead to a tangible and easily perceived result [23], [25]. Given that the MCDA obtained results can be subjective, affected by researchers' opinions, a sensitivity analysis is also performed to verify the reliability of the results obtained [33].

2.1. Criteria Selection

Based on the literature review about FRP waste recycling methods, eight sustainability performance criteria are chosen for carrying out MCDA to compare four alternative polymer matrix composite waste recycling methods (Table 1). Four different criteria groups are chosen for this study, to make sustainability analysis more comprehensive, based on guidelines for sustainability assessment framework [27]. Specific criteria are chosen based on available information obtained by reviewing literature in this paper. Three of eight criteria are qualitative, meaning their values are expressed in linguistic terms not numerical values. The criteria are – quality of recycled fiber surface, recycling method development level and workplace safety.

TABLE 1. CRITERIA FOR MULTICRITERIA ANALYSIS

K_i	Criteria group	K_{ij}	Criteria
K_1	Ecological	K_{11}	Energy consumption
		K_{12}	Global warming potential
K_2	Economical	K_{21}	Recovered fiber price
		K_{22}	Capital costs
K_3	Technological	K_{31}	Recovered fiber tensile strength
		K_{32}	Quality of recycled fiber surface
		K_{33}	Recycling method development level
K_4	Social	K_{41}	Workplace safety

2.2. Analytical Hierarchy Process

Analytical hierarchy process is carried out for criteria weighing. The process of performing an AHP comprises the following steps [31]:

1) Using pairwise comparison, a scale of 1 to 9 decision matrix is formed. 1 means equal importance, but 9 absolute importance over alternative criteria [25]. The decision matrix is shown in Table 2;

TABLE 2. AHP DECISION MAKING MATRIX

1.		K_{11}	K_{12}	K_{21}	K_{22}	K_{31}	K_{32}	K_{33}	K_{41}
2.	K_{11}	1.00	1.00	2.00	4.00	1.00	1.00	1.00	5.00
3.	K_{12}	1.00	1.00	0.50	4.00	3.00	3.00	1.00	5.00
4.	K_{21}	0.50	2.00	1.00	4.00	1.00	1.00	3.00	5.00
5.	K_{22}	0.25	0.25	0.25	1.00	0.25	0.25	1.00	3.00
6.	K_{31}	1.00	0.33	1.00	4.00	1.00	1.00	3.00	5.00
7.	K_{32}	1.00	0.33	1.00	4.00	1.00	1.00	3.00	5.00
8.	K_{33}	1.00	1.00	0.33	1.00	0.33	0.33	1.00	2.00
9.	K_{41}	0.20	0.20	0.20	0.33	0.20	0.20	0.50	1.00

2) To determine criteria weight, comparison matrix and B column vector is generated in Eq. (1) [33]:

$$Bij = \begin{bmatrix} b11 \\ b21 \\ \vdots \\ bn \end{bmatrix}. \tag{1}$$

3) To determine column b, Eq. (2) is utilized:

$$bij = \frac{aij}{\sum_{i=1}^{n} aij}. \tag{2}$$

4) n number of B column vector was combined in a matrix as Eq. (3) shows:

$$Cij = \begin{bmatrix} C11\, C12 \ldots C1n \\ C21\, C22 \ldots C2n \\ \vdots \quad \vdots \; \vdots \; \vdots \\ Cn1\, Cn2 \ldots Cnn \end{bmatrix}. \tag{3}$$

5) Percent importance distribution is derived, and priority vector or W column was calculated using Eq. (4):

$$W = \frac{\sum_{i=1}^{n} Cij}{n}. \tag{4}$$

Obtained criteria weight are shown in Table 3.

TABLE 3. CRITERIA WEIGHTS OBTAINED USING AHP

	K_{11}	K_{12}	K_{21}	K_{22}	K_{31}	K_{32}	K_{33}	K_{41}
Criteria weight, %	17 %	20 %	17 %	5 %	15 %	15 %	8 %	3 %

Table 3 shows that using AHP for weighing criteria used in this paper for MCDA analysis mostly criteria weighs are similar with K_{12} being the most important with 20 %, but three criteria have significantly lower importance, under 10 % with K_{41} being the least important – 3 %. Also, K_{22} and K_{33} have low importance with 5 % and 8 % respectively. K_1 and K_3 are two most important criteria in this study, ecological criteria having 37 % and technological criteria having 38 % importance. That combined is 75 % of all four. It should be noted that this criteria importance is subjective and calculated specifically for this study, while the AHP methodology is well established and applicable for different study fields. As is concluded in various studies [25], [34], [35].

2.3. Criteria Quantification

Quantification in this study is carried out by assigning values ranging from one to five. Where one is the worst grade and five is the best. The scale used is the same for all criteria, despite the fact that qualitative criteria are representative of different fields, for more straight forward obtainable results. This is done subjectively, although in other studies AHP is used for quantification.

Table 4 shows quantified values for all evaluated four methods and three qualitative criteria. Quality of recycled fiber surface is the best for microwave pyrolysis and SC water solvolysis. Both are given value five, based on information in literature review that these polymer matrix composite recycling methods can recover fiber without polymer matrix residue. Recycling method development level is the best for mechanical recycling, as well as workplace safety.

TABLE 4. QUANTIFIED CRITERIA VALUES

FRP recycling method	K_{ij}	K_{ij} quantified value
Mechanical	Quality of recycled fibre surface	1
Pyrolysis		3
Microwave pyrolysis		5
SC water solvolysis		5
Mechanical	Recycling method development level	5
Pyrolysis		4
Microwave pyrolysis		3
SC water solvolysis		2
Mechanical	Workplace safety	4
Pyrolysis		3
Microwave pyrolysis		3
SC water solvolysis		2

2.4. TOPSIS

Chosen criteria with quantified values are analysed using TOPSIS method. This is carried out through the following steps:

1) Design of comparison matrix. Information that is used in TOPSIS is presented in Table 5 about all four compared methods. The matrix is constructed from *m* alternatives and *n* criteria. Each column is each alternative, but each row – criteria. Every unit x_{ij} is criteria K_{ij} real value which belongs to alternative method *J*. Criteria values are obtained from various sources [11], [12], [15], [16], [20], [33], [35];

TABLE 5. DECISION MAKING MATRIX FOR TOPSIS ANALYSIS

Criteria group	Criteria	Mechanical	Pyrolysis	Microwave pyrolysis	SC water solvolysis
Ecological	Energy consumption, kWh/kg waste	0.05	8.33	2.78	17.5
	GWP, $kgCO_2eq$/kg waste	1.8	20.1	21	21.9
Economical	Price of recycled fiber, EUR/kg	5	13	15	19
	Capital cost, EUR	265 000	1 450 000	2 550 000	6 430 000
Technological	Recycled fiber tensile strength, %	1	0.9	0.72	1
	Recycled fiber surface quality	1	3	5	5
	Technology development level	5	4	3	2
Social	Workplace safety	4	3	3	2

2) Normalized value matrix is obtained using Eq. (5) [27]:

$$R_{ij} = \frac{x_{ij}}{(\sum_{J=1}^{n} x_{ij}^{2})^{0.5}}, \quad (5)$$

where
R_{ij} normalized value matrix;
x_{ij} every real value.

3) Weighted normalized matrix V_{ij} is then obtained multiplying every value from R_{ij} with its weight vector W_{ij} [27];
4) Positive ideal and negative ideal solutions are calculated using Eq. (6) and Eq. (7) [27]:

$$V^{+} = \frac{(_1^1(\frac{V_{ij}^{\max}}{j}),(\frac{V_{ij}^{\max}}{j'}))}{(i=1,2,\ldots,n)}, = (V_1^{+}, V_2^{+}, V_3^{+}, \ldots V_m^{+}), \quad (6)$$

$$V^{-} = \frac{(_1^1(\frac{V_{ij}^{\min}}{j}),(\frac{V_{ij}^{\min}}{j'}))}{(i=1,2,\ldots,n)}, = (V_1^{-}, V_2^{-}, V_3^{-}, \ldots V_m^{-}), \quad (7)$$

where
V^{+} positive ideal solution;
V^{-} negative ideal solution;
$j = (j = 1, 2, \ldots, m)$ for indicators that should have larger value for best solution;
$j' = (j = 1, 2, \ldots, m)$ for indicators that should have lower value for best solution.

Every alternatives distance to positive ideal solution and negative ideal solution is calculated using Eq. (8) and Eq. (9) [27]:

$$S_i^+ = S\left(\sum_{j=1}^{m}(V_{ij} - V_j^+)^2\right)^{0.5}, i = 1, 2, \ldots, n \quad (8)$$

$$S_i^- = S\left(\sum_{j=1}^{m}(V_{ij} - V_j^-)^2\right)^{0.5}, i = 1, 2, \ldots, n, \quad (9)$$

where S_i^+ is distance from positive ideal solution and S_i^- is distance from negative ideal solution.

The distance of every alternative to the ideal solution is calculated using Eq. (10) [27]:

$$P_i = \frac{S_i^-}{(S_i^+ + S_i^-)}, \quad (10)$$

where P_i is distance to ideal solution, in range 0 to 1, where 1 is ideal solution.

2.5. Sensitivity Analysis

Sensitivity analysis is applied to verify TOPSIS method result quality. It is done considering that TOPSIS method is affected by researcher's subjectivity. For sensitivity analysis framework proposed specifically for TOPSIS result analysing is used, originally applied for various water samples comparison [30]. It contains the following steps:

1) Supposing that weight vector is disturbed, and each disturbed value is labeled as w_k where $k = 1, 2, ..., n$ and w_k changes into w_k^*, $w_k^* = \gamma_k \cdot w_k$, γ_k is the initial variation ratio of w_k, which is a number larger than zero. As the sum of weight should be equal to one, the disturbance to w_k make other weight change, that can be seen in Eq. (11):

$$\begin{cases} w1' = \dfrac{w1}{w1 + w2 + \ldots + wk^* + \ldots + wn} = \dfrac{w1}{1 + (\gamma k - 1)wk} \\ w2' = \dfrac{w2}{w1 + w2 + \ldots + wk^* + \ldots + wn} = \dfrac{w2}{1 + (\gamma k - 1)wk} \\ \vdots \\ wk' = \dfrac{wk^*}{w1 + w2 + \ldots + wk^* + \ldots + wn} = \dfrac{wk^*}{1 + (\gamma k - 1)wk} \\ \vdots \\ wn' = \dfrac{wn}{w1 + w2 + \ldots + wk^* + \ldots + wn} = \dfrac{wn}{1 + (\gamma k - 1)wk} \end{cases}, \quad (11)$$

where w_1', w_2', …, w_n' are unitary weights for parameters 1, 2, ..., n after w_k is imposed on a disturbance.

2) Unitary variation ratio of w_k after disturbance is defined and expressed as seen in Eq. (12):

$$ßk = \frac{wk'}{wk}, \quad (12)$$

where β_1 is the unitary variation ratio of w_k.

TABLE 6. K_{11} DISTURBED CRITERIA WEIGHT IMPACT ON OTHER CRITERIA WEIGHTS

β_1	0.01	0.05	0.5	1	1.5	2	2.5	3	3.5	4
w_1	0.17	0.85	8.50	17.00	25.50	34.00	42.50	51.00	59.50	68.00
w_2	22.40	22.31	21.21	20.00	18.79	17.57	16.25	14.80	13.10	11.25
w_3	19.40	19.31	18.21	17.00	15.79	14.57	13.25	11.80	10.10	8.2
w_4	7.40	7.31	6.21	5.00	3.79	2.57	1.25	0.00	0.00	0.00
w_5	17.40	17.31	16.21	15.00	13.79	12.57	11.25	9.80	8.10	6.25
w_6	17.40	17.31	16.21	15.00	13.79	12.57	11.25	9.80	8.10	6.25
w_7	10.40	10.31	9.21	8.00	6.79	5.57	4.25	2.80	1.10	0.00
w_8	5.40	5.31	4.21	3.00	1.79	0.57	0.00	0.00	0.00	0.00

This paper for sensitivity analysis uses criteria weights used in MCDA, carried out to evaluate obtained data, 10 different values are assigned. Table 6 shows criteria weights after K_{11} weight vector is being disturbed. Such disturbance was assigned to four criteria in total – K_{11}, K_{31}, K_{32} and K_{41}. Criteria for sensitivity analysis was chosen considering different patterns they show, to make a more comprehensive comparison of how various criteria weigh disturbances affect results obtained by TOPSIS method and how easily these results can be manipulated. All criteria used for MCDA is not utilized for SA because of limited paper volume and similar patterns to those criteria that are evaluated, for example K_{12} pattern and weight is alike to K_{11}, therefore it is not necessary to analyse each of these two criteria. Results obtained by sensitivity analysis are shown in Fig. 1 and discussed in the next chapter.

3. RESULTS AND DISCUSSION

In this paper a comprehensive study is carried out to evaluate and compare different polymer matrix composite material waste recycling methods. In the introduction chapter, literature review is done, analysing available information on FRP waste recycling alternative methods. Based on this information, four recycling methods are chosen for further comparison using chosen sustainability criteria and carrying out multi-criteria decision making analysis, specifically analytical hierarchy process and TOPSIS.

Eight sustainability criteria are chosen for MCDA, as a result of obtained information from literature review of FRP waste recycling methods and methodology guidelines for sustainability assessment framework. Criteria can be divided in four groups, that represent SA main dimensions – economical, ecological, technological and social impact. Criteria selection is restrained by available information, because some FRP recycling methods are more researched than others, but for MCDA comparable data is necessary. Technological criteria group has the most criteria – three, and there is one social criteria – workplace safety.

The next step for criteria weighing is the analytical hierarchy process. This is a subjective MCDA method based on pairwise comparison methodology. Criteria weights are calculated using this method. Ecological and technological criteria groups having almost the same importance on sensitivity analysis carried out in this paper. Most important criteria are GWP (20 %) and energy consumption (17 %). Both of these are ecological criteria, which conclusively in this study is seen as the most important in evaluating sustainability performance and gives the biggest impact on MCDA results. Most of the other criteria have similar importance while workplace safety has 3 % importance rate.

After criteria weighing, three qualitative criteria are quantified so they can be used in the TOPSIS method. Quantification is done subjectively using scale of 1 to 5, assigning numerical values to linguistic ratings to each of qualitative criteria. As a result, the same criteria from alternative FRP waste recycling methods can be compared, because they are transformed to the same scale whereas before linguistic terms would not be applicable for carrying out economical assessment methods such as TOPSIS.

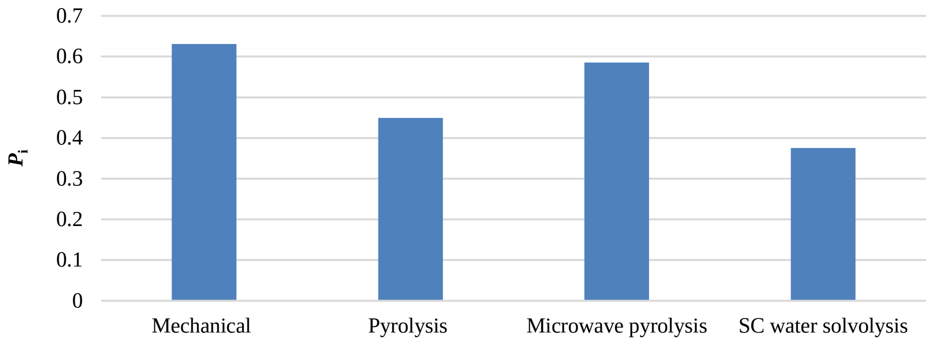


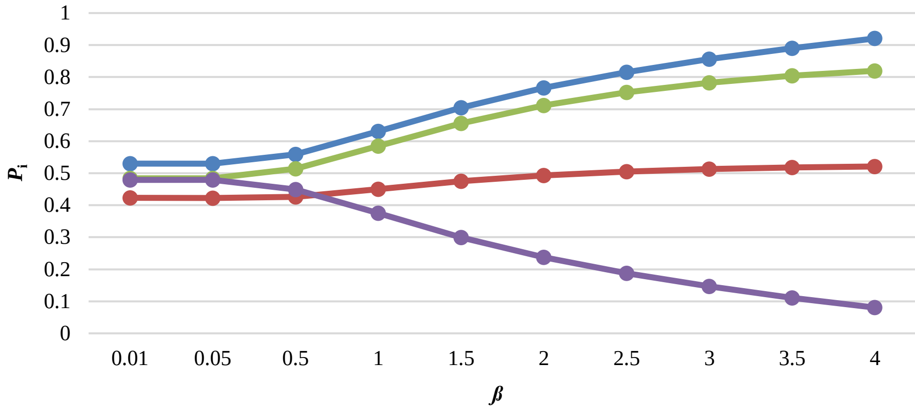
Fig. 1. TOPSIS results.

Finally, the TOPSIS calculations are done to determine the best option for FRP waste recycling in reference to sustainability performance based on the previous steps carried out in this paper. Obtained results are shown in Fig. 1. Results obtained from MCDA evaluating four FRP waste recycling methods show that mechanical recycling method is closest to ideal solution P_i, its obtained score is 0.63. Second best option is microwave pyrolysis with 0.59, then conventional pyrolysis 0.45 and supercritical water solvolysis with 0.38 respectively. All four methods are far from the ideal solution, best option being only 0.13 units higher than the one half of the distance to ideal solution. Furthermore, first and second best performances are close in terms of obtained score. But mechanical recycling shows 40 % better performance than supercritical water solvolysis.

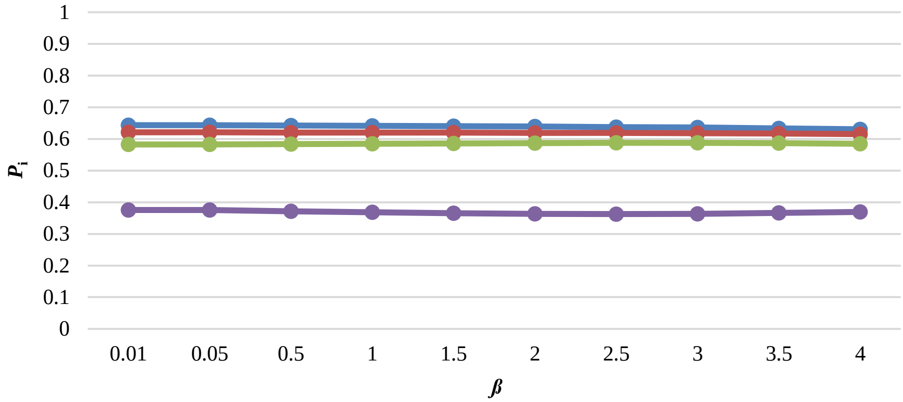
It is far from ideal solution for all of evaluated FRP waste recycling methods which can be explained by analysing criteria value patterns. Some of the compared options are performing much better in some fields while having the worst values in others. For example, mechanical recycling shows the best performance in six out of eight criteria, but has the worst performance in the other two. From this performance it could be anticipated that mechanical recycling is closest to ideal solution out of all four alternatives evaluated in this study, but its performance is hindered by being the worst performing method in other categories. In conclusion, while receiving the best evaluation result, it has some drawbacks which is reflected in its distance to ideal solution.

Second best option shown by results obtained by carrying out multi-criteria decision making analysis for evaluating and comparing sustainability performance of four fiber reinforced plastics waste recycling methods is microwave pyrolysis. This FRP recycling method has the worst performance in recycled fiber tensile strength but is not better than all of the other alternatives in any of the evaluated criteria.

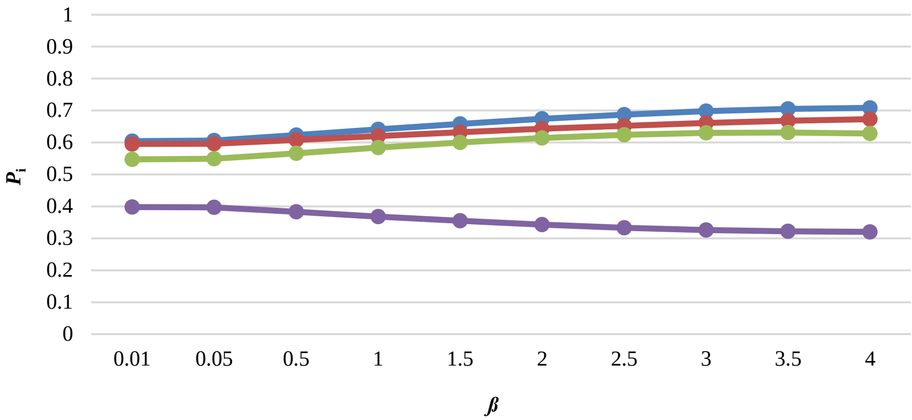
These results suggest that most sustainable option for FRP waste recycling is the most used, based on literature review. It can be argued that such a result is representative of current development level of FRP recycling in general. Mechanical recycling is the most primitive technology with relatively low cost both financial and ecological. At the same time, it does not provide high quality recovered fiber or matrix. Other polymer matrix composite material waste recycling methods have bigger ecological impact, but it should be noted that this study does not take into account indirect impact, meaning lowering global warming potential that could potentially lead to recovering higher quality materials that could be used in high technology composites application lowering the need for new FRP manufacturing, thus saving both material and energy used for virgin fiber and polymer resin production.



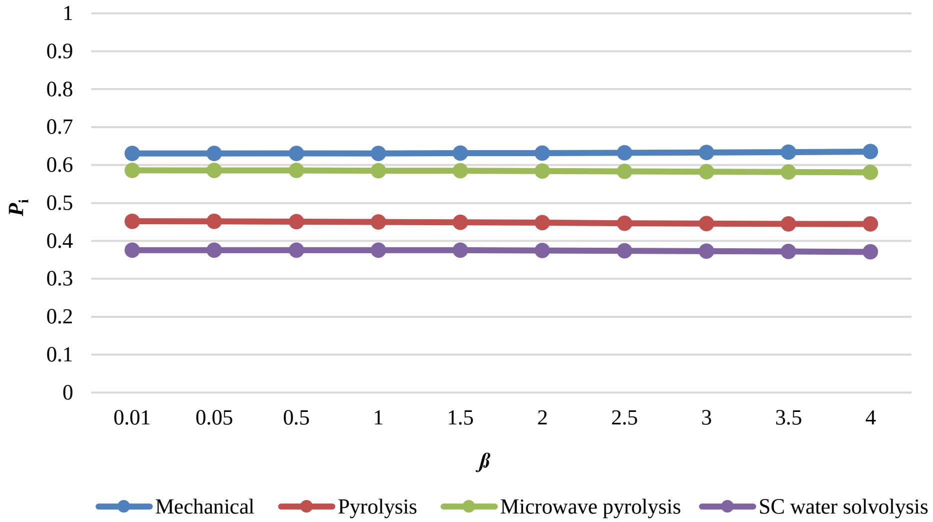
(a)



(b)



(c)



(d)

Fig. 2. Sensitivity analysis results by disturbing criteria weights a) K_{11}; b) K_{31}; c) K_{32}; d) K_{41}.

Further MCDA obtained results are evaluated using sensitivity analysis (SA), where four out of eight criteria weights are changed in order to gain more understanding of how sensitive the multicriteria analysis results are. Fig. 2 shows the obtained results. It is clear that every evaluated criteria in SA by changing its weight shows different impact on TOPSIS results. K_{11} changes do not change the ranking order calculated by MCDA if original criteria K_{11} weight is bigger than 0.5 of its original weight. Results are much more pronounced when talking about distance changes to ideal solution, where K_{11} weight is multiplied by four mechanical recycling method P_i is 0.92, much closer to ideal solution than TOPSIS result, at the same time SC water solvolysis recycling method in these conditions has P_i of 0.08 much lower than original result. When K_{11} weight is only 0.01 of 17 % results are changed significantly, SC water solvolysis and microwave pyrolysis having almost identical, but mechanical recycling still has the best performance. Conventional pyrolysis has the worst result. This shows that greatly lowering values that benefit one specific alternative can have a considerable impact on MCDA results. K_{31} and K_{32} changes in b) and c) graphics show no significant changes. While in both conventional pyrolysis and microwave pyrolysis have switched ranks, mechanical and SC water solvolysis, has the best and the worst performance respectively. K_{41} changes has almost no impact on original TOPSIS results, this can be explained by its only 3 % weight, being multiplied by four it still is only 12 %, furthermore it benefits FRP mechanical recycling method.

4. CONCLUSIONS

Polymer matrix composites are widely used in many different fields of applications, ranging from aircraft to sporting goods. While FRP offers many advantages over homogenous materials, their composite structure makes them complicated to recycle. That is one of the reasons why there is no global scale polymer matrix composite waste recycling system. Fiber reinforced plastics waste recycling is done in only a few recycling plants. Chemical recycling which offers the best results in terms of recovered material quality is not used on an industrial scale [20]. FRP plastic waste is mostly landfilled even though polymer matrix composites manufacturing is an expensive and energy intensive process. Potential recycling of this kind of waste would not only reduce landfilled waste amount but also decrease the need for new FRP manufacturing especially if used in high technology applications. Technological complexity grows with improving results in recycled fiber quality, recovered chemical elements from polymer resin matrix and possibly recovered amount of energy. Studies suggest that chemical recycling of FRP can recover high quality fibers that can be used for the same purpose as virgin fiber [15]. Lack of demand for recycled reinforcement fiber is another restricting factor for implementing global industrial scale FRP recycling system.

Based on literature review, multicriteria decision making analysis and sensitivity analysis in this paper, made to compare FRP waste recycling methods, it is concluded that mechanical recycling method is the most sustainable when specific sustainability criteria used in this paper are applied. But lack of information concerning different FRP waste recycling methods must be acknowledged. More studies about polymer matrix composite waste recycling methods need to be carried out, because available data about other potential sustainability criteria are lacking. A larger range of reliable data would make MCDA results less dependent on few criteria about which information can be found.

Another conclusion is that different methods have very pronounced different strengths that makes them hard to compare. For this reason, MCDA is suitable to make a comparison by using various criteria that represent diverse fields, for example global warming potential and quality of recovered fiber. MCDA offers a system that is easy to follow and carry out. Furthermore, TOPSIS results were examined using sensitivity analysis showing that slight changes do not affect results much, but disturbing criteria weight a lot can greatly impact calculated results, especially if specific criteria weigh is already significant and it benefits or has a negative impact on one alternative much more than others. This shows the importance of professionally done criteria choosing and weighing process.

FRP waste recycling issue is getting more and more pressing. There still is a lot of work to do to establish a widespread effective system for FRP waste recycling that is both economically viable and gives the best results concerning recycled material quality.

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