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CHEMICAL RECYCLING OF CARBON FIBRE-REINFORCED EPOXY COMPOSITES

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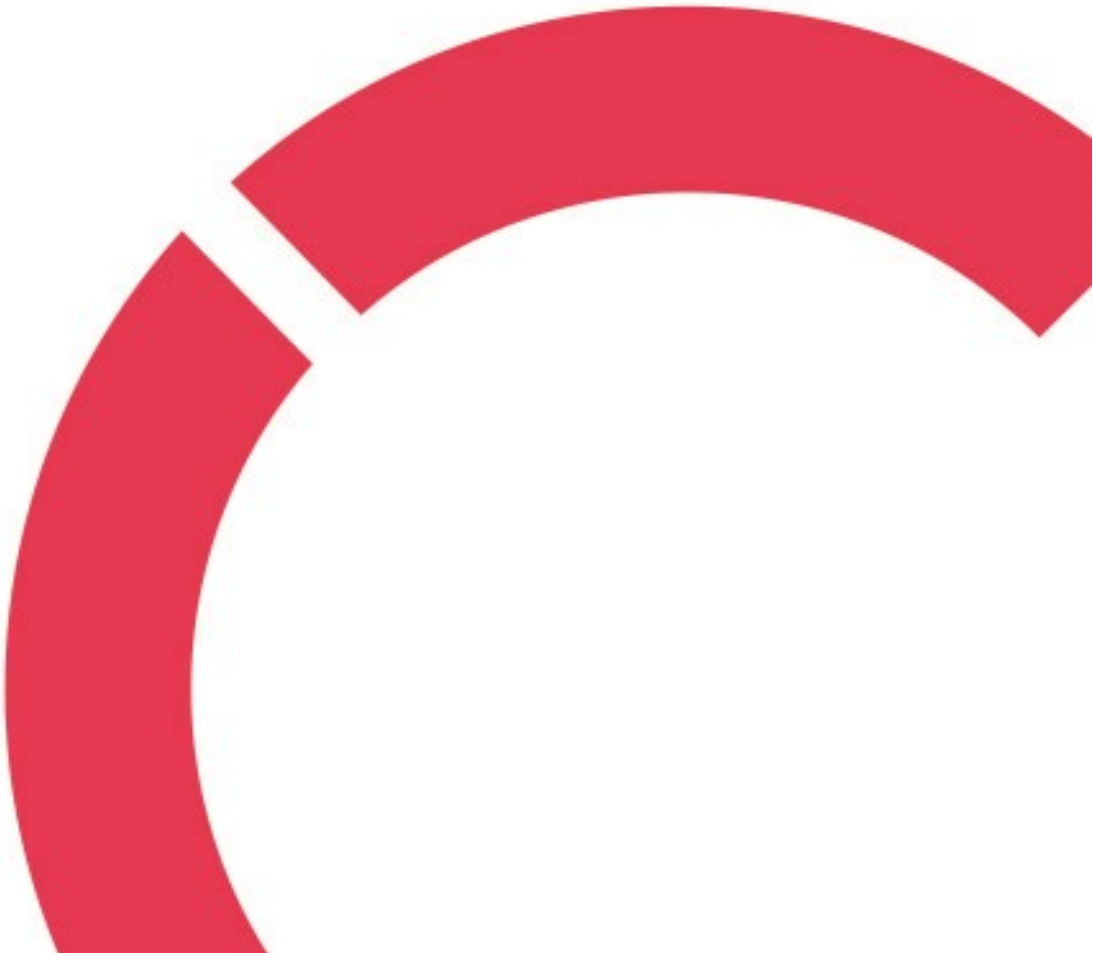

ABSTRACT

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In this literature review the recent studies conducted on chemical recycling using higher and mild reaction conditions are covered. The goal of this thesis was to conduct a review, collect data and compare different chemical recycling methods reported in the literature for recovering carbon fibres with good mechanical properties from carbon fibre-reinforced epoxy composite material, ensuring a good yield on epoxy resin, while promoting the use of organic or green solvents and catalysts.

The use and possibilities of higher reaction condition chemical recycling including solvolysis and hydrolysis are covered and compared. In the reviewed articles, it was found that by using higher temperatures and pressure with the aid of a catalyst the reaction time could be reduced while maintaining the mechanical properties of the recovered carbon fibre. However, it was found that there is insufficient information about the pathways to recover the resin itself and the used solvent. Multiple studies reviewed in this thesis showed that there are possibilities to use a bench plant while using higher reaction conditions while maintaining the woven structure of the fabric.

The reviewed articles covering the use of mild reaction conditions showed good potential for energy efficiency, but on the other hand, seemed to be using more expensive solvents. Although the solvents, for example, ionic liquids and deep eutectic solvents covered in this thesis, are being considered more expensive, they were stated to be green, sustainable, and possess good reusability. The mechanical properties of the recovered fibres are found to be good in both higher and mild reaction condition methods but mild reaction conditions showed more promise in recovering the degradation products from epoxy resin and reusing the solvents.

This thesis covers the most used and promising methods while introducing novel approaches to the topic. The scale and budget of future laboratory testing will help guide the choosing of method and solvent because it might be the determining factor whether to use more expensive solvents with mild reaction conditions, or high reaction conditions with less expensive solvents. The data collected from future laboratory testing will help guide the optimisation of process parameters.

Key words
Carbon fibre, carbon fibre-reinforced epoxy composite, chemical recycling, epoxy resin, higher reaction conditions, mild reaction conditions, solvolysis.

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CONCEPT DEFINITIONS

Curing agent

Is used in certain plastics and resins to initiate the polymerization cross-linking process (Kumar, Pallapa, Rezai & Selvaganapathy 2016).

Flexural strength

A parameter that defines materials bending resistance under load (Pal, Pramanik, Verma, Naqvi, Manna & Kar 2022, 243—270).

Hydrophilicity

Materials property to interact readily with water and letting water be absorbed in the material (Maeda, Endo & Hotta 2015, 213—234).

Prepregs

A composite material where reinforcement materials such as carbon fibres are pre-impregnated with matrix materials such as polymer resin (Seferis, Hillermeier & Buehler 2000, 701—736).

Solvolysis

A solvent reaction, in which the rupture of one or more bonds occurs in the reacting solute. (Muller 1994, 1077—1184).

Strain

“Increase in length per unit original length of the gauge which is expressed as a dimensionless ratio, or as a percentage (%)” (SFS-EN ISO 527-1:2019:en).

Tensile strength

Observed maximum stress, where the test subject breaks or yields (SFS-EN ISO 527-1:2019:en).

Young’s modulus

Indicates an object's stiffness and can be defined based on the object's amount of deformation under an applied force (Grimvall 1999, 46—69).

ABBREVIATIONS

CBT
Cyclic butylene terephthalate

CF
Carbon fibre

CFREC
Carbon fibre-reinforced epoxy composite

DES
Deep eutectic solvent

DGEBA
Bisphenol A Diglycidyl ether

DMF
N, N-dimethylformamide

EG
Ethylene glycol

GF
Glass fibre

HVF
High voltage fragmentation

KOH
Potassium hydroxide

NMP

N-methyl-2-pyrrolidone

MEA

Monoethanolamine

SEM

Scanning electron microscopy

TBD

1,5,7-triazabicyclo [4,4,0] dec-5-ene

TCA

Tricarballylic acid

TER

Thermoset epoxy resin

TABLES

FIGURES

1 INTRODUCTION

Fibre-reinforced polymer composites, specifically carbon fibre (CF)- and glass fibre (GF)-reinforced thermoset polymers are widely used in various industries. Those industries, such as aerospace, energy, and automobile to name a few, prefer the use of these composite materials due to their exceptional properties. The growing need for these composites is also growing the need for more sustainable ways to recycle them. The most common practises which are landfills and incineration should be replaced for their negative impact on the environment. (Karuppannan Gopalraj & Kärki 2020; Zhang, Chevali, Wang & Wang 2020; Shen, Apraku & Zhu 2023, 9644—9658; Danish, Mosaberpanah, Salim, Amran, Fediuk, Ozbakkaloglu & Rashid 2022.) Recent statistics show that by 2021 the amount of composites produced globally was 12.1 million tons. In Europe alone, it was 2962 kilotons of which 1.250 kilotons were thermoset composites that possess better mechanical and thermal properties compared to thermoplastic composites but are also more difficult to recycle. (Witten & Mathes 2022.) Due to their permanent structure after curing, they do not easily degrade. Alongside the research for new recycling methods, there is research about developing innovative thermoset composites with better recyclability and incorporation of bio-based materials. (Wu Klingler, Bifulco, Polisi, Huang & Gaan 2023; Venu, Jayan, Saritha & Joseph 2022; Sharma, Kumar, Rana, Sahoo, Jamil, Kumar, Sharma, Li, Kumar, Eldin & Abbas 2023, 2975—3002; Kumar, Mishra, Verma, Aldosari, Maity, Verma, Patel & Thakur 2023.)

Epoxy resin is a widely used thermoset polymer, which is usually reinforced with CF or GF. The wide use of epoxy thermosets and composites in e.g., wind turbines are driving the epoxy resin market to reach a capacity of USD 37.3 billion by 2025, the installed capacity already being over 651 GW. As the products reach their end-of-life, there will be annually 2 million tons of wind blade waste, with the global amount reaching 43 million tons by 2050. (Zhao, Long, Xu, Liu, Chen & Wang 2023, 72—97; Sommer & Walther 2021, 265—275.) Due to the vast amount of waste, containing CF, it is preferable to recycle it while maintaining good mechanical conditions for the fibres. Finding an optimized method for chemical recycling would benefit the economical aspect because studies have shown that the manufacturing of virgin CF consumes approximately 198—594 MJ/kg of energy and costs 33—66 USD/kg when the energy consumption of recycled carbon fibres are 10.3—35.7 MJ/kg and costs 13—19 USD/kg. (Oliveux, Dandy & Leeke 2015, 61—99; Giorgini, Benelli, Brancolini & Mazzocchetti 2020; Tapper, Longana, Norton, Potter & Hamerton 2020.)

In this literature review type of thesis, the latest studies from the last decade concerning the chemical recycling of carbon fibre-reinforced epoxy composites (CFREC) are collected and reviewed. The thesis is done in consideration of finding methods of chemical recycling and comparing which methods have the lowest environmental impact based on process conditions and used solvents. Process conditions are kept in consideration based on reaction time, temperature, and pressure because those aspects affect energy consumption and therefore environmental impact. The solvents and catalysts are compared if they are considered organic, green, and sustainable. The goal is to collect methods for comparison and discuss the properties which could potentially be further used and optimized to a new method. Optimizing a feasible method for chemical recycling of CFREC could help resolve the environmental issue of the current state of recycling of CFREC.

Review articles in chapter three covers the theoretical background at the subject, in which the different methods including chemical, thermal, mechanical, and other methods are discussed. The databases of Science Direct, ACS, Springer Link and Google Scholar were utilized in the search, with the use of certain keywords. The most used search words for finding studies were carbon fibre, epoxy resin, carbon fibre-reinforced epoxy composites, chemical recycling, solvolysis, mild reaction conditions, and composite waste. One of the criteria used for selecting the articles was the age of the articles, the goal was to keep the range in the years 2013—2023 because the most recent information is essential for research purposes. In addition, other criteria were to use review articles for theoretical background, and peer-viewed research articles for comparing studies. From the review articles studies could also be found and compared. Chapters four and five utilize the peer-viewed research articles for comparison and discussion of the used methods. Chapter four discusses recycling methods with sub- and supercritical fluids and chapter five discusses recycling methods using mild reaction conditions and acid digestion. The most important information for understanding the discussed methods is tabulated in each sub-chapter of chapters four and five. The use of scanning electron microscopy (SEM) images in each discussed study is helping the reader to understand the results based on the visual appearance of recovered CFs. The discussion and comparison of different methods aim to help in concluding an optimized method for future stages of the project and testing for laboratory work.

2 AIM OF THE STUDY

The aim of this thesis was to discuss and compare different methods considering chemical recycling of CFREC. The goal was to compare the reaction conditions of the methods and help optimize a method for future laboratory testing and piloting, which will ultimately contribute to finding an environment-friendly method that can also be utilized in recycling bigger sized CFREC waste in the future. This thesis aims to bring more knowledge in resolving the current state of recycling of CFREC waste and understanding the reuse possibilities of the recovered fibres, resin, and solvents from the process.

The research questions of this literature review type of thesis are: (i) What the current situation on recycling of CFREC is? (ii) What is the status of recent research development activities for ensuring the mechanical properties of recycled CFs obtained through chemical recycling? (iii) What are the new developments in chemical recycling of CF to obtain higher CF yield by using green organic solvents? The structure of this thesis can be seen in the flowsheet presented in FIGURE 1.

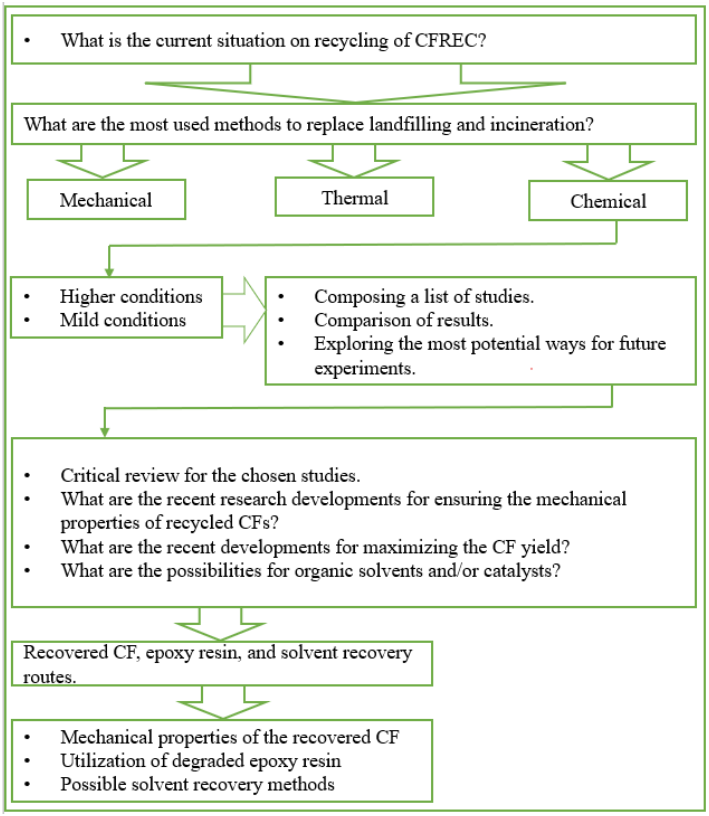


FIGURE 1: The flowsheet of the structure of the thesis.

3 RECYCLING METHODS OF FIBRE-REINFORCED POLYMER COMPOSITES

In this chapter, the most common methods for fibre-reinforced thermoset composite recycling are briefly covered. The information about the basic principles of each method is provided to give the reader an understanding of the variety of techniques and conditions in each method as well as the final condition of the products, polymer resin and fibres, including both CFRPC and GFRPC recycling. With the information provided by this chapter, the reader can better comprehend the methods and conditions of CFRPC recycling, which is the focus of this thesis, and discussed in chapters four and five. Mechanical, thermal, chemical, and other methods utilized in the recycling of CFRPC are briefly covered in this chapter.

3.1 Recycling methods for fibre-reinforced thermoset composites

Recycling of CFRPC waste should have minimal impact on the properties of the fibre and the thermoset resin. It should also be cost-effective and environmentally friendly, which means that there should be minimal possible consumption of energy while ensuring the maximal recyclability of the fibre and resin. The use of organic solvents and catalysts would also be preferable for sustainability. From the three main types of CF waste, this literature review is focused on the fibres that can be recycled from CF-reinforced composites, the other two wastes being prepregs and offcuts from CF production. (Pakdel, Kashi, Varley & Wang 2021; Khalid, Arif, Ahmed & Arshad 2022; La Rosa, Greco, Tosto & Cicala 2021.)

The recycling methods are divided into three main categories which further include different variations with different conditions, machinery and chemicals used. The methods are mechanical, thermal, and chemical recycling, which can be used separately or combined, for example, thermo-chemical recycling methods. Mechanical recycling includes different stages of breaking down the recycled material based on its size and different classification stages based on its composition. Thermal recycling consists of a fluidised bed process and pyrolysis where pyrolysis includes different variations based on the atmosphere and machinery. Chemical recycling is divided into two main methods based on the process conditions. The dividing criteria are considered to be the temperature, where below 200 °C is described as mild reaction conditions, and over that temperature is described as higher reaction condi-

tions including the use of sub- and supercritical fluids in the solvolysis process. (Karuppannan Gopalraj & Kärki 2020; Wu Klingler et al. 2023; Ribeiro, Fiúza, Ferreira, Dinis, Meira Castro, Meixedo & Alvim 2016, 178—193; Krauklis, Karl, Gagani & Jørgensen 2021; Danish et al. 2022.)

3.2 Mechanical recycling of fibre-reinforced thermoset composites

Mechanical recycling is the most commercially used method in the recycling of polymer composite materials. The first step in mechanical recycling is to reduce the size of the waste composites. It is done by shredding or primary cutting before it can be further processed. After cutting the recyclable material into smaller pieces, it can be ground into fine powder. The material also goes through stages like milling, screening, and straining to help get rid of raw materials from composites. (Utekar, V K, More & Rao 2021; Gharde & Kandasubramanian 2019.) The studies show that the use of mechanical recycling is more implemented for GF-reinforced composites rather than CF-reinforced composites. The GFs get sorted by size and reused as additives in composite fabrication as well as in cement to replace the use of fossil fuel-based products. The suggested amount of virgin GF to be replaced with recycled GF in composite fabrication is found to be from 5% to 30% so that the mechanical properties of the product would not be endangered while decreasing the product's overall consumption of energy per kilogram. The energy consumption in mechanical recycling of GF- and CF-reinforced composites is lower than in thermal or chemical recycling, and lower than manufacturing of virgin fibres. Nevertheless, the product from mechanical recycling has a low market value due to its poor mechanical properties after being processed to a small size using techniques that damage the fibre. (Khalid et al. 2022; Rani, Choudhary, Krishnan & Zafar 2021; Shuaib & Mativenga 2016, 198—206; Gharde & Kandasubramanian 2019.)

3.3 Thermal recycling of fibre-reinforced thermoset composites

The most used methods in thermal recycling are pyrolysis and fluidized bed methods. In the pyrolysis method, the composite waste is heated to approximately 300—600 °C in an inert atmosphere. The degradation of the matrix in the composite happens due to the heating, and the polymers break down. After the degradation of the polymers into smaller molecules, they can be further used to generate heat by burning, or for further chemical processing as feedstock. For cleaning the fibres, oxygen can be added to help burn off the resin residue. Pyrolysis is widely used in industry as a batch or continuous process.

(May, Goergen & Friedrich 2021, 70—81; Naqvi, Prabhakara, Bramer, Dierkes, Akkerman & Brem 2018, 118—129.)

In the fluidized bed method, the waste is first cut down into small pieces, the size approximately being around 25 mm. After reducing the size, it is filled in a reactor containing a bed of quartz sand. The fibres are released from the material by volatilizing the polymer with an airflow through the reactor and the sand bed and increasing the temperatures to approximately 450—550 °C. Studies show that the airflow is often generated with a speed of approximately 0.4—1.0 m/s. The fibres get further separated by size from the flowing air, and sorted, via a rotating sieve system and a cyclone. Energy used in the process can be partially recovered by further combusting the remaining organic components, such as the resin, at temperatures of 1000 °C in another chamber. The fibres recovered from the fluidized bed process can be used in different composite applications, for example in injection moulding methods. This method is mainly used in laboratory-sized works, and its downside is the low yield of polymers recovered. (May et al. 2021, 70—81; Pakdel et al. 2021; Rani et al. 2021; Danish et al. 2022.)

3.4 Chemical recycling of fibre-reinforced thermoset composites

In chemical recycling the main goal is to depolymerize the fibre-reinforced thermoset composites and recover the fibres as whole as possible while maximizing the yield of the polymers. Chemical recycling is a good technique for keeping the fibres' mechanical properties intact, although some studies suggest that the composites need to be granulated for batch reactors, therefore shortening the fibres. Still, some studies show that the composites can be recycled in larger pieces using a flow-type reactor. The use of different conditions, solvents, and catalysts are present in chemical recycling. (Khalid et al. 2022.)

The depolymerization is done with different solvents and catalysts, based on the type of polymer in the composite. The process of chemical recycling has been roughly divided into two, based on the affecting factor, temperature. In literature, the process is divided into low-temperature and pressure solvolysis, and high-temperature and pressure solvolysis. The dividing temperature has been around 200 °C. The use of solvents has been another dividing factor based on the use of acids, alcohol, or water with catalysts in mild or higher reaction conditions. (Karuppannan Gopalraj & Kärki 2020; Tian, Wang & Hou 2022, 1021—1041.)

3.5 Other methods for fibre-reinforced thermoset composite recycling

Besides the most used methods mentioned above, there are other methods used in the chemical recycling of fibre-reinforced thermoset composites. In a study found about a microwave-enhanced depolymerization of the composite waste, the process time was short, only 2 min. The GF yield was 51% at 340 °C with the use of an ionic liquid, N-Methyl-N-propylpiperidinium bis(trifluoromethylsulfonyl)imide. (Karuppannan Gopalraj & Kärki 2020.) Studies show that the use of microwave-assisted chemical recycling supports the use of organic synthesis. Another experiment shows that the use of approximately 500 °C microwave irradiation in energy-efficient recycling of CFs from CFRPC shortens the fibre compared to virgin fibres. The recovered fibres can still be used in traditional composite processing methods. (Rani et al. 2021.)

The electrolysis process has also been tested as a recycling method, obtaining good mechanical properties for recycled CFs. The CFRPC waste was used as an anode and the cathode was a stainless-steel plate. The electrolyte was a 3 % NaCl solution. The process took 21 days, so it is suggested that it is more useful on a bigger scale due to the long process time. The energy consumption of this electrolysis process is small, only 2—10 kWh/kg, compared to manufacturing virgin CF, which is 55—165 kWh/kg. The voltage used in the process was 2.6 V. (Karuppannan Gopalraj & Kärki 2020.)

Ultrasonic methods have also been studied. Two different studies show that under ultrasonics CFRPC could be effectively recycled with a 95 % decomposition rate of the resin while maintaining 95 % tensile strength for the CF, compared to virgin CF. The solvent used in both was nitric acid, and the other approach was to use H_2O_2 as a catalyst, while the other used potassium hydroxide (KOH) as a catalyst. The temperatures used for the studies with H_2O_2, and KOH were 60 °C and 160 °C, respectively. (Karuppannan Gopalraj & Kärki 2020.)

Another electrochemical recycling method called high voltage fragmentation (HVF) has been studied. In the HVF method, the waste material gets disintegrated due to the electrical pulses. The electrical pulses are repeated between two electrodes in a short period. The shockwaves that are generated in the process by the electrical discharge are at high temperatures and pressure, which leads to the disintegration of the waste. The approach results in clean and long fibres with low resin content retained in them. (Utekar et al. 2021.)

From these methods, chemical recycling was chosen to be further studied because of its possibilities of ensuring better mechanical properties for the recovered CFs due to its more subtle way of degrading the epoxy resin from the CFREC, compared to mechanical or thermal recycling methods for instance. Also, the possibility of recovering the epoxy with the used solvent gives an advantage for chemical recycling, because it enables to separately reuse the recovered CFs and epoxy resin.

4 RECYCLING METHODS WITH SUB- AND SUPERCRITICAL FLUIDS

In this chapter, recent studies on the chemical recycling of CFREC using solvolysis and hydrolysis using sub- and supercritical fluids as solvents are collected and reviewed. The use of different catalysts and additives is covered case-specifically. The use of sub- and supercritical alcohols, acetic acid and water are reviewed in these collected studies from the last decade, to give the reader a glimpse of possibilities for recycling of CFREC with higher reaction conditions. Case-specific process parameters and used materials are tabulated for quicker and clearer access to specific information. For visual clarification on recycling results based on the condition of recovered fibres, SEM images are added in each reviewed study. Temperatures over 200 °C and pressure varying from atmospheric to over 20 MPa are used in the following reviewed studies, which makes them higher reaction condition recycling processes (Oliveux et al. 2015, 61—99; Karuppannan Gopalraj & Kärki 2020).

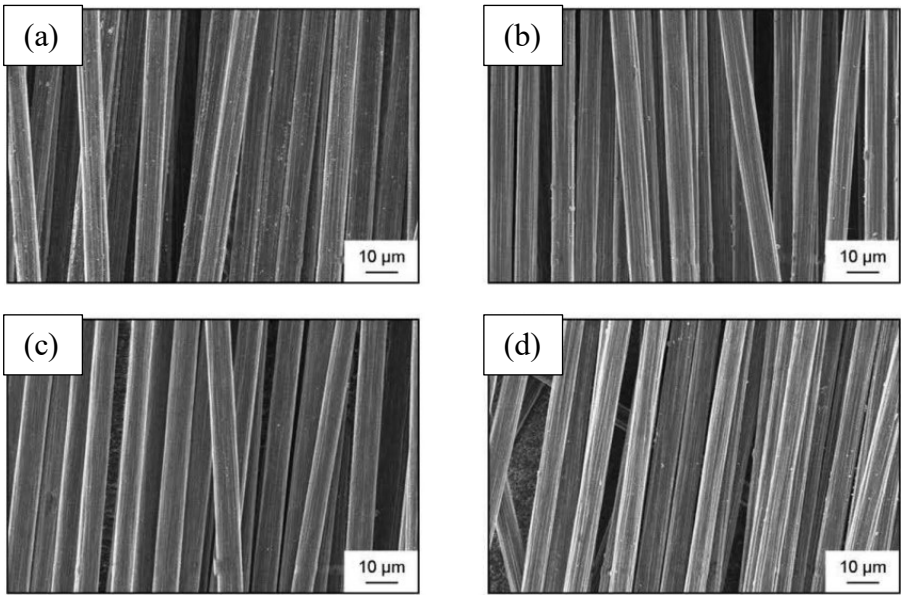
4.1 Supercritical 1-propanol and potassium hydroxide

Yan, Lu, Jing, Chang, Liu and Hou (2016, 46—54) conducted a study to find a way of chemically recycling CFREC using supercritical 1-propanol as a solvent and KOH as a catalyst in different temperatures with different reaction times in a stirred batch reactor. The goal of the study was to find the right amount of catalyst to use, the correct temperature, and the optimal reaction time to degrade the epoxy resin with a good yield while ensuring the best possible mechanical properties of the CFs. The characterization of the recycled CFs was conducted using SEM, thermogravimetric analysis (TGA), X-ray photoelectron spectroscopy (XPS), contact angle measurements and single fibre tensile strength tests, of which SEM analysis and tensile strength will be discussed in this review. The SEM images were used to analyze the visual condition of the CFs to see if they contained any residual waste after degradation of the resin or if they sustained surface damage from recycling in different conditions. The tensile testing was used to compare the tensile strength to the supplied CFs, to find the most suitable recycling parameters while ensuring the best mechanical properties. (Yan et al. 2016, 46—54.) The different parameters and material information of the study are tabulated in TABLE 1.

TABLE 1:Process parameters and material information for recycling with supercritical 1-propanol (adapted from Yan et al. 2016, 46—54).

Materials:	Epoxy resin: tetraglycidyl 4,4′-diaminodiphenylmethane, (TGDDM) Curing agent: 4,4′-diaminodiphenylsulfone, (DDS) Carbon fibres: 7 μm, tensile strength 3.53 GPa (standard: GB/T 26749) Composite material: epoxy resin ~32wt.%, 8 layers of CF fabric, heated at 130 °C for 1 hour, cured at 180 °C for 4 hours. Sample size: 50 mm · 10 mm · 2 mm. Reaction medium: 1-propanol Catalyst: KOH
Temperatures in °C:	Testing: 260, 280, 300, 310, 320, 330, 340 Optimized: 320
Reaction time in min:	Testing: 30, 60, 90, 120 Optimized: 60
Concentration of KOH in wt.%:	Testing: 0.5; 1.0; 2.0 Optimized: 1.0

The recycling study began with the experimentation of different temperatures in supercritical 1-propanol without the addition of KOH. The weight loss was measured at different temperatures, and it was noticed that the epoxy resin was swelling from 1-propanol at 260 °C and 280 °C. At 300 °C the weight loss was 3.4 wt.% which indicates a small decomposition reaction. The weight loss was 8.2 wt.% at 310 °C, 12.8 wt.% at 320 °C, 28.4 wt.% at 330 °C and 30.6 wt.% at 340 °C. The SEM images show good visual condition and clean surface on CFs at 330 °C and above. The tensile testing shows that recycling at 310 °C the tensile strength was the same as the original fibre. At 320 °C and 330 °C the tensile strength retained 95.2 % and 94.6 %, respectively. At 340 °C the tensile strength was only 88.6 % of the original. The study was continued at 320 °C comparing different reaction times with 1 wt.% KOH as a catalyst. It was noticed that by keeping the reaction time below 90 min the CFs retained 90 % of their original tensile strength. The increase of KOH concentration decreased the mechanical properties of the CFs, and it was noticed that recycling at 320 °C for 1 hour with 1 wt.% concentration of KOH the tensile strength was the highest. Also, with these conditions clean fibres were obtained as can be seen in PICTURE 1. (Yan et al. 2016, 46—54.)



PICTURE 1: SEM images of the recycled carbon fibres after the treatment at 320 °C for: (a) 30, (b) 60, (c) 90 and (d) 120 min with 1 wt.% KOH as catalyst (Adapted with permission from Yan et al. 2016, 46—54. Elsevier).

In the study, it was concluded that by increasing the temperature, reaction time, and concentration of KOH, the degradation of the epoxy resin increased, but the mechanical properties of the CFs decreased when passing a certain point. The process parameters were optimized to maximize the strength of the recycled CFs while having the best possible decomposition rate of the resin. The optimal temperature was 320 °C, the optimal reaction time was 60 min, and the optimal concentration of KOH was 1 wt.%. (Yan et al. 2016, 46—54.)

4.2 Acetone in sub- and supercritical conditions

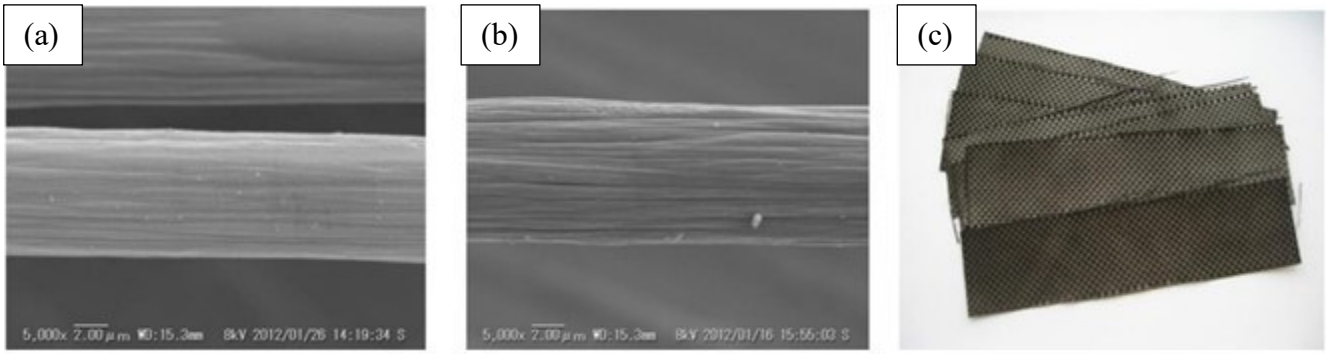
Okajima, Watanabe, Haramiishi, Nakamura, Shimamura and Sako (2017, 44—51) conducted a study in which they compared different solutions for chemical recycling of CF-reinforced plastics including an amine-cured thermosetting epoxy resin. They tested the degradation of the epoxy resin with different alcohols and ketones in a batch reactor and a semi-flow-type bench plant they conducted a study to recover the CFs from composite material using acetone. The testing conducted in the batch reactor for degrading the epoxy resin consisted of different solvents, temperatures, pressures, and reaction times. The goal was to find the most suitable solvent with the capability to degrade the resin in a reasonable time. The more specific parameters of the process and information about the materials used are listed in TABLE 2. (Okajima et al. 2017, 44—51.)

TABLE 2: Process parameters and material information for recycling process with acetone and other solvents (adapted from Okajima et al. 2017, 44—51).

Materials:	Epoxy resin: Glycidyl end-capped poly (bisphenol A-co-epichlorohydrin) Curing agent: Tetraethylenepentamine Carbon fibres: Torayca cloth CO6343 (T300–3000 CFs) Composite material: Epoxy resin, 8 CF cloths, cured at 80 °C for 60 min. CF content ~52 wt.%. Sample size: 200 mm · 45 mm · 3 mm Reaction medium: Methanol, ethanol, 1-propanol, 1-butanol, 2-butanol, tert-butanol, methyl ethyl ketone (>99.8 % purity), and acetone (>99.5 % purity)
Temperatures in °C:	For testing with acetone: 300, 320, 350 For testing with others: 320 Optimized (acetone): 320
Pressure in MPa:	For testing with acetone: 0.1; 0.5; 1; 2; 4; 6; 11.9; 10; 8.7 For testing with others: Methanol 12.2; ethanol 11.2; 1-propanol 9; 1-butanol 6.6; 2-butanol 8.3; tert-butanol 10.8; methyl ethyl ketone 8 Optimized (acetone): 1 MPa
Reaction time in min:	Testing: 0—90 Optimized (acetone): 20
Concentration of solvents in mol/L:	Acetone had different concentrations with different parameters, but the best outcome was 3.64. For other solvents, the same concentration of 3.64 was used.

The study was conducted at 320 °C with a concentration of 3.64 mol/L for all the solvents. In the case of acetone, those parameters were selected as the most competent. Supercritical methanol reached constant decomposition efficiency at 88 % after 20 min, and supercritical ethanol reached 95 % decomposition efficiency after 25 min. Both supercritical 1-propanol and 1-butanol reached a 100 % decomposition efficiency after 25 min. Other supercritical isomers of butanol, 2-butanol and tert-butanol reached decomposition efficiency at 95 % and 88 %, respectively, after 20 min. Supercritical methyl ethyl ketone reached decomposition efficiency at 100 % after 25 min whereas supercritical acetone reached the same efficiency after 20 min. For further studies of temperature and pressure acetone was

selected due to its ability to fully decompose the resin the fastest. The decomposition of the resin was noticed to be 100 % using subcritical acetone as solvent at 320 °C and 1 MPa after 20 min. These parameters for temperature, pressure, and time were studied for recovering the CFs from the composite material in a semi-flow-type bench plant. The tensile strength of the fibres was tested, and they were almost the same as virgin CFs. Also, after investigating the SEM images, it was noticed that the CF sheets retained their form and were clean after the process as can be seen in PICTURE 2. (Okajima et al. 2017, 44—51.)



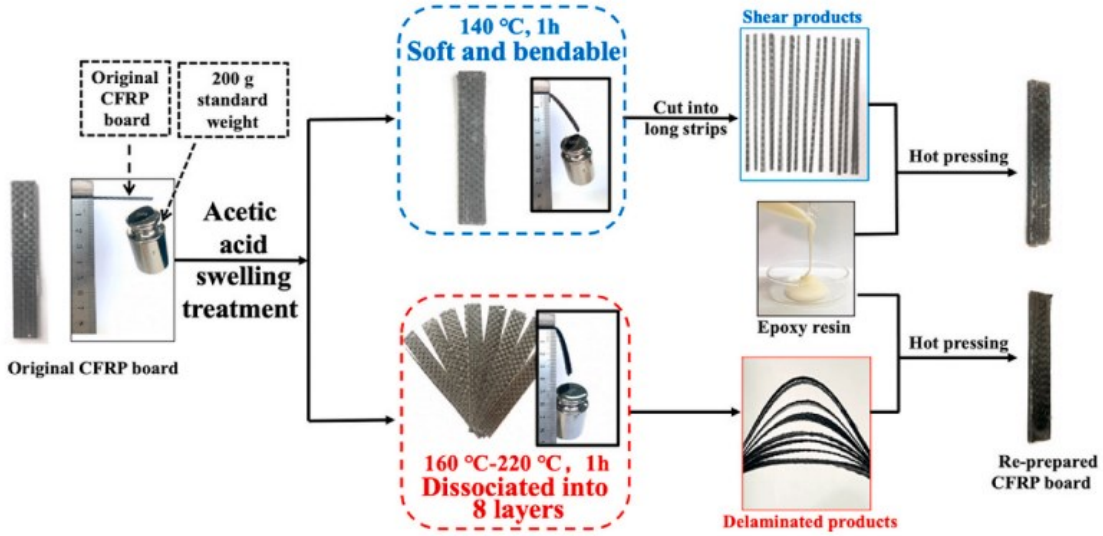
PICTURE 2: SEM images of: (a) virgin carbon fibre, (b) recovered carbon fibre, and an image of (c) recovered whole carbon fibre after decomposition (Adapted with permission from Okajima et al. 2017, 44—51. Elsevier).

In addition, Oliveux, Dandy and Leeke (2015, 96—103) studied the use of a mixture made of water and acetone for solvolysis purposes and the study shows that using 80 vol.% acetone at 350 °C and 23.5 MPa with the resin concentration of 30 mg/mL and a reaction time of 30 min gives a degradation rate of 93 %. The reaction was conducted in a batch reactor. The study itself focused mainly on degrading the epoxy resin, the condition of recovered CFs was not discussed. (Oliveux et al. 2015, 96—103.) Using the same mixture of acetone and water, 80 vol.% and 20 vol.%, respectively, Keith, Román-Ramírez, Leeke and Ingram (2019, 225—234) found that the CFs can be effectively recovered, and the epoxy resin can be fully decomposed using a temperature of 320 °C, a reaction time of 120 min at 19.7 MPa.

4.3 Subcritical acetic acid as solvent

Acetic acid's properties as a solvent have been studied. In the experiments found, an initial swelling stage before the decomposition stage for the polymer resin in the composite has been suggested as well

as a swelling method (Xing, Li, Zheng, Du, Chen & Wang 2021; Ma & Nutt 2018, 307—317.). Xing et al. (2021) proposed a novel method for recycling CFREC by swelling with acetic acid. They studied the swelling and degradation of the polymer and the flexural strength of the re-prepared CFREC boards. In the study, they used a high-pressure reactor at 100—220 °C with a pressure range from 0.07 MPa to 1.03 MPa. The swelling-treated composites were separated into two categories, the ones softened at 140 °C and cut into long strip shear products that were hot pressed into a recycled CFREC board, and the ones dissociated at 160—220 °C and delaminated products that were re-prepared to a CFREC board as is illustrated in PICTURE 3. (Xing et al. 2021.)



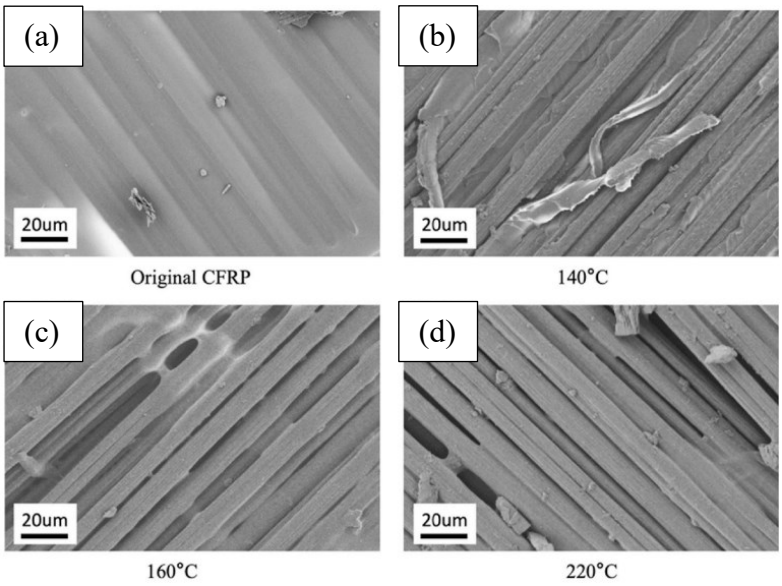
PICTURE 3: Illustration of the acetic acid swelling process and division into two categories of recycled carbon fibres (Adapted with permission from Xing et al. 2021. Elsevier).

In the swelling process, the original CFREC board softened at 140 °C and started to delaminate at 160—220 °C within 60 min. The softened and delaminated products were utilized differently as can be seen in PICTURE 3. The softened products were cut into strips with the size of 80 mm · 2 mm · 2 mm and dried. After drying, ~0.5 g of epoxy resin was added, and the strips were hot pressed into a new CFREC board at a curing temperature of 140 °C for 3 hours and a molding pressure of 0.1 MPa. The size of the new CFREC board was 80 mm · 10 mm · 2 mm. The delaminated products were dried and mixed with ~1.5 g epoxy resin after which they were hot pressed. The parameters for hot pressing were the same as for softened strips as well as the size of the re-prepared CFREC board. The material information and process parameters are compiled in TABLE 3. (Xing et al. 2021.)

TABLE 3: Process parameters and material information for acetic acid swelling method (adapted from Xing et al. 2021).

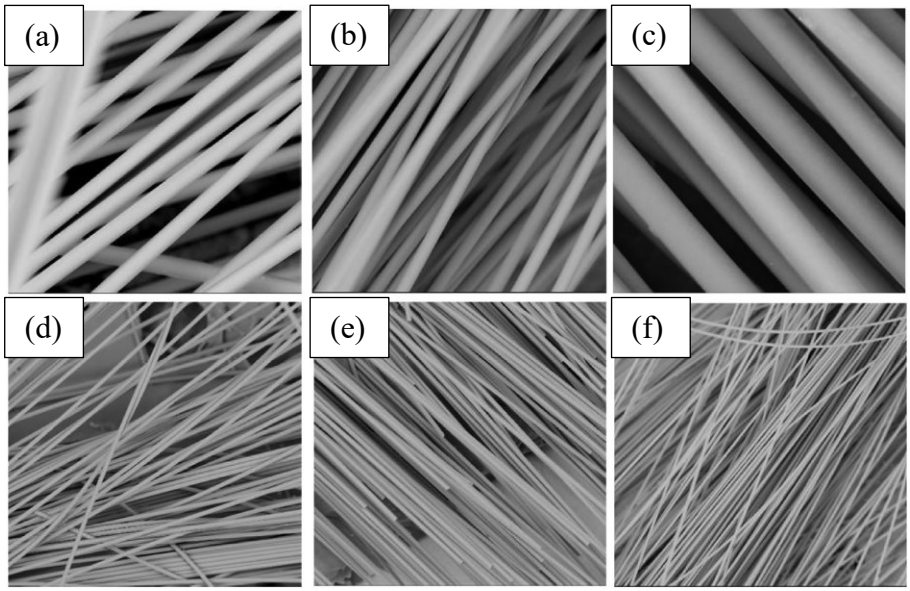
Materials:	Epoxy resin: Bisphenol A type epoxy resin (6508 epoxy resin) Curing agent: Dicyandiamide Carbon fibres: woven cloth (weave angle: 0/90°) Composite material: CFREC prepreg (WP-3011, 0.2 mm thick) 60 wt.% CFs, 40 wt.% epoxy resin, size 80 mm · 10 mm · 0.2 mm. CFREC board: Eight layers of prepregs were used for CFREC board (~2 g, 80 mm · 10 mm · 2 mm.). Hot pressing: cured at 140 °C for 3 hours, molding pressure ~0.1 MPa. Reaction medium: acetic acid
Temperatures in °C:	Testing: 100—220 Optimized: 220
Pressure in MPa:	Corresponding pressure ranged with temperature in autoclave: 0.07—1.03 Optimized: 1.03
Reaction time in hours:	1 to achieve the temperature, 0.5—4 for maintaining the temperature. Optimized: 1

The epoxy resin's purpose was to fill the cracks of the hot-pressed products ensuring the strength of the CFREC board. As can be seen from SEM images in PICTURE 4, the swelling with acetic acid degraded the epoxy resin from the composite samples when the temperature increased. At 220 °C the fibres seem cleaner from epoxy resin which explains the delamination. The re-prepared CFREC board from swelling products obtained at 140 °C did not retain its flexural strength well, only 47 % of that of the original board. The re-prepared CFREC board from swelling products obtained at 220 °C retained its flexural strength well, 89 % of that of the original board. It was concluded that the more rapid degradation of epoxy resin at higher temperatures allowed the added epoxy resin to permeate better in the delaminated products which increased their flexural strength. (Xing et al. 2021.)



PICTURE 4: SEM images of the original CFRP and the swelling products obtained at different temperatures: (a) original CFRP, (b) 140 °C, (c) 160 °C, and (d) 220 °C (Adapted with permission from Xing et al. 2021. Elsevier).

Shetty, Pinkard and Novosselov (2022) conducted a study to recycle CFREC in an acetic acid solution which included 50 wt.% acetic acid and 50 wt.% de-ionized water. The composite material used was unknown, but the resin content was approximately 26—32 %, this was found out using TGA. The reaction was conducted in a batch reactor at 200—300 °C for 0—120 min. (Shetty et al. 2022.) As well as in the study conducted by Xing et al. (2021) about the swelling of the product, Shetty et al. (2022) also studied the swelling, but as an initial stage of the process which was continued with the depolymerization part. The resin removal rate showed swelling of the sample at 200—225 °C, at 250 °C for a residence time of 0—90 min and at 275 °C for a residence time of under 30 min. The heat-up time was approximately 80 min. In this study it was hypothesized that the swelling of samples in acetic acid at lower temperatures could improve the depolymerization in the thermal part of the process, the study of Xing et al. (2021) supports this hypothesis. The optimal parameters were at 300 °C for 30 min, and the autogenic pressure rise was measured to rise at approximately 10 MPa. A limitation of the study with respect to the scope of this thesis is that tensile properties of the recovered fibres were not studied, although the SEM images show extremely clean fibres with an almost total degradation of the polymer resin as seen in PICTURE 5, and it was concluded that based on literature the composition of the fibres is the same as that of the virgin fibres. It was also proven via an EDS analysis that the fibres' surface only included carbon. (Shetty et al. 2022.)



PICTURE 5: SEM images of recovered carbon fibres after exposure at 300 °C for 30 min. (a), (b), and (c) show strands of fibres with 100% resin removal (magnification: 2500x) while (d), (e), and (f) show multiple layers of clean fibres (magnification: 590x) (Adapted with permission from Shetty et al. 2022. Elsevier).

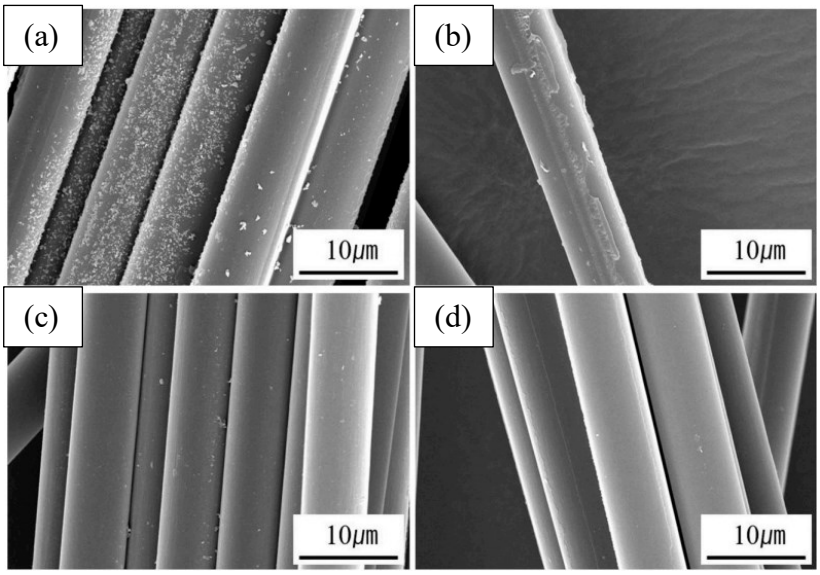
4.4 Recycling using supercritical water

A study was conducted by Kim, Kim, Kim, Park, Yang, Kim, and Jung (2019, 66—72) in which they experimented with a way to recycle CFREC using supercritical water. In the experiment, they removed up to 99.5 % of the epoxy resin from the CFs and fabricated the recycled CFs into composite material by combining a polymerizable low-viscosity thermoplastic resin, cyclic butylene terephthalate (CBT), with them. The recycled CFs showed good results in strength and visual appearance after 30 min of processing and even better after 120 min of processing. The amount of recycled CFs mixed with CBT was 5 wt.%. Both components were in powder form and the melt pressure conditions for the 5 wt.% recycled CF- CBT composite was at 250 °C with 15 MPa of pressure for 2 min. Other process parameters and material information are tabulated in TABLE 4. (Kim et al. 2019, 66—72.)

TABLE 4: Process parameters and material information for the experiment of recycling with supercritical water (adapted from Kim et al. 2019, 66—72).

Materials:	Epoxy resin: KBR-1729 (Cycloaliphatic Glycidyl ether modified type) Curing agent: KBH-1088 (Acid anhydride hardener type) Carbon fibres: TORAYCA® T700S Composite material: Manufactured prepregs were laminated 50 pieces, sample size 1 cm · 1 cm · 1 cm. The wt.% distribution of polymer and CF was not informed. Reaction medium: Water
Temperatures in °C:	Optimized: 405 ± 2
Pressure in MPa:	Optimized: 28 ± 1
Reaction time in min:	Testing: 10, 30, 60, 120 Optimized: 60

The experiment took place in a batch reactor with temperature, pressure and reaction times mentioned in TABLE 4. With reaction times of 10—30 min, the SEM images show remaining resin particles on the surface of the fibres, as can be seen in PICTURE 6. After increasing the reaction time gradually to 60 min and eventually to 120 min, the fibres seem clean with good form with a diameter of 7 ± 1 μm. The tensile strength of the recycled CFs showed a reduction of 18—36 % compared to the pristine fibres. The tensile strength of the pristine fibres was approximately 4.5 GPa with a strain of 1.75%, and the reduced tensile strength of the recycled CFs was 2.9—3.7 GPa. with a strain of 1.6 %. The fabricated composite from the recycled fibres mixed with CBT owed thermal (1.35 ± 0.05 W/mK) and electrical ($1.23 \cdot 10^{-6}$ S/cm) properties after a simple hot-pressing method.



PICTURE 6: SEM images of recycled carbon fibres after supercritical water treatment: (a) 10 min, (b) 30 min, (c) 60 min, and (d) 120 min treatment, respectively (Adapted with permission from Kim et al. 2019, 66—72. Elsevier).

4.5 Supercritical methanol as solvent

A study that was conducted by Okajima, Hiramatsu, Shimamura, Awaya and Sako (2014, 68—76) experiments with the use of supercritical methanol for chemical recycling of CFREC. The recycling process was studied using a batch reactor and a semi-flow-type reactor. The goal of the study was to find the optimal temperature, reaction time and pressure for the best yield of polymer while ensuring the best possible mechanical properties for the CFs. In the study, the recovered epoxy resin was recycled, and the recovered CFs were also recycled as a CFREC using virgin epoxy, and their properties were tested. More information about the parameters and materials used in the study is listed in TABLE 5. (Okajima et al. 2014, 68—76.)

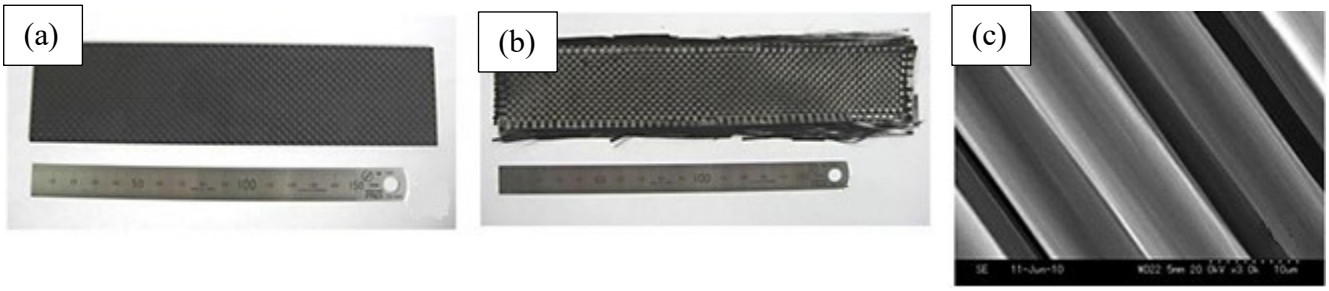
TABLE 5: Process parameters and materials used in the recycling process with supercritical methanol (adapted from Okajima et al. 2014, 68—76).

Materials:	Epoxy resin: Poly (bisphenol A-co-epichlorohydrin) glycidyl end-capped Curing agent: 1,2-Cyclohexane dicarboxylic anhydride Promoter for curing reaction: Triethylamine Carbon cloths: Toray T300-3000 Composite material: 8 CF cloths, CF content 49.2 wt.%. Sample size for batch reactor: 60 mm · 3 mm · 3 mm Sample size for semi-flow-type reactor: 200 mm · 45 mm · 3 mm Reaction medium: methanol (>99.8% purity)
Temperatures in °C:	Testing: 250, 270, 280, 285, 300, 320, 350 Optimized: for the batch reactor at 270 and for the semi-flow-type reactor at 285
Pressure in MPa:	Testing: 5, 8, 10 Optimized: 8 for both reactor types
Reaction time in min:	Testing: 0—120 Optimized: batch reactor for 90 and semi-flow-type reactor for 80

The recovered liquid, which included methanol and methanol-soluble resin from the semi-flow-type reactor, was vaporized, and the resin was recovered. The recovered epoxy resin was re-cured with different ratios of recycled resin mixed with virgin epoxy, and its mechanical properties were tested by a three-point bending test. With virgin epoxy only, the test gave a strength result of 95 MPa and it decreased almost linearly to 35 MPa with 75% of recovered resin. With 50% use of recovered resin, the strength was 55 MPa. A tensile strength test was used for the recovered CFs. The length of the tested CF was 25 mm, and the tensile strength was 3100 MPa and 3000 MPa from the batch reactor and the semi-flow-type reactor, respectively. The tensile strength of the virgin CF was 3400 MPa. There were also visual differences observed with the quality of the fibres based on the parameters and reactors used. PICTURE 7 shows the visual quality of the fibre from the batch reactor. (Okajima et al. 2014, 68—76.)

PICTURE 7: Appearance of recovered carbon fibre at 270 °C, 8 MPa and 90 min: (a) CFRP before decomposition, (b) recovered carbon fibre after decomposition, and (c) SEM photo of recovered carbon fibre (magnification: 3000x), from the batch reactor (Adapted with permission from Okajima et al. 2014, 68—76. Elsevier).

PICTURE 8 shows the visual quality of the fibre from the semi-flow-type reactor. The temperature and reaction time differed with both reactors, but the pressure remained at 8 MPa. The parameters given in PICTURES 7 and 8 are the optimized parameters for both rector types. (Okajima et al. 2014, 68—76.)



PICTURE 8: Appearance of recovered carbon fibre at 285 °C, 8 MPa and 80 min of elapsed time using semi-flow-type reactor: (a) CFRP before decomposition, (b) recovered carbon fibre after decomposition, and (c) SEM photo of recovered carbon fibre (magnification: 3000x) (Adapted with permission from Okajima et al. 2014, 68—76. Elsevier).

5 RECYCLING METHODS WITH MILD REACTION CONDITIONS

In this chapter, recent studies of CFREC recycling are covered, focusing on mild reaction chemical recycling using ionic liquids, deep eutectic solvents (DES), alcohols, amines, and acid digestion. The use of swelling before decomposition was often used in these mild reaction conditions. The collected studies from the last decade are reviewed to give an idea of various solvents and catalysts used in mild reaction chemical recycling. The case-specific parameters and used materials are tabulated in the same fashion as in chapter four to help access specific information clearly and quickly. SEM images are also used in this chapter to clarify the visual conditions of recovered fibres. Temperatures below 200 °C and atmospheric pressure are used which makes these studies mild reaction chemical recycling processes (Oliveux et al. 2015, 61—99; Karuppannan Gopalraj & Kärki 2020).

5.1 Ionic liquids and ethylene glycol as solvent

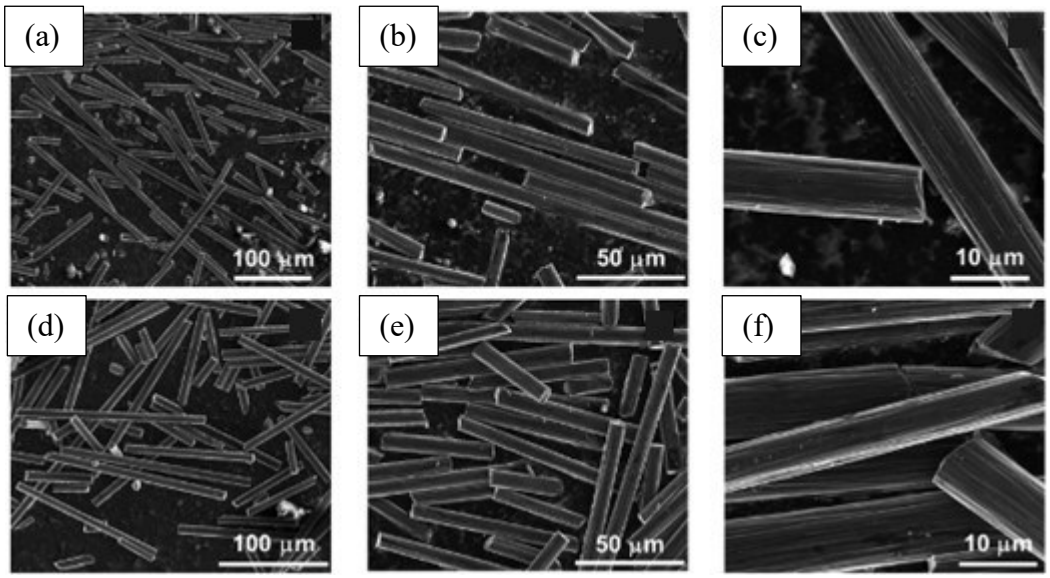
A study was conducted by Pérez, Ayala, Opiri, Ezzir, Li and Warner (2021, 5588—5595) where they experimented with the use of alkyl-methyl-imidazolium ionic liquids and alcohols as solvents with different ratios to swell and dissolute thermoset epoxy resin (TER) and CFREC without the aid of catalytic components, in mild reaction conditions. The ionic liquids used in the study had three different alkyl lengths, they were 1-butyl-3-methyl imidazolium chloride ([BMIm][Cl]), 1-hexyl-3-methyl imidazolium bromide ([HMIm][Br]), and 1-octyl-3-methyl imidazolium bromide ([OMIm][Br]), and the alcohols used were ethylene glycol (EG) and glycerol (Gly). The swelling experiment showed the best results at 150 °C using [BMIm][Cl] and EG. Also, different anion variants of [BMIm][Cl] were tested with EG. The anions were acetate ([$Acet^-$]), propionate ([$Prop^-$]), hexanoate ([Hex^-]), tetrafluoroborate ([$BF4^-$]), and hexafluorophosphate ([$PF6^-$]). More information about the materials and parameters used in this study is listed in TABLE 6. (Pérez et al. 2021, 5588—5595.)

TABLE 6: Process parameters and materials used in the recycling process with alkyl-methyl-imidazolium ionic liquids and alcohols (adapted from Pérez et al. 2021, 5588—5595).

Materials:	Epoxy resin: Bisphenol A Diglycidyl ether (DGEBA) Curing agent: Tricarballylic acid (TCA) Promoter for curing reaction: tetrahydrofuran (THF) Milled carbon fibres: PX-35 milled CFs (length = 150−200μm, diameter = 7.2μm) Composite material: Milled CF (5 wt.%) mixed with a 1:1 ratio of DGEBA and TCA. Sample size of composite: 3000 mg Sample size of epoxy: 500 mg rectangular piece Reaction mediums: [BMIm][Cl], [HMIm][Br], [OMIm][Br], EG and Gly. Optimized reaction medium: 20:80 ratio of [BMIm][Acet] and EG
Temperatures in °C:	Testing: 70, 100, 150 Optimized: 150
Pressure:	Atm
Reaction time in min:	Testing: 0—150 Optimized: 150

EG and Gly were tested with different ratios with each ionic liquid, and it was found that with the mixture of 20% [BMIm][Cl] and 80% EG the swelling of the cured epoxy was the highest, 44.6%. The mixture selected for swelling could not dissolve the TER, so different anions were selected to accompany the [BMIm]- based solvent with EG. The best results for dissolving the epoxy resin were found with the use of a 20:80 ratio of [BMIm][Acet]: EG, which dissolved the epoxy resin in 150 min at 150 °C. Water was used to precipitate the TER from the solution. A 3 g sample of CFREC, which included 5 wt.% of milled CFs, was added to the solution of 20:80 ratio of [BMIm][Acet]: EG, and the dissolution of the epoxy resin was tested at 150 °C for 150 min under atmospheric pressure. The recovered CF's visual condition was evaluated using SEM as can be seen in PICTURE 9 and it was discovered that the surface remained smooth and undamaged. The strength of the single fibre was not tested, but the strength of the recycled CFREC which included 5 wt.% of recovered CFs was compared to the original CFREC and it was discovered that Young's modulus decreased less than 4%, from 2384 +/- 81 MPa to 2299 +/- 42 MPa and the compressive strength decreased only 2.7%, from 75 +/- 6 MPa to

73 +/- 6 MPa. It was concluded that further studies are desired considering aspects such as the CF's direct mechanical condition testing. (Pérez et al. 2021, 5588—5595.) It was not stated if it was due to the length of the recycled milled fibre that it was not conducted in this study.



PICTURE 9: SEM images of (a, b, c) new carbon fibre and (d, e, f) recycled carbon fibre (Adapted with permission from Pérez et al. 2021, 5588—5595. Copyright 2021 American Chemical Society).

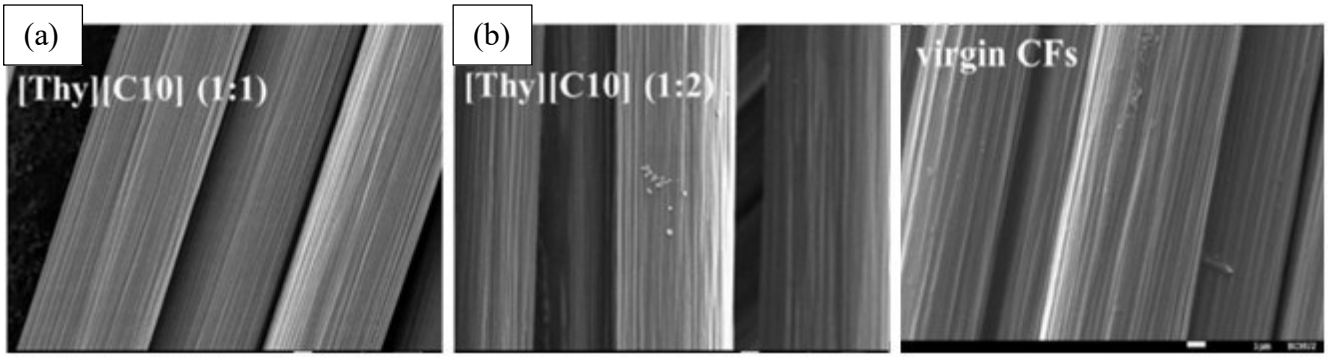
5.2 Recycling using deep eutectic solvents

A study conducted by Liu, Hong, Yang, Lin, Wang, and Chen (2022) studies a novel chemical recycling method of CFREC using DESs with switchable hydrophilicity mixed with metal salt catalysts using mild reaction conditions. The metal salt catalysts examined in this study were $ZnCl_2$, $FeCl_2$ and $FeCl_3$, and the DESs used were mixtures of thymol and different acids. The acids tested were caproic acid, octanoic acid, decanoic acid, and the best result was found with the mixture of thymol and decanoic acid ([Thy][C10]) with the ratio of 1:1. The optimal metal salt catalyst was found to be $ZnCl_2$ (3.3 wt.%). The optimized temperature for complete epoxy decomposition was at 180°C for 1.5 hours in atmospheric pressure. Further information about the materials and parameters used is listed in TABLE 7. (Liu et al. 2022.)

TABLE 7: Process parameters and materials used in the mild chemical recycling with [Thy][C10] and $ZnCl_2$ (adapted from Liu et al. 2022).

Materials:	Epoxy resin: Bisphenol A Diglycidyl ether (DGEBA) Curing agent: Isophorone diamine (IPDA) Carbon fibres: TC36P 12k CF fabric Composite material: CF fabric (40 wt.%) and above resin mixture Optimized reaction medium: thymol and decanoic acid [Thy][C10] 1:1 Catalysts: $ZnCl_2$, $FeCl_2$, $FeCl_3$ Optimized catalyst: $ZnCl_2$ (3.3 wt.%)
Temperatures in °C:	Testing: 160—180 Optimized: 180
Pressure:	Atm
Reaction time in min:	Testing: 0—120 Optimized: 90

First, the swelling of the epoxy resin was tested, following the swelling testing of CFREC, with different parameters and solutions, and it was discovered that the best swelling occurred with above mentioned optimized parameters, solution, and catalyst. The visual condition of the recycled CF was evaluated from different molar ratios of [Thy][C10] compared to the virgin fibre, and the SEM images show no visual damage and no resin residue in the fibres recycled using [Thy][C10] 1:1, as can be seen in PICTURE 10. (Liu et al. 2022.)



PICTURE 10: SEM images of recovered carbon fibres treated at 180 °C for 90 min in ZnCl2/[Thy][C10]- based DESs: (a) [Thy][C10] (1:1), (b) [Thy][C10] (1:2), and SEM image of virgin carbon fibre (Adapted with permission from Liu et al. 2022. Elsevier).

The tensile strength of the CF was tested and compared to the virgin CF. At least 20 fibres of each sample were tested, but their length was not reported. The tensile strength of the recovered fibres was

3.12 GPa. The reduction in strength compared to the virgin fibres was only 5.5 %, which can be considered a good result, and the fibres can be considered well-reusable. It was yet notable, that it was concluded that the studied method can be described as a green method, and the recycled epoxy resin was decomposed into bisphenol A monomers and derivatives. The used solvents and catalysts were regenerated after the process and it was noted that they kept their recycling properties almost entirely, by only increasing the reaction time for full decomposition of epoxy resin in further recycling cycles. (Liu et al. 2022.)

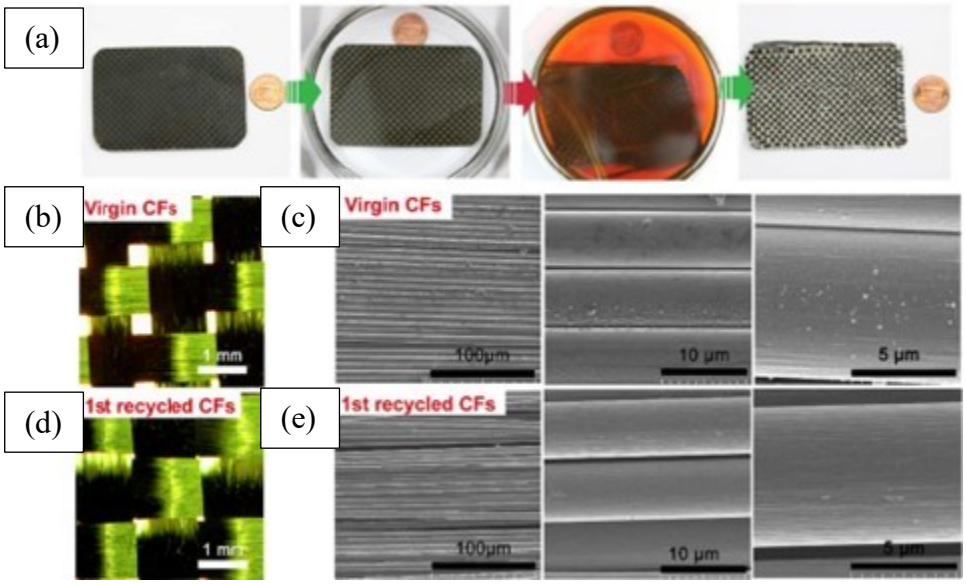
5.3 An organic solvent and catalyst with ethylene glycol

A study conducted by Kuang, Zhou, Shi, Wang, and Qi experimented with the use of n-methyl-2-pyrrolidone (NMP), EG and an organic catalyst 1,5,7-triazabicyclo [4,4,0] dec-5-ene (TBD) for chemical recycling of CFREC using mild reaction conditions, temperature being at 170 °C in atmospheric pressure for 1.5 hours. The ratio of EG/NMP for the recycling of composites was determined to be 10/90, v/v and the amount of the organic catalyst TBD was 0.3 mol/L. It was studied that the NMP solvent does the swelling of the epoxy while guiding the TBD-EG into the polymer and the epoxy could be recycled as oligomers. The mass loss of the epoxy is approximately 95 %. Material information and process parameters are conducted in TABLE 8. (Kuang et al. 2018, 9189—9197.)

TABLE 8: Process parameters and material information for chemical recycling with TBD-EG/NMP solution (adapted from Kuang et al. 2018, 9189—9197).

Materials:	Epoxy resin: EPON resin 828 epoxy oligomer and Hexahydro-4-methylphthalic anhydride (HP) Curing agent: Epikure 3253(2, 4, 6-tris-dimethylaminomethyl phenol) Carbon fibres: plain weave fabric Sample size of CFREC: 13.9 g, 5.5cm · 7.5cm · 3cm Optimized reaction medium: EG/NMP, 10/90, v/v Optimized catalyst: TBD 0.3 mol/L
Temperatures in °C:	Optimized temperature: 170
Pressure:	Atm
Reaction time in min:	Optimized time: 90

After recycling the composite samples in TBD-EG/NMP solution at 170 °C for 90 min, the tensile strength and Young's modulus of the CFs were evaluated and compared to the virgin CFs. The visual appearance was evaluated using optical microscopy and SEM images as can be seen in PICTURE 11. The number of tested fibres per batch was 10 and the length was 10 mm. The Young's modulus results showed no differences, the value being 280 GPa, and the tensile strength improved after recycling. The tensile strength of virgin fibre was 4.9 GPa and after recycling it was 5.5 GPa. (Kuang et al. 2018, 9189—9197.)



PICTURE 11: (a) A picture of CFRP recycling method in 150 mL of solution at 170 °C for 1.5 hours and collected clean fibre fabric. The visual comparison of virgin (b and c) and recycled (d and e) CF images from optical microscope (b and d) and SEM (c and e) (Adapted with permission from Kuang et al. 2018, 9189—9197. Copyright 2018 American Chemical Society).

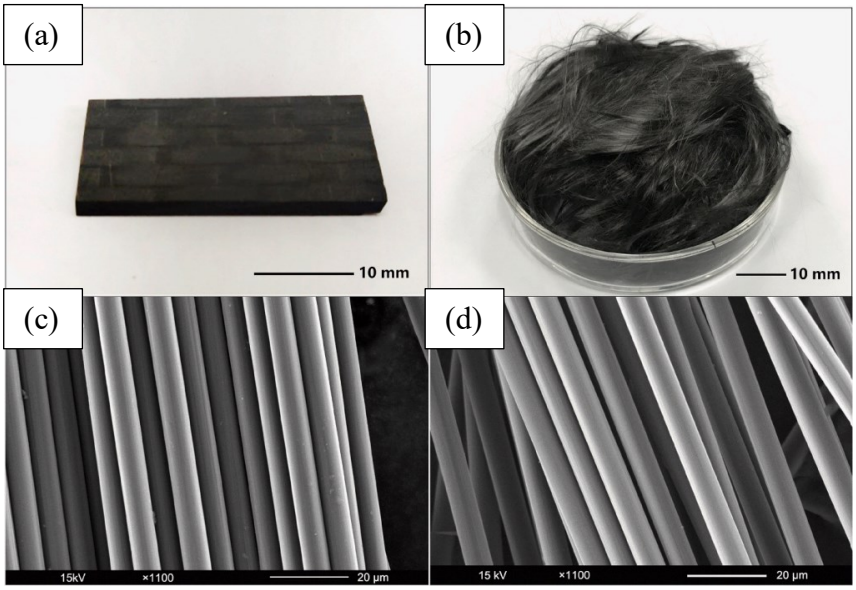
5.4 A monoethanolamine- potassium hydroxide solution

A study shows that with the use of monoethanolamine (MEA) combined with KOH at a temperature of 160 °C for 60 min under atmospheric pressure, CFREC can be decomposed with a ratio of 99 %. In this experiment, MEA is both the solvent and the catalyst while KOH functions as a co-catalyst. For 1 g of CFREC composite 9 mL of MEA and 0.2 g of 0.5 M KOH were used, and it is noted that this recycling method leaves no solid waste. Further process parameters and material information are listed in TABLE 9. (Zhao, An, Li, Zhang, Jiang & Li 2022.)

TABLE 9: Process parameters and material information of chemical recycling process using MEA and KOH under mild conditions (adapted from Zhao et al. 2022).

Materials:	Epoxy resin: Epoxy resin 328 oligomer (bisphenol A epoxy resin) and Methyl tetrahydrophthalic anhydride (MeTHPA) Hardener: 2-ethyl-4-methylimdazole (EMI-2,4) Carbon fibres: Toray T700 carbon cloth CFREC composite: 63 wt.% CF, 37 wt.% epoxy resin Sample size of CFREC: 30mm · 15mm · 3mm Optimized reaction medium: For 1g of CFREC recycling, 9 mL of MEA and 0.2 g of 0.5 M KOH were used. Testing concentrations of KOH: 0—0.7 M Optimized concentration of KOH: 0.5 M
Temperatures in °C:	Testing temperatures: 120—163 Optimized temperature: 160—163
Pressure:	Atm
Reaction time in min:	Testing reaction times: 0—60 Optimized time: 60

The temperatures were tested from 120—121 °C to 160—163 °C for 0 to 60 min and it was discovered that a near 100 % decomposition ratio was achieved at 160—163 °C for 60 min. The concentration was tested for 0 M to 0.7 M, and it was discovered that a nearly 100 % decomposition ratio was achieved with 0.5—0.7 M, so 0.5 M was used. The temperature and reaction time testing were conducted using a 0.5 M concentration of KOH. The visual condition of the CF was tested using SEM images and the strength was tested. Tensile strength testing was done with a single fibre testing method with a speed of 5 mm/min. The length of the tested fibres was not informed, but it was noted that the diameter was decreased by 0.01—0.06 μm after recycling with optimized conditions. The virgin CF tensile strength was 4.22 GPa, and the recovered CF tensile strength was 4.07 GPa, so there was no significant drop in tensile strength, only approximately 5 %. The SEM images show a smooth surface on recycled CFs as can be seen in PICTURE 12. It was concluded that the solution was also recovered, and the depolymerized epoxy resin was reused with good results when mixed with fresh epoxy and curing agent. (Zhao et al. 2022.)



PICTURE 12: Images of (a) CFRP composite sample, (b) recovered carbon fibres, and SEM images of (c) virgin carbon fibres and (d) recovered carbon fibres, from mild chemical recycling at 160—163 °C for 60 min with KOH concentration of 0.5 M (Adapted with permission from Zhao et al. 2022. Elsevier).

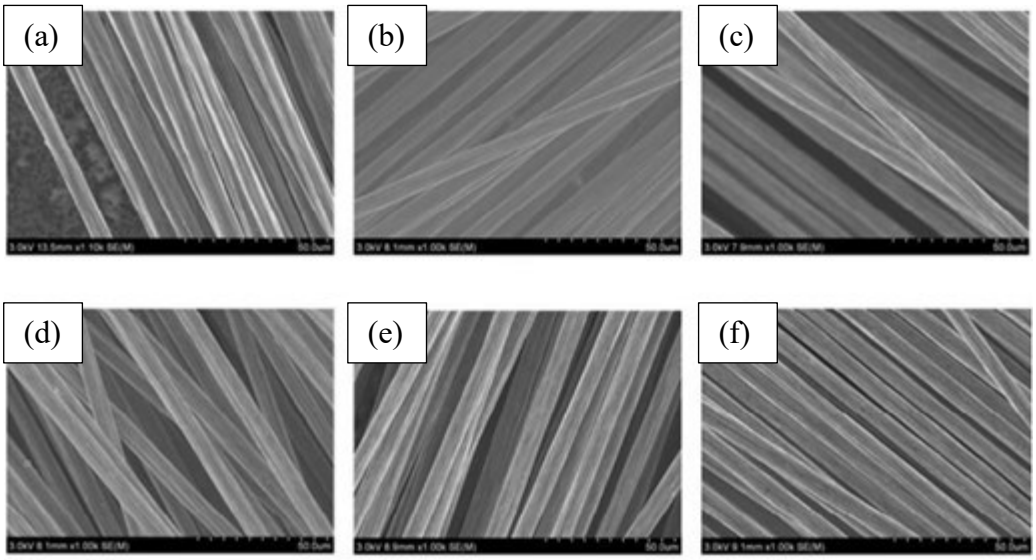
5.5 Recycling of aviation waste using peracetic acid as solvent

A mild oxidative chemical recycling process for CFREC using peracetic acid, which is a mixture of acetic acid and hydrogen peroxide, was studied. The ratio of the peracetic acid solvent was 95 vol% 14 M acetic acid and 5 vol% 9 M hydrogen peroxide (Ac95H5). This mixture was used due to its ability to best retain composition and strength and show the most potential possibilities for reusing. In the experiment, a 97% resin decomposition ratio was achieved, while maintaining good mechanical and visual conditions for the CFs. The process conditions for (Ac95H5) were tested by changing the volume of the solution per one gram of sample, and temperature. By using 60 mL of solvent per one gram of sample at 80 °C, the best outcome was achieved, a 98 % decomposition ratio, and the process time was the shortest, 4 hours. By decreasing the temperature to 65 °C, a 97.2 % decomposition ratio was achieved in 4 hours. The process itself was straightforward, the sample was induced in the solvent and heated. Further process parameters and material information are conducted in TABLE 10. (Das, Chacko & Varughese 2018, 1564—1571.)

TABLE 10: Process parameters and material information for mild chemical recycling with peracetic acid (adapted from Das et al. 2018, 1564—1571).

Materials:	CFREC: Resin, fibre precursor and resin/fibre ratio were unknown in this aviation waste CFREC. Testing with DGEBA type epoxy suggested the resin might be of that type. Sample size of CFREC: 30mm · 25mm · 2mm Optimized reaction medium: 95 vol% 14 M acetic acid and 5 vol% 9 M hydrogen peroxide
Temperatures in °C:	Testing temperatures: 28, 65, 80 Optimized temperature: 80
Pressure:	Atm
Reaction time in hours:	Testing reaction times: 4, 8, 84 Optimized time: 4

The tensile strength of the recovered CFs was tested after various peracetic acid ratios and compared to a virgin CF which was acquired in a mat form. The ratios varied as follows, Ac meaning the vol% of 14 M acetic acid and H the vol% of 9 M hydrogen peroxide, Ac95H5, Ac90H10, Ac80H20, Ac50H50 and Ac20H80. Ac90H10, Ac80H20 and Ac20H80 had the best tensile strengths, close to that of the virgin fibres, but the process parameters were not optimal. With optimized process parameters using Ac95H5, which also had the best reusability, the tensile strength of the CFs was 1.75 GPa, while the virgin CF's tensile strength was 2.3 GPa. The SEM images show a smooth surface with no resin residue for CFs recovered from using Ac95H5 as can be seen in PICTURE 13. (Das et al. 2018, 1564—1571.)



PICTURE 13: High-resolution SEM images of (a) virgin fibres and fibres recovered from using solvent ratios: (b) Ac95H5, (c) Ac90H10, (d) Ac80H20, (e) Ac50H50, and (f) Ac20H80 (Adapted with permission from Das et al. 2018, 1564—1571. Copyright 2018 American Chemical Society).

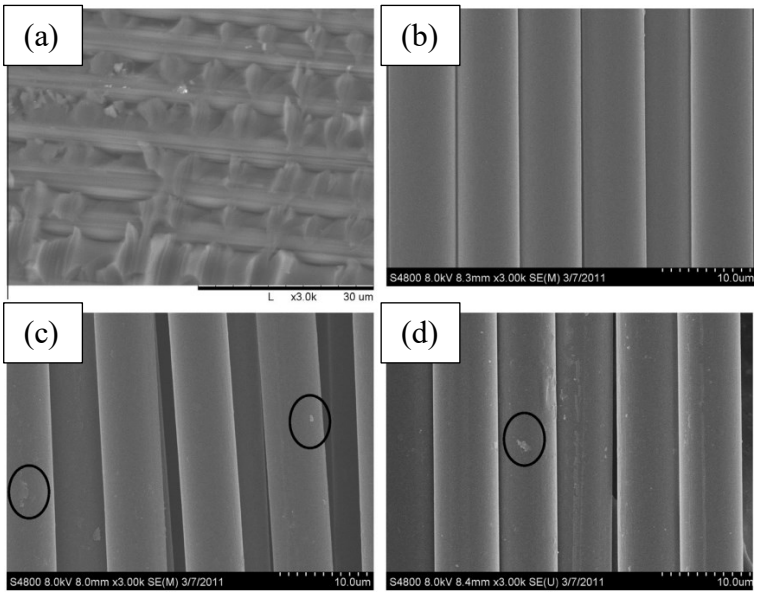
5.6 N, N-dimethylformamide and hydrogen peroxide as solvent

A mild reaction chemical recycling process, which includes the swelling of the CFREC with acetic acid followed by an oxidative degradation using N, N-dimethylformamide (DMF) and hydrogen peroxide (H_2O_2) at 90 °C for 30 min, was studied. The swelling was conducted at 120 °C for 30 min. The amount of recycled composite was 0.2 g, and the amount of solution was optimized to 15 mL of DMF and 15 mL of H_2O_2. The process was conducted at atmospheric pressure. The temperature of the reaction was tested at 80—150 °C, but it was optimized at 90 °C, and it was noted that it would be the efficient amount of energy consumed to acquire a good decomposition ratio for epoxy resin (>90 %) while maintaining good mechanical properties and tensile strength for CFs (>95 %). More process parameters and material information are conducted in TABLE 11. (Xu, Li & Ding 2013, 54—59.)

TABLE 11: Process parameters and material information for mild chemical recycling process using DMF and H_2O_2 (adapted from Xu et al. 2013, 54—59).

Materials:	Epoxy resin: JR-236 EP with the epoxide value of 0.54—0.57 Curing agent: acyclic amine JH-239 Carbon fibres: T-700 12 K Sample size of CFREC: 10 mm · 10 mm · 2.5 mm Optimized reaction medium: For 0.2 g of pretreated CFREC recycling, 15 mL of DMF and 15 mL of hydrogen peroxide (H_2O_2) were used.
Temperatures in °C:	Testing temperatures: 80—150 Optimized temperature: 90
Pressure:	Atm
Reaction time in min:	Testing reaction times: 5—120 Optimized time: 30

The optimized parameters gave good results on the tensile strength of the CFs. It was noted that the best mechanical properties were acquired by recycling under the temperature of 120 °C and that recycling at 90 °C instead of 100 °C was sufficient considering the lower consumption of energy. The tensile strength testing was conducted by using a fibre length of 2.5 cm and a test speed of 20 mm/min. It was noted that the tensile strength after recycling for 30 min at 90 °C was more than 95% of the virgin CFs value of 2.81 GPa. After treating at 100 °C for 30 min the tensile strength was about 97% of the virgin CFs strength. The SEM images show a smooth surface on CF with only a couple of white dots, which is speculated to be resin residue, as can be seen in PICTURE 14. In comparison with CFs recovered at 90 °C, CFs recovered at 150 °C are also shown, and it can be noticed that there is more damage on those CFs due to the higher reaction temperature. (Xu et al. 2013, 54—59.)



PICTURE 14: SEM images of (a) composite sample, (b) virgin carbon fibre, (c) carbon fibre recovered at 90 °C for 30 min, and (d) carbon fibres recovered at 150 °C for 30 min (Adapted with permission from Xu et al. 2013, 54—59. Elsevier).

6 DISCUSSION AND INTERPRETATION

In this chapter, a detailed discussion of the reviewed articles in chapters four and five are included. The discussion will be based on the chemical degradation mechanism, properties of the recovered CFs, reusability of degraded epoxy resin, and possible solvent recovery routes whichever applies to the reviewed article and method.

6.1 Recycling using sub- and supercritical fluids

In the recycling experiment using supercritical 1-propanol from Yan et al. (2016, 46—54) the reaction conditions differed in comparison with the experiment from Okajima et al. (2017, 44—51). Yan et al. (2016, 46—54) used a temperature of 320 °C for one hour, without any mention of used pressure. The reactor was a stirred batch reactor so it can be assumed that it was depressurized, or the pressure raised with temperature. The catalyst used was KOH with the amount of 1.0 wt.%. It was noticed that bigger amounts of KOH resulted in poorer CF conditions due to the reaction between KOH and carbon atoms on the fibre surface. The promoting catalytic effect of an optimized amount of KOH in the degradation of the resin was due to the 1-propanols hydrogen donating property, and it was said that a simple increase of KOH did not aid the degradation of the resin. The sample sizes were almost the same in both studies. Although the composite sample lost its woven structure during the recycling process, it was noted that the good quality recovered fibres could be well reused in the automotive or sporting industry or injection moulding. (Yan et al. 2016, 46—54.)

Okajima et al. (2017, 44—51) had a result of 100% decomposition of epoxy resin at a temperature at similarly 320 °C, but the reaction time was only 25 min and there was no use of a catalyst. It can be assumed that the use of higher pressure at 9 MPa leads to a shorter reaction time than in the study of Yan et al. (2016, 46—54). There was no mention of the recovered CF condition after this supercritical 1-propanol recycling method, most likely because the study was focused on the recycling process using a semi-flow-type bench plant with an optimized subcritical acetone solvent. Subcritical acetone was used at 320 °C for 20 min at 1 MPa of pressure. Acetone was chosen from other solvents for its smaller molecular size which improved for instance the solvents diffusing in epoxy and decomposing the bridge structured resin. In the bench plant, the composite fabric retained its woven structure and

almost full tensile strength. This was assumed to be due to the way of solvent flowing through the reactor where the sample was placed. Eventually, the solvent was collected after cooling and filtration. It can be assumed that in the use of high-temperature and pressure chemical recycling the batch reactor could be used to select the optimized solvent for epoxy decomposition and then it could be tested in a semi-flow-type bench plant to optimize other process parameters. The collected solvent and resin were not tested. (Okajima et al. 2017, 44—51.) From these methods, the reuse of resin and solvent should be further investigated.

Acetone was also studied by Oliveux et al. (2015, 96—103) where they mixed it with water. With a higher pressure of 23.5 MPa used and a higher temperature at 350 °C it suggests that it is not as effective as recycling with only acetone. (Oliveux et al. 2015, 96—103.) Keith et al. (2019, 225—234) also studied the solvent mixture of acetone and water, and it was noticed that the reaction required a higher pressure and a longer reaction time, 19.7 MPa and 120 min, respectively, which also suggests that acetone alone as a solvent was more effective.

Acetic acid as a solvent was studied by Xing et al. (2021) and Shetty et al. (2022). Xing et al. (2021) used a swelling method in temperatures of 100—220 °C, where the layers of the composite sample were delaminated at 160—220 °C. The pressure was 1.03 MPa in an autoclave, and the reaction time for delaminating the composite sample was one hour. Acetic acid was said to be a good solvent for swelling of the epoxy resin due to its hydrogen-donating properties. The epoxy resins and acetic acids' hydrogen bonding are important for the swelling of the resin. The temperature was studied in the swelling treatment, and it was noted that solvent absorbance in the resin increased. It was said that with sufficiently high temperature the energy would be enough to break epoxy bonds. The raising of temperature therefore improved the swelling and mass loss rate. Based on Fourier Transform Infrared Spectroscopy (FTIR) the C-N and C-O-C bonds were cleaved during swelling at 220 °C for one hour. With this method, the composite sample retained its woven structure and was easily remanufactured as a CFREC board. From covered methods, this shows good potential due to its ability to use lower temperature and pressure than other covered solvolysis methods while retaining the woven structure of the composite sample. The downside was the lower mass loss rate which was 25 % at 220 °C after one hour. The remanufactured CFREC board retained 89% tensile strength compared to the virgin CFREC board. This study showed possibilities for delaminating bigger waste, such as wind blades. (Xing et al. 2021.)

Shetty et al. (2022) also experimented with the use of acetic acid mixed 50:50 with de-ionized water but with higher temperatures at 300 °C with a pressure of 10 MPa for 30 min. They came to a result that initial swelling at 200—250 °C for approximately an hour with acetic acid helped degrade the epoxy and the degrading began rapidly in temperatures of 275—300 °C for approximately thirty min. The results showed good visual condition of fibres and analysis showed the presence of reusable aromatic and aliphatic groups in solvent and resin recovery. (Shetty et al. 2022.)

The study of recycling with water in supercritical conditions by Kim et al. (2019, 66—72), using high temperatures at 405 °C with a pressure of 28 MPa for 60 min to decompose the epoxy resin with 99.5 % efficiency from CFREC composite sample, was characterized as a green method. Supercritical water was used instead of subcritical water due to its ability to penetrate quickly into the CFREC microcavities. It was due to its low viscosity, quick heat and mass transfer, and high diffusion coefficient that it degraded epoxy effectively. The recovered resin included o-Cresol, p-Cresol, m-isopropyl phenols, m-xylene, and m-ethyl phenol which were phenol-like petrochemical products. The recovered CFs were impregnated with CBT to form CF-reinforced thermoplastic composites. The method showed potential using water as a solvent and degrading the epoxy effectively, but it caused a comparably high decrease in tensile strength for the CFs. (Kim et al. 2019, 66—72.) It can be assumed that the high consumption of energy due to the high reaction conditions adds costs to the method.

The semi-flow-type bench plant was also experimented with using supercritical methanol at 285 °C and 8 MPa for 80 min, by Okajima et al. (2014, 68—76). The use of this bench plant in this study is showing good results in keeping the woven structure of the CF fabric intact, which assumably makes it easier to remanufacture into a new CFREC composite. It was assumed that in the epoxy resins bridge structure, only the ester bonds were broken. This study also showed the possibilities of reusing the recovered resin by curing it with different mixtures of virgin epoxy resin, and it was noted that the strength decreases with an increase in recovered resin content. For example, the strength of the reused cured epoxy resin was 58% compared to the virgin epoxy resin, when 50% of recovered epoxy resin was used. (Okajima et al. 2014, 68—76.) It can be assumed that the use of this bench plant in higher reaction conditions chemical recycling helps in the recovery of the resin and solution.

It should be noted that the best conditions both visually and tensile strength-wise while keeping the reaction time at a minimum, were obtained when initially swelling with acetic acid or using sub- or supercritical acetone in solvolysis. This could be further studied if it is possible to combine these methods with a bench or pilot plant to recover waste CF in a bigger size with good mechanical conditions

while also reusing the resin and solvent. The bench plant that was used in two of the reviewed studies showed potential in keeping the CF fabric woven structure intact and collecting the resin and solvent for reuse.

6.2 Recycling using mild reaction conditions

In the recycling study using ionic liquids, it was noted that the ionic liquid [BMIm][Cl] combined with EG swelled the composite well but did not have the property to dissolve the epoxy, so a carboxylic anion ([$Acet^-$]) was added. In the swelling study, it was hypothesized that [BMIm][Cl] penetrates the polymer matrix the best of the three candidates due to its shorter alkyl chain. The swelling and dissolving of the epoxy were conducted at 150 °C using atmospheric pressure. Compared to higher reaction conditions, these conditions are safer, but the reaction time was 150 min, which is much longer than in higher reaction conditions. It was hypothesized that the carboxylic anions electrons present in oxygen helped dissolve the epoxy by attacking the ester bonds. This study showed good potential in recyclability for the used solvent, and the recovered monomer could be precipitated from the liquid solvent with water. The solvent itself was tested for reusing and it was noted that the next recycling reaction time was 30 min longer with each reuse. This was assumed to be due to either the presence of residual water or partial decomposition of the solvent. This study showed good visual condition for recovered CFs and it was concluded that the fibres were reused as part of remanufactured CFREC with good results on mechanical properties. By reusability of fibres, solvent and recovered monomers, this study showed great potential, but it was concluded that the materials used in the method would be costly for large-scale testing. The prize for recycling 1 kg of epoxy with this method was compared to a study using TBD-EG/NMP solution. The prize was 1. 25 million USD for this method and 2.2 million USD for the other one. (Pérez et al. 2021, 5588—5595.)

It was speculated in a study using DESs for chemical recycling that the acidic conditions paired with an optimized metal salt promoted good swelling and dissolving of epoxy resin in the composite material. It was speculated that the stronger Lewis acidity of the chosen catalyst improves the navigating and cleaving of C-N and C-O bonds in the epoxy resin, and the longer alkyl-chained fatty acid improved the decomposition of the epoxy resin. (Liu et al. 2022.) Other articles in the literature support the use of metal salts for cleaving the C-N bonds (Liu, Zhang, Guo, Liu, Liu, Xin & Zhang 2017, 20—27; Wang, Cui, Ge, Yang, Wang, Zhang, Li, Deng, Qin & Hou 2015, 3332—3337). In the study, the temperature was 180 °C, and atmospheric pressure was similarly used as in the above-mentioned mild

reaction condition process. This study using DESs showed promising results on solvent recovery and reuse, resin recovery, and CF yield. The CFs showed only a 5.5 % reduction in strength compared to virgin CFs. The resin was decomposed into bisphenol A monomers and derivatives, and the recyclability of the solvent showed similar properties as the recycled ionic liquid solvent. It retained its recycling properties entirely with increased reaction time. (Liu et al. 2022.)

The good reusability of recycled commercial CFREC and epoxy oligomer was discovered in a study using mild reaction conditions including atmospheric pressure and a temperature only at 170 °C. This study using organic solvents showed that the composite samples could retain their woven structure. The reaction time was 90 min, and the experimenting on commercial drone wings showed that they retained their structure and the solvent with the dissolved epoxy was recovered. In the method the organic solvent NMP did the swelling of the epoxy while guiding the TBD-EG solution to dissolve it. The catalyst TBD activates the epoxy resin and with the alcohol breaks the ester bonds. The reusability and properties of the depolymerized epoxy oligomer were tested, and it was found that it had multifunctional hydroxy groups and its reuse possibilities in polyurethane were speculated. The tensile strength of the recured epoxy decreased when the amount of depolymerized epoxy oligomers increased. The CFs kept their mechanical properties almost entirely while maintaining the structure, which shows potential for reuse. (Kuang et al. 2018, 9189—9197.)

The use of KOH as a cocatalyst in a recycling process with MEA was found to be effective in mild reaction conditions. This shows that the use of KOH as a catalyst in higher reaction conditions and mild reaction conditions is promising. This study did not include an initial swelling of the composite sample. The cleaving of the ester bonds in the epoxy was studied to happen due to the amination of the ester bond and the amino group of MEA, and hydrolysis of the ester bonds and KOH. The hydrolysis between the ester bonds and KOH was found to be due to the formation of potassium carboxylate groups. After the recycling, the ester bonds transformed into amide, hydroxyl, and carboxylate groups. The reaction time is comparably low, only 60 min at 160 °C and atmospheric pressure. The recovered fibres showed good visual and mechanical condition, with the downside that the composite sample did not retain its structure. After resizing they could be reused. The solution was successfully recovered as well as the degraded epoxy resin, which was re-cured with different ratios to new epoxy resin. The use of 10 wt.% of recovered epoxy resin showed a 50% decrease in strength. The reuse of the MEA solvent showed a similar nearly 100 % decomposition ratio with similar reaction conditions but with a 60-min increase in reaction time. (Zhao et al. 2022.)

A mild reaction condition recycling process at 80 °C and atmospheric pressure was used to successfully recover CFs, solvent, and resin from the process. The solvent used in the recycling of CFREC was a mixture of acetic acid and hydrogen peroxide. The solvent ratio was optimized to maximize the reusability of the solvent and the CFs recovered with it showed good mechanical and visual properties. The composite sample did not retain its structure. The epoxy degraded due to the cleaving of C-N bonds. Also, another chemical change was observed which was the oxidation from C-OH groups to C=O. The distillation of the recovered solvent broke the polymer further down to aliphatic and aromatic compounds such as phenolics, and it was reused in adhesive-grade epoxy. (Das et al. 2018, 1564—1571.) The use of hydrogen peroxide was also studied with DMF in mild reaction condition recycling of CFREC. This method used acetic acid for swelling at 120 °C for 30 min as an initial stage to accelerate the degradation of the epoxy. The dissolving of the epoxy took 30 min at 90 °C and atmospheric pressure. The epoxy was decomposed in an oxidative way gradually by H_2O_2 and with the catalytic help of DMF. The recovered CFs showed good mechanical and visual properties, and the process itself was speculated to be promisingly scalable. The solvent mixture was successfully recovered but the properties and reusability of the solvent and epoxy resin were not studied. (Xu et al. 2013, 54—59.)

7 CONCLUSIONS AND FUTURE OUTLOOK

In this thesis, a literature review was conducted to discuss and compare the chemical recycling methods of CFREC. The aim was to compare recycling methods for future laboratory testing and help bring more knowledge in resolving the current state of recycling of CFREC while gathering information on the current state of reuse possibilities of the recovered CFs, resin, and solvents. Most of the research found in the literature has optimized chemical recycling methods aiming for a good yield of CF with good mechanical properties. In addition, it was valuable to find and compare methods that also had a good yield of degraded resin and solvent. The research found in the literature was divided into two categories, higher reaction condition chemical recycling and mild reaction condition chemical recycling of CFREC.

The research found in the literature reveals many possibilities to recover CFs with good mechanical properties using higher reaction conditions. The main factors that decreased the mechanical properties were the excessive use of catalysts, such as KOH, and the temperature being too high. With the use of pressure, the temperature was often optimized to maintain the mechanical properties at a low reaction time. The optimized amount of catalyst also helped decrease the reaction time. The reaction times differed from 20 to 120 min. Acid digestion showed promising results as an initial swelling stage before degrading the epoxy from the CFREC with higher reaction conditions. It can be assumed that the acid digestion softens the structure of the composite sample and helps in the epoxy decomposition depending on the solvent and other process parameters.

In the reviewed articles, the most used solvents in higher reaction condition chemical recycling were alcohols, ketones, or water, pointing out the effectiveness of organic solvents in recycling CFREC. Acetic acid was also used as a solvent and in swelling of the composite. From the reviewed studies in higher reaction condition chemical recycling, subcritical acetone showed the most promising results and should be considered a candidate for future studies in higher reaction conditions. With the use of acetone, the decomposition of epoxy was 100 % and the reaction time was the shortest, 20 min. The temperature and pressure did not differ notably from other research with higher reaction conditions, they were 320 °C and 1 MPa. It was due to the small molecular size that it diffused well in epoxy and decomposed the bridge-structured resin. These results were obtained using a bench plant, which gives a promising outlook for piloting. The recovered CF was intact and retained its woven structure which could be promising on behalf of piloting with even bigger CFREC waste. The recovery of resin and

solvent was proven possible with this method. After reviewing studies, it was learned that the initial swelling with acetic acid could decrease the reaction time or lower the process conditions such as temperature and pressure. This is something to take into consideration for future studies.

The research found in the literature for mild reaction condition chemical recycling was also promising. Compared to higher reaction conditions, the temperatures used in mild reaction conditions are much lower, varying from 80 °C to 180 °C in the reviewed studies in this thesis. The pressure used in this research was found to be atmospheric, which is also much lower than in higher reaction condition recycling and can be noticeably safer considering piloting possibilities. The reaction times on the other hand were longer than in higher reaction conditions, varying from 0.5 to 4 hours. The solvents used in the reviewed studies in mild reaction conditions are organic, green, and sustainable, especially the ionic liquids and DESs reviewed in chapter five. This is also an advantage compared to higher reaction conditions, but it should be noted that the green solvents used in mild reaction condition research were considered more expensive. The comparison in cost assessment should be conducted to find out the differences in energy consumption and material expenses between mild and higher reaction conditions, to help find the most affordable method without compromising the results.

The recovered CF fabrics from the recycled CFREC mostly retained their woven structure in mild reaction condition chemical recycling, with good yield and mechanical condition. With the use of lower temperatures and atmospheric pressure, the use of batch reactors could be considered for piloting and bigger waste. The recovery of degraded resins as monomers or oligomers in chemical recycling using mild reaction conditions was found to be more studied. The reusability of the solvent was found promising in the research found on ionic liquids, DESs and MEA-KOH solution. Their ability to maintain the decomposition effectiveness for degrading the epoxy with increased reaction times could be useful for bigger-scale recycling. Research has been focused on finding the applications for recovered epoxy resin components. It was suggested to reuse the recovered resin in small proportion with adhesive grade epoxy due to the recovered resin's brittleness after mild reaction condition recycling. In mild reaction condition recycling the use of initial swelling was found useful in several reviewed studies, following the degrading of the epoxy with H_2O_2, DMF, ionic liquids, or DESs. Depending on the budget these reviewed methods and solvents should be considered for future studies.

In conclusion, for further research the use of acid digestion and swelling of the CFREC should be considered, followed by solvolysis with subcritical acetone or mild reaction condition recycling with

aforementioned solvents. The reviewed research found in the literature gives an idea of the possibilities of chemical recycling and helps in gathering more knowledge on the topic. The scale and budget of the recycling help to guide choosing the fitting parameters, solvents, and catalysts for the optimized method. To optimize a method for chemical recycling of CFREC, considerably more research should be covered, and data should be collected from future laboratory testing.

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