

eingereicht/handed in: 03.07.2020
angenommen/accepted: 25.08.2020

Magnus Gebhardt, M.Sc., Dr. Souvik Chakraborty, Dr. Ioannis Manolakis, Prof. Dr. Dieter Meiners
Institute of Polymer Materials and Plastics Engineering, Agricolastraße 6, TU Clausthal, D-38678 Germany
Precision Engineering, Materials & Manufacturing (PEM) Research Centre & Department of Life Sciences, Institute of Technology Sligo, Ash Lane, Sligo, Ireland

Closed-loop room temperature recycling of Elium CFRPs and its influence on the 2nd generation composite properties

This research work presents a recycling methodology for carbon fibre composites processed with a novel room temperature infusible and curable thermoplastic matrix ELIUM 150 from Arkema. Herein we demonstrate that by dissolving the composites in acetone at room temperature it is possible to separate the matrix from the fibres and to reuse both the matrix and the fibres in form of scrim (preform). A comparison of the thermomechanical, thermal and mechanical properties of the composites shows only marginal performance changes in bulk properties due to the proposed recycling strategy.

Closed-loop Raumtemperatur Recycling von Elium CFK und dessen Auswirkung auf die Eigenschaften der 2. Generation Composites

In diesem Paper wird eine Recyclingmethodik für Carbonfaser-Composites mit einer neuartigen bei Raumtemperatur infundier- und aushärtbaren thermoplastischen Matrix ELIUM 150 des Herstellers Arkema aufgezeigt. Durch das Lösen der Composites in Aceton ist es möglich die Matrix von den Fasern zu trennen und sowohl die Matrix als auch die Fasern in Form eines Geleges wiederzuverwenden. Ein Vergleich der thermomechanischen, thermischen und mechanischen Eigenschaften der Composites zeigt nur geringe Performanceverluste durch das Recycling.

Closed-loop room temperature recycling of Elium CFRPs and its influence on the 2nd generation composite properties

M. Gebhardt, S. Chakraborty, I. Manolakis, D. Meiners

1 INTRODUCTION

1.1 Current status of composite recycling

The current status of composite recycling is generally divided into mechanical, thermal and chemical recycling [1,2,3]. Mechanical recycling is understood to be the crushing of composites by shredding and/or grinding. The use of the recyclate as fillers in concrete to improve its mechanical properties has already been reported [4,5,6]. Furthermore, the use of recycled fibres as short fibre reinforcement in composites is reported to have only minor deviations in mechanical strength compared to 1st generation short-fibre composites [7,8]. However, due to the shredding of the fibres and the matrix partially adhering on to them, the strength values of 1st generation continuous fibre-reinforced composites is seldom achieved [9].

Thermal recycling includes pyrolysis, fluidised-bed pyrolysis and microwave pyrolysis [1,2]. Pyrolysis is currently the most mature industrial scale recycling technology [10,11]. Thermally recycled fibres are mainly chopped or milled and used as fillers, e.g. in injection moulding processes or in compression moulding of non-woven mats [12]. In thermal recycling, a reduction in mechanical properties caused by unavoidable variations in the surface chemistry and texture is often reported regardless of the process [13,14]. In addition, in all industrial pyrolysis applications the recycled fibres are shredded, which, as already mentioned, leads to significantly downsized potential.

Chemical recycling is by far the most fibre-friendly recycling method (least influence on the structural integrity of the fibres) and can be divided into solvolysis and the supercritical fluid method [2]. In solvolysis, the matrix is depolymerized under elevated pressure and temperature [15,16]. The supercritical fluid method works similar to solvolysis while making use of the good mass transfer properties of supercritical fluids [2]. For both methods, to the best of the authors' knowledge, industrial upscaling is only reported in very few attempts by now. Moreover, only a small number of studies have been reported on further processing of these reclaimed fibres. Oliveux et al. [16], for example, used recycled fibres to produce discontinuous carbon fibre tows. In another case, the direct re-infusion of the recycled fibre mats was investigated [17]. With regards to the recovery of long fibres and thus the preservation of

their structural properties, chemical recycling processes probably offer the most promising potential.

1.2 New recyclable matrix materials

In addition to the three recycling options described above, a fourth option can be the dissolution in a common solvent under benign conditions i.e. dissolving the matrix without excessively increasing the temperature and/or pressure. In this regards, the thermoplastic material Elium developed by ARKEMA in 2014 has been investigated here. This material is a room temperature curable and infusible thermoplastic matrix [18]. This feature makes it possible to combine the advantages of a thermoplastic matrix with the simple processing ease of a thermosetting resin without the need for high processing temperature [19]. The material which consists of acrylic acid, methacrylic acid and their methyl esters and other unspecified acrylic copolymers [20], is polymerized in situ using a peroxide initiator and can be processed in the same way as conventional epoxy resins using vacuum assisted resin infusion (VARI) [19,21] and RTM [19,22-25]. From published literature, both the thermal [20,26,27] and mechanical properties [21,24-31] of Elium composites can be considered to be comparable to common epoxy based composites.

Considering the high market potential of this material, it is therefore of crucial importance that within the aegis of circular economy concept to develop recycling strategies for Elium composites right from the market growth phase. However, according to best of our knowledge there is not much work detailing the recycling of Elium composites. Cousins et al. [32] demonstrated limited recyclability of ground Elium glass fibre composites by injection moulding. They also reported on the possibility of dissolving Elium in chloroform to recover the fibres. However, there seems to be a lack of studies that deal with the recovery of undamaged fibre preforms retained in shape by the stitches and subsequently used to manufacture second generation composites.

Due to the obvious risks of using a solvent like chloroform [33,34] for scale up purposes, a common industrial solvent – acetone, is investigated here for dissolution. Acetone is a commonly used solvent to remove PMMA based photoresistor in the nanoscale electronic research industry [35]. Since MMA is a constituent of Elium, it seems to be a natural choice for this work.

This experimental research work details the recyclability of Elium CFRP to recover both matrix (in contrast to thermoset solvolysis [15,16] where the matrix is degraded) and undamaged carbon fibres scrims by dissolution in acetone. The undamaged carbon fibres recovered were used to process 2nd generation laminates with virgin Elium. A range of tests showed that the 2nd generation composites have similar mechanical and thermal properties to the 1st generation ones. These analyses demonstrate that the proposed recycling technology leads to minimal downsizing of bulk properties and hence is advantageous when compared to current reported literature. In addition, this reported process

offers a high potential for industrial applicability, as it requires only a minimum energy input with good scale-up possibility. In our previous study [36], the influence of temperature was investigated as well as the solvent ratio and dissolution time were optimized to achieve the highest possible matrix recovery with a minimal damage to the preform and minimum energy consumption.

2 EXPERIMENTAL SECTION

2.1 Materials

The composites were processed with Elium 150 from Arkema. The resin was polymerized in situ with the addition of 2.5 wt% of the initiator Perkadox CH-50X (dibenzoyl peroxide containing dicyclohexyl phthalate) from AkzoNobel. The 0/90° carbon fibre scrim B-C-636 g/m², width: 1270 mm from SAERTEX was used as reinforcement. The stitches were made of polyester (areal weight: 6 g/m²). Frekote 770NC from Henkel was used as the mould release agent. The dissolution trials were carried out with analytical grade acetone from Henkel (purity: 99.5%).

2.2 Composite processing

For the test specimens, CFRP laminates in the size 400x300 mm² and a thickness of 1.7-2 mm were processed using the VARI method. Two fabric layers were used with a final fibre volume content (V_f) of 45% (approx.). The assembly layup was carried out on a glass plate coated with Frekote 770NC. The infusion process was based on the guidelines detailed by the manufacturer [19]. Elium 150 was mixed manually by gradually adding Perkadox CH 50-X and degassed for 10 minutes under a vacuum of 20 mBar. The subsequent infusion was carried out from the longer side of the reinforcement. After the infusion, the laminate was wrapped in a heat blanket and cured for 1.5 hours at 65°C. In-house observations, as well as reports by Chilali et al. [21] and Kazemia et al. [30], confirmed that the post-cured composite (1 h at 65°C) does not have any noticeable difference in properties compared to a composite cured for 24 h at room temperature. Curing at 65°C was therefore chosen to reduce the curing time from 24 h to 1.5 h. After curing, the specimens were machined to the sizes required for dissolution and further testing.

2.3 Recycling via Dissolution

For recycling via dissolution, CFRP test specimens of the size 200x180 mm² (1st generation composites) were used. The test specimens were measured individually (dimensions, weight) and then placed in a closed container with

acetone at room temperature. The weight ratio was CFRP:Acetone :: 1:20. This was determined by pre-testing with bulk Elium as the best possible ratio for a complete dissolution. The test specimens remained in these enclosed containers for 24 hours at room temperature and were checked for pliability and layer separation immediately after removal. The fibres for the single fibre tensile test were also separated at this stage. The remaining preform was dried for 24 hours under ambient conditions. Contemporarily, the dissolved matrix was recovered by evaporating the acetone at room temperature in a fume hood. The recycled matrix and fibres were then dried at 65°C for 8 hours under vacuum in order to remove any residual acetone.

The described recycling process provides the possibility to reclaim both the matrix and the fibres for future reuse. It is possible to separate the individual scrim layers from each other without damaging the integrity of the layers / preform. In this context, preform is defined as fibres recovered as scrim. The matrix can also be recovered by evaporating the acetone. In contrast to our results on a smaller scale [36], further process optimization and upscaling to the laminates seemed to increase the recovery of the matrix significantly, Fig. 1.

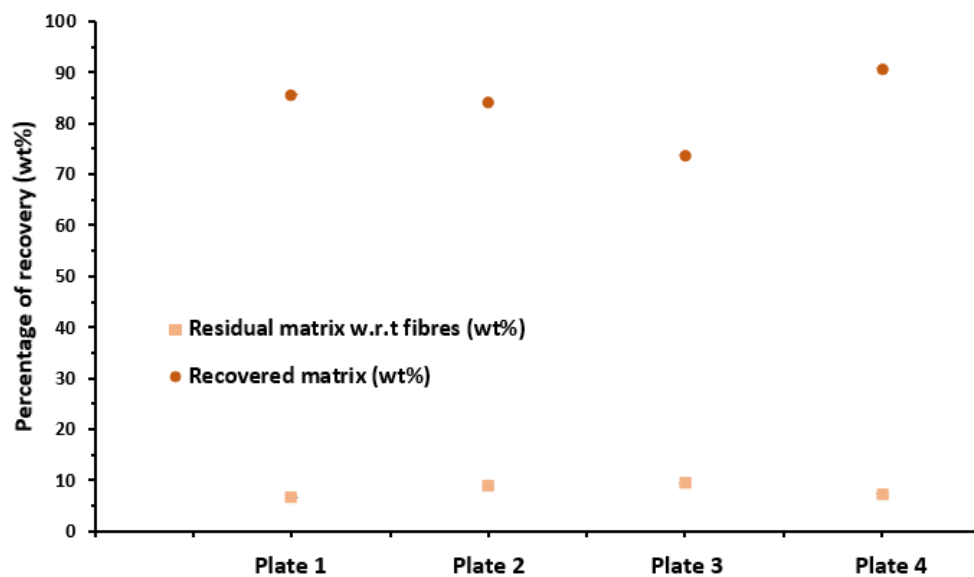


Figure 1: Recovery rate of matrix in the recycling process, size of laminates 200x180 cm²

With the exception of a few fibres at the edge of the scrim - which were unavoidably displaced during the recycling process, the recovered preform was entirely intact and could be reused as it is. The average matrix recovery rate has been calculated at 81%. The amount of matrix remaining on the fibres was approx. 8% (with respect to the fibres). The remaining matrix became visible microscopically when removing individual fibre bundles from the recycled fibres

directly after the dissolution process, Fig. 2. The SEM section of this paper discusses this topic in more detail.

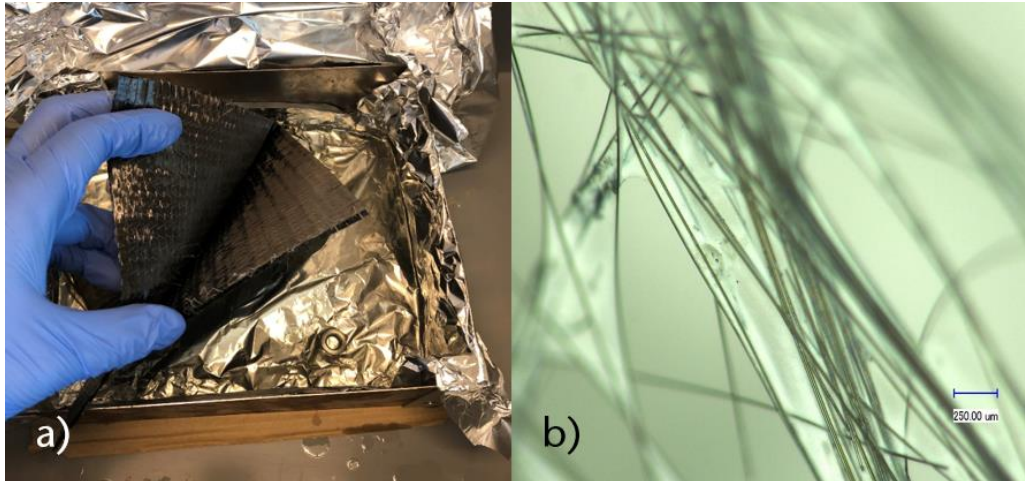


Figure 2: a) Preform after removal from acetone, b) Optical micrographs of fibres after acetone removal

2.4 Processing of 2nd generation test specimens

New composites are hereafter called 1st generation composites and composites produced with recycled fibres are hereafter called 2nd generation composites. With regards to the matrix and the single fibres tested, we refer to “virgin” for new/freshly polymerised material and “recycled” for recovered material.

The 2nd generation composites were processed using the recovered and dried preforms. The preforms were infused with virgin/fresh Elium150 via VARI as described in Section 2.2 and cured for 1.5 h at 65°C in a heat blanket (Fig. 3).

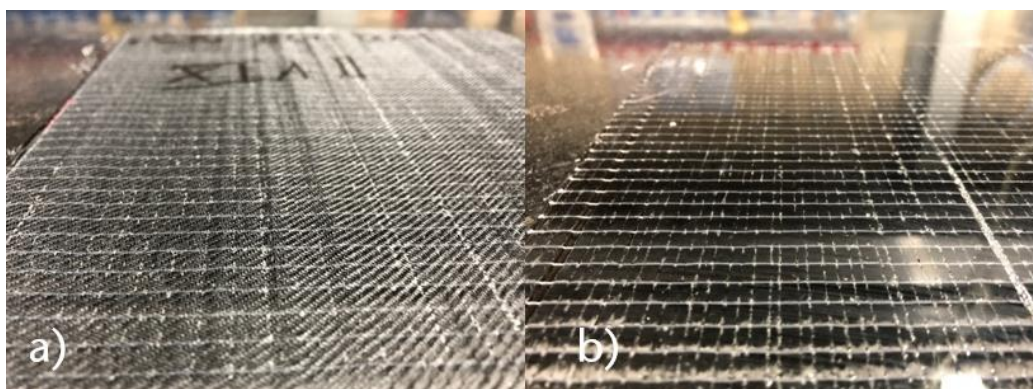


Figure 3: 2nd Generation CFRP made with recycled fibres a) top surface b) bottom surface

Dynamic mechanical analysis (DMA) test specimens in the size 60x10 mm², three-point bending test specimens in the size 150x15 mm² and interlaminar shear strength (ILSS) specimens in the size of 20x10 mm² were machined from these laminates for testing. All samples had a thickness between 1.7 and 2.0 mm. The infusion time was 40 % shorter than for first generation composites.

2.5 Characterization techniques

The chemical composition of the recycled matrix was studied by means of Attenuated Total Reflectance Fourier Transform Infrared Spectroscopy (ATR-FTIR). The device was Alpha T-IR from Bruker and the spectra was recorded in the frequency range of 400 to 4000 cm⁻¹. All values were normalized against the 1720 cm⁻¹ (ester carbonyl) peak.

The thermal analysis of the recycled matrix was carried out by differential scanning calorimetry, DSC (DSC 2920, TA instruments) according to DIN EN ISO 11357-1 [37] and thermogravimetric analysis, TGA (TGA 2950, TA instruments) according to DIN EN ISO 11358 [38], both under nitrogen atmosphere. The DSC was used to determine the midpoint glass transition temperature (T_g). The T_g is strongly dependent on the molecular weight values and distribution, and thus allowed conclusions to be drawn about the characteristics of the recycled material in comparison to the virgin material. This in turn revealed information about the effect of the processing conditions and the thermal history of the material during the recycling process [39]. DSC measurements were conducted following a heat-cool-heat regime from -20 to 200°C (heat up to 200°C at a ramp 10°C/min; cool down to -20°C at a ramp 50°C/min; followed by heating to 200°C, ramp 10°C/min). The T_g values were always taken from the second run to exclude any effect of thermal history [40]. Two measurements were performed for each specimen for repeatability. The temperature range of the TGA measurement was 25-600°C at a ramp of 10°C/min. The thresholds of 5%, 10% and 50% weight loss, which are important for the thermal stability of a material [41] were investigated.

The single fibre tensile test was carried out with fibres with a length of 50-60 mm using both virgin and recycled fibres in accordance to ISO 11566 [44]. The fibres required for the tests were randomly extracted from the individual preforms under a microscope immediately after the dissolution process and fixed between 2 pieces of paper by gluing with Araldite AW4859/HW4859. The free length between the glued areas was 30 mm. This fixture was clamped in a Zwick/Roell ZMART.PRO tensile testing machine operating with a load cell of 20N at a preload of 0.001 N (Fig. 4). The failure load was measured to get an understanding of the influence of the recycling processes on the structural integrity of the fibres. The data obtained was evaluated by Weibull analysis as is common practice [45-48]. At least 20 fibres were always examined for statistical relevance.

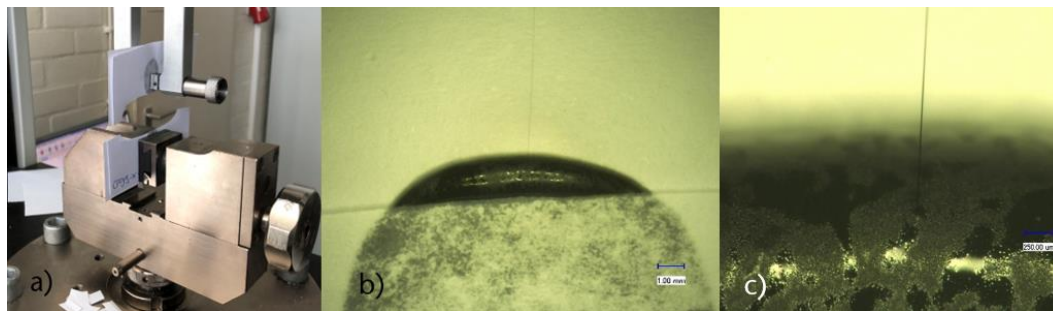


Figure 4: Single fibre tensile Testing a) fixture b) fixed single fibre c) close up single fibre

The surface morphology of the carbon fibres (virgin and recycled) was probed using scanning electron microscopy, SEM (DSM 982 Gemini, Zeiss) with an accelerating voltage of 5 kV in the secondary electron (SE) mode. Energy dispersive X-ray (EDX) analysis of the fibre surfaces was performed in the same instrument.

Dynamic Mechanical Analysis (DMA) was used to compare the thermomechanical properties of the 1st and 2nd generation composites. The measurements were performed with a DMA 2980 from TA Instruments according to DIN EN ISO 6721-1 [42]. The measurements were performed in the temperature range -10°C to 160°C using a heating ramp of 2°C/min in a double cantilever mode at a frequency of 1 Hz. The storage modulus (E'), loss modulus (E'') and loss factor ($\tan \delta$) were recorded. In each case, four measurements were carried out and the mean values reported. Two relevant values were noted: the onset glass transition ($T_{g(\text{Onset})}$) which was calculated from E' , and the maximum of the $\tan \delta$ curve ($T_{g(\tan \delta \text{ max})}$).

The interlaminar shear strength (ILSS) of the 1st and 2nd generation composites were tested according to DIN EN ISO 14130 [43]. The ILSS tests, like the three-point bending tests, were performed at an ambient temperature of 23°C and 45 % humidity. The ratio of test specimen thickness to span was kept at 1:5 and adapted for each specimen.

The three-point bend (3PB) tests to investigate the flexural properties of the 1st and 2nd generation composites were carried out according to DIN EN ISO 14125 [28]. The test was carried out using a Zwick/Roell BZ2-MM100TL.ZW01 universal testing machine operating with a load cell of 100 kN at a preload of 5 N and a thickness to span ratio of 1:16. The specimens were tested with the 90° fibre layers on the outside.

3 RESULTS AND DISCUSSION

3.1 Infrared spectroscopy (FTIR)

Recycled Elium showed a very similar FTIR spectrum to the virgin material, and both polymers to that of standard poly(methyl methacrylate), PMMA [50, 51]. The characteristic absorption bands for methyl and methylene C-H stretching ($2249, 2292\text{ cm}^{-1}$), ester carbonyl (1720 cm^{-1}) and C-O-C (1143 cm^{-1}) are present in both (Table 1), and the overall spectrum remains essentially unchanged between virgin and recycled Elium. Also the peak heights of the values normalized to 1720 cm^{-1} are almost identical (Table 1). Hence it can be postulated that the recovered Elium after dissolution in acetone has no effect in terms of chemical composition in comparison to the virgin Elium matrix.

Vibration	Virgin Elium	Recycled Elium
C-H stretching	2992 cm^{-1}	2992 cm^{-1}
C-H stretching	2949 cm^{-1}	2949 cm^{-1}
C=O	1720 cm^{-1}	1720 cm^{-1}
C-H bending	$1448/1434\text{ cm}^{-1}$	$1448/1434\text{ cm}^{-1}$
C-O-C	1143 cm^{-1}	1141 cm^{-1}

Table 1: Characteristic FTIR Peaks of Elium before and after recycling

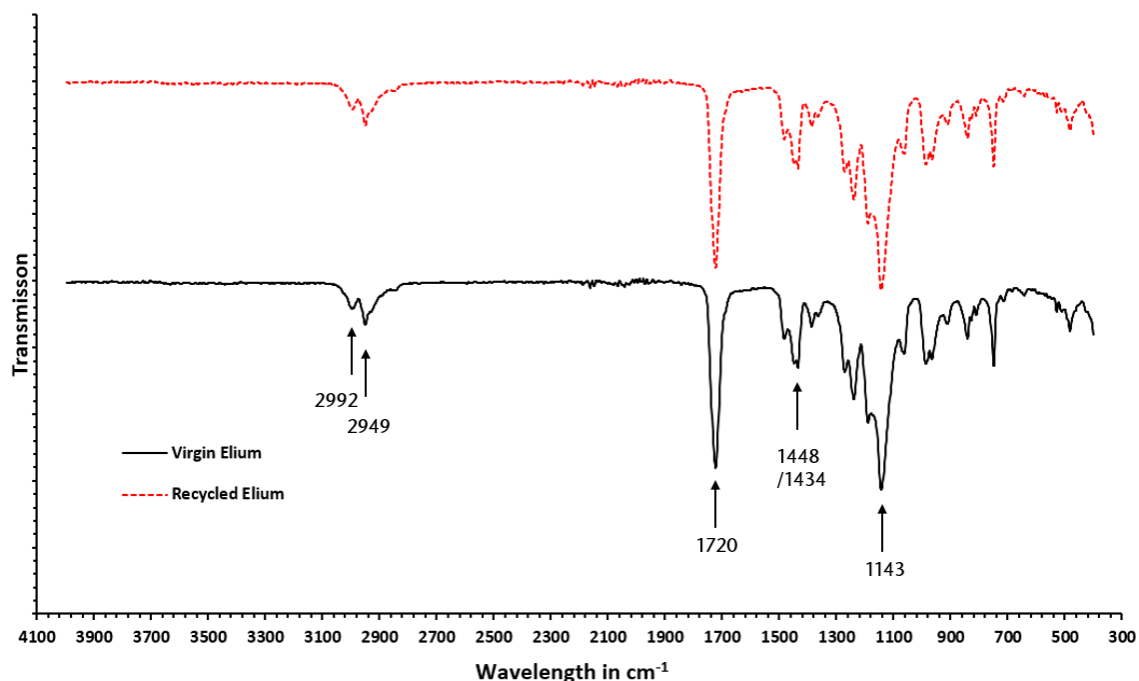


Figure 5: FTIR spectra of Elium matrix before and after recycling (normalized at 1720 cm^{-1} peak)

3.2 Thermal analysis of the recovered matrix (DSC / TGA)

Differential scanning calorimetry (DSC) was used to compare the thermal properties of the recycled Elium with its virgin counterpart and to investigate any alteration in the thermal properties of the matrix upon recycling. Since the maximum service temperature of amorphous thermoplastics such as Elium is below the glass transition temperature, the latter becomes a critical reference value [52]. As can be seen in Table 2 the mean glass transition temperature (T_g) of the recycled material is 16°C higher than that of the virgin material.

Sample name	T_g (°C)
<i>Virgin Elium</i>	98 (± 2)
<i>Recycled Elium</i>	114 (± 3)

Table 2: Mid-point glass transition temperature values before and after recycling

This increase of the glass transition temperature after recycling can be explained by a likely incomplete polymerization during the production of the composites and thus the presence of residual monomers. The polymerisation possibly continues during the dissolution and drying stage of the recycling process. Since the glass transition temperature is strongly dependent on intermolecular interactions between polymer chains and increases with molecular weight [53], a higher T_g indicates an increase in chain length/molecular weight [54] which may have been achieved during the dissolution and the drying stage.

A probable explanation for the presence of residual monomer content after composite manufacture is the increasing viscosity of the material during the in situ polymerisation of Elium after infusion. The viscosity is then too high for the growing chains to react with the remaining monomers (no diffusion possible). When Elium is dissolved, the viscosity is again significantly reduced. This makes it possible for the chains to meet monomer or reactive partners again and to be extended. Although chemically related to Elium, PMMA is usually produced on an industrial scale by dispersion polymerization [55], its production by solution polymerization is also known, even at room temperature [56]. Accordingly, it can be assumed that solution polymerization may occur when Elium is dissolved in acetone. Furthermore, the polymerisation may continue during post-treatment under vacuum (65°C for 8 h) leading to longer chain lengths and a noticeable increase in the T_g . This leads to an extension of the recycled material's service temperature to higher values.

The thermogravimetric analysis in contrast shows a slight reduction in the thermal stability of the matrix after recycling, when referring to the 95% threshold (onset of degradation, T_d) [41]. However, this reduction is noticeably lower compared to the reduction without vacuum drying of the matrix [36]. Moreover, the shoulder observed between 100°C and 200°C as a consequence of absorbed acetone during dissolution [60-62] (explained in our previous work [36]) is no longer visible. T_d , which is an important parameter for thermal

stability, is seen to reduce by 24.8°C from 215.2°C to 190.4°C upon recycling. In comparison, the reduction of T_d for non-vacuum dried matrix is 55.5°C [36]. For other thresholds used in the literature, such as the 90% wt. loss level (17.8°C less) and the 50% wt. loss level (0.5°C increase), the values of the virgin and recycled material are comparable to those of PMMA [63]. Accordingly, from these analyses it can be said that the thermal stability of the matrix is nominally influenced by the proposed recycling technique, Table 3, Fig. 6.

Sample name	95% threshold (°C)	90% threshold (°C)	50% threshold (°C)
Virgin Elium	215 (±4)	273 (±3)	334 (±3)
Recycled Elium	190 (±6)	255 (±10)	334(±7)
PMMA [63]		280	337

Table 3: TGA measurements of virgin and recycled Elium

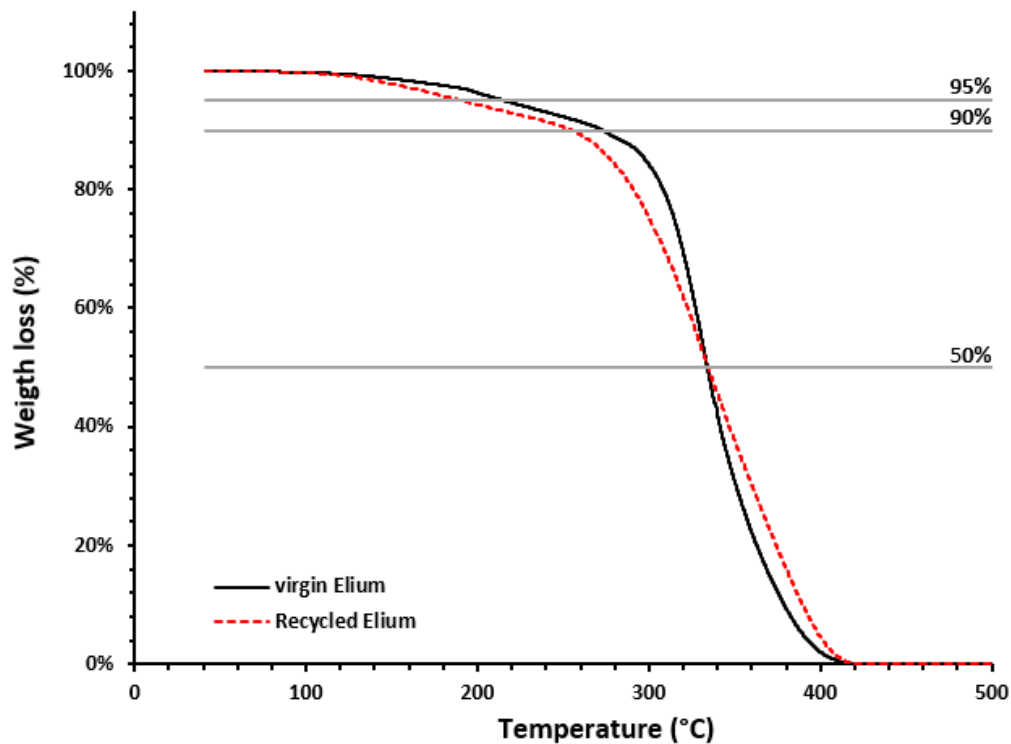


Figure 6: TGA Measurements of virgin and recycled Elium matrix

3.3 Single fibre tensile test (SFT)

Since the breaking strength of carbon fibres changes as a function of fibre length due to its small cross-sectional area (the longer the fibres, the more defects there are and higher the probability of damage leading to failure) [47], the gauge length was set at 30 mm for all single fibre tests. However, due to naturally induced different distribution of defects, a noticeable scattering

occurred even with large sample sizes. For this reason, Table 4 shows the average tensile strength of the individual fibres and the 63% Weibull distribution. The parameters required for the Weibull formula, Formula 1 (scaling parameter σ_0 and the Weibull modulus m), are calculated by linear regression using the modified Formula 2.

$$F(\sigma_f, \sigma_0, m) = 1 - \exp\left(-\frac{\sigma_f}{\sigma_0}\right)^m$$

Formula 1: Derivation Weibull analysis 1

$$\ln\left(\ln\left(\frac{1}{1-F}\right)\right) = m \ln(\sigma_f) - m \ln(\sigma_0)$$

Formula 2: Derivation Weibull analysis 2

In line with published literature dealing with tensile strength of individual carbon fibres, the point considered was 63% of all fibres tested failed according to Weibull distribution [45-47].

Sample name	Weibull 63%-value (MPa)	Ø max breaking strength (MPa)
Virgin fibres	3881	3619 (±673)
Recycled fibres	3719	3482 (±585)

Table 4: Single fibre tensile strength results

As can be seen from Table 4, the tensile strength values according to the Weibull distribution as well as the average breaking strength of the recycled fibres are very close to those of the virgin fibres. The reduction is by 4.2% from 3881 MPa to 3719 MPa (63% Weibull values) and from 3620 MPa to 3482 MPa (average values) respectively is within the uncertainty of the measurement and can therefore be neglected. It can thus be claimed that the presented recycling technique has no influence on the tensile strength and hence the structural integrity of the fibres.

3.4 Scanning electron microscopy (SEM)

The recycled fibres can be visually divided into two groups. Firstly, fibres on the outer side of the preform, Fig. 8 a-b, with very little matrix adhesion, which are sometimes difficult to distinguish from virgin fibres, Fig. 8 c. Secondly, fibres in the middle or inner side of the preform which have a noticeable layer of adhered Elium, Fig. 8 d-e. This can be related to the dissolution behaviour of polymers and the lack of stirring. When dissolving thermoplastics, a rubber-like, highly viscous layer of swollen polymer is formed [66-68], which can also be observed when dissolving pure Elium in acetone, Fig. 7. In a test setup without externally applied flow, as in this case, the dissolution process is determined only by convection flow. This in turn depends on the difference in density between the

solvent and the swollen polymer, as well as its viscosity. The viscosity of this swelling layer is largely dependent on the size and concentration of the dissolved macromolecules [67].

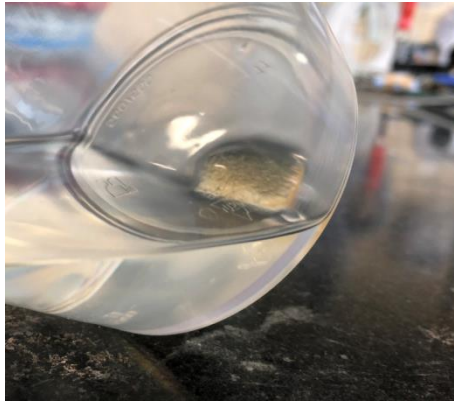


Figure 7: Rubber-like, highly viscous layer of swollen polymer at Elium

When composites are dissolved, a further factor influencing the convection flow needs to be considered, as the solvent flow is hindered by the fibres. It is a general consensus that the polymer is bound to the fibres through the sizing. Accordingly, when the matrix dissolves, the rubbery layer forms at the surface of the fibres.

As the dissolution process progresses, the matrix is dissolved in ever deeper layers of the composite. As a result, the convection flow between the inner parts of the composite and free solvent is hindered by an increasing number of fibres layers and their adherent rubbery layer. The reason for this is the adhesive forces between the rubbery layers and dissolved polymers in the convection flow.

This leads to the fact that the dissolving process proceeds much quicker on the outer sides of the preform, seen as clean surface like the virgin fibre in the SEM pictures, Fig. 8 a-b. The theory of the formation of rubber-like layers around the inner sides of the composite pre-form which do not dissolve due to the hindrance of the convection flow through the outer fibres (and their rubber-like layers) can be substantiated by the pliability tests. When the preform is removed from the solution, it can be bent without any noticeable difficulty (Fig. 2 a). After removal of acetone, considerably more force is required for bending, as if the rubber-like layers have strengthened solid and partly bonded the individual fibres together. Evidence that the Elium on the fibre is dissolved during the recycling process and solidified again when the acetone evaporates can be clearly seen in Figures 8d-f. The observed ridges, Fig. 8 d & 8 f, encircled red, were also reported by Zafeiropoulos et al. [67] after a single washing subsequent to dissolving LDPE on glass fibres by toluene. The connections between the fibres as observed in Figure 8f suggest that these preform layers were connected to the Elium matrix during the dissolution. When the fibres were removed, the minimal separation caused the attached Elium to be pulled apart

(as is commonly observed during crazing in thermoplastics), before the acetone evaporated and the matrix solidified.

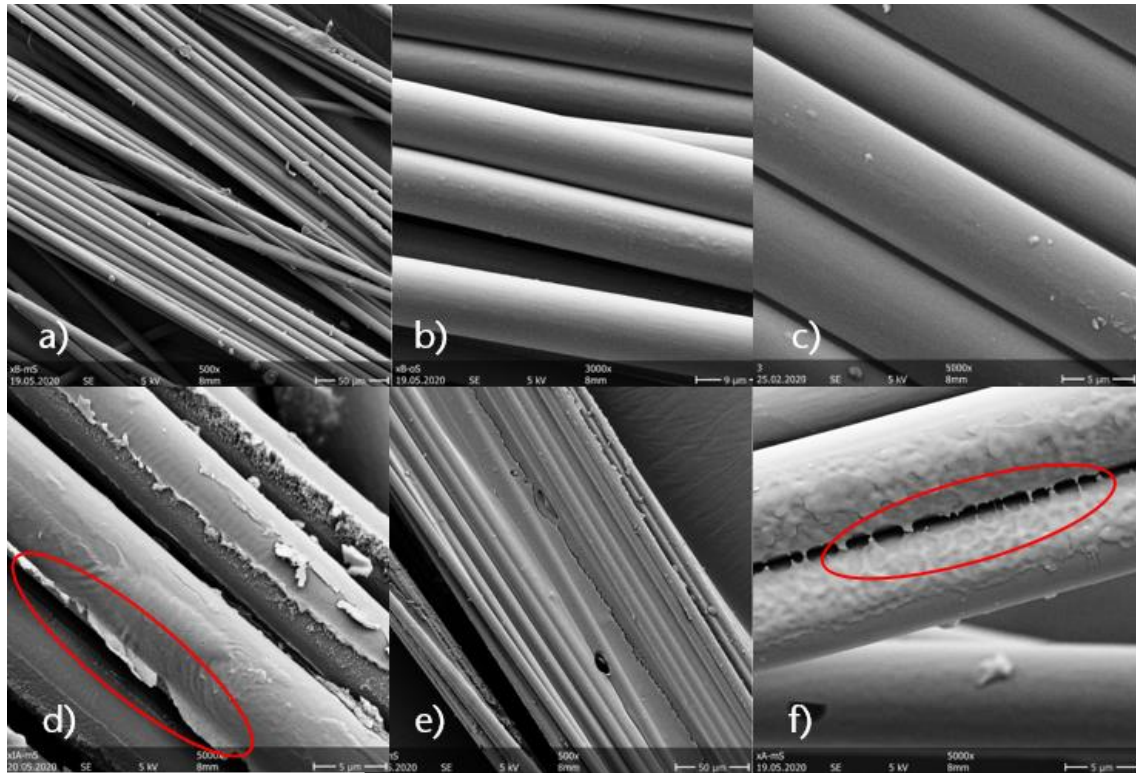


Figure 8: SEM pictures of a-b) recycled fibres at the outer side of the preform c) virgin carbon fibres d-f) recycled fibres at the inner side of the preform showing Elium has been dissolved and is solidified again (encircled red)

As shown in Figure 9, a thin layer is always observed on the surface after dissolution.

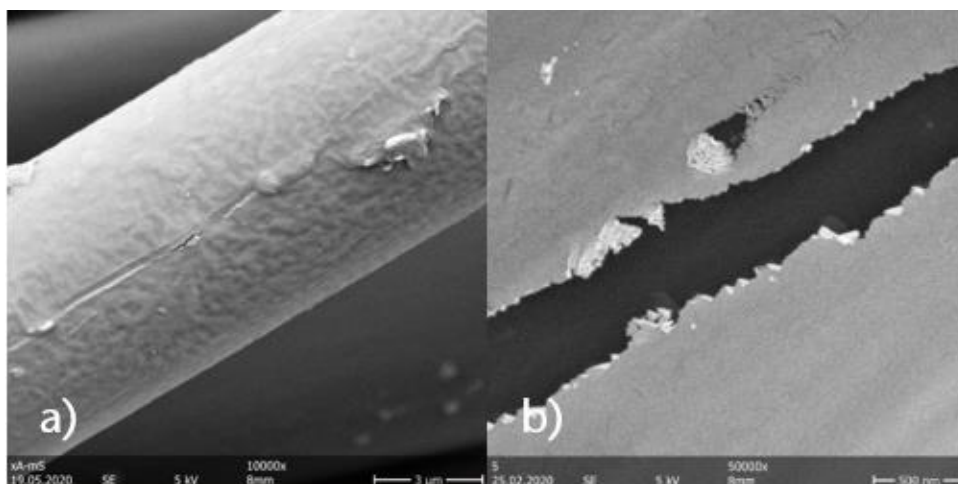


Figure 9: SEM pictures of carbon fibres with surface coating after dissolution

Since for the chemical bonding of the matrix to the fibres the presence of the sizing is essential [70-72], EDX was used to assess whether sizing signatures can be detected in the remaining layer. The layer can consist either of a thin adherent layer of Elium without sizing or, as in the case of supercritical solvolysis by Dauget et al. [73], the original sizing. The EDX measurements revealed that the fibre surface always contains a low atomic percentage of silicon (Si), which indicates the presence of the sizing [71] (Fig. 10, Table 5). This is a decisive advantage over recycling by pyrolysis or solvolysis, which generally strips away the sizing leaving bare fibres after recycling [1]. This retained layer, on the other hand, can be regarded as particularly beneficial if an Elium matrix is used to process composites with these recycled and reclaimed carbon fibre preforms.

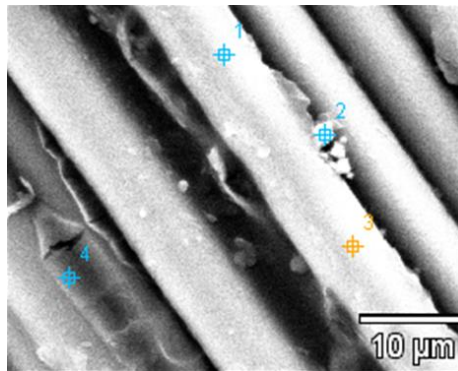


Figure 10: EDX measuring points

Measuring point	C (%)	O (%)	Si (%)	Pt (%)
PT1	49.4	39.8	10.5	0.3
PT2	61.0	28.8	9.9	0.3
PT3	52.3	34.8	12.4	0.5
PT4	73.3	22.1	4.3	0.2

Table 5: Elemental composition in atomic% from the EDX measurements

The presence of both sizing and Elium on the fibres could be potentially linked to the mechanical responses observed by DMA and ILSS and has been discussed in the following sections.

3.5 Dynamic mechanical analysis (DMA)

The 2nd generation composites produced with virgin Elium and recycled fibres were tested by DMA for their thermomechanical properties from which information about fibre-matrix adhesion can be derived [74, 75]. Gerard et al. [76], reported that the glass transition temperature and the storage modulus can be good indicators for fibre-matrix adhesion. The $T_{g(\text{Onset})}$ (+4.7°C) and $T_{g(\text{tan}\delta \text{max})}$ (+3.1°C) were observed to increase slightly in comparison to the 1st

generation composites, Fig. 11,12, Table 6. This could be potentially attributed to post-polymerisation of the reclaimed matrix during vacuum drying (8 h, 65°C) to remove residual acetone (8% residual Elium w.r.t. fibres in the preform). However, it should be noted that the increase in T_g is marginal and therefore the thermal stability of the recycled composites can be considered comparable to their virgin counterparts.

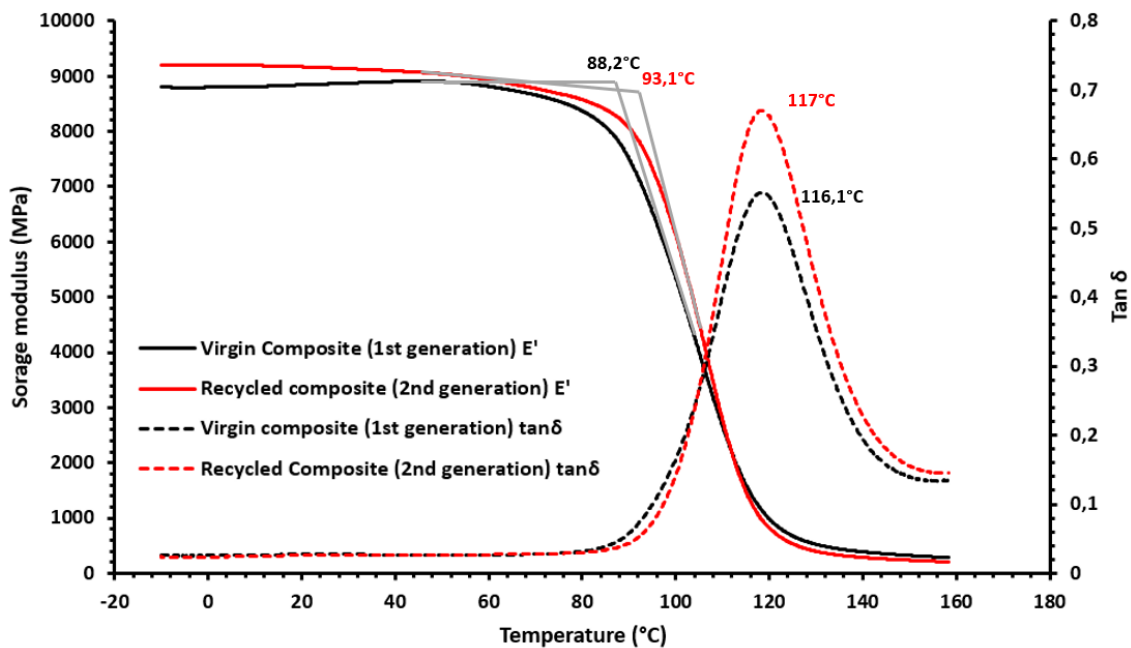


Figure 11: Storage modulus and $\tan\delta$ of typical DMA curves for 1st and 2nd generation composites

Sample name	$\varnothing T_{g(Onset)} (^\circ\text{C})$	$\varnothing T_{g(\tan(\delta)_{max})} (^\circ\text{C})$	Stor. mod. (MPa)
1 st Generation	87.7 (± 3.9)	116.0 (± 5.0)	8952 (± 969)
2 nd Generation	92.4 (± 2.7)	119.2 (± 2.4)	8210 (± 763)

Table 6: Glass transition temperatures by DMA for first and second generation composites

However, despite the signature of sizing remnants, the storage modulus (E') at room temperature (25°C) is seen to reduce by 8.2% from 8952 MPa to 8210 MPa indicating a slight degradation in interfacial adhesion. In addition, an increase in the $\tan\delta$ peak is observed, which indicates an increase in damping and is generally interpreted as a degradation of interfacial properties as well [75, 77]. Nevertheless, the reduction in thermomechanical properties is still relatively small and can be considered as acceptable, given the background that recycled fibres that are used.

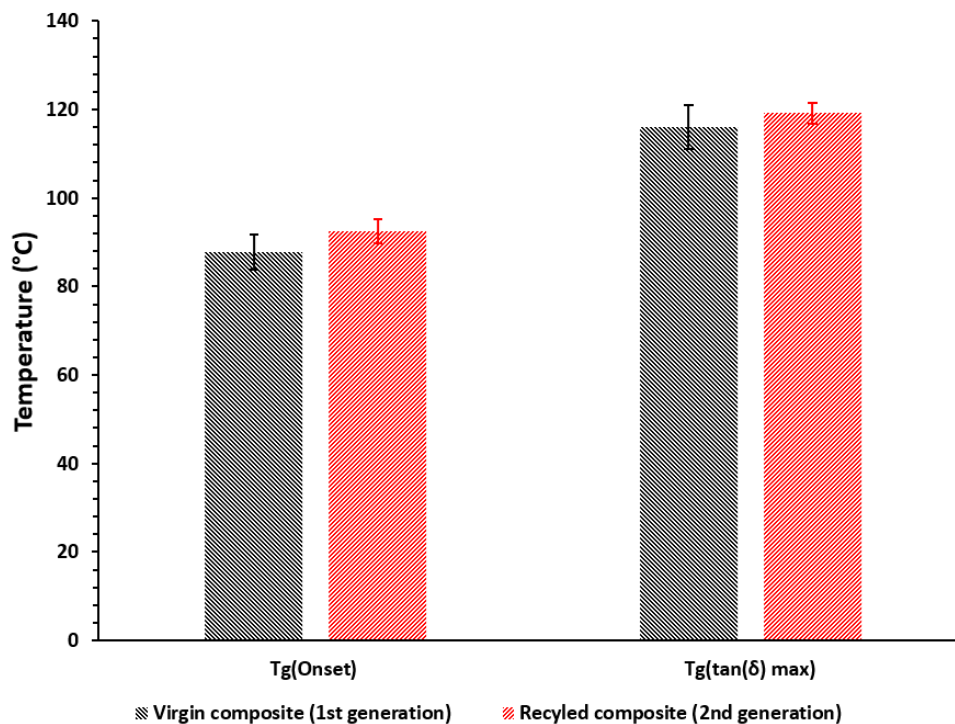


Figure 12: Glass transition temperatures by DMA for first and second generation composites

In conclusion, it can be considered that the thermomechanical properties of the recycled material detected on the basis of the DMA data are relatively near to the properties of the virgin material.

3.6 Interlaminar shear strength (ILSS)

Since the mechanical properties of composites are strongly dependent on the fibre-matrix interfacial interaction through the sizing which influences load transfer between the fibres and the matrix [78,79], ILSS testing was used to probe the fibre-matrix interface. According to Singh et al. [80], crack initiation and propagation under bending stress in composites occur mainly along the interlaminar surface, which in turn leads to delamination and failure. Especially in the case of reused fibres, the shear properties can be especially affected upon recycling [17].

From Table 7, it is observed that the ILSS results do not reflect the slight degradation observed earlier from the DMA measurements. The averaged shear strength is practically unchanged with the recycled composites at 31.4 GPa is 99.8% of the virgin composite with 31.5 GPa, Table 7. The scatter in the data in both cases is relatively high, but that is common for ILSS measurements [81, 82].

Sample name	∅ Shear stress at break (GPa)
1 st Generation	31.5 (±5.3)
2 nd Generation	31.4 (±5.9)

Table 7: ILSS results of the Elium CFRPs

Thus, it can be concluded that the proposed recycling has no noticeable influence on the fibre-matrix adhesion which can be explained by the viscoelastic behaviour of thermoplastics. In contrast to thermoset systems, the thermoplastic matrix is not cross-linked which means that the chains shear/slide against each other during crack propagation. However, the interactions between the chains are still present, leading to plastic deformation and a reduction in crack propagation [24]. Bhudolia et al. [31] observed during shear strength tests of Elium composites that the fibre-matrix adhesion is strong which usually leads to matrix-dominated failure under shear. Since in the present situation there is still sizing remaining on the recycled fibres (for further explanations please refer to Section 3.4), it can be assumed that sufficient fibre-matrix adhesion can be achieved even in the composites processed with recycled fibres and fresh Elium. In areas where Elium remnants are still present on the fibres, the solubility of polymerized Elium in Elium monomers can also lead to a very good bond between the old and virgin matrix.

3.7 Three-point bending (3PB)

The results of the three-point bending tests, Table 8, complement the assertion of a high fibre-matrix adhesion deduced from the ILSS (please refer to Section 3.6). Macroscopically, none of the samples tested showed interfacial failure. Both 1st and the 2nd generation specimens failed by compression on the upper side, as has been reported earlier [25]. However, the modulus seemed to decrease by 12.2% and the flexural strength by 27.3%, Figure 13, Table 8.

Sample name	∅ Bending modulus (GPa)	∅ Flexural strength (MPa)
1 st Generation	12.5 (±2.5)	480 (±48)
2 nd Generation	10.9 (±1.3)	349 (±50)

Table 8: 3PB results of the Elium CFRPs

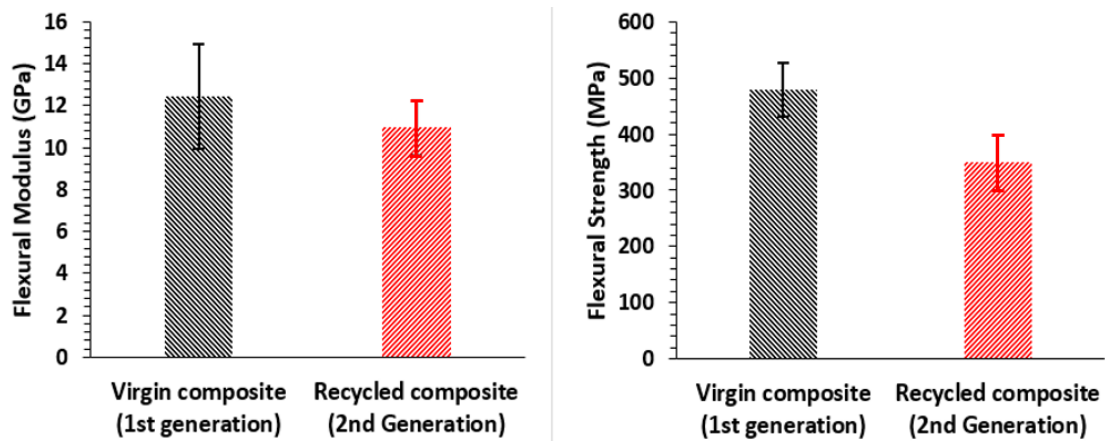


Figure 13: 3PB results for 1st and 2nd generation composites

The reduction in the flexural properties of the 2nd generation composites after recycling can be related to the observed compressive failure. As shown in Figure 14, during the infusion of the recycled fibres, small air bubbles were partially formed on the top ply of the laminate (in the direction of the infusion). A possible explanation is the higher infusion rate when using recycled fibres. The infusion time has been observed to reduce by about 40%. This may be caused by a lower compression of the preform due to the Elium remnants on the preform. Unfortunately, despite the change in the directional media length as proposed by Bhulodia et al [23], it has to date proved impossible to avoid this issue (which is currently being investigated for future reports). Since the 3PB samples are machined at 90° to the direction of infusion, the air inclusions are located transversely in the top ply of the specimen. This results to less matrix on the top of the specimen which fails under pressure at this point during testing possibly resulting in reduced flexural properties of these materials as observed.



Figure 14: Bubbles on the top ply of the 2nd generation composite (bubbles encircled red)

4 CONCLUSION

This work presents a recycling strategy that enables reclamation of undamaged fibres in the form of scrims and reuse them almost completely in a similar form and quality. In addition, it is possible to recover 81% of the matrix with properties quite near to the virgin material.

According to FTIR analysis, there is no significant change in the chemical structure of the matrix material due to recycling. Furthermore, no decrease in the upper application temperature of the recycled matrix could be observed by means of DSC measurements (T_g increases by 16°C). TGA measurements revealed a certain reduction of the degradation temperature (T_d decreases by 24.8°C); but overall, the properties of the recycled matrix were found to be quite near to the level of the virgin matrix properties.

The tests carried out showed that reuse of the recycled fibres in the form of scrims is feasible. The suggested presence of the sizing on the fibres and retaining the preform structure are the main advantages of this recycling method. The recycled fibres, as has been demonstrated here, could be processed quite similar to the virgin ones using VARI. SEM/EDX observations suggested that sizing remains on the fibres which results in adequate fibre-matrix adhesion and only a minor degradation of the interlaminar properties. The interlaminar shear strength according to the ILSS measurements was almost identical in both generation of the composites (negligible 0.2% reduction for the 2nd generation composites). This being said, a noticeable reduction in the flexural performance of the composite with recycled fibres was observed (bending modulus decreased by 12.2%). This was mainly attributed to small bubbles observed in the 2nd generation composites during processing.

Based on the recycling process presented in this paper, a nearly quantitative recovery of the fibres is feasible. The recovered fibres can be subsequently re-used for identical applications with comparable performance to pristine systems. Therefore, this work can serve as proof of concept of a Closed-Loop Recycling approach for infusible thermoplastic composites.

Acknowledgement

The authors would like to thank Prof. Jörg Adams from the Institute of Physical Chemistry at Clausthal University of Technology for carrying out the FTIR measurements. We also thank Mrs. Peggy Knospe from the Institute of Mechanical Process Engineering for the SEM and EDX measurements.

Literature

- [1] Oliveaux, G.; Dandy, L.O.; Leeke, G.A. Current status of recycling of fibre reinforced polymers: Review of technologies, reuse and resulting Properties
Progress in Materials Science 72 (2015) p. 61-99
DOI: <https://doi.org/10.1016/j.pmatsci.2015.01.004>
- [2] Chen, J.; Wang, J.; Ni, A. Recycling and reuse of composite materials for wind turbine blades: An overview
Journal of Reinforced Plastics and composites 28 (2019) p. 567-577
DOI: <https://doi.org/10.1177/0731684419833470>
- [3] Beauson, J.; Lilholt, H.; Brøndsted, P. Recycling solid residues recovered from glass fibre-reinforced composites – A review applied to wind turbine blade materials
Journal of Reinforced Plastics and Composites 33 (2014) p. 1542-1556
DOI: <https://doi.org/10.1177/0731684414537131>
- [4] Ribeiroa, M.C.S.; Meira-Castrob, A.C.; Silvab, F.G.; et al. Re-use assessment of thermoset composite wastes as aggregate and filler replacement for concrete-polymer composite materials: a case study regarding GFRP pultrusion wastes
Resources, Conservation & Recycling 104 (2015) p. 417-426
DOI: <https://doi.org/10.1016/j.resconrec.2013.10.001>
- [5] Ogi, K.; Shinoda, T.; Mizui, M. Strength in concrete reinforced with recycled CFRP pieces
Composites Part A: Applied Science and Manufacturing 36 (2005) 7, p. 893-902
DOI: <https://doi.org/10.1016/j.compositesa.2004.12.009>
- [6] Correia, J.; Almeida, N.; Figueira, J. Recycling of FRP composites: reusing fine GFRP waste in concrete mixtures
Journal of Cleaner Production 19 (2011) 15, p. 1745-1753
DOI: [10.1016/j.jclepro.2011.05.018](https://doi.org/10.1016/j.jclepro.2011.05.018)

- [7] Palmer, J.; Ghita, L.; Savage, L.; et al. Successful closed-loop recycling of thermoset composites
Composites Part A: Applied Science and Manufacturing 40 (2009) 4, p. 490-498
DOI: <https://doi.org/10.1016/j.compositesa.2009.02.002>
- [8] Beauson, J.; Madsen, B.; Toncelli, C.; et al. Recycling of shredded composites from wind turbine blades in new thermoset polymer composites
Composites Part A: Applied Science and Manufacturing 90 (2016) p. 390-399
DOI: <https://doi.org/10.1016/j.compositesa.2016.07.009>
- [9] Pickering, S.J.; Recycling technologies for thermoset composite materials – current status
Composites Part A: Applied Science and Manufacturing 37 (2006) 8, p. 1206-1215
DOI: [10.1016/j.compositesa.2005.05.030](https://doi.org/10.1016/j.compositesa.2005.05.030)
- [10] N.N. ELG Carbon Fibre Ltd. website
<http://www.elgcf.com/de/about/what-we-do> (2020)
- [11] N.N. CFK Valley Stade Recycling GmbH & Co. KG website
<https://www.cfk-recycling.de/index.php?id=6> (2020)
- [12] Meng, F.; McKechnie, J.; Turner, T.; et al. Environmental aspects of use of recycled carbon fibre composites in automotive applications
Environmental Science & Technology 51 (2017) p. 12727-12736
DOI: <https://doi.org/10.1021/acs.est.7b04069>
- [13] N.N. Launching the carbon fibre recycling industry
Reinforced plastics (2010), p. 33-37
- [14] Limburg, M.; Stockschröder, J.; Quicker, P. Thermal treatment of carbon fibre reinforced polymers
Waste Management & Research 37 (2019), p. 73-82
DOI: [10.1177/0734242X18820251](https://doi.org/10.1177/0734242X18820251)

- [15] Xu, P.;
Li, J.;
Ding, J.; Chemical recycling of carbon fibre/ epoxy components
in a mixed solution of peroxide hydrogen and N,N-
dimethyl formamide

Composites Science and Technology 82 (2013), p.
54-59

DOI:
<http://dx.doi.org/10.1016/j.compstech.2013.04.002>
- [16] Oliveux, G.;
Bailleul, J.;
Gillet, A.;
et al. Recovery and reuse of discontinuous carbon fibres by
solvolysis: Realignment and properties of
remanufactured materials

Composites Science and Technology 139 (2017), p.
99-108

DOI:
<http://dx.doi.org/10.1016/j.compstech.2016.11.001>
- [17] Knight, C.C.;
Zeng, C.;
Zhang, C.;
et al. Fabrication and properties of composites utilizing
reclaimed woven carbon fiber by sub-critical and
supercritical water recycling

Materials Chemistry and Physics 149-150 (2015) p.
317-323

DOI:
<http://dx.doi.org/10.1016/j.matchemphys.2014.10.023>
- [18] N.N. Arkema ELIUM Webpage

<https://www.arkema.com/en/products/product-finder/range-viewer/Elium-resins-for-composites/>
(2020)
- [19] N.N. Vacuum Infusion of ELIUM® Resin Process Guide to
Vacuum Infuse ELIUM® Thermoplastic Composite
Parts

ARKEMA (2018)
- [20] N.N. Material Safety Data Sheet ELIUM

ARKEMA (2014)
- [21] Chilali, A.;
Zouari, W.;
Assarar, M.;
et al. Analysis of the mechanical behaviour of flax and glass
fabrics-reinforced thermoplastics and thermoset
resins

Journal of Reinforced Plastics and Components 35
(2019) 16, p. 1217-1232

DOI: 10.1177/0731684416645203

- [22] N.N. Combining the Best of two polymer worlds
Kunststoffe International (2016) p. 49-51
- [23] Bhudolia, S.K.;
Perrotey, P.P.;
Joshi, S.C. Optimizing Polymer Infusion Process for Thin Ply
Textile Composites with Novel Matrix System
MDPI Materials 10 (2016) 293 p. 1-19
DOI: 10.3390/ma10030293
- [24] Barbosa, J.;
Bortoluzzi, B.;
Toncelli, C.;
et al. Analysis of fracture toughness in mode II and
fractographic study of composites based on Elium®
150 thermoplastic matrix
Composites Part B 175 (2019) 107082 p. 1-10
DOI:
<https://doi.org/10.1016/j.compositesb.2019.107082>
- [25] Bhudolia, S.K.;
Joshi, S.C.;
Bert, A.;
et al. Flexural characteristics of novel carbon
methylmethacrylate composites
Composites Communications 13 (2019) p. 129-133
DOI: <https://doi.org/10.1016/j.coco.2019.04.007>
- [26] Bhudolia, S.K.;
Perrotey, P.;
Joshi, S.C.;
et al. Enhanced vibration damping and dynamic mechanical
characteristics of composites with novel pseudo-
thermoset matrix system
Composite Structures 179 (2017) p. 502-513
DOI:
<http://dx.doi.org/10.1016/j.compstruct.2017.07.093>
- [27] Obande, W.;
Ray, D.;
Ó Brádaigh, C.; Viscoelastic and drop-weight impact properties of an
acrylic-matrix composite and a conventional
thermoset composite – A comparative study
Materials Letters 238 (2019) p. 38-41
DOI: <https://doi.org/10.1016/j.matlet.2018.11.137>
- [28] Kinvi-Dossou,
G.;
Matadi
Boumbimba, R.;
Bonfoh, N.;
et al. Innovative acrylic thermoplastic composites versus
conventional composites: Improving the impact
performances
Composite Structures 217 (2019) p. 1-13
DOI: <https://doi.org/10.1016/j.compstruct.2019.02.090>

- [29] Bhudolia, S.K.; Joshi, S.C. Low-velocity impact response of carbon fibre composites with novel liquid Methylmethacrylate thermoplastic matrix
Composite Structures 203 (2018) p. 696-708
DOI: <https://doi.org/10.1016/j.compstruct.2018.07.066>
- [30] Kazemia, M.E.; Shanmugama, L.; Lub, D.; et al. Mechanical properties and failure modes of hybrid fiber reinforced polymer composites with a novel liquid thermoplastic resin, Elium®
Composites Part A 125 (2019) 105523
DOI: <https://doi.org/10.1016/j.compositesa.2019.105523>
- [31] Bhudolia, S.K.; Perrotey, P.; Joshi, S.C.; Mode I fracture toughness and fractographic investigation of carbon fibre composites with liquid Methylmethacrylate thermoplastic matrix
Composites Part B 134 (2018) p. 246-253
DOI: <https://doi.org/10.1016/j.compositesb.2017.09.057>
- [32] Cousins, D.S.; Suzuki, Y.; Murray, R.E.; et al. Recycling glass fiber thermoplastic composites from wind turbine blades
Journal of Cleaner Production 209 (2019) p. 1252-1263
DOI: <https://doi.org/10.1016/j.jclepro.2018.10.286>
- [33] N.N. Trichlormethan Datenblatt
Institut für Arbeitsschutz der Gesetzlichen Unfallversicherung GESTIS-Stoffdatenbank (2020)
- [34] Alder, C.M.; Hayler, J.D.; Henderson, R.K.; et al. Updating and further expanding GSK's solvent sustainability guide
Green Chemistry (2016) 18, p. 3879-3890
DOI: 10.1039/c6gc00611f
- [35] N.N. Electron Beam Lithography in Nanoscale Fabrication: Recent Development
IEEE Transactions on electronics packaging manufacturing 26 (2003) p. 141–149
DOI: 10.1109/TEPM.2003.817714

- [36] Gebhardt, M.; Chakraborty, S.; Manolakis, I.; et al. Recycling of CFRP composites based on a thermoplastic matrix (ELIUM 150) which can be infused and cured at room temperature – Recovery of the matrix by dissolution processes and testing of fibre damage by means of SEM and single fibre tensile tests
Forschungsfeld Rohstoffsicherung und Ressourceneffizienz, Herausgeber: Oliver Langefeld, Asja Mrotzek-Blöß, Papierflieger Verlag GmbH, Clausthal-Zellerfeld, 2020.
- [37] N.N. DIN EN ISO 11357-1: Kunststoffe – Dynamische Differenz-Thermoanalyse (DSC) Teil 1: Allgemeine Grundlagen. (2008)
- [38] N.N. DIN EN ISO 11358-1: Kunststoffe – Thermogravimetrie (TG) von Polymeren Teil 1: Allgemeine Grundsätze. (2008)
- [39] Ehrenstein, G.W.; Riedel, G.; Trawiel, P. Praxis der Thermischen Analyse von Kunststoffen 2. Auflage
Carl Hanser Verlag, München, 2003
- [40] Schawe, J.; Riesen, R.; Widmann, J.; et al. DSC-Kurven interpretieren Teil 1: Dynamische Messungen
UserCom (2000)
- [41] Gornicka, B.; Gorecki, L. TGA/DTG/DSC investigation of thermal ageing effects on polyamide–imide enamel
Journal of Thermal Analysis and Calorimetry 101 (2010) p.647-650
DOI: 10.1007/s10973-010-0883-9
- [42] N.N. DIN EN ISO 6721-1:2019-09 Kunststoffe - Bestimmung dynamisch-mechanischer Eigenschaften - Teil 1: Allgemeine Grundlagen (2019)
- [43] N.N. DIN EN ISO 14130 Faserverstärkte Kunststoffe - Bestimmung der scheinbaren interlaminaren Scherfestigkeit nach dem Dreipunktverfahren mit kurzem Balken (1998)
- [44] N.N. ISO 11566 Kohlenstofffasern - Bestimmung der Zugeigenschaften von Probekörpern aus Einzelfilamenten (1996)

- [45] Jiang, G.; Pickering, S.J.; Lester, E.H.; et al. Characterisation of carbon fibres recycled from carbon fibre/epoxy resin composites using supercritical n-propanol
Composites Science and Technology 69 (2009) 2, p. 192-198
DOI: 10.1016/j.compscitech.2008.10.007
- [46] Morenton, R.; Watt, W.; Johnson, W.; Carbon Fibres of High Strength and High Breaking Strain
Nature (1967) p. 690-691
- [47] Paiva, M.C.; Bernardo, C.A.; Nardin, M.; Mechanical, surface and interfacial characterisation of pitch and PAN-based carbon fibres
Carbon 38 (2000) p. 1323-1337
DOI: 10.1016/S0008-6223(99)00266-3
- [48] Weibull, W. A Statistical Distribution Function of wide applicability
Journal of applied mechanics (1951) p. 293-299
- [49] Hesse, M.; Meier, H.; Zeeh, B.; Spektroskopische Methoden in der organischen Chemie 8. Auflage
Thieme Georg Verlag, Stuttgart, 2012
- [50] Duan, G.; Zhang, C.; Li, A.; et al. Preparation and Characterization of Mesoporous Zirconia Madeby Using a Poly (methyl methacrylate) Template
Nanoscale Research Letters 3 (2008) p. 118-122
DOI: 10.1007/s11671-008-9123-7
- [51] Ramesh, S.; Leen, K.H.; Kumutha, K.; et al. FTIR studies of PVC/PMMA blend based polymer electrolytes
Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy 66 (2016) 4-5, p. 1237-1242
DOI: 10.1016/j.saa.2006.06.012
- [52] Bonnet, M. Kunststoffe in der Ingenieur Anwendung: verstehen und zuverlässig auswählen
Vieweg+Teubner Verlag, 2009
- [53] Menges, G.; Haberstroh, E.; Michaeli, W.; et al. Menges Werkstoffkunde Kunststoffe 6. Auflage
Carl Hanser Verlag, 2011

- [54] O'Driscoll, K.; Sanayei, R.A.; Chain-Length Dependence of the Glass Transition Temperature
Macromolecules 24 (1991) p. 4479-4480
DOI: 10.1021/ma00015a038
- [55] Roos, E.; Maile, K.; Seidenfuß, M.; Werkstoffkunde für Ingenieure Grundlagen, Anwendung, Prüfung 6. Auflage
Springer Vieweg (2017)
- [56] Lyoo, W.S.; Noh, S.K.; Kang, G.C.; et al.; Preparation of High Molecular Weight Poly(methyl methacrylate) with High Yield by Room Temperature Suspension Polymerization of Methyl Methacrylate
Fibers and Polymers 5 (2004) 1, p. 75-81
DOI: <https://doi.org/10.1007/BF02875498>
- [57] Tabtiang, J.; Lumlong, B.; Venables, C.; The influence of preparation method upon the structure and relaxation characteristics of poly(methyl methacrylate) / clay composites
European Polymer Journal 36 (2000) 12, p. 2559-2568
DOI: [https://doi.org/10.1016/S0014-3057\(00\)00061-6](https://doi.org/10.1016/S0014-3057(00)00061-6)
- [58] Brockhaus, A.; Jenckel, E.; Über die Kinetik des thermischen Abbaues von Polymethacrylsäuremethylester
Institut für theoretische Hüttenkunde und physikalische Chemie der Technischen Hochschule Aachen (1955) p. 1-32
- [59] Holland, B.J.; Hay, J.N.; The effect of polymerisation conditions on the kinetics and mechanisms of thermal degradation of PMMA
Polymer Degradation and Stability 77 (2002) p. 435-439
DOI: 10.1016/S0141-3910(02)00100-3
- [60] Loos, M.R.; Coelho, L.A.F.; Pezzin, S.H.; The effect of acetone addition on the properties of epoxy
Polímeros: Ciência e Tecnologia 18 (2008) p. 76-80
DOI: <https://doi.org/10.1590/S0104-14282008000100015>
- [61] Sharmin, E.; Imo, L.; Ashraf, S.M.; et al.; Acrylic-melamine modified DGEBA-epoxy coatings and their anticorrosive behaviour
Progress in Organic Coatings 50 (2003) p. 47-54
DOI: 10.1016/j.porgcoat.2003.10.003

- [62] Ehrenstein, G.W.; Riedel, G.; Trawiel, P. Praxis der Thermischen Analyse von Kunststoffen 2. Auflage
Carl Hanser Verlag, München, 2003
- [63] Van Krevelen, D.W.; Nijenhuis, K. Properties of Polymers, 3rd edition
Elsevier, 1997
- [64] Gouli, S.; Poulakis, J.G.; Papaspyrides, C.D. Solvent Recycling of Poly(methyl Methacrylate) Decorative Sheets
Advances in Polymer Technology 13 (1994) 3, p. 207-211
DOI: <https://doi.org/10.1002/adv.1994.060130303>
- [65] Arostegui, A.; Sarrionandi, M.; Aurrekoetxe, J.; et al. Effect of dissolution-based recycling on the degradation and the mechanical properties of acrylonitrile butadiene styrene copolymer
Polymer Degradation and Stability 91 (2006) p. 2768-2774
DOI: [10.1016/j.polymdegradstab.2006.03.019](https://doi.org/10.1016/j.polymdegradstab.2006.03.019)
- [66] Evchuk, I.Y.; Musii, R.I.; Makitra, R.G.; et al. Solubility of Polymethyl Methacrylate in Organic Solvents
Russian Journal of Applied Chemistry 78 (2005) p. 1576-1580
DOI: <https://doi.org/10.1007/s11167-005-0564-9>
- [67] Ueberreiter, K.; Asmussen, F. Die Auflösungsgeschwindigkeit von Polymeren 1. Formulierung des Vorganges und seine Temperaturabhängigkeit
Macromolecular Chemistry and Physics (1961) p. 1-14
DOI: [10.1002/macp.1961.020440128](https://doi.org/10.1002/macp.1961.020440128)
- [68] Bristow, G.M.; Watson, W.F. Cohesive energy densities of polymers Part 2.- Cohesive energy densities from viscosity measurements
Transactions of the Faraday Society Vol. 54, p.1742-1747
DOI: <https://doi.org/10.1039/TF9585401742>
- [69] Zafeiropoulos, N.E.; Varelidis, P.C.; Papaspyrides, C.D.; et al. Characterisation of LDPE residual matrix deposited on glass fibres by a dissolution/reprecipitation recycling process
Composites: Part A 30 (1999) p. 831–838
DOI: [https://doi.org/10.1016/S1359-835X\(98\)00198-5](https://doi.org/10.1016/S1359-835X(98)00198-5)

- [70] Greisel, M.; Horn, S. MAI Polymer - Zentrum für Faser-Matrix-Anbindung: Anwendungsangepasste thermoplastische Matrix/Schlichte-Systeme : Abschlussbericht (2016)
- [71] Wetjen, D.; Wechselwirkung von Carbonfasern, Schlichte und epoxidbasierter polymerer Matrix in carbonfaserverstärkten Kunststoffen
Dissertation Universität Augsburg, 2016
- [72] Jancar, J. Review of the role of the interphase in the control of composite performance on micro- and nano-length scales
Journal of Materials Science 43 (2008) p. 6747–6757
DOI: <https://doi.org/10.1007/s10853-008-2692-0>
- [73] Dauguet, M.; Mantaux, O.; Nicolas, N.; et al. Recycling of CFRP for high value applications: Effect of sizing removal and environmental analysis of the SuperCritical Fluid Solvolysis
Procedia CIRP 29 (2015) p. 734-739
DOI: <https://doi.org/10.1016/j.procir.2015.02.064>
- [74] Mulligan, D.R.; Gnaniah, S.J.P.; Sims, D.G. Characterisation of the Fibre-Matrix Interface by Thermal Analysis
NPL Measurement Note CMMT(MN)060, National Physical Laboratory, Queens Road, Teddington, Middlesex, United Kingdom, 2000
- [75] Afaghi-Khatibi, A.; Mai, Y. Characterisation of fibre/matrix interfacial degradation under cyclic fatigue loading using dynamic mechanical analysis
Composites: Part A 33 (2002) p. 1585–1592
DOI: [https://doi.org/10.1016/S1359-835X\(02\)00117-3](https://doi.org/10.1016/S1359-835X(02)00117-3)
- [76] Gérard, J.F.; Perret, P.; Chabert, B. Study of Carbon/Epoxy Interface (or Interphase): Effect of Surface Treatment of Carbon Fibers on the Dynamic Mechanical Behavior of Carbon/Epoxy Unidirectional Composites
Controlled Interphases in Composite Materials (1990) p. 449-456
DOI: https://doi.org/10.1007/978-94-011-7816-7_44
- [77] Gérard, J.F.; Perret, P.; Chabert, B. Viscoelastic study of carbon/epoxy unidirectional composite materials
Makromolekulare Chemie. Macromolecular Symposia (1989) p. 139-147
DOI: <https://doi.org/10.1002/masy.19890230111>

- [78] Macherauch, E.; Zoch, H.W. *Praktikum in Werkstoffkunde*
Springer Vieweg, 2014
DOI: 10.1007/978-3-658-05038-2
- [79] Mallick, J. *Fiber-reinforced composites: materials, manufacturing, and design 3rd edition*
CRC Press, 2007
DOI: 10.1201/9781420005981
- [80] Singh, K.K.; Rawat, P.; Rai, A.K. *Mechanical characterization of GFRP laminate reinforced with short carbon fiber fillers under ILSS test and 3-point bend test*
ARPN Journal of Engineering and Applied Sciences 11 (2016) p. 10091-10096
- [81] Zahid, S.; Nasir, M.A.; Nauman, S.; et al. *Experimental analysis of ILSS of glass fibre reinforced thermoplastic and thermoset textile composites enhanced with multiwalled carbon nanotubes*
Journal of Mechanical Science and Technology 33 (2019) p. 197-204
DOI: 10.1007/s12206-018-1219-0
- [82] Rahman, M.M.; Zainuddin, S.; Hosur, M.V.; et al. *Effect of NH₂-MWCNTs on crosslink density of epoxy matrix and ILSS properties of e-glass/epoxy composites*
Composite Structures 95 (2013) p. 213–221
DOI:
<http://dx.doi.org/10.1016/j.compstruct.2012.07.019>

Bibliography

DOI 10.3139/O999.02052020
Zeitschrift Kunststofftechnik / Journal of Plastics
Technology 16 (2020) 5; page 179–210
© Carl Hanser Verlag GmbH & Co. KG
ISSN 1864 – 2217

Stichworte:

CFK Recycling, 2. Generation Composites, Closed-Loop Recycling, Thermomechanische Eigenschaften

Keywords:

CFRP Recycling, 2nd generation composites, Closed-Loop Recycling, Thermomechanical properties

Autor / author:

Magnus Gebhardt (1. Autor)
Dr. Souvik Chakraborty (2. Autor)
Prof. Dr. Dieter Meiners (4. Autor)
Institut für Polymerwerkstoffe und Kunststofftechnik
TU Clausthal
Agricolastraße 6
38678 Clausthal-Zellerfeld

E-Mail: magnus.gebhardt@gmx.net
Tel.: +49 (0)176/28571842

Dr. Ioannis Manolakis (3. Autor)
Department of Life Sciences
School of Science
Institute of Technology Sligo
Ash Lane, Sligo, Ireland

E-Mail: Manolakis.ioannis@itsligo.ie
Tel.: +353 (0)71 9305427

Herausgeber / Editors:

Editor-in-Chief
Prof. em. Dr.-Ing. Dr. h.c. Gottfried W. Ehrenstein
Lehrstuhl für Kunststofftechnik
Universität Erlangen-Nürnberg
Am Weichselgarten 9
91058 Erlangen
Deutschland
Tel.: +49 (0)9131/85 - 29703
Fax: +49 (0)9131/85 - 29709
E-Mail: ehrenstein@ikt.uni-erlangen.de

Europa / Europe
Prof. Dr.-Ing. Dietmar Drummer, verantwortlich
Lehrstuhl für Kunststofftechnik
Universität Erlangen-Nürnberg
Am Weichselgarten 9
91058 Erlangen
Deutschland
Tel.: +49 (0)9131/85 - 29700
Fax: +49 (0)9131/85 - 29709
E-Mail: drummer@ikt.uni-erlangen.de

Amerika / The Americas
Prof. Prof. hon. Dr. Tim A. Osswald, verantwortlich
Polymer Engineering Center, Director
University of Wisconsin-Madison
1513 University Avenue
Madison, WI 53706
USA
Tel.: +1 608/263 9538
Fax: +1 608/265 2316
E-Mail: osswald@enr.wisc.edu