# THE INFLUENCE OF PYROLITIC DEGRADATION ON MECHANICAL PROPERTIES OF CARBON FIBRES WITHIN RECYCLING COMPOSITE MATERIALS

*Michal NÁVRAT[1], Jaroslav ZÁVADA[1], Veronika GLOGAROVÁ[1]*
*[1] Institute of Environmental Engineering, VŠB-Technical University of Ostrava, Czech Republic*
*e-mail: jaroslav.zavada@vsb.cz*

**Abstract**

The article deals with the influence of thermal pyrolytic degradation on mechanical properties of carbon fibres used in the production of composite material. The carbon fibre has been chosen as the reinforcement of composite and the resin formed a matrix (binder). During the pyrolysis process, the resin was eliminated and the carbon fibre was separated. Pyrolysis was carried out at temperatures of 450 °C, 550 °C and 650 °C. Subsequently also tensile tests were performed on the treated material to compare the mechanical properties of the fibres prior to pyrolysis and after decomposition. The results showed negative influence at the selected temperatures during the pyrolysis treatment on the mechanical properties of the carbon fibres.

**Keywords:** carbon fibre; composite material; pyrolysis; mechanical properties

## 1 INTRODUCTION

The fundamental of composite materials is their composition. Composites consist of two or more components which differ from each other in their mechanical, physical and chemical properties. They are produced by mechanical blending of individual components of a composite [3], [4]

The use of the most up-to-date types of composite materials can be observed in astronautics, aeronautics or automobile industries. They are fibrous materials from boron, glass, aramid or carbon embedded in the composite matrix which is mostly made of metal, pottery or polymer in the form of resin or bismaleimide.

The main advantage of composite materials compared to traditional steel structures is their industrial process, mainly the simpler manufacturing of elements of a more complicated shape, a lower amount of parts produced as well as a smaller amount of produced waste. Another unquestionable advantage consists in arbitrary setting layers during manufacturing a composite, thanks to which the whole industrial process is simplified. This arbitrary setting of layers also facilitates changing the thickness of a product and thus simplifies manufacturing elements more complicated in shape. [1], [2], [3] On the other hand, an exact ratio of matrix and reinforcement must be kept during composite hardening, as well as the correct direction of orientation of fibres and the course of manufacture. [3]

## 2 PYROLYSIS TESTS

The pyrolysis tests of a thermosetting composite with fibre reinforcement were implemented in the company MSV Studénka s. r. o. in Bílovec near Ostrava. The pyrolytic decomposition of its polymer matrix took place in an electric oven provided by the company Elektrické pece Svoboda. The pyrolysis process took 5 hours. Totally, there were three pyrolytic decompositions done, namely at the temperatures of 450 °C, 550 °C and 650 °C so that it is possible to find out which temperature is ideal and leads to the perfect removal of polymer matrix in composite. Consequently, individual recyclate samples were observed under an electron microscope which revealed whether the polymer matrix was really fully removed from a fibre so that it is possible to find out changes in mechanical properties of the given recyclate, the tensile strength of which was tested on a tensile testing machine in laboratories of the Faculty of Materials Engineering at VŠB – TU Ostrava.

### 2.1 Sample characteristics

The samples for pyrolytic decomposition were provided by the Czech Aerospace Research Centre. They were rectangular carbon plates of 40 mm × 20 mm and a thickness of 2 mm (Fig 1). The composite was formed from the carbon fibre which functioned as reinforcement and the epoxy resin which functioned as a matrix or bonding compound. The used carbon fibre fabric is named HEXCEL G1157 D. The type of carbon fibre used for fabric manufacturing is called TENAX E HTA40 E 13 6K. The tensile strength of the fibre, specified by the producer, is 3950 MPa. This value will be compared to the value of the tensile strength of recycled fibre which was measured on a walter+bai tensile testing machine LFV 100 kN.


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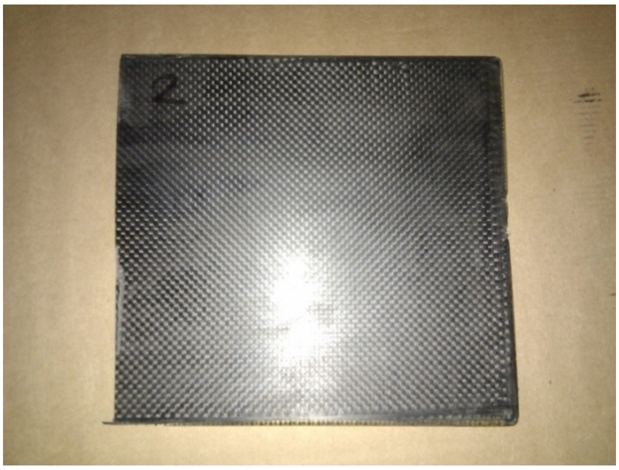


**Fig. 1 Sample of composite plate from carbon fibre filled with epoxy resin.**

## 2.2 Characteristics of pyrolytic decomposition

The epoxy used during the production of a composite plate begins to degrade thermally at about 400 °C – 420 °C. The largest smoke generation is at a temperature between 420 °C and 450 °C. At temperatures over 460 °C smoke does not occur anymore which means that the main part of decomposition has finished. [44] The processing temperature denoted by Tz is considered the minimum sufficient temperature for the complete removal of polymer components of the composite. For the processed composite material of carbon – epoxy type, the sufficient processing temperature is Tz = 550 °C. At higher temperatures, the carbon fibres are over-decomposed which can lead to a significant deterioration of their mechanical properties. A short period of temperature persistence can cause insufficient warming of the processed composite, thus also incomplete removal of polymer matrix. [5] The chosen temperature procedure of pyrolytic decomposition depending on time up to the value of minimum temperature required for the removal of polymer matrix is shown on a graph in Figure 2. The temperature increase dependent on time was the same for all three samples. The only difference was in the final processing temperature.

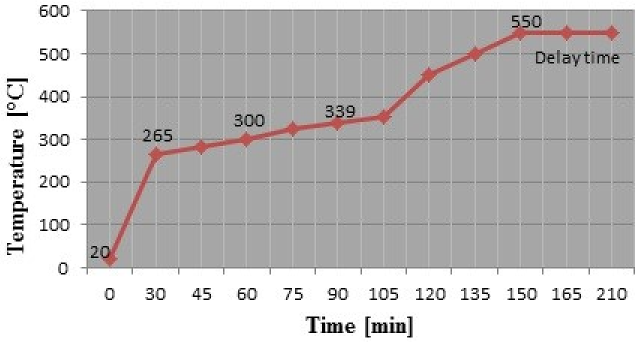


**Fig. 2 Temperature curve of universal processing mode.**

The relevant length of temperature persistence depends on the specifications of filling – the processed material. All three samples tested had the same thickness of 2 mm and were therefore kept at their processing temperature for 120 minutes.

## 2.3 Characteristics of tensile tests

After the pyrolytic decomposition, the separated carbon fabric was hairsprayed to make the fibres stick together and to make it easier for operating. Then individual clusters of fibre were dissected from the carbon fabric and glued between two prepared carbon plates of 3 × 4 cm with fast-bonding glue. The plates were prepared because of better gripping the sample between the tensile testing machine jaws (Fig. 3). Four testing fibres were prepared from each sample obtained at different pyrolysis temperatures so that it is possible to calculate an average tensile strength of the recycled carbon fibre. The tensile test was performed on a multifunctional servo-hydraulic testing machine LFV 100 kN by the company w+b in accordance with the standard ČSN EN ISO 527-1. The load rate was 10 mm/min. Only one cluster with 6000 fibres was tested. From the known diameter of one fibre dv [mm], the initial section of a fibre $S_v$ [$mm^2$] and the initial section of the tested cluster $S_0$ [$mm^2$] were calculated:

$$S_v = \frac{\pi d_v^2}{4} \quad (1)$$


Volume LXIII (2017), No. 4
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$$S_0 = 6000 \cdot S_v \tag{2}$$

The load F [N] was converted to stress σ [MPa].

$$\sigma = \frac{F}{S_0} \tag{3}$$

From the calculated stress, the maximum value which corresponds to the ultimate strength $\sigma_m$ [MPa] was calculated. The strain ΔL [mm] was converted to the specific elongation at ultimate strength $\varepsilon_m$ [-] where $L_0 = 120$ mm:

$$\varepsilon = \frac{\Delta L}{L_0} \tag{4}$$

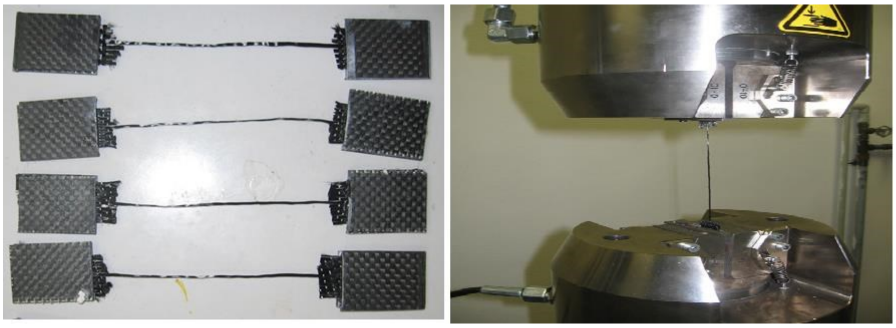

**Fig. 3 Preparing the sample of carbon fibre and fastening the sample in a tensile testing machine.**

## 3 RESULTS OF ANALYSES

### 3.1 Sample 1 – 450 °C

At processing temperature of 450 °C, the polymer matrix was removed very well (Fig. 4) and it is possible to ground the recyclate to a required size fraction and use its properties again, e.g. by adding it into construction materials. [4] However, during thorough observations under an electron microscope, it can be seen that the polymer matrix is not completely removed and the carbon fibres are still sporadically covered with the epoxy resin (Fig 5). The mechanical properties of sample 1 are given in Table 1.


**Fig. 4 Appearance of recyclate after pyrolytic decomposition at processing temperature of 450 °C.**

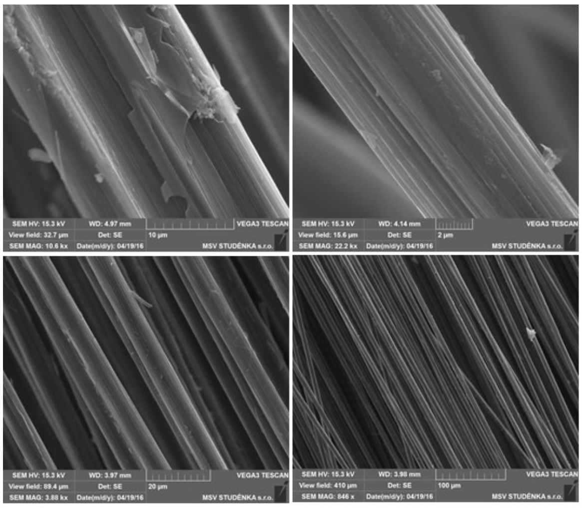

**Fig. 5: Detailed appearance of fibres at processing temperature of 450 °C.**

**Tab. 1 Mechanical properties of sample 1 - 450 °C, d0 = 0.0075 mm.**

| Sample 1 | Ultimate strength $\sigma_m$ [MPa] | Elongation $\Delta L$ [mm] | Strain $\varepsilon_m$ [-] |
|---|---|---|---|
| 1_1 | 1699.10 | 1.70 | 0.014 |
| 1_2 | 1705.92 | 1.57 | 0.013 |
| 1_3 | 1829.24 | 1.87 | 0.016 |
| 1_4 | 1143.46 | 1.64 | 0.014 |
| Diameter | **1594.43** | **1.70** | **0.014** |
| Stand.deviation | **265.47** | **0.11** | **0.001** |

As the results show, the average tensile strength of the recycled carbon fibres TENAX E HTA40 E 13 6K after the pyrolytic decomposition (450 °C) is 1594.43 MPa. Compared to the value stated by the producer, 3950 MPa, the tensile strength deteriorated by 59.63 %.

## 3.2 Sample 2 – 550 °C

At processing temperature of 550 °C, the polymer matrix was almost perfectly removed which was obvious from electron microscope images. The mechanical properties of sample 2 are stated in Table 2

**Tab. 2 Mechanical properties of sample 2 (550°C), d0 = 0.0066 mm.**

| Sample 2 | Ultimate strength $\sigma_m$ [MPa] | Elongation $\Delta L$ [mm] | Strain $\varepsilon_m$ [-] |
|---|---|---|---|
| 2_1 | 707.34 | 1.37 | 0.011 |
| 2_2 | 642.04 | 1.13 | 0.009 |
| 2_3 | 587.30 | 1.23 | 0.010 |
| 2_4 | 587.32 | 1.25 | 0.010 |
| Diameter | **645.56** | **1.25** | **0.010** |
| Stand.deviation | **49.06** | **0.10** | **0.001** |

As the results show, the average tensile strength of the recycled carbon fibres TENAX E HTA40 E 13 6K after the pyrolytic decomposition (550 °C) is 645.56 MPa. This means that the tensile strength deteriorated by 83.66 %.

## 3.3 Sample 3 – 650 °C

Due to the absolute degradation of the composite sample during the pyrolytic processing (650°C), the tensile tests cannot be performed.

## 4 CONCLUSION

The article deals with the pyrolytic processing of a composite material made by combining carbon fibre and epoxy resin. The influence of thermal pyrolytic decomposition on the mechanical properties of the carbon fibre was examined, particularly on its tensile strength. Three pyrolytic decompositions were performed at different temperatures (450 °C, 550 °C and 650 °C) so that it is possible to verify which temperature leads to the complete removal of polymer matrix and find out what influence the temperature has on the mechanical properties of carbon fibre.

In case of Sample 1, which was processed at the temperature of 450 °C, the polymer matrix was almost completely removed but observations under an electron microscope proved that the carbon fibres are still slightly covered with the resin. The tensile tests proved that the tensile strength deteriorated by 59.63 %.

Sample 2 was processed at the temperature of 550 °C. The electron microscope showed that the polymer matrix was almost completely removed and the fibres are not covered with the resin. The tensile tests proved that the tensile strength deteriorated by 83.66 %.

Sample 3 was processed at the temperature of 650 °C. At this temperature, the sample was absolutely degraded, therefore, the carbon fibre could not be tested.

Due to the obtained results, it can be concluded that the chosen temperature for pyrolytic processing has a negative influence on the mechanical properties of the carbon fibre. Therefore, it is not recommended to reuse the carbon recyclate as reinforcement in the production of a new carbon element. However, the carbon recyclate can be grounded into a required size fraction and used e.g. by adding it into construction materials or into the glass fibre from which conveyors for track vehicles in MSV Studénka are made. [6]

The authors [7] tried to thermally recycle carbon fibres with super-heated steam. In order to manufacture high quality recycled carbon fibres (R-CFs), carbon fibre-reinforced composite wastes were pyrolysed with super-heated steam at 550 °C in a fixed bed reactor for varying reaction times. The surface analysis showed that there was no matrix char residue on the fibre surfaces. The recycling efficiency of the R-CFs from the composites was strongly dependent on the pyrolysis temperature, reaction time, and the super-heated steam feeding rate. Based on the results, increasing the steam pyrolysis conditions was favourable for increasing the removal of char. During pyrolysis, the tensile strength deteriorated only by 9.58 %, which was the highest value among the obtained clean carbon fibres (CFs), compared to that of the virgin carbon fibres (V-CFs).

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