

# Influence of Some Technological Parameters on the Content of Voids in Composite during On-Line Consolidation with Filament Winding Technology

M. Stefanovska, B. Samakoski, S. Risteska, G. Maneski

**Abstract**—In this study was performed in situ consolidation of polypropylene matrix/glass reinforced roving by combining heating systems and roll pressing. The commingled roving during hoop winding was wound on a cylindrical mandrel. The work also presents the advances made in the processing of these materials into composites by conventional technique filament winding. Experimental studies were performed with changing parameters – temperature, pressure and speed. Finally, it describes the investigation of the optimal processing conditions that maximize the mechanical properties of the composites. These properties are good enough for composites to be used as engineering materials in many structural applications.

**Keywords**—Commingled fiber, consolidation heat, filament winding, voids.

## I. INTRODUCTION

CONTINUOUS fiber-reinforced polymers (CFRP) are complex materials composed of fibers and polymers as matrix. Thermoplastic matrices are rarely used in these so-called CFRP composites. There is a lot of interest in fiber composite materials with thermoplastic polymer matrix, because the thermoplastic polymers compared with thermosetting polymers offer a potential for greater fracture toughness, large elongation at fracture, faster and more automatic processing, unlimited shelf life of the raw material, recycling and cleaner working environment [1]. The most important differences in the production technology of fiber composites with thermoplastic matrices compared with composites with thermosetting matrices arise from the much higher viscosity and higher processing temperature of the thermoplastic composites. The melt viscosity of the thermoplastic matrices is high, 100-1000 Pa·s which means that it is difficult for the molten plastic to penetrate into the fiber bundles and ensure a complete wetting of all individual fibers. In comparison, the viscosity of thermosetting matrices used for fiber composites is in the range of 0.1-10 Pa·s [2], [3]. For thermoplastic matrix materials which can be used

continuously at elevated temperature, such as 100-250°C, the processing temperature is in the range of 220-390°C and, of course, special care is required in designing tools and selecting accessories regarding thermal expansion and thermal resistance. Thermoplastic composites are expected to undergo substantial growth over the next 5 to 20 years. There is particularly strong growth in automotive and aerospace sectors. The main advantages compared to thermosetting composites are rapid processing, reduced volatiles, improved reuse/recycling, and in some cases reduced material cost (e.g. polypropylene). In the new material process development additional effort should be put in place to reduce the percentage of void content in composites according to standards [4], [5].

## II. EXPERIMENTAL PART

The roving used in this work was Twintex® R PP 60 B 1870, dry prepreg made by commingling continuous glass roving and polypropylene (PP) filaments supplied by Fiber Glass Industry, Inc. The glass weight content was 60% and the nominal lineal weight was 1870 tex. Chemical composition of this product is given in Table I.

TABLE I  
THE CONTENT OF COMMINGLED FIBER TWINTEX® R PP 60 1870 B

Twintex® R PP 60 1870 B	Glass content (%wt)	Glass roving (tex)	Matrix	B
R-roving	60	1870	Polypropylene (PP)	Black color, heat and UV stabilization

Experimental part was performed in laboratory conditions on prototype filament winding machine (FB 6/1) made by Mikrosam AD. The machine was integrated with specially designed laboratory head for in situ winding of thermoplastics. Schematic layout of designed laboratory head with all parts can be seen on Fig. 1.

As can be seen from Fig. 1, the constructed machine head for in situ consolidation with filament winding technology consisted from several parts: steel mandrel, first heated roller with temperature T1, second heated roller with temperature T2, consolidation heated roller with temperature T3 and maximal pressure P of 10 bar, heat air-gun unit with temperature T4 and roving heating unit with temperature T5.

The roving was preheated in the roving heating unit and passed through tension system made of first and second heated

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roller. The consolidation of the roving was seen directly on the rotating mandrel with help of consolidation roller, which apply temperature and pressure on the roving. The mentioned rollers were heated to be prevented thermal shock of the PP matrix. Heat air-gun was used to help the consolidation between thermoplastic layers.

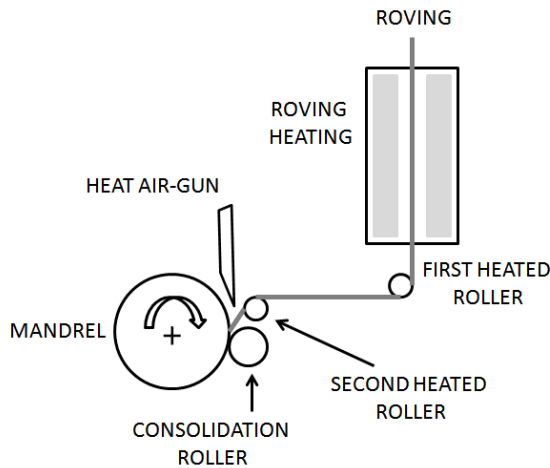


Fig. 1 Schematic layout of designed laboratory head

At the beginning, were determined nine experiments with five technological parameters to see how these parameters affect the percent of void in the final composite. Following parameters were chosen for the research: temperature of second heated roller, temperature and pressure of consolidation roller, temperature of heat air-gun and speed of winding V (Table II).

TABLE II  
 TECHNOLOGICAL PARAMETERS OF SAMPLE PRODUCTION

N <sup>o</sup>	T1 (°C)	T2 (°C)	T3 (°C)	T4 (°C)	T5 (°C)	P (bar)	V (m/min)
1	100	120	230	/	70	1	1.5
2	100	120	230	/	70	3	1.5
3	100	120	230	/	70	5	1.5
4	100	120	250	/	70	5	1.5
5	100	120	260	/	70	5	1.5
6	100	120	270	/	70	5	1.5
7	100	120	300	/	70	5	3
8	100	100	300	/	70	5	6
9	100	100	250	150-200	70	5	1.5

The temperature of the second heated roller T2 had values 100°C or 120°C, whereas the consolidation roller had temperature T3 in the range from 230°C to 300°C and pressure from 1 bar to 5 bars. Further, heat air-gun temperature was between 150-200°C and wasn't involved in all tests. Finally, the minimal and maximal speeds of roving winding were 1.5 m/min and 6 m/min, respectively and 3 m/min was middle speed of winding. The temperature of the first heated roller T1 and the temperature of roving heating unit T5, were held constant in all experiments. The specimens were manufactured by filament winding with different number of layers, from one

to three, as schematically shown on Fig. 2.

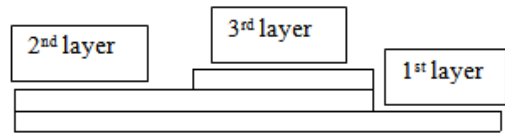


Fig. 2 Method of winding a sample with one, two or three layers

### III. RESULTS AND DISCUSSION

Similar equipment for in situ filament winding of Twintex® TR PP 60 B 1870 was reported in [6]. To prevent crystallization of PP matrix they have used air gun as a heat source. Also, in [7] has been investigated novel process technology, where PP-homopolymer type Novolen 1100® VC was on line impregnated with filament winding technology. Consolidation of PP-homopolymer was made with help of hot air gun for heating the nip-point, whereas the nip-point temperature was controlled with IR-thermometer.

Parameters for production of Twintex® tubes by filament winding technique were Taguchi approach reported in [8]. Fibers temperature, winding speed, number of layers and roving were used as parameters. With help of Taguchi method experimental parameters were reduced and effective parameters were determined. Further, [9] performed fatigue life analysis of GFR/epoxy filament wound composites pipe according to Taguchi experiment design (L<sub>9</sub>) for three different levels of fatigue parameters: filament angles, surface crack-depth ratios and stress levels. It was concluded, that stress levels among fatigue test parameters showed the highest effect to results.

#### A. Differential Scanning Calorimetry (DSC)

Differential scanning calorimetry (DSC) was done to discover the melt temperature of the Twintex®. This information will determine the lowest processing temperature of the polypropylene matrix during the filament winding process.

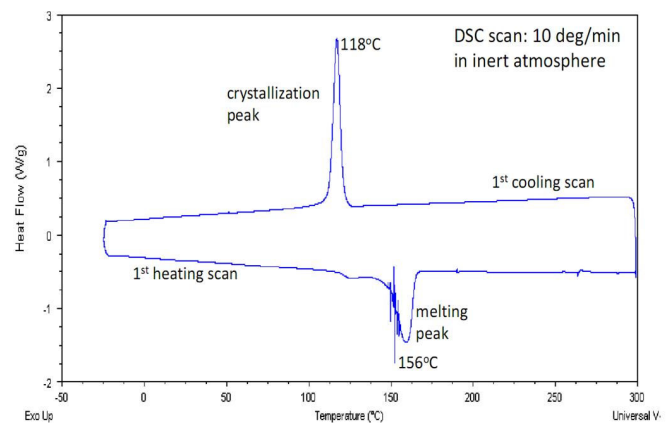


Fig. 3 DSC curve of PP used in commingled fibers

For this purpose Perkin Elmer differential scanning calorimeter was used with maximal temperature 300°C and

heating ramp 10°C/min in inert atmosphere. Fig. 3 shows the DSC curve of the Twintex®, where two peaks can be observed. First peak is crystallization peak of PP at 118°C and the second peak at 156°C corresponds to the melt temperature of the PP. Similar melt and crystallization temperatures of PP are reported in [10]-[14].

### B. Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) were performed to be determined maximal processing temperature of PP. Sample from Twintex® was heated up to 600°C with heating ramp 30°C/min in inert atmosphere. Fig. 4 shows the TGA curves of the Twintex®. As seen in Fig. 3, until 300°C there cannot be seen any mass change of the Twintex®, which means there is no PP degradation to be observed. However, with increasing temperature at 420°C the TGA curve starts to fall and Twintex® losses almost 43% weight at 450°C. With further rise of the temperature there is no change in the mass percent to be observed. Similar results of TGA for PP are reported in [15], [16].

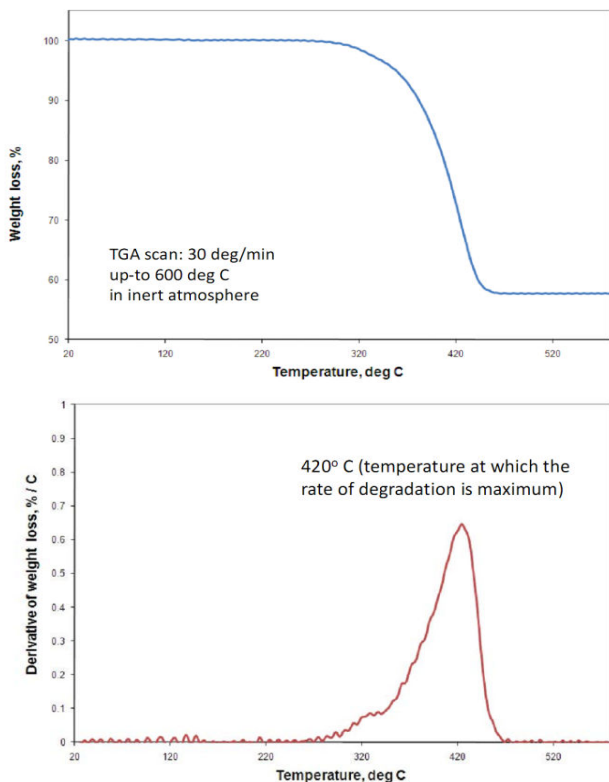


Fig. 4 TGA curve of PP used in commingled fibers

### C. Void Content

Void content in the composite was determined with help of experimental and theoretical density of composite according to ASTM D 3171 standard [4]. This characteristic of composite product is very important to characterize the quality of the produced part. Void content in the composite was used to characterize the quality of the produced part and it was calculated using the experimental and theoretical density of the composite according to (1):

$$V_{void} = \frac{(\rho_{ct} - \rho_{ce})}{\rho_{ct}} \quad (1)$$

where  $V_{void}$  is void content of the composite,  $\rho_{ct}$  is theoretical density of the composite ( $\text{kg/m}^3$ ) and  $\rho_{ce}$  is experimental density of the composite ( $\text{kg/m}^3$ ). As stated above, method A of ASTM D 792 standard was used to calculate the experimental density  $\rho_{ce}$  of the composite. Five samples with dimensions 2.54 cm x 2.54cm were cut from the wound composite with diamond blade saw. After the samples were cut, their weight was measured in air and beaker of ethanol to obtain the specific gravity. Sartorius (model) density determination kit was used to weigh the sample. Specific gravity of the sample was calculated according to (2):

$$y = \frac{W_{ca}}{W_{ca} - W_{ce}} \quad (2)$$

In (2)  $y$  is experimental specific gravity of the composite,  $W_{ca}$  is weight of composite sample in air (kg) and  $W_{ce}$  is weight of composite sample in ethanol (kg). The density of the composite  $\rho_{ce}$  is determined using the specific gravity and density of ethanol according to (3).

$$\rho_{ce} = y\rho_e \quad (3)$$

In (3)  $y$  is experimental specific gravity of composite,  $\rho_e$  is density of ethanol ( $\text{kg/m}^3$ ).

The experimental density was determined according to method A from ASTM D 792 standard [5] and the theoretical density was calculated according to (4).

$$\rho_{ct} = \frac{100}{\left(\frac{w_f}{\rho_f} + \frac{w_r}{\rho_r}\right)} \quad (4)$$

where  $w_f$  is weight percent of fiber in the composite (%),  $w_r$  is weight percent of resin in the composite (%),  $\rho_r$  is density of resin ( $\text{kg/m}^3$ ) and  $\rho_f$  is density of fiber ( $\text{kg/m}^3$ ).

In Table III are given the percents of voids in the wound composite's samples. It can be seen, that void percent in all samples is between 3.45% and 0.5%. The best results were achieved in sample 9 were head air-gun unit with temperature between 150-200°C was used. This was the only sample in which this unit was used in the consolidation process. Also, the speed of winding and consolidation pressure had shown an influence in the percent of voids. The winding speed had minimal value of 1.5 m/min, which gave enough time for the PP to be melted and consolidated, whereas maximal consolidation pressure of 5 bar helped in removing the trapped air bubbles into the structure. The biggest percent of void 3.45% was noticed in sample 1, where no air-gun was used and even though the winding speed was minimal, the consolidation pressure of 1 bar was too small to decrease the percent of void. In the rest samples where different temperatures and no air-

gun were used for consolidation, the percent of void was 1.5% to 2.5%.

TABLE III  
 EXPERIMENTAL RESULTS FROM THE SAMPLES

N <sup>o</sup>	W <sub>f</sub> %	W <sub>r</sub> %	ρ <sub>ct</sub> g/cm <sup>3</sup>	V <sub>void</sub> %
1 <sup>st</sup> 1-5	60	40	1.428	3.45
2 <sup>nd</sup> 1-5	60	40	1.428	2.53
3 <sup>rd</sup> 1-5	60	40	1.428	2.50
4 <sup>th</sup> 1-5	60	40	1.428	2.34
5 <sup>th</sup> 1-5	60	40	1.428	2.10
6 <sup>th</sup> 1-5	60	40	1.428	1.32
7 <sup>th</sup> 1-5	60	40	1.428	1.50
8 <sup>th</sup> 1-5	60	40	1.428	1.80
9 <sup>th</sup> 1-5	60	40	1.428	0.50

According to these results, it is obvious that the temperature of the air-gun T4 plays a major role in the consolidation process. Reference [17] have reported 3.4% voids in Twintex® laminates manufactured with help of compression molding and vacuum molding. Around 2% void content at 4 bar pressure was determined in [7]. Beyond this pressure no further decrease in void percent is observed.

#### D. Constituents Weight Percentage

The weight percentages of the components of the composites were needed to calculate the theoretical density. As guidelines were used measurements according to ASTM D792 [5] and ASTM D 2584 [18] standards. In both standards are mentioned different methods for resin removal from the composite. After resin removal, weight of remaining fibers was measured. The weight percentage of the remaining fiber was calculated according (5), where W<sub>f</sub> is weight percent of fiber in the composite (%), W<sub>r</sub> is weight percent of resin in the composite (%), W<sub>fc</sub> is weight of fibers in the composite sample (g) and W<sub>c</sub> is weight of composite sample (g).

$$W_f = 100 \left( \frac{W_{fc}}{W_c} \right) = (1 - W_r) \quad (5)$$

To determine the weight percentage of the components in the final composite burn-off method was used. Five samples with dimension 2.54cm x 2.54cm were cut from the fabricated composite pipes and were placed in furnace with temperature of 600°C in duration of 1 hour. When burning process was completed, the weight of remaining glass fibers was recorded at room temperature. The results are shown in Table III, where fiber volume percent in the final composite is 60% and calculated density of the composite is 1.428g/cm<sup>3</sup>. The same percent volume of fibers was reported for the Twintex® in the technical specification by the manufacturer (Table I).

#### E. Scanning Electron Microscopy (SEM)

Small samples were cut from manufactured GF/PP samples and were placed and observed under scanning electron microscope (Hitachi S-800) without any surface preparation to determine the impregnation quality.

SEM analyses were used to verify the impregnation depth of resin into the fibers and to determine the porosity of the commingled fibers. In Fig. 5 are shown some of SEM images of the samples.

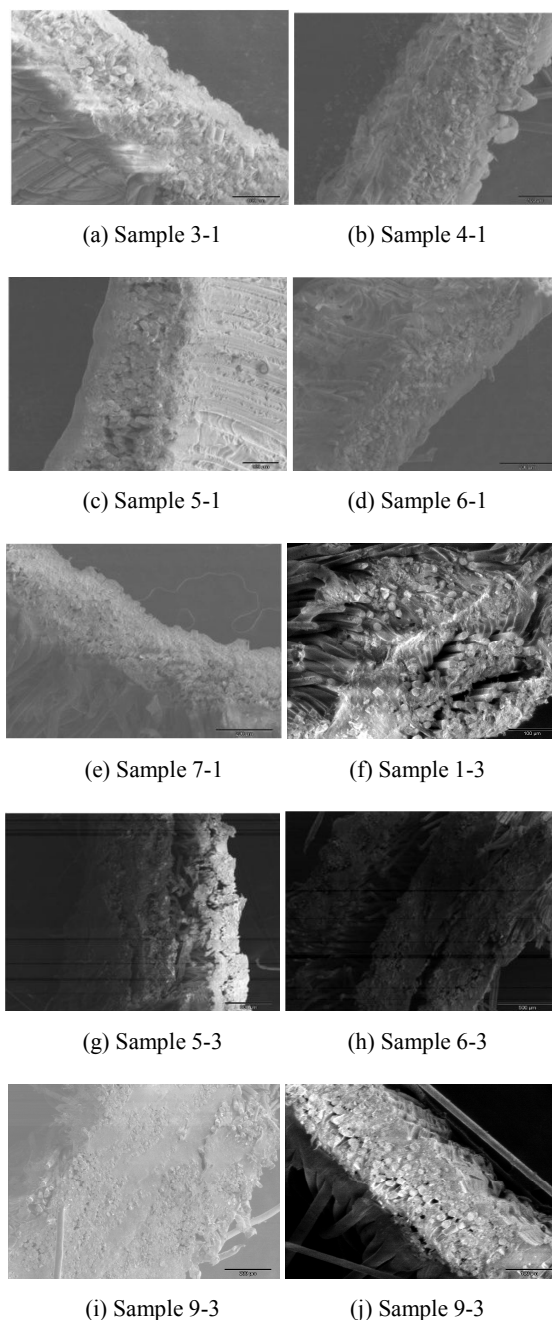


Fig. 5 Impregnation quality of prepared samples

Description of SEM images is based on the sample number and number of layers from which the sample is made. For example, sample 2-3 means probe 2 made with 3 layers according to the parameters given in Table II for sample 2. From SEM analysis of the samples can be seen good impregnation of PP into the fibers when the number of layers is smaller than 3. When number of layers is 3, some dry fiber arias can be observed by some of the samples, which lead to

interlaminar shear failure. Very good impregnation due to the intimate contact between the individual dry glass and PP fibers was reported by [19] in design and manufacture of low-cost full scale pultrusion prototype equipment. The conclusion was based on samples observation made by reflected light optical microscopy with surface preparation of the samples. Also, some larger dry spots between the glass fibers at larger magnifications have been reported. Experimental procedure to study the consolidation of Twintex® T PP and Comfil® G was done in [20]. Reference [20] have indicated, that consolidation behavior of Comfil® composite is influenced by the viscosity of the matrix and by the distribution of bundles, whereas compaction of Twintex® takes place after melting of the matrix and applied pressure increases consolidation efficiency. According to this research matrix fiber coalescence in Twintex® is the main mechanism involved in consolidation.

#### IV. CONCLUSION

The experimental procedure described in the present work is suitable to study the consolidation behavior of thermoplastic matrix composite. The results shown, that good interaction between the layers is strongly dependent on the temperature of air-gun, which should be greater than the thermoplastic melt temperature. Rising of the temperature from 210°C to 300°C is followed by sharp fall of voids percent due to the good melting of the PP, which is characterized by low viscosity and good wetting of the glass fibers. Temperature higher than 300°C is not desired, due to degradation of thermoplastics. Also, higher pressure of consolidation roller will decrease the percent of voids in the final composite, together with 1.5 m/min speed of winding.

From the obtained SEM images is to be seen good merger between composite layers in all samples from 1 to 8. But when three layers are coming in connection there are large cavities between the layers, which lead to conclusion that there is not good merger.

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