THE POROSITY OF PLAIN WEAVE CARBON CARBON COMPOSITES AS AN INPUT PARAMETER FOR EVALUATION OF MATERIAL PROPERTIES

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Abstract

This paper introduces the preliminary study of the void system evolved in carbon carbon (C/C) composites due to the individual steps of technological process. The void cross-sections, observed on planar microphotographs of the composite specimen surface, are analyzed with the modern methods of image analysis.

1 Introduction

Generally, plain weave C/C composites are produced from a moulded fabric/resin preform (C/P composite) with a long series of carbonization/densification treatment. This technology, matrix precursor pyrolysis in particular, causes extensive development of voids in the composite structure. The experimental measurements have shown that the porosity of C/C composite, ensued from carbonization of fabric/resin preform, may reach up to 50%, porosity of final product ranges from 20 to 30%. As non-separable part of the C/C composite structure it becomes an important characteristic of material determining the further material properties. The voids, as a great barrier of the heat flow, decrease e.g. thermal conductivity of composites [1], on the other hand porosity is necessary for composites biocompatibility as it enables the living tissue penetration etc [2,3].

The structure of voids is complex and not well described. Voids may be characterized according to their size (macropores, mesopores, micropores), position (transversal cracks, delamination cracks, crimp voids, matrix pores etc.) or shape (rod-like shape, thin plates, rough branch structures) [4,5].

In any case the proper knowledge of porosity and void structure is necessary whether we aim to control the porosity progress or create the models describing physical properties of C/C composites. For this purpose the detailed analysis of voidness due to the individual steps of technological process is developed.

2 Experiment

2.1 Examined material

Composite plate has been processed with three series of carbonization/densification treatment and final graphitization of carbonized composite. C/P preform has been prepared with hot pressing of eight layers of carbon fabric (Hexcel G 1169 created from the carbon multifilament Torayca T 800 HB) impregnated with phenolic resin UMAFORM LE [6].

The influence of technology on composite structure has been analyzed on selected structural specimen set up of cross-section cuts of composite plate in every step of technological process. The selected set of specimen contains these composites: *green **CP**, carbonized **C**, carbonized/densified **CI**, recarbonized **CIC**, recarbonized/densified **CICI**, twice recarbonized **CICIC**, and graphitized **G**.*

2.2 Image analysis of microstructure

The structure of the composites has been examined on the planar microphotographs of specimen surface. To receive high quality structural images, the surface of specimen have been ground and polished (using standard metallographic techniques) prior to scanning of specimen structure viewed by polarized light. Image analysis device, employed for data acquisition, consists of the following components:

- NIKON ECLIPSE E 600 microscope, Märzhäuser motorized scanning stage, digital monochrome camera VDC 1300C, software LUCIA G [7]

The main part of composite image processing has been provided using image analyser LUCIA G, which offers wide range of functions enabling complex image processing from image adjustment prior to actual analysis up to complex image data analysis [7].

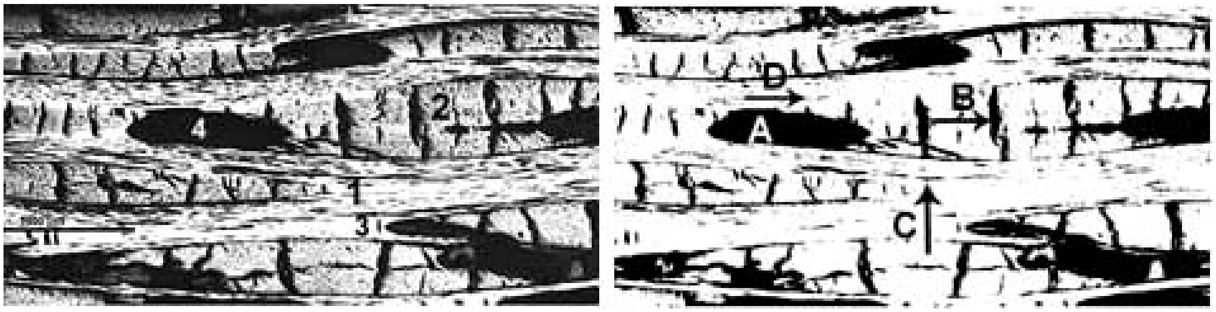
Structural measurement is provided using the options of Measurement menu (measurement, calibration, object and field measurements, measurement frame, measurement features). Measurement features give a wide range of functions to evaluate structural objects (Area, AreaFraction, Circularity, Elongation, Length, MaxFeret, MeasuredArea, MinFeret, etc).

2.3 Methodology of the void morphology description

The reaming part of this paper concentrates on the investigation of the void development in the composite structure due to the technological process. The studied voids are observed on planar microphotographs of composite specimen surface. The least void that can be recognized has a size of one pixel. The real length of one pixel in studied images is equal to 1.7 μm. Example of scanned composite structure is shown in Fig.1a).

Previous study of the binary images of composite structure led to the hypothesis there are four main systems of the voids [8], see Fig.1b):

- The vacuoles appearing among the carbon tow reinforcement, A
- Transversal cracks perpendicular to the orientation of carbon tow, B
- Elongated voids in the direction of carbon tow (delamination cracks in particular), C
- Small holes of rather circular cross-section, D



a) **b)**

Figure 1. a) Scanned structure of C/C composite: *1-carbon tow,longitudinal cut, 2-carbon tow, cross-section cut, 3-carbon matrix, 4-void* b) Main system of voids: *A-vacuoles, B-transversal crack, C-elongated void, D- small hole*

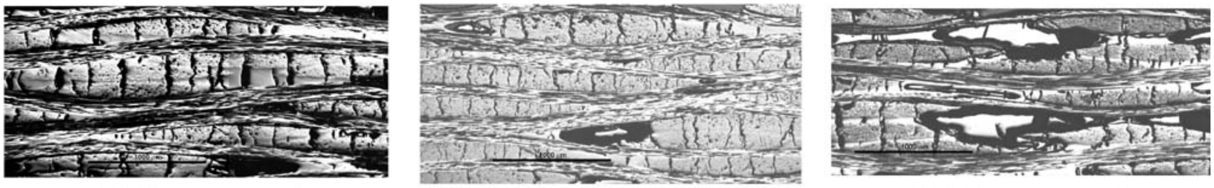
3D visualization of the most extensive voids shows they possess complex structure [8], not in the least reminding of the globular shape typical for pores, which are mostly defined with pore cross-section diameter [9]. In case of C/C composites‘ voids their cross-section area appears as more suitable morphological characteristic than its diameter. As supplement parameters have been chosen void area fraction, maximum and minimum Feret’s diameters.

These morphological characteristics have been determined for all four void groups in every composite specimen, and their statistical parameters have been evaluated. The results have shown the wide range of obtained values for measured characteristics, and the overlay of separately examined groups.

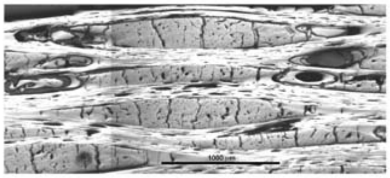
Although the analysis of Feret’s diameters in the study [8] proved the two-directional orientation of void goups depending on the course of carbon tows, in woven reinforcement interlaced in two nearly orthogonal directions, the evaluation of obtained data indicated the unsuitability of statistical processing of the characteristics obtained for divided void systems. Therefore the histograms of void area distribution were processed for integrated sample of all measured voids as a definite description of the system. Size of one sample containing all four void types is about 50000.

3 Results and discussions

CP composite C composite CI composite



CIC composite CICI composite CICIC composite



G composite

Figure 2. The structure of individual specimen after every step of technological process

The microphotographs of composite specimen structure, shown in Fig.2, indicate the progress of voids due to the technological process. It is obvious that the initial voids develop already during the C/P composites‘ curing. These voids form due to pressure of water vapour which molecules eliminate as a result of polycondensation reaction of the phenolic resin in the temperature of 117 °C [10]. The cross-linking of resin and increasing viscosity of the system avoid the escape of vapour bubbles and fix them inside the composite structure in the form of extensive vacuoles. These voids remind of the cavity system in the rocks.

Carbonization of fabric/resin preform (C composite) starts rapid progress of voids in composite structure, the porosity may reach up to 40%. Its main part is on behalf of delamination cracks that spread in fabric-resin interface and connect with the vacuoles. It creates the complex system of extensive voids oriented in the direction of carbon tows in woven reinforcement. The other significant system of voids is located in carbon tows, and these voids are described as transversal cracks perpendicular to the tow axis. It is obvious this void systems continuously pass one another.

Further densification/carbonization treatment is applied to decrease the porosity and improve the volume density of final graphitized composite, see Fig.3.

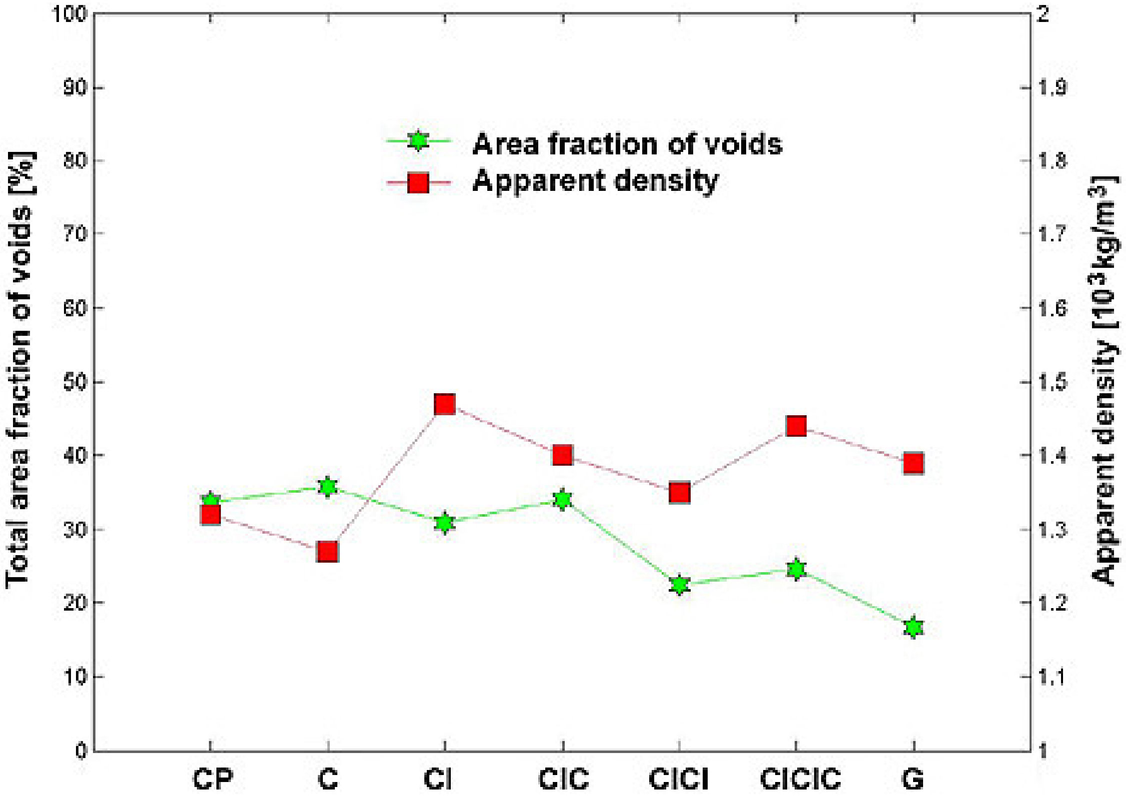


Figure 3. The development of porosity and apparent density[*1] due to technological process

[*1] The real density of system cannot be specified with the current penetration methods as it does not capture the closed pores. The number of closed pores increase due to densification process, and those cannot be filled with matrix precursor again [11]. It indicates there is no efficiency of too many cycles of carbonization/densification treatment, as the chance to densify the composite structure is limited with the number of open pores.

As mentioned above the results of measured void areas vary in wide range of values. The basic histograms for relative frequency of void area proved there is necessary to evaluate the void areas separately according to their size. The area of 5000 μm^2 have been experimentally chosen as a dividing value. Comparing this value to the definition of macropores suitable for living tissue penetration in [2] we consider the group of pores greater then 5000 μm^2 may be defined as macropores from the viewpoint of biocompatibility of composite. The distribution of macropores is shown in Fig.4. The statistical characteristics of measured void areas are shown in Fig.5.

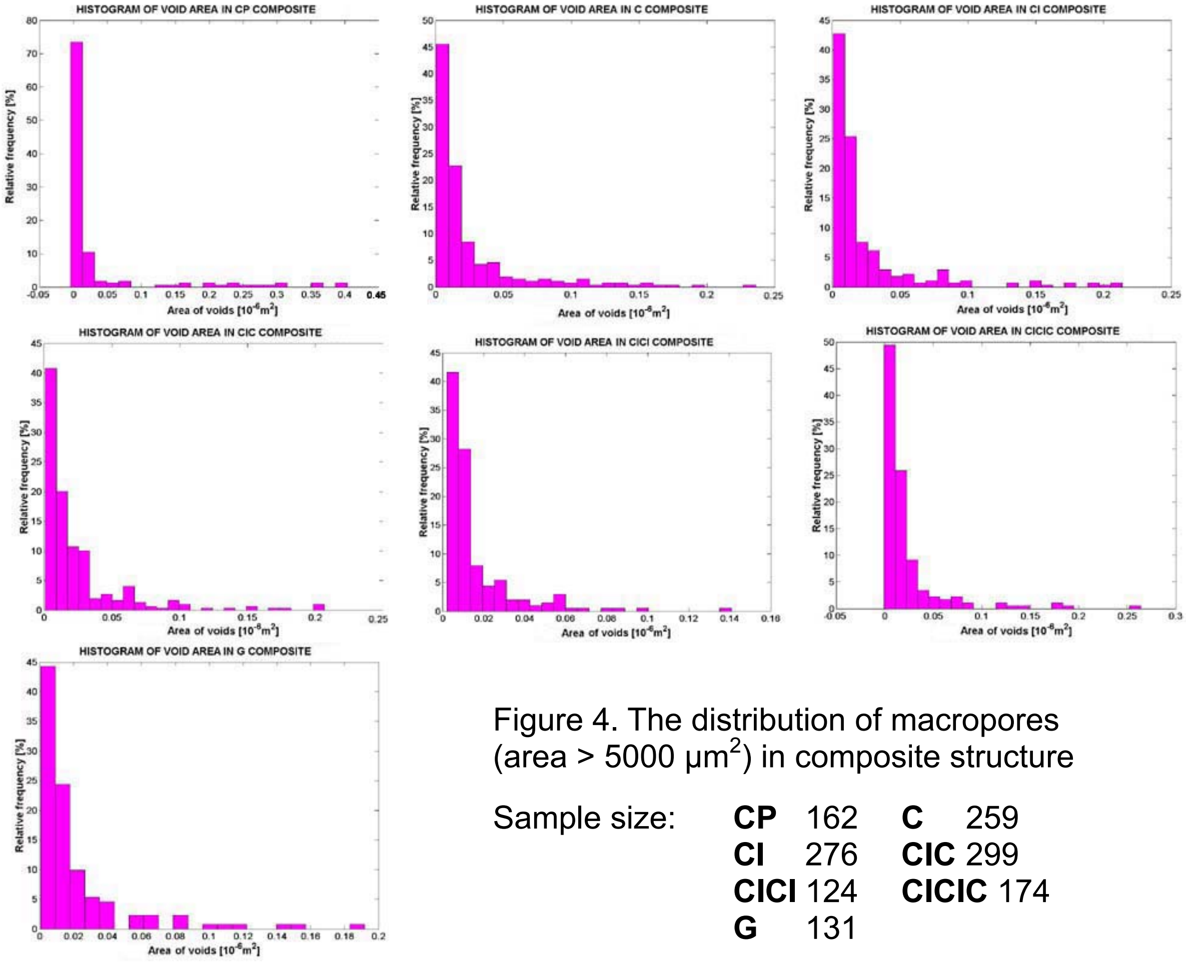
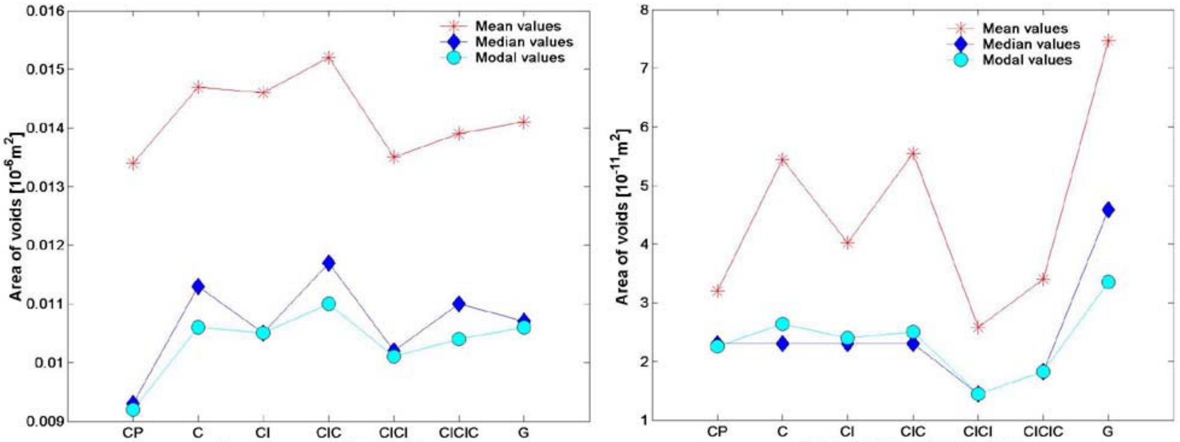


Figure 4. The distribution of macropores (area > 5000 μm^2) in composite structure

Sample size:				
	CP	162	**C**	259
	CI	276	**CIC**	299
	CICI	124	**CICIC**	174
	G	131		



a) **b)**

Figure 5. Statistical characteristics of measured void areas: a) for the group of voids with area greater than 5000 μm^2, b) for the group of voids with area under 5000 μm^2

Once the obtained results are evaluated the development of porosity and void structure may be described. There is obvious area reduction of macropores present in composite structure which proves the hypothesis of most extensive voids elimination due to technological process. This elimination also appears in decrease of statistical characteristics of macropores area, see Fig.5a).

On the other hand the results shown Fig.5b) display the increase of mean area of small pores in final graphitized composite. This indicates the graphitization of C/C composite improves the compactness of material in microscopic level.

The comparison of the vacuoles and delamination cracks occurence displays the influence of heating/cooling process on the morphology of most extensive voids, see Fig.6. These voids mostly effect the properties of final C/C composite.

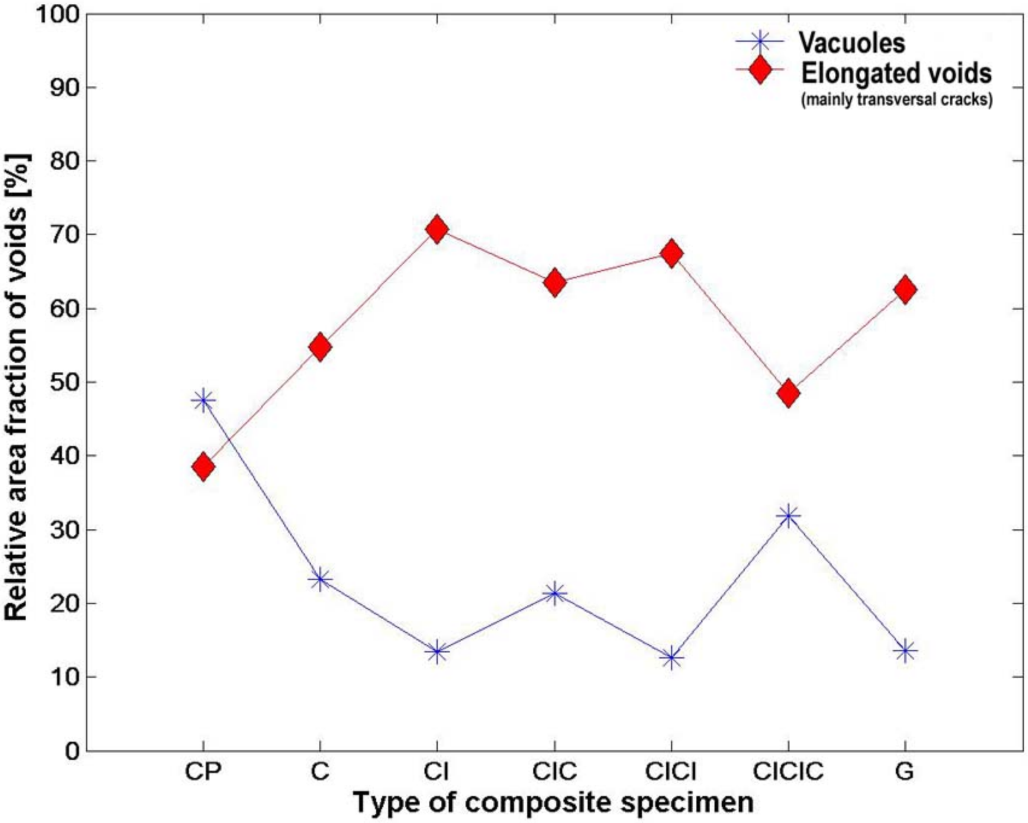


Figure 6. The occurence of transversal cracks and vacuoles
in individual composite specimen

Fig.6 shows the progress of delamination cracks, which are induced with the cooling processes, due to different thermal expansion of carbon fibers and carbon matrix. These voids are the most dangerous during the mechanical loading of C/C composites. Therefore its reduction is required.

On the other hand the growth of delamination cracks leads to the opening of the vacuoles, and enables its reimpregnation. The increase of open porosity is although significant for biocompatibility of composites, where certain amount of open macropores of defined size (100-450 μm in diameter for globular pores) is necessary for living tisue penetration [2,3].

Acquired knowledge of the extensive void progress indicates the indispensable influence of cooling process on composite material structure and signifies it as one of the feasible way to the control of the C/C composites porosity.

4 Conclusions

This paper presents the results of the initial study of void progress in C/C composites due to the technological process. It investigates the influence of composite technology on voids morphology and suggest basic characteristics to its description. The obtained results demonstrate the connection among the individual technological steps and development of various type of voids, mainly the extensive ones. Here belong the vacuoles, evolved due to the curing of CP composite, or delamination cracks, as a product of composite cooling, necessary consequence of heat treatment of composite material. Proper knowledge of C/C composites porosity may help the advancement of processes specialized in the control of void progress in material structure.

The improvement of porosity controlling increase the chances of the tayloring of C/C materials according to the application requirements. This may increase the fatigue life of C/C composites in technical or medical applications and save a lot of future costs connected to the use of C/C composites.

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