Defects in carbon plastics detected by acoustic microscopy

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Abstract. The paper shows the experience of using scanning pulsed acoustic microscopy (SIAM) to solve practical problems of quality control of polymer composite materials (PCM), to identify flaws and in-service damage, and to study changes in their internal microstructure under external influences. Acoustic images of structural defects and damages typical for carbon fiber reinforced composites are presented.

1. Introduction
Currently, polymer composite materials (PCM) based on carbon and glass fibers are widely used in mechanical engineering. Strong fibers serve as reinforcing elements and the polymer matrix as a binder, thus ensuring lighter weight of construction and high mechanical characteristics. The presence of multiple interfaces in the material leads to the fact that destruction processes occur under their influence and begin at the micro level [1-2]. Quality control of adhesion at the interfaces is one of the key tasks in the design of composite media and their production. Various microscopy methods are widely used to study the microstructure of PCM – optical, electron, confocal laser and atomic force microscopy, etc. [3-4]. These tools allow us to perform high-resolution studies of both surface and near-surface layers [5]. Damage analysis of the material is performed by fractography on specially prepared slices and fractures [6-8].

The presence of large and critical defects as well as premature aging of the material is detected by ultrasonic and x-ray non-destructive methods. High-resolution methods such as acoustic microscopy and x-ray computer tomography are used for the research purposes in the design of new PCM and improving their existing properties [9, 10]. Computer tomography (CT) allows obtaining high-resolution 3D images of the internal structure; however, there are restrictions on the samples size under investigation. For example, a 3D x-ray scanning tomograph TDM1000-IS/SP (Yamato Scientific Co., Ltd, Japan) allows getting images of the internal structure with resolution of 10 microns on samples of 20×10×2 mm [11]. For a number of practical tasks, such as studying the dynamics of material degradation processes under the effect of external factors, the size restriction makes CT method actually inapplicable.

Acoustic microscopy (AM) is an easy-to-use and safe method to observe the individual reinforcements and the binder pockets, to identify technological defects - pores and microscopic areas of adhesion losses. Pulsed acoustic microscopes with an operating frequency of 50-100 MHz provide a...
spatial resolution of about 50 microns at a depth of up to 5 mm. The design of an acoustic microscope does not imply a protective chamber and the size restrictions of the observed samples. The AM method requires immersion, but this technical problem can be solved by using water gels and other modern immersion media.

The paper presents the experience of using the acoustic microscopy method to detect technological defects and operational damages in the PCM structures. This review is relevant because the acoustic microscopes are not widely used in research laboratories. In addition, there is no document or standard for regulation such research yet. However, in modern machines and aircraft structures this method allows solving a number of problems related to identifying the causes of damage and describing defects at the microscopic level.

2. Method and materials

2.1. Acoustic microscope. The basic principles of the acoustic microscopy method are described in [3, 12 and 13]. A scanning pulsed acoustic microscope is used to study both surface and volume of material. The monitoring depth depends on frequency of the probing ultrasound, the acoustic lens aperture and the properties of the observed object. Signals reflected from internal structural elements, including defects and damages, are saved at each point of the scanning field and stored together with its coordinate. The resulting data allow getting the layer-by-layer acoustic images as the cross-sections which are orthogonal to the object surface (B-scan) or parallel to it and lying at a certain depth (C-scan).

The size of carbon fiber microstructural elements varies from 10-15 (fiber) to 200-300 microns (fiber layer thickness). Ultrasound with a frequency of 50-100 MHz perceives the carbon fiber laminate as a system of anisotropic layers, at the boundaries of which the radiation has reflection and refraction. The reflection at the interfaces depends actually on a thickness of thin binder (matrix) layer, which is varied in the range of 30-70 microns. It was shown [14] that the reflection from binder layer has a quasi-resonant character, which leads to acoustical contrast formation in the images of carbon fiber internal structure. Besides the reflection coefficient maximum occurs at the interface of various air voids and exfoliations as well with normal incident of the ultrasonic probe beam axis to the reflecting boundary. Focus diameter of high-frequency (50–100 MHz) acoustic lenses with a small angular aperture is several tens of microns. Thus, voids with a lateral size of about 100 microns or more are confidently detected in images.

In the paper, the scanning pulsed acoustic microscope (SIAM, IBCF RAS) was used. Acoustic lenses with operating frequencies of 50-100 MHz and the angular aperture of 11° were applied. The imaging resolution was 30-60 microns for the studied carbon fiber materials. The duration of the probing pulse was 25-40 ns, which allowed confidently distinguishing the signals from the laminate layers by the delay time. Result of the studying are presented in the form of layer by layer acoustic images in depth (C-scans).

2.2. PCM structural defects. Manufacturing of PCM products is accompanied by various defects which can adversely affect the material properties [15]. The most typical process defects and in-service damages of carbon fiber plastics as well the source of their occurrence are discussed below. There is information about the property of different PCM and their individual components - their hardness, strength, toughness, which is associated with their durability, reliability and stability [16]. But it is not the quality of the components that matters, but also the quality of the production technology. In the paper the images of carbon fiber reinforced plastics made with prepreg and infusion technology are presented. The last tendency is the winding technology application [17] and probably its combination with the prepreg one. Since the composite material consists of a matrix and reinforcing elements the adhesion loss between them is a common defect for all technologies.

For any methods of PCM production there is a probability that foreign objects or substances that are not regulated by the manufacturing technology (films, dust, hair, fat, etc.) may penetrate into the material. Inclusions can disrupt the orientation of fibers and laminate layers, form waviness, pockets of binder or air cavities.
In operation, the PCM failure can occur through various mechanisms but the several types of defects are formed as a result: delamination between PCM layers, matrix cracking and adhesion loss at the fibers-to-matrix boundary, fibers breaking. With prolonged exposure, there is a gradual accumulation of damages which leads to the total fracture. The whole spectrum of structural changes can occur simultaneously in the PCM material as it is caused by damage.

3. Results and discussion

The internal structure of carbon fiber reinforced laminate with binder pockets is shown in figure 1a. Our calculation reveals correlation between the amplitude of the reflected signal and thickness of the binder layer [19]. This dependence is one of the main mechanisms of acoustic contrast formation for displaying the internal structure of carbon fiber reinforced plastics without defects of material integrity. The echo signal acquires noticeable amplitude even at five-micron binder layer. Thickening of the binder around the individual carbon threads leads to increasing the contrast in acoustic images and has resonance behavior depend on wave length of probing radiation. The formation of folds of the reinforcing layers and pockets of the binder is thus clearly observed in acoustic images for woven composites. In our case it is defect of fiber packing and the binder pockets (1) around it.

In figure 1b the defects of adhesion loss between the composite layers are presented – extended delamination (2) and partial contact (3). The defects were the result of ingress of anti-adhesive lubricant and insufficient amount of binder with high volatile elements content that is characteristic for the layered composites structure made by prepreg technology. Due to the high reflection coefficient of ultrasound at the interfaces with air the AM method enables to detect delaminating well even with a small gap. The extended defect (2) is a big area with a high acoustic contrast. The second defected area (3) consists of multiple microscopic zones of partial adhesion loss or kissing contact. Due to the microscopic sizes of the reflectors they don’t be detected with usual ultrasonic equipment (5-25 MHz). At low-frequency the echo-signal amplitude will be decreased depend on microdefects clustering that needs of an additional interpretation and adjustment of the equipment sensitivity. Due to the focusing and high frequency (>50 MHz) using acoustic microscopy allows revealing the partial contact area that is hidden and dangerous in operation [20].

Figure 1. Acoustic microscopy of carbon fiber reinforced (CFR) composite internal microstructure, technological defects: (a) – binder folds (1), (b) – delaminating (2) and partial contact (3), (c) – local adhesion losses at fiber-to-matrix boundary (4) and (d) – carbon black inclusions (5).

Insufficient impregnation of the fiber reinforcing layers and curing conditions of the polymer binder in the process of composite manufacturing can cause weak adhesion at fiber-to-matrix boundary. Local detachments can growth under mechanical loads afterwards [17, 18]. In figure 1c initial structure of carbon fiber reinforced plastic (CFRP) with partial adhesion loss at fiber-to-matrix boundary is presented. Defects are located at the matrix along the reinforcing components and are seen as multiple bright elongated spots (4).

In figure 1d the internal structure of CFRP with the carbon black particles is presented. Inclusions are detected in acoustic images depending on the ratio of their size to sound wavelength and value of
acoustic impedance at the reflecting boundary [14]. In the image (figure 1(d)) there are reinforcing structure of the composite sample with grey scale and several particles of 100–300 microns size as bright spots (5).

Our extensive practice in CFRP structural studying with scanning pulsed acoustic microscopy shows that the method is the effective non-destructive tool for the assessment of the composite structure quality, in the process of the new materials design and their tests [21]. In operation, the PCM failure can occur through various mechanisms. In fact the several types of damaging are formed: delamination between composite layers, matrix cracking and adhesion loss at the fibers-to-matrix boundary, fibers breaking. With prolonged exposure, there is a gradual accumulation of damages which leads to the total fracture.

The whole spectrum of structural changes can occur simultaneously in the PCM material under damaging impact. For example, the low-speed impact results in matrix cracking, fiber breaking and delaminating. In figure 2(a) shows damage after the low-speed impact with energy 30 J. The surface indentation of 450 \( \mu \)m deep was hardly visible and extended multiple delaminating with maximal lateral size of 40 mm was found in the composite bulk. Delaminating in each layer extends accordingly to filaments orientation. Damages are distributed throughout the entire volume 32\textsuperscript{6} layers specimen (6.7 mm). Results were given by the authors previously in [22].

Figure 2. Acoustic microscopy of CFR composite internal microstructure, operation damages: (a) – multiple delamination after low-speed impact (1), (b) – matrix cracking in [45,90]\textsuperscript{2S} laminate under the tensile test (2), (c) – cross section (B-scan) and (d) – longitudinal section (C-scan) of large inclined matrix crack in unidirectional composite after mechanical test (3).

In the literature, there are several terms that denote defects in the matrix – matrix cracking, transverse cracks, etc. [10, 17]. The microscopic cracks in matrix could be initiators of the composite structure failure and effect on thermomechanical properties, including degradation in modules, the Poisson's ratio and a thermal expansion coefficient. Acoustic imaging of the matrix cracks are presented in figure 2(b). They are formed in CFR specimen with fiber package [90/45]\textsuperscript{2S} under the tensile load 1250 N/mm\textsuperscript{2}. Cracks are visible as the dark lines of 100 \( \mu \)m opening. There is no acoustic reflection from such cracks caused by their boundaries are vertical. It is possible also that they are a cluster of microcracks with 1–2 \( \mu \)m size that allows considering them as point source scatters and in the case the scattered signal wasn’t enough to make contrast in the image.

Actually, the matrix cracks can be the potential reservoirs for gases and liquids and can be the cause of rapid irreversible changes in the material structure. In figures 2c and 2d the large matrix crack (3) in CFR composite structure is shown. The sample was prepared as unidirectional composite by winding technology with 65% of fibers bulk content and 20x200x4 mm size. It was loaded for the tensile along filaments with stops for microstructural investigation. In the image of longitudinal section (C-scan, figure 2d) the shape of the crack is seen. It is inclined and developed in depth with rounded front. The extended cracks located along the reinforcing elements inside the binder between fibers layers are characteristic for the last stage of composite destruction process that is before the integrity loss. Damage shown in figures 2c and 2d was located in the zone of maximal mechanical stress.
concentration. Acoustic contrast in the area of damage is enhanced for planar elements, which are perpendicular to the axis of the probing beam. Clustering of such elements increases the reflected signal amplitude significantly. The inclined cracks could be observed in acoustic images due to relief on the crack borders and an acceptable angle of their inclination to the axis of the probing beam.

**Conclusion**

Experiments show that the scanning pulse acoustic microscopy method is an effective non-destructive tool for evaluating the quality of composites in the process of designing and testing new material. The experimental results for carbon fibers reinforced plastics is presented in the paper. In the range of ultrasonic frequencies of 50-100 MHz both characteristic technological and operational defects with a lateral size of more than 100 microns are accurately detected – delamination between the composite layers, matrix cracking and loss of adhesion at the fiber-matrix interface, fiber rupture.

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