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EVALUATION OF THE POSSIBILITY OF APPLYING SPATIAL 3D IMAGING USING X-RAY COMPUTED TOMOGRAPHY RECONSTRUCTION METHODS FOR QUANTITATIVE ANALYSIS OF MULTIPHASE MATERIALS

RENTGENOWSKA ANALIZA ILOŚCIOWA MATERIAŁÓW WIELOFAZOWYCH Z WYKORZYSTANIEM PRZESTRZENNEGO OBRAZOWANIA (3D) PRZY UŻYCIU METOD REKONSTRUKCJI TOMOGRAFII KOMPUTEROWEJ

In this paper the possibility of using X-ray computed tomography (CT) in quantitative metallographic studies of homogeneous and composite materials is presented. Samples of spheroidal cast iron, Fe-Ti powder mixture compact and epoxy composite reinforced with glass fibers, were subjected to comparative structural tests. Volume fractions of each of the phase structure components were determined by conventional methods with the use of a scanning electron microscopy (SEM) and X-ray diffraction (XRD) quantitative analysis methods. These results were compared with those obtained by the method of spatial analysis of the reconstructed CT image. Based on the comparative analysis, taking into account the selectivity of data verification methods and the accuracy of the obtained results, the authors conclude that the method of computed tomography is suitable for quantitative analysis of several types of structural materials.

Keywords: computed tomography, electron microscopy, X-ray diffraction, stereology, phase analysis, quantitative research,

W pracy przedstawiono możliwość wykorzystania rentgenowskiej tomografii komputerowej (CT) w ilościowych badaniach metalograficznych materiałów jednorodnych oraz kompozytowych. Badaniom porównawczym poddano wypraskę mieszaniny proszkowej Fe – Ti, żeliwo sferoidalne, a także kompozyt epoksydowy zbrojony włóknami szklanymi. Udział objętościowy poszczególnych składników strukturalnych określano za pomocą klasycznych metod stereologicznych z wykorzystaniem skaningowej mikroskopii elektronowej (SEM) oraz metodami rentgenowskiej analizy ilościowej (XRD). Uzyskane typowymi metodami wyniki porównano z rezultatami otrzymanymi za pomocą metody przestrzennej analizy obrazu tomografii komputerowej. Na podstawie przeprowadzonych analiz, biorąc pod uwagę selektywny sposób weryfikacji danych oraz dokładność uzyskiwanych wyników, można stwierdzić przydatność wykorzystania metody tomografii komputerowej, w obszarze ilościowej analizy strukturalnej szerokiego spektrum materiałów konstrukcyjnych.

1. Introduction

Modern structural materials, which currently include a wide range of materials - metals, polymers, ceramics and composites, as well as those being in the research phase, like for example fullerenes, thanks to a set of specific physico-chemical characteristics are finding diverse, ever more specialized use in all sectors of economy. At the same time, wider and wider range of materials available in the market forces the use of new research techniques that enables faster, more versatile, and unambiguous definition of the structural and functional properties, that would help in the proper selection of materials.

One of these modern methods are 3D techniques used, among other things, in modern computer numerical control (CNC) tooling machines, 3D scanners and printers, coordinate measuring machines (CMM) or incremental technologies like

RP (rapid prototyping). In addition to the typical manufacturing applications, 3D techniques are also widely used in laboratory structural studies [1-10], qualitative and quantitative analysis [6-10], or other high-speed measurements of specific materials' properties, made for the sole purpose of manufacturing processes. The versatility of these research methods allows its use in many industries, from timber industry (quality assessment of wood at sawmills) [4], to the highly specialized electrotechnic and electronic companies [2], to medical applications [5]. The X-ray computed tomography (CT) is also a 3D technique method, which popularity is evident mainly in the spatial imaging of material defects, cracks, different types of discontinuity or latent defects. It is used primarily in the automotive industry, where, in factories of some vehicle manufacturers, engine blocks and critical structural components of vehicles or bulky products are subjected to tomographic examinations [11]. X-ray methods are also

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used in stationary and mobile stations at border crossings (in Europe, parts of cars and even entire trailers and vehicles are inspected in this manner).

The devices used in the spatial imaging reconstruction method, are not limited to research areas mentioned above. They are also adapted for a wide group of recipients, that need information about the state of the object, product or material, concerning both the state of the surface and changes inside the volume [12-16].

Such attempts in application of CT in metallographic research were also undertaken in the Department of Advanced Materials and Technologies, MUT, however, due to the current lack of any normative documents in this regard, some problems were encountered concerning the extent and manner in which the standardized materials' tests should be conducted with this type of equipment. Numerous research works [1-17] and the documents of the VDI/VDE 2630 association from the year 2009 [17] allow only for the determination of the accuracy associated with the geometry measurement errors, scanning errors, determination of scan resolution or dimensions-material dependence.

Therefore, in this study, to examine the usefulness of CT techniques for qualitative and quantitative analysis of phase structure in material science, a group of typical construction materials, ranging from classic cast iron, to powders compact made of iron and titanium powder, to typical polymer composite reinforced with glass fiber, was selected. Such a choice of materials belonging to different material groups, due to the different contents of phase components, matrix and precipitations, or matrix and reinforcing fibers [18], enforces the search for universal and accurate research methods, which would allow for rapid assessment of the state of materials. As suggested by the authors of papers [18-22], there is still a need to find and implement standardized testing methods that could be used in different technological processes for the study of new material properties. Equally important is to find methods of non-destructive testing, allowing the detection of possible sources of damage, threatening the security of future exploitation, especially in the case of different types of materials, including metallic and non-metallic, that must meet the high performance requirements.

Therefore, in this paper the measurement accuracy of tomographic technique quantitative analysis has been presented in comparison to conventional techniques such as RIR (Reference Intensity Ratio) quantitative X-ray phase analysis and stereological analysis technique with the use of a scanning electron microscope (SEM).

2. Material and methods

Sets of samples of three different materials, with the parameters listed below, were prepared for this study. First was the spheroidal cast iron in the form of a cylinder with a diameter of 4 mm and a height of 6 mm. Second material was cylindrical compact of technically pure iron and titanium elemental powders mixtures, prepared by uniaxial pressing. Pellets had a diameter of 3 mm, height of 4 mm and were prepared from powder mixtures with three

different content of iron (20, 50 and 80 wt.%). The last material was epoxy composite reinforced with glass fiber and with a ceramic filler element. The sample had the shape of a rectangular prism, with the dimensions of 10x10x10 mm, and contained 19 layers with differently oriented glass fiber reinforcement.

The identification of the observed phase structure was made on a Rigaku Ultima IV diffractometer, using $\text{CoK}\alpha 1$ radiation (voltage 40 kV, current 40 mA). The 2θ angle scanning range of a parallel beam ranged from 30 to 150 degrees, at a speed of 1 deg/min. With the help of XRD technique a qualitative and quantitative analysis of cast iron and powder mixtures compacts was carried out, and the volume fractions of the phase structure components were specified by the use of the reference standard method RIR.

Due to the impossibility of the use of XRD technique for quantitative structural analysis of the epoxy composite, tests were performed by observing the sample with the use of the Quanta 3D FEG Dual Beam (SEM) microscope under lowered vacuum conditions, with the magnification x70. In order to determine the ratio of matrix and reinforcement volume fractions, SEM pictures were analyzed with the use of the microscope's stereological software.

The results of XRD and SEM analysis was then compared with the results of the spatial analysis of reconstruction images performed by the computer microtomograph Nikon / Metris XT H 225 ST, equipped with open tube ultrafocus reflection target radiation source, with a maximum accelerating voltage of 225 kV. Several thousand CT pictures (accurately 3125), taken with the 4 mln pixels detector matrix, were reconstructed in to 3D representations, with the voxel resolution (linear dimension of 1 cubic 3D pixel) of 1.5 μm .

3. Results and discussion

3.1. Spheroidal cast iron

The X-ray phase analysis performed on a sample of the cast iron revealed the phase structure components typical for this type of material, namely: a solid solution of carbon in ferrite - $\text{Fe}(\alpha)$; iron carbide, Fe_3C – cementite; and the free form of carbon - graphite (Fig.1). The content of the individual components specified by RIR was as follows: $\text{Fe}(\alpha)$ - 87 wt.% (70% vol.), cementite – 3 wt.% (3% vol.) and graphite - 10 wt.% (27% vol.). In the case of tomographic analysis only two structural components were identified, namely the matrix (composed of ferrite and cementite) in an amount of 88 wt.% (68% vol.), and 12 wt.% of graphite (32% vol.). One can already see significant differences in research capabilities of compared techniques. While the XRD analysis allow to determine precisely the type and the contribution of individual structural components, the CT due to the similar X-ray density of the ferrite and cementite (the difference is of about 2%) are not able to separate the matrix components. However, tomographic technique allows to determine the morphology of graphite precipitations, in this case spheroidal and homogeneity of their distribution in the matrix (Fig.2), which is impossible in the case of XRD studies.

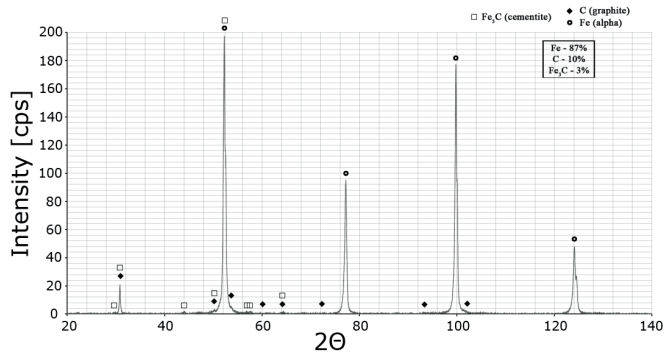


Fig. 1. XRD diffractogram for sample of cast iron with weight percentages of individual phase components calculated via RIR methodology

3.2. Metal powders compact

The problem with distinguishing between cast iron matrix components, which do not differ significantly in terms of X-ray density, prompted the authors to perform the CT - XRD comparative analysis for two-phase materials (Fe-Ti powder compacts), made from ingredients of relatively similar atomic masses, while varying their mutual ratio in the structure. As in the case of a cast iron sample, the XRD technique confirmed its effectiveness in qualitative and quantitative analysis of titanium and iron samples. Results obtained from diffraction patterns (Fig.3) are consistent with the designed composition of the studied compacts, with a maximum error of 6 wt.% (Table 1). Comparable and

even a little better values, with maximum 4 wt.% error were obtained for the CT technique analysis (Table 1), however, as mentioned earlier, spatial analysis of CT reconstructions, in addition to specifying the volume fraction of individual components, allowed also for the analysis of the morphology and uniformity of distribution of the individual elements used to produce the compacts (Fig.4). It is also possible to determine the size distribution of the precipitates of the individual components in a volume of the structure (various colors that appear in the reconstruction image - figure 4c - correspond to the different sizes of the iron powder particles) and to determine whether they are dispersed or if they form conglomerates.

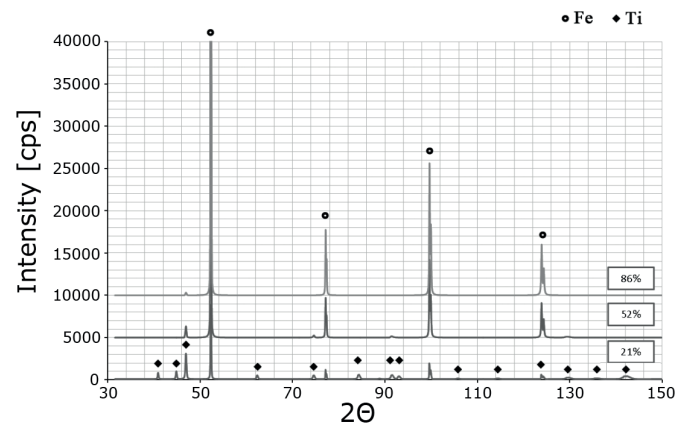


Fig. 3. XRD diffractogram for samples of Fe-Ti compacts with weight percentages of iron calculated via RIR methodology

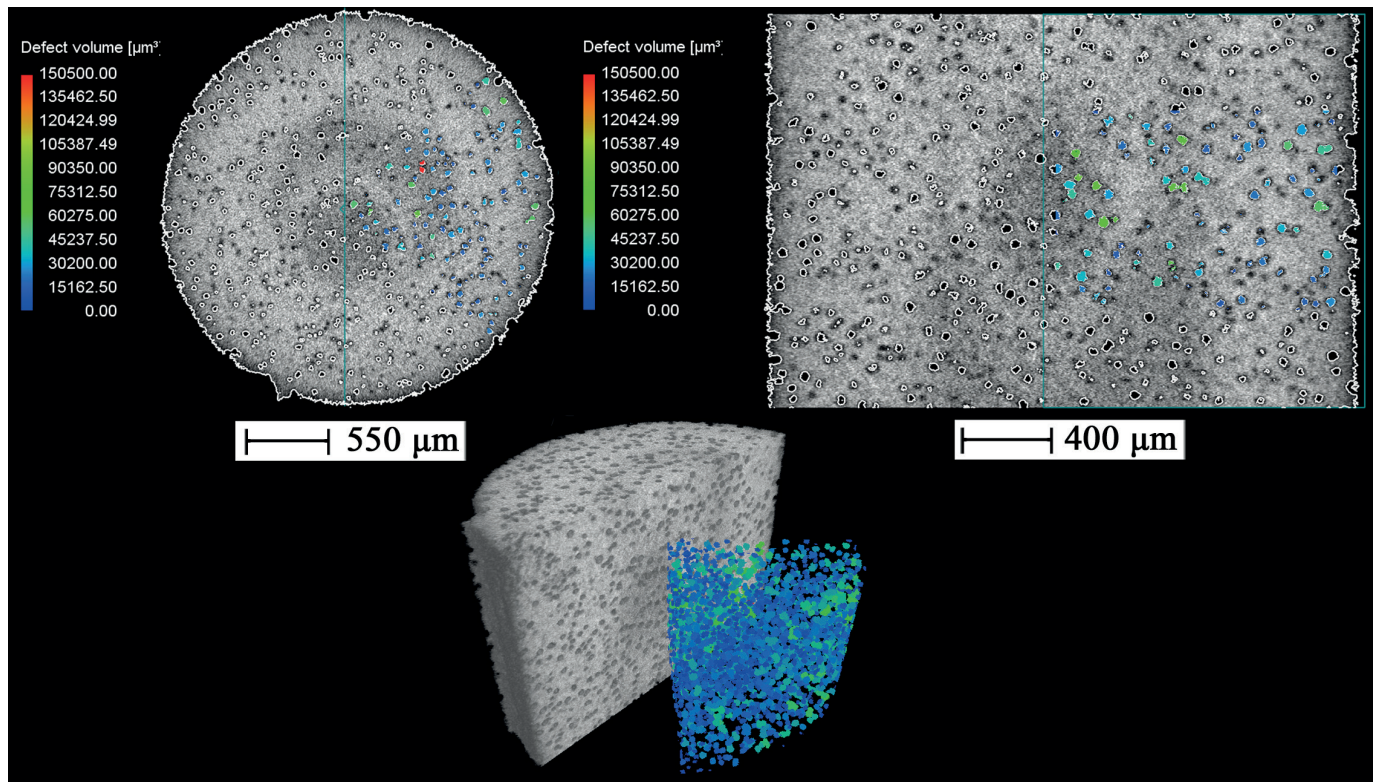


Fig. 2. CT reconstruction quad image with three side view cross-sections and 3D half-clipped representation of spheroidal cast iron sample with highlighted graphite precipitations

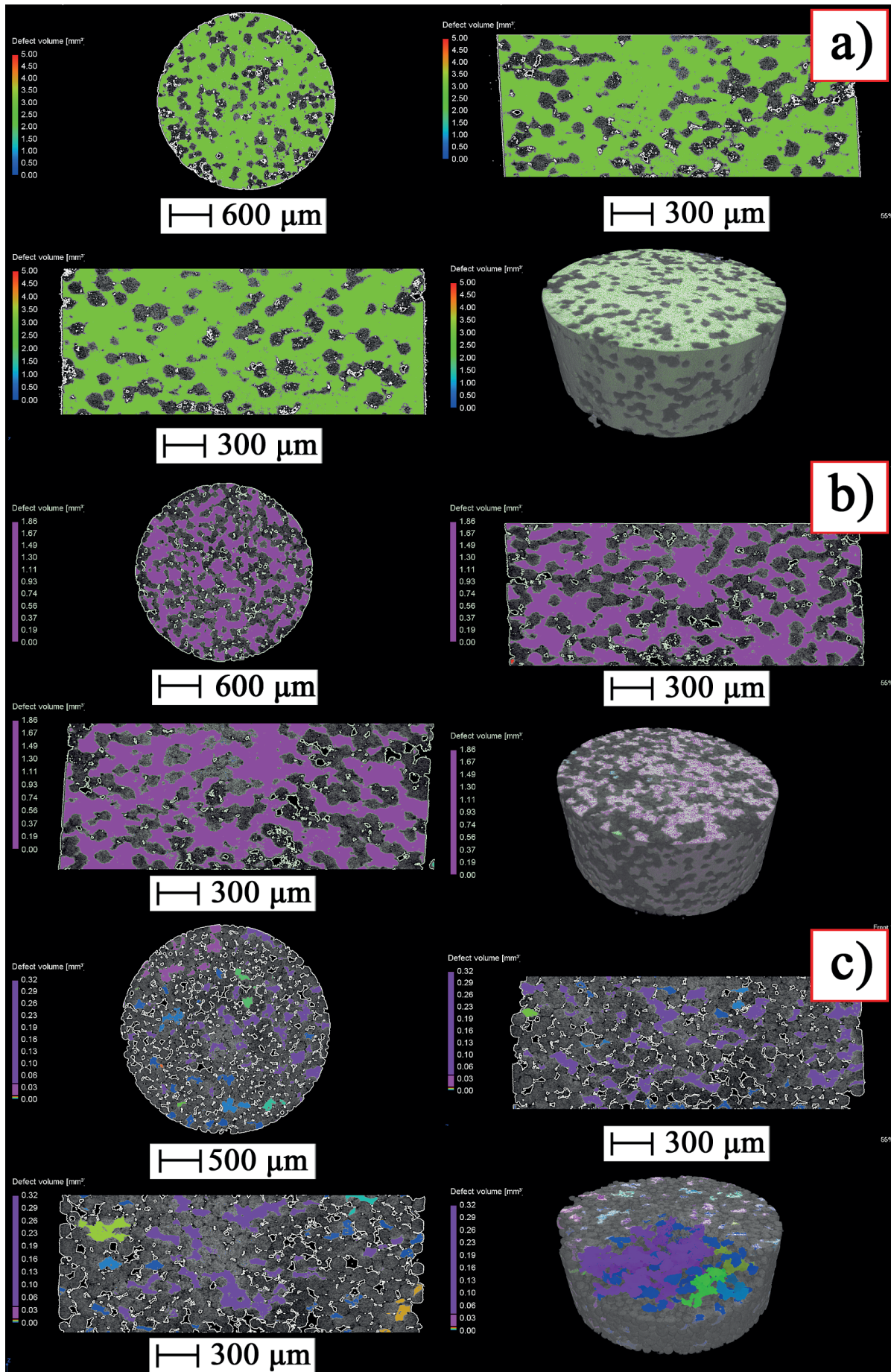


Fig. 4. CT reconstruction quad images of Fe-Ti compacts with a) 80 wt.% of iron (green color), b) 50 wt.% of iron (pink color), and c) 20 wt.% of iron (with different colors representing different sizes of iron particles)

It is also worth mentioning that in the case of XRD phase analysis one scan can only reveal the structure of material in relatively thin surface layer of the cross-sectioned material. And only by preparing another cross-sections, deeper into the material volume, and repeating diffraction tests one can analyze eventual changes in material's phase structure in different parts of its volume (semi-tomography). Whereas CT technique allows for analysis of entire sample volume in one scan process, which greatly reduces time and cost of metallographic research.

3.3. Epoxy - fiberglass composite

In the case of materials with non-crystalline structure (like proposed fiberglass reinforced epoxy composite,) the use of X-ray diffraction qualitative and quantitative analysis is not possible. Therefore, the assessment of the quantity of each phase component was based on the stereological analysis of the structure observed by scanning electron microscopy (Fig.5). From the manufacturing process data, it is known that the analyzed composite is composed of 19 zones (layers), in which the reinforcement, in the form of alternately arranged fibers and glass mat, is characterized by an increasingly smaller diameter of the fibers. The area of zones from 1 to 10 comprises of four glass mats with fibers' diameter of 600 μm , interspersed with layers of 450 μm diameter filaments. Zone 11 is an area of the filler with small ceramic particles, while from 12 to 18 are zones made from loose glass fibers with varying diameters of sequentially 450, 300 and 150 μm . The last 19th zone is a gelcoat protective layer.

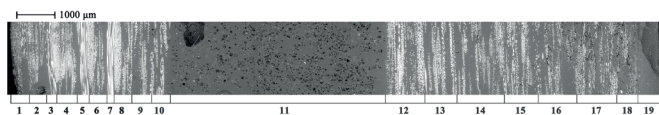


Fig. 5. SEM image of layered structure of glass fiber reinforced epoxy composite with indicated zones of different reinforcement configurations

Microscopic analysis of the cross-section of the composite confirms the layered, highly heterogeneous structure of this engineering material. The observed inhomogeneous distribution of the reinforcement and the matrix constitutes a fundamental difficulty in assessment of the contribution of different structural components in the composite volume. The correctness of this verification depends on many factors, which include, among other things, the orientation of cross-section plane, the resolution of the analyzed image, the direction of the fibers in layers. Nevertheless, with the use of computer software (Nicon NIS Elements BR) for stereological analysis of the SEM image, the ratios of glass fibers to epoxy matrix volume fraction was determined for individual zones (Fig.6). Values of volume fraction was computed by using binarized image evaluation algorithms. This computer software after selecting the appropriate elements of the observed microstructure was automatically executed all the calculations.

In the case of CT techniques, despite obtaining similar (to SEM) image of the cross section of the analyzed material (Fig.7) the results for the mutual participation of

the reinforcement and the matrix are considerably different (Fig.8). The average volume fraction of glass fibers in zones 1-10 and 12-18 are 33.9% and 18.6%, respectively, for SEM analysis, which are within 1% deviation from the production data of the composite. Whereas the same areas analyzed by means of computed tomography, yielded results of 54.4% and 60.9%, which are, respectively, about 1.6 and 3.3 times higher.

The primary reason for such a significant differences in the obtained results is the measurement methodology. In the case of SEM stereology, analyzed image is two-dimensional and it is assumed that tested material has similar structure deeper into the volume, and this assumption allows to determine, with a certain error, the volumetric parameters. Whereas in the case of CT techniques, observed 2D image, as mentioned earlier in paragraph 3.1, is derived from three-dimensional reconstruction (Fig.9a,b) which allows for the selection of any representative or characteristic area or volume for analysis. As in previous reported cases CT technique allows to determine the morphology, shape and other characteristics of the geometry of the structures in the volume of the test material, which is impossible in the case of X-ray diffraction XRD, or severely restricted in the case of microscopic observation. This is a powerful advantage of computed tomography in various types of material research and characterization. Unfortunately, the correctness of analysis of CT reconstructions depends on several factors, the most important of which are: resolution of reconstructed image (which is much lower than that of SEM images), degree of reconstructed geometry deformation, quality and contrast of base X-ray transmission images and material phases' X-ray density contrast. These parameters strongly influences the ability to differentiate material from background and one material phase from other in the reconstructed volume. However, the information about the surface of the fibers, glass mat and quality of the composite in different parts of entire analyzed volume obtained by computed tomography allow for more reliable (from stereological) assessment of the quality the material production process.

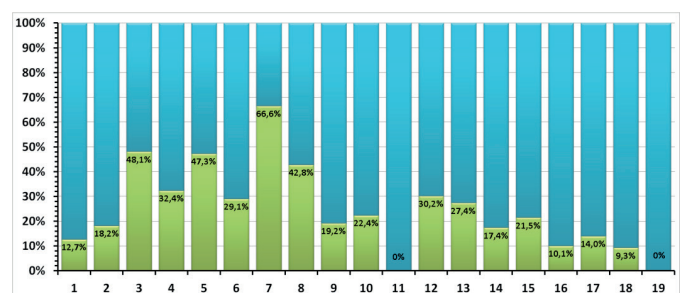


Fig. 6. Volume fraction of glass fibers in different zones of composite's epoxy matrix obtained by stereological analysis of SEM images

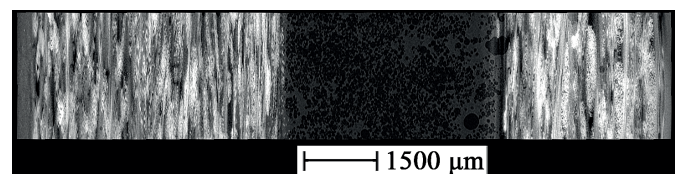


Fig. 7. CT reconstruction image of layered structure of glass fiber reinforced epoxy composite

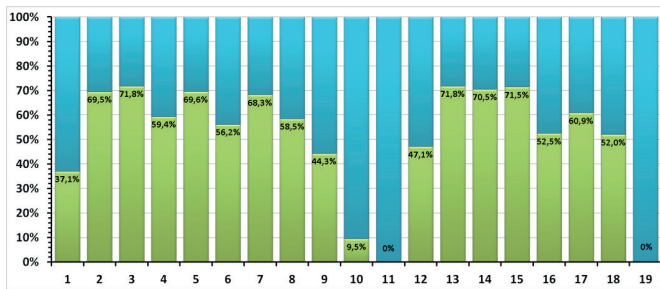


Fig. 8. Volume fraction of glass fibers in different zones of composite's epoxy matrix obtained by spatial analysis of CT reconstruction

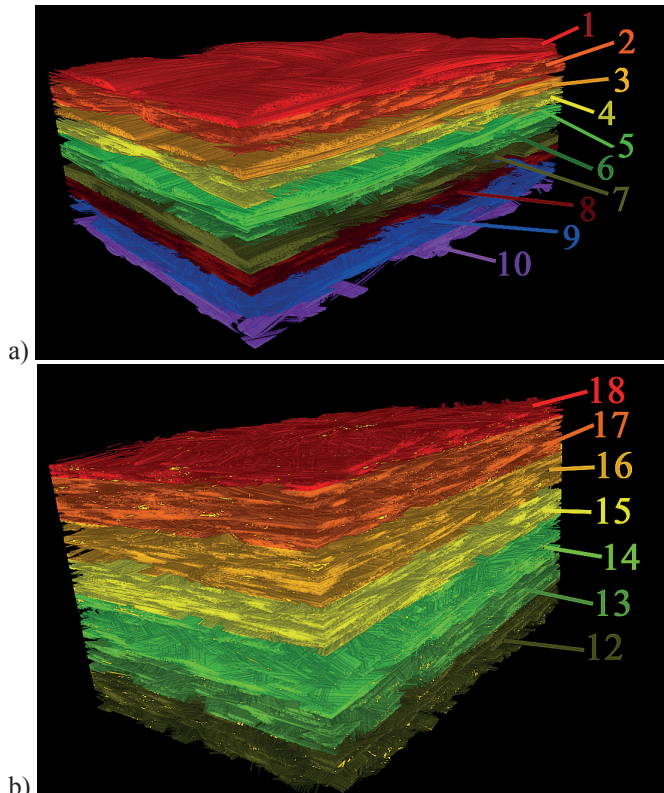


Fig. 9. CT reconstruction 3D image of composite's zones 1-10 (a) and 12-18 (b) with separated fiberglass reinforcement layers

4. Conclusions

Unfortunately the X-ray CT technique can't match the capabilities of the phase detection that a X-ray diffraction (XRD) has, or isn't able to achieve such image resolution that is obtainable by scanning electron microscopy (SEM). However, based on the experimental results, authors conclude that this methodology is suitable for the rapid assessment of the basic structural properties of construction materials and reduces time and cost of metallographic research.

In case of comparing phase quantitative analysis made by XRD and CT techniques for two metallic materials, the differences between components' volume fractions obtained from those techniques oscillates in range of 2-3 wt.% for samples of cast iron, and 1-7 wt.% for compacts made of Fe-Ti powders mixtures. Significantly greater variations were observed in comparison of the results of SEM observations with spatial analysis of the reconstructed composite sample.

Those discrepancies are mainly connected with the difficulties with proper separation of fiberglass reinforcement from epoxy matrix due to very inhomogeneous structure of the composite and insufficient resolution of tomographic image. Nevertheless, the ability to observe sample's entire internal structure, within fully interactive 3D environment, constitutes a great advantage of computed tomography.

Additional information about the size of the particles, their orientation relative to each other, deformation, and information on porosity, cracks should soon be reflected in the normative documents [16-17]. Similarly to stereological, comparative with standards, measurements performed nowadays, that allow you to conduct quantitative analysis, the standards for computed tomography, in the future, will allow you to quickly collect standardized quantitative information, which are absent in the present day metallographic CT tests, which may become more common as a result of combining different research techniques - cumulative techniques.

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